TOTAL SYNTHESIS OF 2-AMINOIMIDAZOLE ALKALOIDS FROM *LEUCETTA* AND *CLATHRINA* SPONGES

by

PANDUKA B KOSWATTA

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ABSTRACT

TOTAL SYNTHESIS OF 2-AMINOIMIDAZOLE ALKALOIDS

FROM *LEUCETTA* AND *CLATHRINA* SPONGES

Panduka B. Koswatta, PhD

The University of Texas at Arlington, 2010

Supervising Professor: Carl J. Lovely

Sponges of the major class, *Calcarea* can be found in nearly all marine habitats. Two *Calcarea* genera, *Leucetta* and *Clathrina* have found be rich with 2-aminoimidazole alkaloids. The first alkaloid of these groups was isolated in 1987 from a bright yellow sponge of the class *Calcispongiae (calcarea) and Leucetta chagosensis dendy (Leucetta)* collected near Na'ama in the Gulf of Eilat (Aqaba) in the Red Sea. The nomenclature for this whole series of compounds is based on the location of the initial source material. The first Chapter describes the isolation, biological activities, possible biosyntheses and total synthesis of these alkaloids. The last section of this Chapter describes the retrosynthetic approach to three of these alkaloids, calcaridine A, spirocalcaridine A and spirocalcaridine B *via* oxidative reaction of corresponding 4,5-dibenzylicimidazole derivatives.

The second Chapter describes the studies of oxidative reactions of Davis, Aryl *N*-sulfonyloxaziridine with tetrahydrobenzimidazole (THB) derivatives to provide

corresponding spiro-fused 5-imidazolones or in some cases bis-addition products, depending on the nature of the C2-substituent and the solvent employed. The comparison of oxidative abilities of DMDO and oxaziridine is also described later in this Chapter, and it has been found that while stereoselectivity of both reagents are the same, oxaziridine shows relatively slow reactivity towards complex molecules thereby requiring higher reaction time probably due to the steric clashes between the oxidant and the reductant. It has also been found that both DMDO and oxaziridine can be used for the oxidative addition across imidazole C4-C5 double bond to synthesize corresponding dihydroxy imidazolidines.

In Chapter three, total synthesis of (±)-*epi*-calcaridine A and (±)-calcaridine A is described using position selective metalations of 4,5-diiodo-1-methyl-1*H*-imidazole to provide a 14-methoxynaamine A derivative, which was subjected to oxidative rearrangement with 3-(4-nitrophenyl)-2-phenylsulfonyloxaziridine to construct the imidazolone core of calcaridine A, and this was the first examples of rearrangements involving substrates other than a tetrahydrobenzimidazole type derivative. In addition to the total synthesis, relative stereochemistry of the natural product has been determined through X-ray crystal structure of the epimeric congener.

Total syntheses of six other *Leucetta* derived alkaloids, preclathridine A, clathridine A, naamidine G, 14-methoxynaamidine G, naamine G and naamidine H are described in Chapter four, by extrapolation of the approach described in the second Chapter. In these two chapters we demonstrated that 4,5-dihaloimidazoles can be functionalized in a sequential and controlled manner in the order of C5→C4→C2 using Grignard reagents (for the 4- and 5-positions) and *n*-BuLi (for C2).

The studies described in the fifth Chapter are directed toward the total synthesis of spirocalcaridine A and B. Several approaches developed based on the biosynthetic consideration, starting from 4,5-diiodoimidazole and *ipso*-cyclization of 4-aryl trienone *via de novo* synthesis of imidazole ring, are described in this Chapter.

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LIST OF ABBREVATIONS

AcOH	Acetic acid
AIBN	Azobisisobutyronitrile
Boc	*tert*-Butyloxycarbonyl
DEPT	Distortionless Enhasment by Polarization Transfer
DIBAL	Diisobutylaluminum hydride
DIEA	Diisopropylethyl amine
DMAP	4-Dimethylaminopyridine
DMAS	*N,N*-Dimethylaminosulfonyl
DMDO	Dimethyldioxirane
DMEDA	*N,N'*-Dimethylethylenediamine
DMF	*N,N*-Dimethylformamide
DMP	Dess-Martin periodinane
FT	Fourier Transform
HMPA	Hexamethylphosphorictriamide
HRMS	High resolution mass spectroscopy
LDA	Lithium diisopropylamide
LTMP	Lithium tetramethylpiperidide
MOM	Methoxymethyl
Ms	Methanesulfonyl
NBS	*N*-Bromosuccinimide
n-BuLi	n-Butyl lithium

NIS	*N*-Iodosuccinimide
NMR	Nuclear magnetic resonance
NOE	Nuclear overhauser effect
NOESY	Nuclear overhauser effect spectroscopy
PCC	Pyridinum chlorocromate
Phth	Phtalimide
Py	Pyridine
SEM	2-Trimethylsilylethoxymethoxy
TBAF	Tetra-n-butylammonium fluoride
TBS	*tert*-Butyldimethyl silyl
t-BuLi	*tert*-Butyl lithium
Tf	Trifluoromethylsulfonate
TFA	Trifluoroacetic acid
THF	Tetrahydrofuran
TMEDA	*N,N,N',N'*-Tetramethylethylenediamine
TMS	Trimethyl silyl
Tris	2,4,6-Triisopropyl phenyl
Ts	*p*-Toluenesulfonyl
δ	Chemical shift (NMR)

CHAPTER 1

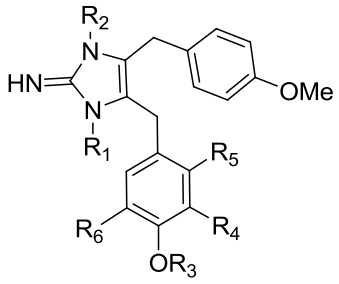
INTRODUCTION

Marine sponges are a rich source of alkaloids with interesting biological activities and often exhibiting unique structural frameworks. Among these, sponges of the major class, *Calcarea* can be found in nearly all marine habitats. Two *Calcarea* genera, *Leucetta* and *Clathrina* have found to contain more than sixty examples of imidazole alkaloids during last three decades (Figures 1.1-1.7).[1]

1.1 Isolation of 2-aminoimidazole alkaloids from *calcarea* sponges

The majority of these alkaloids have been isolated during the last thirty years and they show close similarities in their structural features. For example, the vast majority contains a central 2-aminoimidazole ring, substituted by one or two functionalized benzyl groups in some combination of the *N*1, C4 and C5 positions and in some cases the 2-amino moiety is further substituted with a hydantoin or hydantoin derivative. In most cases the 4,5-dibenzylic derivatives are known as naamines or naamidines (Figures 1.1 - 1.3) and the 1,4-dibenzylic derivatives are known as isonaamines or isonaamidines (Figure 1.4). To date, no studies have been reported detailing the biosynthetic origin of these natural products (see in section 1.2), and thus it is not clear if naamines are precursors for naamidines or *vice versa*, or whether they are formed from completely different pathways. Analogous mono benzylic derivatives are shown in Figure 1.5. In addition to these benzylic alkaloids, a few other examples

containing a naphtha[2,3-*d*]imidazole ring system, presumably biogenetic relatives of above alkaloids, have been isolated in the course of these investigations (Figure 1.6).



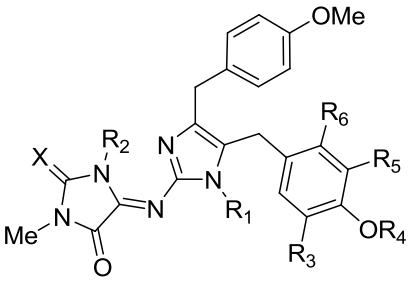
1a-h

Number	R^1	R^2	R^3	R^4	R^5	R^6	Name
1a	Me	H	H	H	H	H	Naamine A
1b	Me	Me	Me	OH	H	H	Naamine B
1c	Me	H	Me	OMe	OH	H	Naamine C
1d	H	H	Me	H	H	H	Naamine D
1e	Me	Me	Me	H	H	H	*N*,*N*-Dimethyl naamine D
1f	Me	H	Me	OH	H	OH	Naamine E
1g	Me	H	H	H	H	OMe	Naamine F
1h	Me	H	H	OMe	H	OMe	Naamine G

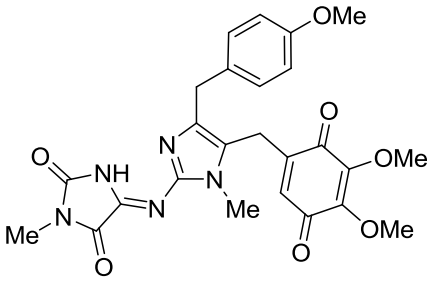
Figure 1.1: Naamines: 4,5-Dibenzylic alkaloids isolated from *Leucetta* and *Clathrina* sponges.

In 1987 Kashman reported the isolation of naamine A (**1a**), naamidine A (**2a**), isonaamine A (**5a**) and isonaamidine A (**6a)** from a bright yellow sponge of the class *Calcispongiae (calcarea) and Leucetta chagosensis dendy (Leucetta)* collected near Na'ama in the Gulf of Eilat (Aqaba) in the Red Sea.[2] The nomenclature for this whole series of compounds is based on the location of the initial source material. Although, at the time of the initial isolation, the biological activities were not reported for these novel alkaloids, in 1998 Ireland reported that naamine A (**1a**) acts as an antagonist of the epidermal growth factor (EGF) receptor, which plays an important role in the

development of several human tumors.[3] It was subsequently determined that this alkaloid exhibits selective antagonism of the EGF-mediated mitogenic response. *In vivo* evaluation of **1a** against EGF-dependent A431 tumors in athymic mice indicated that this alkaloid has modest antitumor activity, in which it intensifies the phosphotransferase activity of extracellular signal-regulated kinases causing A431 cells to arrest in the G_1-phase of the cell cycle.[4] Also, it was found to promote caspase-dependent apoptosis in tumor cells.[5]



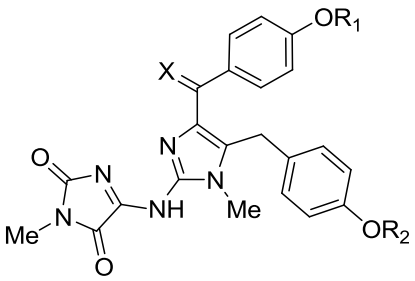
2a-i



3: Naamidine F

Number	R^1	R^2	R^3	R^4	R^5	R^6	X	Name
2a	Me	H	H	H	H	H	O	Naamidine A
2b	Me	H	H	Me	OH	H	O	Naamidine B
2c	Me	Me	H	H	H	H	O	Naamidine C
2d	H	H	H	Me	H	H	O	Naamidine D
2e	Me	H	OH	Me	OMe	OH	O	Naamidine E
2f	Me	H	H	Me	H	H	O	Naamidine G
2g	Me	H	OMe	H	OMe	H	O	Naamidine H
2h	Me	H	OMe	H	OMe	H	NMe	Naamidine I
2i	Me	H	H	Me	OMe	OH	O	Pyronaamidine

Figure 1.2: Naamidines: 4,5-Dibenzylic alkaloids isolated from *Leucetta* and *Clathrina* sponges.



4a-e

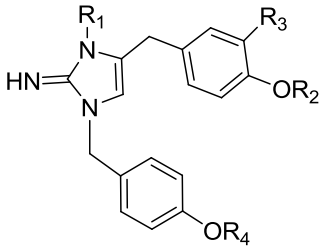
Number	R^1	R^2	X	Name
4a	Me	H	H, OH	14-Hydroxynaamidine A
4b	Me	Me	H, OH	14-Hydroxynaamidine G
4c	Me	H	H, OMe	14-Methoxynaamidine A
4d	Me	Me	H, OMe	14-Methoxynaamidine G
4e	Me	Me	O	14-Oxonaamidine G

Figure 1.3: Naamidines with mono benzylic oxidation isolated from *Leucetta* and *Clathrina* sponges.

Kashman and co-workers found a nudibranch, *Notodoris citrine*, feeding on the *Leucetta chagosensis* sponge, contains new imidazole alkaloids, naamine B (**1b**), naamidines B-D (**2b-d**) and isonaamidine B (**6b**) together with **1a**, **2a**, **5a** and **6a** in 1989.[6] In the same year, Fattorusso and coworkers reported the isolation of clathridine A (**8b**) from the sponge *Clathrina clathrus* collected the Procida Channel, near Napoli, Italy.[7] This mono substituted imidazole alkaloid was found to show *in vitro* antimycotic activity against *Candida albicans* and *Saccharonryces cerevisiae* (40 µg/disk). In addition to clathridine A (**8b**), a small quantity of its stable Zn-complex has been found in the same fraction.

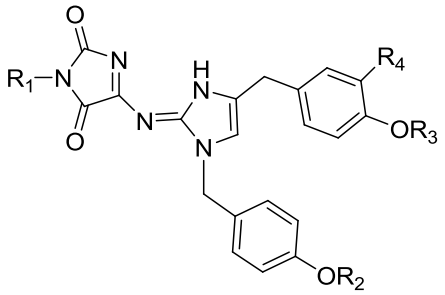
In 1990, Scheuer reported pyronaamidine (**2i**) and a new napthoquinone alkaloid kealiiquinone (**10b**), which biosynthetically is probably derived from the intramolecular oxidative coupling of pyronaamidine [or possibly naamidine E and F], from a Micronesian sponge, *Leucetta sp*.[8,9] Pyronaamidine (**2i**) was reported to be

mildly cytotoxic to KB cells with a minimum inhibitory concentration (MIC) value of 5 µg/mL.



5a-c

Number	R^1	R^2	R^3	R^4	Name
5a	H	H	H	H	Isonaamine A
5b	Me	Me	H	H	Isonaamine B
5c	H	Me	OMe	Me	Isonaamine C



6a-e

Number	R^1	R^2	R^3	R^4	Name
6a	Me	H	H	H	Isonaamidine A
6b	Me	H	Me	H	Isonaamidine B
6c	Me	Me	Me	H	Isonaamidine C
6d	H	H	Me	H	Isonaamidine D
6e	Me	Me	Me	OMe	Isonaamidine E

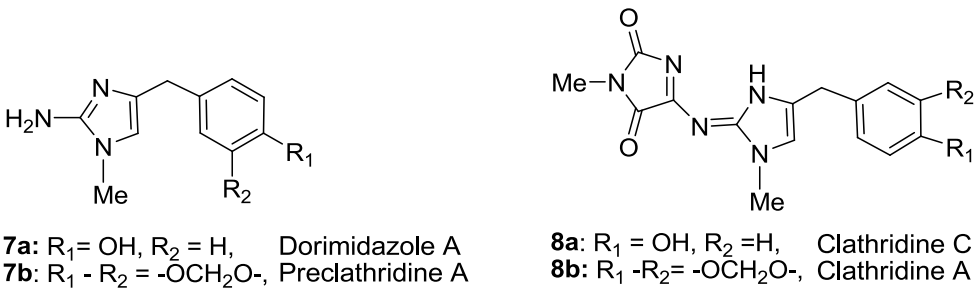
Figure 1.4: 1,4-Dibenzylic alkaloids isolated from *Leucetta* and *Clathrina* sponges.

In the following year, Crews and co-workers reported the isolation of dorimidazole A (**7a**), which was isolated from an Indo-Pacific sponge, *Notodoris gardineri.*[10] The crude extract of this sponge was found to exhibit *in vitro* activity

against the parasite, *Nippostrongylus brasilienris,* at 50 µg/mL. This group reported the first isolation of preclathridine A (**7b**), a Zn-complex of isonaamidine C and clathridine B (clathridine B is the analogous 4,5-dibenzylic derivative of clathridine A) in 1993 from a *Leucetta* sponge collected from Fiji, just offshore from Thang-galai Island.[11] Presumably, preclathridine A and dorimidazole A are the precursors for clathridine A and clathridine C respectively. However, at the moment there is no evidence which one forms first.

Just after reporting these new alkaloids, Call and co-workers reported the isolation of new imidazole alkaloids, naamidine E (**2e**), naamidine F (**3**) and clathridine C (**8a**) from the *Calcareous* sponge *Leucetta* sp. and a predatory nudibranch, *Notodoris gardineri* collected from caves on Flynn Reef in the Cairns' section of the Great Barrier Reef.[9] Isolation of naamidines E and F is suggestive of a biosynthetic relationship between them and kealiiquinone (**10b**).

In 1995 Pietra reported the existence of naamidine G (**2f**), 14-hydroxynaamidine A (**4a**), 14-hydroxynaamidine G (**4b**), 14-methoxynaamidine A (**4c**), 14-methoxynaamidine G (**4d**) and 14-oxanaamidine G (**4e**) in a *Leucetta* sponge species, collected from the Grand Passage Reef, north from the lagoon of New Caledonia.[12] These are the first alkaloids isolated from *Calcareous* sponges with mono benzylic oxidation and their individual biological activities have not been reported. Notably, however, the crude extract has been found to be mildly cytotoxic towards KB cells and anti-yeast *(Candida albicans*).[13]



7a: R_1= OH, R_2 = H, Dorimidazole A
7b: R_1 - R_2 = -OCH_2O-, Preclathridine A

8a: R_1 = OH, R_2 =H, Clathridine C
8b: R_1 -R_2= -OCH_2O-, Clathridine A

Figure 1.5: Mono benzylic alkaloids isolated from *Leucetta* and *Clathrina* sponges.

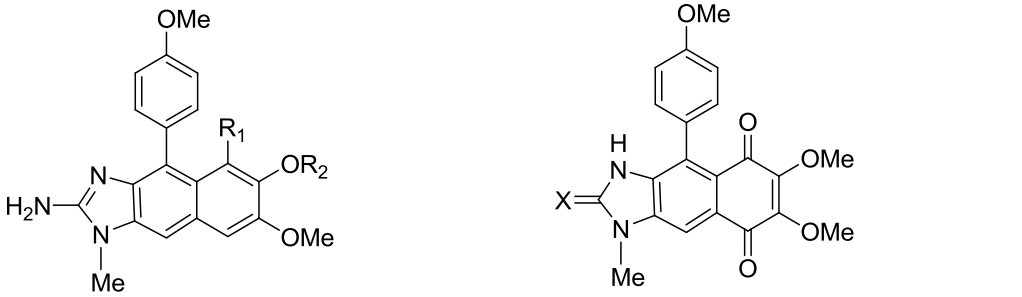
From the bright, lemon yellow sponge *Leucetta chagosensis*, this time collected from Chuuk State, Federated States of Micronesia, naamine C (**1c**) and 2-deoxy-2-aminokealiiquinone (**10a**) were isolated in 1997 by Schmitz and co-workers.[14] The isolation of these structurally interrelated alkaloids from the same sponge suggested that maybe **10a** can be formed from **1c** biosynthetically. In 1998, the same group reported the isolation of isonaamine B (**5b**), isonaamidine D (**6d**) with two Zn-complexes of isonaamidine B and isonaamidine D from the sponge *Leucetta cf. chagosensis* isolated from the same place.[15] These alkaloids have been tested against *P. aeruginosa*, *S. aureus*, *A. niger*, and *S. cerevisiae* at concentrations of 200, 100, 10, and 1 µg/mL to determine the MIC, however only isonaamidine D has been found to show inhibitory activity against *A. niger* at 100 µg/mL. The same sponge isolated from the Red Sea in 1995 was found to have new alkaloid, naamine D (**1d**), which is active against the AIDS OI pathogen *Cryptococcus neoformans* (6.25 µg/mL), and weak inhibitor of inducible nitric oxide synthase (iNOS) at 1.0 µg.mL.[16] This alkaloid was isolated along with four known alkaloids, naamidines A (12.5 µg/mL), B (6.25 µg/mL), D (not tested) and G (12.5 µg/mL), which are also active against the *C. neoformans.*

Two sponges, *Leucetta chagosensis* and *Leucetta* cf. *chagosensis*, collected from the Great Barrier Reef and the Fiji Islands respectively, have been reported to contain new alkaloids, naamine E (**1f**), isonaamine C (**5c**) and isonaamidine E (**6e**).[17] The cytotoxic effects of these compounds against HM02 (stomach carcinoma), HepG2 (liver carcinoma) and Huh7 (liver carcinoma with mutated p53) cell lines have been investigated and only compound **6e** [GI_{50} 7.0 µg/mL (HM02 and HepG2) and 1.3 µg/mL (Huh7)] and **5c** [GI_{50} 5.3 (HM02), 2.2 (HepG2), and 2.1 µg/mL (Huh7)] were found to be active.

In 2003, Crews reported the isolation of *N,N*-dimethyl naamidine D (**1e**) from the sponge *Leucetta avocado* and it was found to exhibit only mild activity in an antimicrobial panel consisting of *E. coli*, *S. aureus*, *B. subtilis*, and *C. albicans* (100 µg/disk).[18] The Crews group also reported the isolation of the first spiro-linked imidazole alkaloids, (-)-spirocalcaridine A (**12a**), (-)-spirocalcaridine B (**12b**) and the imidazolone, (+)-calcaridine A (**13**).[19] These compounds have unprecedented skeletons and functionality and are the first chiral non-organometalic 2-aminoimidazoles isolated from *Calcareous* sponges to be reported. During this isolation neither relative nor absolute stereochemistry was reported, also their biological activities have not been investigated.

Naamine F (**1g**), naamine G (**1h**) and kealiinine A-C (**9a-c**) have been isolated from the sponge, *Leucetta Chagosensis dendy* collected from Indonesia.[20] Among these, naamine G has been found to exhibit strong antifungal activity against the phytopathogenic fungus *Cladosporium herbarum* (20 µg/disk) and mild cytotoxicity against mouse lymphoma (L5178Y) and human cervix carcinoma (HeLa) cell lines at a concentration of 10 µg/mL. Kealiinine A has been found to more active than

naamine G against brine shrimp, *Artemia salina* (mortality rate of 50% vs. 10% at 20 µg/mL). Structural similarities of these kealiinines (**9a-c**) to kealiiquinone (**10**) suggest that they may be intermediates in the biosynthetic conversion of naamines or naamidines to kealiiquinones. However, it is not clear whether they form *via* the same or different routes from the naamines or naamidines.



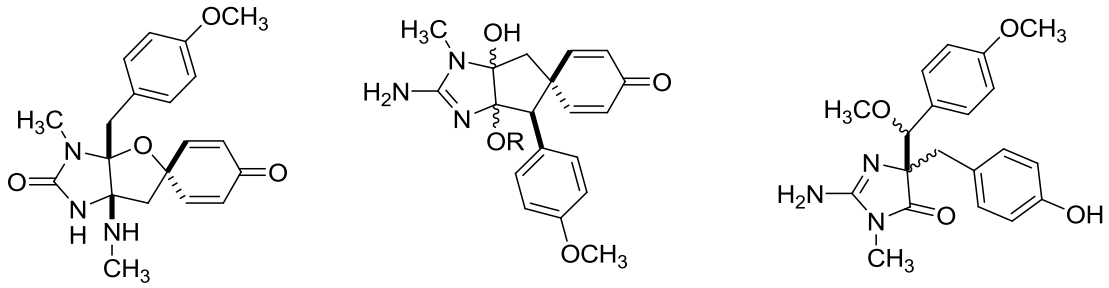
9a: R_1 = H, R_2 = H, Kealiinine A
9b: R_1 = H, R_2 = Me, Kealiinine B
9c: R_1 = OMe, R_2 = Me, Kealiinine C

10a: X = NH, 2-Deoxy-2-aminokealiiquinone
10b: X = O, Kealiiquinone

Figure 1.6: Naphthimidazole alkaloids isolated from *Leucetta* and *Clathrina* sponges.

Again, the Indonesian sponge *Leucetta chagosensis* has been reported to possess two new imidazoles, naamidine H (**2g**) and naamidine I (**2h**), which are cytotoxic against HeLa cells with IC_{50} values of 5.6 and 15 µg/mL respectively. Although, these two alkaloids show very similar structures, the latter has a guanidine instead of urea and this may influence the cytotoxicity of two alkaloids.[21] The first description of the isolation of (-)-spiroleucettadine (**11**) was reported by Crews in 2004 and it was found to be moderately active against *E. coli* and *Staphylococcus epidermitis* (200 µg/mL) and has good antibacterial activity against *Enterococcus durans* (6.25 µg/mL).[22] After reporting the isolation of this alkaloid, several research groups have directed their attention toward this molecule; however, none of these

efforts resulted in the synthesis of the reported sructure.[23, 24, 25] Subsequently, further analysis of the structure by the Crews lab, including re-isolation and obtaining a X-ray structure, led to the proposed structural revision depicted in Figure 1.7.[26]



11: (-)-Spiroleucettadine

12a: R = H, (-)-Spirocalcaridine A
12b: R = Me, (-)-Spirocalcaridine B

13: (+)-Calcaridine A

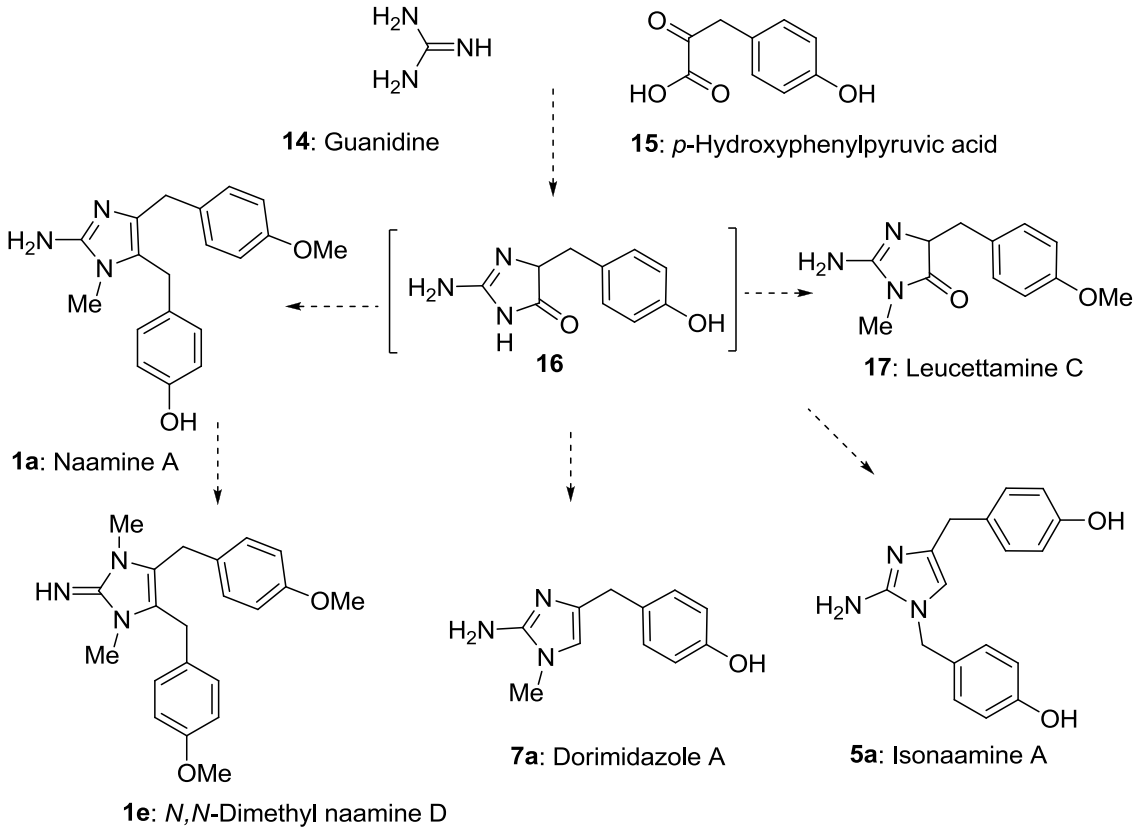
Figure 1.7: Oxygenated imidazole alkaloids isolated from *Leucetta* and *Clathrina* sponges.

Each of these molecules has characteristic structural and chemical properties, although they have similar substitution pattern. The position of the oxidation, the number of oxidized atoms and substitution at C2-amino group, bring variation to each of these molecules and these deviations also bring unique chemical properties to each of them. Due to these structural complexities, some of them have as yet relative and or absolute stereochemical relationship undefined (e.g. spirocalcaridines, calcaridines, 14-hydroxynaamidines and 14-methoxynaamidines), thus synthetic programs may be able to elucidate some of these issues. Despite the structural differences among these natural products, an approach to one of them may provide intermediate that can be used en route to other members of the family. Furthermore, since these molecules have been isolated in low percentages of the sponges, and unreported biological

activitie, total synthesi of these provides material for both biological investigation and structural elucidations.

1.2 Biosynthesis of 2-aminoimidazoles

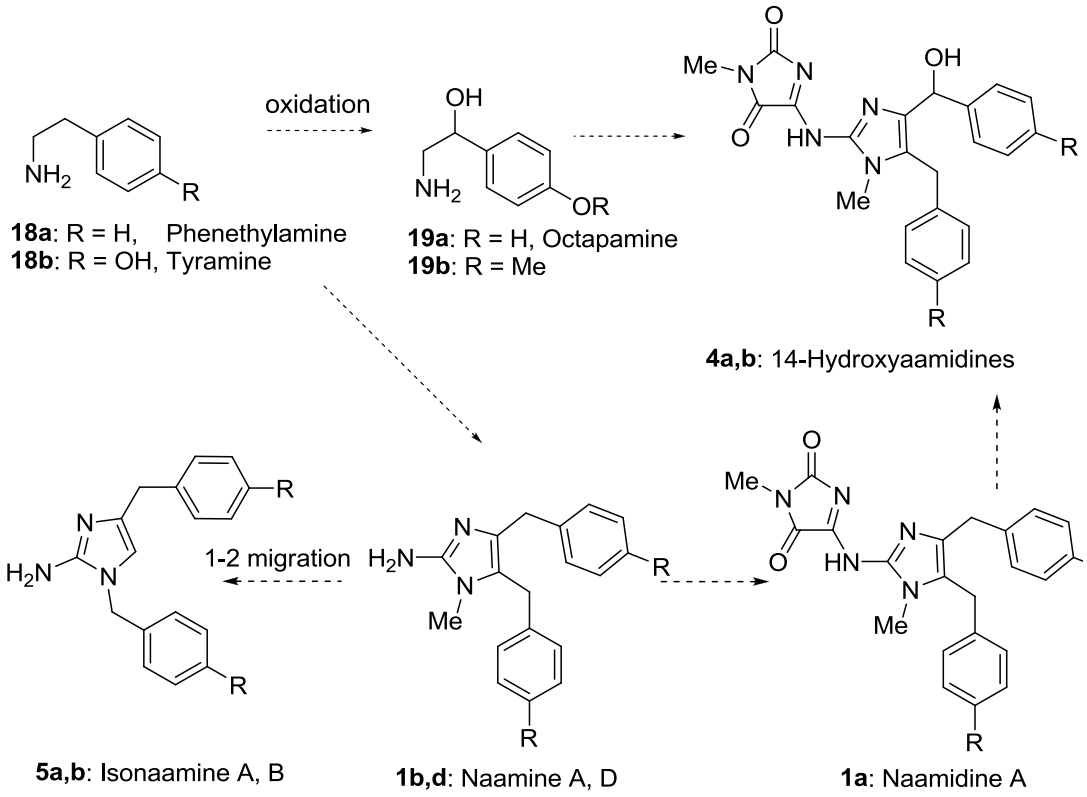
A clear definition of biosynthetic origin of 2-amino alkaloids has not been established at the present time. Although a number of hypothetical possibilities have been described, there is still no experimental verification has been found.[6,12, 15] Crews has proposed the biogenetic origin of intermediate **16** from guanidine (**14)** and *p*-hydroxyphenylpyruvic acid (**15**) (Scheme 1.1), which then serves as the precursor for dorimidazole A (**7a**), isonaamine A (**5a**) and other related compounds.[10,18]



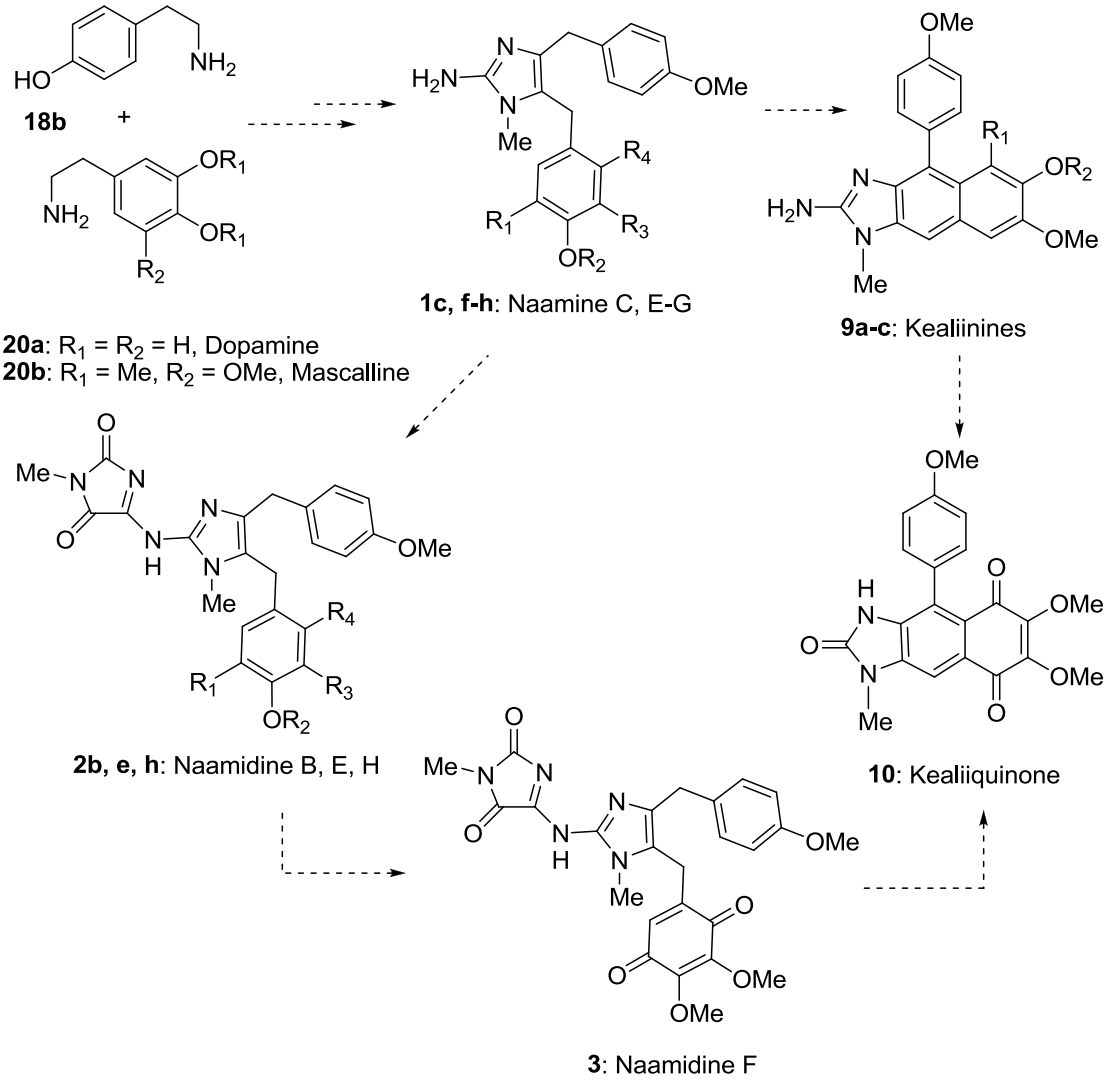
Scheme 1.1

Another suggestion provided by Kashman and coworkers is that 4,5-disubstituted imidazoles of naamines may originate from two phenethylamines (**18a**)

and the isonaamines are obtained from either 1,2-migration of a benzyl moiety from carbon to the neighboring nitrogen (Scheme 1.2).[6] Alternatively, two tyramine units **18b** may serve as the initial building blocks since the 4-hydroxy group is already present in it. However, it is not clear, from these pathways, whether the mono oxidation at benzylic position present in 14-hydroxynaamidines takes place through an enzymatic route *via* naamines[12] (i.e. **1a**→**4a**,**c**) or whether they are directly formed from 2-amino-1-arylethanol derivatives (i.e. **19a**,**b** →**4a**,**c**).



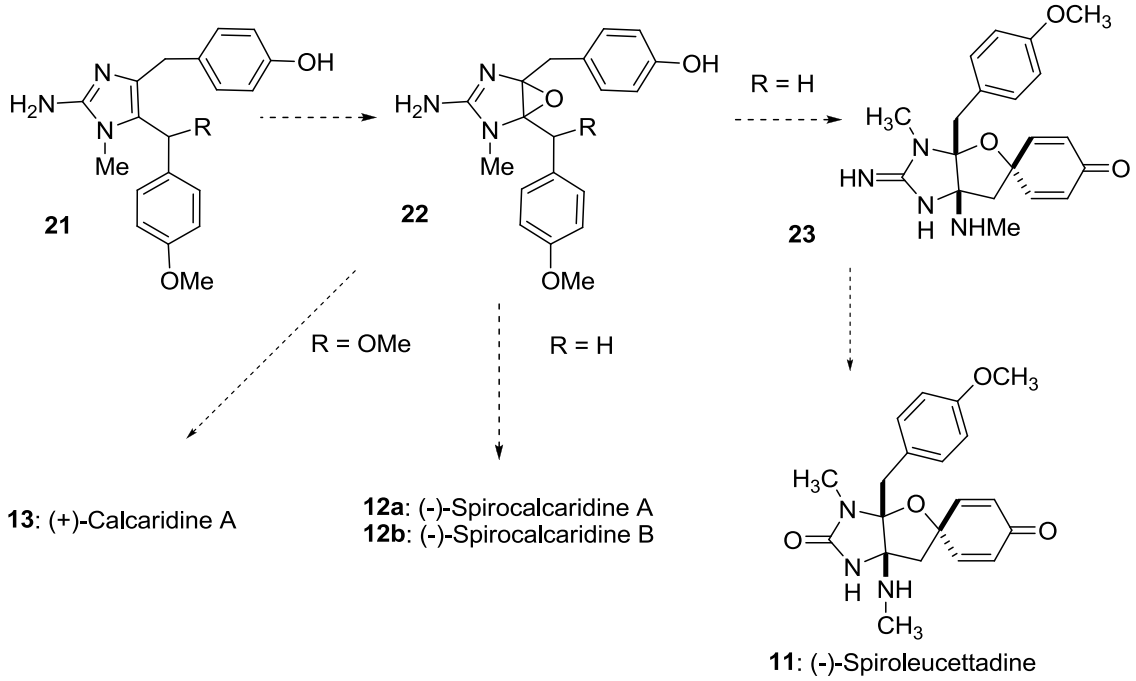
Scheme 1.2



Scheme 1.3

Similarly, other alkaloids in this family may be derived from suitably substituted amines such as mascalline[27] and dopamine (Scheme 1.3). For example, naamines E and G may be derived from combination of mascalline and tyramine derivatives. Although there is no clear evidence, formation of naamidines might occur *via* derivatization of these naamines. It can be postulated that the formation of kealiiquinone (**10**) might take place *via* naamidine F (**3**), which is, probably, an intermediate in the biosynthetic transformation of naamidine E (**2e**) to kealiiquinone

(**10**).[9] It has also been suggested that the formation of kealiiquinone (**10**) might take place *via* kealiinines (**9**), which in turn are formed from naamidine derivatives by ring closure and aromatization. Subsequent oxidation of the pyrogallol dimethyl ether ring to the quinone, plus hydrolytic loss of the aminoimidazoledione part of the molecule would lead support to this suggestion.[8] This idea is supported by the isolation of naamine C and 2-deoxy-2-aminokealiiquinone (**10a**) from the same sponge.[14]



Scheme 1.4

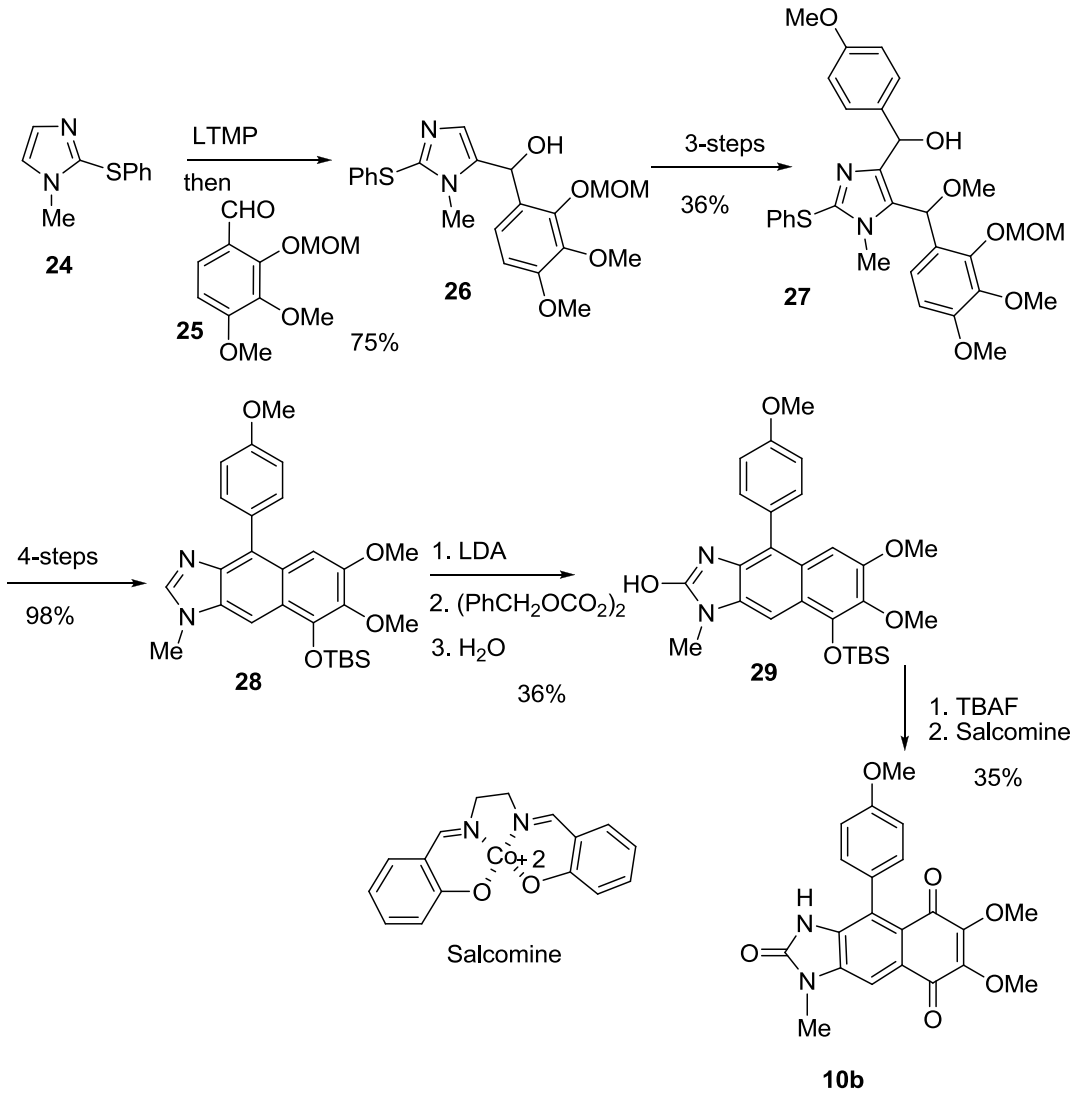
Highly oxygenated alkaloids such as spirocalcaridine A and B, calcaridine A and spiroleucettidine might also be derived from either some of above naamines (e.g. **4c**, **1a** etc.) after enzymatic oxidative addition, oxidative rearrangement and *ipso*-cyclization *via* a common intermediate **22**[28, 29] or they may originate from completely different route starting from tyrosine derivatives (Scheme 1.4).[24, 25, 30]

Since presently there have been no experimental studies performed to elucidate the biosynthetic origin of these 2-aminoimidazole alkaloids, it is difficult to predict their exact origin. However, it is very clear that they are formed from a similar sequence of reactions from derivatives of simple building block. The structural relationships among these alkaloids suggest that they are probably the various intermediates of multi-step biosynthesis processes. It is this principal that our lab has utilized as paradigm in the design of synthetic approaches towards this family of natural products. As will be described later in this dissertation, the rearrangement of **21** to calcaridine A is feasible, and this leads some support to the hypothesis that these molecules result from biosynthetic interconversion.

1.3 Synthetic approaches to *Leucetta* and *Clathrina* alkaloids

The structural complexities and biological activities of *Calcarea* alkaloids have captured the attention of synthetic chemists over past two decades. In principal, there are two possible general approaches for the total synthesis of these imidazole alkaloids; one in which the imidazole ring is constructed in a *de novo* fashion, or a second in which a pre-existing imidazole is elaborated; both methods have been used for the synthesis of these alkaloids.

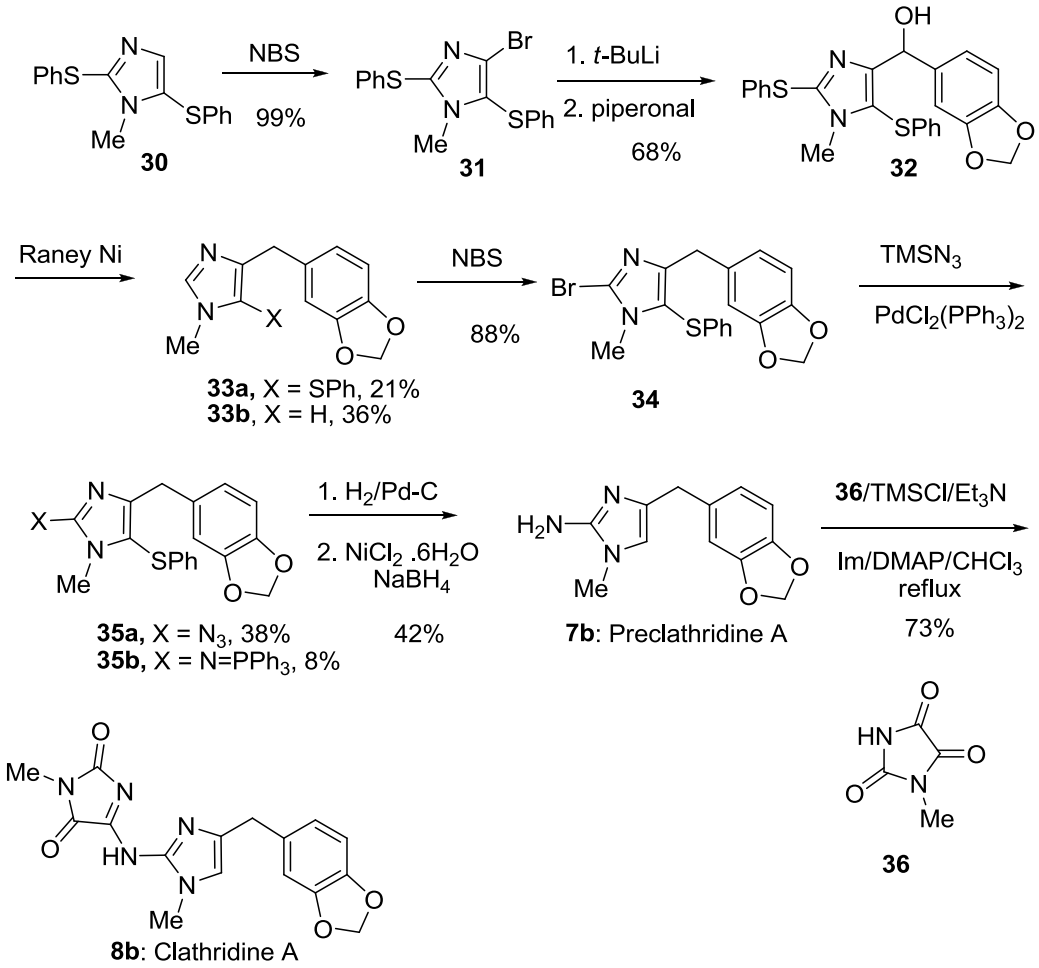
One of the earliest examples of activity in this area is the total synthesis of kealiiqinone (**10b**), starting with 2-thiophenylimidazole **24** (Scheme 1.5).[31] In this method, the imidazole C2-position has been protected and the functionalization is carried out C5→C4 sequence followed by removal of C2-protection. Our approach, to be described in this dissertation, avoids the need to protect the C2-position. The imidazole derivative is treated with LTMP followed by aldehyde **25** to afford the alcohol **26**. Then the methylation of hydroxyl group followed by the bromination of C4-position is carried out with NBS to facilitate the introduction of the C4-substituent. The resulting bromide is treated with *tert*-butyl lithium (*t*-BuLi) and then *p*-anisaldehyde to provide **27** as a mixture of diastereomers. Addition of TFA initiates intramolecular Friedel-Crafts of this alcohol to provide the naphthimidazole framework. A sequence of deprotection and re-protection reactions provides naphthimidazole **28.** Finally, the introduction of C2-oxygenation is accomplished by metalation and treatment with dibenzyl peroxydicarbonate and the TBS removal provided the dihydroxy derivative **29**. Subjection of **29** to autooxidation in the presence of salcomine is carried out to complete the total synthesis of kealiiqinone (**10b**) in 11 steps from **24** and in 2.6% overall yield.



Scheme 1.5

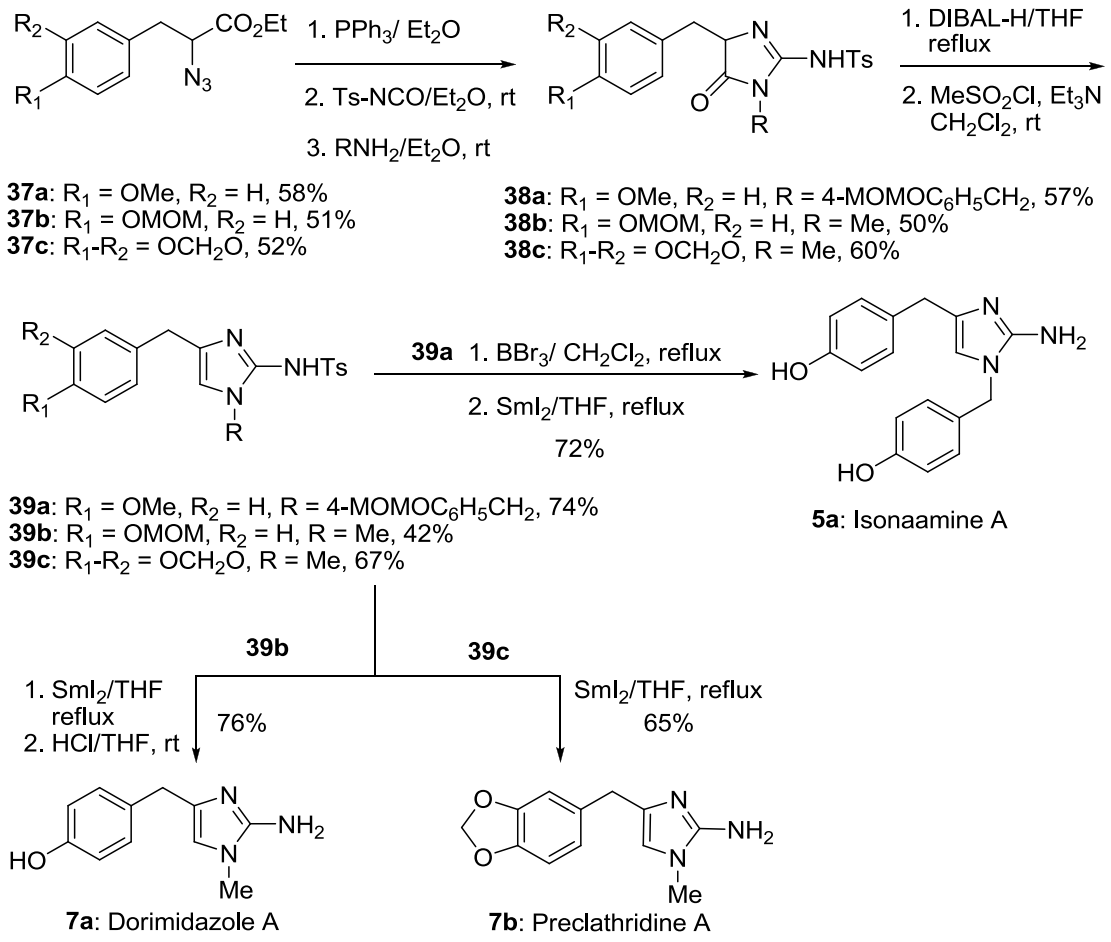
Total synthesis of preclathridine A (**7b**) and clathridine A (**8b**) was carried out by Ohta's group starting from the 1-methyl-2,5-diphenylthio-1*H*-imidazole **30** as shown in Scheme 1.6.[32, 33] Imidazole **30** was prepared by 1-methyl-1*H*-imidazole and phenyldisulfide in two steps, and bromination of **30** at the C4-position with NBS followed by treatment with *t*-BuLi and piperonal provided alcohol **32**. Desulfuration of this alcohol with Raney nickel provided a mixture of sulfide **33a (**21%) and imidazole **33b (**36%). Treatment of the former with NBS provided the bromide **34**,

which was treated with trimethylsilyl azide to form the azide **35a** (38%) as a mixture with iminophosporane **35b** (8%). Then, azide **35a** was reduced with H_2/Pd-C followed by the reductive removal of thiophenyl substituent at the C5-position to synthesize preclathridine A (**7b**) in 3% overall yield. In order to synthesize clathridine A (**8b**), preclathridine A was treated with 1-methylparabanic acid (**36**), which was activated with trimethylsilyl chloride and triethylamine in chloroform at reflux prior to the addition of **7b**.[34]

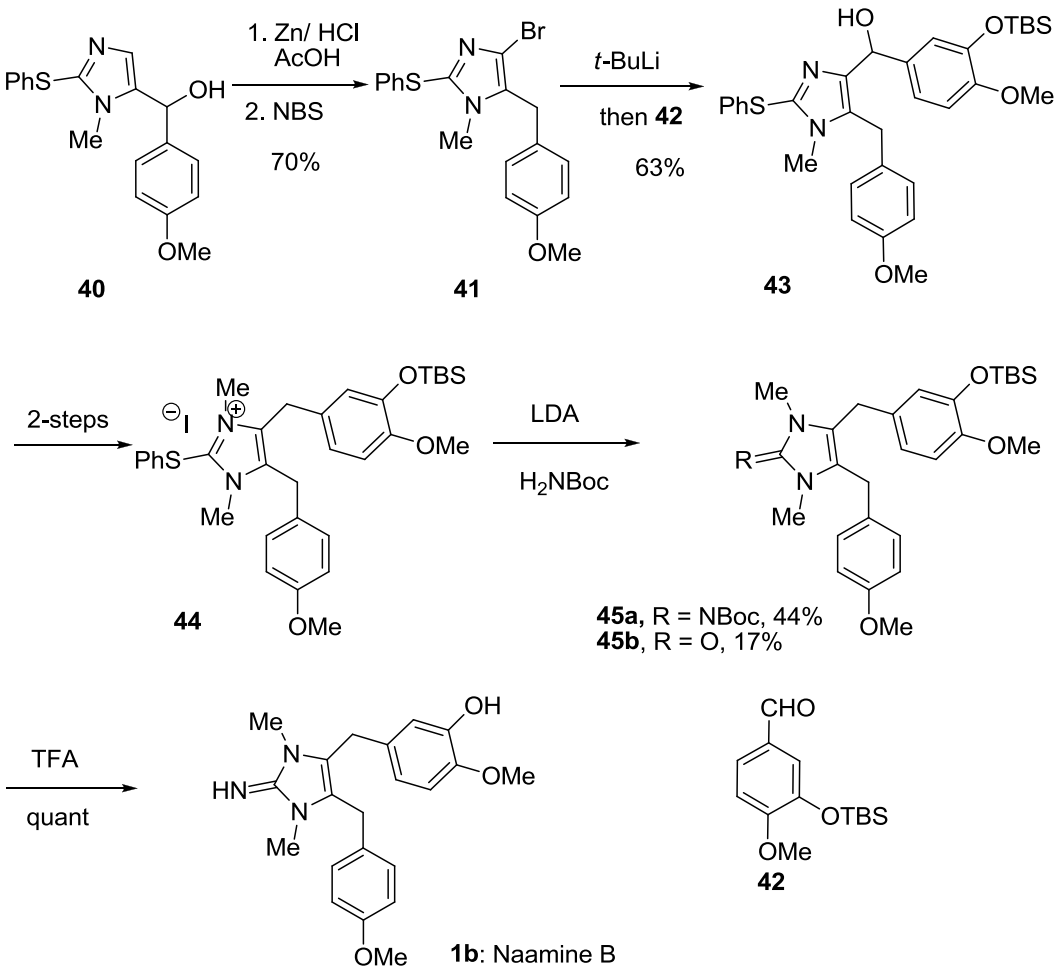


Scheme 1.6

In 1999 Molina and Fresneda reported the first total syntheses of isonaamine A (**5a**), dorimidazole A (**7a**), and preclathridine A (**7b**) (Scheme 1.7).[34] Here, they used α-azido esters **37a-c**, which were synthesized from appropriate ethyl 3-arylpropionates, to prepare 2-amino-1,4-disubstituted imidazoles through application of the aza-Wittig reaction of iminophosphorane derivatives with tosyl isocyanate followed by treatment with primary amines providing **38a-c**. Semi-reduction with DIBAL, dehydration with methanesulfonyl chloride and deprotection of *N*-tosyl group in these substrates provided the target molecules, **5a**, **7a, b** in reasonable yields.



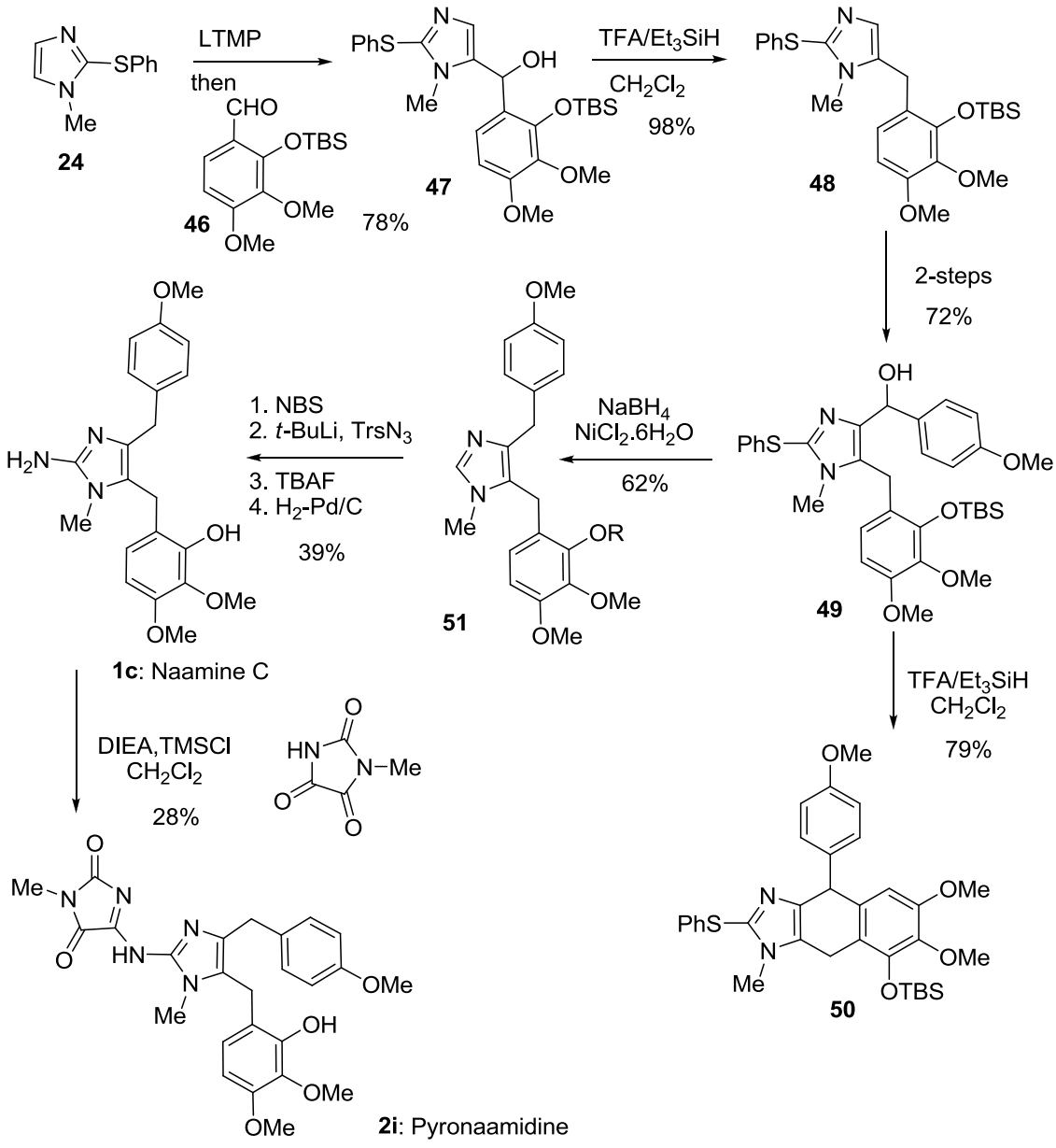
Scheme 1.7



Scheme 1.8

The first total synthesis of naamine B (**1b**) was carried out by the Ohta's lab with imidazole **40** (Scheme 1.8), which was prepared in a similar fashion to **26** as described in Scheme 1.5.[35] The benzylic hydroxy group of **40** was removed by reduction with zinc powder–*conc.* HCl and the C4-position of the product was treated with NBS to provide bromide **41**. Conversion of this bromide to alcohol **43** was accomplished on treatment of *t*-BuLi and aldehyde **42**. Reduction of this alcohol with Et_3SiH and TFA followed by refluxing in ethyl acetate in the presence of MeI provided imidazolium salt **44**. This imidazolium salt was treated with the anion

derived from *tert*-butyloxycarbamate and LDA to afford the desired *N*-Boc imino compound **45a** accompanied by the 2-oxoimidazoline **45b**. Treatment of **45a** with TFA led to removal of the Boc protecting group and thus completing the first total synthesis of naamine B in 20% overall yield from **40**.

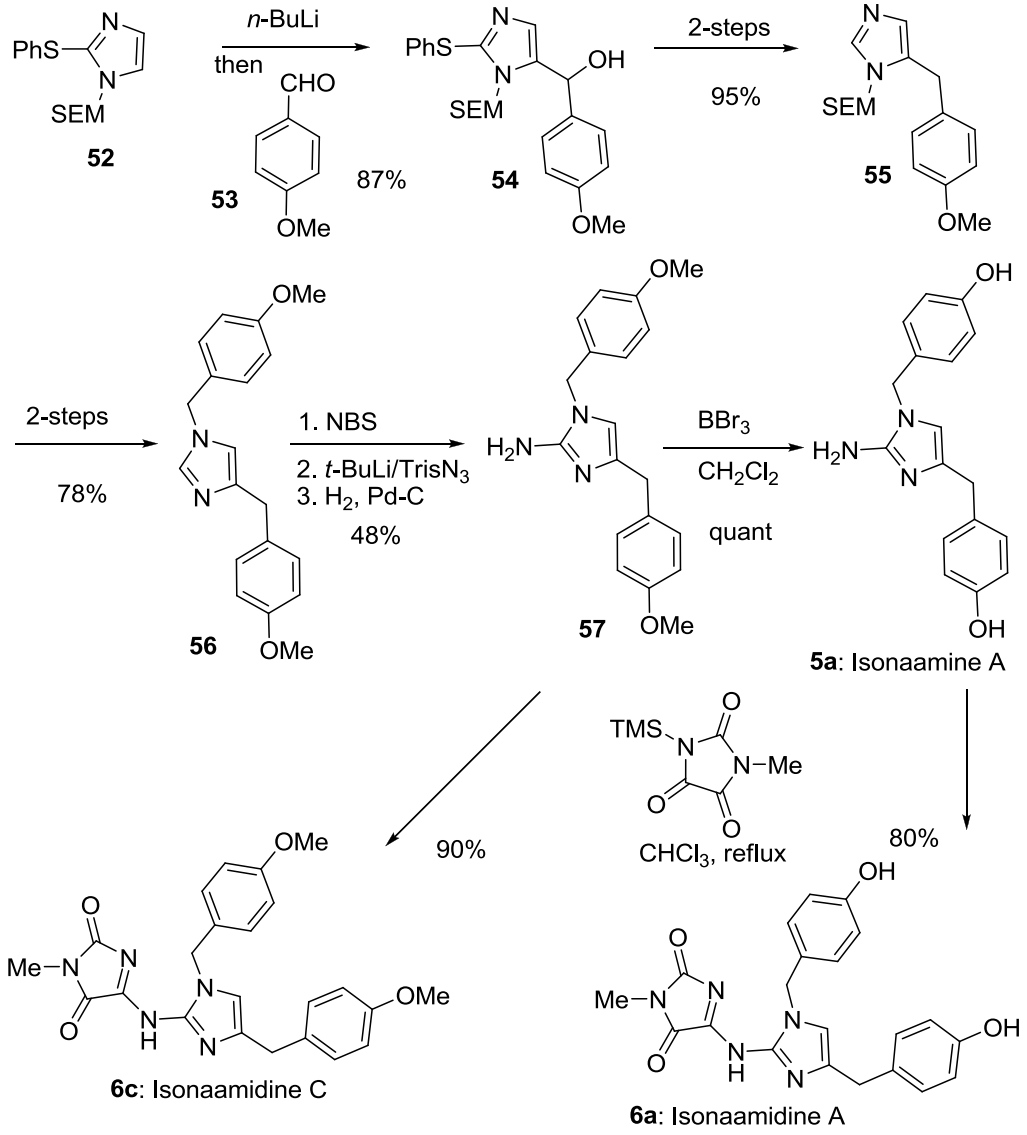


Scheme 1.9

Naamine C (**1c**) and pyronaamidine (**2i**) have been synthesized, from 7 and 8 steps respectively, starting from **24** (Scheme 1.9)**.**[36] In this approach, a similar sequence of reactions described for total synthesis of kealiiqinone (**10b**), in Scheme 1.5, was used. Imidazole **24** was treated with LTMP followed by aldehyde **46** to obtain alcohol **47** in 78% yield and the removal of the benzylic hydroxyl group was achieved with TFA and Et_3SiH to provide **48** in 98% yield. This disubstituted imidazole was treated with NBS to afford the 4-bromoimidazole, which was subjected to lithiation with *t*-BuLi at -78 ºC followed by quenching with *p*-anisaldehyde to produce alcohol **49** in 72% overall yield.

Removal of the hydroxyl group from **49** using an ionic reduction protocol was problematic (TFA and Et_3SiH); since it formed tricyclic imidazole **50** through an intramolecular Friedel-Craft like reaction. This has been overcome by the addition of a combination of $NaBH_4$ and $NiCl_2.6H_2O$ resulting in the formation of the 4,5-dibenzylimidazole **51** in 62% yield. Conversion of **51** to naamine C (**1c**) was accomplished with C2-bromination (NBS), azidation (*t*-BuLi, $TrsN_3$), removal of the TBS group (TBAF) followed by reduction of azide, in 35% overall yield. Pyronaamidine (**2i**) has been synthesized by treating naamine C with 1-methylparabanic acid in the presence of TMSCl and DIEA.

Ohta and co-workers have used a relatively short and efficient method to synthesize isonaamidines A (**6a**) and C (**6c**) starting from disubstituted imidazole **52** (Scheme 1.10).[37]

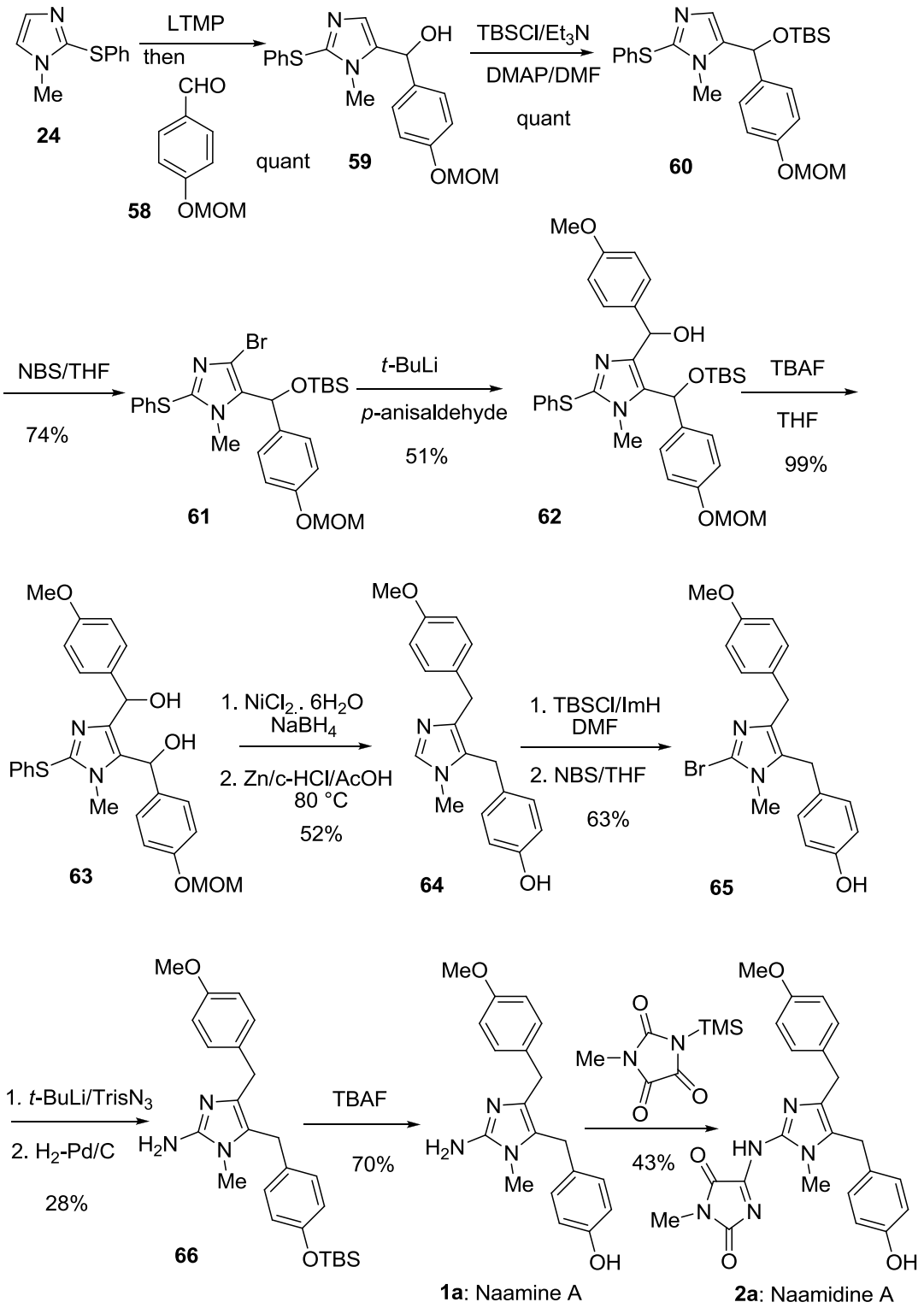


Scheme 1.10

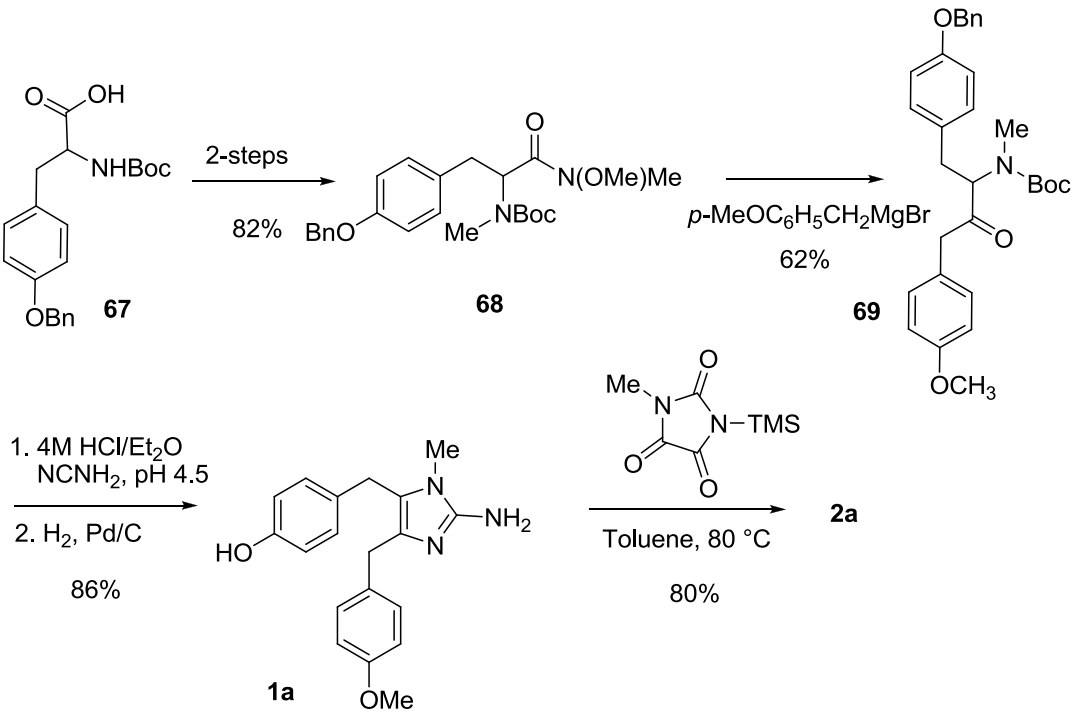
Lithiation of the sulfide **52** with *n*-BuLi followed by treatment with *p*-anisaldehyde **53** provided alcohol **54** in 87% yield. The removal of the hydroxyl group with TFA and Et_3SiH, followed by desulfuration with $NaBH_4$ provided imidazole **55** in 95% overall yield. Treatment of this imidazole with 4-methoxybenzyl bromide in refluxing EtOAc provided an imidazolium salt, which was treated with 10% HCl to provided 1,4-disubstituted imidazole **56** in 78% overall yield.

Bromination of **56** at the C2-position followed by azidation and then reduction of resulting azide were carried out to produce amine **57** in 48% overall yield. Subsequent demethylation of **57** with BBr_3 provided isonaamine A (**5a**), which provided isonaamidine A (**6a**) upon treatment with 1-methylparabanic acid in the presence of TMSCl and Et_3N. Repetition of the above step with amine **57** provided isonaamidine C (**6c**).

The first total synthesis of naamine A (**1a**) and naamidine A (**2a**) was reported by Ohta and co-workers in 2000 (Scheme 1.11).[38] Imidazole **24** was treated with LTMP and aldehyde **58** to afford alcohol **59**, which was protected with TBSCl followed by bromination at the C4-position providing **61** in 74% overall yield. Lithiation at C4 position followed by quenching with *p*-anisaldehyde (**53**) provided diol **62.** A series of protecting group maneuvers and functional group manipulations including, removal of the TBS, the thiophenyl, and the MOM groups, deoxygenation of the diols provided **64** in 26% starting from **61.** Attempts to introduce C2 azide after protecting the phenolic hydroxylic group with TBS failed due to the lithiation at benzylic proton of the C5 position. Therefore, subsequent bromination at the C2-position permitted the bromide **65**, which was converted to the amine **66** in usual manner. Removal of TBS group with TBAF completed the synthesis of naamine A (**1a**) in 2.6% overall yield. Introduction of the hydantoin unit furnished the total synthesis of naamidine A.

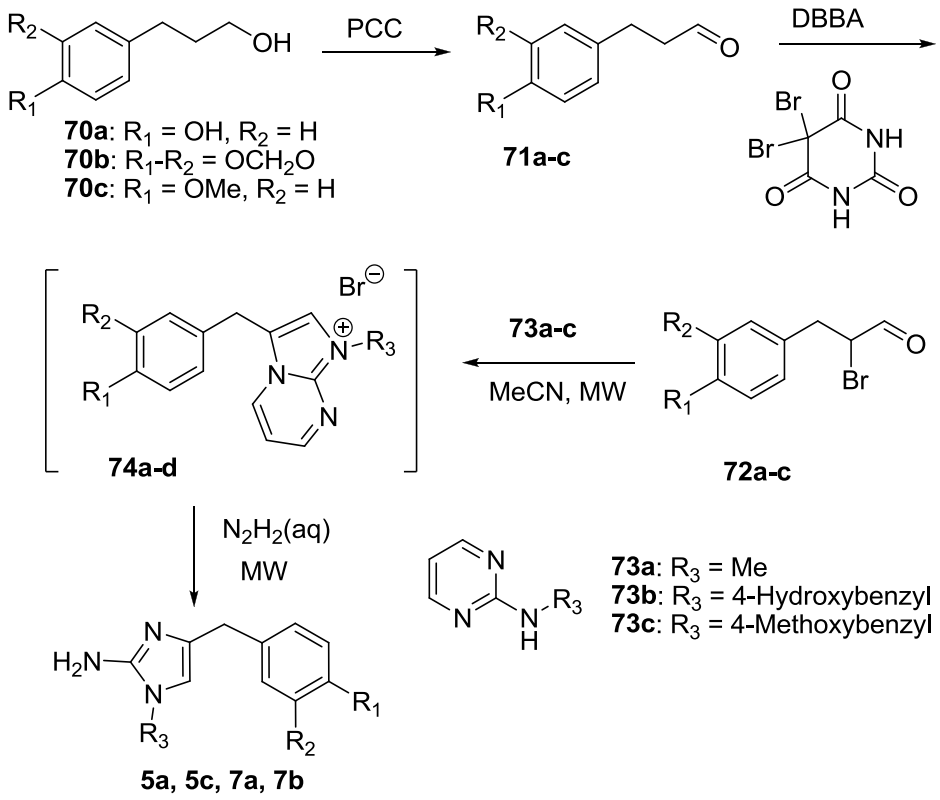

LTMP
then
24
58
quant
59
TBSCl/Et3N
DMAP/DMF
quant
60
NBS/THF
74%
61
t-BuLi
p-anisaldehyde
51%
62
TBAF
THF
99%
63
1. NiCl2. 6H2O
NaBH4
2. Zn/c-HCl/AcOH
80 °C
52%
64
1. TBSCl/ImH
DMF
2. NBS/THF
63%
65
1. t-BuLi/TrisN3
2. H2-Pd/C
28%
66
TBAF
70%
1a: Naamine A
43%
2a: Naamidine A

Scheme 1.11



Scheme 1.12

In contrast, Watson and co-workers have developed a short and efficient synthesis of naamine A and naamidine A starting with commercially available tyrosine derivative **67** (Scheme 1.12).[30] Selective *N*-methylation of the Boc-protected amino acid **67** using MeI and NaH followed by *in situ* formation of the acid fluoride and reaction of this fluoride with *N,O*-dimethylhydroxylamine provided the Weinreb amide **68**. This was then treated with *p*-$MeOC_6H_5CH_2MgBr$ to provide the α-amino ketone **69**. After removing the Boc-protecting group with 4 M HCl, the resulting salt was subjected to condensation with cyanamide, affording the 2-aminoimidazole, which provided naamine A (**1a)** after removing benzylic protection under catalytic hydrogenation. Introduction of the hydantoin moiety onto **1a** completed the second total synthesis of naamidine A (**2a**), in 6 steps with 35% overall yield.



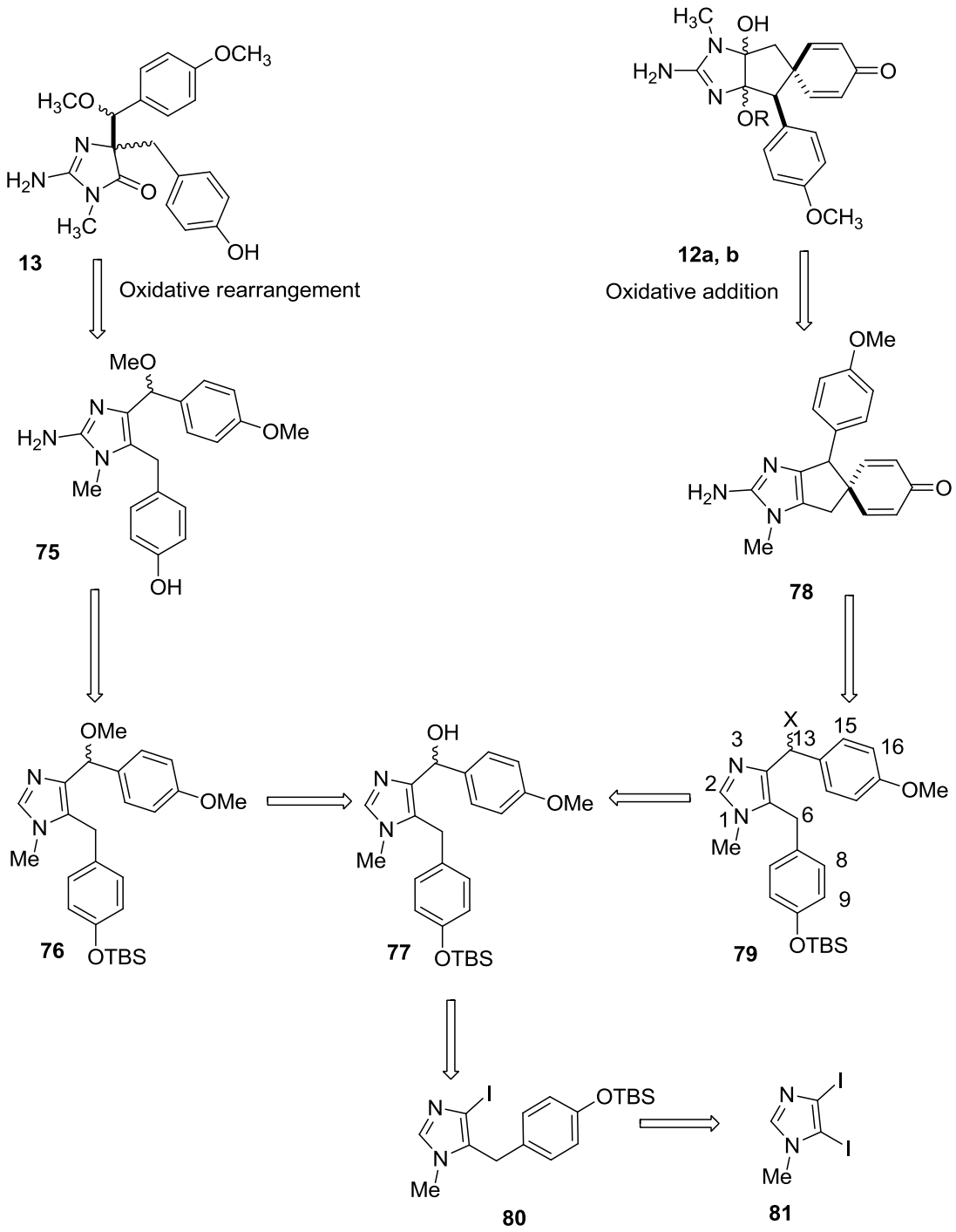
Scheme 1.13

In 2008, Eycken reported the total synthesis of preclathridine A (**7b**), dorimidazole A (**7a**), isonaamine A (**5a**) and isonaamine C (**5c**) using 2-alkylaminopyrimidines (**73**) and 2-bromoaldehydes (**72**) as shown in Scheme 1.13.[39] This syntheses was commenced by oxidizing 3-phenylpropanols **70a-c**, which were obtained from corresponding cinnamic acid derivatives, to the aldehyde derivatives **71a-c** using PCC. The 2-bromination of these aldehydes were accomplished using a mild brominating reagent, 5,5-dibromobarbituric acid (DBBA) at room temperature to isolate **72**, which were irradiated together with 2-aminopyrimidine derivatives **73a-c** in acetonitrile at 80 ^{o}C to provide the intermediate **74**. These intermediates were irradiated with hydrazine hydrates to form corresponding natural products **5a, 5c, 7a** and **7b.**

1.4 Our approach to *Leucetta* and *Clathrina* alkaloids

Although more than sixty 2-aminoimidazole alkaloids have been isolated from *Calcarea* sponges, there have been no experimental investigations of the biosynthesis or biosynthetic relationships between these molecules. The forgoing, notwithstanding some hypothetical relationship can be investigated, and thus an approach to one of these natural products may provide intermediates that can be used en route to other family members. Due to the structural (relative and absolute stereochemistry) and biological ambiguities of calcaridine A (**13**), spirocalcaridine A (**12a**) and spirocalcaridine B (**12b**), our initial attempts were directed towards the total syntheses of these alkaloids. Ideas evolved out of these investigations for developing approaches to other members of this family by suitable modifications of the developed synthetic methods.

We have for some time had an interest in the development of new methods and strategies for the construction of complex imidazole-containing natural products from simple imidazole derivatives, rather than *de novo* synthesis of the imidazole ring.[40] Towards this end, we and others have demonstrated that 4,5-dihaloimidazoles can be functionalized in a sequential and controlled manner at C5- and then C4-positions by treatment with Grignard reagents, and then electrophilic trapping.[41-45] Subsequent C2-functionalization can then be accomplished by lithiation, and electrophile quench, for example, with an "N_3^+" equivelent,[46] thus providing a flexible and expedient approach to a large number of these 2-aminoimidazole alkaloids.



Scheme 1.14

Consequently, the retrosynthetic analysis of these molecules started with 4,5-diiodoimidazole **81** (Scheme 1.14), which can be converted in to monoiodoimidazole

80 with TBS-protected benzyl bromide using transmetalation.[41] A second Grignard reaction of **80** could be used to synthesize alcohol **77** using *p*-anisaldehyde (**53**) as the electrophile. This alcohol can be converted to **76** or **79** (X is a suitable leaving group) to synthesize precursors for calcaridine A (**13**) and spirocalcaridines (**12a**, **b**) respectively. The critical reaction en route to the spirocalcaridines is an intramolecular dearomatizing alkylation, and although this reaction has not been reported with imidazole derivatives, similar *ipso*-cyclization (C7→C13) of silyl protected phenols are known with alkyl bromides to provide spiro[4.5]decadienone derivaties.[47-49] Application of these conditions might provide the precursor for **78**, which will provide **78** after C2-amination. Similarly, C2-amination of **76** can be used to synthesize 14-methoxynaamine A, **75**. After this, **75** and **78** will be subjected to oxidative chemistry rearrangement and addition respectively to provide calcaridine A and spirocalcaridines A and B. Toward this end, our group has found that 4,5-disubstituted imidazoles on reaction with DMDO undergo a rapid rearrangement to provide 5-imidazolones or, in some cases, oxidative additions.[50, 51] We anticipated that these reactions might prove useful en route to *Leucetta* alkaloids. Our efforts towards these ends are detailed in the remainder of the dissertation.

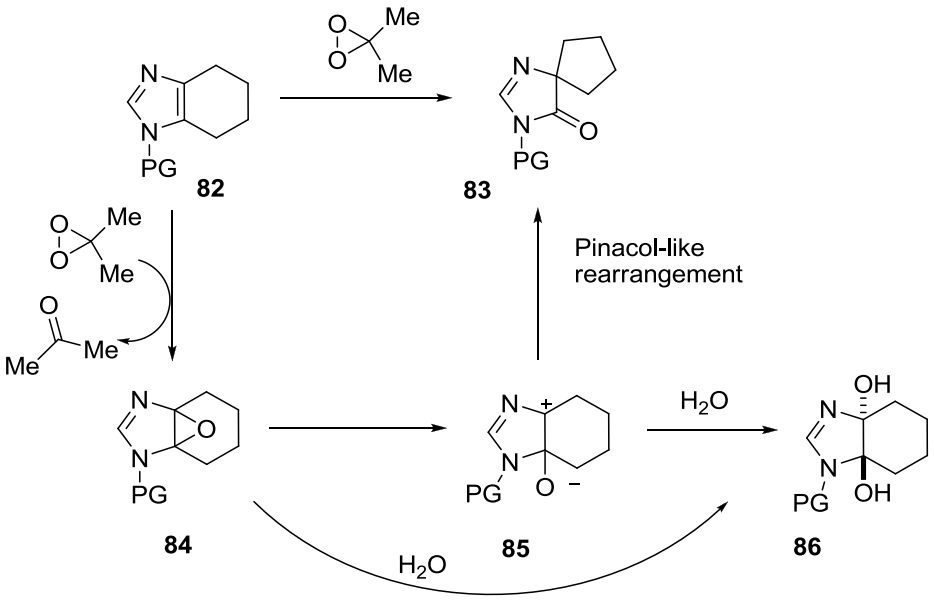
CHAPTER 2

OXIDATIVE REACTION OF IMIDAZOLE DERIVATIVES WITH N-SULFONYL OXAZIRIDINES

The oxidation chemistry of tetrahydrobenzimidazole has been studied earlier by our group, and it has been found, that the DMDO oxidation of *N*1-substituted tetrahydrobenzimidazole derivatives (THB's, **82**) lead to oxidative rearrangement to the corresponding spiro-imidazolones **83** (Scheme 2.1). In one case there is some evidence that dihydroxylation occurs.[52] While no detailed mechanistic studies were conducted, the reaction was formulated as proceeding *via* epoxide **84** in analogy with the corresponding rearrangement of *N*-acyl indoles with DMDO.[53-57] Subsequent ring opening of intermediate **84** leads to the formation of the more stable zwitterion **85**, which rearranges presumably *via* a pinacol-type process to provide **83**. Dihydroxyl derivative **86** may be formed upon addition of water to either **84** or **85**. However, no experimental evidence has been collected at the moment to support this hypothesis.

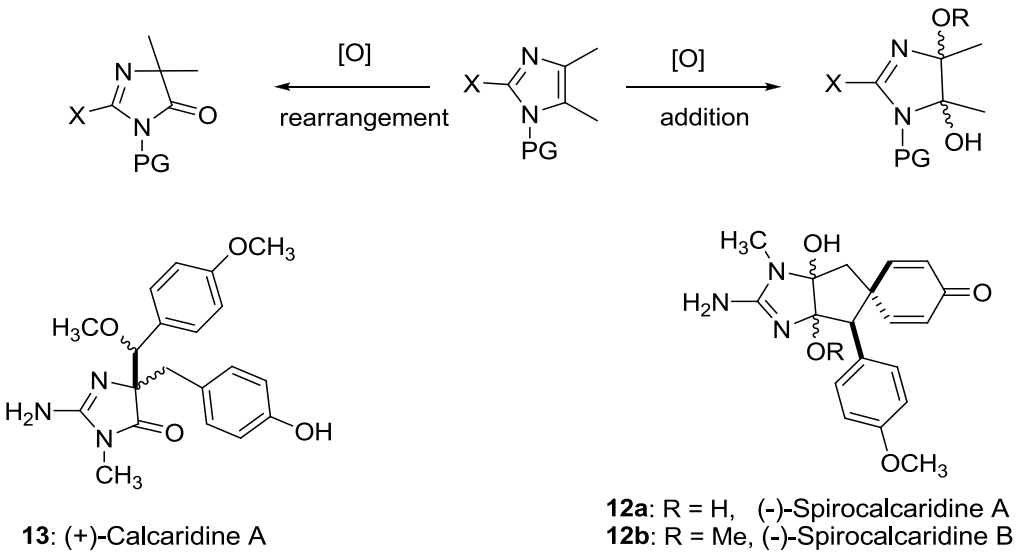
While this chemistry was quite satisfactory in most respects, there were practical aspects of using DMDO that rendered it somewhat inconvenient, particularly for small scale scouting experiments. Chief among the deficiencies was the need to prepare isolated DMDO solutions, which often were of variable concentration and would contain trace (and variable) amounts of water. Therefore, we sought to identify alternative oxidants that would effect this rearrangement. Among several possibilities, Davis' reagents, *N*-sulfonyloxaziridines,[58-61] attracted our attention as they share many common characteristics with dioxiranes, therefore it occurred to us that this

class of reagent may offer a shelf-stable alternative to DMDO. These reagents are readily accessible by oxidation of the *N*-sulfonylimine, which in turn can be obtained from condensation of the corresponding benzaldehyde derivative and a sulfonamide.[62-65] An additional attraction of these reagents is that chiral, non-racemic variants are known and thus the possibility of asymmetric versions of this chemistry is feacible.[66]



Scheme 2.1

There are two general types of oxidative transformations required in order to develop synthetic approaches to these alkaloids. An oxidative rearrangement that can be used to construct the imidazolone moiety found in **13** while, oxidative addition that can be used to synthesize dihydroxy moiety found in **12a** and **12b** (Scheme 2.2), by changing the conditions used in the oxidative transformation.



Scheme 2.2

Thus, the initial goals of this study were to establish the utility of *N*-sulfonyloxaziridines in the oxidative chemistry of imidazoles. In order to explore these two transformations, 3-Phenyl-2-phenylsulfonyloxaziridine (**87**) and the more electrophilic 3-(4-Nitrophenyl)-2-phenylsulfonyloxaziridine (**88**) (Figure 2.1) were used.[64]

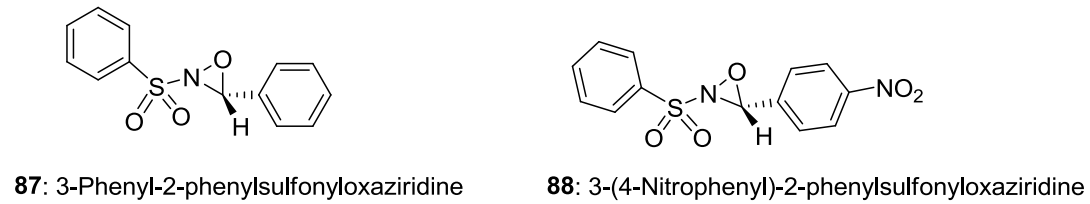
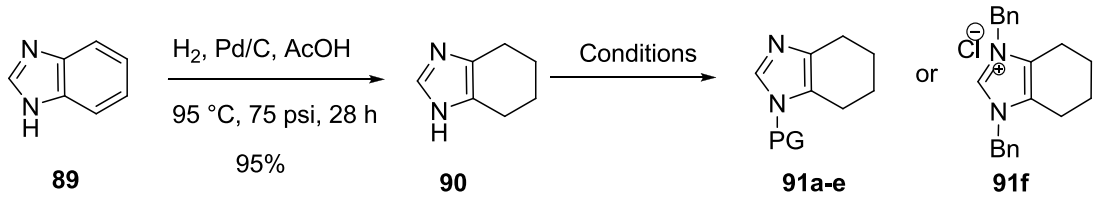


Figure 2.1: Davis' Oxaziridines

2.1 Oxidative transformations with oxaziridines

When we began this investigation there were no examples of oxidative reactions of imidazole derivatives with oxaziridines, therefore a series of model studies have been performed using substrates based on their potential to mimic precursors for natural products shown in Scheme 1.14. First of all, 4,5,6,7-tetrahydro-1*H*-benzimidazole **90** (THB) was converted in to a series of *N*1-protected THB derivatives as shown in Scheme 2.3 and Table 1, Where the nature of the protecting group would influence the electronic character of the imidazole.[52]



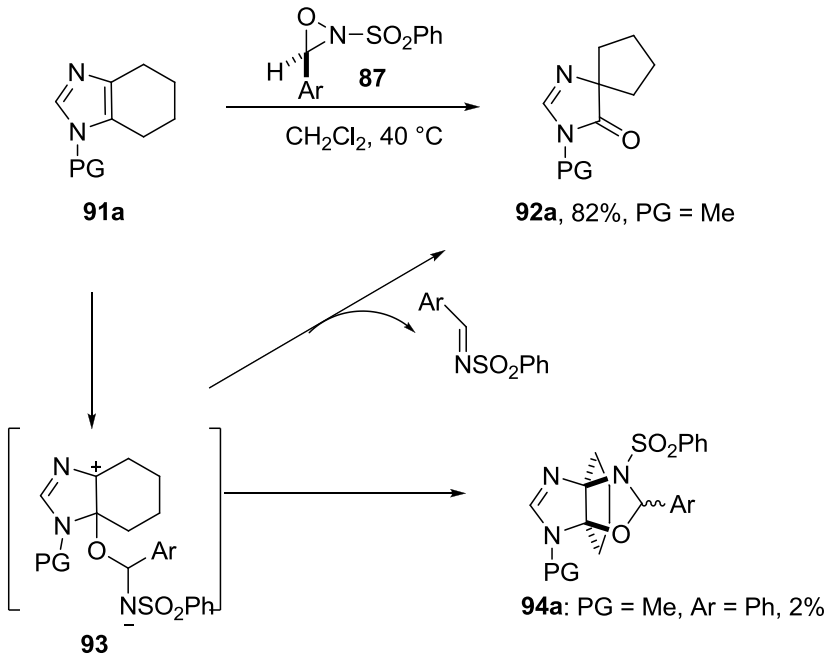
Scheme 2.3

Table 2.1: Conditions and yields for the preparation of THB derivatives, **91a-f**

Entry	PG	Substrate	Conditions	Yield/%
1	Me	**91a**	NaH/THF, MeI, rt.	85
2	Bn	**91b**	NaH/THF, BnCl, rt.	95
3	MOM	**91c**	NaH/THF, MOMCl, rt.	30
4	SEM	**91d**	NaH/THF, SEMCl, rt.	71
5	DMAS	**91e**	Et_3N/CH_2Cl_2, DMASCl, rt.	91
6	Bn_2	**91f**	NaH/DMF, BnCl, reflux	85

In our previously reported studies,[52] it was found that the THB (**91**) needed to be reasonably electron rich for this reaction to occur, and so in preliminary experiments, the Me-substituted THB **91a** was employed. After a few scouting reactions, we were delighted to find that exposure of **91a** to 2.0 equiv of phenyl

sulfonyloxaziridine **87** in CH_2Cl_2 led to a smooth rearrangement reaction, providing the spiro fused 5-imidazolone **92a** (Scheme 2.4).



Scheme 2.4

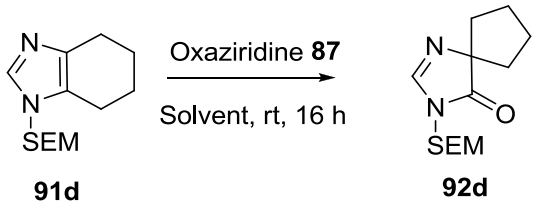
Interestingly, in addition to the major rearrangement product **92a**, a small amount of second product **94a** was isolated in ~2% yield. It was clear from the NMR data and HRMS analysis that this byproduct contained fragments derived from the oxaziridine. A clue to the identity of this product was found in a study by Dmitrienko of the utility of *N*-sulfonyloxaziridines as oxygen transfer agents with indoles.[67] Instead of the desired oxindoles, formal [3+2] oxaziridine–alkene cyclo-adducts (across the indole 2,3-bond) were isolated as the major products (diastereomeric mixture) when simple benzaldehyde-derived oxaziridines were used.[67, 68] This type of reaction has been generalized with a variety of olefins leading to the formation of isoxazoles. A similar adduct, **94a** is formed in this case through net addition of the

oxaziridine across the imidazole 4,5-bond. Although NOE and NOESY experiments were conducted, the relative stereochemistry of the benzylic center awaits assignment and the regiochemistry of this adduct has been proposed based on the putative mechanism of formation (Scheme 2.4), and in analogy with Dmitrienko's study. The isolation of **94a** has implications for the mechanism of this rearrangement reaction. As described above in Scheme 2.1, the DMDO-mediated rearrangement was proposed to proceed *via* 3a,7a-epoxide **84**, which opens to form **85**, and then rearranges.[52] The isolation of **94a** when an *N*-sulfonyloxaziridine is employed suggests an alternative mechanistic pathway involving the direct formation of zwitterion **93**, and then either elimination of imine and rearrangement to spiro imidazolone (**93→92**, Scheme 2.4), or intramolecular trapping to generate **94** (**93→94**, Scheme 2.4). While extrapolation of this mechanistic proposal to the DMDO-mediated process should be made with caution, the direct formation of the zwitterionic intermediate **85** (or the corresponding DMDO adduct related to **93**) cannot be ruled out on the basis of the experimental data available.[53, 54]

2.1.1 Selection of the best solvent

Given the success of this initial reaction we began to investigate the scope and limitations of this reaction, first by selecting a suitable solvent using **91d** as the substrate (Scheme 2.5). The reason for selecting SEM derivative is that it contains a moderately electron withdrawing group and therefore, it gives a relatively low-yield in the oxidative reaction.[52]

As shown in Table 2 (identical conditions other than the solvent), it appears that this rearrangement works well in moderately polar solvents such as chloroform and dichloromethane, but not in the non-polar or highly polar solvents. However, rather than being an intrinsic polarity issue we suspect that this may be due to the poor solubility of the oxaziridines in these solvents. As a result, in subsequent experiments dichloromethane and chloroform were used as the solvents.



91d **92d**

Scheme 2.5

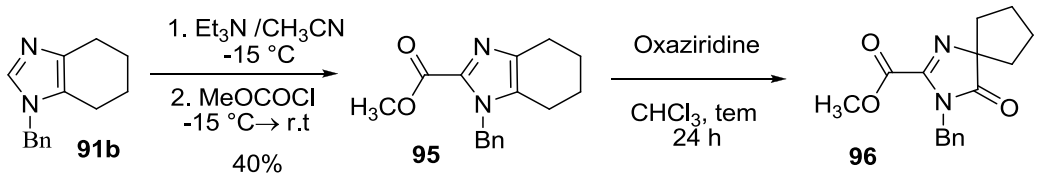
Table 2.2: Yield of the oxidation reaction of **91d** in different solvents

Entry	**Solvent**	**Yield/%**
1	Benzene	24
2	Acetonitrile	41
3	Chloroform	60
4	Dichloromethane	64

2.1.2 The best conditions for oxidative reactions

After selecting a suitable solvent, further optimization was performed to identify the best conditions for the oxidative transformations. Therefore, methyl 2-(1-benzyltetrahydrobenzimidazole) carboxylate **95**[69] was prepared as shown in Scheme 2.6 and it was used to optimize the reaction under the conditions shown Table 3, and the yield shown is an average of duplicates. It was observed that oxaziridine **87** did

not provide an improved yield by increasing the reaction temperature. However, oxaziridine **88** provided a better yield, while providing the best yield at 40 ºC.



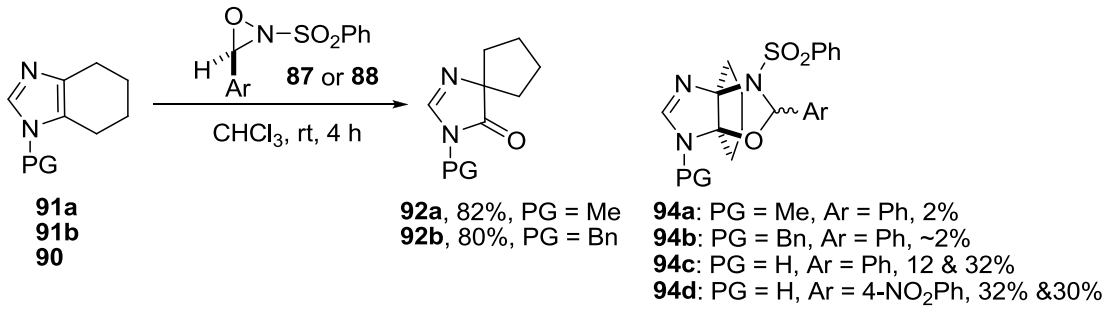
Scheme 2.6

Table 2.3: Yield of the rearrangement of **95** to **96** under different conditions

Entry	Oxaziridine	At rt/%	At 40 ºC/%
1	**87**	12	13
2	**88**	21	55

After optimizing the oxidation reaction, a collection of THB derivatives that differed in the nature of the *N*-substituent **91a–f** were investigated in the rearrangement chemistry with Davis' reagents (**87** or **88**) as discussed in following sections.

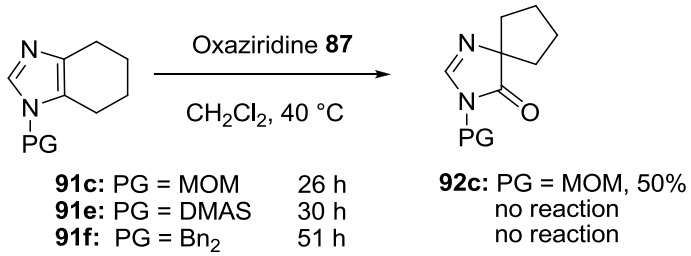
2.1.3 Oxidation of electron rich imidazole derivatives



Scheme 2.7

The rearrangement proceeds smoothly with the same types of substrates that undergo reaction with DMDO.[52] With an electron donating group on the substrate, the reaction provides the corresponding rearranged product as the major product, with a small amount of oxaziridine adduct as described in section 2.1. However, we were unable to fully purify **94b** to determine an accurate yield, but it was the same order as **94a**. We also investigated the reaction of unsubstituted tetrahydrobenzimidazole, that is, PG = H and found that it reacts with both Davis' reagents (**87** or **88**), but provides the addition product **94c** or **94d** as a separable mixture of as yet unassigned diastereomers, rather than rearrangement products.

2.1.4 Oxidation of electron deficient imidazole derivatives

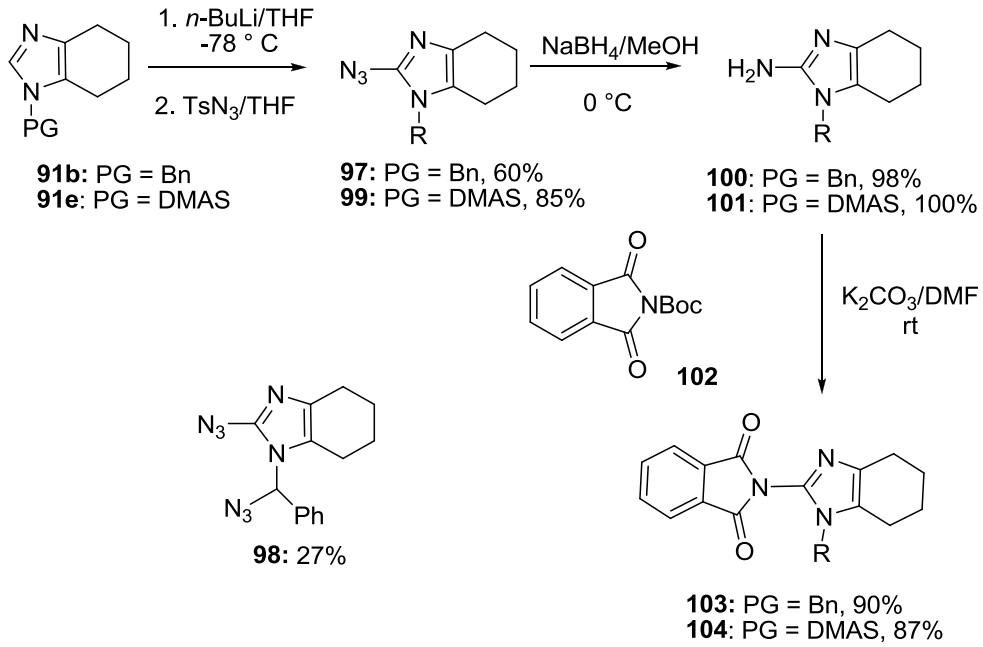


Scheme 2.8

With a moderately electron withdrawing group (**91c**), the rearrangement occurs but the yield was decreased and it required a higher temperature and a longer reaction time. Furthermore, the same set of unreactive substrates with DMDO fails to rearrange, including the sulfonyl urea **91e** and imidazolium salt **91f**. These results are consistent with the outcome of analogous reaction with DMDO; the electrophilic addition of oxaziridine across the C=C of imidazole is facilitated by electron donating group on *N*1-position, which is consistent with mechanism proposed in Scheme 2.12.

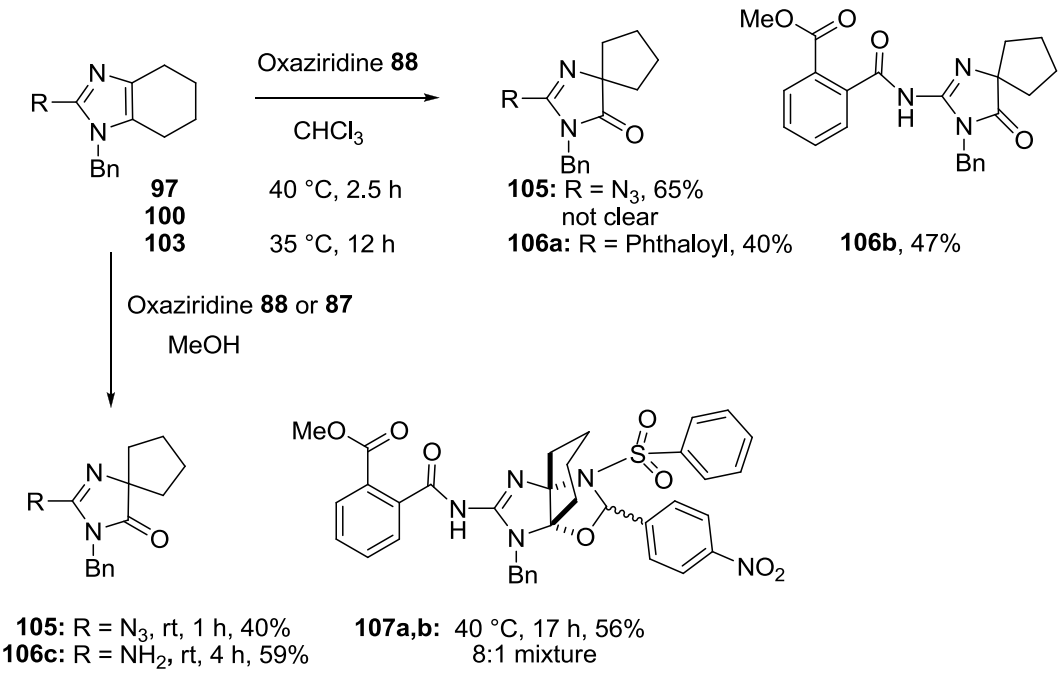
2.2 Evaluation of 2-amino-containing substrates

One major deficiency of the DMDO-mediated rearrangements was application of this reaction to 2-amino substituted derivatives.[70] It was our intention to use this transformation in approaches to a variety of marine natural products in the oroidin and related families.[70-73] Therefore, we decided to investigate the use of Davis' reagents with substrates of this type. Since use of the Davis' reagent led to rearrangement of the benzyl-protected THB, and as some of our on-going total synthesis efforts utilized benzyl-protected derivatives, we prepared both the 2-azido, and the *N*-phthalimidoyl derivatives **97** and **103**, respectively, for investigation (Scheme 2.9). The 2-azido derivative **97** was obtained by lithiation of **91b** at C2, followed by treatment with TsN_3, which provided **98** also as a byproduct. $NaBH_4$ reduction of **97** and treatment of **100** with the modified Nefkens' reagent **102** provided phthalimide derivative **103**.[74] We had found in our initial studies with Davis' reagent that the DMAS-protected THB **91e** did not undergo rearrangement, but we speculated that if the electron density of the imidazole moiety could be increased, a rearrangement might ensue. Accordingly, the 2-amino derivative **104** was prepared as above *via* metalation and trapping with TsN_3 providing the 2-azido congener **99** (Scheme 2.9), which was easily reduced to amine **101**, which in turn was protected with a phthaloyl moiety on treatment with Nefkens' reagent, providing **104**.



Scheme 2.9

2.2.1 Oxidation of imidazole with benzyl group at *N*1-position



Scheme 2.10

Gratifyingly, the benzyl-protected derivatives **97** underwent smooth rearrangement to the corresponding 5-imidazolones **105** on treatment with the nitrophenyl substituted oxaziridine **88** in chloroform (Scheme 2.10). Treatment of **103** under the same conditions provided two imidazolones **106a** (minor) and **106b** (major); however, it was surprising to obtain methanol adduct **106b** from this reaction since there was no obvious methanol source in the reaction media. Repetition of this reaction, under the same conditions, result the same outcome although the yields were little different. Stirring of a solution of **106a** in methanol, at room temperature did not result in the opening of the phthalimide ring. Then, we stirred a solution of **106a** in the same source of chloroform in the presence of silica gel, which was used for the column purification of above reaction; from this test we found the conversion of **106a** to **106b** after few minutes. After changing the chloroform source, it was observed a smooth conversion of **103** to **106a** without any detectable methanol derivative **106b**. Therefore, we think the formation of **106b** may be a result of a combination of contaminated solvents and silica gel, which facilitates the ring opening of the phthalimide.

We suspected from our experience with DMDO mediated reactions of a 2-amino substituted derivative,[70] that dimerization might be occurring in CH_2Cl_2 and $CHCl_3$. In fact, we did not observe the imidazolone **106c** when amine **100** was treated with oxaziridine in chloroform as this reaction was not clean enough to isolate a pure product. Therefore, the reaction of **100** was performed in a more polar solvent, methanol as shown in Scheme 2.10. Interestingly, in this case amine **100** underwent the rearrangement as expected providing **106c** in 60%, meanwhile azide **97** provided relatively low amount of spiro-imidazolone **105**.[75] Interestingly, treatment of **103** with

oxaziridine **88** in methanol resulted in cleavage of phthaloyl moiety while providing [3+2] adducts **107a,b**, of oxaziridine and imidazole.

2.2.2 Oxidation of imidazole with DMAS group at *N*1-position

108a: R' = Me, 68%
108b: R' = H, 65%

Scheme 2.11

Table 2.4: Result of the oxidation reactions of imidazole with DMAS group at *N*1-position

Entry	R	Substrate	Conditions	Product
1	N_3	**98**	MeOH, 40 ºC, 12 h	No reaction
2	NH_2	**101**	MeOH, rt, 4 h	**108a**(X-ray)
3	Phthaloyl	**104**	MeOH, rt, 60 h	No reaction
4	NH_2	**101**	Acetone-H_2O, rt, 4 h	**108b**

In DMAS derivatives, neither the azide **99** nor the protected substrate **104** rearranged (Table 4, entries 1 and 3), however, we discovered that when the 2-amino derivative **101** was reacted with oxaziridine **88** in methanol a rapid reaction occurred, but not the anticipated rearrangement to a 5-imidazolone. On isolation of the product it was clear from the NMR data that it contained a methoxy group, and the typical signal due to the imidazolone carbonyl in the ^{13}C NMR spectrum at around δ_C = 180 ppm was absent.[52] The NMR and MS data pointed to the formation of the hydroxy methyl ether **109** (Scheme 2.12), in which the stereochemistry was assigned on the basis of methanol trapping the (incipient) carbocation on the opposite face from the oxaziridine approach.[76] Subsequently, an X-ray crystal structure determination

(Figure 2.2) proved that both our stereochemical proposal and our constitutional assignment were incorrect. The methoxy and hydroxy groups were in fact *cis* to one another and there was a net migration of the DMAS moiety to the exocyclic nitrogen.

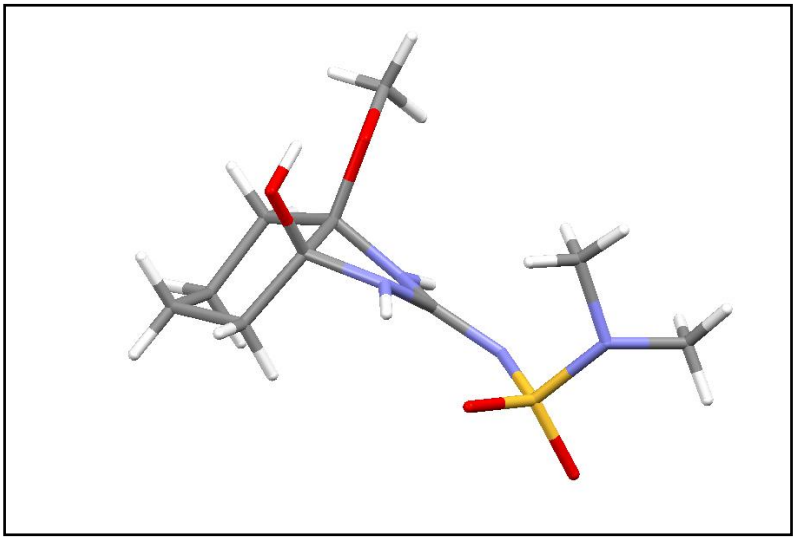
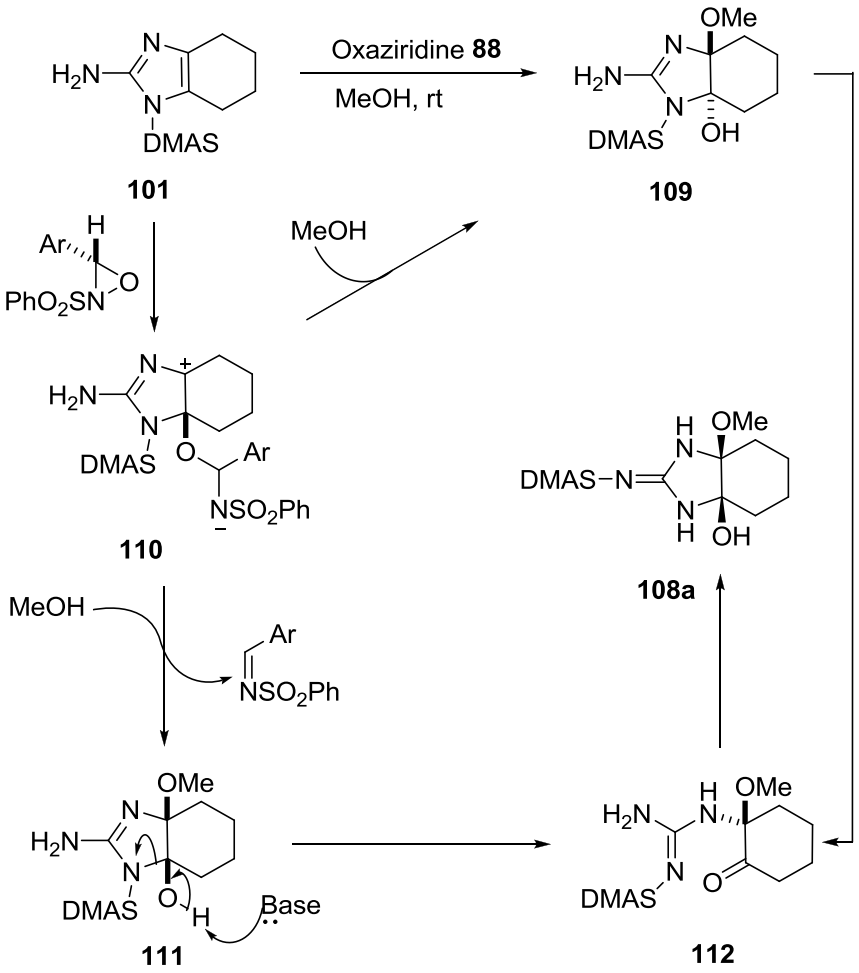


Figure 2.2: X-ray crystal structure of **108a.**

Presumably, the locus of the *N*-protecting group changes as a result of a ring opening/reclosure sequence *via* **112** as depicted in Scheme 2.12. We cannot distinguish at this time whether the initial formation of the expected adduct **109** occurs, and that this rearranges to form the observed adduct **108a**, or the formation of **111** occurs, which then rearranges to **108a**. It was also found that a similar reaction could be performed in aqueous acetone, leading to the formation of dihydroxylation product **108b** (Scheme 2.11 and Table 4, entry 4). The constitution was assigned on the basis of the ^{13}C NMR spectrum, in which only five unique carbon signals were observed and the stereochemical assignment is by analogy to **108a**. A sample of **108b** was recystallized from hot water and subjected to X-ray crystallographic analysis.

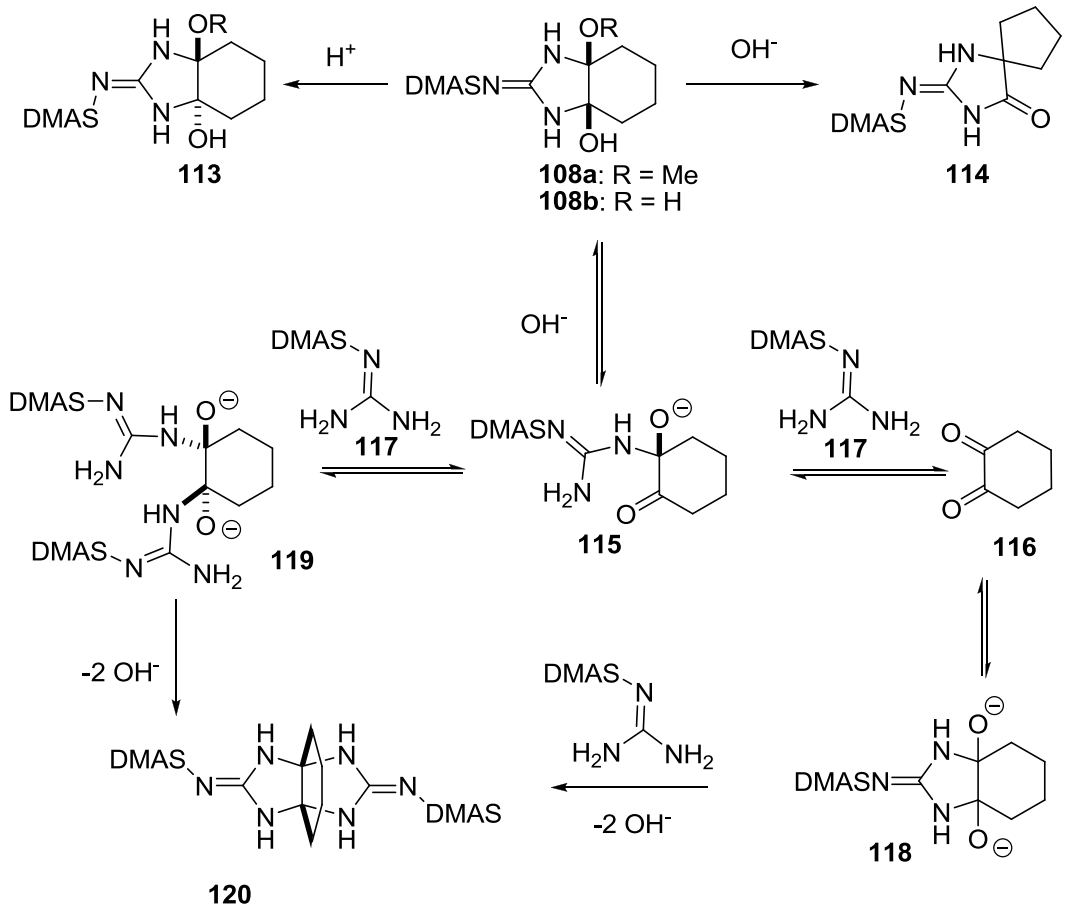
Somewhat surprisingly, we found that the initial product had undergone reaction to produce the propellane-like bis-guanidine **120** (Scheme 2.13).



Scheme 2.12

This spectrum of reactivity is interesting and presumably reflects the relative stabilities of the zwitterionic intermediate **110**. In the case of **100**, this is highly electron rich and so the carbocationic center is quite stable (*via* delocalization of the amine lone pair), which in turn permits collapse of the carbinolamine (with expulsion of the imine) and rearrangement. On the other hand, with the more electron deficient

system **101**, presumably the zwitterion **110** is relatively unstable, and is heavily solvated leading to trapping of the carbocation with methanol or water.



Scheme 2.13

In order to investigate whether the epimerization of **108a,b** occurs to form **113** (Scheme 2.13), they were heated in a solution of methanol and catalytic amount of HCl, and there was extra compound appearing from **108a**, without the methyl peak. Attempted purification of this compound was not successful as the product was not clean enough for the further characterization. However, from **108b** no indication of epimerization observed even at 60 ºC as only the starting material was recovered.

Next, both substrates were heated at 70 ºC in a basic solution of acetonitrile to see whether they rearrange to corresponding imidazolone **114.**[77] Interestingly, **108b** provided a product having almost identical ^{13}C NMR spectrum to the starting material; however the appearance of ^{1}H NMR spectrum was little bit different as it exhibits two equivalent DMAS groups and one cyclohexane ring. After taking the HRMS of this material, we determined that the structure is the same bis-guanidine **120** shown in Scheme 2.13, from the information we gathered during the attempted X-ray crystallographic analysis of **108b**. We believe that **108b** upon treatment with base tend to open the imidazolidine ring providing **115**, which eventually forms diketone **116** and DMAS protected-guanidine **117** and they exist as an equilibrium system. Formation of glycoluril derivative **120** may take place either by condensation of **116** with two guanidine derivatives (*via* **118**) or condensation of **115** with a guanidine derivative to form **119**, which provides **120** with loss of two hydroxide groups.[78, 79] However, we do not have any evidence to support either of these hypothesis, or another alternatives for this mechanism.

2.3 Comparison of oxidative ability of DMDO and oxaziridine

After observing this interesting oxidative ability of oxaziridines **87** and **88** with THB derivatives, their oxidation reactions were further observed by using more complex molecules used earlier in the DMDO chemistry.[52] The outcome of these studies is summarized in Table 5, which shows only the isolated yield of products.

While all the oxaziridine reactions were carried out in chloroform, DMDO reactions have been carried out in dichloromethane/acetone mixture and it should be

noted that the un-reacted starting materials were recovered from all the oxaziridine reactions; providing lower yields compared to DMDO reactions (Table 5, entry 2-5).

Table 2.5: Comparison of oxidative reactions of DMDO and oxaziridine **88**

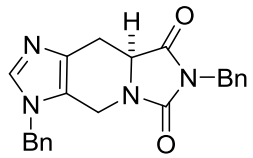
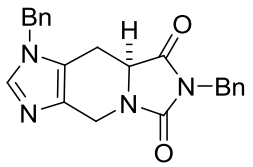
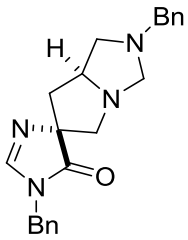
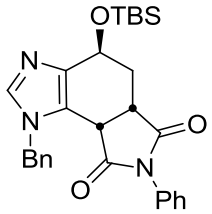
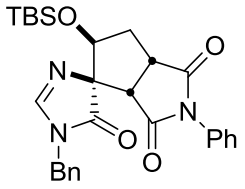
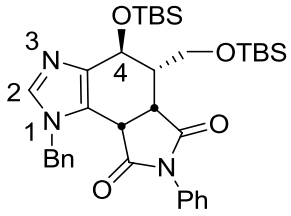
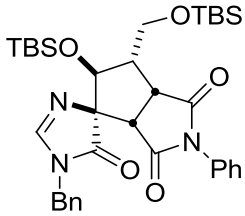
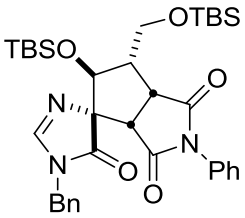
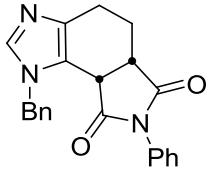
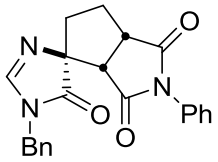
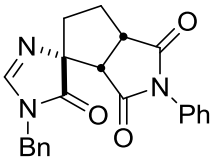
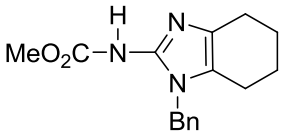
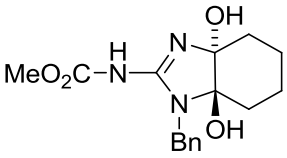
Entry	Substrate	Products	Yield/% DMDO	Yield/% **88**
1	  **121a/b**	 **122**	45	48
2	 **123**	 *endo*-**124**	82	70
3	 **125**	 *endo*-**126a**  *exo*-**126b**	14 56	13 20

Table 2.5 Continued

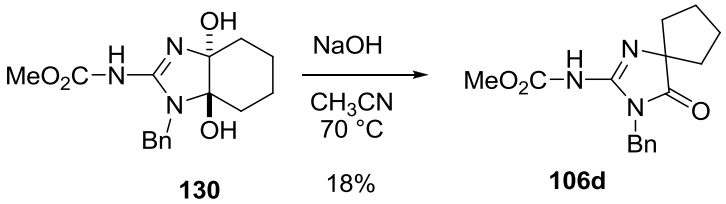
4	 **127**	 *endo*-**128a**  *exo*-**128b**	27 56	17 39
5	 **129**	 **130**	70	17

Although, the stereoselectivity of both oxidizing reagents are approximately the same, the product ratio in the case of the oxidation reaction of **125** with oxaziridine **88** is different from that of DMDO. This may be due to the presence of the 4-OTBS moiety, which results in a steric clash with the larger oxidant. As it was mentioned in the beginning of this chapter, some evidence has been collected about the dihydroxylation of **129** when treated with DMDO; however this was not investigated further during that time.[70] Therefore, during this study a solution of amide **129** in acetone-water mixture was treated with oxaziridine **88** to isolate dihydroxy derivative **130** in 17% with some unreacted starting material (Table 5, entry 5). Since this was an interesting finding, **129** was also treated with a freshly prepared DMDO solution to isolate **130** in 70%, confirming the dihydroxylation of

amide **129**, with both oxidizing reagents. Relative stereochemistry of this product was assigned on the basis of water trapping the carbocation on the opposite face from the oxidative reagent approach.[76]

Since we found DMDO is also feasible for the oxidative addition of 2-aminoimidazole derivatives, amine **101** was reacted with acetone-water solution of DMDO, and we were delighted to isolate the dihydroxy derivative **108b** in 17% yield.

Once the dihydroxy derivative **130** was isolated from the reaction of amide **129** with DMDO, we were interested to observe the possibility of rearrangement of **130** to **106d** (Scheme 2.14). In fact, we found that the treatment of **130** with a basic solution of acetonitrile tends to smooth rearrangement providing **106d**. Although the yield was low, this was an interesting result and the yield can be improved once the reaction conditions are optimized.

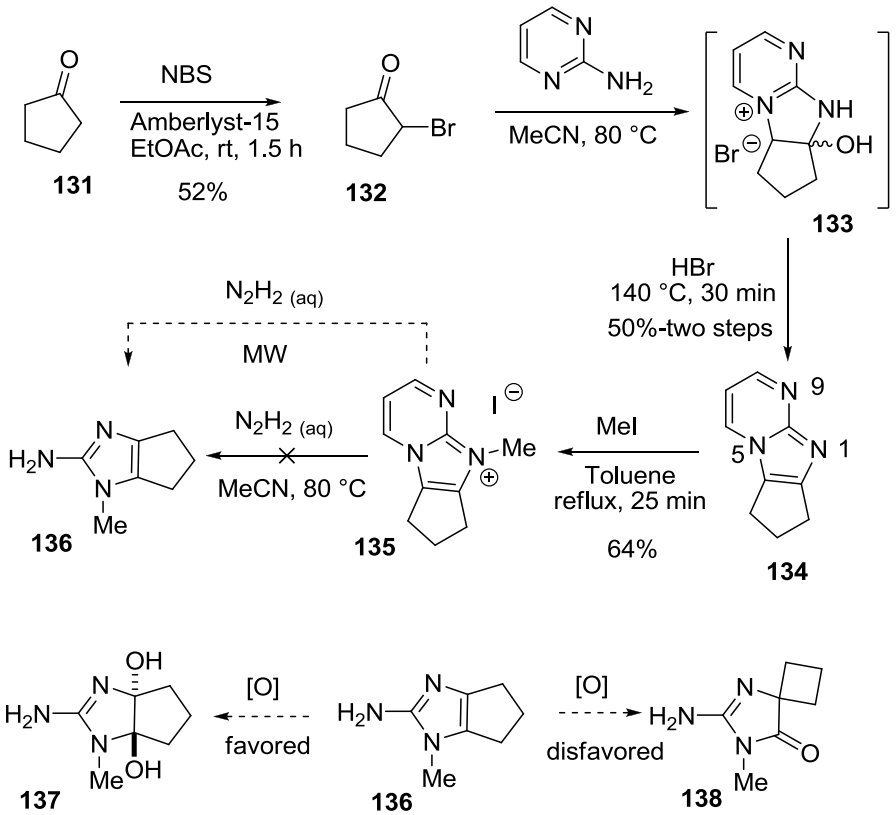


130 **106d**

Scheme 2.14

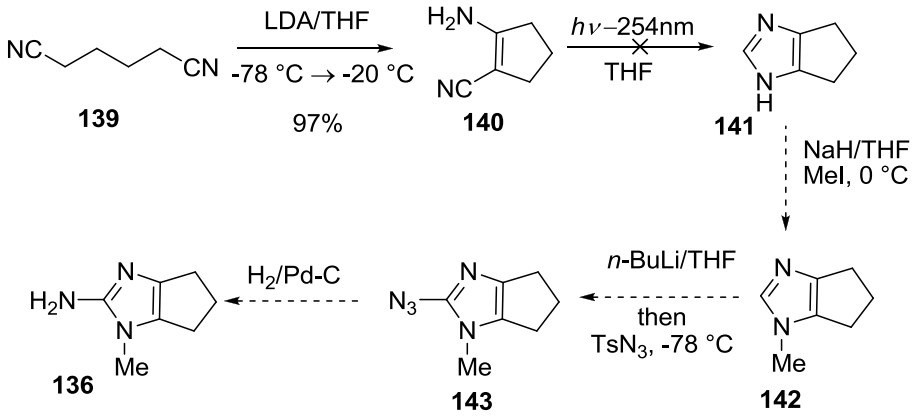
2.4 Synthetic attempt to 2-Amino-1-methyl-1,4,5,6-tetrahydrocyclopentaimidazole (**136**)

Although we observed the oxidative addition with amine **101** to produce dihydroxy derivatives **108**, there is a net migration of DMAS group from the *N*1-position to exocyclic C2-amine of the imidazole. Since some of our eventual target molecules, spirocalcaridine A (**12a**) and spirocalcaridine B (**12b**) contain cyclopenta[*d*]imidazole, we wanted to establish whether five-five ring system undergoes oxidative addition favoring **137** (Scheme 2.15) since there is highly energetic an unfavorable ring compression if it undergoes oxidative rearrangement to form **138**.



Scheme 2.15

First, cyclopentanone (**131**) was brominated with NBS and catalytic amount of Amberlyst-15® to isolate 2-bromocyclopentanone **132**,[80] which was treated with 2-aminopyrimidine directly under refluxing acetonitrile to produce hydroxyl salt **133**.[81] Treatment of **133** with concentrated HBr at 140 ºC provided imidazo[1,2-*a*]pyrimidine **134** in one pot. It has been shown that the methylation of imidazo[1,2-*a*]pyrimidine takes place at *N*1-position rather than at *N*9-position,[82, 83] therefore a solution of **130** and MeI in toluene was heated to reflux to yield methiodide **135** in 64% yield. Initial attempt to convert **135** to 2-aminoimidazole **136** with aqueous hydrazine failed as only a trace amount of the product was isolated.[81] Next, this reaction was attempted using microwave irradiation; however, this sequence could not be finished as the micro-wave instrument was not functioning well.



Scheme 2.16

Alternatively, Thorpe–Ziegler cyclization of the adiponitrile **139** was carried out using LDA to synthesize **140** (Scheme 2.16),[84] which was subjected to photochemical irradiation at 254 nm for 40 h.[85] However, no conversation of **140** to imidazole **141** was observed form this reaction, and the repeated attempts pursue this

strategy were also failed. Therefore, rather than spending time continuing this study, it was decided to advance to next step; the total synthesis of calcaridine A (**13**), which is discussed in Chapter 3 in details.

2.5 Summary

We have demonstrated that aryl *N*-sulfonyloxaziridines are effective reagents for the oxidative rearrangement of tetrahydrobenzimidazoles to the corresponding spiro-fused 5-imidazolones or in some cases to bis-addition products under mild conditions. There are some fairly subtle effects in play here that lead to various outcomes depending on the nature of the 2-substituent and the solvent employed. We found that the presence of electron donating group at imidazole *N*1-position facilitates the oxidative rearrangement of imidazole derivatives in consistence with DMDO. Also of note is the fact that a free amine can be tolerated with these substrates leading to either rearrangement or addition of solvent depending upon the *N*1-protecting group.

We have also compared the oxidative abilities of *N*-sulfonyloxaziridines and DMDO, and found that while stereoselectivity of both reagents are the same, oxaziridine shows relatively slow reactivity towards complex molecules thereby requiring higher reaction time. This might be due to the difference in steric clashes between oxidant and the substrates. However, oxaziridines provide a shelf-stable alternative to DMDO, which has to be prepared in isolated form to effect this rearrangement. We also found that both DMDO and oxaziridine can be used for the oxidative addition of imidazole derivatives to synthesize corresponding dihydroxy

derivative, and some of these dihydroxyderivatives can be rearranged to corresponding spiro-imidazolones under basic conditions.

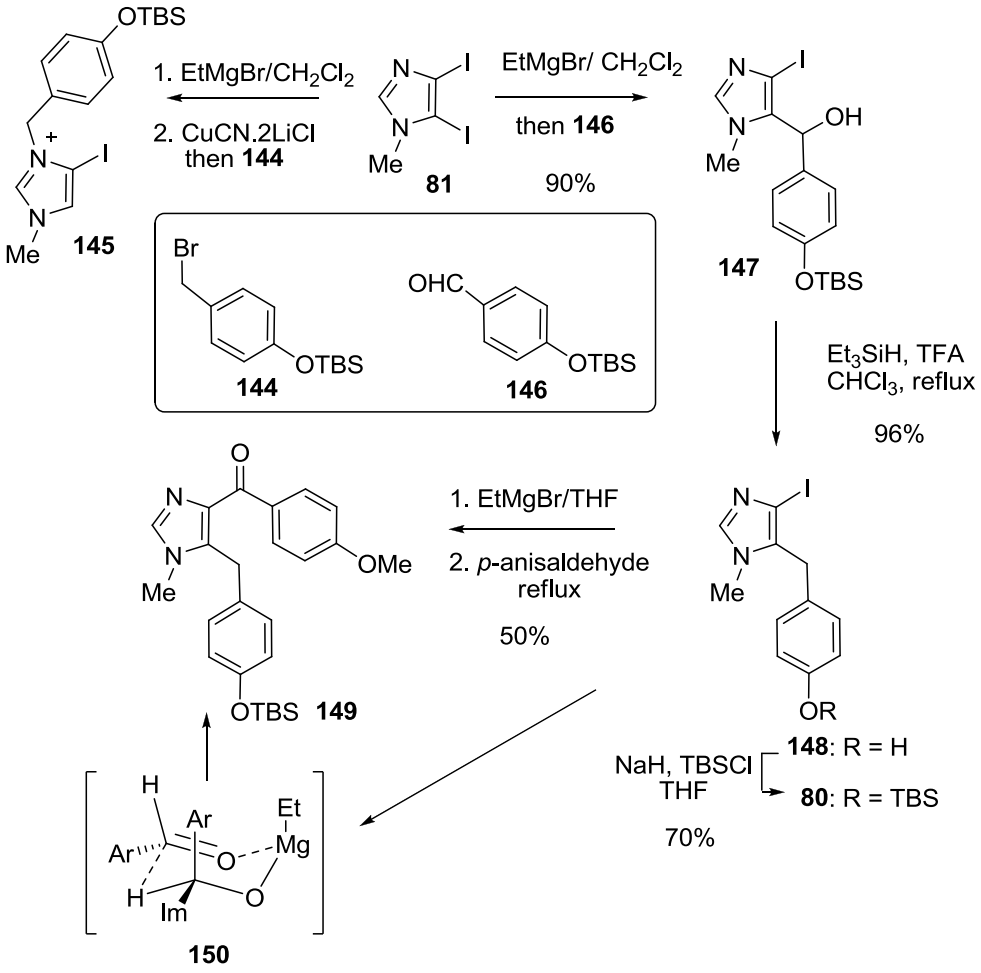
CHAPTER 3

TOTAL SYNTHESIS OF THE *LEUCETTA*-DERIVED ALKALOID CALCARIDINE A

3.1 First generation approach to calcaridine A

As described in the retrosynthetic analysis of calcaridine A in Chapter 1, our initial approach to calcaridine A (**13**) was started by metalating the 4,5-diiodoimidazole **81** with EtMgBr in CH_2Cl_2 at C5, and then treating the resulting Grignard with a 1.0 M solution of CuCN.2LiCl followed by the siloxybenzyl bromide derivative **144**.[41-45] However, rather than the required substituted product **80**, we obtained the imidazolium salt **145** (Scheme 3.1). Presumably, the organometallic species and the alkylating agent do not react and upon quenching the reaction, the reduced heterocycle undergoes *N*-alkylation. To circumvent this issue we modified the approach to employ an aldehyde, the product of which would then be reduced to **80**. Thus, metalation of **81** with EtMgBr and then reaction with the siloxybenzaldehyde **146**[86] provided the expected doubly benzylic alcohol **147** (Scheme 3.1). Ionic reduction of **147** with Et_3SiH in the presence of TFA provided the reduced product with concomitant desilylation (**148**),[36] thus the reduction product was treated with TBSCl/NaH to afford **80**.[87] The resulting heterocycle was subjected to a second round of metalation and treatment of the *in situ* formed Grignard with *p*-anisaldehyde (**53**). However, rather than obtaining the expected alcohol, we either obtained a complex mixture of products when the reaction was conducted in CH_2Cl_2, or the

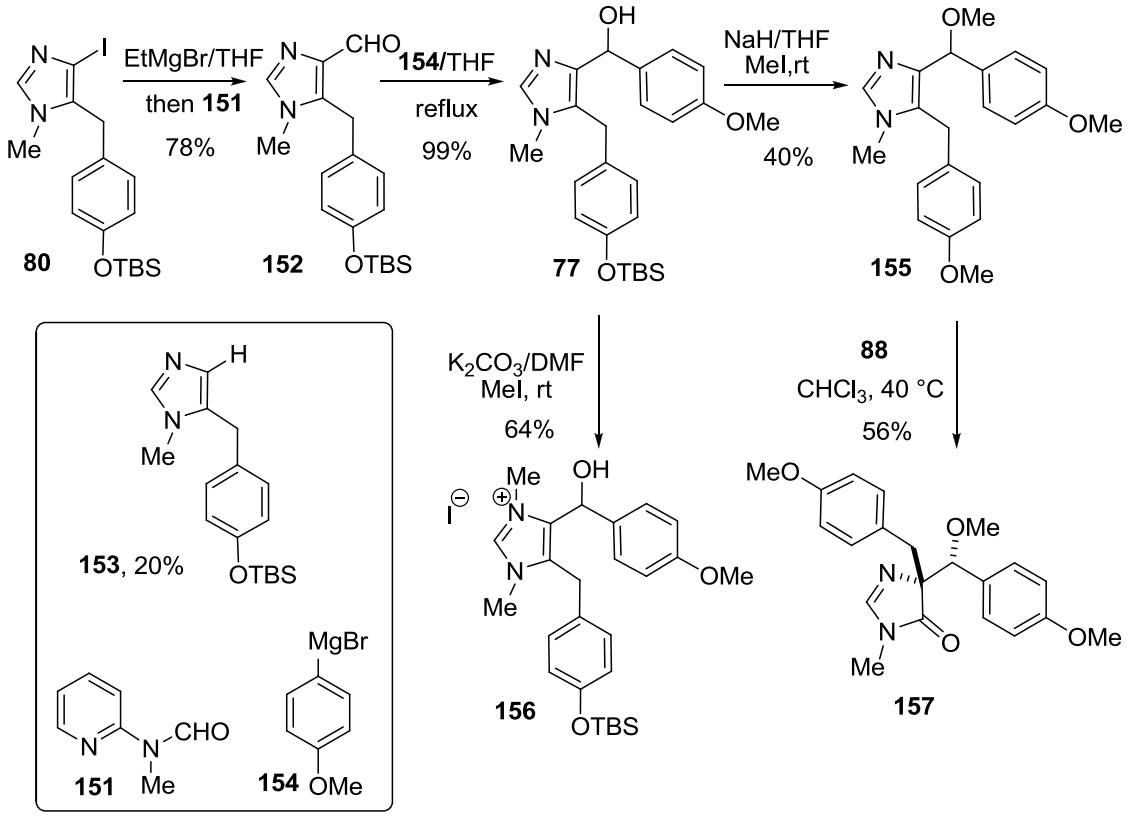
corresponding ketone derivative **149** in approximately 50% yield along with a similar amount of the *p*-methoxybenzyl alcohol when the reaction was performed in THF. The latter outcome suggests that the initial (and required) alcohol is formed but then undergoes an Oppenhauer-type oxidation (*via* **150**), providing ketone **149**.[88-91]



Scheme 3.1

Unfortunately we were unable to find conditions that prevented the oxidation, and therefore we adopted a slightly different tactic, in which the imidazole was first

formylated and then the remaining aromatic fragment introduced *via* Grignard chemistry (Scheme 3.2).

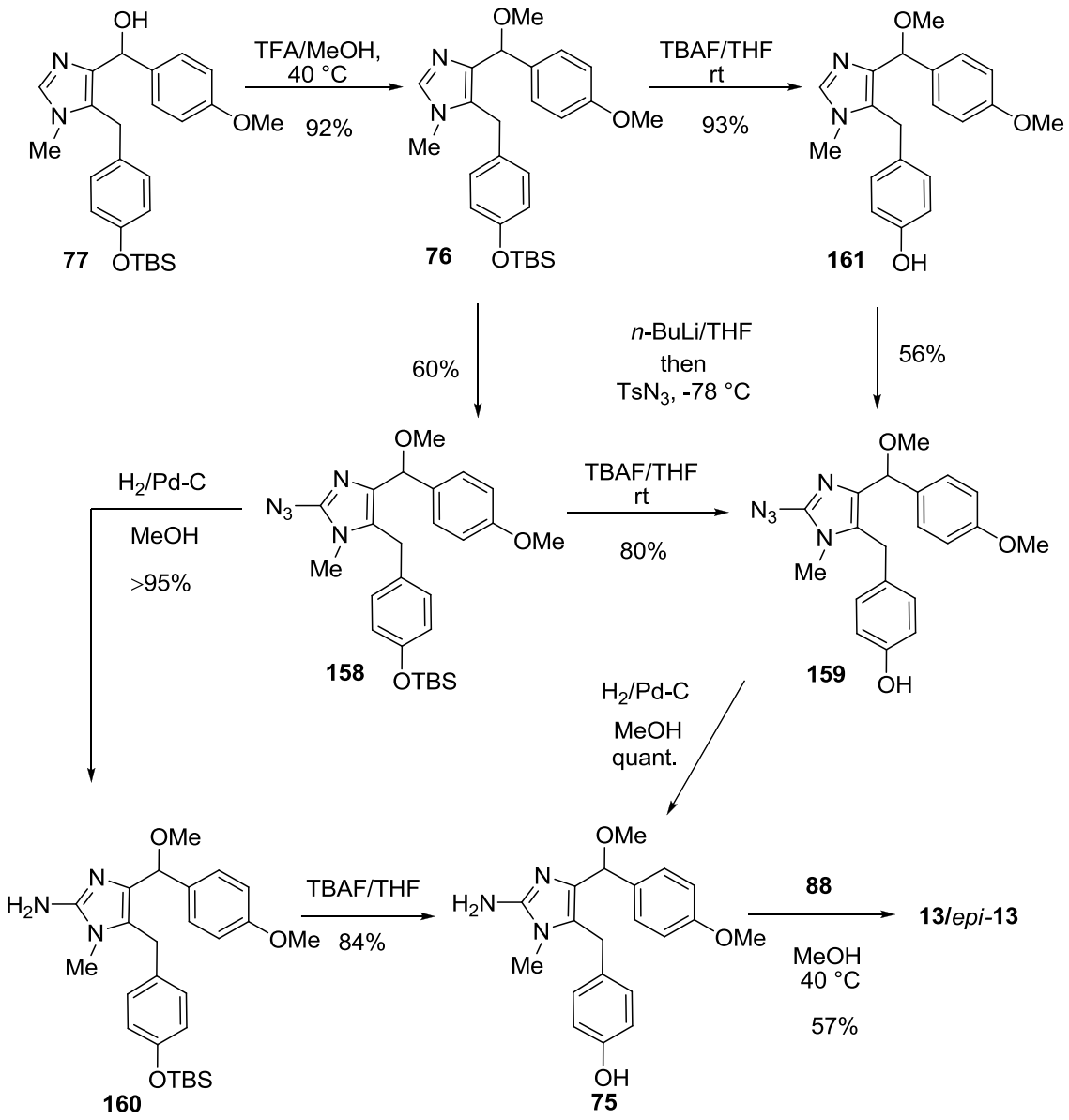


Scheme 3.2

Metalation was performed as before with EtMgBr and then reaction with the *N*-methyl-*N*-(2-pyridyl)formamide (**151**)[92] provided the corresponding aldehyde **152** in addition to de-halogenated product **153** (Scheme 3.2). Addition of 4-methoxyphenylmagnesium bromide (**154**)[93] to **152** led to the formation of alcohol **77** in excellent yield. Initially attempts to methylate **77** with K_2CO_3/MeI were compromised by *N*-alkylation and the formation of the imidazolium salt **156**. Then, attempted methylation of alcohol **77** with NaH and MeI also failed as this resulted the replacement of silyl protection **77** providing **155**, with 10% of desired methyl ether

76. At this point we decided to test whether the oxidative rearrangement was feasible on this type of substrate, as all of the previously investigated examples were tetrahydrobenzimidazole derivatives (Chapter 2).[51] Gratifyingly, it was found that upon treatment of **155** with 2.5 equivelent of *N*-sulfonyloxaziridine **88** in chloroform at 40 °C a smooth rearrangement took place providing 2:1 mixture of imidazolone **157** in a modest, but unoptimized, yield. Only one pure diastereomer was isolated after the recystallization of the mixture with EtOAc and benzene, and although we have not rigorously assigned the relative stereochemistry, we have tentatively assigned it as indicated in Scheme 3.2 based on the similarity of its ^{1}H NMR data compared to *epi*-calcaridine A (*epi*-**13**, see Scheme 3.4).

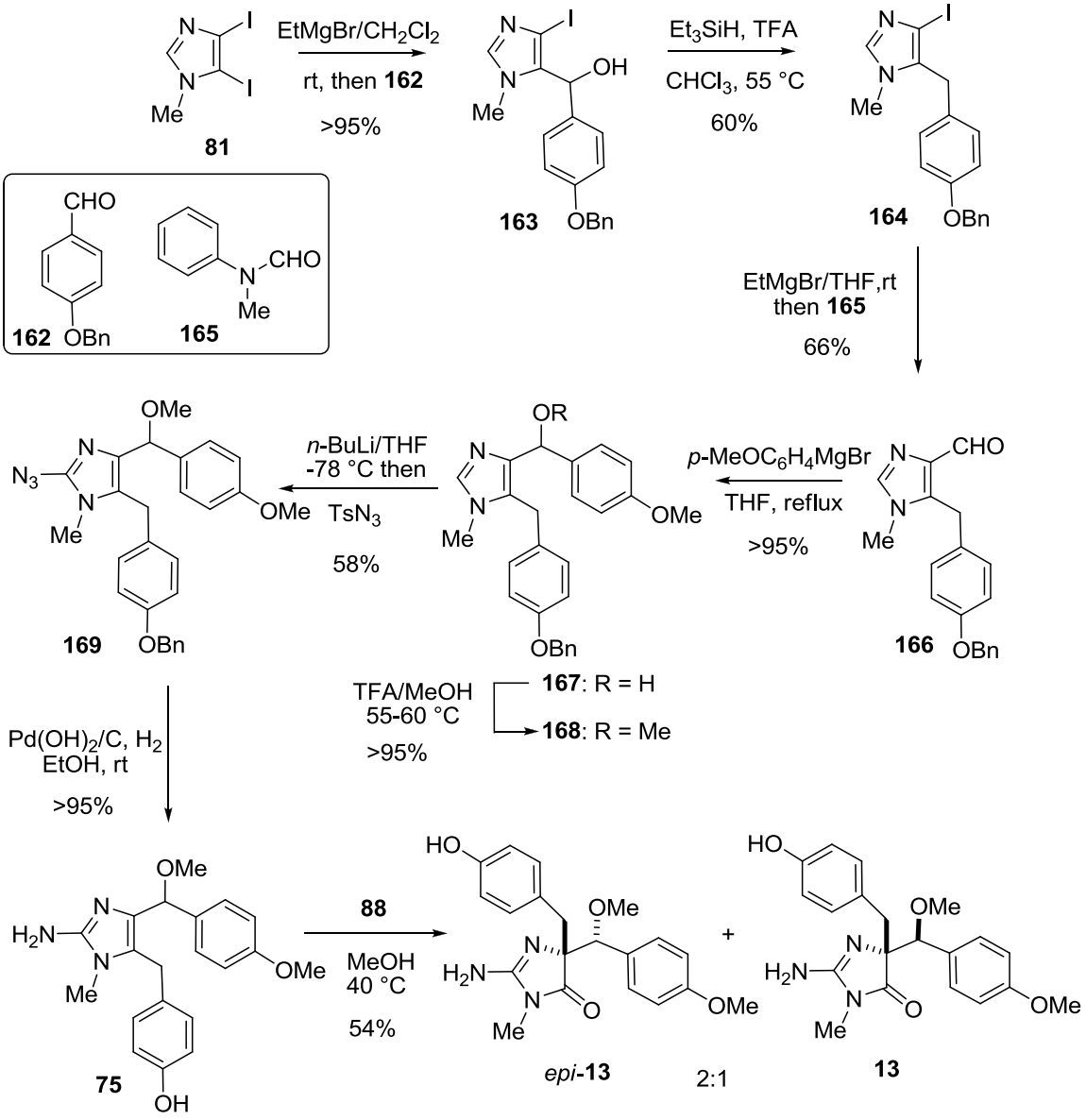
As above methylation attempts failed, **77** was converted to the methyl ether **76** by acid-catalyzed substitution (Scheme 3.3).[94] Next, the *C*2-azidation of **76** was carried out using *n*-BuLi and TsN_3 at -78 ºC to obtain **158** in 60% yield.[46] Catalytic reduction of this azide with H_2/Pd-C provided amine **160** in excellent yield.[95] Then, the attempted removal of TBS protection was problematic, since the purification was difficult with highly polar amine **75**. Therefore, by switching these two steps, removal of the TBS protection from **158** was carried out using TBAF to provide azide **159**, which was also synthesized *via* phenol derivative **161** as shown in Scheme 3.3. The catalytic reduction of this azide with H_2/Pd-C provided amine **75**, which was treated with 2-(phenylsulfonyl)-3-(4-nitrophenyl)oxaziridine (**88**) in methanol at 40 ºC leading to the formation of calcaridine A (**13**) and *epi*-calcaridine A (*epi*-**13**) as a 1:2 mixture in 57% yield (Scheme 3.3). Unfortunately, at this stage we were unable to separate the two diastereomers, but it was clear from the NMR data that the minor of the two diastereomers was calcaridine A (**13**).



Scheme 3.3

Although the general feasibility of the strategy had been demonstrated, issues with regard to the deprotection-reprotection sequence (**147**→**80**, Scheme 3.1) and a subsequent deprotection prompted us to make changes to the protecting group on the phenolic oxygen and to use a Bn-group. The choice of this protecting group had the added advantage that it could be removed by catalytic hydrogenation, which would be

used in the late-stage reduction of the 2-azido moiety and therefore would telescope the sequence to some extent.



Scheme 3.4

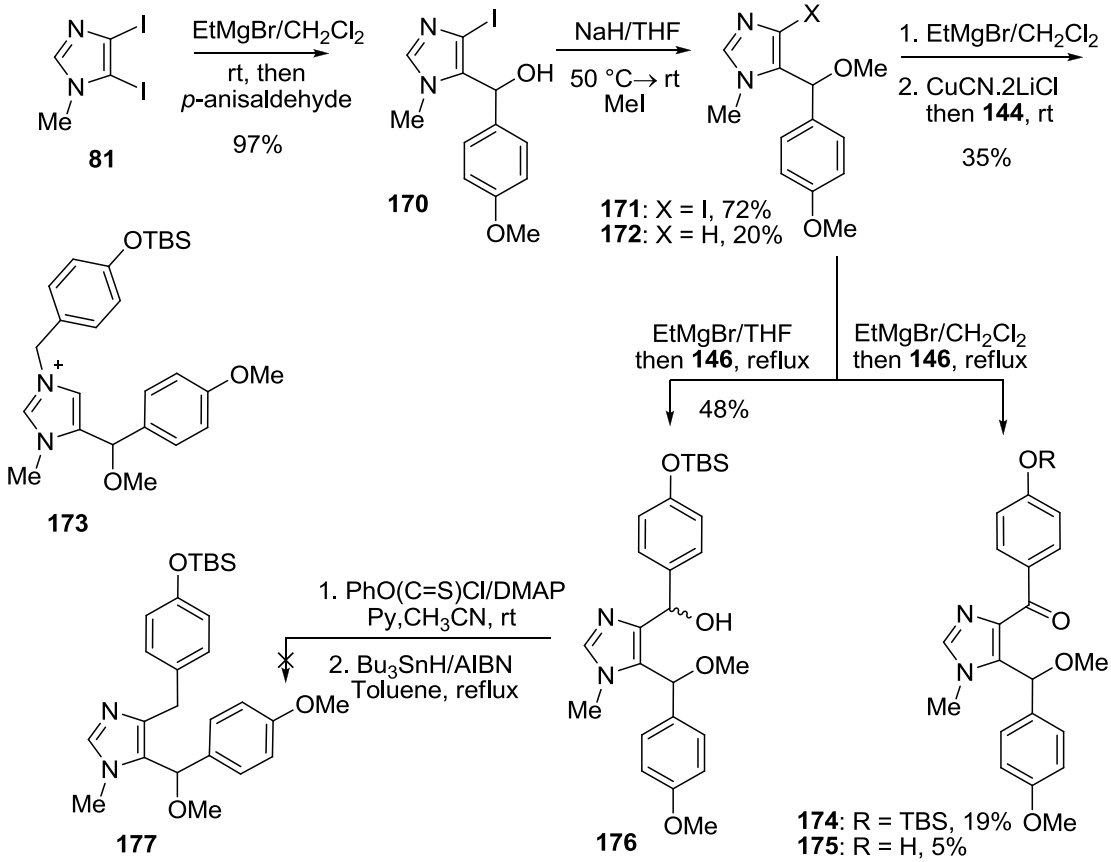
Consequently, diiodoimidazole **81** was converted to alcohol **163** through metalation and reaction with the benzyl-protected aldehyde **162**,[96] Et_3SiH-mediated reduction provided the net substitution product **164** (Scheme 3.4). Metalation at C4,

and reaction with *N*-methylformanilide (**165**) provided the aldehyde **166**, which was reacted with *p*-methoxyphenylmagnesium bromide (**154**) to product the required alcohol **167**. Acid catalyzed ether formation provided **168** in excellent yield.

At this stage the 2-amino substituent was introduced by deprotonation of **168** at C2 with *n*-BuLi and reaction with TsN_3 affording **169** (Scheme 3.4). Treatment of **169** with $Pd(OH)_2/C$, H_2 resulted in conversion of the azido moiety into the amine and hydrogenolysis of the benzyl protecting group providing 14-methoxynaamine A (**75**).[97] Exposure of this substrate to **88** in MeOH at 40 °C led to oxidative rearrangement and the formation of a 2:1 mixture of *epi*-calcaridine A (*epi*-**13**) and calcaridine A (**13**), and the attempted separation of this mixture using HPLC, MPLC and preparative TLC failed to provide the individual diastereomers.

3.2 Second generation approach to calcaridine A

On analysis of stereochemical outcome of the oxidative rearrangement, we hypothesized that it might be possible to improve the diastereoselectivity by switching the location of the two benzylic substituents, thereby placing the pre-existing stereo center closer to the point of the oxygen transfer at C5 of the imidazole.[51] To accomplish this necessitated a minor re-engineering of the synthetic sequence, but in reality required modifying the order of addition, this is in fact one of the strengths of our approach to these natural products.

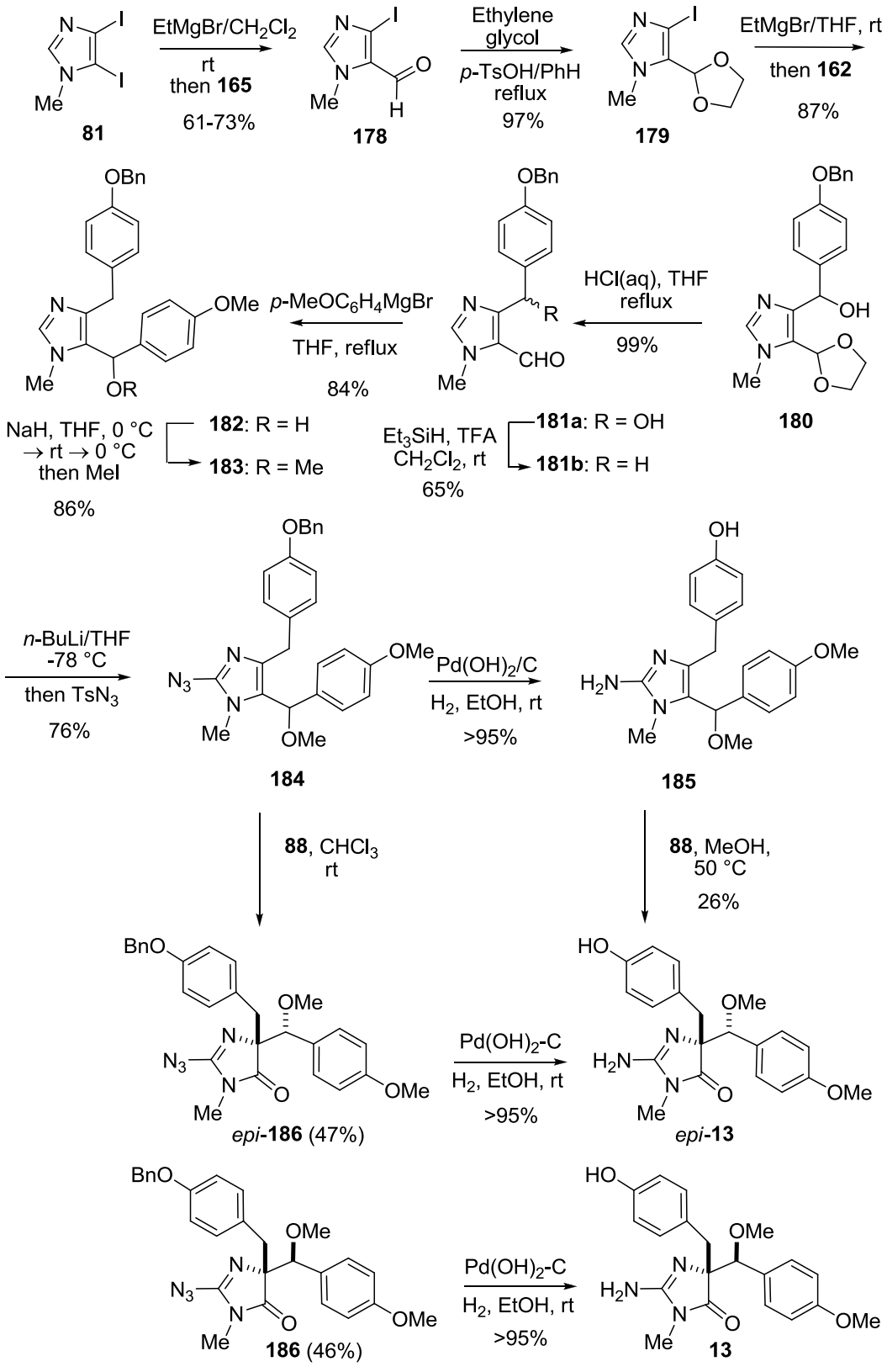


Scheme 3.5

Accordingly, **81** was metalated at C5 and reacted with *p*-anisaldehyde (**53**) to provide the alcohol **170** in excellent yield (Scheme 3.5), which was converted into the methyl ether **171** by treatment with NaH followed by methyl iodide. Initially we attempted to introduce the remaining benzyl fragment by metalation with EtMgBr, conversion to the cuprate (CuCN.2LiCl) and then trapping with the TBS-protected benzyl bromide derivative **144**, but this strategy again led to the formation of the imidazolium salt **173**. Using benzaldehyde **146** as electrophile with the Grignard derived from **171** in CH_2Cl_2 was partially successful providing the ketone **174** and the desilylated ketone **175**. The desilylated ketone **175** was subjected to reduction under Wolff-Kischner conditions, but this experiment was unsuccessful. Interestingly, repetition of the Grignard chemistry in THF provided the anticipated diol **176** as a 2:1 mixture of diastereomers (stereochemistry unassigned) in addition to 43% of the de-halogenated imidazole **172**. Unfortunately, attempts to remove the hydroxyl group in **176** by radical based deoxygenation methods were also unsuccessful.[98]

As these direct approaches were not successful we adopted the approach delineated in Scheme 3.6 to assemble the inverted substrate. Thus, metalation of **81** and trapping with *N*-methylformanilide (**165**) provided the corresponding aldehyde **178**, which was protected as an acetal with ethylene glycol, providing **179**.[99] Metalation at C4 and reaction of the resulting Grignard with **162** provided the alcohol **180**. Attempted ionic reduction of the alcohol at this stage was complicated by reduction of the acetal, but it was found that deprotection of the acetal followed by reduction of the alcohol produced **181b**.[100] Addition of *p*-methoxyphenylmagnesium bromide provided the expected alcohol **182**. Attempts to convert this alcohol to the methyl ether **183** with TFA and methanol were not successful at this stage; therefore

this was achieved by treating with NaH and MeI.[101] Installation of the 2-amino substituent was accomplished through lithiation at C2 and reaction with TsN_3 (Scheme 3.6). Catalytic hydrogenation led to reduction of both the azide moiety to the amino group and hydrogenolysis of the O-benzyl protecting group providing **185**, the regioisomer of 14-methoxynaamine A. Subjection of **185** to oxidative rearrangement with the Davis reagent **88** provided a 1:1 mixture of calcaridine A (**13**) and *epi*-calcaridine (*epi*-**13**) in low but unoptimized yield. Although there had been some change in the diastereoselectivity of the rearrangement, the separation problem persisted. As shown in Chapter 2, our earlier studies with the oxidative rearrangement chemistry had demonstrated that 2-azido substituted imidazoles would participate in the rearrangement and so we subjected **184** to oxidation in the hope that the diastereomers resulting from this reaction might be more easily separable. We were gratified to find that although there was no change in the diastereoselectivity (1:1 mixture), the resulting 2-azido imidazolones **186** and *epi*-**186** were separable by chromatography. Both diastereomers were individually subjected to catalytic hydrogenation to provide in essentially quantitative yield, calcaridine A (**13**) and *epi*-calcaridine A (*epi*-**13**) as solids to complete the total synthesis of (±)-calcaridine A in 10 steps with 8.4% overall yield. The NMR spectra of one of the diastereomers matched that reported in the literature.[19] Once pure, as an added bonus, the non-natural diastereomer provided suitable crystals for X-ray analysis (Figure 3.1), which in addition to confirming the connectivity provided the relative stereochemistry of this diastereomer and by analogy that of the natural product. We were also able to use the crystalline non-natural diastereomer to seed and effect fractional crystallizations to purify all of the previously prepared diastereomeric mixtures.


EtMgBr/CH2Cl2
rt
then 165
61-73%
81
178
Ethylene glycol
p-TsOH/PhH
reflux
97%
179
EtMgBr/THF, rt
then 162
87%
180
HCl(aq), THF
reflux
99%
p-MeOC6H4MgBr
THF, reflux
84%
NaH, THF, 0 °C
→ rt → 0 °C
then MeI
86%
182: R = H
183: R = Me
Et3SiH, TFA
CH2Cl2, rt
65%
181a: R = OH
181b: R = H
n-BuLi/THF
-78 °C
then TsN3
76%
184
Pd(OH)2/C
H2, EtOH, rt
>95%
185
88, CHCl3
rt
88, MeOH,
50 °C
26%
epi-186 (47%)
Pd(OH)2-C
H2, EtOH, rt
>95%
epi-13
186 (46%)
Pd(OH)2-C
H2, EtOH, rt
>95%
13

Scheme 3.6

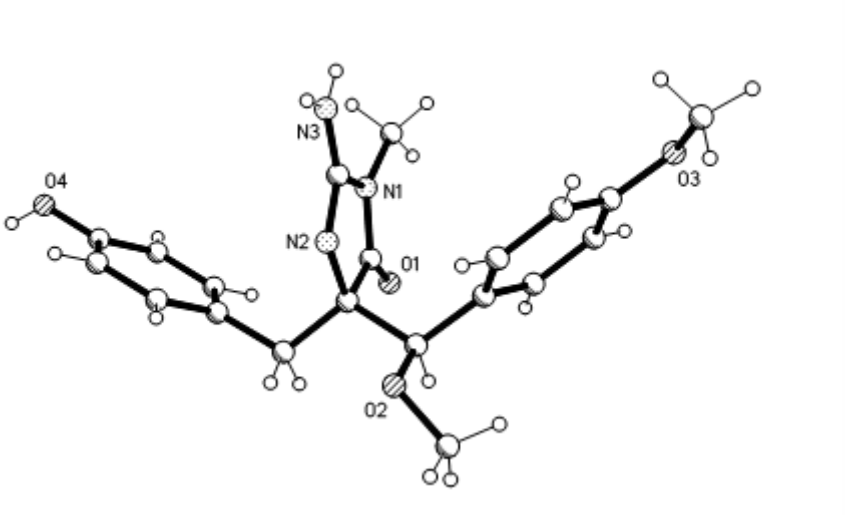


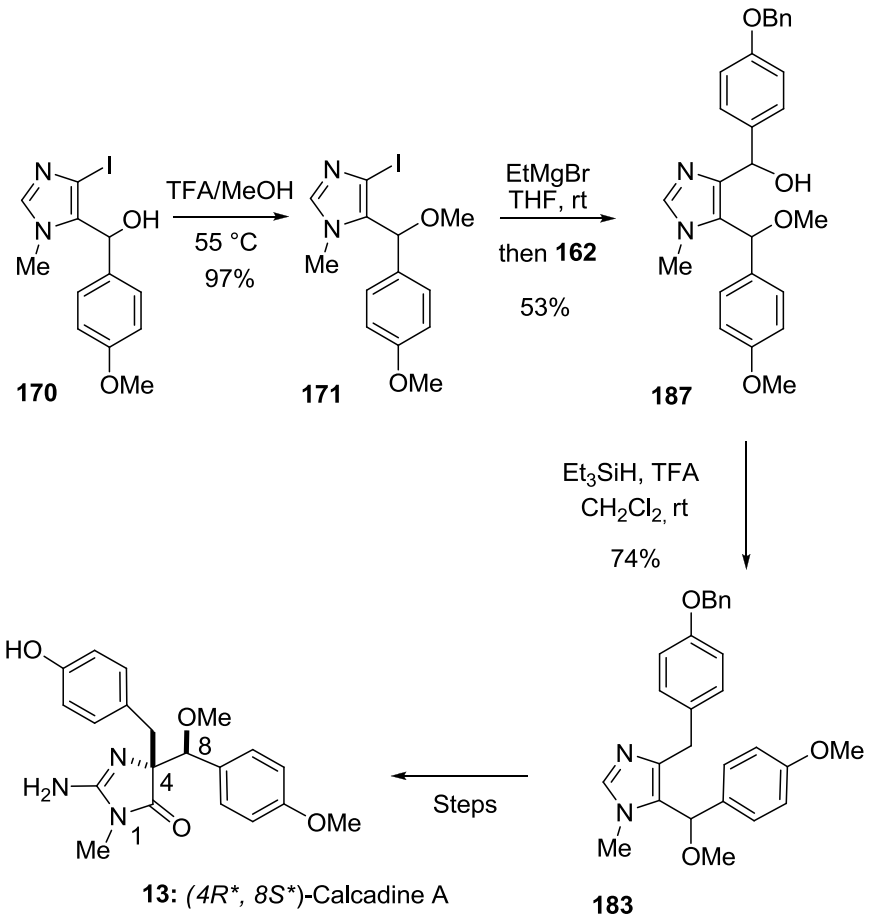
Figure 3.1: X-ray crystal structure of *epi*-calcaridine A (*epi*-**13**)

In one last experiment to improve the overall efficiency we attempted to establish whether either of the individual diastereomers of **13** or *epi*-**13** could be converted to the other through acid catalyzed ether exchange. It was hypothesized that the carbocation would be stabilized by the *p*-methoxy group and thus may facilitate an epimerization. Unfortunately, we found treatment of either diastereomer with cat. HCl in MeOH at various temperatures up to reflux led to no discernable epimerization.

3.3 A shorter approach to calcaridine A *via* a modified method

Although we had completed the total synthesis of calcaridine A as shown in Scheme 3.5, several aspects of the approach detracted considerably from the synthesis. Specifically two features were less than optimal. First, the fact that the *p*-methoxybenzyl moiety was introduced in two steps (**81→178** and **181b→182**) was not optimal and second the necessity to protect and subsequently deprotect the carboxyaldehyde moiety in **178** was particularly disturbing. Indeed, the synthesis of

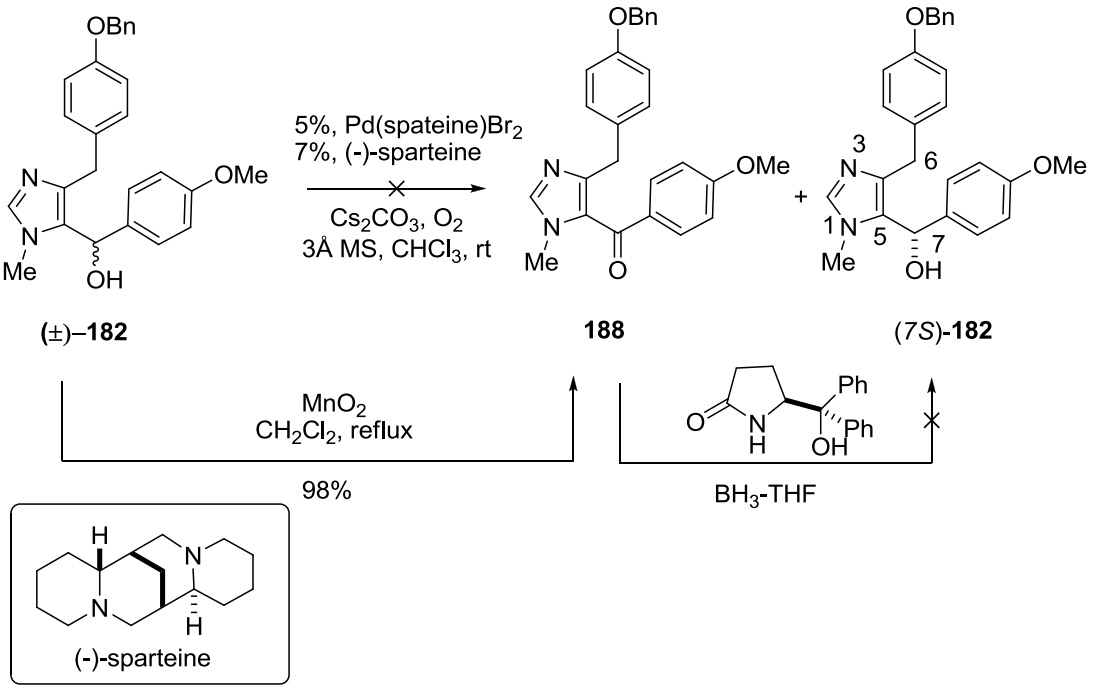
imidazole **183** required a total of seven steps and provided the substrate in 25% overall yield. Therefore, we modified the synthetic route as shown in Scheme 3.7 by treating **170** with methanol and TFA to produce methyl-ether **171** which was reacted with EtMgBr and 2.5 equivelent of aldehyde **162** at room temperature for 3 days to provide the alcohol **187** in 53% as a 2:1 mixture of diastereomers. When this reaction was repeated with 4 equivelent of the aldehyde at elevated temperature for 3 days, no improvement of the yield was observed, and we have found earlier this reaction is problematic when 1.1 equivelent of aldehyde is used (Scheme 3.1). The next transformation in the sequence is where the major challenge lay, that is the chemoselective deoxygenation of the hydroxyl group in the presence of the methyl ether, both of which are doubly benzylic. Gratifyingly, we found that the benzylic alcohol was reduced selectively at room temperature simply through the addition of Et_3SiH and TFA to a solution of alcohol **187** in dry dichloromethane providing imidazole **183** in four steps with 36% overall yield in contrast to the sequence shown in Scheme 3.6. Continuation of the sequence finished the second total synthesis of (±)-calcaridine A in 7 steps with 12.5% overall yield.



Scheme 3.7

3.4 Attempts towards an asymmetric synthesis of calcaridine A

Since we had determined the relative stereochemistry of calcaridine A to be (*4R*, 8S**) through an X-ray crystal structure determination, we wished to carry out the asymmetric synthesis of natural occurring enantiomer of calcaridine to assign the absolute stereochemistry. In this regard, first we attempted the palladium-catalyst enantioselective oxidation of the alcohol **182** using the method described recently by Stoltz and co-workers (Scheme 3.8).[102] However, in this case no oxidized product was detected in the ^{1}H NMR spectrum of the crude material. This result might be due to the steric hindrance of the doubly benzylic alcohol used this attempt.

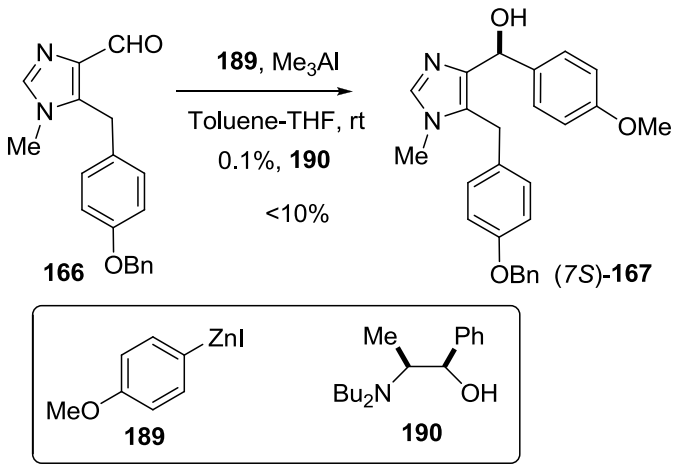


Scheme 3.8

Next, we began to investigate asymmetric reduction of ketone **188**, hoping to isolate the (*7S*)-**182**, with CBS catalyst (Scheme 3.8).[103] In this regard, racemic alcohol **182** was oxidized to ketone **188**, which was subjected to CBS asymmetric reduction.[104] Unfortunately, this reduction did not work providing only un-reacted **182**, although the catalyst was found to work well with benzophenone in a trial experiment. Perhaps the imidazole behaves as Lewis base with the reducing agent.

Next we tried direct asymmetric 1,2-addition of organozinc species **189** to aldehyde **166** following the recent report by Woodward (Scheme 3.9).[105] As most of these asymmetric additions have been done in toluene, it was difficult for us to carry out this reaction in toluene due to the poor solubility of the aldehyde **166**. Despite

several screens of solvents, we were unable to find a suitable solvent for this reaction, and therefore we chose not to pursue this approach any further.



Scheme 3.9

3.5 Summary

We have developed a biomimetically-guided total synthesis of the *Leucetta*-derived alkaloid calcaridine A using position selective metalations of 4,5-diiodo-1-methyl-1*H*-imidazole to provide a 14-methoxynaamine A derivative, which was subjected to oxidative rearrangement with 3-(4-nitrophenyl)-2-phenylsulfonyloxazirid-ine to construct the imidazolone core of calcaridine A. Although the oxidative rearrangement of tetrahydrobenzimidazole was discussed in Chapter 2, oxidative rearrangements discussed in this chapter are the first examples of rearrangements involving substrates other than a tetrahydrobenzimidazole type derivative. While these experiments do not prove that calcaridine A is formed biosynthetically *via* an oxidative rearrangement of 14-methoxynaamine A, they do demonstrate that it is a

feasible pathway. Although chemically the rearrangement exhibits poor diastereoselectivity, an enzyme mediated process might be expected to proceed with substantially higher levels of selectivity. In addition to the total synthesis, we have determined the relative stereochemistry of the natural product through X-ray crystallography of the epimeric congener. In the course of this project we have prepared quite large quantities of both diastereomers which have been submitted for biological testing through the NCI Developmental Therapeutics Program, and the NIH Molecular Libraries Screening Centers Network.

During this synthetic studies we have also found *N*1-methyl protected imidazole derivative does not undergo cross coupling reaction after metalation (EtMgBr), trans-metalation (CuCN.2LiCl) followed by the addition of aryl bromide to provide 4 (5)-substituted imidazole derivative.[41]

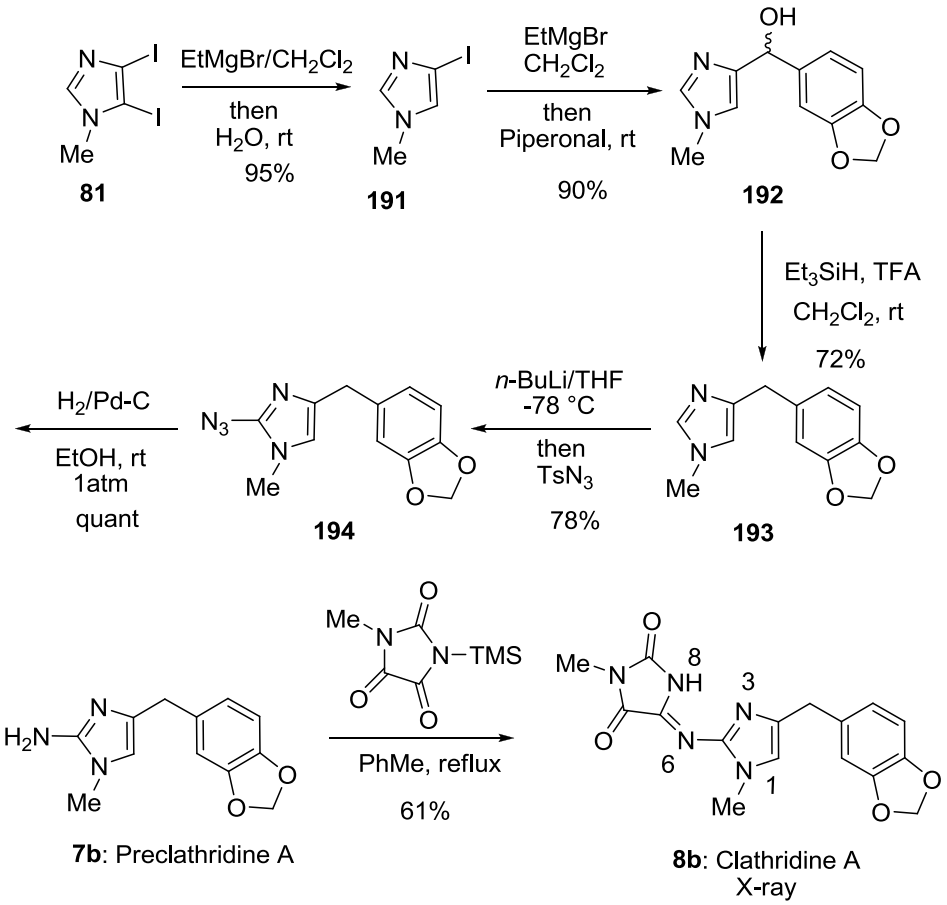
CHAPTER 4

TOTAL SYNTHESIS OF OTHER *LEUCETTA* ALKALOIDS

As described in the Chapter 3, we have developed methodology based on the elaboration of polyhaloimidazoles *via* Grignard reagents which does not require protection (and subsequent deprotection) of the more reactive 2- and 5-positions, which is necessary using deprotonation strategies or halogen–lithium exchange.[31-33, 35-37] These approaches to calcaridine A, described in the previous chapter provided us a better understanding of synthetic routes, and intermediates that can be used for the total synthesis of other members of both the *Leucetta* and *Clathrina* families of alkaloids. In this chapter we describe the total synthesis of several other *Leucetta* and *Clathrina* alkaloids by expanding on this strategy.

4.1 Total synthesis of preclathridine A (**7b**) and clathridine A (**8b**)

There are three previously reported syntheses of preclathridine A and clathridine A (Chapter 1, Schemes 1.6, 1.7 and 1.13).[32, 39, 106] The first report by Ohta's laboratory employs the sequential metalation (Br→Li) of an imidazole derivative, but requires blocking groups (thiophenyl groups) to prevent unwanted metalation at the 2- and 5-positions.[32] Both the Molina[106] and the Eycken[39] groups have utilized methods that involve the *de novo* synthesis of the 2-aminoimidazole system *via* iminophosphoranes and masked guanidine derivatives, respectively. While strategically our approach is closely related to that reported by the Ohta's laboratory, the use of Grignard reagent avoids the use of protecting groups on the imidazole ring.



Scheme 4.1

Our synthetic studies commenced with the conversion of 4,5-diiodo-l-methyl-1*H*-imidazole (**81**) to 4-iodoimidazole **191** by treating with EtMgBr, followed by water (Scheme 4.1) in 95% yield.[50] The resulting 4-iodoimidazole was reacted with EtMgBr to effect formation of imidazol-4-yl Grignard which was treated with piperonal [3,4-(methylenedioxy)benzaldehyde] to provide the alcohol **192** in 90% yield.[32] Removal of the doubly benzylic alcohol was accomplished by reducing with Et_3SiH and TFA at room temperature affording **193**. Lithiation at C2 was accomplished with *n*-BuLi and the resulting species was treated with TsN_3 to install an azide moiety **194** in 52% overall yield for two steps. The synthesis of

preclathridine A (**7b**) was completed in 5 steps in 48% overall yield by reducing azide **194** on catalytic hydrogenation. Finally, the procedure reported by Watson and co-workers with TMS-activated methyl parabanic acid was used to convert preclathridine A (**7b**) to clathridine A (**8b**) in 30% overall yield from six steps.[30] The two synthetic compounds displayed spectroscopic data generally consistent with the structures of the natural product. The ^{1}H NMR data were in excellent agreement, the ^{13}C NMR data were generally in good agreement but we noted small differences in the ^{13}C NMR spectrum to that reported in both the isolation papers and in synthetic material.[7,107,108] All three sets of reported data were acquired in different solvents, but there is apparently little solvent dependence on the chemical shifts, and so we do not believe that this is the cause of the discrepancies. Fortunately, our synthetic material provided a nicely crystalline product, which was subjected to X-ray crystallography, and this provided confirmation of the connectivity (Figure 4.1). We also observed, that in the crystalline state, **8b** exists as the N(8)H tautomer (Scheme 4.1).

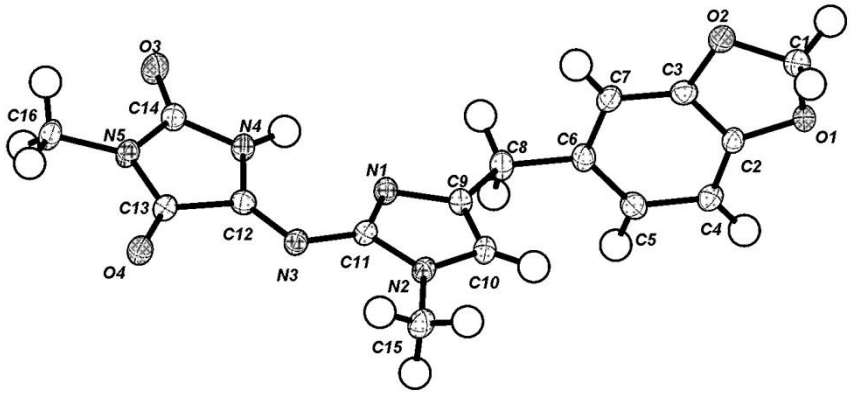
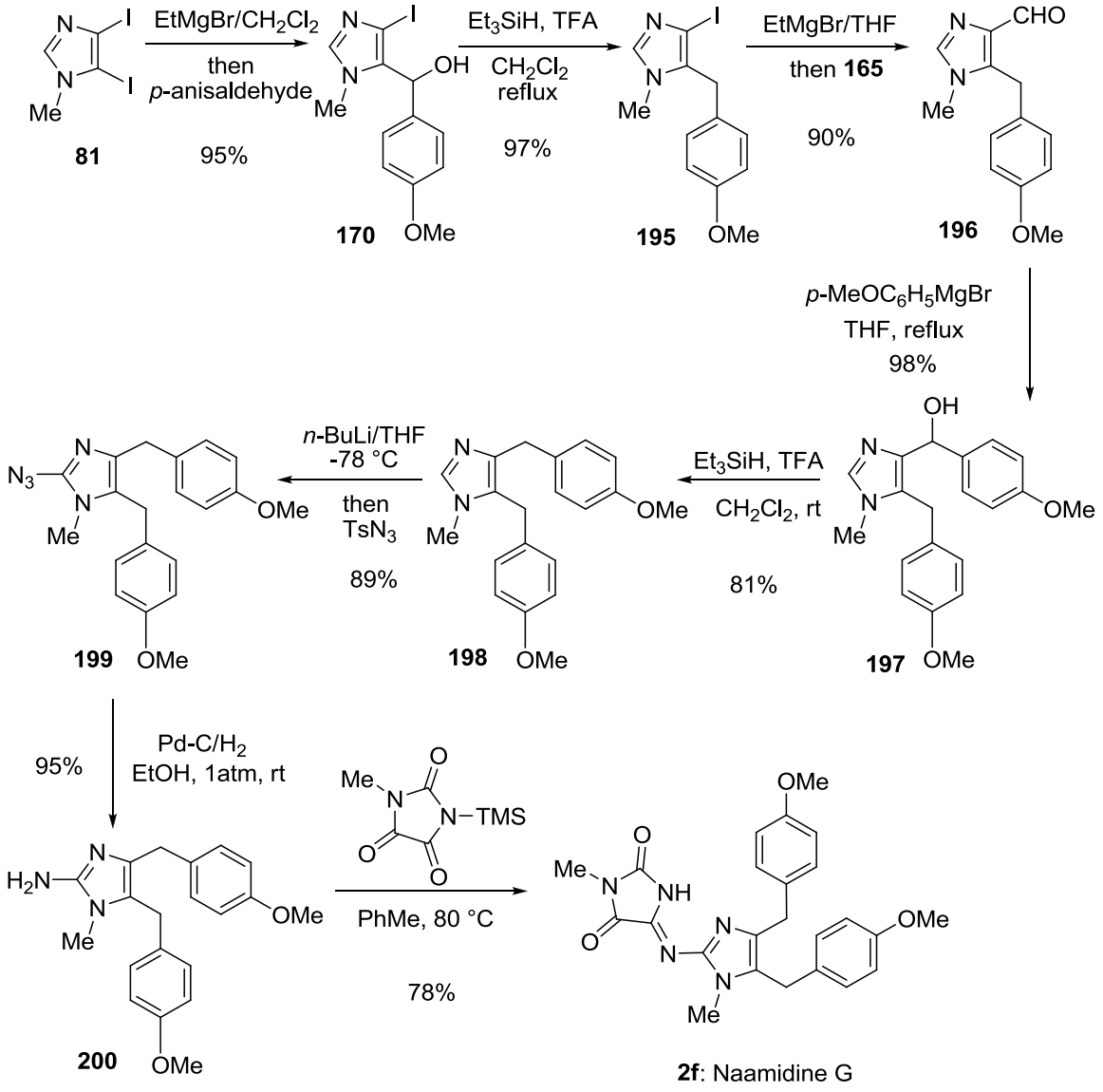


Figure 4.1: X-ray crystal structure of clathridine A (**8b**)

4.2 Total synthesis of naamidine G (**2f**) and 14-methoxynaamidine G (**4d**)



Scheme 4.2

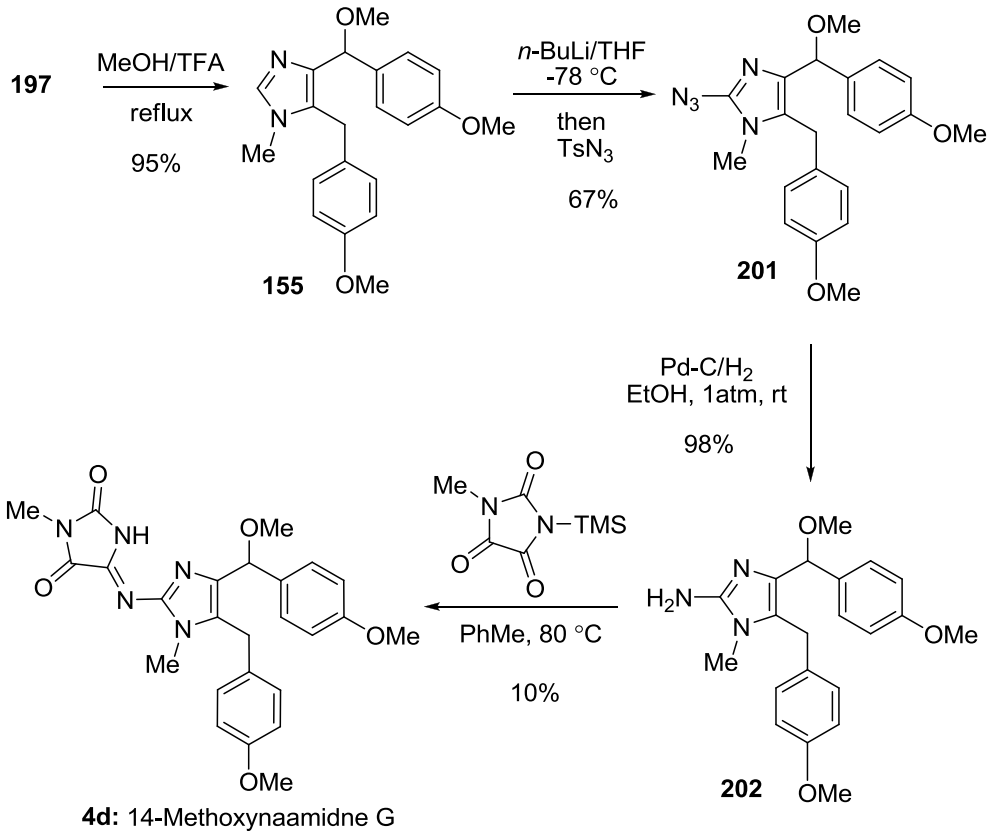
Our approach to these natural products was patterned after a similar sequence of reactions used as described in section 4.1 starting from 4,5-diiodoimidazole **81** (Scheme 4.2). Alcohol **170** was obtained as described earlier in Scheme 3.4. After the

ionic reduction of the benzylic hydroxyl group of **170** was performed with Et_3SiH and TFA, the resulting 4-iodoimidazole was reacted with EtMgBr to effect formation of the imidazol-4-yl Grignard reagent which was treated with *N*-methylformanilide (**165**) to provide the aldehyde **196**. Reaction of this aldehyde with *p*-$MeOC_6H_4MgBr$ provided the desired alcohol **197**, which is the key intermediate for naamidine G and 14-methoxynaamidine G (Scheme 4.2). Removal of the doubly benzylic alcohol was accomplished by treating with Et_3SiH and TFA at room temperature affording **198**. Lithiation at C2 was accomplished with *n*-BuLi and the resulting organolithium species was treated with TsN_3 to install an azide moiety, which was subjected to the catalytic hydrogenation to provide 1-methylnaamine D (**200**). This was then converted to naamidine G (**2f**) using the procedure used in the total synthesis of clathridine A (Scheme 4.1) to complete the total synthesis of **2f** in 41% overall yields over 8 steps.

The total synthesis of 14-methoxynaamidine G (**4d**) commenced with the conversion of alcohol **197** to methyl ether **155** with methanol and TFA (Scheme 4.3). Introduction of C2 azide was accomplished by metalation and reaction with TsN_3 provided **202**, which was converted to amine **203** by catalytic hydrogenation. Following the same reaction as above, TMS-activated methyl parabanic acid provided 14-methoxynaamidine G (**4d**) in low yield.

Initially it was assumed that the TMS source might cause the decomposition of the starting material. Therefore, the introduction of hydantoin moiety was attempted using other reported methods.[32, 37, 108] However, all methods failed to provide the product and only the decomposition of the starting material was observed; thereby unable to get improved yield. From this approach, we have developed eight-step

syntheses of the *Leucetta* alkaloids naamidine G (41% overall yield) and 14-methoxynaamidine G (5% overall yield).

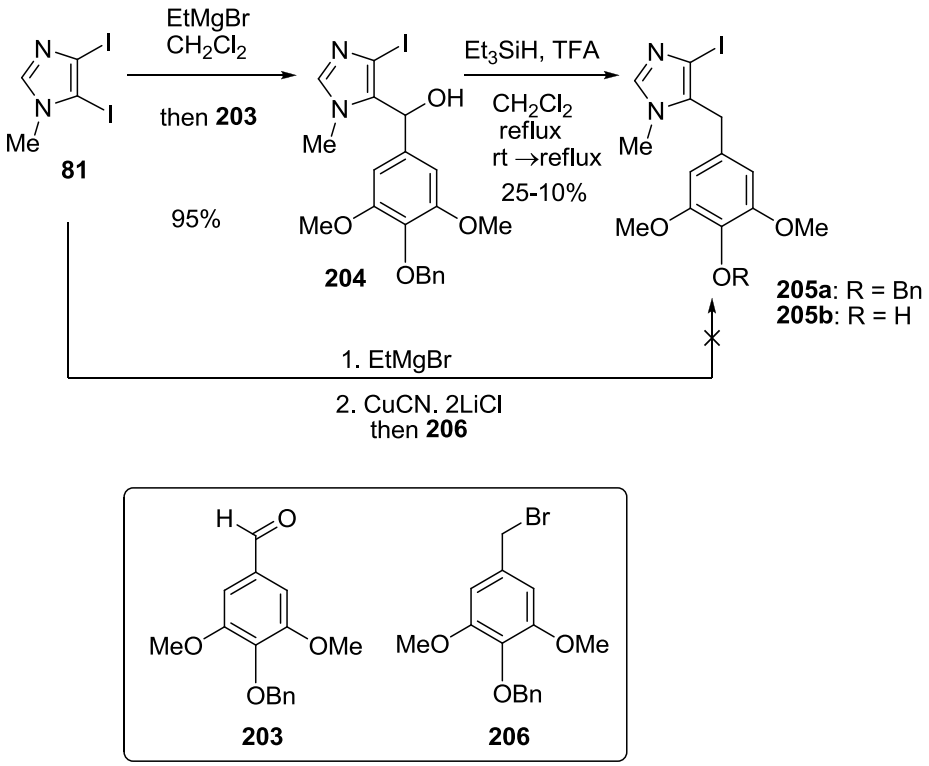


Scheme 4.3

The spectroscopic data of the synthetic compounds were in agreement with the data reported for the natural products. Interestingly, the melting point of the synthetic naamidine G was 195-196 ºC in contrast to the natural product (94 ºC).[12] Attempts to synthesize 14-hydroxynaamidine G (**4b**) starting from **197** were unsuccessful as the problem persists during the introduction of hydantoin moiety as discussed above.

4.3 Total synthesis of naamine G (**1h**) and naamidine H (**2g**)

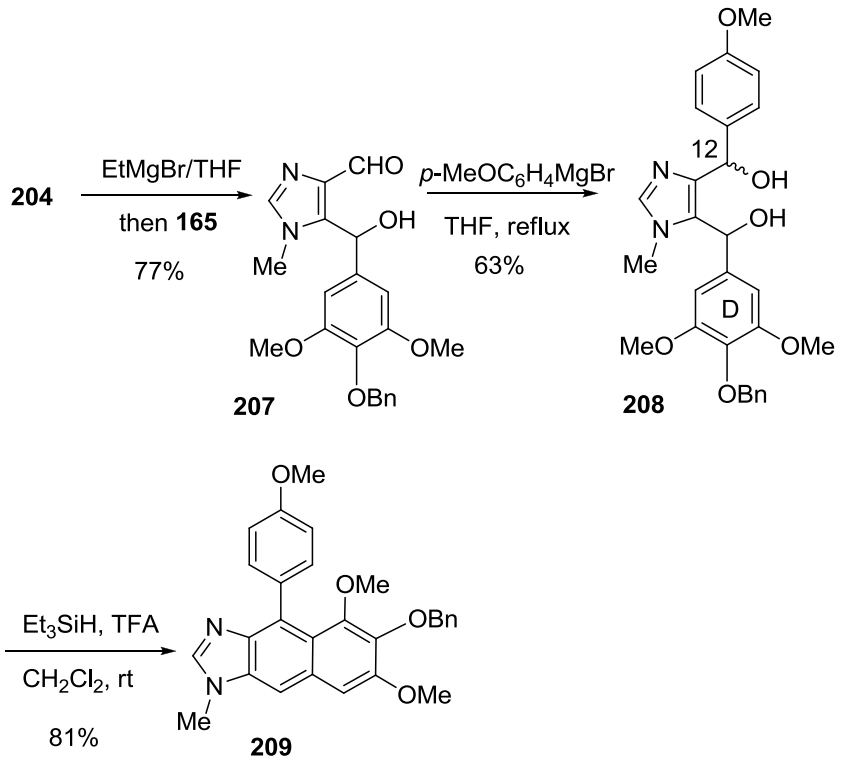
Again our synthetic endeavors begin by treating 4,5-diiodo-1-methyl-1*H*-imidazole **81** with EtMgBr followed by benzaldehyde **203**[109] to provide alcohol **204** in 95% yield (Scheme 4.4). Ionic reduction of this alcohol was attempted using well known conditions with Et_3SiH and TFA, conditions that we have used previously in total synthesis of calcaridine A and as discussed in sections 4.1 and 4.2. Surprisingly, this reaction did not work well at room temperature, nor did increasing the reaction temperature to reflux (CH_2Cl_2 or $CHCl_3$). We have also used $BF_3.OEt_2$ in combination with Et_3SiH to effect related reductions, but this tactic failed to provide the desired product. It has been observed in the course of related reactions that the solution typically becomes red or orange when the carbocation formed and it slowly disappears upon reacting with Et_3SiH. However, in this case the reaction turned only pale yellow, which presumably indicates the absence or little formation of carbocation from the alcohol. All of these attempts resulted in desired product **205a** and de-benzylated product **205b** in low but variable yields.



Scheme 4.4

Rather than spend time finding conditions to reduce the alcohol at this stage, it was decided to push the synthesis forward by introducing the C4 *p*-methoxybenzyl group. To accomplish this, alcohol **204** was formylated by treatment with 2 equivelent of EtMgBr and then 3 equivelent of *N*-methylformanilide (**165**) affording aldehyde **207** in 77% (Scheme 4.5). Subsequent treatment of **207** with excess *p*-$MeOC_6H_4MgBr$ produces the diol **208**, which was then subjected to ionic reduction with Et_3SiH/TFA. In contrast to alcohol **204**, the diol was readily soluble in CH_2Cl_2 and after the addition of TFA, the solution became highly colored. Analysis of the reaction mixture indicated complete consumption of diol **208**, but rather than reduction to provide **214** (Scheme 4.6), the naphtho[2,3-*d*]imidazole derivative **209** was formed. The formation of **209** can be readily understood in terms of an

intramolecular Friedel-Crafts cyclization of the electron rich aromatic D-ring onto the carbocation produced by ionization of the C12 alcohol and then dehydration. Similar cyclizations have been reported previously also under acidic conditions,[36] and this sequence can be used to synthesize kealiinine B, C and other related alkaloids (Scheme 1.6).

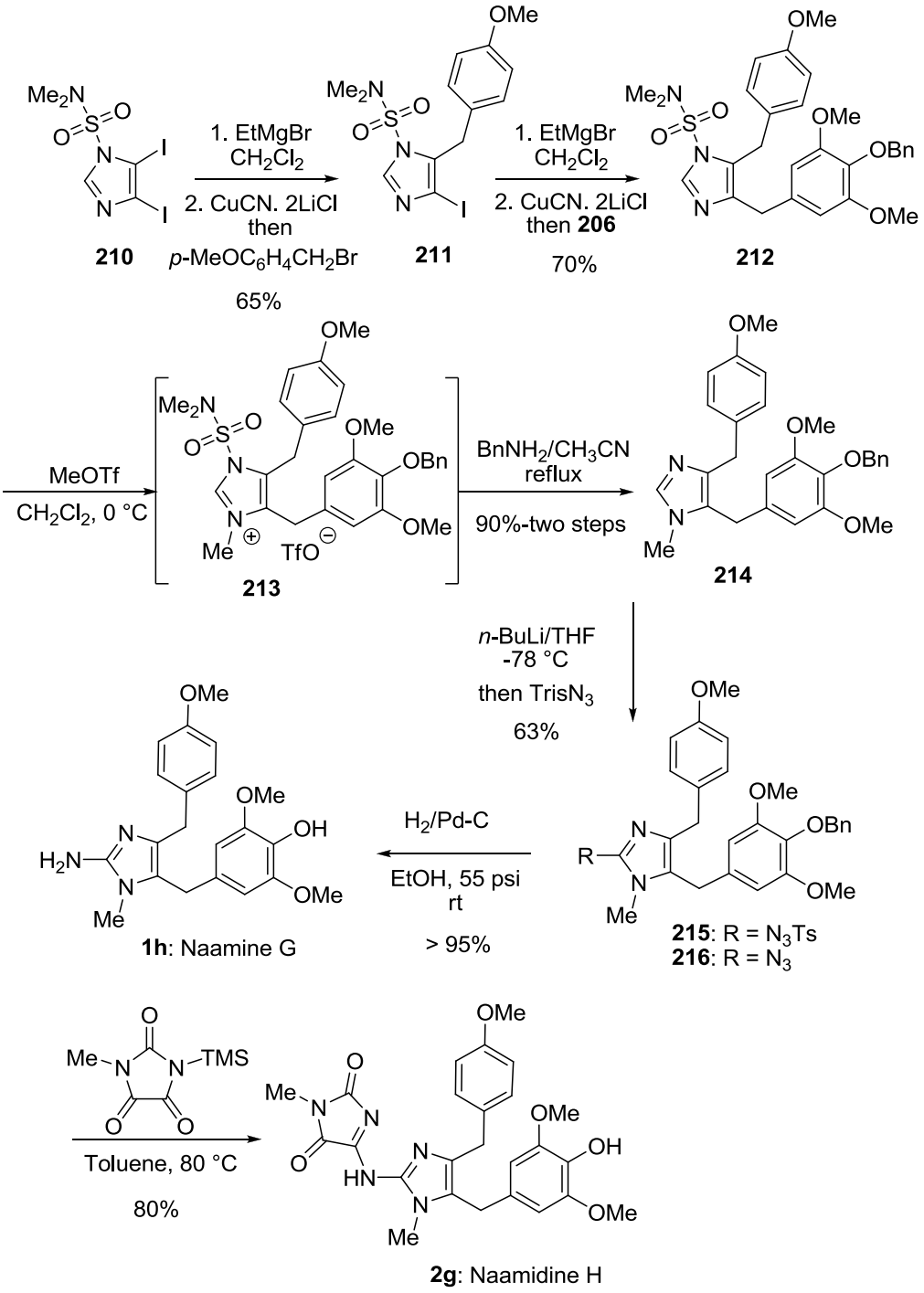


Scheme 4.5

Since all attempts *via* alcohol **204** failed to produce key intermediate **214** en route to naamidine H, the synthetic strategy required redesigning to avoid the intermediacy of benzylic alcohols related to **204**. Lindell and co-workers have reported that it is possible to directly alkylate imidazolyl cuprate derivatives with benzylic bromides and thus we pursued this strategy.[41] However, treating imidazole

81 with EtMgBr, then transmetalation with 1.0 M CuCN.2LiCl followed by 4-benzyloxy-3,5-dimethoxybenzyl bromide[109] (**206**, Scheme 4.4) did not provide an isolable product. We have observed previously that methyl substituted imidazoles do not appear to be good substrates in this reaction (Chapter 3, Section 3.1), and we noted in the Lindell reports that electron withdrawing protecting groups were employed, therefore we decided to switch protecting groups. The DMAS group at *N*1-position has been used in our lab for a variety of imidazoles and in a variety of chemistries, thus it was anticipated that this would be suitable in the present context.[110-112] An additional factor driving this choice was the recent report describing an efficient method for methylating DMAS-protected imidazoles with transposition, which we envisioned being useful for our synthesis.[113-115] Accordingly, imidazole **210** was treated with EtMgBr followed by freshly prepared 1.0 M solution of CuCN.2LiCl and *p*-methoxybenzyl bromide[116] to produce the iodoimidazole **211** in 65% yield (Scheme 4.6). Repetition of this sequence of reactions; EtMgBr, CuCN.2LiCl followed by benzyl bromide **206** was performed to produce the 4,5-dibenzylimidazole **212** in 70% yield. At this stage the DMAS-protecting group needed to be removed and replaced with an *N*-methyl group in a position specific manner. This could be easily achieved *via* formation of the methyl imidazolium salt **213**, which facilitates the removal of DMAS protection at *N*1-position. Following literature precedent, disubstituted imidazole **212** was treated with methyl triflate in dichloromethane to provide the imidazolium ion **213**,[114] the formation of which was observed by TLC and confirmed by ^{1}H NMR spectroscopy of the crude reaction mixture. After removing the solvent, first attempts made to remove the DMAS group with the help of aniline

failed.[113] Therefore, intermediate **213** was reacted with benzylamine in refluxing acetonitrile to afford the methylimidazole **214** in 90% overall yield.



Scheme 4.6

When **214** was treated with *n*-BuLi and TsN_3, tosyl derivative **215** was isolated as the only substitution product in addition to large amount of recovered starting material.[117] This was a somewhat surprising result since we and others have used this approach for the azidation of imidazole C2 position. Fortunately the preparation of azide **216** was achieved in 63% yield simply by treating imidazole **214** with *n*-BuLi and followed by trisyl azide at -78 ºC. Catalytic hydrogenation of this material converted the azide to the amine and removed the benzyl protecting group and provided of naamine G (**1h**) in 95% yield. Conversion of **1h** to naamidine H (**2g**) was accomplished using Watson's completing the total synthesis of naamidine H in 20% overall yields from 6 steps.

NMR spectroscopic data of the synthetic naamidine H matched with the natural product; however the melting point of the natural product has not been reported during the isolation and it was found that the synthetic material has a very sharp melting point of 204-205 ºC. While NMR data of the natural naamine G was reported in DMSO-D6, those of synthetic material was taken in Methanol-D3, as a result ^{13}C NMR signals of two product show little deviation, but not significant. We think this may be due to two different solvents used to take the NMR spectroscopic data.

4.4 Summary

We have developed high yielding five- and six-step syntheses of the *Leucetta* alkaloids preclathridine A (**7b**) and clathridine A (**8b**), respectively. The syntheses rely on chemoselective halogen–magnesium exchange, permitting the selective

installation of the C4-benzyl group without protection of the more acidic C2-and C5-positions. Subsequent lithiation at C2-position and electrophilic trapping of the disubstituted imidazole derivative lead to the introduction of 2-amino moiety in very good yield.

By extrapolating the above strategy, we have developed high yielding, eight-step sequence for the total synthesis of naamidine G (**2f**), and the application of the same sequence of reactions provided related alkaloid, 14-methoxynaamidine G (**4d**) in low yield due to the unsolved problem occurred during the introduction of hydantoin moiety.

Initial attempts to syntheses of naamine G (**1h**) and naamidine H (**2g**) using the same approach failed as a result of uncontrollable, but useful side reaction, which could lead to total synthesis of another *Leucetta* alkaloid family. As a result of this failure, we have developed high yielding, practical approach to above alkaloids using consecutive two cross coupling reactions of a DMAS-substituted diiodoimidazole derivative, which was later converted in to the required intermediate for the target molecules. In fact this is one of the advantages in our strategy.

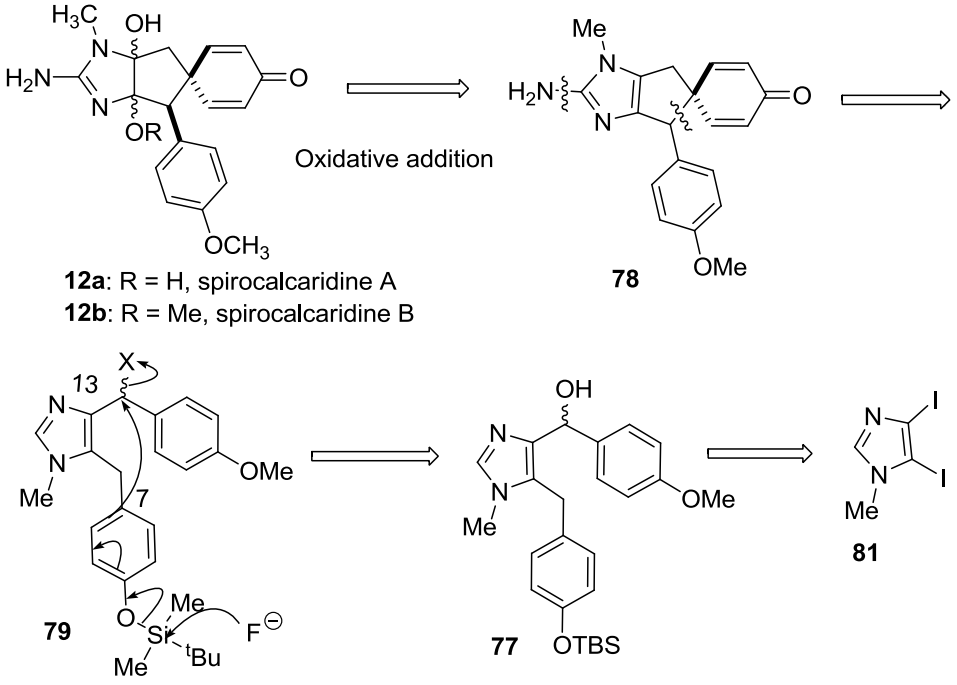
All the synthetic materials were tested for their biological activities and we have found interesting outcome form these tastings and they will be reported in due course.

This chemistry is very easy to perform and leads to the preparation of these natural products on the gram-scale. This chemistry can be used for the synthesis of other members of the *Leucetta* family of alkaloids, and it is also amenable for the synthesis of analogs for medicinal chemistry programs. We are actively exploring these areas.

CHAPTER 5

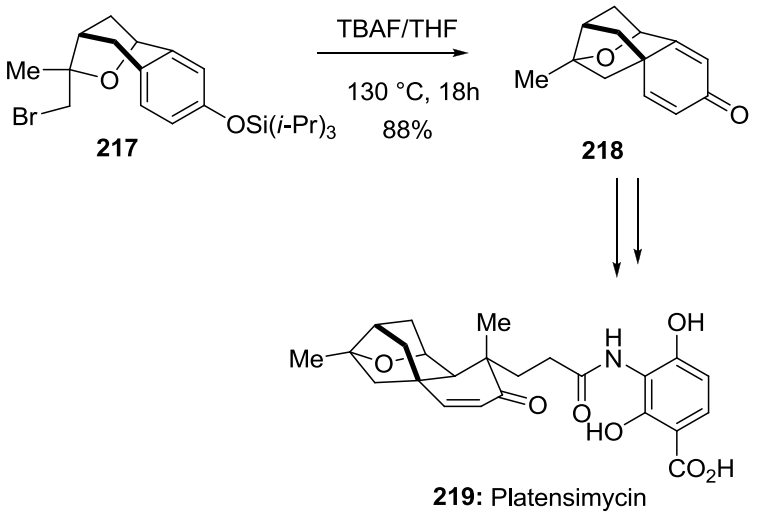
SYNTHETIC STUDIES TOWARDS SPIROCALCARDINE A AND SPIROCALCARIDINE B

As described in Chapter 1, our approach to spirocalcaridine A (**12a**) and spirocalcaridine B (**12b**) began with 4,5-diiodoimidazole **81** (Scheme 5.1). The crucial disconnection in this approach is the conversion of 4,5-disubstituted imidazole **79** to spiro-fused cyclopentaimidazole **78**. We proposed to accomplish this through the *ipso*-cyclization of the imidazole C4-benzylic group to form C7-C13 bond by treating **79** with an appropriate desilylating reagent based on the recent advances in this type of transformation.[118-120]



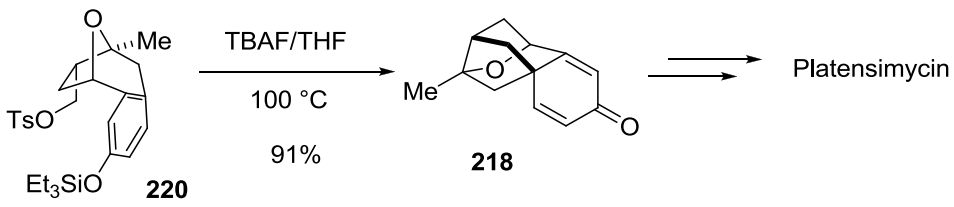
Scheme 5.1

During the synthesis of platensimycin (**219)**, a product of *Streptomyces platensis*,[121] Corey reported the enantioselective approach to tetracyclic α,β-dienone **218** *via* bromoether **217**, which was heated with 1.03 equivalent of TBAF in THF at 130 ºC for 4 h leading to the isolation of **218** (Scheme 5.2).[118]



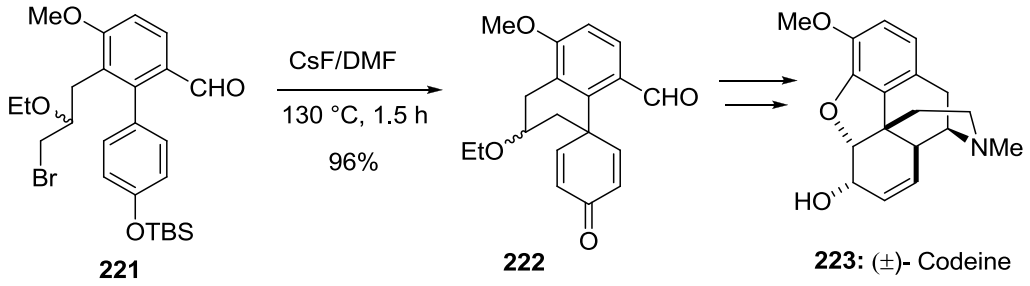
Scheme 5.2

A similar reaction was reported by Njardarson as an approach to the compact core of platensimycin as shown in Scheme 5.3.[119] In this transformation, he used the tosylate **220** instead of a bromide derivative to construct the tetracyclic core; otherwise both approaches are the same.



Scheme 5.3

During a recent total synthesis of (±)-codeine (**223**), Magnus and co-workers reported the *ipso*-cyclization of bromoether **221** (Scheme 5.4) using CsF in DMF to isolate the dienone **222** in an excellent yield.[120]



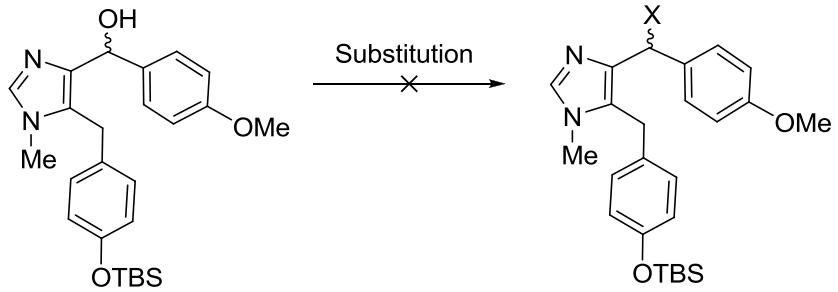
221 **222** **223:** (±)- Codeine

Scheme 5.4

5.1 First generation attempt *via* secondary alcohol **77**

Based on above examples we started our approach to spirocalcaridines A and B (**12a**, **b)** form alcohol **77**, which we attempted to convert to bromide **79a** as shown in Scheme 5.5, using the conditions shown Table 1. Although the use of PBr_3 is well known for converting alcohols to the corresponding bromide, **77** did not provide the anticipated bromide, as the starting material was recovered under the conditions shown in entries 1 and 2.[122] Next, this alcohol was treated with HBr under refluxing conditions; however, neither product nor starting material was isolated from this reaction (Entry 3).[123, 124] Since the bromination attempts with CBr_4/PPh_3 (Entry 4)[125] and NBS/PPh_3 (Entry 5)[126] also failed, it was thought that some of these conditions might simply form the imidazolium salt by protonating the imidazole *N*3-position, since most of these conditions produce HBr as a byproduct, thereby it is removed with aqueous layer during the workup. Attempts to convert this alcohol into mesylate

derivative **79b** also failed although the HCl produced from this condition is neutralized by the added amine (Entry 6 and 7).[127] Finally, when the alcohol **77** was treated with 4-nitrophenylsulfonyl chloride and DIEA, only 10% of the desired product **79c** was isolated and this was not sufficient to continue the sequence (Entry 8).



Scheme 5.5

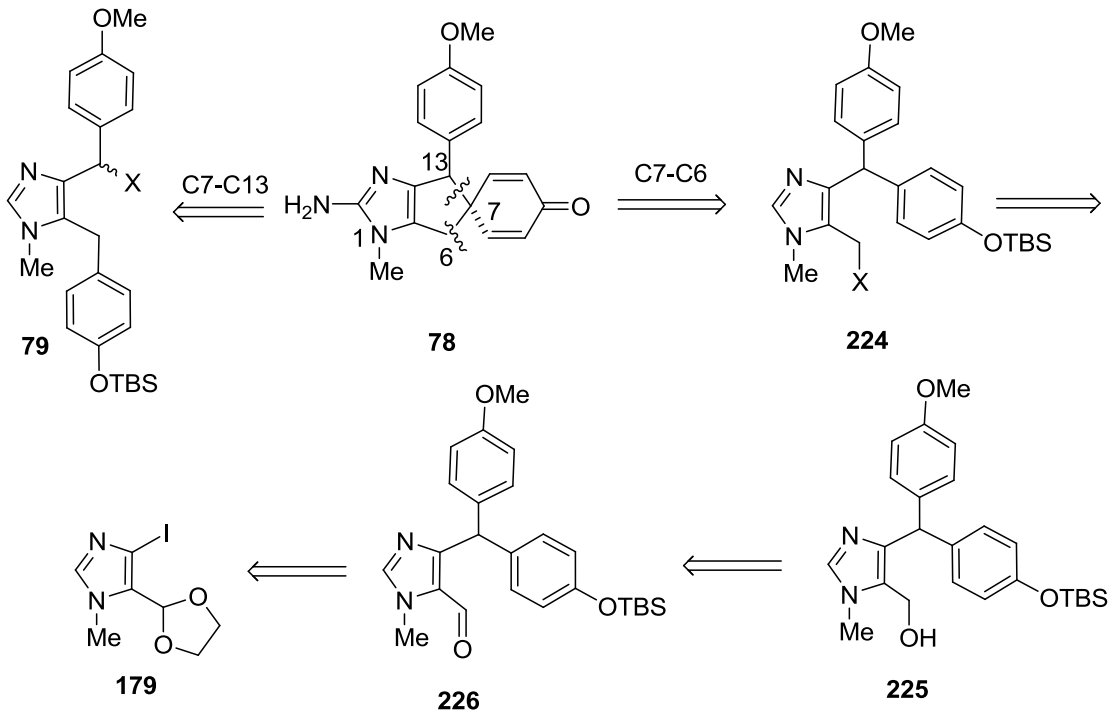
Table 5.1: Reaction conditions for the substitution reactions of **77**

Entry	Substitution-conditions	X	Product	Yield/%
1	PBr_3/CH_2Cl_2, 0 ºC	Br	**79a**	-
2	PBr_3, Py/Et_2O, -5 ºC	Br	**79a**	-
3	HBr/Reflux	Br	**79a**	-
4	PPh_3, CBr_4/CH_2Cl_2, 0 ºC	Br	**79a**	-
5	NBS, PPh_3/CH_2Cl_2, 0 ºC	Br	**79a**	-
6	MsCl, Et_3N/THF, 0 ºC	OMs	**79b**	-
7	MsCl, Et_3N/CH_2Cl_2, 0 ºC	OMs	**79b**	-
8	4-$(NO_2)C_6H_4SO_2Cl$, DIEA/CH_2Cl_2, rt	NPS	**79c**	~10

NPS: 4-Nitrophenylsulfonyl

5.2 Second generation approach *via* primary alcohol **225**

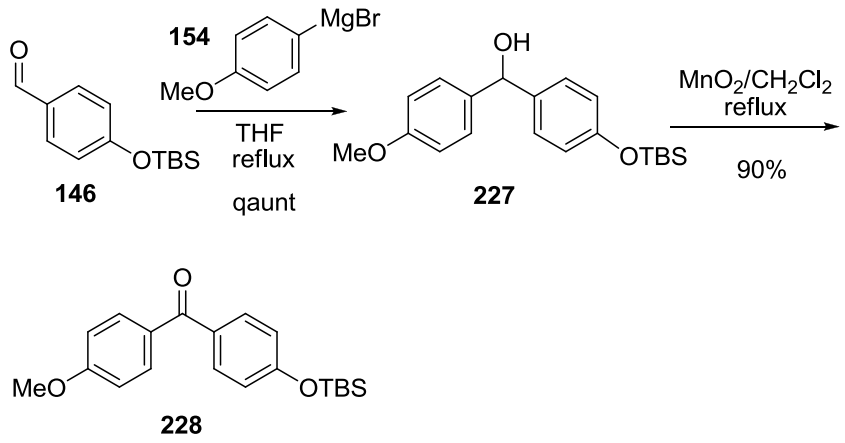
As above attempts failed to provide the required substrate **79** to continue the sequence *via* the secondary alcohol **77,** it was decided to attempt these transformations with primary alcohol **225**, which is less crowded than **77**, thus it might easily undergo nucleophilic substitution (Scheme 5.6).



Scheme 5.6

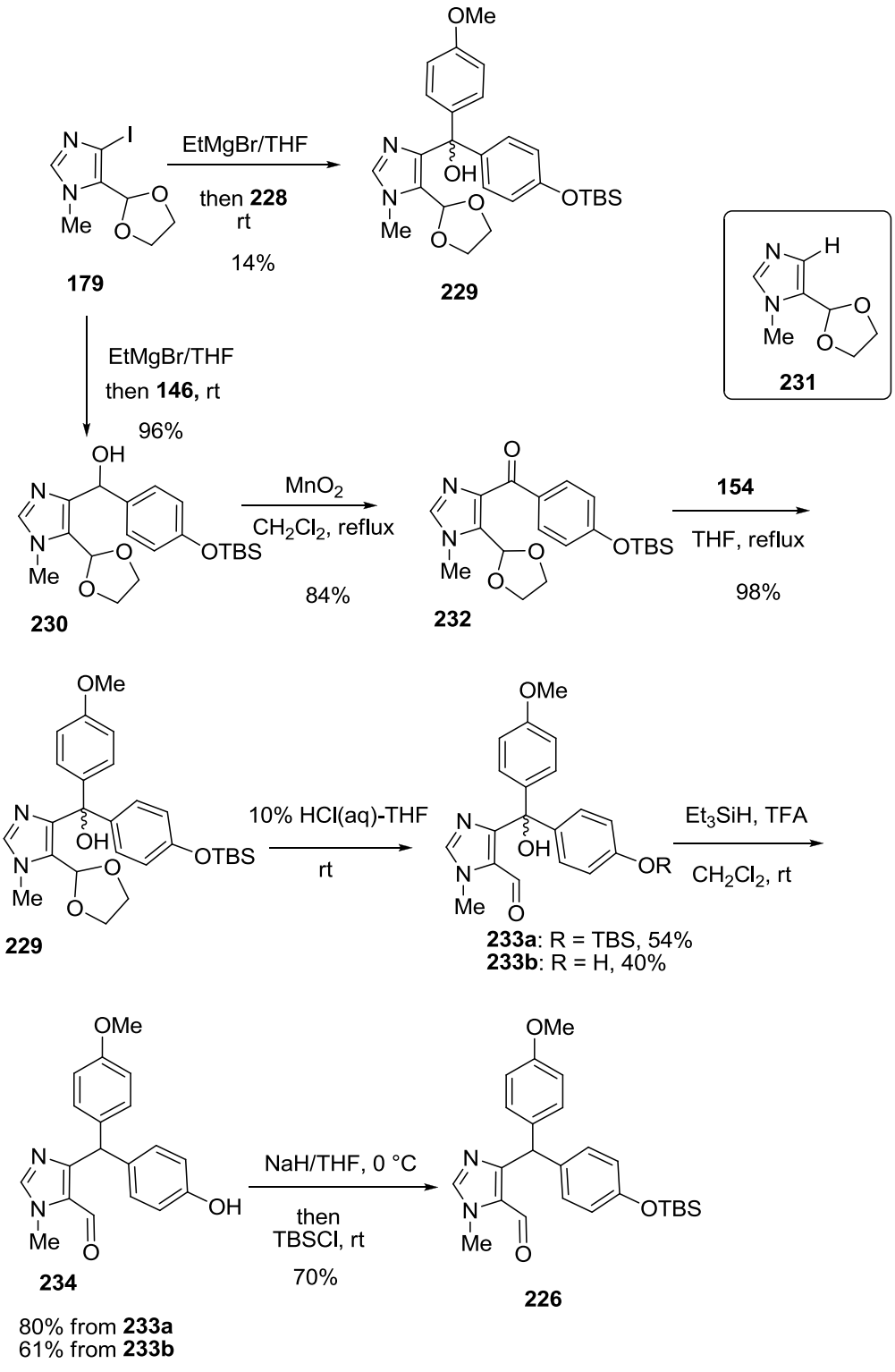
As was described above, most of the examples described in literature involved the participation of a primary alkyl halide or sulfonate.[119, 120] During the above attempts, intermediate **78** was disconnected through the C7-C13 bond to provide **79**. Alternatively, it is possible to disconnect **78** through C7-C6 bond to provide intermediate **224**, where X is a suitable leaving group. This is accessible from the

alcohol **225**, which will be obtained by reducing aldehyde **226**. Conversion of acetal **179** to aldehyde **226** can be accomplished with EtMgBr and the corresponding benzophenone derivative **228** (Scheme 5.7). In other words, such an intermediate should be accessible through chemistry already established in our lab.



Scheme 5.7

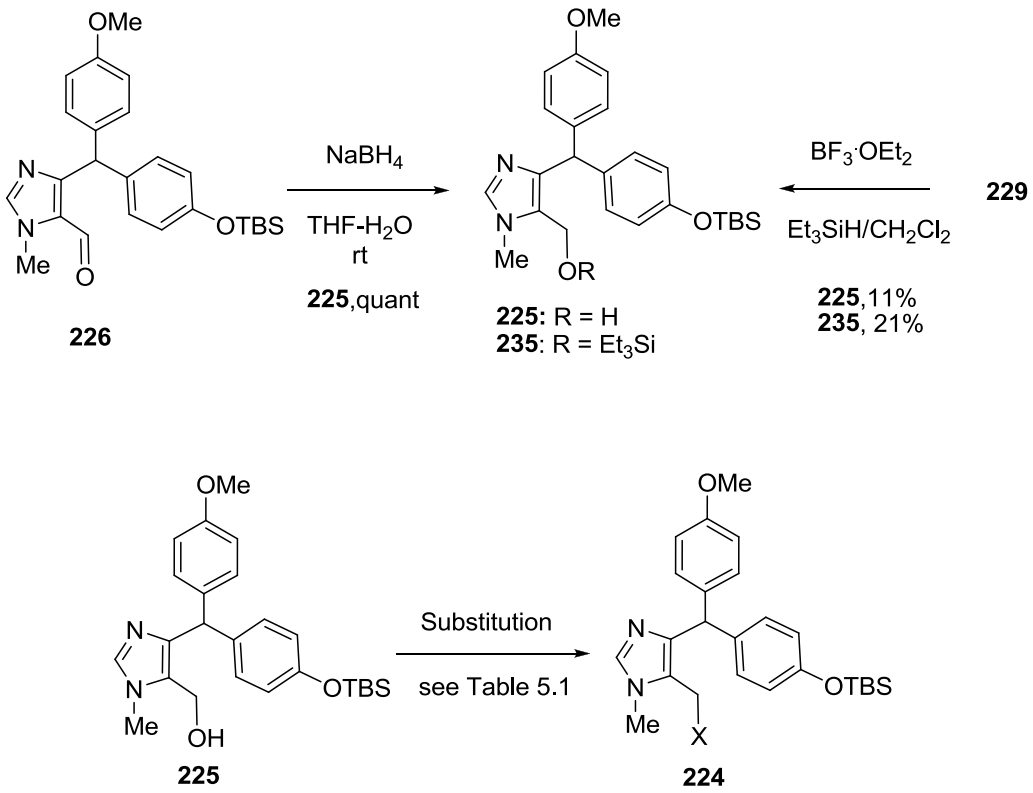
In order to commence this synthetic approach, ketone **228** was synthesized as shown in Scheme 5.7 *via* alcohol **227** in 90% overall yield in three steps. Then, acetal **179** was treated with EtMgBr and ketone **228** to isolate alcohol **229** only in 14% yield (Scheme 5.8). We anticipated that, this might be due to the steric clash between the Grignard species generated from **179**, and the diaryl ketone **228**. In addition, benzophenones of this type would be expected to be less reactive due to electronic considerations. Therefore, this alcohol was prepared in two steps; first by treating acetal **179** with EtMgBr and aldehyde **146** to isolate alcohol **230** in combination with de-halogenated product **231**. Attempted separation of these two compounds was unsuccessful, since they have almost identical R_f values.


OMe
EtMgBr/THF
then 228
rt
14%
OH
OTBS
179
229
231
EtMgBr/THF
then 146, rt
96%
MnO2
CH2Cl2, reflux
84%
154
THF, reflux
98%
230
232
10% HCl(aq)-THF
rt
Et3SiH, TFA
CH2Cl2, rt
233a: R = TBS, 54%
233b: R = H, 40%
NaH/THF, 0 °C
then
TBSCl, rt
70%
234
226
80% from 233a
61% from 233b

Scheme 5.8

Therefore, rather than spending time to purify at this stage, it was carried through to the next step, which will change the polarity of alcohol **230**, and this might ease the separation of ketone **232** form **231**. Accordingly, alcohol **230** was oxidized with MnO_2 to provide ketone **232**, which was treated with Grignard reagent **154** to synthesize alcohol **229** in 98% yield.

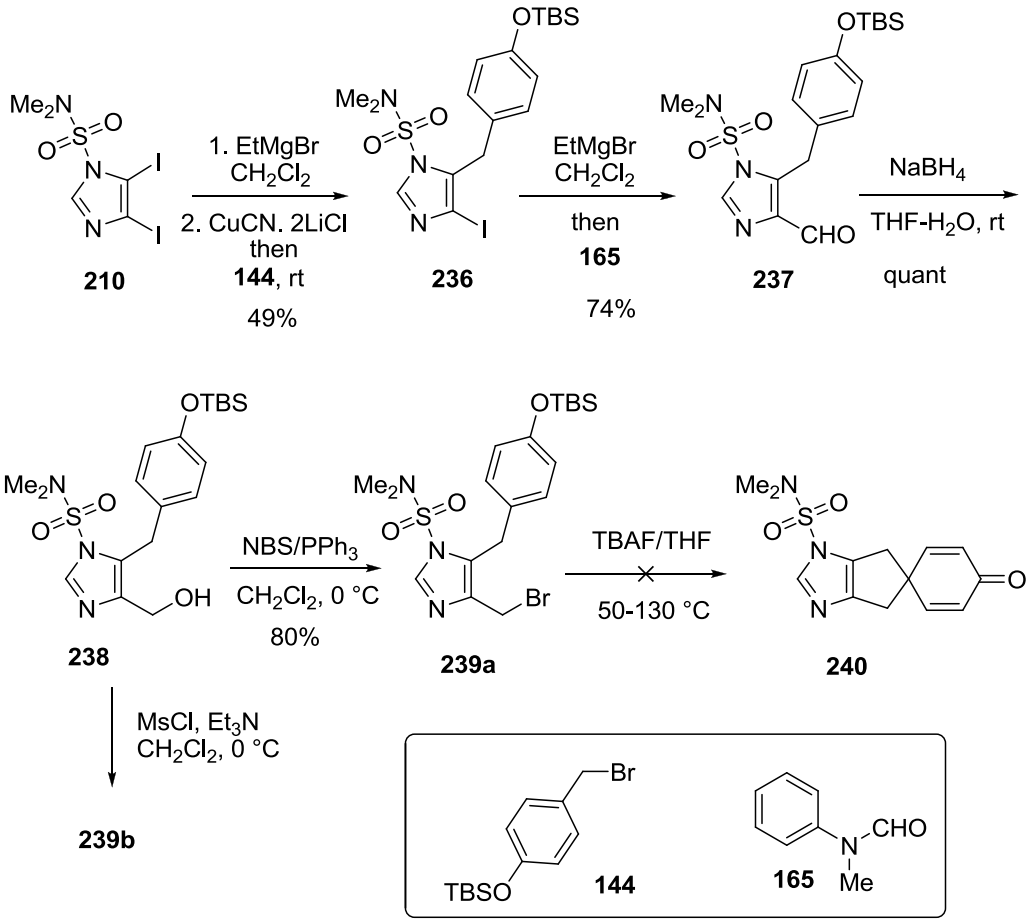
Removal of the tertiary hydroxyl group from **229** was first attempted with $BF_3{\cdot}OEt_2$ and Et_3SiH, and it was found to provide a mixture of **225** and **235** in low yield (Scheme 5.9). Therefore, this alcohol was treated with 10% aq. HCl solution in THF to remove the acetal protection, and this led to removal both acetal and TBS moieties of the aromatic ring providing **233a** and **233b**. After separating these two compounds, **233a** was subjected to ionic reduction to remove the tertiary hydroxyl group; however, it was also found the subsequent removal of TBS protection from this alcohol providing **234**. Accordingly, **233b** was also subjected to the ionic reduction to isolate **234** in 61% yield. Since the substrate **226** required TBS-substituted phenol for the cyclization attempt (Scheme 5.6, **224→78**), **234** was treated with NaH and TBSCl to provide the aldehyde **226** in 67% yield (Scheme 5.8). Reduction of this aldehyde with $NaBH_4$ in 1:30 mixture of H_2O:THF provided alcohol **225** in 5 minutes (Scheme 5.9). Attempts to subject this alcohol to substitution, under the conditions shown in Table 1 failed, which suggest that imidazole alcohols **77** and **225** are either susceptible to the formation of imidazolium salts under some of above conditions or they do not react with the nuleophiles used. It is considerable that the formation of imidazolium salts under those conditions takes place due to the presence of an electron donating substitution at imidazole *N*1-position, which increases the nucleophilicity of imidazole *N*3-position.



Scheme 5.9

In fact, we have found earlier that *N*1-methyl substituted imidazoles tend to form imidazolium salt with benzyl bromide and MeI even at rt (Schemes 3.1, 3.2 and 3.5). In principle, this could be prevented by reducing the electron density of the imidazole with electron withdrawing substitution at *N*1-position, which would then be exchanged with methyl group later in the synthesis (see section 4.3). Before commencing this approach with real substrate, we investigated the workability of this hypothesis with DMAS-protected imidazole **210** as shown in Scheme 5.10.

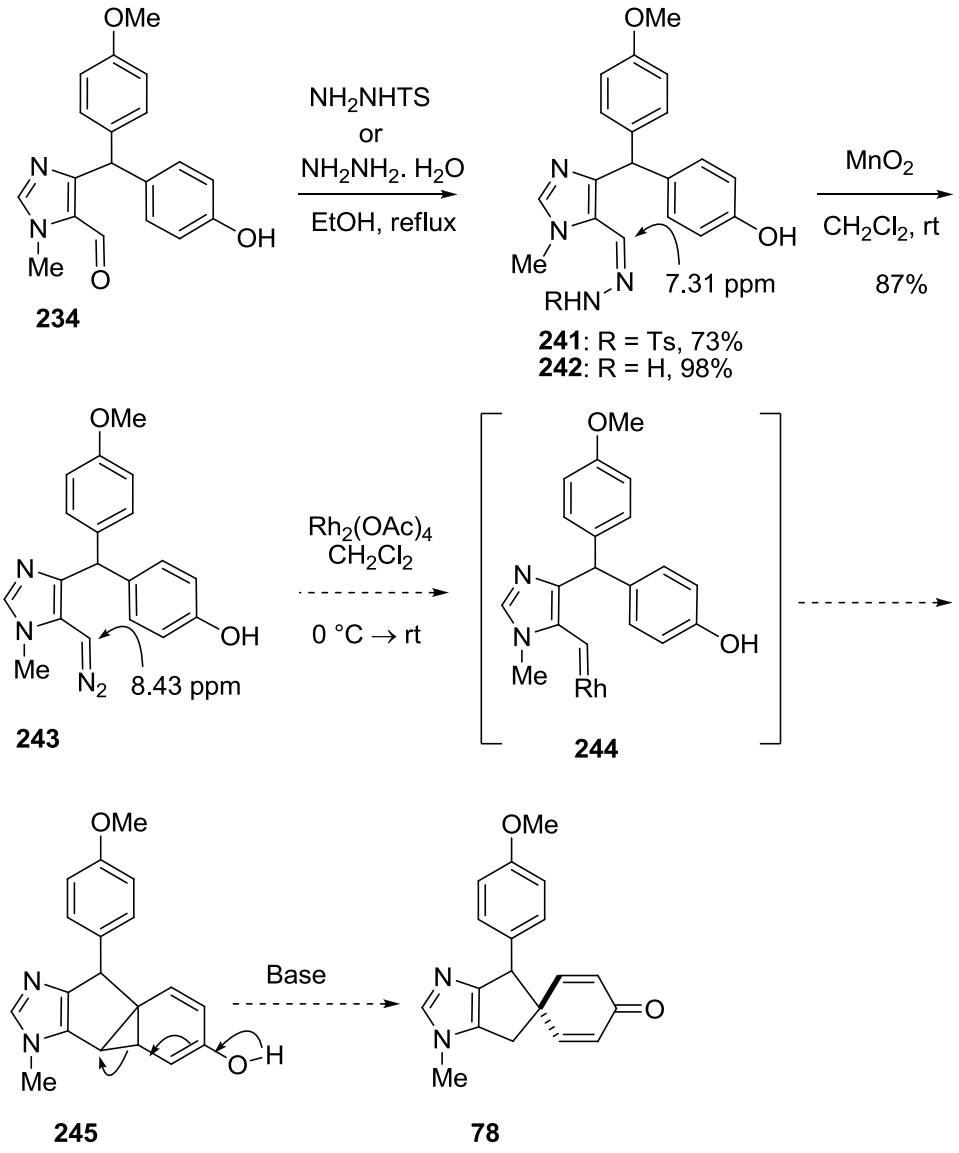
Accordingly, the treatment of imidazole **210** with EtMgBr, CuCN.2LiCl followed TBS-protected benzyl bromide **144** provided the mono-substituted imidazole **236** in 49% yield. Second Grignard exchange, followed by the addition of *N*-methylformanilide (**165**) provided 74% of the desired aldehyde **237**.



Scheme 5.10

Then, **237** was reduced to alcohol **238** with $NaBH_4$ in THF-H_2O mixture and this alcohol was treated with NBS and PPh_3 to synthesize bromide **239a** in very good yield. We have found earlier that 4-substituted benzyl bromides are not stable for an extended period of time; therefore this bromide was subjected to *ipso*-cyclization without further delay, under the conditions described by Corey (Scheme 5.2). Unfortunately, this bromide was found to decompose under these conditions and attempted cyclization as low as at 50 ºC also failed due to the decomposition of the bromide. An attempt to synthesize mesyl ester from the alcohol **238**, was not successful as this led to the formation of an unknown product **239b** (Scheme 5.10).

5.3 Third generation approach *via* intramolecular Büchner reaction



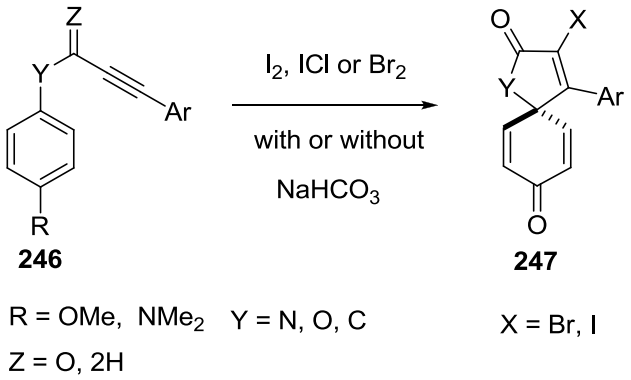
Scheme 5.11

Since all the attempts to synthesize the cyclopenta[d]imidazole **78** failed *via* the *ipso*-cyclization of **79** and **225**, we recognized the possibility of constructing this intermediate using an intramolecular Büchner reaction.[128, 129] Towards this end, aldehyde **234** was converted into the tosylhydrazone **241** with tosylhydrazide in

ethanol (Scheme 5.10).[130] Treatment of this hydrazone, either with Et_3N in methanol[131] or with KOH in methanol[132] failed to provide the diazo-imidazole **243** as reported. Thus we moved to the preparation of **242** by treating aldehyde **234** with hydrazone hydrate in ethanol.[133] Stirring of this hydrazone in dichloromethane with MnO_2[133] provided a reddish solid which we ancicipated as the diazo compound **243**. Although the physical appearance of this material was consistence with literature,[134] the azo-methane proton appears at 8.44 ppm in in the ^{1}H NMR spectrum of **243** in contrast to the corresponding proton in **242**, which appears at 7.31 ppm, and there were differences in the signals of ^{13}C NMR spectra of two compounds. This was somewhat doubtfull, and usually azo-methane proton has a lower chemical shift than starting hydrazone.[131] We were unable to confirm this structure since this material was not stable for further analysis and the attempted intramolecular Büchner reaction did not provide the anticipated product **245**, although there was a slight color change during the reaction.[128, 129] The ^{1}H-NMR spectrum of the crude product resembled the starting material, **243**. At this moment we were unable to confirm whether the Rh-carbenoid **244** is formed from this reaction. Repeated attempts also failed to provide any evidence regarding this conversion.

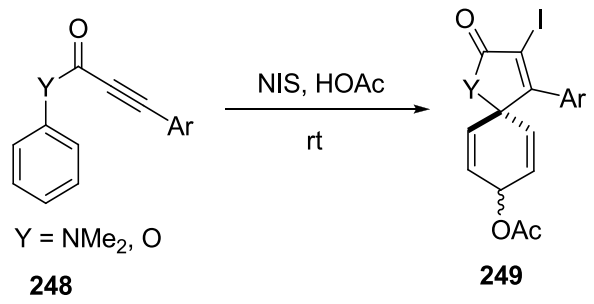
5.4 Fourth generation approach using *ipso*-halocyclization of 4-aryl-1-alkynes

When we were searching in the literature precedence for alternate route to **78**, we noted that Larock and co-workers have reported the intramolecular *ipso*-halocyclization of para-substituted aryl-alkynes **246** to spiro[4,5]trienone **247** under mild conditions (Scheme 5.12).[135] This novel approach caught our attention since this chemistry leads to the formation of a framework coupled with similar ring system we are interested, mild reaction conditions and easy workup make this approach worth further exploration.

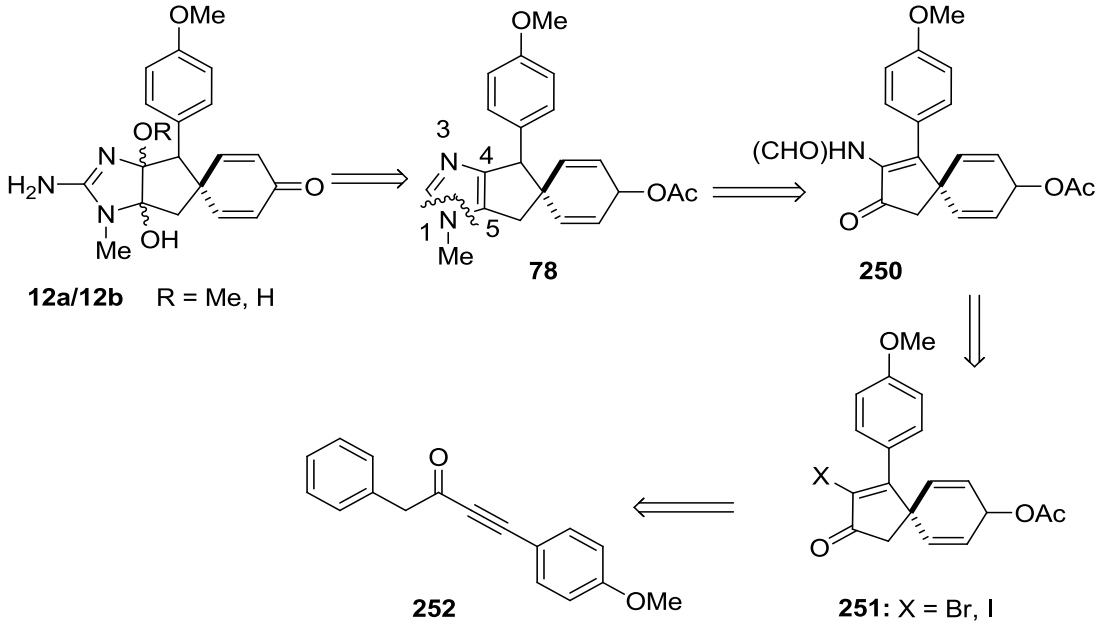


Scheme 5.12

After this report, Jin-Heng and co-workers reported the similar *ipso*-cyclization of unsubstituted arylalkynes **248** to synthesize spiro[4,5]trienyl acetates **249** with NIS in acetic acid (Scheme 5.13).[136]



Scheme 5.13

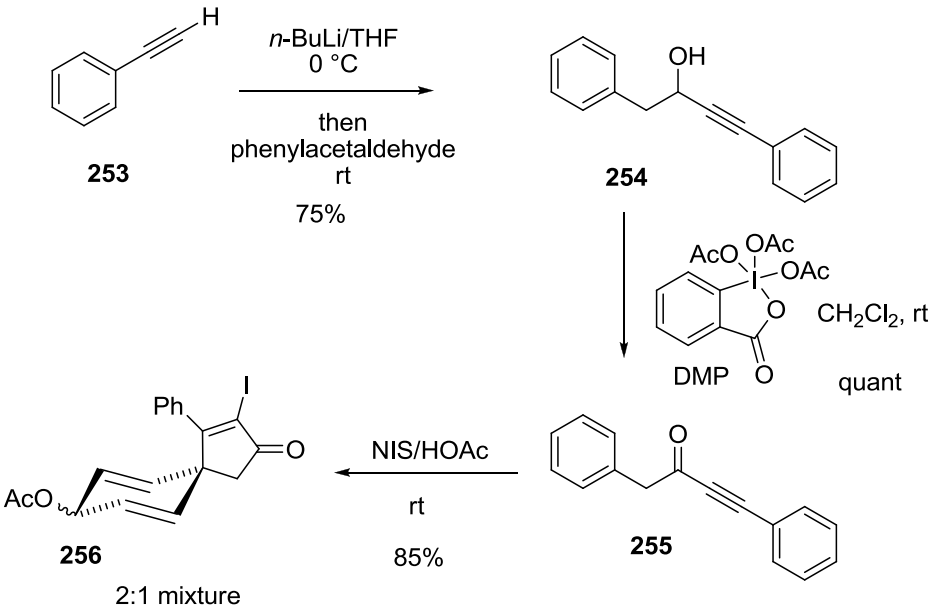


Scheme 5.14

If a reasonable disconnection approach can be developed *via* this type of intermediate, it would necessitate the *de novo* synthesis of the imidazole ring to provide the spiro-imidazole **78**. Regarding this idea we came up with a series of disconnections approach as shown in Scheme 5.14, which lead to the known alkynone **252**. Spiro[4,5]trienyl acetates **251** would be the ideal intermediate compared to spiro[4,5]triennone **247**, since it has only one carbonyl group allowing us to construct

the imidazole ring of **78** at a late stage *via* corresponding methylimine of **250**. A cross-coupling reaction with **251** will be used to introduce the formamide unit found in **250** using the Ullmann-type reaction.[137]

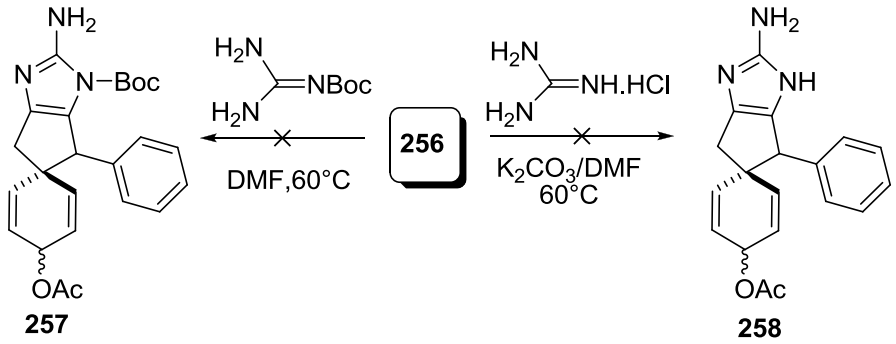
In order to understand the feasibility of formation of the imidazole ring, a model substrate was prepared by treating phenyl acetylene with *n*-BuLi at 0 °C, followed by the addition of phenyl acetaldehyde ($PhCH_2CHO$) to form alkynol **254**, in 75% (Scheme 5.15).[138] This alcohol was oxidized with DMP to produce alkynone **255** quantitatively.[135] Subsequent treatment of **255** with NIS in acetic acid, at room temperature, provided **256** as a 2:1 mixture of diastereomers.[136]



Scheme 5.15

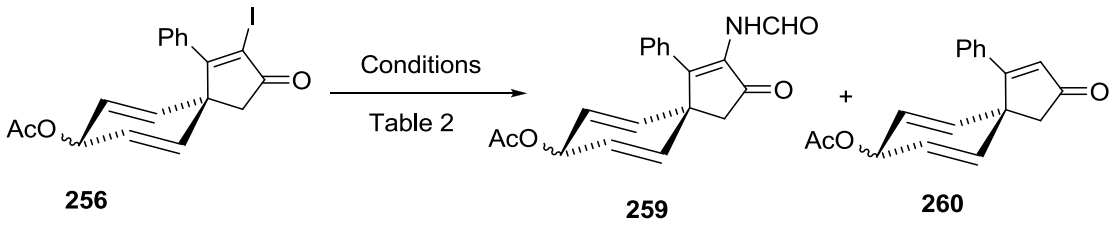
Before we carried out the next step, the cross coupling reaction, **256** was treated with two different guanidine derivatives to establish whether the direct formation of imidazole ring might be feasible. It was not surprising however, to

recover the starting material from the two reactions (Scheme 5.16),[139-141] since sp^2-hybrid centers bearing halogens have not been previously used to construct imidazole ring under these conditions.



Scheme 5.16

Next, we focused on an Ullmann-type cross coupling reaction of **256** to replace the iodide with formamide (Scheme 5.17). Toward this end, we performed several screening reactions as shown in Table 2, by changing the ligand, solvent, base, catalyst and the temperature of the reaction.[137, 142-145]



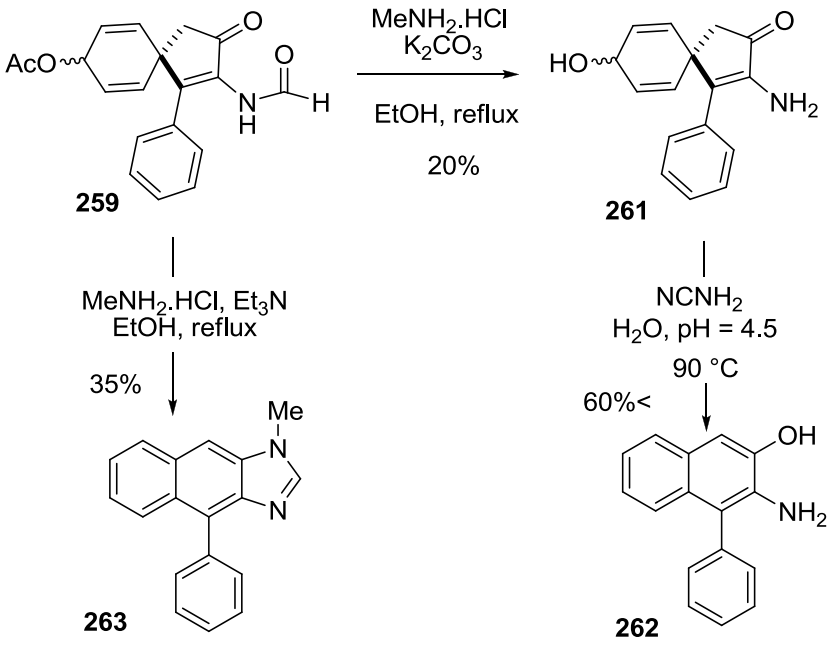
Scheme 5.17

Table 5.2: Optimization of cross-coupling reaction of **256**

Entry	Solvent	Ligand[a]	Base[b]	Catalyst	Amide	Tem/°C	Time/h	%-259	%-260
1	Isopropanol	2 eq L2	2 eq B3	5% CuI	Formamide	80	12	NR[c]	-[d]
2	1,4-dioxane	10% L1	2 eq B1	5% CuI	Formamide	110	28	NR	-
3	Toluene	10% L1	1.5 eq B1	5% CuI	Formamide	80	24	NR	-
4	THF	10% L1	1.5 eq B1	5% CuI	Formamide	70	22	50	25
5	DMF	10% L1	1.5 eq B1	5% CuI	Formamide	70	30	10	-
6	CH_3CN	10% L1	1.5 eq B1	5% CuI	Formamide	70	30	45	40
7	THF	1 eq L1	1.5 eq B1	1 eq CuI	Formamide	25	15	-	10
8	THF	2 eq L1	2 eq B1	1 eq CuI	Formamide	80	48	52	30
9	THF	2 eq L1	2 eq B1	1 eq CuI	Formamide	40	10	21	20
10	THF	10% L3	2 eq B2	5% CuI	Formamide	25	12	-	-
11	THF	10% L1	2 eq B2	5% CuI	Formamide	70	15	40	30
12	THF	10% L1	2 eq B1	5% CuBr	Formamide	70	15	50	30
13	THF	10% L1	2 eq B1	5% CuI	Formamide	70	15	30	-
14	THF	10% L3	2 eq B1	5% CuCN	Formamide	70	15	-	-
15	THF	10% L1	2 eq B1	5% CuI	Urea	70	15	-	30
16	THF	10% L1	2 eq B1	5% CuI	*N*-Methylurea	70	15	-	30

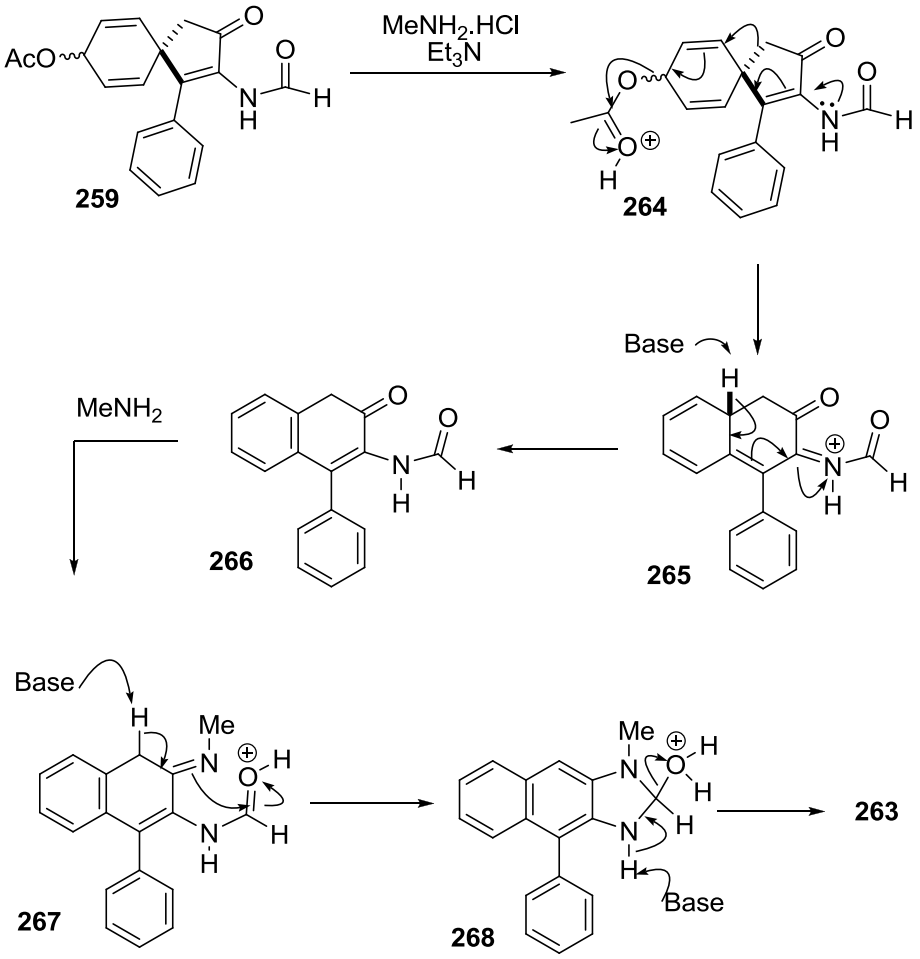
[a] L1 = DMEDA, L2 = Ethyleneglycole, L3 = TMEDA
[b] B1 = Cs_2CO_3, B2 = K_2CO_3, B3 = K_3PO_4
[c] NR = No reaction or the starting material was recovered
[d] - = Not isolated

No cross-coupling reaction was observed in isopropanol, 1,4-dioxane and toluene, as shown in entries 1, 2 and 3. When the solvent was changed to THF, this reaction was found to work smoothly providing amide **259** (Entry 4). In addition, this reaction was found to work DMF and CH_3CN; forming the required product in relatively low yields (Entries 5 and 6). As a result THF was used as the solvent in subsequent reactions. Among the ligands tested, DMEDA (L1) was found to be the only ligand to facilitate the cross coupling (Entry 1, 9 and 10). Although, three possible bases were explored, Cs_2CO_3 found to be superior to the others, and 1.5 equivalent of the base was found to be enough for the reaction as the yield did not change by changing the amount of the base added (Entry 4, 8 and 12). CuI and CuBr were found to be equally efficient at catalyzing this cross-coupling reaction (Entry 4 and 12), although we used CuI as the catalyst in our synthesis, and CuBr was not tested thoroughly during this study. Despite the fact that urea derivatives have been used in the cross coupling reactions,[146, 147] they did not seems to work with **256** under the optimum conditions (Entry 15, 16) as only reductive dehalogenation was observed from these reactions providing **260**.



Scheme 5.18

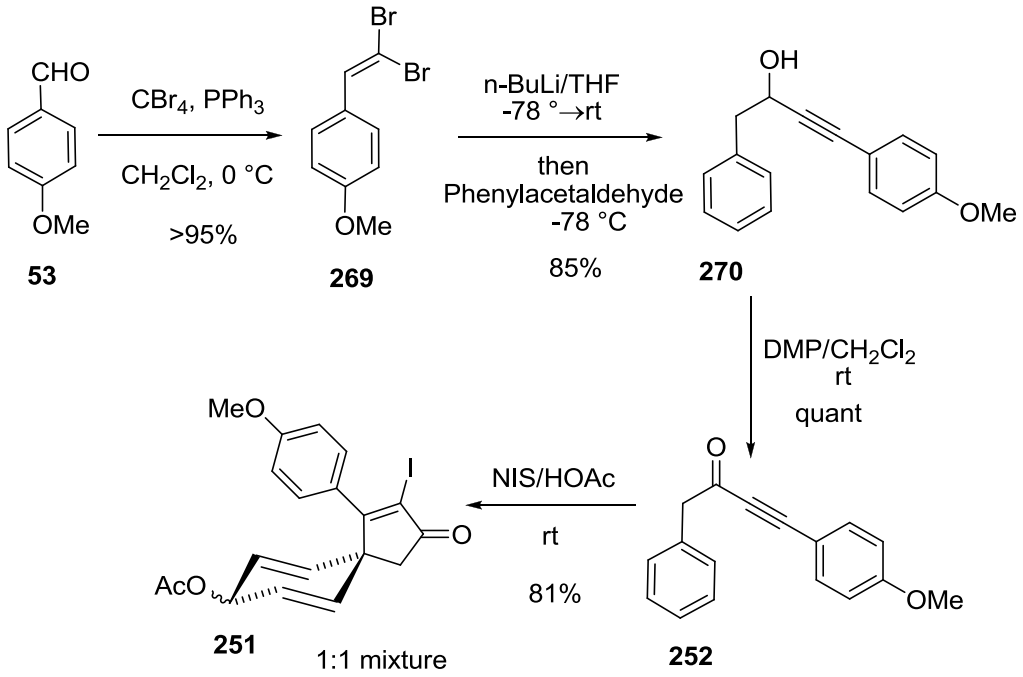
Since the cross coupling reactions did not work with urea derivatives, the keto-amide **259** was treated with methylamine hydrochloride and K_2CO_3 in refluxing ethanol in the expectation forming the imidazole (Scheme 5.18). However, these conditions provided **261**, resulting in the removal of the acetyl and formyl groups of the starting material. Although this was not the expected product, in principal it could be used to construct the imidazole of the target intermediate by treatment with cyanamide under acidic condition.[30] Instead of the desired imidazole derivative, we found the rearrangement of **261** to **262** under this condition. Next, the substrate **259** was treated with methylamine hydrochloride and triethylamine in refluxing ethanol to see the outcome of this reaction, and in this case the formation of the imidazole was observed while the starting material rearranged to **263**.[144]



Scheme 5.19

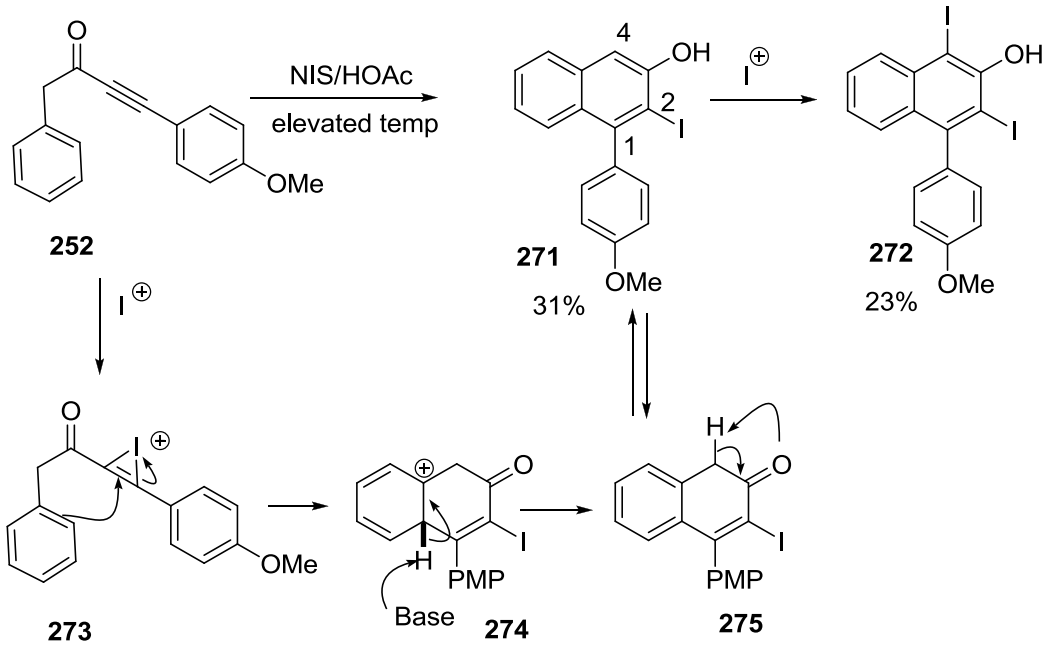
These rearrangements can be understood as depicted in Scheme 5.19, by protonating **259** to provide **264**, which rearranges to form **265** liberating AcOH. Removal of a proton from this intermediate results in the formation of more stable diphenyl derivative **266**. As the methyl amine is available in the reaction mixture, the carbonyl is then converted into the *N*-methylimine **267**. Lost of a proton form this intermediate forms the corresponding 2,3-diaminonaphthalene derivative, which is converted into **268** eventually. Elimination of water form this intermediate provides naphtho[2,3-*d*]imidazole **263**.

It can be assumed that in an acidic media, **261** follows a similar pathway to form **262**. However, it does not form the imidazole ring, may be due to the formation of alcohol **262** is faster than the formation of cyanimine from a **266**-like intermediate under these conditions.



Scheme 5.20

At this moment it was thought that further studies should be done with the actual substrate **252**, having 4-methoxy group in the aromatic ring, which was synthesized in 80% overall yield as shown in Scheme 5.20.[138] Portionwise addition of NIS to a solution of alkynone **252** in acetic acid, at room temperature provided **251** as a 1:1 mixture of diastereomers. It was found that there is a necessity to control the rate of addition of NIS in order to maintain ambient temperature as the products from this reaction are totally different at higher temperature (Scheme 5.21).



Scheme 5.21

The cyclization reaction of **252** at higher temperature can be understood as the intramolecular electrophilic addition of iodonium ion **273** forms intermediate **274**, which forms **275** after the de-protonation. Tautomerization of **275** provides naphthol **271**,[148] which is converted to diiodonaphthol **272** by the electrophilic substitution of iodonium ion at C4-position. The structure **272** was assigned based on the spectroscopic data of these two products; while **271** has 9 protons in the aromatic region in its ^{1}H-NMR spectrum, **272** has only 8 protons. The hydroxyl proton of **271** appears at 5.67 ppm and that of **272** appears at 6.37 ppm, which suggests more shielding, which is consistence with being in the middle of two iodine atoms. The ^{13}C-NMR spectrum of **271** has 15-signals with 7-methylene carbons, whereas **272** has 15-signals with 6-methylene carbons. The other strong evidence is that the C4 and C2 signals of **271** appear at 108.9 ppm and 97.7 ppm respectively. While **272** is missing the peak at 108.9 ppm, its C4 and C2 signals now appear at 93.5 ppm and 83.3 ppm

respectively. If the electrophilic addition takes place in the second aromatic ring of naphthol **271**, **272** should still have the C4 signal around 108 ppm, and while this is not the case, the possibility to have the second iodine at C4 position. Later we were able to confirm this hypothesis by taking an X-ray crystal structure of **272** (Figure 5.1).

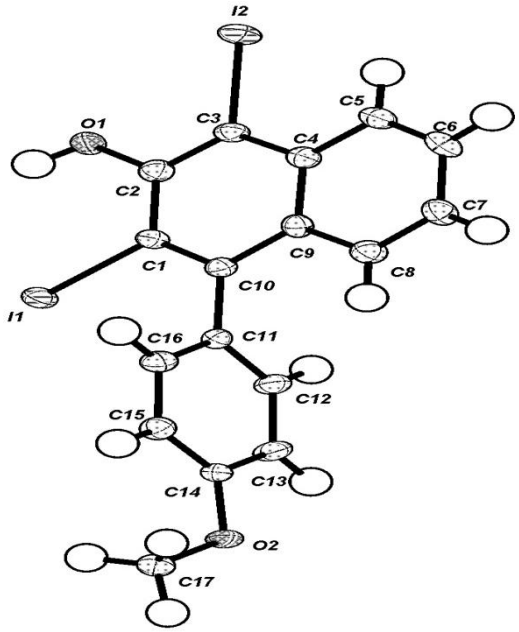
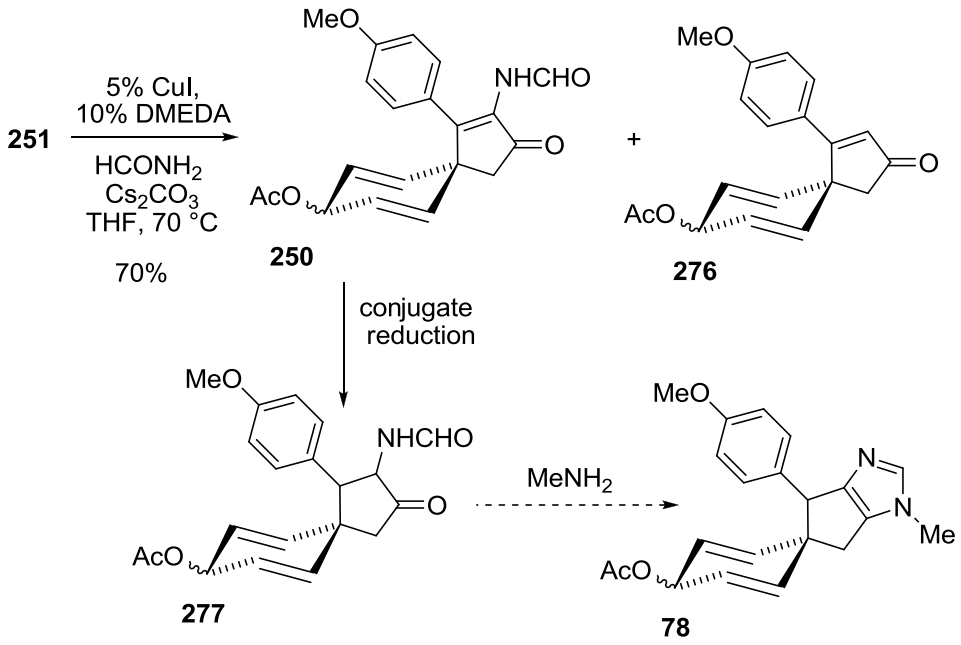


Figure 5.1: X-ray crystal structure of **272**

Next, the cross-coupling reaction of **251** was initiated under optimal conditions to provide keto-amide **250** in 70% yield (Scheme 5.22). From an analysis of rearrangement mechanism shown in Scheme 5.19, it clear that the presence of conjugated enamine moiety of **259** facilitates this pathway and this could be prevented by reducing the conjugation. The reduction of conjugated double bond of the cyclopentenone moiety is the appropriate step in this case, since this would provide the correct unsatuartion required for the imidazole ring. Consequently, before

continuing the synthesis with unsaturated keto-amide **250**, it was subjected to various methods for conjugate reductions, anticipating isolating saturated keto-amide **277** as shown in Scheme 5.22 and Table 3. Treatment of **277** with methylamine hydrochloride (as in Scheme 2.18) would provide spiro-imidazole **78**.

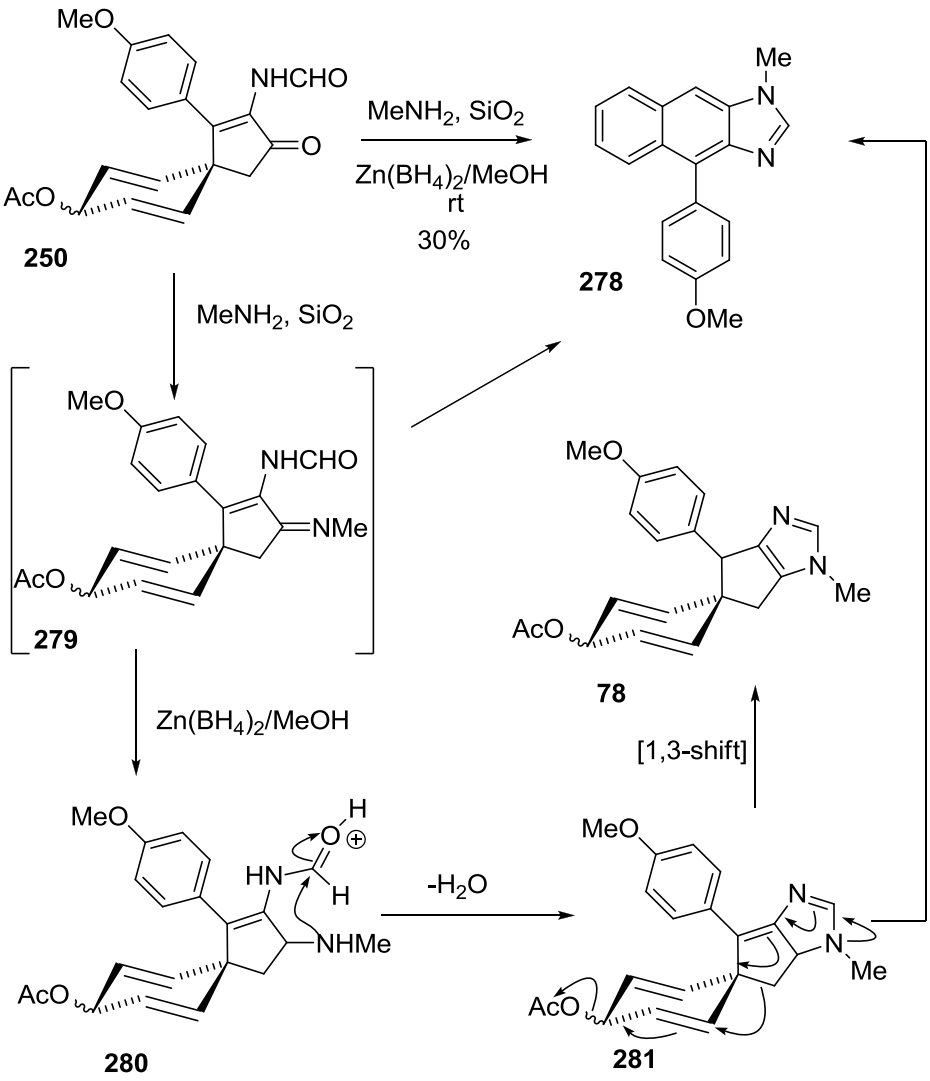


Scheme 5.22

Table 5.3: Conditions for the conjugate reduction of **250** and **251**

Entry	Substrate	Condition	Additive	Silane	yield
1	**250**	$Rh_2(PPh_3)_3Cl$/Benzene, rt	-	Et_3SiH	0
2	**250**	$Pd(PPh_3)_4$ /$CHCl_3$, rt	$ZnCl_2.H_2O$	Ph_2SiH_2	Not clean
3	**250**	K-Selectride/THF, -78 ºC	-	-	0
4	**250**	K-Selectride/THF, rt	-	-	Not clean
5	**250**	MeCu/DIBAL/THF, -50 ºC	HMPA	-	Not clean
6[149]	**250**	HMPA/CH_2Cl_2, 0 ºC→rt		Cl_3SiH	Not clean
7	**251**	$Rh(PPh_3)_3Cl$/Benzene, rt	-	Et_3SiH	0
8	**251**	MeCu/DIBAL/THF, -50 ºC	HMPA	-	0

When the conjugate reduction was attempted with the Rh-catalyst, only the starting material was recovered from the reaction (Entry 1).[150] However, when Pd- or Cu-catalysts were used for the reaction, they were not clean enough to provide clear evidence for the outcome (Entry 2 and 5).[151, 152] Although, potassium *tri*-sec-butylborohydride (K-selectride) has been used previously for the conjugate reduction, no progress was observed with our substrate at -78 ºC, and the reaction was not clean enough to see any result by increasing the reaction temperature.[153] Since all conditions failed to reduce the conjugate double bond of amide **250**, we thought this might be due to the steric hindrance of the conjugate double bond. Therefore, two attempts were made to reduce the conjugate double bond of 2-iodocyclopentenone **251**, hoping this might be reduced easily. However, no reduction was observed from these two reactions also (entries 7 and 8).



Scheme 5.23

As the above conjugate reductions were unsuccessful, **250** on pre-absorbed silica was treated with a solution of $MeNH_2$ and $Zn(BH_4)_2$ in methanol (Scheme 5.23),[149] hoping methylamine **279**, formed during the reaction, would be reduced *in-situ* to produce the amine **280**, and this will provide the right oxidation of the imidazole ring; two unsaturated bonds (Scheme 5.23). Then, this would react with amide to form dihydroimidazole **281**, which will tautomerize to form imidazole **78**. However, naphtho[2,3-*d*]imidazole **278** was isolated from this reaction and at the

moment we do not have evidence to conclude whether this rearrangement take place from imine **279** or from dihydroimidazole **271**.

5.5 Summary

We have evaluated two basic approaches to the spirocalcaridines, which have failed to provide the natural products. The first approach, based on biosynthetic consideration, failed as a result of failure during the bromination of the secondary alcohol **77**. Then we attempted to perform this bromination with the secondary alcohol **225**, and this attempt also failed to provide the desired bromide. Although, we were able to perform the bromination later with a model substrate, with an electron withdrawing group at imidazole *N*1-position, to synthesize bromide **239**, this approach was not successful as the *ipso*-cyclization was problematic from this intermediate. Then, the attempts made to synthesize the intermediate **78** *via* intramolecular Büchner reaction also failed leading us to an alternative approach.

Then the second approach, which is exactly opposite to our first approach in which we focused on the *de novo* synthesis of imidazole ring, also failed as the problem result of failure during the synthesis of imidazole ring with required substitution. However, from this approach we were successfully able to optimize a cross coupling reaction of vinyl-iodide derivatives and formamide. This study also provided some insight in to other *Leucetta* derived alkaloids.

CHAPTER 6

EXPERIMENTAL SECTION

6.1 General methods

All of the reagents were purchased from commercial suppliers and were used without purification otherwise specified. All reactions involving air- or water-sensitive compounds were conducted in oven-dried (overnight) glassware under atmosphere of dry nitrogen. All the solvents were purified by Innovative Technology's Pure-Solve solvent purification system.

NMR spectra were recorded on JEOL Eclipse+ 500 MHz and ECX 300 MHz spectrometers. The ^{1}H NMR spectra were recorded in $CDCl_3$ (unless otherwise indicated) at a spectrometer frequency of 500.13 MHz and 300.53 MHz using residual $CHCl_3$ (δ = 7.26) as reference. The ^{13}C spectra were recorded in $CDCl_3$ (unless otherwise indicated) at a spectrometer frequency of 125.76 MHz and 75.57 MHz using residual $CDCl_3$ (δ = 77.2) as internal reference.

Melting points were recorded on a Laboratory Devices Inc. Melt. Temp apparatus and are uncorrected.

Infrared (IR) spectra were obtained on a Bruker Vector 22 FT-IR spectrometer, using KBr pellets for solid or neat films between NaCl for liquids and oils, and Bruker ALPHA FT-IR Spectrometer using neat samples. All spectra are reported in cm^{-1}.

Mass spectra were recorded by the Department of Chemistry and Biochemistry, University of Florida, Gainesville by electrospray ionization (HR-ESIMS) unless otherwise indicated. All mass spectral data are reported as *m/z* (relative intensity).

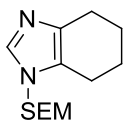
Analytical thin layer chromatography (TLC) was performed on Whatman silica gel 60F254 aluminum backed precoated plates (0.25 mm layer). All liquid chromatography separation (LCS) was performed using ICN silica gel (200-400 mesh).

6.2 Synthesis

3-Phenyl-2-phenylsulfonyloxaziridine (87) and **3-(4-Nitrophenyl)-2-phenylsulfonyloxaziridine (88)** were prepared from corresponding aldehydes and phenylsulfonamide according to the literature procedures.[64, 65]

All of the simple tetrahydrobenzimidazoles **90**, **91a-c, e, f**, **95** and imidazolones **92a-c, 96** have been reported and characterized previously by our group.[52, 70]

1-Trimethylsilylethoxymethyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (91d):



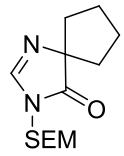
Sodium hydride (60% oil dispersion, 788 mg, 19.7 mmol) was added in small portions to a stirred solution of tetrahydrobenzimidazole **90** (2.00 g, 16.4 mmol) in THF (20 mL) with cooling (ice/water). After 10 min, the reaction mixture was allowed to warm to room temperature and stirred for 2 h. The grey solution was then re-cooled (ice/water) and SEMCl (3.50 mL, 19.7 mmol)

was added dropwise by syringe. The reaction mixture was stirred at room temperature for 25 h and quenched with water (2 mL). The solvent was removed by rotary evaporation and the residue was dissolved in EtOAc. The organic solution was washed with water (50 mL), brine (50 mL) and dried (Na_2SO_4). Concentration gave the crude product which was purified by chromatography (EtOAc/ hexane, 3:7) to provide pure **91d** (2.78 g, 71%) as a pale yellow oil: ^{1}H NMR: δ = 7.43 (s, 1H), 5.13 (s, 2H), 3.46 (t, *J* = 8.2 Hz, 2 H), 2.60 (m, 4H), 1.80 (m, 4H), 0.89 (t, *J* = 8.2 Hz, 2 H), -0.03 (s, 9H); ^{13}C NMR δ = 137.5, 135.8, 125.7, 73.8, 65.8, 24.3, 23.3, 22.9, 20.5, 17.7, -1.3; IR (neat, cm^{-1}): = 3374, 2931, 2853, 1680, 1494, 1447, 1248, 1090, 859, 836. HR-ESIMS: Calcd. for $C_{13}H_{24}N_2NaOSi$ $[M+Na]^+$ 275.1550, found 275.1549.

General procedure for oxidative rearrangements:

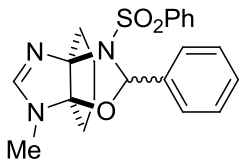
3-Phenyl-2-phenylsulfonyl- (**87**) or 3-(4-Nitrophenyl)-2-phenylsulfonyloxaziridine (**88**) (1.20-2.00 mmol, 1.2-2.0 equiv.) was added to a stirred solution of the tetrahydrobenzimidazole derivative (1.00 mmol, 1 equiv.) in HPLC grade chloroform (15 mL) at the indicated temperature and the mixture was stirred for the indicated time while monitoring the reaction time by TLC. On completion of the reaction, the solvent was removed by rotary evaporation and the crude product was purified through a short plug of silica gel using (mixtures of EtOAc and hexanes) to provide the desired product. We were unable to assign the relative stereochemistry of the [3+2] adducts *via* NOESY experiments, as no diagnostic NOE interactions emerged from these experiments.

3-Trimethylsilylethoxymethyl-1,3-diazaspiro[4.4]non-2-ene-4-one (92d):



3-Phenyl-2-phenylsulfonyloxaziridine (188 mg, 0.72 mmol, 2.0 equiv.) was added to a solution of **91d** (91.0 mg, 0.4 mmol) in $CHCl_3$ (8 mL) at room temperature, which was stirred for 16 h at the same temperature. The solvent was removed by rotary evaporation at and the residue was purified using MPLC (EtOAc/hexanes, 1:4) to produce imidazolone **92d** (58 mg, 60%) as a pale yellow oil: ^{1}H NMR: δ = 7.70 (s, 1H), 4.84 (s, 2H), 3.49 (t, *J* = 8.2 Hz, 2 H), 1.98-1.93 (m, 6H), 1.79 (m, 2H), 0.89 (t, *J* = 8.2 Hz, 2 H), -0.03 (s, 9H); ^{13}C NMR: δ = 185.3, 150.8, 78.2, 70.0, 66.5, 37.5, 26.0, 17.8, -1.4; IR (neat, cm^{-1}): = 2954, 1738, 1610, 1347, 1248, 1088, 837, 754, 693; HR-ESIMS: Calcd. for $C_{13}H_{25}N_2O_2Si$ $[M+H]^+$ 269.1680, found 269.1682.

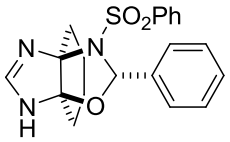
(1*R,6*S**,8*R**/8*S**)-9-benzenesulfonyl-12-methyl-7-oxa-8-phenyl-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (94a):**

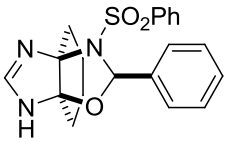


3-Phenyl-2-phenylsulfonyloxaziridine (545 mg, 2.10 mmol, 1.2 equiv.) was added to a solution 1-methyl-4,5,6,7-tetrahydro-1*H*-benzimidazole **91a** (240 mg, 1.75 mmol, 1 equiv.) in chloroform (15 mL) at rt. The residue was purified using MPLC (EtOAc/Hexanes = 1:4) to give the known spiroimidazolone **92a**[52] (218 mg, 82%) and oxaziridine adduct **94a**, the latter as a pale yellow oil (13 mg, 2%): ^{1}H NMR: δ = 7.52 (d, *J* = 7.4 Hz, 2H), 7.34 (t, *J* = 7.4, 1H), 7.22-7.14 (two triplets overlap, *J* = 7.2, 7.4 Hz, 5H), 7.06 (t, *J* = 7.2 Hz, 2H), 6.76 (s, 1H), 6.14 (s, 1H), 2.75 (s, 3H), 2.09-1.92 (m, 3H), 1.86-1.76 (m,1H), 1.70-1.56 (m, 2H), 1.50-1.22 (m, 2H); ^{13}C NMR (DEPT-135): δ = 155.6 (CH), 141.9 (C), 136.5 (C), 131.8 (CH), 129.0 (CH), 128.4 (CH),

128.2 (CH), 127.7 (CH), 127.3 (CH), 100.6 (C), 92.6 (C), 91.1 (CH), 31.7 (CH_2), 28.2 (CH_3), 26.0 (CH_2), 18.3 (CH_2), 17.5(CH_2); IR (neat, cm^{-1}): = 2946, 1597, 1447, 1344, 1162, 754, 724, 689, 606, 566; HR-ESIMS (*m/z*): Calcd. for $C_{42}H_{46}N_6NaO_6S_2$ $[2M+Na]^+$ 817.2812, found 817.2871.

(1*R,6*S**,8*R**)- and (1*R**,6*S**,8*S**)-9-Benzenesulfonyl-7-oxa-8-phenyl-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (94c-i) and (94c-ii):**[a]





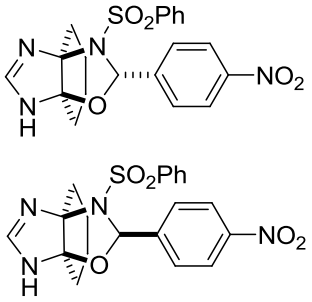
Oxaziridine **87** (0.60 g, 2.30 mmol, 1.2 equiv.) was added to a stirred solution of tetrahydrobenzimidazole **90** (0.235 g, 1.92 mmol, 1.0 equiv.) in $CHCl_3$ (25 mL), and the reaction mixture was stirred for 2 h at rt. At this point the reaction was stopped and the solvent was removed by rotary evaporation. The crude products were separated by MPLC using EtOAc:hexanes (1:1) to provide two diastereomeric products:

Adduct 94c-i, pale yellow solid (90 mg, 12%): m.p = 79-82 °C; ^{1}H NMR: δ = 7.35 (s, 1H), 7.33-7.29 (m, 1H), 7.22-7.04 (m, 10H), 5.82 (s, 1H), 2.61-2.55 (m, 1H), 2.33-2.24 (m, 2H), 1.81-1.56 (m, 5H); ^{13}C NMR (DEPT-135): δ = 152.0 (CH), 140.4 (C), 135.4 (C), 132.2(CH), 129.4 (CH), 128.9 (CH), 128.3 (CH), 127.8 (CH), 127.2 (CH), 104.3 (C), 89.9 (CH), 86.1 (C), 32.9 (CH_2), 31.0 (CH_2), 20.4 (CH_2), 19.3 (CH_2); IR (neat, cm^{-1}) = 3384 (brd), 3067, 2950, 1594, 1447, 1340, 1159, 1090, 1032, 754, 687, 610; HR-ESIMS: Calcd. for $C_{20}H_{21}N_3NaO_3S$ $[M+Na]^+$ 406.1201, found 406.1201.

[a] Each group of spectroscopic data has been assigned arbitrarily to a particular epimer, however, the assignments may be reversed.

Adduct 94c-ii: pale yellow solid (230 mg, 32%): m.p = 84-86 °C; ^{1}H NMR: δ = 7.32-7.26 (m,2H), 7.20-7.02 (m, 10 H), 5.80 (s, 1H), 2.59-2.53 (m, 1H), 2.33-2.21 (m, 2H), 1.80-1.53 (m, 5H); ^{13}C NMR (DEPT-135): δ = 154.3 (CH), 140.6 (C), 134.1 (C), 132.1 (CH), 129.7 (CH), 129.2 (CH), 128.3 (CH), 128.1 (CH), 126.9 (CH), 106.0 (C), 89.7 (CH), 83.2 (C), 31.5 (CH_2), 31.3 (CH_2), 18.4 (CH_2), 17.0 (CH_2); IR (neat, cm^{-1}): = 3385 (brd), 3067, 2949, 1594, 1447, 1340, 1159, 1090, 1032, 754, 610; HR-ESIMS: Calcd. for $C_{40}H_{43}N_6O_6S_2$ $[2M+H]^+$ 767.2680, found 767.2615.

(1*R,6*S**,8*R**)- and (1*R**,6*S**,8*S**)- 9-Benzenesulfonyl-8-(4-nitrophenyl)-7-oxa-9,10,12- triazatricyclo[4.3.3.0]dodec-10-ene (94d-i) and (94d-ii):**



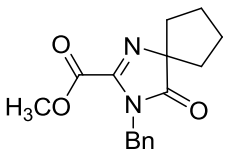
Oxaziridine **88** (2.51 g, 8.19 mmol, 2.0 equiv.) was added to a stirred solution of tetrahydrobenzimidazole **90** (0.50 g, 4.09 mmol, 1.0 equiv.) in chloroform (25 mL) at rt. Then reaction mixture was stirred at rt. for 4.5 h, and then concentrated by rotary evaporation. The crude products were separated by a short plug of silica gel using gradient column (3:7,acetone:hexanes →Acetone) to provide two separable diastereomers.

Adduct 94d-i: pale yellow solid (0.56 g, 32%): m.p = 90-93 °C; ^{1}H NMR: δ = 7.86-7.84 (d, *J* = 8.8 Hz, 2H), 7.46 (d, *J* = 8.8 Hz, 2H), 7.35 (d, *J* = 7.4 Hz, 3H), 7.21 (s, 1H), 7.176 (d, *J* = 7.4 Hz, 2H), 5.97 (s, 1H), 2.63 (m, 1H), 2.23 (m, 2H), 1.71-1.24 (m, 5H); ^{13}C NMR: δ = 152.2, 148.3, 143.4, 140.3, 132.9, 129.8, 128.6, 127.3, 122.9, 105.1, 88.5, 86.1, 32.7, 30.6, 20.1, 18.9; IR (neat, cm^{-1}) = 3370 (brd), 3068, 2945, 1597, 1524, 1347, 1160, 1090, 911, 859, 732, 688; HR-ESIMS (*m/z*): Calcd. for

$C_{20}H_{21}N_4O_5S$ $[M+H]^+$ 429.1227, found 429.1217; Calcd. for $C_{20}H_{20}N_4NaO_5S$ $[M+Na]^+$ 451.1047, found 451.1076.

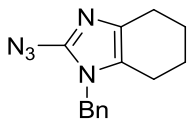
Adduct 94d-ii pale yellow solid (0.52 g, 30%): m.p = 99-101 °C; ^{1}H NMR: δ = 7.91-7.88 (d, *J* = 8.3 Hz, 2H), 7.36-7.33 (d, 4H), 7.22-7.11 (m, 4H), 6.19 (brd, 1H), 5.84 (s, 1H), 2.66-2.59 (m, 1H), 2.35-2.20 (m, 2H), 1.80-1.73 (m, 2H), 1.66-1.60 (m, 3H); ^{13}C NMR (DEPT-135): δ = 154.3 (CH), 148.7 (C), 141.4 (C), 140.2 (C), 132.8 (CH), 130.2 (CH), 128.5 (CH), 127.0, 123.1 (CH), 106.6 (C), 88.1 (CH), 83.6 (C), 31.5 (CH_2), 31.2 (CH_2), 18.3 (CH_2), 16.9 (CH_2); IR (neat, cm^{-1}) = 3392 (brd), 3069, 2950, 1592, 1525, 1347, 1160, 1089, 911, 857, 732; HR-ESIMS (*m/z*): $C_{20}H_{21}N_4O_5S$ $[M+H]^+$ 429.1227, found 429.1212; Calcd. for $C_{20}H_{20}N_4NaO_5S$ $[M+Na]^+$ 451.1047, found 451.1071.

Methyl (3-Benzyl-4-oxo-1,3-diaza-spiro[4.4]non-1-ene-2)-formate (96):



Oxaziridine **87** or **88** (2.0 mol, 2 equiv.) was added to a solution of **95** (1.0 mmol) in dry chloroform (10 mL) at rt. Then, the mixture was stirred at the indicated temperature for 24 h. After removing the solvent, the crude material was purified over silica gel (35% EtOAc in hexanes) to isolate **96** as a pale yellow solid in 12-55% yield.

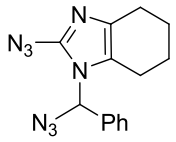
2-Azido-1-benzyl-4,5,6,7-tetrahydro-1*H* -benzimidazole (97):



n-BuLi (16 mmol, 8 mL, 2 M solution in cyclohexane) was added dropwise to a pre-cooled (-78 °C) solution of **91b** (3.15 g, 14.8 mmol) in anhydrous THF (60 mL) and the reaction mixture was allowed to stir for 1.5 h at that temperature. Tosyl azide (3.34 g, 17.0 mmol) was

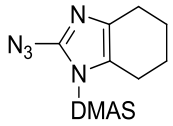
added at -78 °C to the resulting reaction mixture. The suspension was stirred for 2 h at -78 °C and gradually allowed to reach room temperature and stirred for additional 2 h. Saturated aqueous solution of NH_4Cl was added to the reaction and extracted with EtOAc (3x 50 mL). The combined organics were dried over anhydrous Na_2SO_4 and the solvent was removed in *vacuo*. The resulting residue was purified over silica gel using 20% EtOAc in hexanes to isolate **97** (2.27 g, 60%) and **98** (1.17 g, 27%): ^{1}H NMR: δ = 7.32 (t, *J* = 7.3 Hz, 2H), 7.28 (d, *J* = 7.3 Hz, 1H), 7.08 (d, *J* = 7.3 Hz, 2H), 4.82 (s, 2H), 2.54 (m, 2H), 2.33 (m, 2H), 1.75 (m, 4H); ^{13}C NMR δ = 138.8, 136.5, 134.6, 128.9, 127.8, 126.8, 125.0, 46.2, 24.2, 23.3, 22.8, 20.8; IR (neat, cm^{-1}) = 2932, 2851, 2132, 1494, 1473, 728, 695; HR-ESIMS: Calcd. for $C_{14}H_{16}N_5$ $[M+H]^+$ 254.1400, found 254.1399; Calcd. for $C_{14}H_{16}N_2$ $[M-N_3+H]^+$ 226.1339, found 226.1335.

2-Azido-1-(azidophenylmethyl)-4,5,6,7-tetrahydro-1*H*-benzimidazole (98):



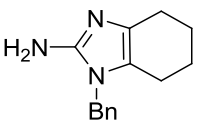
Dark brown solid: m.p = 90-93 °C; ^{1}H NMR δ = 7.37-7.32 (m, 3H), 7.25-7.21 (m , 2H), 6.72 (s, 1H), 2.52-2.49 (m, 2H), 2.37-2.28 (m, 1H), 1.75-1.57 (m, 5H); ^{13}C NMR: δ = 138.9, 136.0, 135.0, 129.1, 128.8, 126.1, 124.5, 70.9, 24.2, 22.9, 22.8, 21.8; DEPT (135°) δ =: 129.1 (CH), 128.8 (CH), 126.1 (CH), 70.7 (CH), 24.2 (CH_2), 22.9 (CH_2), 22.7 (CH_2), 21.8 (CH_2); IR (KBr, cm-1) = 3317, 3051, 3030, 2934, 2852, 2147, 2103, 1503, 1440, 1242, 738, 697; HRMS (*m/z*): Calcd. for $C_{14}H_{15}N_8$ $[M+H]^+$ 295.1414, found 295.1414.

2-Azido-1-dimethylaminosulfonyl-4,5,6,7-tetrahydrobenzimidazole (99):



n-BuLi (7.3 mL, 11 mmol of 1.5 M solution in hexanes) was added dropwise to a pre-cooled (-78 °C) solution of **91e** (2.29 g 10.0 mmol) in anhydrous THF (50 mL), followed by stirring for 1.5 h at that temperature. Tosyl azide (2.35 g, 12.0 mmol) was added in one portion at -78 °C to the reaction mixture. The suspension was stirred for 2 h at -78 °C and gradually allowed to reach room temperature and stirred for additional 2 h. The reaction mixture was quenched with dilute NH_4Cl (15 mL) and water (50 mL) was added, followed by extraction with CH_2Cl_2 (2x50 mL). The combined organic extracts were dried (Na_2SO_4) and concentrated in *vacuo* below 30 °C. The resulting crude residue was purified by column chromatography (hexane/ethyl acetate, 1:9) to yield **99** as a yellow solid (2.30 g, 85%): m.p = 101-103 °C; ^{1}H NMR δ = 2.97 (s, 6H), 2.71-2.69 (m, 2H), 2.50-2.48 (m, 2H), 1.78-1.76 (s, 4H), ^{13}C NMR: δ = 139.2, 135.0, 126.8, 38.4, 24.3, 23.4, 22.9, 22.5; IR (KBr, cm^{-1}) = 2940, 2857, 2149, 1609, 1510, 1395, 1188, 1057, 967; HR-ESIMS: Calcd for $C_9H_{15}N_6O_2S$ [M+H]+ 271.0972, found 271.0972; Calcd for $C_9H_{15}N_4O_2S$ $[M+H-2N]^+$ 243.0910, found 243.0889.

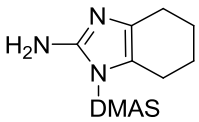
2-Amino-1-benzyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (100):



$NaBH_4$ (519 mg, 13.4 mmol, 2.1 equiv.) was added portionwise to a solution of **97** (1.62 g, 6.39 mmol, 1 equiv.) in anhydrous MeOH (50 mL) with cooling (ice/water). The mixture was stirred 10 min at the same temperature, at which time TLC analysis indicated the completion of the starting material. The reaction was quenched by adding saturated NH_4Cl (20 mL) and the aqueous layer was extracted with CH_2Cl_2 (3x30 mL), combined layer

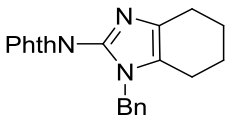
was dried (Na_2SO_4) and concentrated to isolate **100** (1.45 g, 100%) as a pale yellow solid: m.p = 134-137 °C; ^{1}H NMR: δ = 7.53- 7.26 (m, 3H), 7.10-7.08 (m, 2H), 4.84 (s, 2H), 3.77 (br, 2H), 2.49 (m, 2H), 2.37 (m, 2H), 1.80- 1.78 (m, 4H); ^{13}C NMR δ = 146.6, 136.7, 131.2, 129.1, 127.8, 126.4, 121.6, 45.9, 24.0, 23.5, 23.1, 20.7; IR (KBr, cm^{-1}) = 3363, 3290, 3087 (brd) 2942, 2917, 2854, 1643, 1539, 1453, 726, 698; HR-ESIMS (*m/z*): Calcd for $C_{14}H_{18}N_3$ $[M+H]^+$ 228.1495, found 228.1511.

2-Amino-1-dimethylaminosulfonyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (101):



Following the above procedure, $NaBH_4$ (204 mg, 5.27 mmol, 2.1 equiv.) and **99** (0.68 g, 2.51 mmol, 1 equiv.) were used to isolate **101** (0.620 g, 100%) as a pale yellow solid: m.p = 192-195 °C; ^{1}H NMR: δ = 5.20 (br, 2H), 2.92 (s, 6H), 2.62-2.58 (m, 2H), 2.41-2.37 (m, 2H), 1.78-1.70 (m, 4H); ^{13}C NMR δ =148.2, 133.6, 120.3, 38.4, 24.2, 23.2, 23.0, 22.8; IR (neat, cm^{-1}): = 3423, 3282, 3122, 2932, 2855, 1638, 1562, 1454, 1377, 1185, 1163, 1055, 969, 724; HR-ESIMS: Calcd. for $C_9H_{17}N_4O_2S$ $[M+H]^+$ 248.1072, found 245.1062.

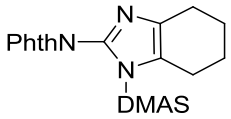
1-Benzyl-2-phthalimidoyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (103):



The amine **100** (1.36 g, 6.0 mmol), potassium carbonate (1.65 g, 12.0 mmol) and the modified Nefkens' reagent **102** (2.97 g, 12.0 mmol) were added simultaneously to dichloromethane (75 mL) and the reaction mixture was allowed to stir at rt. for 24 h. The reaction mixture was washed with 10% $NaHCO_3$ solution and the organic layer was separated. The aqueous phase was further extracted with CH_2Cl_2 (25 mL). The combined organic extracts were washed with brine and dried (Na_2SO_4). The organic solution was

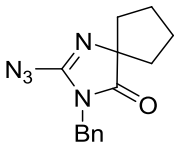
concentrated by rotary evaporation and the residue was purified by column chromatography (hexane/ethyl acetate, 7:3) to obtain the product **103** (1.92 g, 90%) as a solid: m.p = 174-175 °C ; ^{1}H NMR: δ = 7.93-7.89 (dd, *J* = 3.0, 5.5 Hz, 2H), 7.80-7.75 (dd, *J* = 3.0, 5.5 Hz, 2H), 7.27-7.19 (m, 3H), 7.08-7.06 (m, 2H), 4.91 (s, 2H), 2.66 (m, 2H), 2.35 (m, 2,H), 1.80 (m, 4H); ^{13}C NMR δ = 166.9, 136.6, 135.5, 134.8, 131.6, 130.5, 128.9, 127.9, 127.7, 126.9, 124.2, 47.5, 24.2, 23.1, 22.8, 21.2; IR (neat, cm^{-1}): = 2933, 2850, 1731, 1504, 1469, 1434, 1362, 1080, 697; HRESIMS: Calcd. for $C_{22}H_{20}N_3O_2$ $[M+H]^+$ 2358.1550, found 358.1529; Calcd. for $C_{44}H_{39}N_6O_4$ $[2M+H]^+$ 715.3027, found 715.3051.

2-Phthalimidoyl-1-dimethylaminosulfonyl-4,5,6,-7-tetrahydro-1*H*-benzlimidazole (104):



Amine **101** (732 mg, 3.0 mmol), potassium carbonate (0.826 g, 6.0 mmol) and the modified Nefkens' reagent **102** (1.485 g, 6.0 mmol) (45 mL) were stirred in dichloromethane, following the above procedure to obtain the product **104** (1.12 g, 100%) as a pale yellow solid after the purification by column chromatography (hexane/ethyl acetate, 7:3): m.p = 197-200 °C; ^{1}H NMR: δ = 7.95 (dd, *J* = 3.0, 5.5 Hz, 2H, 2H), 7.81 (dd, *J* = 3.0, 5.5 Hz, 2H, 2H), 2.88 (s, 6H), 2.76 (m, 2H), 2.64 (m, 2H), 1.86 (m, 4H);); ^{13}C NMR δ = 166.8, 137.1, 134.9, 131.8,131.0, 128.4, 124.3, 38.1, 24.1, 22.9, 22.7, 22.5; IR (neat, cm^{-1}) = 2941, 2856, 1735, 1721, 1526, 1381, 1176, 975, 882, 725, 619, 572; HR-ESIMS (*m/z*): Calcd. for $C_{34}H_{36}N_8NaO_8S_2$ $[2M+Na]^+$ 771.1990, found 771.1919.

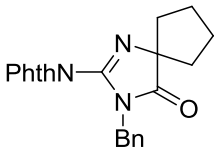
2-Azido-3-benzyl-1,3-diazaspiro[4.4]non-2-ene-4-one (105):



Oxaziridine **88** (765 mg, 2.50 mmol, 2.5 equiv.) was added to a solution of **97** (253 mg, 1.0 mmol) in $CHCl_3$ (15 mL) at room temperature. The reaction mixture was heated at 35 °C for 4 h, at which time TLC analysis indicated the completion of the reaction. The solvent was removed under vacuum at room temperature, and the residue was purified through a silica gel column (hexane/ethyl acetate, 8:2) to afford **105** (175 mg, 70%) as a pale yellow solid: m.p= 83-85 °C. ^{1}H NMR: δ = 7.46-7.44 (m, 2H), 7.36-7.25 (m, 3H), 4.97 (2H, s), 2.36-2.28 (m, 2H), 2.24-2.13 (m, 2H), 2.13-2.03 (m, 4H); ^{13}C NMR: 177.1, 156.7, 134.2, 129.2, 128.84, 128.80, 74.6, 45.6, 37, 6, 25.5; IR (KBr, cm^{-1}): = 2968, 2937, 1750, 1591,1501, 1437, 1343, 1220, 1120, 747, 704, 627; HR-ESIMS: Calcd. for $C_{14}H_{16}N_5O$ $[M+H]^+$ 270.1349, found 270.1346.

This product was isolated in 40% yield when this reaction was repeated in methanol underotherwise indicated conditions.

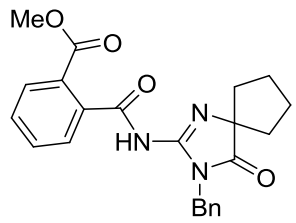
2-Phthalimidoyl-3-benzyl-1,3-diazaspiro[4.4]non-2-ene-4-one (106a):



Oxaziridine **88** (1.11 g, 3.64 mmol, 2 equiv.) was added to a stirred solution of **103** (651 mg, 1.8 mmol) in anhydrous chloroform (27 mL) at 40 °C, and it was stirred for 12 h. After that, the solvent was removed *in vacuo* and the crude product was purified by a short plug of silica gel using (EtOAc/Hexanes = 15/85) to provide **106a** as a pale yellow solid (269 mg, 40%): m.p =159.5-162.5 ˚C; ^{1}H NMR: δ = 7.83-7.79 (m, 2H), 7.79-7.75 (m, 2H), 7.08-7.04 (m, 3H), 7.98-7.95 (m, 2H), 4.68 (s, 2H), 2.19-2.16 (m, 2H), 2.04-2.01 (m, 6H); ^{13}C NMR: δ = 184.4, 164.5, 146.5, 135.0, 131.3, 128.8, 127.9,

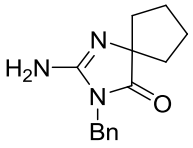
127.0, 124.3, 79.0, 44.3, 37.8, 26.0; IR (neat, cm^{-1}) = 2965, 1733, 1635, 1408, 1337, 1150,882, 718; HRMS (*m/z*): Calcd. for $C_{22}H_{20}N_3O_3$ $[M+H]^+$ 374.1491, found 374.1491; Calcd. for $C_{22}H_{19}N_3NaO_3$ $[M+Na]^+$ 396.1319, found 396.1310.

***N*-(3-Benzyl-4-oxo-1,3-diaza-spiro[4.4]non-1-en-2-yl)-phthalamic acid methyl ester (106b):**



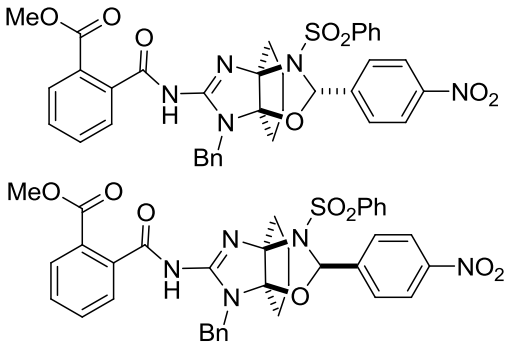
Oxaziridine **88** (616.3 mg, 2.01 mmol, 2 equiv.) was added to a stirred solution of **103** (360 mg, 1.0 mmol) in anhydrous chloroform (15 mL) at 40 °C followed by stirring for 23 h. After removing the solvent *in vacuo*, crude product was purified by a short plug of silica gel using (EtOAc/Hexanes = 2/8) to isolate the **106b** (although no methanol was added, this product was isolated from repetition of this reaction) as a pale yellow solid (194.4 mg, 47%): m.p =127-129 ˚C; ^{1}H NMR: δ = 9.45 (s 1H, disappear upon addition of D_2O), 8.08-8.05 (m, 1H), 7.54-7.47 (m, 3H), 7.43-7.41 (m, 2H), 7.34-7.26 (m, 3H), 4.85 (s, 2H), 3.83 (s, 3H), 2.18-2.12 (m, 2H), 1.92-1.81 (m, 6H); ^{13}C NMR: δ = 179.3, 177.4, 170.5, 159.6, 137.0, 136.1, 133.8, 130.9, 130.1, 129.9, 128.7, 128.5, 128.0, 127.9, 69.0, 52.5, 43.0, 38.0, 25.4; DEPT (135°) δ =179.3(C), 177.4(C), 170.5(C), 159.6(C), 137.0(C), 136.1(C), 133.8(C), 130.9(CH), 130.1(CH), 129.9(CH), 128.7(CH), 128.5(CH), 128.0(CH), 127.9(CH), 69.0(C), 52.5(CH_3), 43.0(CH_2), 38.0(CH_2), 25.4(CH_2); IR (neat, cm^{-1}) = 3314 (brd), 3030, 2952, 2874, 1731, 1633, 1578,1465, 1343, 1122, 1056, 754, 700; HRMS (*m/z*): Calcd. for $C_{23}H_{24}N_3O_4$ $[M+H]^+$ 406.1761, found 406.1720.

2-Amino-3-benzyl-1,3-diazaspiro[4.4]non-2-ene-4-one (106c):



Oxaziridine **87** (522 mg, 2.00 mmol, 2 equiv.) was added to **100** (227 mg, 1.0 mmol) in MeOH (30 mL) followed by stirring at room temperature for 4 h. The solvent was removed by rotary evaporation and the residue was purified through a short plug of silica gel (EtOAc/Hexanes, 1:1→EtOAc→MeOH/EtOAc, 0.5:9.5) to isolate **106c** (143 mg, 59%) as a pale yellow solid: m.p = 106-108 °C; 1H NMR: δ = 7.35-7.32 (m, 2H), 7.30-7.28 (d, 1H), 7.27-7.23 (m, 2H), 4.70 (s, 2H), 4.65 (br, 2H), 2.10-2.05 (m, 2H), 1.90-1.73 (m, 6H); 13C NMR δ = 181.2, 155.5, 135.5, 129.0,128.1, 127.2, 71.9, 42.7, 38.1, 25.6. IR (neat, cm^{-1}): = 3364, 3032, 2958, 1665, 1455, 1353, 1074, 754, 666; HR-ESIMS: Calcd. for $C_{14}H_{18}N_3O$ [M+H]+ 244.1444, found 244.1463.

1*R,6*S**,8*R**)- and (1*R**,6*S**,8*S**)- *N*-[9-Benzenesulfonyl-12-benzyl-8-(4-nitrophenyl)-7-oxa-9,10,12-triaza-tricyclo[4.3.3.0]dodec-10-en-11-yl]-phthalamic acid methyl ester (107a) and (107b):**



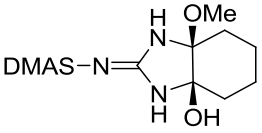
Oxaziridine **88** (320 mg, 1.04 mmol, 2 equiv.) was added to **103** (187 mg, 0.5 mmol) in MeOH (8 mL) followed by stirring at 40 ºC for 17 h. The solvent was removed by rotary evaporation and the residue was purified through a short plug of silica gel (EtOAc/Hexanes, 1:4) providing two products.

Adduct 107a, pale yellow solid (22 mg, 6%): m.p = 90-93 °C; ^{1}H NMR: δ = 9.72 (s, 1H), 8.05 (m, 1H), 7.93 (m, 1H), 7.84 (d, *J* = 8.8 Hz, 2H), 7.57-7.39 (m, 8H), 7.34 (m,

2H), 7.27-7.28 (m, 4H), 6.18 (s, 1H), 4.64 (d, *J* = 15.1 Hz, 1H), 4.09 (d, *J* = 15.1 Hz, 1H), 3.88 (s, 3H), 2.81 (m, 1H), 2.14 (m, 1H), 1.89-1.84 (m, 1H), 1.66-1.49 (m, 3H), 1.38 (m, 2H); ^{13}C NMR: δ = 178.7, 170.5, 160.1, 148.2, 142.2, 140.4, 137.8, 137.6, 133.8, 133.2, 130.6, 130.1, 129.8, 129.2, 129.0, 128.7, 128.6, 127.9,127.2, 126.5, 123.2, 98.4, 89.6, 82.9, 52.6, 43.7, 30.9, 30.4, 18.3, 17.3; IR (neat, cm^{-1}) = 3335 (brd), 3064, 2951, 2852, 1729, 1614, 1561, 1483, 1348, 1290, 1161, 1091, 1034, 735; HR-ESIMS (*m/z*): Calcd. for $C_{36}H_{34}N_5O_8S$ $[M+H]^+$ 696.2122, found 696.2114; Calcd. for $C_{36}H_{33}N_5NaO_8S$ $[M+Na]^+$ 718.1948, found 718.1931.

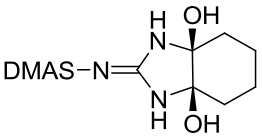
Adduct 107b, pale yellow solid (182 mg, 50%): m.p = 79-82 °C; ^{1}H NMR: δ = 9.24 (s, 1H), 8.33 (d, *J* = 8.8 Hz, 2H), 8.13 (d, *J* = 8.3 Hz, 2H), 8.00 (d, *J* = 8.8 Hz, 2H), 7.91-7.76 (m, 4H), 7.71-7.53 (m, 4H), 7.51-7.47 (m, 4H), 7.47 (d, *J* = 7.2 Hz, 2H), 7.05 (s, 1H), 4.92 (d, *J* = 15.1 Hz, 1H), 4.60 (d, *J* = 15.1 Hz, 1H), 4.06 (s, 3H), 2.48-2.41 (m, 1H), 2.28-2.23 (m, 1H), 1.92-1.80 (m, 2H), 1.66-1.14 (m, 4H); ^{13}C NMR: δ = 178.4, 170.3, 159.8, 148.1, 144.6, 141.1, 137.9, 137.2, 133.9, 133.3, 130.5, 130.0, 129.8, 129.5, 128.8, 128.5, 127.8, 127.7, 127.1, 126.9, 123.5, 98.7, 90.2, 84.3, 52.4, 42.8, 28.2, 25.0, 15.3, 14.1 ; IR (neat, cm^{-1}) = 3335 (brd), 3054, 2951, 2836, 1729, 1601, 1561, 1478, 1348, 1285, 1161, 1078, 1034, 735; HR-ESIMS (*m/z*): Calcd. for $C_{36}H_{34}N_5O_8S$ $[M+H]^+$ 696.2122, found 696.2120; Calcd. for $C_{36}H_{33}N_5NaO_8S$ $[M+Na]^+$ 718.1948, found 718.1933.

2-Dimethylaminosulfonylimino-3a-hydroxy-7a-methoxyoctahydrobenzimidazole (108a):



Oxaziridine **88** (615 mg, 2.00 mmol, 2.0 equiv.) was added to a stirred solution of **101** (245 mg, 1.0 mmol) in anhydrous MeOH (15 mL) at room temperature and it was stirred for 4 h. The solvent was removed *in vacuo* and the crude product was purified by a short plug of silica gel using (EtOAc/hexanes = 6/4) to provide a white solid which was recrystallized from dichloromethane to isolate **108a** as white crystalline solid (198 mg, 68%): m.p =149-151 °C; ^{1}H NMR (DMSO-d6): δ = 8.16 (s, 1H), 7.71 (s, 1H), 5.51 (s, 1H), 3.19 (s, 3H), 2.53 (s, 6H), 2.05-1.93 (m, 2H), 1.53-1.17 (m, 6H); ^{13}C NMR: δ = 157.8, 89.0, 87.8, 50.1, 38.8, 36.9, 29.1, 21.2, 19.9; IR (KBr, cm^{-1}) = 3463, 3350, 3251 (brd), 2956, 2866, 1611, 1449, 1313, 1201, 1140, 1087, 1056, 950, 863, 716; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{40}N_8NaO_8S_2$ $[2M+Na]^+$ 607.2303, found 607.2286.

2-Dimethylaminosulfonylimino-3a,7a-dihydroxyhexahydrobenzimidazole (108b):



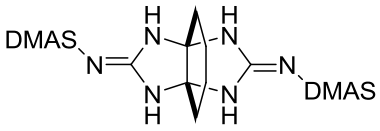
Oxaziridine **88** (366 mg, 1.2 mmol, 1.1 equiv.) was added to a stirred solution of **101** (270 mg, 1.1 mmol) in an acetone-water (2:1) mixture (15 mL) followed by stirring for 2 h. The solvent was removed *in vacuo* and the crude product was purified by a short plug of silica gel using (acetone/hexanes = 3/7) to isolate a white solid, which was recrystallized using methanol-acetone mixture to give **108b** as a colorless crystalline solid (199 mg, 65%): m.p =100-103 °C; ^{1}H NMR (DMSO-d6): δ = 7.60 (s, 2H), 5.61 (s, 2H), 2.52 (s, 6H), 1.80-1.55 (m, 4H), 1.40-1.20 (m, 4H); ^{13}C NMR: δ = 157.9,

86.5, 38.8, 34.3, 20.8; IR (KBr, cm^{-1}) = 3417, 3364, 3348, 3258, 2942, 2868, 1611, 1465, 1298, 1202, 1146, 1049, 953, 874,859, 717 ; HR-ESIMS (*m/z*): Calcd for $C_9H_{19}N_4O_4S$ $[M+H]^+$, 279.1121, found 279.1135; Calcd for $C_9H_{18}N_4NaO_4S$ $[M+Na]^+$ 301.0940, found 301.0949.

Synthesis of 108b using DMDO:

A 0.032 M solution of DMDO (28.80 mL, 0.9 mmol) in acetone was added to amine **101** (150 mg, 0.6 mmol) and water (1 mL) at rt. After stirring for overnight, solvent was removed and the product was recrystallized with water to isolate **108b** (29 mg, 17%) as a white solid.

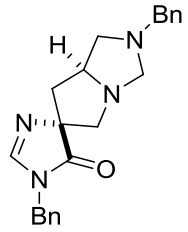
8,11-Bis(*N,N*-dimethylsulfonylimino)-7,9,10,12-tetraazatricyclo[4.3.3.0]dodecane (120):



A solution of **108b** (115 mg, 0.4 mmol) and NaOH (82 mg, 2.05 mmol, 5 equiv.) in acetonitrile was heated at 70 ºC overnight (The white solid was soluble in the reaction mixture while providing a brown colored mixture over 1 h period). After cooling to rt., the reaction was neutralized with diluted HCl and extracted with EtOAc. Evaporation of the solvent under vacuum provided the title compound as a brown solid: m.p = > 250 ºC; ^{1}H NMR (DMSO-d6): δ = 8.16 (s, 4H), 2.51 (s, 12H), 1.82 (m, 4H), 1.36 (m, 4H); ^{13}C NMR: δ = 157.9, 77.4, 38.7, 30.3, 17.0; IR (KBr, cm^{-1}) = 3385, 3108, 2969, 1610, 1520, 1347, 1286, 1134, 1110, 894, 716; HR-ESIMS (*m/z*): Calcd for $C_{12}H_{25}N_8O_4S_2$ $[M+H]^+$, 409. 1414, found 409.1378; Calcd for $C_{12}H_{24}N_8NaO_4S_2$ $[M+Na]^+$ 431.1262, found 431.1194.

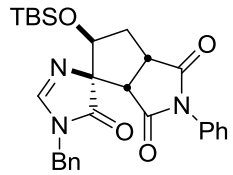
All other compounds **121-130** have been prepared from our lab earlier.[52, 70]

(3*R,5'*S**,7'a*S**)-2',3-Dibenzyl-7',7'a-dihydro-spiro[5*H*-imidazole-5,6'(5'H)-[1*H*]pyrolo[1,2-c]imidazole1',3',4(2'*H*,3*H*)-trione *endo*-(122):**



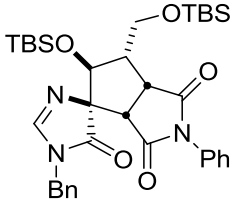
Following the general procedure, **121a,b** (90 mg, 0.2 mmol) and oxaziridine **88** (89 mg, 0.3 mmol, 1.2 equiv.) in chloroform were stirred at rt. for 4 h. Chromatographic separation using (3:7 EtOAc:hexanes→ EtOAc) provided **122** (43 mg, 48%).

(3'a,*S,4*R**,5'*S**,6'a*S**)-1-Benzyl-5'-*tert*-butyldimethylsilyloxy-2'-phenyl-1',3',3'a,6'a-tetrahydrospiro[5-imidazolone-4,4'-cyclo[3'a,6'a-*c*]pyrrole-1,3-dione]** (*endo*-**124**)**:**

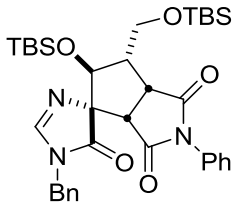


Following the general procedure, **123** (152 mg, 0.3 mmol) and oxaziridine **88** (197 mg, 0.37 mmol, 2 equiv.) in chloroform were stirred at 40 ºC for 40 h. Chromatographic separation using (1:3 EtOAc: hexanes) provided *endo*-**124** (115 mg, 70%) as a white solid; m.p = 187-190 ºC.

(3'a,*S,4*S**,5'*S**,6'*R*,6'a*S**)-1-Benzyl-5'-*tert*-butyldimethylsilyloxy-6'-*tert*-butyl dimethylsilyloxymethyl-2'-phenyl-1',3',3'a,6'a-tetrahydrospiro[5-imidazolone-4,4'-cyclo[3'a,6'a-*c*]pyrrole-1,3-dione]** (*endo*-**126a** and *exo*-**126b):**



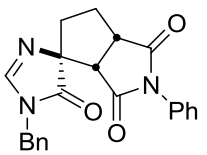
endo-**126a**



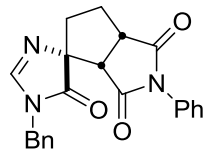
exo-**126b**

125 (134 mg, 0.2 mmol) and oxaziridine **88** (262 mg, 0.85 mmol, 4 equiv.) in chloroform were stirred at 40 ºC for 36 h. Chromatographic separation (1:1 EtOAc: hexanes) provided *endo*-**126a** (18 mg, 13%), *exo*-**126b** (27 mg, 20%) and starting material **125** (22 mg, 16%).

(3'a,*S,4*R**,6'a*S**)-1-Benzyl-2'-phenyl-1',3',3'a,6'a-tetrahydrospiro[5-imidazolone-4,4'-cyclo[3'a,6'a-*c*]pyrrole-1,3-dione]** (*endo*-**128a** and *exo*-**128b):**



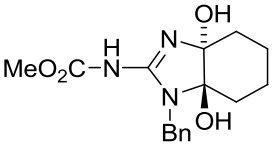
endo-**128a**



exo-**128b**

Following the general procedure, **127** (158 mg, 0.4 mmol) and oxaziridine **88** (410 mg, 1.33 mmol, 3 equiv.) in chloroform were stirred at 40 ºC for 96 h. Chromatographic separation using (2:3 EtOAc: hexanes) provided *endo*-**128a** (28 mg, 17%), *exo*-**128b** (64 mg, 39%) and starting material **127**.

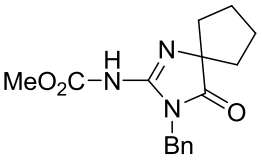
Methyl 2-(1-Benzyl-3a,7a-dihydroxy-3a,4,5,6,7,7a-hexahydro-1*H*-benzoimidazolyl)carbamate (130):[70]



Oxaziridine **88** (185 mg, 0.60 mmol, 1.5 equiv.) was added to a stirred solution of **129** (115 mg, 0.4 mmol) in an acetone-water (10:1) mixture (11 mL) followed by stirring for 20 h. The solvent was removed *in vacuo* and the crude product was purified over a short plug of silica gel (1:4 →1:1, EtOAc: hexanes) to provide **130** (23 mg, 17%): ^{1}H NMR: δ = 8.20 (s, 1H), 7.37 (m, 2H), 7.29-7.21 (m, 3H), 4.66 (d, *J* = 15.4 Hz, 1H), 4.42 (d, *J* = 15.4 Hz, 1H), 3.69 (s, 3H), 1.91-1.81 (m, 3H), 1.58-1.41 (m, 2H), 1.38-1.11 (m, 3H).

Repeating the above procedure, DMDO in acetone (0.03 M, 17.50 mL, 0.5 mmol) and water (1 mL) were added to **129** (100 mg, 0.4 mmol) at . After overnight stirring, solvent was removed and purified as above to isolate **130** (73 mg, 70%) was isolated from column chromatography.

1-Benzyl-2-methylcarbamato-1,3-diazaspiro[4,4]non-1-en-4-one (106d):

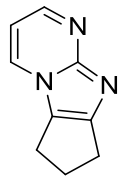


An aqueous solution of NaOH (4.0 M, 0.2 mL) was added to a solution of **130** (100 mg, 0.3 mmol) in acetonitrile (2.5 mL) and the mixture was heated at 70 ºC for 4 h. After cooling to rt., the solution was acidified with 2% aqueous HCl solution and the product was extracted in to EtOAc. The dried solvent was removed in *vacuo* to afford a crude product, which was purified over silica gel (1:1, EtOAc: hexanes) to isolate 106d (13 mg, 18%) as a light brown solid: ^{1}H NMR: δ = 8.63 (br, 1H), 7.40-7.37 (m,

2H), 7.33-7.27 (m, 3H), 4.79 (s, 2H), 3.74 (s, 3H), 2.21-2.09 (m, 2H), 1.94-1.81 (m, 6H); ^{13}C NMR: δ = 177.3, 164.5, 160.3, 135.9, 128.7, 128.5, 127.9, 68.8, 53.0, 42.8, 38.0, 25.3.

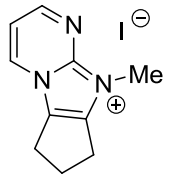
2-Bromocyclopentanone (132) was prepared from cyclopentanone (**131**) following Yadav's protocol, in 48% yield.[80]

4,5,6-trihydrocyclopentaimidazo[1,2-*a*]pyrimidine (134):



2-aminopyrimidine (1.43 g, 15.1 mmol) was added to a solution of bromide **132** (3.19 g, 19.6 mmol) in dry acetonitrile (30 mL) at rt. and the mixture was heated at 80 ºC for 10 h. Then, HBr (10 mL) was added to the mixture and heated at 140 ºC for 30 min. After cooling the reaction to rt., solid K_2CO_3 was sued to neutralize the reaction mixture and the product was extracted in to CH_2Cl_2 (2x20 mL) solvent was removed to isolate **134** (1.20 g, 50%) as a dark brown solid: m.p = 176-179 ºC; ^{1}H NMR: δ = 8.39 (dd, *J* = 1.9, 4.1 Hz, 1H), 8.15 (dd, *J* = 1.9, 6.8 Hz, 1H), 6.81 (dd, *J* = 4.1, 6.8 Hz, 1 H), 2.95 (two overlapped triplets, *J* = 7.2, 6.9 Hz, 4H), 2.59 (two overlapped triplets, *J* = 7.2, 6.8 Hz, 2H); ^{13}C NMR: δ = 155.2, 151.9, 146.5, 130.6, 125.1, 108.0, 26.9, 25.9, 22.4; IR (KBr, cm^{-1}) = 3066, 3016, 2965, 2918, 2855, 1619, 1514, 1438, 1358, 1238, 1188, 1071, 788, 761 ; HR-DARTMS (*m/z*): Calcd. for $C_9H_{10}N_3$ $[M+H]^+$ 160.0869, found 160.0865.

1-Methyl-4,5,6-trihydrocyclopentaimidazolium[1,2-*a*]pyrimidine (135):



MeI (0.52 mL, 8.3 mmol) was added to a solution of **134** (1.20 g, 7.54 mmol) in toluene (30 mL) at rt., and the resulting mixture was heated to reflux for 25 min. Reaction was cooled to rt. and the solvent was decanted to isolate the brown solid, which was washed once with EtOAc (50 mL) to provide the pure product, **135** (1.42 g, 64%) as a light brown solid: m.p = 158-160 ºC; ^{1}H NMR (D_2O): δ = 8.78 (dd, *J* = 1.6, 7.0 Hz, 1H), 8.75 (dd, *J* = 1.6, 4.7 Hz, 1H), 6.46 (dd, *J* = 4.7, 7.0 Hz, 1 H), 3.87 (s, 3H), 2.95 (t, *J* = 7.2 Hz, 4H), 2.60 (quintet, *J* = 7.2 Hz, 2H); ^{13}C NMR: δ = 154.0, 145.1, 143.4, 135.8, 127.4, 113.3, 31.1, 26.2, 23.2, 22.7; IR (KBr, cm^{-1}) = 3066, 3003, 2955, 1646, 1518, 1448, 1397, 1362, 1331, 1293, 1146, 816, 772, 749; HR-ESIMS (*m/z*): Calcd for $C_{10}H_{12}N_3$ $[M]^+$ 174.1026, found 174.1023.

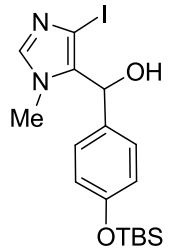
2-amino-1-cyclopentene-1-carbonitrile (140) was prepared using Martinez's protocol in 97% yield.[84]

4-(*t*-butyldimethylsilyloxy)benzyl bromide (144) was prepared in two steps, first by reducing the aldehyde **146** to corresponding benzyl alcohol, second by brominating this alcohol with PBr_3 in Et_2O.[122, 154]

4-(*t*-butyldimethylsilyloxy)benzaldehyde (146) was prepared following the Sundberg's procedure.[86]

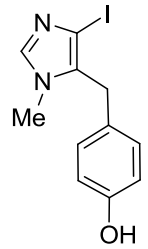
4,5-Diiodo-1-methyl-1*H*-imidazole (**81**) was prepared starting from imidazole in two steps following literature report.[155]

5-(4-*t*-Butyldimethylsiloxyphenyl)hydroxymethyl-4-iodo-1-methyl-*1H*-imidazole (147):



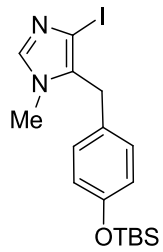
A 3.0 M solution of EtMgBr in ether (1.10 mL, 3.3 mmol) was added to a solution of **81** (1.00 g, 3.0 mmol) in dry CH_2Cl_2 (10 mL) at rt over 5 min. The resulting mixture was stirred at rt. for 20 min and benzaldehyde **146** (0.78 g, 3.3 mmol) was added. After stirring for 24 h, half saturated NH_4Cl (20 mL) was added to the reaction and the resulting white solid was dissolved in EtOAc and the layers were separated. The organic layer was dried over anhyd. Na_2SO_4 and concentrated to provide a pale yellow solid which was recrystallized from CH_2Cl_2/MeOH to isolate **147** (1.32g, 99%) as a white solid: m.p = 202-205 °C; ^{1}H NMR (DMSO-d_6): δ = 7.58 (s, 1H), 7.13 (d, *J* = 8.7 Hz, 2H), 6.83 (d, *J* = 8.7 Hz, 2 H), 6.19 (d, *J* = 4.1 Hz, 1H), 5.79 (d, *J* = 4.1 Hz, 1H), 3.33 (s, 3H), 0.93 (s, 9H), 0.17 (s, 6H); ^{13}C NMR (DMSO-d_6): δ = 154.5, 141.9, 135.3, 135.2, 127.0, 120.1, 85.7, 66.4, 33.1, 26.1, 18.5, -4.0; IR (KBr, cm^{-1}) = 3418 (br), 3128, 2956, 2930, 2858, 1605, 1508, 1258, 1157, 1040, 916, 840, 784; HR-ESIMS (*m/z*): Calcd. for $C_{17}H_{26}IN_2O_2Si$ $[M+H]^+$ 445.0803, found 445.0790; Calcd. for $C_{17}H_{25}IN_2NaO_2Si$ $[M+Na]^+$ 467.0622, found 467.0608; Calcd. for $C_{34}H_{50}I_2N_4NaO_4Si_2$ $[2M+Na]^+$ 911.1352, found 911.1354.

5-(4-hydroxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (148):



Et_3SiH (8.00 mL, 50.2 mmol) and TFA (4.64 mL, 60.2 mmol) were added to a solution of **147** (4.46 g, 10.0 mmol) in anhydrous $CHCl_3$ (50 mL) at rt. Then the resulting mixture was heated at 55-60 °C for 30 h under nitrogen and quenched by addition of satd. aq. solution of $NaHCO_3$. The resulting yellow solid was then dissolved in acetone and the acetone layer was extracted with $CHCl_3$ several times until the yellow color disappeared in aqueous layer. The combined organic solutions were dried over anhyd. Na_2SO_4 and concentrated to give a pale yellow solid, which was triturated with hexanes to give the desilylated phenol **148** (3.15 g, 96%): m.p = 179-181 °C; ^{1}H NMR (DMSO-d_6): δ = 9.26 (s, 1H), 7.56 (s, 1H), 6.89 (d, *J* = 8.5 Hz, 2H), 6.65 (d, *J* = 8.5 Hz, 2 H), 3.78 (s, 2H), 3.41 (s, 3H); ^{13}C NMR (DMSO-d_6): δ = 156.4, 140.5, 133.8, 129.4, 128.4, 115.9, 85.2, 32.6, 29.3; IR (KBr, cm^{-1}) = 3422 (br), 2802, 2680, 2602, 1611, 1514, 1380, 1275, 1240, 1151, 990, 830, 766; HR-ESIMS (*m/z*): Calcd. for $C_{11}H_{12}IN_2O$ $[M+H]^+$ 314.9989, found 314.9992; Calcd. for $C_{11}H_{11}IN_2NaO$ $[M+Na]^+$ 336.9808, found 336.9800.

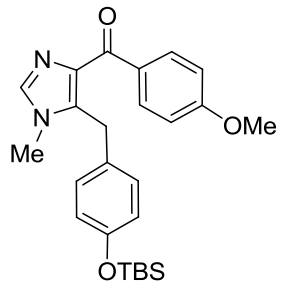
5-(4-*t*-Butyldimethylsilyloxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (80):



NaH (60%, 1.67 g, 10.6 mmol) was added to a solution of **148** (3.02 g, 9.6 mmol) in dry THF (30 mL) at rt. and the mixture was heated at 50 °C for 1.5 h, then the reaction was allowed to come to rt. *t*-Butyldimethylsilyl chloride (TBSCl) (1.59 g, 10.6 mmol) was added and the mixture stirred for 24 h at rt. The reaction mixture was diluted with hexanes and washed once with water, and brine. The organic layer was dried over anhyd.

Na_2SO_4 and concentrated to give a pale yellow solid, which was recrystallized from $CHCl_3$/MeOH to afford **80** as a white solid (2.86 g, 70%): m.p = 202-205 °C; ^{1}H NMR (DMSO-d_6): δ = 7.61 (s, 1H), 7.00 (d, *J* = 8.7 Hz, 2H), 6.78 (d, *J* = 8.7 Hz, 2 H), 3.87 (s, 2H), 3.44 (s, 3H), 0.93 (s, 9H), 0.16 (s, 6H); ^{13}C NMR (DMSO-d_6): δ = 154.2, 140.6, 133.6, 131.1, 129.6, 120.5, 85.2, 32.6, 29.3, 26.1, 18.4, -4.0; IR (KBr, cm^{-1}) = 3077, 2929, 2857, 1610, 1510, 1471, 1269, 1174, 983, 838, 781; HR-ESIMS (*m/z*): Calcd. for $C_{17}H_{26}IN_2OSi$ $[M+H]^+$ 429.0854, found 429.0857.

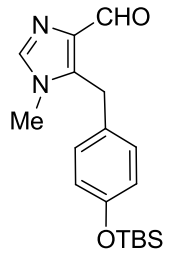
{5-(4-*t*-Butyldimethylsilyloxybenzyl)-1-methyl-1*H*-imidazol-4-yl}-(4-methoxy)phenylmethanone (149):



A 3.0 M solution of EtMgBr in ether (0.09 mL, 0.3 mmol) was added dropwise to a solution of **80** (104 mg, 0.2 mmol) in dry THF (2 mL) at rt. The resulting mixture was stirred at rt. for 15 min and *p*-anisaldehyde **53** (0.20 mL, 1.6 mmol) was added. After stirring at 40 °C for 32 h, half saturated NH_4Cl was added to quench the reaction and the organic layer was extracted with EtOAc, dried (Na_2SO_4) and concentrated to afford the crude product which was purified a short plug of silica gel (EtOAc/hexanes, 2:3) to isolate **149** (48.2 mg, 46%) as a pale yellow oil: ^{1}H NMR: δ = 8.28 (d, *J* = 8.7 Hz, 2H), 7.39 (s, 1H), 7.05 (d, *J* = 8.2 Hz, 2H), 6.95 (d, *J* = 8.7 Hz, 2H), 6.73 (d, *J* = 8.2 Hz, 2H), 4.40 (s, 1H), 3.85 (m, 3H), 3.44 (s, 3H), 0.95 (s, 9H), 0.15 (s, 6H); ^{13}C NMR δ = 188.2, 162.8, 154.4, 138.7, 138.1, 136.6, 132.9, 131.6, 130.0, 129.4, 120.3, 113.3, 55.5, 31.7, 29.2, 25.7, 18.2, -4.35; IR (neat, cm^{-1}) = 2956, 2930, 2858, 1630, 1601, 1538, 1509, 1464, 1367, 1254, 1170, 903, 841, 783; HR-ESIMS (*m/z*): Calcd. for $C_{25}H_{33}N_2O_3Si$ $[M+H]^+$ 437.2255,

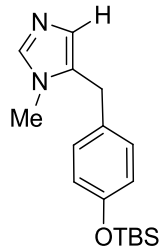
found 437.2306, Calcd. for $C_{25}H_{32}N_2O_3SiNa$ $[M+Na]^+$ 459.2074, found 459.2108, Calcd. for $C_{50}H_{64}N_4NaO_6Si_2$ $[2M+Na]^+$ 859.4257, found 859.4179.

5-(4-*t*-Butyldimethylsilyloxybenzyl)-1-methyl-1*H*-imidazole-4-carbaldehyde (152):



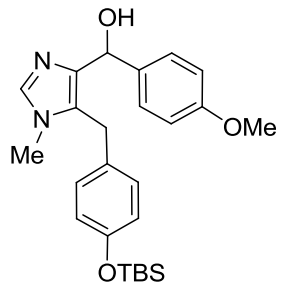
A 3.0 M solution of EtMgBr in ether (1.43 mL, 4.3 mmol) was added to a solution of **80** (1.79 g, 4.2 mmol) in dry THF (40 mL) at rt. The resulting mixture was stirred at rt. for 30 min and then *N*-methyl-*N*-(2-pyridyl)formamide, **151** (0.50 mL, 4.3 mmol) was added. After stirring for 2 h at rt., half saturated NH_4Cl was added to quench the reaction and the organic layer was extracted with EtOAc, dried (Na_2SO_4) and concentrated to provide a crude product, which was purified through a short plug of silica gel (EtOAc/hexanes, 2:3) to isolate **152** as an off-white solid (1.18 g, 78%): m.p = 78-80 °C; ^{1}H NMR: δ = 10.00 (s, 1H), 7.42 (s, 1H), 6.97 (d, *J* = 8.5 Hz, 2H), 6.73 (d, *J* = 8.5 Hz, 2 H), 4.33 (s, 2H), 3.44 (s, 3H), 0.96 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 187.7, 154.7, 138.8, 138.3, 137.8, 129.3, 129.0, 120.5, 31.6, 28.6, 25.7, 18.3, -4.3; IR (KBr, cm^{-1}) = 2929, 2891, 2821, 1680, 1605, 1552, 1508, 1466, 1254, 1206, 902, 845, 782; HR-ESIMS (*m/z*): Calcd. for $C_{18}H_{27}N_2O_2Si$ $[M+H]^+$ 331.1836, found 331.1906; Calcd. for $C_{18}H_{26}N_2NaO_2Si$ $[M+Na]^+$ 353.1656, found 353.1692; Calcd. for $C_{36}H_{52}N_4NaO_4Si_2$ $[2M+Na]^+$ 683.3419, found 683.3401.

5-(4-*t*-Butyldimethylsilyloxybenzyl)-1-methyl-1*H*-imidazole (153):



From above reaction, **153** was isolate as a light brown solid (250 mg, 20%): m.p = 78-80 °C; ^{1}H NMR: δ = 7.34 (s, 1H), 6.96 (d, *J* = 8.5 Hz, 2H), 6.78 (s, 1H), 6.73 (d, *J* = 8.5 Hz, 2 H), 3.823 (s, 2H), 3.35 (s, 3H), 0.95 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 154.3, 138.1, 130.8, 130.5, 129.4, 127.7, 120.3, 31.5, 29.5, 25.7, 18.2, -4.3; IR (KBr, cm^{-1}) = 2955, 2930, 2857, 1509, 1258, 916, 839, 781; HR-ESIMS (*m/z*): Calcd. for $C_{17}H_{27}N_2OSi$ $[M+H]^+$ 303.1887, found 303.1896.

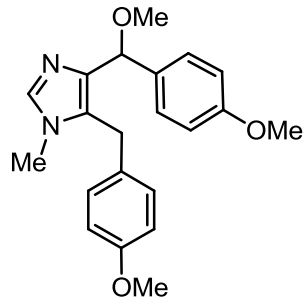
5-(4-*t*-Butyldimethylsilyloxybenzyl)-4-[hydroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (77):



A few drops of *p*-bromoanisole (from 3.25 mL, 26.0 mmol) was added dropwise to a two-necked round-bottom flask containing freshly-crushed, oven-dried magnesium turnings (0.62 g, 26.0 mmol) and a small crystal of iodine in THF (20 mL). This mixture was then heated at 45 °C under nitrogen until the iodine color faded. The remaining *p*-bromoanisole was added dropwise over 10 min while heating at the same temperature. After the addition was completed, the mixture was heated to reflux for 1 h and cooled to rt. Then, a solution of **152** (1.08 g, 3.3 mmol) in THF (10 mL) was added. The resulting mixture was stirred at reflux for 7 h and cooled to 0 °C; saturated aqueous NH_4Cl (20 mL) was added cautiously and the organic layer was extracted with EtOAc (3x30 mL), washed once with brine, and dried over anhyd. Na_2SO_4, concentrated to give thick brown oil, which was purified by a short plug of silica gel (EtOAc/hexanes, 4:1) to isolate **77**

(1.42 g, 100%) as a pale yellow solid: m.p = 108-109 °C; ^{1}H NMR: δ = 7.36 (s, 1H), 7.33 (d, *J* = 8.5 Hz, 2H), 6.83-6.82 (d, *J* = 8.5 Hz, 4 H), 6.68 (d, *J* = 8.5 Hz, 2H), 5.74 (s, 1H), 3.81 (s, 2H), 3.77 (s, 3H), 3.32 (s, 3H), 0.96 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 158.8, 154.3, 141.3, 136.9, 136.3, 130.6, 129.1, 127.9, 126.0, 120.3, 113.7, 69.5, 55.3, 31.7, 28.3, 25.8, 18.3, -4.3; IR (neat, cm^{-1}) = 3189 (br), 3001, 2955, 2858, 1609, 1509, 1467, 1251, 1172, 1037, 1007, 840, 756; HR-ESIMS (*m/z*): Calcd. for $C_{25}H_{35}N_2O_3Si$ $[M+H]^+$ 439.2411, found 439.2390; Calcd. for $C_{25}H_{34}N_2NaO_3Si$ $[M+Na]^+$ 461.2207, found 461.2207; Calcd. for $C_{50}H_{70}N_4NaO_6Si_2$ $[2M+Na]^+$ 899.4570, found 899.4611.

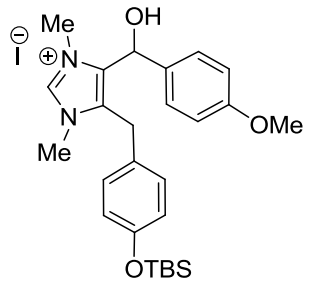
5-(4-Methoxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (155):



NaH (60% in mineral oil, 45.23 mg, 1.1 mmol) was added to a solution of alcohol **77** (451 mg, 1.0 mmol) in THF (10 mL) at rt. After stirring for 1.5 h, the reaction was cooled to 0 ºC and MeI (0.07 mL, 1.1 mmol) was added. Then, the reaction was allowed to come to rt. slowly and stirred overnight; water (2 mL) was added to the reaction and the organic layer was extracted with EtOAc (3x5 mL). Combined organic layers were dried (Na_2SO_4) and concentrated to provide crude material, which was purified over silica gel (1:3 hexane: EtOAc) to isolate **155** (146 mg, 40%) as a pale yellow oil: ^{1}H NMR: δ = 7.33 (d, *J* = 8.7 Hz, 2H), 7.22 (s, 1H), 6.86 (d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.3 Hz, 2H), 6.71 (d, *J* = 8.3 Hz, 2H), 5.26 (s, 1 H), 3.87 (s, 2H), 3.67 (s, 3H), 3.66 (s, 3H), 3.28 (s, 3H), 3.19 (s, 3H); ^{13}C NMR: δ = 158.8, 158.2,

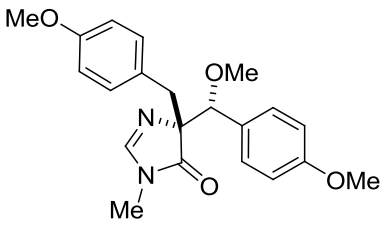
139.4, 137.1, 133.8, 130.2, 129.1, 128.2, 127.1, 114.0, 113.6, 79.5, 56.7, 55.2, 55.2, 31.5, 28.2; IR (neat, cm^{-1}) = 2934, 2834, 1611, 1510, 1246, 1176, 1087, 1033, 109; HR-ESIMS (*m/z*): Calcd. for $C_{21}H_{25}N_2O_3$ $[M+H]^+$ 353.1860, found 353.1864.

4-(4-*t*-Butyldimethylsilanyloxybenzyl)-1,3-dimethyl-5-[hydroxy-(4-methoxyphenyl)]methyl- 3*H*-imidazol-1-ium iodide (156):



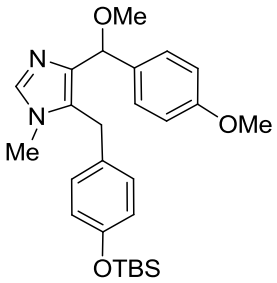
MeI (0.03 mL, 0.4 mmol) was added to a solution of **77** (83 mg, 0.2 mmol) and K_2CO_3 (52 mg, 0.4 mmol) in dry DMF (1 mL) at rt. and the reaction was stirred for 12 h. Water was added to the resulting yellow solution and the organic layer was extracted CH_2Cl_2 (3x5 mL). Combined organic layers were dried over anhyd. Na_2SO_4, concentrated in *vacuo* and resulting crude material was purified over silica gel (EtOAc) to afford **156** (110 mg, 64%) as a light brown oil: ^{1}H NMR: δ = 9.59 (s, 1H), 7.15 (d, *J* = 8.5 Hz, 2H), 6.95 (d, *J* = 8.5 Hz, 2H), 6.79 (d, *J* = 8.5 Hz, 2H), 6.73 (d, *J* = 8.5 Hz, 2H), 6.23 (d, *J* = 4.4 Hz, 1H), 4.94 (d, *J* = 4.4 Hz, 1H), 4.22 (d, *J* = 17.1 Hz, 1H), 3.94 (d, *J* = 17.1 Hz, 1H), 3.74 (s, 3H), 3.70 (s, 3H), 3.65 (s, 3H), 0.93 (s, 9H), 0.14 (s, 6H); ^{13}C NMR: δ = 159.4, 155.0, 136.8, 133.6, 131.8, 130.8, 129.4, 127.8, 127.2, 120.8, 114.2, 64.1, 55.4, 35.8, 34.4, 28.7, 25.7, 18.3, -4.3; IR (neat, cm^{-1}) = 3302, 2932, 2858, 1609, 1579, 1510, 1254, 1174, 1033, 913, 839; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{37}N_2O_3Si$ $[M]^+$ 453.2568, found 453.2607.

5-(4-Methoxy-benzyl)-5-[methoxy-(4-methoxy-phenyl)-methyl]-3-methyl-3,5-dihydro-imidazol-4-one (157):



Oxaziridine **88** (283 mg, 0.9 mmol) was added to a solution of **155** (132 mg, 0.4 mmol) in chloroform at rt. and the reaction was stirred at 40 ºC overnight. After removing the solvent, crude material was purified over silica gel (4:6→4:3, EtOAc: hexanes) to isolate the mixture of diastereomers and the recystallization of the mixture with EtOAc: benzene (1:1) provided **157** (78 mg, 56%) as a white solid: m.p = 166-168 ºC; ^{1}H NMR: δ = 7.21 (d, *J* = 8.5 Hz, 2H), 7.12 (s, 1H), 7.04 (d, *J* = 8.5 Hz, 2H), 6.79 (d, *J* = 8.8 Hz, 2H), 6.69 (d, *J* = 8.8 Hz, 2H), 4.49 (s, 1 H), 3.78 (s, 3H), 3.73 (s, 3H), 3.35 (d, *J* = 13.2 Hz, 1H), 3.28 (s, 3H), 3.07 (d, *J* = 13.2 Hz, 1 H), 2.45 (s, 3H); ^{13}C NMR: δ = 181.8, 159.5, 158.4, 153.7, 131.4, 129.5, 127.9, 127.2, 113.1, 85.4, 80.2, 57.5, 55.2, 55.2, 39.0, 26.7; IR (KBr, cm^{-1}): = 3060, 2980, 2930, 2829, 1720, 1609, 1531, 1463, 1291, 1251, 1178, 1098, 841, 719,619; HR-ESIMS (*m/z*): Calcd. for $C_{21}H_{24}N_2NaO_4$ $[M+Na]^+$ 391.1628, found 391.1577.

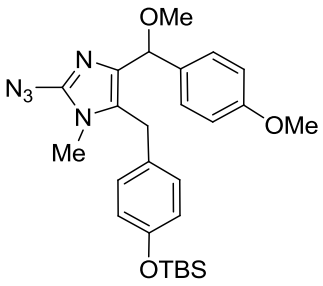
5-(4-*t*-Butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (76):



TFA (0.80 mL, 10.2 mmol) was added to a solution of **77** (1.12 g, 2.6 mmol) in anhyd MeOH (30 mL) at rt. and the resulting solution was then heated at 40 °C for 7 h. On completion of the reaction, the organic layer was washed

with saturated aqueous solution of $NaHCO_3$ (2x 20 mL). After ensure the pH of the solution was neutral, the organic layer was extracted with EtOAc, washed once with water, brine and dried over anhyd. Na_2SO_4, concentrated to give **76** (1.15 g, quantitative) as a colorless oil: ^{1}H NMR: δ = 7.37 (d, *J* = 8.7 Hz, 3H), 6.87-6.82 (m, 4H), 6.70 (d, *J* = 8.7 Hz, 2H), 5.30 (s, 1H), 3.93 (s, 2H), 3.77 (s, 3H), 3.34 (s, 3H), 3.30 (s, 3H), 0.96 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 158.8, 154.2, 139.6, 137.1, 133.7, 130.9, 129.1, 128.2, 127.1, 120.2, 113.7, 79.5, 56.8, 55.3, 31.6, 28.4, 25.7, 18.3, -4.4; IR (neat, cm^{-1}) = 2930, 2856, 1609, 1509, 1465, 1251, 1172, 1090, 1035, 916, 839, 782; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{37}N_2O_3Si$ $[M+H]^+$ 453.2568, found 453.2534.

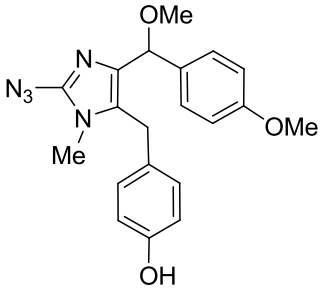
2-Azido-5-(4-*t*-butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (158):



n-Butyl lithium (1.6 M solution in hexanes, 1.42 mL, 2.3 mmol) was added in portions to a stirred solution of **76** (920 mg, 2.1 mmol) in dry THF (20 mL) at -78 °C (dry ice/ acetone). The reaction was stirred for 40 min at the same temperature. Then TsN_3 (485 mg, 2.5 mmol) in THF (2 mL) was added dropwise and stirred for 1 h at the same temperature. Then the reaction was quenched with satd. aq. solution of NH_4Cl (5 mL). Aqueous layer was extracted with EtOAc (3x15 mL), combined layers were dried (Na_2SO_4) and concentrated to provide a pale brown oil, which was purified over a short plug of silica gel (EtOAc: hexanes = 1:9) to isolate **158** (637 mg, 63%) as a pale brown oil: ^{1}H NMR: δ = 7.41 (d, *J*= 8.5 Hz, 2H), 6.87-6.83 (m, 4 H), 6.70 (d, *J*= 8.5

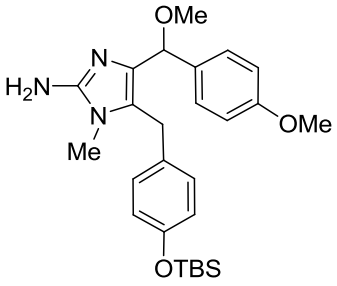
Hz, 2H), 5.22 (s, 1H), 3.85 (s, 2H), 3.78 (s, 3H), 3.35 (s, 3H), 3.04 (s, 3H), 0.97 (s, 9H), 0.17 (s, 6H); ^{13}C NMR: δ = 159.0, 154.3, 139.8, 136.8, 133.4, 130.6, 129.0, 128.4, 126.1, 120.3, 113.7, 79.1, 56.9, 55.3, 29.4, 28.7, 25.8, 18.3, -4.3; IR (neat, cm^{-1}) = 2931, 2857, 2138, 1609, 1508, 1253, 1170, 1091, 913, 838; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{35}N_5NaO_3Si$ $[M+Na]^+$ 516.2401, found 516.2364.

2-Azio-5-(4-hydroxybenzyl)-4-[methoxy-(4-methoxy)phenyl]methyl-1-methyl-1*H*-imidazole (159):



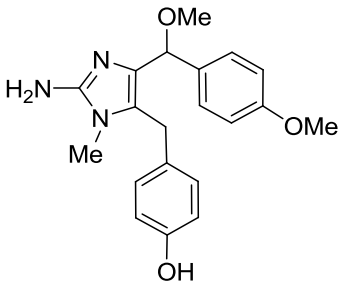
TBAF (1.0 M solution in THF, 0.85 mL, 0.9 mmol) was added to a solution of azide, **158** (380 mg, 0.8 mmol) in THF (20 mL) at rt. and stirred until the starting material was consumed as indicated by TLC. Then, an aqueous saturated solution of NH_4Cl (10 mL) was added to the above reaction, and the aqueous layer was extracted with EtOAc. The organic layer was washed with water (2x10 mL), once with saturated aqueous solution of brine, dried with Na_2SO_4, and concentrated in *vacuo*. Crude material was purified through a short plug of silica gel using 30% EtOAc in hexanes to isolate a reddish brown solid, **159** (231 mg, 80%): m.p = 71-74 °C; ^{1}H NMR: δ = 7.37 (d, *J* = 8.7 Hz, 2H), 6.81 (d, *J* = 8.7 Hz 4H), 6.67 (d, *J* = 8.2 Hz, 2H), 5.25 (s, 1H), 3.83 (s, 2H), 3.77 (s, 3H), 3.33 (s, 3H), 3.04 (s, 3H); ^{13}C NMR: δ = 160.0, 155.1, 139.9, 136.6, 133.2, 129.2, 129.1, 128.3, 126.3, 115.8, 113.8, 79.1, 56.9, 55.3, 29.5, 28.6; IR ($CHCl_3$, cm^{-1}) = 2932, 2858, 2139, 1611, 1510, 1248, 1172, 1080, 1033, 833, 756; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{21}N_5NaO_3$ $[M+Na]^+$ 402.1537, found 402.1573.

2-Amino-5-(4-*t*-butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)] methyl-1-methyl-1*H*-imidazole (160):



Azide **158** (454 mg, 0.9 mmol) was mixed with 10% Pd-C (35 wt%) in MeOH in a round bottom flask sealed with a rubber septum. Then, a balloon filled with H_2 was used to briefly purge the reaction, and the reaction was stirred at rt. for 1 h. Then, the mixture was filtered through a pad of Celite, and the Celite pad was washed with EtOAc (20 mL), and the solvent was removed in *vacuo* to isolate amine **160** as a pale yellow solid: (428 mg, quantitative): m.p = 121-124 °C; ^{1}H NMR: δ = 7.35 (d, *J* = 8.7 Hz, 2H), 6.88 (d, *J* = 8.7 Hz, 2H), 6.81 (d, *J* = 8.7 Hz, 2H), 6.70 (d, *J* = 8.2 Hz, 2H), 5.16 (s, 1H), 4.42 (br, 1H), 3.82 (s, 2H), 3.76 (s, 3H), 3.32 (s, 3H), 3.02 (s, 3H), 0.96 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 158.9, 154.2, 147.5, 133.7, 132.7, 131.1, 129.0, 128.2, 122.6, 120.2, 113.7, 78.6, 56.7, 55.3, 29.2, 28.7, 25.7, 18.3, -4.3; IR (KBr, cm^{-1}) = 3392, 3301, 3119, 2932, 2858, 1646, 1611, 1554, 1509, 1255, 1172, 1092, 915, 835, 782; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{38}N_3O_3Si$ $[M+H]^+$ 468.2677, found 468.2697; Calcd. for $C_{26}H_{37}N_3NaO_3Si$ $[M+Na]^+$ 490.2496, found 490.2542; Calcd. for $C_{52}H_{74}N_6NaO_6Si_2$ $[2M+Na]^+$ 957.5101, found 957.5173.

2-Amino-5-(4-hydroxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (14-methoxynaamine A) (75):

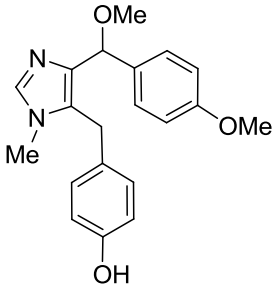


Following the above procedure azide **159** (537 mg, 1.4 mmol) and 10% Pd-C (30 wt%) were stirred in methanol (10 mL) to synthesize **75** (495 mg, quant.) as a pale yellow solid; m.p = 91-95 °C; ^{1}H NMR: (CD_3OH): δ = 7.29 (d, *J* = 8.7 Hz, 2H), 6.90 (d, *J* = 8.7 Hz 2H), 6.89 (d, *J* = 8.7 Hz, 2H), 6.70 (d, *J* = 8.7 Hz, 2H), 5.32 (s, 1H), 3.87 (d, *J* = 3.8 Hz, 2H), 3.77 (s, 3H), 3.34 (s, 3H), 3.19 (s, 3H); ^{13}C NMR: δ = 159.9, 156.3, 147.0, 130.8, 128.9, 127.8, 126.9, 124.3, 123.3, 115.4, 113.8, 75.1, 55.7, 54.5, 28.7, 27.0; IR (KBr, cm^{-1}) = 3548, 3475, 3417, 2996, 2934, 1614, 1512, 1247, 1174, 1114, 823, 618.; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{24}N_3O_3$ $[M+H]^+$ 354.1812, found 354.1827; Calcd. for $C_{20}H_{23}N_3NaO_3$ $[M+Na]^+$ 376.1632, found 376.1645.

Synthesis of 75 from 160:

A 1.0 M solution TBAF (1.0 M, 0.11 mL) in THF was added to a solution of amine **160** (42 mg, 0.1 mmol) in THF (1 mL) at rt. After stirring for 1 h, satd. aq. solution of NH_4Cl (3 mL) was added to the reaction and the organic layer was extracted with EtOAc (2x3 mL). The combined organic layers were washed with water (2x5 mL) and brine and dried over Na_2SO_4 workup described in the synthesis of **159** provided the crude material, which was purified over silica gel (EtOAc →MeOH) to isolate amine **75** (27 mg, 84%).

5-(4-Hydroxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (161):



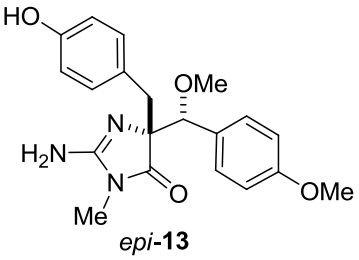
A 1.0 M solution of TBAF (3.78 mL, 3.8 mmol) was added to a solution of **76** (1.56 g, 3.4 mmol) in THF (30 mL) at rt. After stirring overnight and using the workup described in the synthesis of **75** from **160**, isolated the crude material, which was purified over silica gel (1:9, EtOAc: MeOH) to isolate **161** (1.08 g, 93%) as an off-white solid: m.p = 168-170 °C; ^{1}H NMR (Acetone-D6): δ = 8.43 (br, 1H), 7.36 (d, *J* = 8.5 Hz, 2H), 7.36 (s, 1H), 6.87 (d, *J* = 8.5 Hz 2H), 6.80 (d, *J* = 8.5 Hz, 2H), 6.69 (d, *J* = 8.5 Hz, 2H), 5.30 (s, 1H), 3.93 (s, 2H), 3.73 (s, 3H), 3.37 (s, 3H), 3.23 (s, 3H); ^{13}C NMR: δ = 158.8, 156.0, 139.0, 136.8, 134.5, 129.5, 129.2, 128.3, 127.6, 115.3, 113.1, 79.3, 55.6, 54.6, 30.8, 27.7; IR (KBr, cm^{-1}) = 3107, 2932, 2817, 2671, 2588, 1611, 1512, 1450, 1275, 1247, 1174, 1092, 820; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{23}N_2O_3$ $[M+H]^+$ 339.1703, found 339.1717; Calcd. for $C_{20}H_{22}N_2NaO_3$ $[M+Na]^+$ 361.1523, found 361.1537.

Synthesis of 159 from 161:

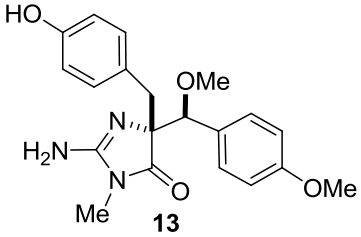
n-Butyl lithium (1.6 M solution in hexanes, 3.45 mL, 5.5 mmol) was added in portions to a stirred solution of **161** (845 mg, 2.5 mmol) in dry THF (25 mL) at -78 °C (dry ice/ acetone). The reaction was stirred for 30 min at the same temperature and the ice bathe was removed for 10 min. Then, the reaction was re-cooled to -78 ºC and TsN_3 (1.14 g, 5.8 mmol) in THF (5 mL) was added dropwise. The resulting mixture was stirred for 1 h at the same temperature followed by the same workup as above provided **159** (538 mg, 56%).

(*4R*, 8S) and (*4R*, 8R**)-2-Amino-4-(4-benzyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1,5-dihydroimidazol-5-one:**

[Calcaridine A (13) and *epi*-calcaridine A (*epi*-13)]



epi-**13**



13

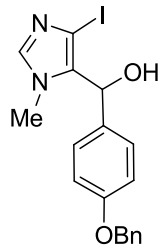
Amine **75** (175 mg, 0.5 mmol) and oxaziridine **88** (311 mg, 1.0 mmol) were dissolved in methanol (5 mL) at rt. Then, the mixture was heated at 40 °C for overnight. After cooling to rt. solvent was removed and crude material was purified by flash column chromatography using 10% methanol in EtOAc to isolate calcaridines (106 mg, 56%) as a 2:1 mixture of diastereomers (These two product could not be separated for full characterization at this stage);

[major product (*epi*-**13**)]: ^{1}H NMR (CD_3OH): δ = 7.19 (d, *J* = 8.8 Hz, 2H), 6.93 (d, *J* = 8.5 Hz, 2H), 6.83 (d, *J* = 8.8 Hz, 2H), 6.60 (d, *J* = 8.5 Hz, 2H), 4.46 (s, 1 H), 3.74 (s, 3H), 3.35 (d, *J* = 13.8 Hz, 1H), 3.26 (s, 3H), 3.06 (d, *J* = 13.8 Hz, 1 H), 2.40 (s, 3H); ^{13}C NMR δ = 175.3, 160.1, 157.4, 156.3, 130.9, 129.2, 127.2, 125.3, 114.5, 113.1, 84.9, 74.0, 56.2, 54.3, 38.7, 23.8.

[minor product (**13**)]: ^{1}H NMR (CD_3OH): δ = 7.33 (d, *J* = 8.8 Hz, 2H), 6.94 (d, 2H), 6.84 (d, *J* = 8.5 Hz, 2H), 6.57 (d, 2H), 4.44 (s, 1 H), 3.80 (s, 3 H), 3.34 (d, *J* = 13.8 Hz, 1H), 3.13 (s, 3H), 3.05 (d, *J* = 13.8 Hz, 1 H), 2.69 (s, 3H); ^{13}C NMR δ = 176.9, 160.3, 158.0, 156.4, 130.8, 129.4, 127.3, 124.5, 114.5, 113.5, 85.0, 73.8, 55.9, 54.4, 38.6, 24.1.

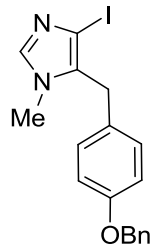
4-Benzoyloxybenzaldehyde (162) was prepared from 4-hydroxybenzaldehyde as reported by Haung.[96]

5-[(4-Benzyloxyphenyl)-hydroxy]methyl-4-iodo-1-methyl-1*H*-imidazole (163):



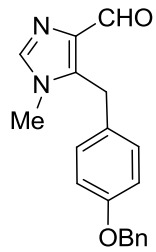
EtMgBr (3.0 M solution in ether, 2.31 mL, 6.9 mmol) was added to a solution of **81** (2.20 g, 6.6 mmol) in dry CH_2Cl_2 (15 mL) at rt. over ~5 min. The resulting mixture was stirred at rt. for 20 min, and 4-benzoyloxybenzaldehyde, **162** (1.54 g, 7.3 mmol) in dry CH_2Cl_2 (25 mL) was added and stirring was continued for overnight. Sat. aq. NH_4Cl (10 mL) was added to the reaction mixture and the resulting pale yellow solid was filtered and the filtrate was partitioned with CH_2Cl_2. The organic layer was dried (Na_2SO_4) and concentrated to provide a pale yellow solid. The resulting solid was triturated with hexanes, which was decanted from the residue to provide **163** (2.80 g, quant) as a white solid: m.p = 195-198 °C; ^{1}H NMR (DMSO-d_6): δ = 7.58 (s, 1H), 7.43 (d, *J* = 7.3 Hz, 2H), 7.38 (t, *J* = 7.3 Hz, 2 H), 7.32 (t, *J* = 7.3 Hz, 1H), 7.17 (d, *J* = 8.7 Hz, 2H), 6.99 (d, *J* = 8.7 Hz, 2H), 6.22 (d, *J* = 4.1 Hz, 1H), 5.80 (d, *J* = 4.1Hz, 1H), 5.07 (s, 2H), 3.38 (s, 3H); ^{13}C NMR: δ = 157.9, 141.9, 137.6, 135.4, 134.6, 129.0, 128.4, 128.3, 127.0, 115.1, 85.7, 70.0, 66.5, 33.2; IR (KBr, cm^{-1}): 3189 (br), 3034, 2948, 2874, 1607, 1506, 1387, 1236,1166, 1006, 971, 744, 699; HR-ESIMS (*m/z*): Calcd. for $C_{18}H_{18}IN_2O_2$ $[M+H]^+$ 421.0408, found 421.0392; Calcd. for $C_{18}H_{17}IN_2NaO_2$ $[M+Na]^+$ 443.0227, found 443.0194.

5-(4-Benzoyloxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (164):



Et_3SiH (5.36 mL, 33.6 mmol) and TFA (2.58 mL, 33.6 mmol) were added to a solution of **163** (2.82 g, 6.7 mmol) in anhydrous $CHCl_3$ (50 mL) at rt., then the resulting mixture was heated at 55-60 °C for 24 h. After cooling to rt., the reaction was quenched by the addition of sat. aq. solution of $NaHCO_3$. The resulting mixture was extracted with $CHCl_3$ several times and the combined extracts were dried (Na_2SO_4) and concentrated. The residue was purified by chromatography (hexane/EtOAc, 1:1) to isolate **164** as a thick colorless oil (1.61 g, 60%); ^{1}H NMR: δ = 7.42-7.30 (m, 6H), 7.03 (d, *J* = 8.7 Hz, 2H), 6.89 (d, *J* = 8.7 Hz, 2 H), 5.02 (s, 2H), 3.91 (s, 2H), 3.41 (s, 3H); ^{13}C NMR: δ = 157.8, 139.4, 137.0, 133.4, 129.6, 129.1, 128.7, 128.1, 1127.5, 115.2, 84.8, 70.2, 32.6, 30.0; IR (neat, cm^{-1}): = 3031, 2918, 1609, 1509, 1418, 1239, 1175, 1013, 816, 740, 697; HR-ESIMS (*m/z*): Calcd. for $C_{18}H_{18}IN_2O$ $[M+H]^+$ 405.0458, found 405.0470; Calcd. for $C_{18}H_{17}IN_2NaO$ $[M+Na]^+$ 427.0278, found 427.0247.

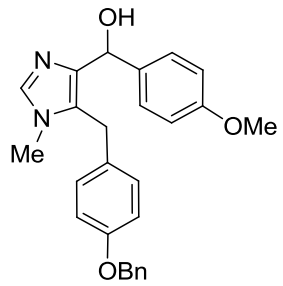
5-(4-Benzoyloxybenzyl)-1-methyl-1*H*-imidazole-4-carboxaldehyde (166):



EtMgBr (3.0 M in ether, 1.90 mL, 5.7 mmol) was added to a solution of **164** (2.19 g, 5.4 mmol) in dry THF (30 mL) at rt., and the resulting mixture was stirred for 20 min. *N*-methylformanilide, **165** (0.74 mL, 6.0 mmol) was added and the resulting mixture was stirred at rt. overnight. Then, satd. aq. NH_4Cl (10 mL) was added to quench the reaction and the organic layer was extracted with EtOAc, dried (Na_2SO_4) and concentrated to provide the crude product, which was purified through a short plug of

silica gel (hexane/EtOAc, 3:2) to isolate **166** as an off-white solid (1.09 g, 66%): m.p = 148-150 °C; ^{1}H NMR: δ = 10.01 (s, 1H), 7.51 (s, 1H), 7.41-7.30 (m, 5H), 7.05 (d, *J* = 8.5 Hz, 2H), 6.88 (d, *J* = 8.5 Hz, 2H), 5.02 (s, 2H), 4.34 (s, 2H), 3.47 (s, 3H); ^{13}C NMR: δ = 187.5, 157.9, 138.7, 138.0, 137.8, 136.9, 129.4, 128.7, 128.6, 128.1, 127.5, 115.3, 70.1, 31.7, 28.5; IR (KBr, cm^{-1}): = 3107, 3032, 2859, 1674, 1510, 1244, 1175, 799, 780, 740, 698; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{19}N_2O_2$ $[M+H]^+$ 307.1441, found 307.1462; Calcd. for $C_{19}H_{18}N_2NaO_2$ $[M+Na]^+$ 329.1260, found 329.1258.

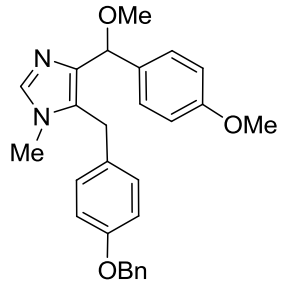
5-(4-Benzoyloxybenzyl)-4-[hydroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (167):



A few drops of *p*-bromoanisole (from 2.21 mL, 17.6 mmol) was added dropwise to a two-necked round-bottom flask containing freshly-crushed oven-dried, magnesium turnings (0.42 g, 17.6 mmol) and a small crystal of iodine in THF (20 mL). This mixture was then heated at 45 °C under nitrogen until the iodine color faded. The remainder of the *p*-bromoanisole was added dropwise over 10 min while maintaining the same temperature. After the addition was complete, the mixture was heated to reflux for 1 h and cooled to rt., then a solution of **166** (1.08 g, 3.5 mmol) in THF (10 mL) was added followed overnight stirring. After cooling to 0 °C, satd. aq. NH_4Cl (20 mL) was added carefully and the resulting mixture was extracted with EtOAc (3x50 mL), washed once with brine, dried (Na_2SO_4), and concentrated to give thick, brown oil. The crude product was purified through a short plug of silica gel (EtOAc) to isolate **167** as a pale yellow solid (1.47 g, 100%): m.p = 124-127 °C; ^{1}H NMR: δ = 7.42-7.29 (m, 8H), 6.89-6.76 (m, 6H), 5.78

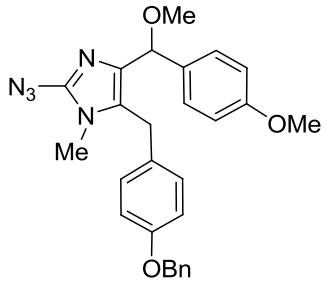
(s, 1H), 5.00 (s, 2H), 4.38 (br, 1H), 3.78 (s, 2H), 3.73 (s, 3H), 3.28 (s, 3H); ^{13}C NMR: δ = 158.8, 157.5, 140.9, 137.1, 137.0, 136.2, 130.2, 129.1, 128.7, 128.1, 128.1, 127.6, 125.9, 115.1, 113.7, 70.1, 69.5, 55.3, 31.8, 28.2; IR (KBr, cm^{-1}): = 3198 (br), 3031, 2932, 2835, 1611, 1584, 1509, 1454, 1302, 1244, 1175, 1035, 801, 752, 698; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{27}N_2O_3$ $[M+H]^+$ 415.2016, found 415.2016; Calcd. for $C_{26}H_{26}N_2NaO_3$ $[M+Na]^+$ 437.1856, found 437.1817.

5-(4-Benzoyloxybenzyl)-1-methyl-4-[methoxy-(4-methoxyphenyl)]methyl-1*H*-imidazole (168):



TFA (0.53 mL, 6.8 mmol) was added to a solution of **167** (1.42 g, 3.4 mmol) in anhyd. MeOH (20 mL) at rt. and the mixture was heated at 55 °C for overnight. Sat. aq. $NaHCO_3$ (20 mL) was added to the above reaction mixture and the aqueous layer was extracted with EtOAc (3x30 mL) and the organic layer was washed once with aq. $NaHCO_3$, once with water and brine. After drying (Na_2SO_4), the organic layer was concentrated to provide **168** (1.58 g, quant) as a pale yellow oil; ^{1}H NMR: δ = 7.51 (s, 1H), 7.42-7.29 (m, 7H), 6.92-6.76 (m, 6H), 5.30 (s, 1 H), 5.02 (s, 2H), 3.95 (s, 2H), 3.77 (s, 3H), 3.35 (s, 3H), 3.33 (s, 3H); ^{13}C NMR: δ = 158.9, 157.5, 139.5, 137.2, 137.1, 133.7, 130.5, 129.2, 128.7, 128.2, 128.1, 127.6, 127.1, 115.0, 113.7, 79.5, 70.1, 56.8, 55.3, 31.7, 28.3; IR (neat, cm^{-1}) = 3032, 2971, 2916, 1610, 1509, 1244, 1174, 1011, 804, 742; HR-ESIMS (*m/z*): Calcd. for $C_{27}H_{29}N_2O_3$ $[M+H]^+$ 429.2117, found 429.2176; Calcd. for $C_{27}H_{28}N_2NaO_3$ $[M+Na]^+$ 451.1992, found 451.1955.

2-Azido-5-(4-benzoyloxybenzyl)-1-methyl-4-[methoxy-(4-methoxyphenyl)]methyl-1*H*-imidazole (169):

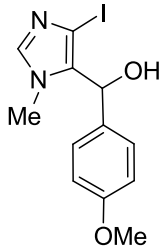


n-Butyl lithium (1.6 M solution in hexanes, 0.72 mL, 1.1 mmol) was added dropwise to a stirred solution of **168** (445 mg, 1.0 mmol) in dry THF (8 mL) at -78 °C. The reaction was stirred for 30 min at the same temperature. The cooling bath was removed for 10 min, then the reaction mixture was re-cooled to -78 °C and TsN_3 (246 mg, 1.3 mmol) in THF (1 mL) was added dropwise. After stirring for additional 20 min at -78 °C, the reaction mixture was allowed to come to rt. and stirred for 10 min. The reaction was quenched by the careful addition of satd. aq. NH_4Cl (3 mL). The aqueous layer was extracted with EtOAc (3x15 mL), and the combined organic extracts were dried (Na_2SO_4) and concentrated to form a pale brown oil, which was purified through a short column of silica gel (hexane/EtOAc, 4:1) to isolate unreacted starting material and **169** (283 mg, 58%) as a reddish brown oil: ^{1}H NMR: δ = 7.49-7.34 (m, 7H), 6.97 (d, *J* = 8.8 Hz, 2H), 6.89 (d, *J* = 8.8 Hz, 4H), 5.23 (s, 1 H), 5.02 (s, 2H), 3.85 (s, 2H), 3.78 (s, 3H), 3.35 (s, 3H), 3.04 (s, 3H); ^{13}C NMR: δ = 159.1, 157.6, 139.8, 137.1, 136.9, 133.4, 130.3, 129.2, 128.7, 128.5, 128.1, 127.6, 126.2, 115.1, 113.8, 79.2, 70.1, 56.9, 55.3, 29.5, 28.7. IR (neat, cm^{-1}): = 3032, 2933, 2835, 2137, 1610, 1509, 1244, 1173, 1088, 1033, 832, 744, 697; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{24}N_5O_2$ $[M+H-MeOH]^+$ 438.1925, found 438.1898; Calcd. for $C_{27}H_{27}N_5NaO_3$ $[M+Na]^+$ 492.2006, found 492.1977.

Synthesis of 75 from 169:

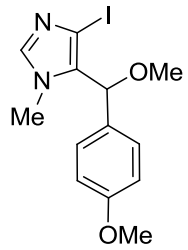
Azide **169** (258 mg, 0.6 mmol) was dissolved in EtOH (3 mL) and stirred under a hydrogen atmosphere (55 psi) in the presence of 20% $Pd(OH)_2$ on charcoal (77 mg) at rt. for overnight. The catalyst was filtered through a pad of Celite and the filtrate was concentrated to provide amine **75** (196 mg, quant) as a pale yellow solid; m.p = 91-95 °C.

4-[Hydroxy-(4-methoxyphenyl)]methyl-4-iodo-1-methyl-1*H*-imidazole (170):



A 3.0 M solution of EtMgBr in ether (3.31 mL, 9.9 mmol) was added into a solution of **81** (3.01 g, 9.0 mmol) in dry CH_2Cl_2 (30 mL) at rt. over 5 min. The resulting mixture was stirred at rt. for 45 min under nitrogen and *p*-anisaldehyde, **53** (0.86 mL, 9.9 mmol) in dry CH_2Cl_2 (10 mL) was added dropwise. After stirring at rt. for 16 h, half saturated NH_4Cl (50 mL) was added to it and the resulting white solid was dissolved in EtOAc and the layers were separated. The organic layer was dried over anhyd. Na_2SO_4 and concentrated and the resulting solid was triturated with hexanes to isolate **170** as a white solid (2.79 g, 90%): m.p = 124-125 °C; ^{1}H NMR (DMSO-d_6): δ = 7.55 (s, 1H), 7.13 (d, *J* = 8.8 Hz, 2H), 6.88 (d, *J* = 8.8 Hz, 2 H), 6.18-6.17 (d, *J* = 4.1 Hz, 1H), 5.78-5.77 (d, *J* = 4.1 Hz, 1H), 3.70 (s, 3H), 3.34 (s, 3H); ^{13}C NMR: δ = 158.7, 141.9, 135.4, 134.3, 126.9, 114.2, 85.7, 66.5, 55.6, 33.1; IR (KBr, cm^{-1}): = 3424 (br), 3158, 1608, 1510, 1242, 1032, 867, 838; HR-ESIMS (*m/z*): Calcd. for $C_{12}H_{14}IN_2O_2$ $[M+H]^+$ 345.0095, found 345.0086; Calcd. for $C_{12}H_{13}IN_2O_2Na$ $[M+Na]^+$ 366.9914, found 366.9904; Calcd. for $C_{24}H_{26}I_2N_4NaO_4$ $[2M+Na]^+$ 710.9936, found 710.9927.

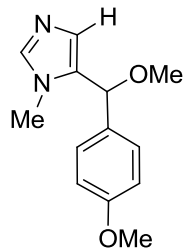
4-Iodo-1-methyl-5-[methoxy-(4-methoxyphenyl)]methyl-1*H*-imidazole (171):



NaH (60%, 0.21 g, 5.2 mmol) was added to a solution of **170** (1.65 g, 4.8 mmol) in dry THF (40 mL) at rt. and the mixture was heated at 60 ° C for 2.5 h. Then the reaction was allowed to come to rt. and MeI (0.33 mL, 5.2 mmol) was added dropwise followed by 24 h stirring. The reaction mixture was quenched by adding water (20 mL) and extracted with EtOAc (3x50 mL). The combined organic extracts were dried over anhyd. Na_2SO_4 and concentrated to provide a thick brown oil, which was purified through a short plug of silica gel (EtOAc/hexanes, 3:7) to isolate **171** and **172**.

Data for 171: a pale yellow oil (1.24 g, 72%); ^{1}H NMR: δ = 7.38 (s, 1H), 7.19 (d, *J* = 8.7 Hz, 2H), 6.85 (d, *J* = 8.7 Hz, 2 H), 5.52 (s, 1H), 3.79 (s, 3H), 3.38 (s, 3H), 3.33 (s, 3H); ^{13}C NMR: δ = 159.0, 141.4, 131.8, 131.2, 126.9, 113.9, 88.0, 76.4, 56.9, 55.4, 33.0; IR (KBr, cm^{-1}): = 2932, 1612, 1510, 1463, 1303, 1248, 1087, 1033, 951, 837, 784; HR-ESIMS (*m/z*): Calcd. for $C_{13}H_{16}IN_2O_2$ $[M+H]^+$ 359.0251, found 359.0261; Calcd. for $C_{13}H_{15}IN_2O_2Na$ $[M+Na]^+$ 381.0055, found 381.0076; Calcd. for $C_{26}H_{30}I_2N_4NaO_4$ $[2M+Na]^+$ 739.0249, found 739.0244.

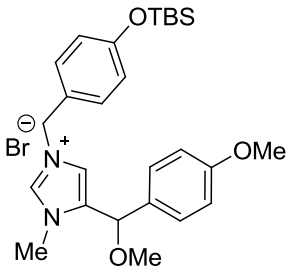
5-[Methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (172):



172: a light brown oil (0.22 g , 20%); ^{1}H NMR: δ = 7.65 (s, 1H), 7.28 (d, *J* = 8.7 Hz, 2H), 6.92 (d, *J* = 8.7 Hz, 2 H), 6.59 (s, 1H), 5.35 (s, 1H), 4.89 (s, 1H), 3.77 (s, 3H), 3.57 (s, 3H), 3.30 (s, 3H); ^{13}C NMR: δ = 159.4, 139.6, 131.5, 130.7, 130.1, 128.2, 113.8,

76.5, 56.4, 55.4, 32.2; IR (KBr, cm^{-1}): = 3373, 3109, 2934, 2833, 1610, 1511, 1463, 1248, 1174, 1079,1032, 828; HR-ESIMS (*m/z*): Calcd. for $C_{13}H_{17}N_2O_2$ $[M+H]^+$ 233.1285, found 233.1265.

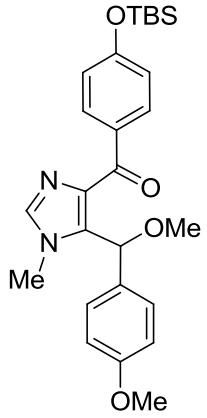
1-(4-*t*-Butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-3-methyl-3*H*-imidazol-1-ium bromide (173):



A 3.0 M solution of EtMgBr in ether (1.00 mL, 3.0 mmol) was added to a solution of **171** (1.00 g, 2.8 mmol) in dry CH_2Cl_2 (10 mL) at rt. The resulting mixture was stirred for 45 min, and 1.0 M solution of CuCN.2LiCl (3.20 mL) was added followed by bromide, **144** (1.00 g, 3.3 mmol) in dry CH_2Cl_2 (5 mL). The yellow reaction mixture was stirred at rt. for 15 h and poured into half saturated NH_4CI (30 mL) followed by stirring for 10 min. The resulting precipitated solid was filtered and the filtrate was extracted with CH_2C1_2. The combined organic extracts were dried (Na_2SO_4) and evaporated. The residue was purified by flash chromatography (2:3 EtOAc/hexanes) to yield **172** (0.38 g, 57%) and imidazolium bromide **173** (0.54 g, 35%) as a dark-green solid, which liquifies upon exposure to air; ^{1}H NMR: δ = 10.45 (br, 1H), 7.27 (d, *J* = 7.8 Hz, 2H), 7.22 (d, *J* = 8.2 Hz, 2H), 6.90 (d, *J* = 8.2 Hz, 2H), 6.79 (d, *J* = 7.8 Hz, 2H), 6.57 (s, 1H), 5.39 (d, *J* = 11.9 Hz, 1H), 5.30 (d, *J* = 11.9 Hz, 1H), 5.23 (s, 1H), 3.90 (s, 2H), 3.80 (s, 3H), 3.44 (s, 3H), 3.28 (s, 3H), 0.94 (s, 9H), 0.15 (s, 6H); ^{13}C NMR: δ = 160.4, 156.7, 138.5, 136.3, 130.6, 128.8, 127.2, 125.6, 121.0, 119.9, 114.6, 75.3, 56.8, 55.5, 53.0, 35.1, 25.7, 18.2, -4.4; IR (neat, cm^{-1}) = 3392 (br), 2932, 2858, 1610, 1512, 1463,

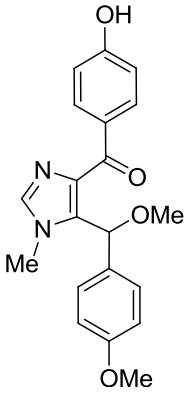
1252, 1173, 1083, 913, 838, 754; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{37}N_2O_3Si$ $[M]^+$ 453.2568, found 453.2591.

4-(*t*-Butyldimethylsilyloxyphenyl)-{5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazol-4-yl}methanone (174):



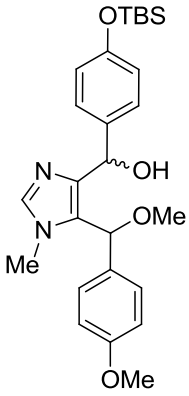
A 3.0 M solution of EtMgBr in ether (1.16 mL, 3.5 mmol) was added to a solution of **171** (1.14 g, 3.2 mmol) in dry CH_2Cl_2 (10 mL) at rt. over 5 min. After stirring the reaction for 30 min, benzaldehyde **146** (1.49 g, 6.3 mmol) in dry CH_2Cl_2 (5 mL) was added at rt. Then, the mixture was heated to reflux for 45 h, after which water was added to quench the reaction and the organic layer was separated, washed once with 10% Na_2SO_3 solution and brine. The organic layer was dried over anhyd. Na_2SO_4 and concentrated to provide the crude product, which was purified by column chromatography using gradient elution (EtOAc/hexanes 1:9→EtOAc) to isolate **174** (284 mg, 19%) as a pale brown oil: ^{1}H NMR: δ = 8.23 (d, *J* = 8.5 Hz ,2H), 7.39 (s,1H), 7.33 (d, *J* = 8.5 Hz, 2H), 6.91 (d, *J* = 8.5 Hz, 2 H), 6.85 (d, *J* = 8.5 Hz, 2 H), 6.54 (s, 1H), 3.79 (s, 3H), 3.49 (s, 3H), 3.44 (s, 3H), 0.99 (s, 9H), 0.23 (s, 6H); ^{13}C NMR: δ = 188.4, 159.8, 158.9, 140.2, 138.3, 137.8, 133.0, 131.7, 131.3, 127.1, 119.6, 113.8, 74.7, 57.4, 55.4, 33.4, 25.7, 18.3, -4.2. HR-DARTMS (m/z): Calcd. for $C_{26}H_{35}N_2O_4Si$ $[M+H]^+$ 467.2363, found 467.2363

4-Hydroxyphenyl-{5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazol-4-yl}methanone (175):



From above reaction, the desilylated product **175** was isolated as a white solid (60 mg, 5%); m.p = 197-200 °C; ^{1}H NMR (Acetone-D6): δ = 8.85 (d, *J* = 8.7 Hz, 2H), 8.06 (s, 1H), 7.80 (d, *J* = 8.7 Hz, 2H), 7.39-7.36 (m, 4H), 7.00 (s, 1H), 4.23 (s, 3H), 4.00 (s, 3H), 3.86 (s, 3H); ^{13}C NMR: δ = 187.0, 161.3, 159.0, 140.5, 138.6, 137.3, 133.5, 131.7, 130.4, 127.0, 114.5, 113.6, 74.7, 56.5, 54.7, 32.6; IR (KBr, cm^{-1}) = 2932, 1633, 1606, 1510, 1439, 1248, 1163, 1081, 910; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{20}N_2NaO_4$ $[M+Na]^+$ 375.1315, found 375.1314.

4-(4-*t*-Butyldimethylsilyloxyphenyl)hydroxymethyl-5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (176):

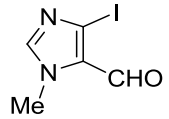


A 3.0 M solution of EtMgBr in ether (0.5 mL, 1.5 mmol) was added to a solution of **171** (420 mg, 1.2 mmol) in dry THF (5 mL) at rt. over 5 min. The resulting mixture was stirred for 15 min and benzaldehyde **146** (691 mg, 2.9 mmol) in dry THF (1 mL) was added. After completion of the addition, the mixture was heated to reflux for 45 h, at which time water was added to quench the reaction mixture at rt. and the organic layer was extracted with EtOAc, dried over anhyd. Na_2SO_4 and concentrated to provide the crude product which was purified through a short plug of silica gel (EtOAc/hexanes, 3:1) to isolate 2:1 diastereomeric **176** as a yellow solid (264 mg, 48%); m.p = 102-104 °C; ^{1}H NMR (common protons are integrated together, corresponding peak from minor product is underlined): δ =

7.35 (s, 1H), 7.29 (d, *J* = 8.5 Hz, 2H), 7.09, 7.00 (d, *J* = 8.8 Hz, 2H), 6.80 (d, *J* = 8.5 Hz, 2 H), 6.77-6.74 (m, 2 H), 5.82, 5.81 (s, 1H), 5.41, 5.39 (s, 1H), 3.78, 3.76 (s, 3H), 3.29 (s, 3H), 3.32, 3.07 (s, 3H), 0.95 (s, 9H), 0.14 (s, 6H); ^{13}C NMR (minor product shown within brackets): δ = 159.0 (158.9), 155.1 (155.0), 144.2 (144.5), 138.0 (138.2), 136.9, 131.6 (131.5), 128.1 (127.9), 127.4 (127.3), 125.1 (125.2), 120.0, 113.8 (113.7), 74.8 (74.5), 69.9 (69.7), 56.7 (56.7), 55.3 (55.3), 32.8, 25.8, 18.3, -4.3; IR (neat, cm^{-1}) = 3175 (br), 2932, 2858, 1609, 1508, 1251, 1087, 914; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{37}N_2O_4Si$ $[M+H]^+$ 469.2517, found 469.2534, Calcd. for $C_{26}H_{36}N_2O_4SiNa$ $[M+Na]^+$ 491.2337, found 491.2319, Calcd. for $C_{52}H_{72}N_4$ NaO_8Si_2 $[M+Na]^+$ 959.4781, found 959.4781.

In addition to the alcohol, reductive dehalogenated product **172** was obtained from the above reaction as light brown oil, (119 mg, 43%).

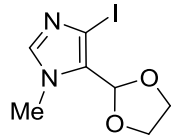
4-Iodo-1-methyl-1*H*-imidazole-5-carboxaldehyde (178):



A solution of EtMgBr (3.0 M in ether, 2.62 mL, 7.9 mmol) was added into a solution of **81** (2.50 g, 7.5 mmol) in dry CH_2Cl_2 (20 mL) at rt. over 10 min, under nitrogen atmosphere. After all starting material was reacted with the Grignard reagent (TLC), *N*-methylformanilide, **165** (1.01 mL, 8.2 mmol) was added dropwise to above mixture and stirred at rt. for a further 16 h. Half saturated NH_4Cl (10 mL) was added to the reaction and the resulting suspension was extracted with CH_2Cl_2 (3x20 mL). The organic layer was dried (Na_2SO_4) and concentrated; the resulting residue was purified by flash chromatography (EtOAc/hexanes, 1:4) to isolate **178** (1.09 g, 61%) as a white solid: m.p = 69-72 °C; ^{1}H NMR: δ = 9.62 (d, *J* = 0.5 Hz, 1H), 7.55 (s, 1H), 3.91 (s, 3H); ^{13}C

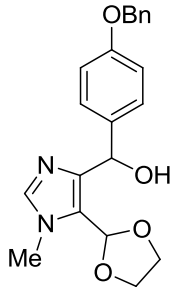
NMR: δ = 181.3, 145.0, 130.0, 100.3, 34.5; IR (KBr, cm^{-1}): = 2811, 1666, 1504, 1338, 1243, 964, 782, 707; HR-ESIMS (*m/z*): Calcd. for $C_5H_6IN_2O$ $[M+H]^+$ 236.9519, found 236.9527; Calcd. for $C_5H_5IN_2NaO$ $[M+Na]^+$ 258.9339, found 258.9362.

5-[1,3]Dioxolan-2-yl-4-iodo-1-methyl-1*H*-imidazole (179):



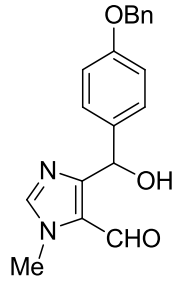
p-Toluenesulfonic acid monohydrate (140 mg, 0.7 mmol) and ethylene glycol (95%, 4.10 mL, 73.7 mmol) were added to a solution of **178** (3.48 g, 14.7 mmol) in toluene (75 mL). The reaction mixture was heated to reflux for 22 h with a Dean-Stark condenser fitted. The mixture was cooled to rt., and then the reaction mixture was washed with sat. $NaHCO_3$ (3x25 mL) and water. The resulting toluene solution was dried (Na_2SO_4), concentrated and purified by chromatography (hexane/EtOAc, 65:35) to provide **179** (4.02 g, 97%) as an off-white solid; m.p = 115-118 °C; ^1H NMR: δ = 7.39 (s, 1H), 5.79 (s, 1H), 4.15 (m, 2H), 4.04 (m, 2H), 3.69 (s, 3H); ^{13}C NMR: δ = 141.9, 127.0, 98.8, 87.8, 65.3, 33.2; IR (KBr, cm^{-1}): = 2951, 2887, 1578, 1494, 1473, 1370, 1245, 1217, 1085, 952, 815; HR-ESIMS (*m/z*): Calcd. for $C_7H_{10}IN_2O_2$ $[M+H]^+$ 280.9782, found 280.9791; Calcd. for $C_7H_9IN_2NaO_2$ $[M+Na]^+$ 302.9610, found 302.9612.

4-(4-Benzyloxyphenyl)hydroxymethyl-5-([1,3]dioxolan-2-yl)-1-methyl-1*H*-imidazole (180):



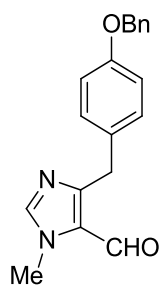
A solution of EtMgBr (3.0 M in ether, 2.67 mL, 8.0 mmol) was added to a solution of **179** (2.14 g, 7.6 mmol) in dry THF (20 mL) at rt. over 10 min. The resulting mixture was stirred at rt. until all the starting material reacted (TLC analysis, ca. 30 min) and then 4-benzyloxybenzaldehyde, **162** (1.78 g, 8.4 mmol) in dry THF (10 mL) was added followed by stirring for 38 h. Saturated aq. NH_4Cl (10 mL) was added to quench the reaction and the organic layer was extracted with EtOAc (3x30 mL), washed once with brine. The EtOAc solution was dried (anhyd. Na_2SO_4) and concentrated to provide the crude product, which was purified by column chromatography (EtOAc→EtOH/EtOAc, 1:9) to isolate **180** as a white solid (2.46 g, 87%): m.p = 160-162 °C; ^{1}H NMR: δ = 7.40-7.27 (m, 8H), 6.92 (d, *J* = 8.3 Hz, 2H), 5.90 (s, 1H), 5.84 (s, 1H), 5.03 (s, 2H), 4.00 (m, 2H), 3.92 (m, 2H), 3.66 (s, 3H); ^{13}C NMR: δ = 158.1, 145.0, 138.8, 137.2, 136.2, 128.6, 128.0, 127.9, 127.5, 121.8, 114.7, 97.3, 70.1, 69.5, 65.1, 33.0; IR (KBr, cm^{-1}): = 3176 (br), 3120, 2918, 1606, 1511, 1419, 1226, 1076, 1035, 951, 843, 698; HR-ESIMS (*m/z*): Calcd. for$C_{21}H_{23}N_2O_4$ $[M+H]^+$ 367.1652, found 367.1662; Calcd. for $C_{21}H_{22}N_2NaO_4$ $[M+Na]^+$ 389.1472, found 389.1469.

4-(4-benzyloxyphenyl)hydroxymethyl-1-methyl-1*H*-imidazole-5-carboxaldehyde (181a):



A solution of 10% HCl (10 mL) was added to the acetal **180** (2.23 g, 6.1mmol) in THF (100 mL) and the resulting cloudy reaction was heated at 55 °C (The reaction became clear while dissolving all solid after 10 min) while the reaction progress was monitored by taking 0.5 mL aliquots and neutralizing with satd. aq. $NaHCO_3$. The aqueous layer was extracted with EtOAc, and the organic layer was dried over anhyd. Na_2SO_4 and concentrated to provide the crude product, which was evaluated by ^1H NMR spectroscopy. After all starting material was consumed (2 h), the reaction was worked-up following the above procedure affording the pure aldehyde (1.95 g, quant), as a cream colored solid: m.p = 135-136 °C; ^1H NMR: δ = 9.90 (s, 1H), 7.46 (s, 1H), 7.41-7.31 (m, 7H), 6.94 (d, *J* = 8.7 Hz, 2 H), 6.01 (s, 1H), 5.03 (s, 2H), 3.87 (s, 3H); ^{13}C NMR: δ = 180.4, 158.7, 156.8, 142.1, 137.0, 135.1, 128.7, 128.1, 128.0, 127.5, 126.6, 115.1, 70.9, 70.1, 34.5; IR (KBr, cm^{-1}) = 3327 (br), 3088, 3009, 2862, 1655, 1513, 1352, 1297, 1245, 1045, 1014, 807, 786, 741, 715; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{19}N_2O_3$ $[M+H]^+$ 323.1390, found 323.1382; Calcd. for $C_{19}H_{18}N_2NaO_3$ $[M+Na]^+$ 345.1210, found 345.1198.

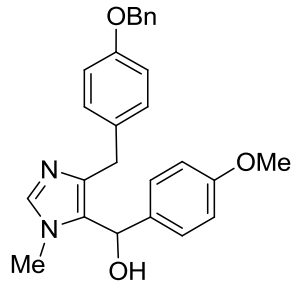
4-(4-Benzyloxybenzyl)-1-methyl-1*H*-imidazole-5-carbaldehyde (181b):



Et_3SiH (3.86 mL, 24.2 mmol) and TFA (2.80 mL, 36.3 mmol) were added to a solution of **181a** (1.95 g, 6.1 mmol) in anhydrous $CHCl_3$ (100 mL) under nitrogen atmosphere at rt. Then the resulting

mixture was stirred for 24 h while monitoring the reaction progress by TLC. The reaction was quenched with saturated aqueous $NaHCO_3$ solution. The organic layer was extracted with $CHCl_3$ (3x50 mL). Combined organic extracts were dried over anhyd. Na_2SO_4 and concentrated to provide a yellowish white solid, which was purified over silica gel with (EtOAc/hexanes, 3:1) to isolate **181** (1.21 g, 65%) as a pale yellow solid: m.p = 85-86 °C; ^{1}H NMR: δ = 9.85 (s, 1H), 7.47 (s, 1H), 7.40-7.28 (m, 5H), 7.18 (d, *J* = 8.7 Hz, 2 H), 6.90 (d, *J* = 8.7 Hz, 2 H), 5.01 (s, 2H), 4.12 (s, 2H), 3.85 (s, 3H); ^{13}C NMR: δ = 179.1, 157.6, 155.8, 142.9, 137.1, 131.3, 129.7, 128.6, 128.0, 127.5, 127.0, 115.2, 70.1, 34.4, 33.2; IR (KBr, cm^{-1}) = 3121, 3058, 3028, 2915, 2826, 2746, 1763, 1665, 1520, 1332, 1247, 1171, 1009, 845, 744, 699, 633; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{19}N_2O_2$ $[M+H]^+$ 307.1441, found 307.1444; Calcd. for $C_{19}H_{18}N_2NaO_2$ $[M+Na]^+$ 329.1266, found 329.1207.

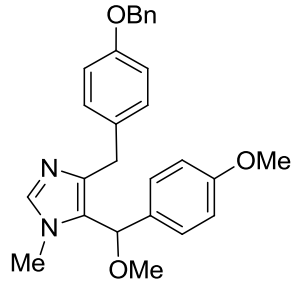
4-(4-Benzyloxybenzyl)-5-[hydroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (182):



A few drops of *p*-bromoanisole (from 1.98 mL, 15.8 mmol) were added dropwise to a two neck round-bottom flask containing freshly-crushed, oven-dried magnesium turnings (0.38 g, 15.8 mmol) and a small crystal of iodine in THF (25 mL). This mixture was then heated at 45 °C under nitrogen until the iodine color faded. The rest of the *p*-bromoanisole was added dropwise over 10 min while maintaining the same temperature. After the addition was completed, the mixture was heated at reflux for 1 h and cooled to rt. Then, a solution of **181b** (1.21 g, 3.9 mmol) in THF (10 mL) was added, and stirred at reflux for

overnight. After cooling the reaction mixture to 0 °C, sat. aq. NH_4Cl (20 mL) was added and the organic layer was extracted with EtOAc (3x50 mL), washed once with brine and dried over anhyd. Na_2SO_4. Concentration of the organic layer provided thick brown oil, which was purified by a short plug of silica gel (EtOAc) to isolate **182** (1.64 g, 84%) as a white solid: m.p = 148-149 °C; ^{1}H NMR: δ = 7.41-7.25 (m, 6H), 7.16 (d, *J* = 8.5 Hz, 2 H), 7.10 (d, *J* = 8.5 Hz, 2 H), 6.85 (d, *J* = 8.5 Hz, 2 H), 6.81 (d, *J* = 8.5 Hz, 2 H), 6.05 (s, 1H), 4.99 (s, 2H), 3.88 (s, 2 H), 3.78 (s, 3H), 3.34 (s, 3H); ^{13}C NMR: δ = 159.0, 157.3, 138.4, 137.2, 132.6_1, 132.5_5, 129.7, 129.0, 128.6, 128.0, 127.5, 127.0, 115.1, 113.9, 70.1, 65.5, 55.4, 33.4, 32.1; IR (KBr, cm^{-1}) = 3200 (br), 3115, 2998, 2908, 2834, 1611, 1510, 1459, 1238, 1173, 1032, 804, 697; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{27}N_2O_3$ $[M+H]^+$ 415.2016, found 415.2034; Calcd. for $C_{26}H_{26}N_2NaO_3$ $[M+Na]^+$ 437.1836, found 437.1819.

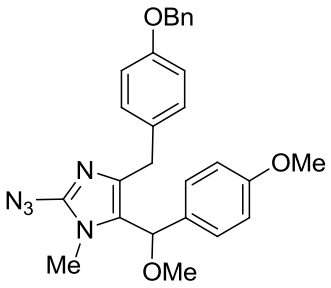
4-(4-Benzyloxybenzyl)-5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (183):



NaH (60%, 162 mg, 4.1 mmol) was added portionwise to a stirred mixture of alcohol **182** (1.12 g, 2.7 mmol) in anhydrous THF (25 mL) at 0 °C. After completion of the addition, the resulting mixture was stirred for 10 min at the same temperature. After the reaction mixture was stirred for 1.5 h at rt. MeI (0.20 mL, 3.2 mmol) was added at 0 °C and stirred for 10 min, then the reaction was stirred for 36 h at rt. Water (20 mL) was added and the aqueous layer was extracted with EtOAc (3x30 mL), the organic solution was dried (Na_2SO_4) and concentrated. the residue was purified through a short plug of silica gel

with (EtOAc/hexanes, 3:1) to provide **183** (0.97 g, 84%) as a pale yellow oil: ^{1}H NMR: δ = 7.40 (m, 2H), 7.36 (t, *J* = 7.3 Hz, 3H), 7.30 (t, *J* = 7.3 Hz, 1H), 7.20 (d, *J* = 8.3 Hz, 2 H), 7.10 (d, *J* = 8.7 Hz, 2 H), 6.88 (d, *J* = 8.3 Hz, 2 H), 6.81 (d, *J* = 8.7 Hz, 2 H), 5.48 (s, 1H), 5.01 (s, 2H), 3.94 (s, 2 H), 3.76 (s, 3H), 3.27 (s, 3H), 3.23 (s, 3H); ^{13}C NMR: δ = 158.9, 157.2, 142.1, 138.4, 137.3, 133.3, 132.0, 129.7, 128.6, 127.9, 127.5, 127.3, 125.2, 114.9, 113.8, 75.0, 70.1, 56.6, 55.3, 33.0, 32.7; IR (neat, cm^{-1}): = 3032, 2931 1609, 1509, 1246, 1174, 1086, 1031, 805, 741, 698; HR-ESIMS (*m/z*): Calcd. for $C_{27}H_{29}N_2O_3$ $[M+H]^+$ 429.2173, found 429.2181; Calcd. for $C_{27}H_{28}N_2NaO_3$ $[M+Na]^+$ 451.1992, found 451.1951.

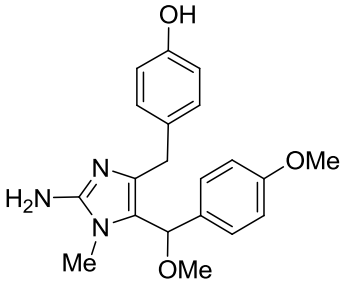
2-Azido-4-(4-benzyloxybenzyl)-5-[methoxy-(4-methoxy)phenyl]methyl-1-methyl-1*H*-imidazole (184):



n-Butyl lithium (1.33 M solution in hexane, 1.87 mL, 2.5 mmol) was added dropwise to a stirred solution of **183** (888 mg, 2.1 mmol) in dry THF (10 mL) at -78 °C. The reaction mixture was stirred for 45 min at the same temperature. Then, the dry ice/acetone bath was removed for 5 min followed by re-cooling to -78 °C and dropwise addition of TsN_3 (491 mg, 2.5 mmol). After 1 h stirring at -78 °C, the reaction was quenched by the careful addition of satd. aq. NH_4Cl (3 mL). The aqueous layer was extracted with EtOAc (3x25 mL), and the combined organics were dried (Na_2SO_4) and concentrated to afford a pale brown oil, which was purified through a short column of silica gel (hexane/EtOAc, 4:1) to isolate **184** (972 mg, 76%) as a thick, pale yellow oil: ^{1}H NMR: δ = 7.42-7.31 (m, 5H), 7.20 (d, *J* = 8.7 Hz, 2H), 7.07 (d, *J* = 8.7 Hz, 2H), 6.88

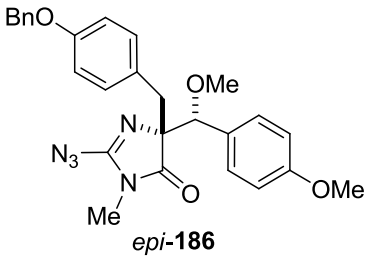
(d, *J* = 8.7 Hz, 2H), 6.79 (d, *J* = 8.7 Hz, 2 H), 5.37 (s, 1H), 5.03 (s, 2H), 3.90 (s, 2H), 3.78 (s, 3H), 3.21 (s, 3H), 3.02 (s, 3H); ^{13}C NMR: δ = 158.9, 157.3, 140.8, 139.6, 137.3, 133.0, 131.8, 129.7, 128.6, 128.0, 127.5, 127.3, 124.3, 114.9, 113.8, 75.0, 70.1, 56.5, 55.3, 32.8, 30.4; IR (neat cm^{-1}): = 2932, 2835, 2136, 1610, 1509, 1248, 1172, 1085, 1033, 833, 738, 697; HR-ESIMS (*m/z*): Calcd. for $C_{27}H_{28}N_5O_3$ $[M+H]^+$ 470.2187, found 470.2191; Calcd. for $C_{27}H_{27}N_5NaO_3$ $[M+Na]^+$ 492.2006, found 492.1969.

2-Amino-4-(4-hydroxybenzyl)-5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (185):

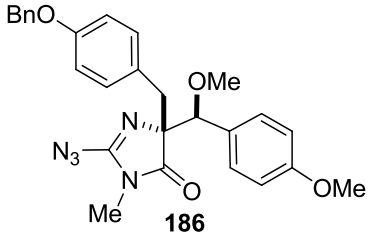


Azide **184** (100 mg, 0.3 mmol) was dissolved in EtOH (5 mL) and stirred overnight under a hydrogen atmosphere (55 psi) in the presence of 20% $Pd(OH)_2$ on charcoal (30 mg) at rt. The catalyst was filtered through a pad of Celite and the filtrate was concentrated to isolate amine **185** (91 mg, 97%) as pale yellow solid: m.p = 91-95 °C. ^{1}H NMR (CD_3OH): δ = 7.20 (d, *J* = 7.8 Hz, 2H), 7.06 (d, *J* = 7.3 Hz 2H), 6.88 (d, *J* = 7.8 Hz, 2H), 6.74 (d, *J* = 7.3 Hz, 2H), 5.49 (s, 1H), 3.78 (s, 2H), 3.75 (s, 3H), 3.37 (s, 3H), 3.18 (s, 3H); ^{13}C NMR: δ = 159.6, 156.3, 147.2, 130.0, 129.3, 128.0, 127.3, 125.9, 121.6, 115.4, 113.8, 74.2, 56.0, 54.5, 29.7, 28.3; IR (KBr, cm^{-1}) = 3400, 1611, 1512, 1248, 1175, 1030; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{24}N_3O_3$ $[M+H]^+$ 354.1812, found 354.1827; Calcd. for $C_{20}H_{23}N_3NaO_3$ $[M+Na]^+$ 376.1632, found 376.1645.

(*4R*, 8S) and (*4R*, 8R**)-2-Azido-4-(4-benzyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1,5-dihydroimidazol-5-one (186 and *epi*-186):**



epi-**186**



186

Oxaziridine **88** (254 mg, 0.8 mmol) was added to a stirred solution of azide **184** (255 mg, 0.5 mmol) in $CHCl_3$ (3 mL) at rt. and stirred overnight. On completion of the reaction (TLC), the solvent was removed and the yellow residue was purified by gravity column chromatography (CH_2Cl_2/toluene, 1:1, no pressure was used) to isolate *epi*-**186** (123 mg, 47%) as a pale yellow semi-solid and **186** (121 mg, 46%) as a pale yellow solid.

Data for *epi*-186

^{1}H NMR: δ = 7.36-7.26 (m, 5H), 6.95 (d, *J* = 8.7 Hz, 2H), 6.78 (d, *J* = 8.7 Hz, 2H), 6.72 (d, *J* = 8.7 Hz, 2H), 6.68 (d, *J* = 8.7 Hz, 2H), 4.90 (s, 2H), 4.78 (s, 1H), 3.92 (d, *J* = 14.2 Hz, 1 H), 3.72 (s, 3H), 3.35 (d, *J* = 14.2 Hz, 1 H), 3.33 (s, 3H), 2.76 (s, 3H); ^{13}C NMR: δ = 173.7, 160.2, 158.4, 158.3, 136.7, 130.7, 128.9, 128.6, 128.1, 127.6, 125.4, 124.5, 115.0, 114.0, 84.1, 79.0, 70.0, 57.7, 55.3, 38.3, 27.1; IR (neat, cm^{-1}) = 2931, 1764, 1599, 1512, 1455, 1250, 1177, 1098, 1029, 834, 797, 738, 698; HR-ESIMS (*m/z*): Calcd. for $C_{27}H_{28}N_5O_4$ $[M+H]^+$ 486.2136, found 486.2141.

Data for 186

m.p = 54-56 °C; ^{1}H NMR: δ = 7.35-7.26 (m, 7H), 6.99 (d, *J* = 8.7 Hz, 2H), 6.65 (m, 4 H), 4.90 (s, 2H), 4.77 (s, 1H), 3.85 (s, 3H), 3.25 (d, *J* = 14.2 Hz, 1 H), 3.13 (s, 3H), 3.05 (s, 3H), 2.94 (d, *J* = 14.2 Hz, 1 H); ^{13}C NMR: δ = 175.4, 160.7, 159.1, 158.3, 136.7, 130.5, 130.1, 128.6, 128.1, 127.6, 125.3, 123.6, 114.9, 114.3, 84.2, 79.0, 69.9,

57.1, 55.4, 38.4, 27.4; IR (neat, cm^{-1}): = 2931, 1764, 1599, 1512, 1455, 1250, 1177, 1098, 1029, 834, 797, 738, 698; HR-ESIMS (*m/z*): Calcd. for $C_{27}H_{28}N_5O_4$ $[M+H]^+$ 486.2136, found 486.2138.

(*4R*, 8R)-*epi*-Calcaridine *epi*-(13):**

Azide *epi*-**186** (94 mg, 0.2 mmol) was dissolved in EtOH (3 mL) and stirred under a hydrogen atmosphere (55 psi) in the presence of 20% $Pd(OH)_2$ on charcoal (40 mg) at rt. overnight. The catalyst was filtered through a pad of Celite and the filtrate was concentrated to isolate *epi*-calcaridine A, *epi*-**13** (73 mg, quant) as an off-white solid: m.p = 218-220 °C; ^{1}H NMR (CD_3OH): δ = 7.20 (d, *J* = 8.7 Hz, 2H), 6.94 (d, *J* = 8.7 Hz, 2H), 6.89 (d, *J* = 8.7 Hz, 2H), 6.66 (d, *J* = 8.7 Hz, 2H), 4.59 (s, 1 H), 3.77 (s, 3H), 3.43 (d, *J* = 14.2 Hz, 1 H), 3.31 (s, 3H), 3.18 (d, *J* = 14.2 Hz, 1 H), 2.51 (s, 3H). ^{13}C NMR: δ = 172.2, 160.5, 157.8, 156.8, 130.9, 129.0, 126.2, 124.40 114.9, 113.5, 84.1, 73.5, 56.3, 54.4, 38.3, 24.1; IR (KBr, cm^{-1}): = 3311 (br), 3001, 2830, 1770, 1693, 1613, 1560, 1513, 1440, 1309, 1256, 1089, 1032, 832, 793, 718; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{24}N_3O_4$ $[M+H]^+$ 370.1761, found 370.1761; Calcd. for $C_{20}H_{23}N_3NaO_4$ $[M+Na]^+$ 392.1586, found 392.1512.

(*4R*, 8S)-Calcaridine A (13):**

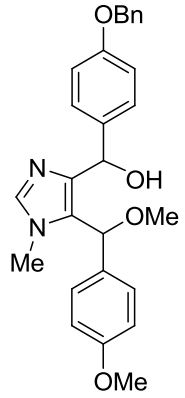
Following the procedure above, azide **186** (102 mg, 0.2 mmol) and 20% $Pd(OH)_2$ on charcoal (40 mg) in EtOH (3 mL) gave calcaridine A, **13** (78 mg, quant) as a pale yellow solid: m.p = 163-165 °C; ^{1}H NMR (CD_3OH): δ = 7.37 (d, *J* = 8.3 Hz, 2H), 7.03 (d, *J* = 8.3 Hz, 2H), 6.86 (d, *J* = 8.3 Hz, 2H), 6.64 (d, *J* = 8.3 Hz, 2H), 4.58 (s, 1H), 3.82 (s, 3H), 3.16 (s, 3H), 3.16 (d, *J* = 14.2 Hz, 1H), 2.83 (s, 3H), 2.50 (d, *J* =

14.2 Hz, 1H); ^{13}C NMR: δ = 173.3, 160.7, 158.7, 156.9, 130.8, 129.4, 126.1, 123.0, 114.9, 114.1, 84.2, 73.1, 56.1, 54.6, 37.9, 24.6; IR (KBr, cm^{-1}): = 3265 (br), 2833, 1781, 1692, 1612, 1560, 1513, 1449, 1346, 1250, 1093, 1023, 836, 799; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{24}N_3O_4$ $[M+H]^+$ 370.1761, found 370.1761.

Synthesis of 171 from 170: an alternative method

TFA (2.77 mL, 36.0 mmol) was added to a solution of **170** (4.15 g, 12.o mmol) in Methanol (40 mL) at rt. and the mixture was heated at 60 °C overnight. The reaction was allowed to come to rt. and was quenched with saturated $NaHCO_3$ (30 mL). The aqueous layer was extracted with EtOAc (3x50 mL). The combined organic extracts were dried (Na_2SO_4) and concentrated to isolate a thick pale yellow oil **171** (4.16 g, 97%).

4-(4-Benzyloxyphenyl)hydroxymethyl-5-[methoxy-(4-methoxy)phenyl]methyl-1-methyl-1*H*-imidazole (187):



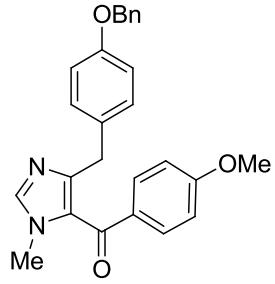
A 3.0 M solution of EtMgBr in Et_2O (3.36 mL, 10.1 mmol) was added to a solution of **171** (3.30 g, 9.2 mmol) in anhyd THF (30 mL) at rt. The resulting mixture was stirred at the same temperature for 30 min and 4-benzyloxybenzaldehyde, **162** (4.86 g, 22.9 mmol) was added. After stirring the reaction for 72 h, water (10 mL) was added to quench the reaction. The organic solution was separated and washed with saturated brine solution (30 mL). The organic solution was dried (Na_2SO_4) and concentrated to provide the crude product, which was purified by column chromatography using gradient elution (EtOAc/hexanes,

1:9→EtOAc) to isolate **187** as a 2:1 mixture of the diastereomers (2.16 g, 53%): ^{1}H NMR (the signals due to the aromatic protons are overlapping, the underlined signals correspond to the minor product and the relevant signals are integrated together with the major product): δ = 7.41 (m, 2H), 7.36 (m, 2H), 7.3 (m, 2H), 7.25 (m, 2H), 7.18 (m, 2H), 7.08 (d, *J* = 8.7 Hz, 1H), 6.86 (d, *J* = 8.3 Hz, 1H), 6.77 (d, *J* = 8.7 Hz, 2H), 5.94, 5.91 (s, 1H), 5.62, 5.56 (s, 1H), 4.95, 4.94 (s, 2H), 3.67, 3.66 (s, 3H), 3.20, 3.17 (s, 3H),3.15, 3.07 (s, 3H). ^{13}C NMR (minor product shown within brackets): δ = 158.9 (158.8), 157.9 (157.8), (145.0) 144.7, 138.3 (138.2), 137.3 (137.3), 131.9, 128.6 (128.3), 128.1, 127.9 (127.8), 127.5, 127.4, 125.6 (125.5), 114.6 (114.6), (113.9) 113.7, 74.7 (74.5), 70.0, 69.6 (69.5), 56.7 (56.6), (55.3) 55.2, 32.7; IR (neat, cm^{-1}): = 3101 (br), 2992, 2930, 2833, 6109, 1501, 1453, 1236, 1169, 1081, 1013, 806, 750; HR-ESIMS (*m*/*z*): Calcd for $C_{27}H_{29}N_2O_4$ [M+H]+: 445.2122, found: 445.2125; calcd for $C_{54}H_{57}N_4O_8$ [2M+H]+:889.4147, found: 889.4196.

Synthesis of 183 from 187:

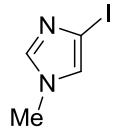
TFA (0.73 mL, 9.4 mmol) and Et_3SiH (1.51 mL, 9.4 mmol) were added to a solution of **187** (1.40 g, 3.1 mmol) in dichloromethane at rt. The resulting mixture was stirred at the same temperature for 24 h and was quenched by adding sat $NaHCO_3$. The layers were separated and the aqueous layer was extracted with CH_2Cl_2 (2x 50 mL). The combined organic layer was dried ($NaSO_4$) and concentrated to give the crude material which was purified over silica gel with EtOAc to provide the title compound (1.00 g, 74%).

[4-(4-Benzyloxybenzyl)-1-methyl-1*H*-imidazol-5-yl]-(4-methoxyphenyl)-methanone (188):



MnO_2 (0.26g, 30.3 mmol) was added to a solution of alcohol **182** (1.26 g, 3.0 mmol) in dichloromethane at rt., and the resulting mixture was heated at reflux for overnight. After cooling to rt., the reaction mixture was filtered through a pad of Celite. Filtrate was concentrated to isolate ketone **188** (1.23 g, 98%) as a pale yellow oil: ^{1}H NMR: δ = 8.30 (d, *J* = 8.8 Hz, 2H), 7.421-7.29 (m, 6H), 7.14 (d, *J* = 8.5 Hz, 2 H), 6.97 (d, *J* = 8.8 Hz, 2 H), 6.89 (d, *J* = 8.5 Hz, 2 H), 5.01 (s, 2H), 4.42 (s, 2H), 3.87 (s, 3 H), 3.47 (s, 3H); ^{13}C NMR: δ = 188.2, 162.8, 157.6, 138.7, 138.1, 137.1, 136.7, 132.9, 131.5, 129.8, 129.5, 128.7, 128.0, 127.6, 115.2, 113.3, 70.1, 55.5, 31.7, 29.2; IR (KBr, cm^{-1}): = 3111, 3007, 2905, 2839, 1597, 1534, 1504, 1336, 1310, 1230, 1171, 1154, 1024, 948, 847, 808, 770; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{25}N_2O_3$ $[M+H]^+$ 413.1860, found 413.1858.

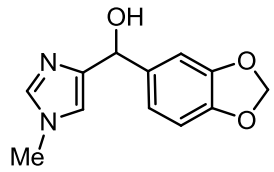
4-Iodo-1-methyl-1*H*-imidazole (191):[155]



EtMgBr (3.0 M solution in ether, 11.00 mL, 32.9 mmol) was added to a solution of **81** (10.00 g, 29.9 mmol) in dry CH_2Cl_2 (150 mL) at rt. over ~15 min. The resulting mixture was stirred for 20 min and water (20 mL) was added to the reaction mixture after running a TLC experiment to confirm all the starting material is consumed. Then, the layers were separated. The aqueous layer was extracted with CH_2Cl_2 (3x50 mL) and the combined organic layers were dried (Na_2SO_4) and concentrated to isolate **190** as pale yellow oil (5.86 g, 94%).

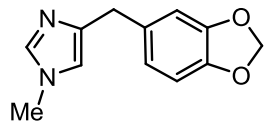
.

4-(Benzo[1,3]dioxol-5-yl)hydroxymethyl-1-methyl-1*H*-imidazole (192):



EtMgBr (3.0 M solution in ether, 9.76 mL, 29.3 mmol) was added to a solution of **191** (5.80 g, 27.9 mmol) in dry THF (100 mL) at 0 ºC over ~10 min. The resulting mixture was stirred at rt. for 30 min and piperonal (4.65 mL, 30.7 mmol) was added to the reaction slowly at rt. After stirring overnight at rt., satd. aq. NH_4Cl (20 mL) was added to the reaction and the layers were separated. The aqueous layer was extracted with EtOAc (50 mLx3) and the combined organic layers were dried (Na_2SO_4) and concentrated to provide a crude product, which was purified over silica gel (100% EtOAc → 100% Acetone) to isolate **192** (5.82 g, 90%) as an orange semi-solid: ^{1}H NMR: δ = 7.35 (s, 1H), 6.90 (s, 1H), 6.87 (d, *J* = 7.8 Hz, 1H), 6.72 (d, *J* = 7.8 Hz, 1H), 6.45 (s, 1H), 5.88 (s, 2H), 5.66 (s, 1H), 5.27 (br, 1H), 3.54 (s, 3H); ^{13}C NMR: δ = 147.5, 146.8, 146.0, 137.8, 137.4, 120.1, 117.2, 107.9, 107.4, 101.0, 70.0, 33.5; IR (neat, cm^{-1}): = 3114, 2891, 1493, 1442, 1243, 1037, 927, 794, 756; HR-ESIMS (*m/z*): Calcd. for $C_{12}H_{13}N_2O_3$ $[M+H]^+$ 233.0921, found 233.0925.

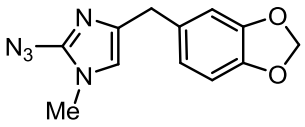
4-(Benzo[1,3]dioxol-5-yl)methyl-1-methyl-1*H*-imidazole (193):



TFA (7.00 mL, 91.4 mmol) and Et_3SiH (17.00 mL, 106.4 mmol was added to a solution of **192** (5.31 g, 22.9 mmol) in dry CH_2Cl_2 (150 mL) at rt. The resulting mixture was stirred for 48 h and sat. $NaHCO_3$ solution was added to it. The resulting mixture was extracted with CH_2Cl_2 several times and the combined extracts were dried (Na_2SO_4) and concentrated. The residue was purified by chromatography (Acetone/EtOAc, 3/7) to isolate a pale yellow semi-solid: **193** (3.55 g, 72%); ^{1}H NMR: δ = 7.27 (s, 1H), 6.71

(s, 1H), 6.67 (s, 2H), 6.46 (s, 1H), 5.82 (s, 2H), 3.76 (s, 2H), 3.52 (s, 3H); ^{13}C NMR: δ = 147.6, 145.8, 142.8, 137.4, 134.4, 121.6, 117.0, 109.4, 108.2, 101.0, 34.7, 33.2; IR (neat, cm^{-1}): = 3367, 2900, 2778, 1492, 1441, 1246, 1185, 1038, 928, 815, 773; HR-ESIMS (*m/z*): Calcd. for $C_{12}H_{13}N_2O_2$ $[M+H]^+$ 217.0972, found 217.0991.

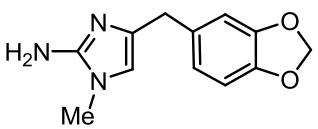
2-Azido-4-(benzo[1,3]dioxol-5-yl)methyl-1-methyl-1*H*-imidazole (194):



Following the general procedure for the azidation, *n*-Butyl lithium (1.4 M solution in hexanes, 11.34 mL, 15.9 mmol), **193** (3.12 g, 14.4 mmol) and TsN_3 (3.41 g, 17.3 mmol) were used to afford the crude product, which was purified over silica gel (15% → 20% EtOAc in hexanes) to isolate **194** (2.93 g, 78%) as a reddish brown oil: ^{1}H NMR: δ = 6.76 (s, 1H), 6.72 (s, 2H), 6.24 (s, 1H), 5.90 (s, 2H), 3.74 (s, 2H), 3.31 (s, 3H); ^{13}C NMR: δ = 147.7, 146.0, 140.2, 140.0, 133.6, 121.7, 116.1, 109.5, 108.2, 100.9, 34.8, 31.5; IR (neat, cm^{-1}): = 2896, 2154, 2125, 1494, 1441, 1245, 1039, 929, 812; HR-ESIMS (*m/z*): Calcd. for $C_{12}H_{12}N_5O_2$ $[M+H]^+$ 258.0986, found 258.0987.

2-amino-4-(benzo[1,3]dioxol-5-yl)methyl-1-methyl-1*H*-imidazole (7b):

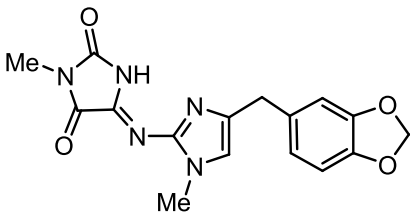
Preclathridine A



Following the general procedure for reduction, Azide **194** (2.80 g, 10.9 mmol) was dissolved in EtOH (30 mL) and stirred under a hydrogen atmosphere (1 atm) in the presence of 10% Pd-C on charcoal (0.30 g) at rt. overnight to afford **7b** (2.59 g, quant) as a yellow solid: m.p = 116-118 °C; ^{1}H NMR ($CDCl_3$): δ = 6.73 (s, 1H), 6.69 (s, *2*H), 6.08 (s, 1H), 5.86 (s, 2H), 4.14 (br, 2H), 3.63 (s, 2H), 3.26 (s, 3H); ^{13}C NMR

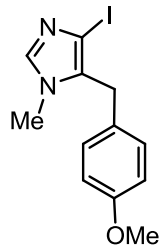
δ = 148.0, 147.5, 145.8, 137.1, 134.5, 121.7, 112.8, 109.5, 108.1, 100.8, 34.7, 31.3; IR (KBr, cm^{-1}): = 3432, 3299, 3109, 1643, 1547, 1505, 1442, 1244, 1034, 921, 814 ; HR-ESIMS (*m/z*): Calcd. for $C_{12}H_{14}N_3O_2$ $[M+H]^+$ 232.1081, found 232.1094.

2-(3-methylimidazolidine-2,4-dione)imino-4-(4-Benzo[1,3]dioxol-5-ylmethyl-1-methyl-1*H*-imidazole (8b): Clathridine A



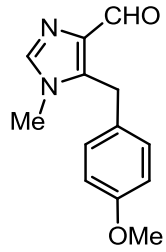
N,O-Bis(trimethysilyl)acetamide (5.18 mL, 21.2 mmol) was added to a solution of 1-methylparabanic acid (2.26 g, 17.7 mmol) in dry CH_3CN (30 mL) under an N_2 atmosphere and the resulting mixture was heated at reflux temperature for 1.5 h. Then, the solvent was removed by vacuum distillation, and to the resulting yellow residue in dry toluene (5 mL) was added amine **7b** (817 mg, 3.5 mmol) under N_2 atmosphere. After, this mixture was heated at 85 º C overnight (product starts to form after 10 min as a yellow solid at the bottom of the flask), the product was filtered and solid was washed with methanol (10 mL) to isolate **8b** as a yellow solid (730 mg, 61%): m.p = 253-254 ºC (lit.[7] m.p = 260-262 ºC); ^{1}H NMR: δ = 6.75 (d, *J* = 8.3 Hz, 1H), 6.71 (s, 1H), 6.70 (d, *J* = 8.3 Hz, 1H), 6.52 (s, 1H), 5.93 (s, 2H), 3.80 (s, 2H), 3.71 (s, 3H), 3.18 (s, 3H); ^{13}C NMR: δ = 162.0, 155.0, 147.8, 147.1, 146.3, 144.3, 139.7, 132.7, 121.7, 117.7, 109.3, 108.4, 101.0, 34.5, 32.2, 24.8; IR (KBr, cm^{-1}): = 3204, 3122, 2914, 1789, 1738, 1665, 1444, 1391, 1323, 1249, 1209, 1114, 1034, 925, 732; HR-ESIMS (*m/z*): Calcd. for $C_{16}H_{16}N_5O_4$ $[M+H]^+$ 342.1197, found 342.1201.

4-Iodo-5-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (195):



Et_3SiH (13.9 mL, 87.2 mmol) and TFA (5.60 mL, 72.6 mmol) were added to a solution of **170** (5.00 g, 14.5 mmol) in anhydrous CH_2Cl_2 (100 mL) at rt., then the resulting mixture was heated at reflux for 60 h under N_2 atmosphere. After cooling to rt., the reaction was quenched by the addition of sat. aq. solution of $NaHCO_3$. The resulting mixture was extracted with CH_2Cl_2 several times and the combined extracts were dried (Na_2SO_4) and concentrated. The residue was purified by chromatography (hexane/EtOAc, 1/3) to isolate **195** (4.65 g, 97%) as a pale yellow solid: m.p = 97-99 ºC; ^{1}H NMR (Acetone –D6): δ = 7.50 (s, 1H), 7.09 (d, *J*= 8.7 Hz, 2H), 6.85 (d, *J*= 8.7 Hz, 2 H), 3.92 (s, 2H), 3.74 (s, 3H), 3.51 (s, 3H); ^{13}C NMR: δ = 158.6, 139.4, 133.4, 129.3, 129.1, 114.3, 84.9, 55.4, 32.6, 30.0; IR (KBr, cm^{-1}): = 3005, 3001, 2957, 2834, 1608, 1508, 1444, 1280, 1247, 1176, 1030, 981, 821, 770, 694; HR-ESIMS (*m/z*): Calcd. for $C_{12}H_{14}IN_2O$ $[M+H]^+$ 329.0145, found 329.0142.

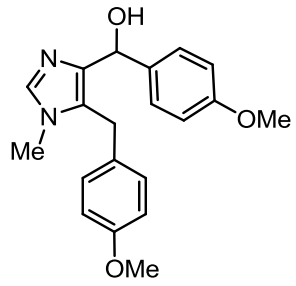
5-(4-Methoxybenzyl-1-methyl-1*H*-imidazole-4-carbaldehyde (196):



EtMgBr (3.0 M in ether, 5.2 mL, 15.7 mmol) was added into a solution of **195** (4.90 g, 14.9 mmol) in dry THF (50 mL) at 0 ºC, and the resulting mixture was stirred at rt. for 2 h. Then, *N*-methylformanilide, **165** (2.23 mL, 17.9 mmol) was added at 0 ºC and the resulting mixture was stirred at rt. overnight. Saturated aq. NH_4Cl (20 mL) was added to quench the reaction and the organic layer was extracted with EtOAc, dried (Na_2SO_4) and concentrated to provide the crude product, which was purified through a short plug of silica gel (EtOAc) to isolate **196** (3.10 g, 90%) as

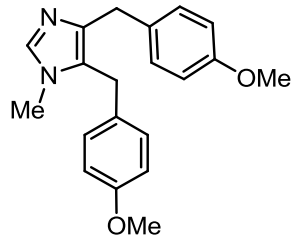
a white solid: m.p = 120-121 °C; ^{1}H NMR: δ = 9.99 (s, 1H), 7.40 (s, 1H), 7.04 (d, *J* = 8.3 Hz, 2H), 6.79 (d, *J* = 8.3 Hz, 2H), 4.32 (s, 2H), 3.3.75 (s, 3H), 3.45 (s, 3H); ^{13}C NMR: δ = 187.7, 158.6, 138.8, 138.4, 137.7, 129.3, 128.5, 114.4, 55.3, 31.6, 28.5; IR (KBr, cm^{-1}): = 3105, 3022, 2957, 2818, 2770, 1669, 1552, 1511, 1344, 1244,1022, 824, 789; HR-ESIMS (*m/z*): Calcd. for $C_{13}H_{15}N_2O_2$ $[M+H]^+$ 231.1128, found 231.1139.

4-[hydroxy-(4-methoxyphenyl)]methyl-5-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (197):



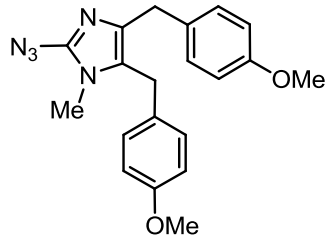
Following the general procedure for the Grignard reaction, *p*-bromoanisole (6.67 mL, 52.1 mmol), magnesium turnings (1.25 g, 52.1 mmol) and a solution of **196** (3.00 g, 13.0 mmol) in THF (20 mL) were used to afford a crude product, which was purified through a short plug of silica gel (EtOAc) to isolate **197** (4.34 g, 98%) as an off-white solid: m.p = 134-135 °C; ^{1}H NMR: δ = 7.33 (d, *J* = 8.7 Hz, 2H), 7.33 (s, 1H), 6.88 (d, *J* = 8.7 Hz, 2H), 6.81 (d, *J* = 8.7 Hz, 2H), 6.75 (d, *J* = 8.7 Hz, 2H), 5.74 (s, 1H), 3.81 (s, 2H), 3.75 (s, 6H), 3.30 (s, 3H); ^{13}C NMR: δ = 158.5, 158.1, 141.4, 136.8, 136.7, 130.2, 129.2, 127.8, 126.2, 113.9, 113.4, 69.4, 55.1, 55.1, 31.4, 28.0; IR (KBr, cm^{-1}): = 3112 (br), 2835, 2710, 1608, 1582, 1511, 1461, 1298, 1250, 1173, 1042, 846, 820, 787; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{23}N_2O_3$ $[M+H]^+$ 339.1703, found 339.1701.

4,5-Bis(4-methoxybenzyl)-1-methyl-1*H*-imidazole (198):



Et_3SiH (1.88 mL, 11.8 mmol) and TFA (0.73 mL, 9.5 mmol) were added to a solution of **197** (800 mg, 2.4 mmol) in anhydrous CH_2Cl_2 (20 mL) at rt, then the resulting mixture was stirred overnight. The reaction was quenched by the addition of satd. aq. solution of $NaHCO_3$ and the resulting mixture was extracted with CH_2Cl_2 several times and the combined extracts were dried (Na_2SO_4), concentrated and the residue was purified by chromatography (acetone) to provide **198** (615 mg, 81%) as a pale yellow oil: ^1H NMR: δ = 7.32 (s, 1H), 7.16 (d, *J* = 8.7 Hz, 2H), 6.90 (d, *J* = 8.7 Hz, 2H), 6.78 (d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 3.86 (s, 2H), 3.85 (s, 2H), 3.75 (s, 3H), 3.74 (s, 3H), 3.30 (s, 3H); ^{13}C NMR: δ = 158.3, 157.9, 139.0, 136.7, 133.2, 130.4, 129.6, 129.0, 125.7, 114.1, 113.9, 55.3, 33.1, 31.7, 28.3; IR (neat, cm^{-1}): = 3000, 2908, 2835, 1610, 1509, 1461, 1245,1178, 1034,819; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{23}N_2O_2$ $[M+H]^+$ 323.1754, found 323.1774.

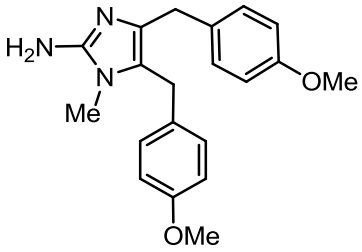
2-Azido-4,5-bis(4-methoxybenzyl)-1-methyl-1*H*-imidazole (199):



n-Butyl lithium (1.4 M solution in hexanes, 1.50 mL, 2.1 mmol) was added dropwise to a stirred solution of **198** (613 mg, 1.9 mmol) in dry THF (15 mL) at -78 °C. The reaction was stirred for 1 h at the same temperature. The cooling bath was removed for 10 min, then the reaction mixture was re-cooled to -78 °C, and TsN_3 (0.80 g, 4.1 mmol) in THF (3 mL) was added dropwise. After stirring for 40 min at -78 °C, the reaction

mixture was allowed to come to rt. and was quenched by the addition of satd. aq. NH_4Cl (3 mL). The aqueous layer was extracted with EtOAc (3x20 mL), and the combined organic extracts were dried (Na_2SO_4) and concentrated to provide a pale brown oil, which was purified through a short column of silica gel (hexane/EtOAc, 4:1) to isolate **199** (615 mg, 89%) as a pale yellow oil: ^{1}H NMR: δ = 7.17 (d, *J* = 8.3 Hz, 2H), 6.90 (d, *J* = 8.3 Hz, 2H), 6.79 (d, *J* = 8.3 Hz, 2H), 6.77 (d, *J* = 8.3 Hz, 2H), 3.84 (s, 2H), 3.79 (s, 2H), 3.76 (s, 6H), 3.06 (s, 3H); ^{13}C NMR: δ = 158.3, 158.0, 139.0, 136.3, 132.8, 130.2, 129.5, 129.0, 125.0, 114.1, 113.9, 55.3, 32.8, 29.5, 28.7; IR (neat, cm^{-1}): = 3000, 2944, 2835, 2132, 1609, 1508, 1247, 1176, 1034, 821; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{22}N_5O_2$ $[M+H]^+$ 364.1773, found 364.1768.

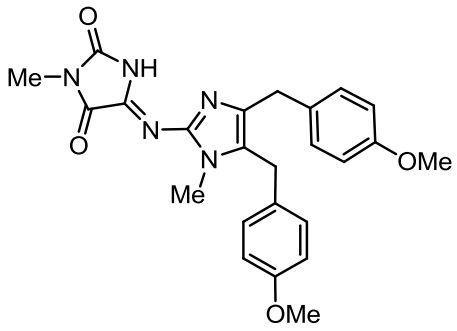
2-Amino-4,5-bis(4-methoxybenzyl)-1-methyl-1*H*-imidazole (200):



Azide **199** (475 mg, 1.3 mmol) was dissolved in EtOH (10 mL) and stirred under a hydrogen atmosphere (1 atm) in the presence of 10% Pd-C on charcoal (47 mg) at rt. overnight. The catalyst was filtered through a pad of Celite and the filtrate was concentrated to isolate amine, **200** (440 mg, quant) as a pale yellow solid: m.p = 175-176 °C; ^{1}H NMR: δ = 7.09 (d, *J* = 8.3 Hz, 2H), 6.93 (d, *J* = 8.3 Hz, 2H), 6.77 (d, *J* = 8.3 Hz, 2H), 6.74 (d, *J* = 8.3 Hz, 2H), 5.79 (br, 2H), 3.75 (s, 2H), 3.74 (s, 3H), 3.70 (s, 3H), 3.68 (s, 2H), 3.03 (s, 3H); ^{13}C NMR: δ = 158.4, 158.0, 147.4, 132.4, 130.4, 130.4, 129.6, 128.9, 121.0, 114.1, 113.9, 55.3, 55.3, 31.8, 29.5, 28.5; IR (KBr, cm^{-1}): = 3374, 3296, 3123, 2954, 2838, 1764, 1610, 1550, 1510, 1462, 1249, 1178, 1033,

819, 758; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{24}N_3O_2$ $[M+H]^+$ 338.1863, found 338.1871.

4,5-Bis(4-methoxybenzyl)-1-methyl-2-(3-methylimidazolidine-2,4-dione)imino--1*H*-imidazole (2f): Naamidine G

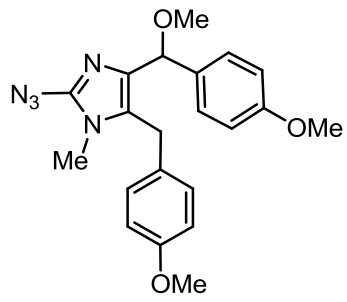


N,*O*-Bis(trimethysilyl)acetamide (1.00 mL, 4.1 mmol) was added to a solution of 1-methylparabanic acid (526 mg, 4.1 mmol) in dry CH_3CN under an N_2 atmosphere and the resulting mixture was heated at reflux temperature for 1h. Then, the solvent was removed by distillation and to the resulting yellow residue in toluene (3 mL) was added amine **200** (277 mg, 0.8 mmol) under N_2. After, this mixture was heated at 85 º C overnight, water (5 mL) was added and the organic layer was extracted in to EtOAc. The dried organic layer (Na_2SO_4) was concentrated to afford a yellow residue, which was purified over silica gel (EtOAc/hexanes, 3/7) to provide **2f** (287 mg, 78%) as a yellow powder: m.p = 195-196 ºC (lit.[12] m.p = 94 ºC); ^{1}H NMR: δ = 10.04 (br, 1H), 7.10 (d, *J* = 8.7 Hz, 2H), 6.90 (d, *J* = 8.7 Hz, 2H), 6.80 (d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 3.89 (s, 2H), 3.88 (s, 2H), 3.76 (s, 6H), 3.47 (s, 3H), 3.15 (s, 3H); ^{13}C NMR: δ = 162.3, 158.6, 158.3, 155.7, 146.5, 145.0, 135.7, 131.5, 129.4, 129.1, 129.0, 127.0, 114.3, 114.1, 55.4, 32.3, 30.0, 28.7, 24.7; IR (KBr, cm-1): = 3241, 2931, 2835, 1788, 1738, 1656, 1511, 1451, 1302, 1248, 1177, 1036, 744, 697; HR-ESIMS (*m/z*): Calcd. for $C_{24}H_{26}N_5O_4$ $[M+H]^+$ 448.1979, found 448.1984.

Synthesis of 155 from 197:

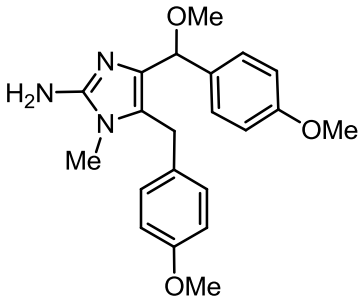
TFA (0.60 mL, 7.8 mmol) was added to a solution of **197** (1.31 g, 3.9 mmol) in anhyd MeOH (20 mL) at rt. and the mixture was then heated at 55 °C overnight. Satd. aq. solution of $NaHCO_3$ was used to neutralize the above reaction mixture, and the aqueous layer was extracted with EtOAc (30 mLx3). Combined organic layers were washed with water and brine, then dried (Na_2SO_4) and concentrated to provide **155** (1.37 g, >95%) as a pale yellow oil: ^{1}H NMR: δ = 7.34 (d, *J* = 8.7 Hz, 2H), 7.23 (s, 1H), 6.86 (d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 6.71 (d, *J* = 8.7 Hz, 2H), 5.26 (s, 1 H), 3.93 (s, 2H), 3.75 (s, 6H), 3.34 (s, 3H), 3.29(s, 3H).

2-Azido-5-[4-methoxybenzyl]-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (201):



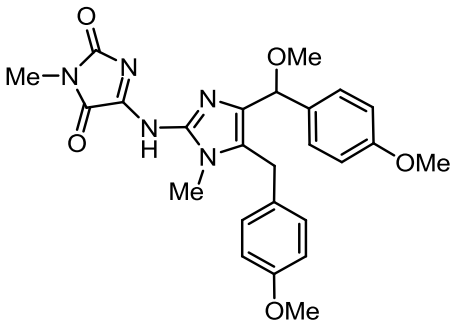
Following the general procedure for azidation, *n*-Butyl lithium (1.4 M solution in hexanes, 2.66 mL, 3.7 mmol), **155** (1.45 g, 3.4 mmol) in dry THF (20 mL) and TsN_3 (0.80 g, 4.1 mmol) were used to obtain the crude product, which was purified through a short column of silica gel (hexane/EtOAc, 4:1) to isolate **201** (891 mg, 67%) as a reddish brown oil: ^{1}H NMR: δ = 7.41 (d, *J* = 8.7 Hz, 2H), 6.90 (d, *J* = 8.7 Hz, 2H), 6.84 (d, *J* = 8.7 Hz, 2H), 6.76 (d, *J* = 8.7 Hz, 2H), 5.23 (s, 1 H), 3.85 (s, 2H), 3.78 (s, 3H), 3.76 (s, 3H), 3.35 (s, 3H), 3.03 (s, 3H); ^{13}C NMR: δ = 159.0, 158.4, 139.8, 136.9, 133.4, 129.9, 129.1, 128.4, 126.1, 114.1, 113.7, 79.1, 56.8, 55.3, 55.3, 29.4, 28.6; IR (neat, cm^{-1}) = 2937, 2834, 2138, 1508, 1246, 1174, 1087, 1033, 829; HR-ESIMS (*m/z*): Calcd. for $C_{21}H_{23}N_5NaO_3$ $[M+Na]^+$ 416.1693, found 416.1677.

2-Amino-5-(4-methoxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (202):



Following the general procedure for catalytic reduction, azide **201** (800 mg, 2.0 mmol) in EtOH (10 mL) and 10% Pd-C on charcoal (80 mg) at 1 atm hydrogen were used to afford amine **202** (745 mg, quant) as a pale yellow solid: m.p = 139-140 °C; ^{1}H NMR (CD_3OH): δ = 7.29 (d, *J* = 8.7 Hz, 2H), 6.95 (d, *J* = 8.7 Hz, 2H), 6.82 (d, *J* = 8.7 Hz, 2H), 6.78 (d, *J* = 8.7 Hz, 2H), 5.21 (s, 1H), 3.86 (s, 2H), 3.74 (s, 3H), 3.73 (s, 3H), 3.30 (s, 3H), 3.08 (s, 3H); ^{13}C NMR: δ = 159.0, 158.4, 148.9, 133.8, 131.8, 130.7, 128.7, 127.9, 123.0, 113.6, 113.1, 78.5, 55.4, 54.4, 54.4, 28.0, 27.7; IR (KBr, cm^{-1}) = 3129, 1671, 1510, 1459, 1248, 1176, 1084, 1031, 830, 746; HR-ESIMS (*m/z*): Calcd. for $C_{21}H_{26}N_3O_3$ $[M+H]^+$ 368.1969, found 368.1970.

5-(4-Methoxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-2-(3-methylimidazolidine-2,4-dione)imino-1*H*-imidazole (4d): 4-Methoxynaamidine G

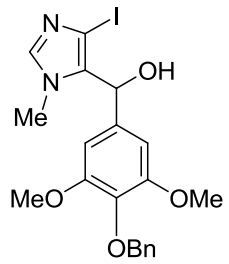


Following the general procedure, *N,O*-Bis(trimethysilyl)acetamide (0.33 mL, 1.4 mmol) and 1-methylparabanic acid (174 mg, 1.4 mmol) in dry CH_3CN (15 mL) were used to produce a yellow residue, which was treated with amine **202** (100 mg, 0.3 mmol) to obtain the product **4d** (13 mg, 11%) as a yellow solid after the purification over

silica gel (EtOAc/hexanes, 2/3): ^{1}H NMR: δ = 7.34 (d, *J* = 8.8 Hz, 2H), 6.92 (d, *J* = 8.8 Hz, 2H) , 6.87 (d, *J* = 8.8 Hz, 2H) , 6.78 (d, *J* = 8.8 Hz, 2H) , 5.29 (s, 1H), 3.98 (s, 2H), 3.79 (s, 3H), 3.77 (s, 3H), 3.45 (s, 3H), 3.35 (s, 3H), 3.17 (s, 3H);

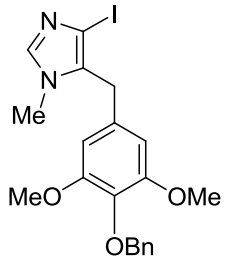
4-Benzoyloxy-4,5-dimethylbenzaldehyde (203) was prepared following Tanaka's procedure from syringaldehyde.[109]

5-(4-Benzyloxy-3,5-dimethoxyphenyl)hydroxymethyl-4-iodo-1-methyl-1*H*-imidazole (204):



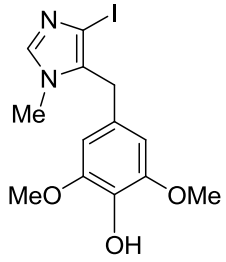
EtMgBr (3.0 M solution in ether, 7.54 mL, 22.6 mmol) was added to a solution of **81** (7.19 g, 21.5 mmol) in dry CH_2Cl_2 (100 mL) at rt. over ~10 min. After stirring at rt. for 20 min, aldehyde **203** (6.46 g, 23.7 mmol) was added to the reaction followed by 48 h stirring. Then, satd. aq. NH_4Cl (10 mL) was added to the reaction and the resulting pale yellow solid was filtered and the filtrate was partitioned with CH_2Cl_2. The organic layer was dried (Na_2SO_4) and concentrated to provide a pale yellow solid. The resulting solid was triturated with hexanes, recrystallized with CH_2Cl_2 to isolate **204** (9.68 g, 95%) as a white solid: m.p = 173-175 ºC; ^{1}H NMR: δ = 7.44 (d, *J* = 7.8 Hz, 2H), 7.31-7.24 (m, 4H), 6.57 (s, 2H), 5.98 (s, 1H), 5.18 (s, 1H), 4.98 (s, 2H), 3.75 (s, 6H), 3.43 (s, 3H); ^{13}C NMR: δ = 153.6, 141.1, 137.8, 136.6, 135.9, 135.2, 128.6, 128.2, 127.9, 102.7, 84.6, 75.0, 67.1,56.3, 33.5; IR (neat, cm^{-1}): = 3253 (br), 3104, 1585, 1501, 1411, 1338, 1226, 1140, 1033, 908 ; HR-DARTMS (*m/z*): Calcd. for $C_{20}H_{21}IN_2O_4$ $[M]^+$ 480.0546; found: 480.0546; Calcd. for $C_{20}H_{22}IN_2O_4$ $[M+H]^+$ 481.0624, found 481.0624.

5-(4-Benzyloxy-3,5-dimethoxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (205a):



Et_3SiH (1.00 mL, 6.2 mmol) and TFA (0.40 mL, 5.2 mmol) were added to a solution of **204** (0.50 g, 1.0 mmol) in anhydrous $CHCl_3$ (20 mL) at rt. and the resulting mixture was heated at reflux temperature for 24 h under nitrogen atmosphere. After cooling to rt., reaction was quenched by the addition of satd. aq. $NaHCO_3$. The aqueous layer was extracted several times with $CHCl_3$ and the combined organic extracts were dried (Na_2SO_4) and concentrated. The residue was purified by chromatography (EtOAc) to isolate **205a** (0.14 g, 28%) as a pale brown semi solid: ^{1}H NMR: δ = 7.44 (d, *J* = 6.9 Hz, 2H), 7.36 (s, 1H), 7.30 (t, *J* = 6.9 Hz, 2H), 7.25 (d, *J* = 6.9 Hz, 1H), 6.31 (s, 2H), 4.95 (s, 2H), 3.87 (s, 2H), 3.73 (s, 6H), 3.40 (s, 3H); ^{13}C NMR: δ = 153.8, 139.6, 137.9, 135.9, 133.1, 128.5, 128.2, 127.9, 105.2, 85.0, 75.1, 56.3, 32.7, 31.0; IR (neat, cm^{-1}): = 3107, 2939, 1588, 1494, 1460, 1420, 1237, 1215, 1187, 1100, 978,758 ; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{22}IN_2O_3$ $[M+H]^+$ 465.0670, found 465.0669.

5-[(3,5-Dimethoxy-4-hydroxy)benzyl]-4-iodo-1-methyl-1*H*-imidazole (205b):

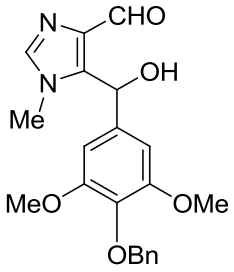


From above reaction, **205b** (0.11 g, 27%) as a pale yellow solid: m.p = 210-212 ºC; ^{1}H NMR (DMSO-D6): δ = 7.40 (s, 1H), 6.34 (s, 2H), 5.47 (br, 1H), 3.90 (s, 2H), 3.82 (s, 6H), 3.45 (s, 3H); ^{13}C NMR: δ = 148.6, 140.6, 134.7, 133.7, 128.4, 106.1, 85.1, 56.5, 32.6, 30.0; IR (neat, cm^{-1}): = 3107, 2936 (br), 1595, 1514, 1499, 1413, 1242, 1214, 1113, 1037, 838, 811, 765, 737; HR-DARTMS (*m/z*): Calcd. for $C_{13}H_{16}IN_2O_3$ $[M+H]^+$ 375.0200, found 375.0226.

4-Benzoyloxy-4,5-dimethylbenzyl bromide (206):

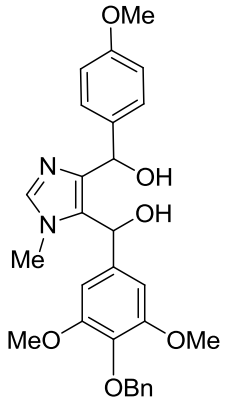
Aldehyde **203** was reduced to 4-benzoyloxy-4,3-dimethylbenzylalcohol with $NaBH_4$,[154] and this alcohol was brominated using PBr_3 and pyridine in Et_2O.[122]

5-(4-Benzyloxy-3,5-dimethoxyphenyl)hydroxymethyl-1-methyl-1*H*-imidazole-4-carbaldehyde (207):



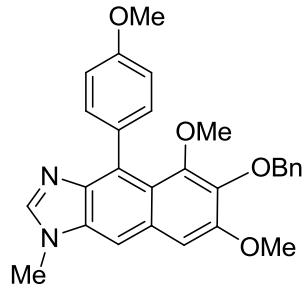
EtMgBr (3.0 M in ether, 13.8 mL, 41.4 mmol) was added into a solution of **204** (9.03 g, 18.8 mmol) in dry THF (200 mL) at rt., and the resulting mixture was stirred for 20 min. Then, *N*-methylformanilide, **165** (2.78 mL, 22.6 mmol) was added to the reaction mixture followed by stirring for 33 h. Half saturated NH_4Cl (30 mL) was added to quench the reaction and the organic layer was extracted with EtOAc, dried (Na_2SO_4) and concentrated to provide the crude product, which was purified through a short plug of silica gel (EtOAc→Acetone) to isolate **207** as a pale yellow solid (5.52 g, 77%): m.p = 132-134 ºC; ^{1}H NMR: δ = 9.82 (s, 1H), 7.42 (d, *J* = 7.3 Hz, 2H), 7.38 (s, 1H), 7.29 (t, *J* = 7.3 Hz, 2H), 7.25 (m, 1H), 6.50 (s, 2H), 6.23 (s, 1H), 4.94 (s, 2H), 3.71 (s, 6H), 3.47 (s, 3H); ^{13}C NMR: δ = 188.4, 153.8, 141.7, 139.9, 138.0, 137.7, 136.6, 136.2, 128.5, 128.2, 127.9, 103.4, 75.1, 66.7, 56.3, 33.1; IR (neat, cm^{-1}): = 3253 (br), 3106, 2938, 1680, 1587, 1502, 1450, 1415, 1230, 1100, 1056, 824 ; HR-DARTMS (*m/z*): Calcd. for $C_{21}H_{23}N_2O_5$ $[M+H]^+$ 383.1601, found 383.1597.

5-(4-Benzyloxy-3,5-dimethoxyphenyl)hydroxymethyl-4-[hydroxyl-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (208):



Following the general procedure for this Grignard reaction, *p*-bromoanisole, (7.23 mL, 56.5 mmol), magnesium turnings (1.35 g, 56.5 mmol) and a small crystal of iodine in THF (100 mL) and a solution of **207** (5.40 g, 14.1 mmol) in THF (50 mL) were used to obtain a crude product, which was purified through a short plug of silica gel (EtOAc) to isolate **208** (4.38 g, 63%) as a pale yellow oil, which was used in the next step directly without further characterization.

6-Benzyloxy-5,7-dimethoxy-4-(4-methoxyphenyl)-1-methyl-1*H*-naphtho[2,3-*d*]imidazole (209):



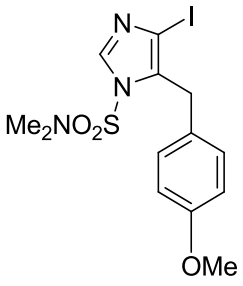
Et_3SiH (11.41 mL, 71.4 mmol) and TFA (4.81 mL, 62.5 mmol) were added to a solution of **208** (4.38 g, 8.9 mmol) in anhydrous CH_2Cl_2 (100 mL) at rt. and the resulting mixture was stirred for 24 h under nitrogen atmosphere. Then, the reaction was quenched by the addition of satd. aq. solution of $NaHCO_3$. The aqueous layer was extracted with CH_2Cl_2 several times and the combined extracts were dried (Na_2SO_4), and concentrated. The residue was purified by chromatography (EtOAc→ acetone) to provide **209** (3.30 g, 81%) as a pale brown solid; m.p = 194-197 ºC; ^{1}H NMR: δ = 7.87 (s, 1H), 7.61 (s, 1H), 7.53 (d, *J* = 8.3 Hz, 2H), 7.45 (d, *J* = 8,7 Hz, 2H), 7.36 (t, *J* = 7.3 Hz, 2H), 7.30 (t, *J* = 7.3 Hz, 1H), 7.08 (s, 1H), 7.05 (d, *J* = 8.7 Hz, 2H), 5.10 (s,

2H), 3.96 (s, 3H), 3.89 (s, 3H), 3.74 (s, 3H), 3.37 (s, 3H); ^{13}C NMR: δ = 158.2, 152.1, 150.6, 146.6, 142.9, 139.8, 138.0, 134.4, 132.4, 131.6, 130.9, 129.7, 128.4, 128.3, 127.9, 119.6, 112.7, 104.0, 102.3, 75.2, 60.9, 55.8, 55.4, 31.0; IR (neat, cm^{-1}): = 2929, 2831, 1607, 1514, 1451, 1330, 1274, 1236, 1145, 1075, 1027, 826, 740 ; HR-DARTMS (*m/z*): Calcd. for $C_{28}H_{27}N_2O_4$ $[M+H]^+$ 455.1971, found 455.1983.

***p*-Methoxybenzylbromide** was prepared from the bromination of *p*-methoxybenzyl alcohol using Schiller's protocol.[122]

4,5-diiodo-1-(*N*,*N*-dimethylsulfonyl)-1H-imidazole (210) was prepared from 4,5-diiodo-1*H*-imidazole following previous work done by our group.[50]

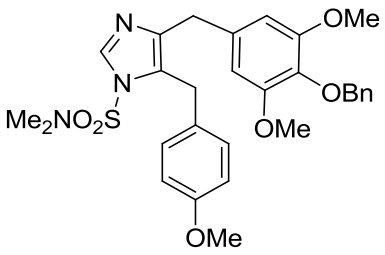
1-(*N*,*N*-dimethylsulfonyl)-4-iodo-5-(4-methoxybenzyl)-1*H*-imidazole (211):



EtMgBr (3.0 M solution in ether, 8.60 mL, 25.8 mmol) was added to a solution of **210** (7.19 g, 21.5 mmol) in dry CH_2Cl_2 (150 mL) at rt. The resulting mixture was stirred at rt. for 20 min and 1.0 M solution of CuCN.2LiCl in dry THF (26.0 mL, 26.0 mmol) was added followed by *p*-methoxybenzyl bromide (3.80 mL, 25.8 mmol). The orange reaction solution was stirred at rt. for 48 h and poured into half sat. NH_4Cl containing 2% concentrated NH_3 (50 mL). After stirring for 20 min, the resulting solid was filtered off and the filtrate was partitioned with CH_2Cl_2 (3x50 mL). The organic layer was dried (Na_2SO_4), concentrated and purified by chromatography (EtOAc/hexane, 3:7) to afford **211** (6.41 g, 65%) as a pale yellow solid: m.p = 76-78 ºC; ^{1}H NMR: δ = 7.87 (s. 1H), 6.99

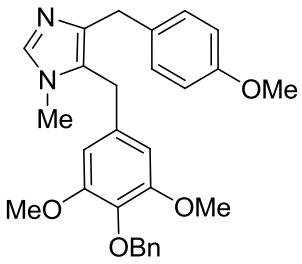
(d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 6.37 (s, 2H), 4.09 (s, 2H), 3.71 (s, 3H), 2.49 (s, 6H); ^{13}C NMR: δ = 158.5, 139.7, 137.9, 132.5, 129.1, 114.0, 90.6, 55.4, 37.6, 29.9; IR (neat, cm^{-1}): = 3111, 2919, 1514, 1459, 1415, 1240, 1173, 1174, 1095, 960, 802 ; HR-DARTMS (*m/z*): Calcd. for $C_{13}H_{17}IN_3O_3S$ $[M+H]^+$ 422.0030, found 422.0047.

4-(4-Benzyloxy-3,5-dimethoxybenzyl)-1-(*N*,*N*-dimethylsulfonyl)-5-(4-methoxybenzyl)-1*H*-imidazole (212):



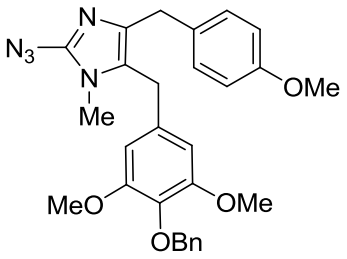
Following the above procedure, EtMgBr (3.0 M solution in ether, 4.98 mL, 14.9 mmol), **211** (5.72 g, 13.6 mmol) in dry CH_2Cl_2 (150 mL), 1.0 M solution of CuCN.2LiCl in dry THF (16.3 mL, 16.3 mmol) and **206** (6.87 g, 20.4 mmol) were used to synthesize **212** (5.18 g, 70%) as a pale yellow oil after the purification by chromatography (EtOAc:hexane, 1:1): ^{1}H NMR: δ = 7.94 (s. 1H), 7.48 (d, *J* = 7.3 Hz, 2H), 7.33 (t, *J* = 7.3 Hz, 2H), 7.27 (t, *J* = 7.33 Hz, 1H), 6.94 (d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 6.37 (s, 2H), 4.94 (s, 2H), 4.14 (s, 2H), 3.78 (s, 2H), 3.75 (s, 3H), 3.71 (s, 6H), 2.57 (s, 6H); ^{13}C NMR: δ = 158.4, 153.4, 141.3, 138.1, 138.0, 135.6, 134.8, 130.3, 129.9, 128.9, 128.5, 128.2, 127.8, 114.0, 106.0, 75.1, 56.1, 55.4, 37.5, 34.0, 28.1; IR (neat, cm^{-1}): = 2929, 2857, 1691, 1507, 1393, 1252, 1124, 909, 836, 779 ; HR-DARTMS (*m/z*): Calcd. for $C_{29}H_{34}N_3O_6S$ $[M+H]^+$ 552.2163, found 552.2180.

5-(4-Benzyloxy-3,5-dimethoxybenzyl)-4-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (214):



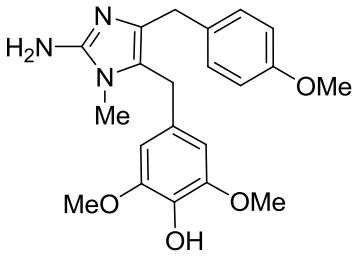
Methyl trifluoromethanesulfonate (0.95 mL, 8.7 mmol) was added dropwise to a solution of **212** (3.96 g, 7.2 mmol) in CH_2Cl_2 (50 mL), at 0 ºC under N_2 and stirred for 4 h at the same temperature.[114] Then, the solvent was evaporated under reduced pressure and the crude pale yellow oil was dissolved in dry acetonitrile (30 mL), and benzylamine (0.95 mL, 8.7 mmol) was added to it. After heating at 80 ºC for 10 h,[113] solvent was evaporated to provide a crude oil, which was purified with a gradient column (EtOAc:hexanes, 3:1→EtOAc:acetone; 1:1) to isolate **214** (2.96 g, 90%) as a pale brown oil: ^{1}H NMR: δ = 7.45 (d, *J* = 7.3 Hz, 2H), 7.40 (s, 1H), 7.31 (t, *J* = 7.3 Hz, 2H), 7.27 (t, *J* = 7.3 Hz, 1H), 7.19 (d, *J* = 8.7 Hz, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 6.15 (s, 2H), 4.96 (s, 2H), 3.88 (s, 2H), 3.87 (s, 2H), 3.75 (s, 3H), 3.64 (s, 6H), 3.34 (s, 3H); ^{13}C NMR: δ = 157.9, 153.8, 138.8, 137.9, 136.8, 135.5, 134.1, 133.1, 129.5, 128.5, 128.2, 127.9, 125.5, 113.9, 105.1, 75.1, 56.1, 55.3, 32.8, 31.9, 29.5; IR (neat, cm^{-1}): = 2929, 2857, 1691, 1507, 1391, 1251, 1176, 1150, 910; HR-ESIMS (*m/z*): Calcd. for $C_{28}H_{31}N_2O_4$ $[M+H]^+$ 459.2278, found 459.2278.

2-Azido-5-(4-benzyloxy-3,5-dimethoxybenzyl)-4-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (216):



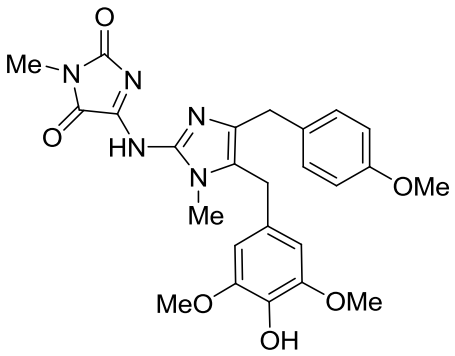
n-Butyl lithium (1.6 M solution in hexanes, 1.31 mL, 2.1 mmol) was added dropwise to a stirred solution of **214** (870 mg, 1.9 mmol) in dry THF (20 mL) at -78 °C, and the reaction was stirred for 1h. The cooling bath was removed for 10 min, then the reaction mixture was re-cooled to -78 °C, and then $TrisN_3$ (706 mg, 2.3 mmol) was added. After stirring for an additional 45 min at -78 °C, the reaction mixture was quenched by the addition of satd. aq. NH_4Cl (5 mL). The aqueous layer was extracted with EtOAc (3x15 mL), and the combined organic extracts were dried (Na_2SO_4) and concentrated to provide a pale brown oil, which was purified through a short column of silica gel (hexane/EtOAc, 7:3) to isolate azide **216** (600 mg, 63%) as a pale brown oil: ^{1}H NMR: δ = 7.46 (d, *J* = 7.4 Hz, 2H), 7.38-7.26 (m, *J* = 7.4 Hz, 3H), 7.21 (d, *J* = 8.5 Hz, 2H), 6.79 (d, *J* = 8.5 Hz, 2H), 6.16 (s, 2H), 4.95(s, 2H), 3.85 (s, 2H), 3.80 (s, 2H), 3.76 (s, 3H), 3.64 (s, 6H), 3.08 (s, 3H); ^{13}C NMR: δ = 158.0, 153.7, 139.2, 137.9, 136.5, 135.5, 134.0, 132.9, 129.5, 128.6, 128.2, 127.9, 124.6, 113.9, 105.0, 75.0, 56.1, 55.3, 32.7, 30.0, 29.6; IR (neat, cm^{-1}): = 2929, 2857, 2129, 1691, 1507, 1391, 1252, 1150, 1124, 909, 836, 779 ; HR-ESIMS (*m/z*): Calcd. for $C_{28}H_{30}N_5O_4$ $[M+H]^+$ 500.2292, found 500.2290.

2-Amino-5-(3,5-dimethoxy-4-hydroxybenzyl)-4-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (1h): Naamine G



Azide **216** (600 mg, 1.2 mmol) was dissolved in EtOH (15 mL) and stirred overnight under a hydrogen atmosphere (55 psi) in the presence of 20% $Pd(OH)_2$ on charcoal (100 mg) at rt. The catalyst was filtered through a pad of Celite and the filtrate was concentrated to isolate naamine G, **1h** (430 mg, 95%) as a greenish-yellow solid; m.p = 218-220 °C; ^{1}H NMR (CD_3OH): δ = 7.17 (d, *J* = 8.7 Hz, 2H), 6.84 (d, *J* = 8.7 Hz, 2H), 6.34 (s, 2H), 3.92 (s, 2H), 3.84 (s, 2H), 3.74 (s, 3H), 3.69 (s, 6H), 3.23 (s, 3H); ^{13}C NMR: δ = 158.8, 148.3, 146.5, 134.3, 129.8, 129.2, 127.3, 122.8, 122.4, 114.0, 105.2, 55.5, 54.5, 28.8, 28.3, 27.9; IR (neat, cm^{-1}): = 3244 (br), 3004, 2836, 1667, 1654, 1609, 1500, 1461, 1429, 1245, 1216, 1110, 1022; HR-DARTMS (*m/z*): Calcd. for $C_{21}H_{26}N_3O_4$ $[M+H]^+$ 384.1918, found 384.11907.

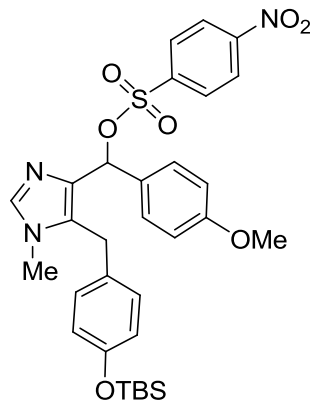
5-(3,5-Dimethoxy-4-hydroxybenzyl)-4-(4-methoxybenzyl)-1-methyl-2-(3-methylimidazolidine-2,4-dione)imino-1*H*-imidazole (2g): Naamidine H



Following the general procedure for this reaction, *N*,*O*-Bis(trimethysilyl)acetamide (0.63 mL, 2.6 mmol) and 1-methylparabanic acid (331 mg, 4.1 mmol) in dry CH_3CN (10 mL) were used to produce 3-trimethylsilyl-1-methylparabanic acid. After removing the solvent, naamine G, **1h** (198 mg, 0.5 mmol) was added and the mixture was heated at

80 º C overnight in dry toluene (5 mL). Usual workup and purification over silica gel (EtOAc/hexanes, 4/6) provided naamidine H (**2g**) as a yellow amorphous solid (205 mg, 80%): m.p = 204-205 ºC; ^{1}H NMR: δ = 7.14 (d, *J* = 8.7 Hz, 2H), 6.98 (br, 1H), 6.78 (d, *J* = 8.7 Hz, 2H), 6.14 (s, 2H), 3.89 (s, 2H), 3.88 (s, 2H), 3.75 (s, 3H), 3.69 (s, 6H), 3.49 (s, 3H), 3.47 (s, 3H), 3.16 (s, 3H); ^{13}C NMR: δ = 162.3, 158.3, 155.5, 147.4, 146.6, 144.7, 136.1, 133.7, 131.7, 129.4, 128.1, 126.7, 114.1, 104.7, 56.3, 55.4, 32.3, 30.0, 29.7, 24.8; IR (neat, cm^{-1}): = 3501, 3212, 2929, 2837, 1784, 1718, 1652, 1511, 1392, 1113, 1039, 1020, 918 ; HR-DARTMS (*m/z*): Calcd. for $C_{25}H_{28}N_5O_6$ $[M+H]^+$ 494.2034, found 494.2049.

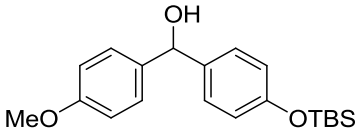
4-Nitrobenzenesulfonyl {5-[4-*tert*-butyldimethylsilanyloxybenzyl]-1-methyl-1*H*-imidazol-4-yl}-(4-methoxyphenyl)methanoate (79c):



DIEA (0.12 mL, 0.7 mmol) and 4-nitrophenylsulfonyl chloride (152 mg, 0.7 mmol) were added to a solution of **77** (200 mg, 0.5 mmol) in dry CH_2Cl_2 (8 mL) at rt. and stirred for 22 h. After evaporating the solvents, the residue was diluted with water and extracted with EtOAc. The organic phase was washed with dil. HCl, half satd. aq. $NaHCO_3$ and brine respectively. After drying over anhyd. Na_2SO_4, it was concentrated and the residue was purified over silica gel using 100% EtOAc to isolate **79c** (36 mg, 10%) as a pale yellow solid: m.p = 182-185 °C; ^{1}H NMR : δ = 8.17 (d, *J* = 8.8 Hz, 2H), 7.72 (d, *J* = 8.8 Hz, 2H), 7.51 (d, *J* = 8.8 Hz, 2H), 7.43 (s, 1H), 6.87 (d, *J* = 8.5 Hz, 2H), 6.78 (d, *J* = 8.8 Hz, 2H), 6.71 (d, *J* = 8.5 Hz, 2H), 5.25 (s, 1H), 3.98 (d, *J* = 17.1 Hz, 1H), 3.90 (d, *J* = 17.1 Hz,

1H), 3.78 (s, 3H), 3.37 (s, 3H), 0.96 (s, 9H), 0.16 (s, 1H); ^{13}C NMR: δ =160.2, 154.7, 150.5, 143.8, 137.8, 132.3, 131.5, 131.0, 130.6, 129.4, 129.3, 123.8, 123.2, 120.5, 113.8, 69.3, 55.4, 32.1, 28.6, 25.7, 18.3, -4.3; IR (neat, cm^{-1}) = 2931, 2858, 1607, 1531, 1510, 1349, 1305, 1255, 1146, 914, 841, 782, 733; HR-ESIMS (*m/z*): Calcd. for $C_{31}H_{38}N_3O_7SSi$ $[M+H]^+$ 623.2121, found 623.2130.

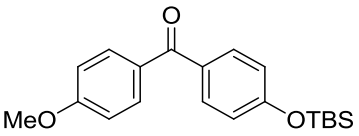
4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxybenzyl alcohol (227):[156]



A few drop of *p*-bromoanisole (from 3.17 mL, 25.4 mmol) was added drop wise to a two neck round bottom flask containing, freshly crushed oven dried, magnesium turnings (0.61 g, 25.4 mmol) and a small crystal of iodine in THF (30 mL). This mixture was then heated at 45 °C under nitrogen until the iodine color faded. The rest of the *p*-bromoanisole was added drop wise while maintaining a gentle reflux. After addition was completed, the mixture was heated to reflux for 1 h and then, cooled to rt. Then, a solution of **146** (1.50 g, 6.3 mmol) in THF (10 mL) was added to it followed by stirring at reflux for 12 h. Then, it was cooled to 0 ° C; 20 mL of satd. aq. NH_4Cl was added and the organic layer was extracted with EtOAc (3x25 mL), washed once with brine, dried over anhyd. Na_2SO_4 and concentrated to afford thick yellow oil, which was purified by a short plug of silica gel with 10% EtOAc in hexanes to isolate above alcohol (2.18 g, quant) as a yellow solid: m.p = 76-79 ºC; ^{1}H NMR: δ = 7.27 (d, *J* = 8.8 Hz, 2H), 7.26 (s, 1H), 7.21 (d, *J* = 8.8 Hz, 2H), 6.86 (d, *J* = 8.8 Hz, 2H), 6.79 (d, *J* = 8.8 Hz, 2H), 5.75 (d, *J* = 3.3 Hz, 1H), 3.79 (s, 3H), 2.12 (d, *J* = 3.3 Hz, 1H), 0.97 (s, 9H), 0.18 (s, 6H); ^{13}C NMR: δ = 159.0, 155.1, 136.9, 136.4, 127.9, 127.8, 120.0, 113.9, 75.5, 55.4, 25.8, 18.3, -4.3; IR (neat, cm^{-1}): = 3379(br),

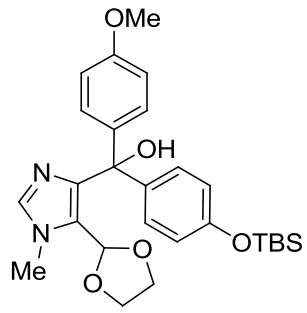
2955, 2930, 2858, 1608, 1509, 1251, 1169, 1035, 838, 780: HR-ESIMS (*m/z*): = Calcd. for $C_{20}H_{22}O_2Si$ $[M-H_2O]^+$ 327.1775 found 327.1777; Calcd. for $C_{20}H_{28}O_3NaSi$ $[M+Na]^+$ 367.1700, found 367.1678.

4-*tert*-Butyldimethylsilanyloxyphenyl-(4-methoxyphenyl)methanone (228):



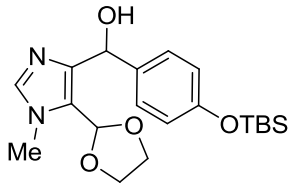
Manganese dioxide (2.27 g, 26.1 mmol) was added to a solution of **227** (0.90 g, 2.6 mmol) in dichloromethane (15 mL) and the resulting suspension was heated to reflux for 18 h. After cooling, the suspension was filtered through a pad of Celite and the Celite pad was washed with dichloromethane (50 mL). Then, the combined organics were concentrated in *vacuo* to isolate ketone **228** (0.80 g, 90%) as a pale yellow oil: ^{1}H NMR: δ = 7.79 (d, *J* = 8.8 Hz, 2H), 7.72 (d, *J* = 8.5 Hz, 2H), 6.95 (d, *J* = 8.8 Hz, 2H), 6.89 (d, *J* = 8.5 Hz, 2H), 3.88 (s, 3H), 1.00 (s, 9H), 0.25 (s, 6H); ^{13}C NMR: δ = 194.7, 162.9, 159.6, 132.3, 132.2, 131.4, 130.8, 119.7, 113.5, 55.6, 25.7, 18.3, -4.2; IR (neat, cm^{-1}): = 2956, 2930, 2858, 1650, 1601, 1507, 1304, 1257, 1163, 1031, 911, 840, 783; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{27}O_3Si$ $[M+H]^+$ 343.2693, found 343.1693.

4-*tert*-Butyldimethysilanyloxyphenyl-(5-[1,3]dioxolan-2-yl-1-methyl-1*H*-imidazol-4-yl)-(4-methoxy-phenyl)-methanol (229):



A solution of EtMgBr (3.0 M in ether, 0.74 mL, 2.2 mmol) was added to a solution of **179** (596 mg, 2.1 mmol) in dry THF (5 mL) at rt. over 3 min. The resulting mixture was stirred at rt. until all the starting material consumed, and then **228** (802 mg, 2.3 mmol) in dry THF (3 mL) was added followed by stirring for 24 h. Saturated aq. NH_4Cl (2 mL) was added to quench the reaction and the organic layer was extracted with EtOAc (2x5 mL) and washed once with brine. The EtOAc solution was dried (anhyd. Na_2SO_4) and concentrated to provide the crude product, which was purified by a short plug of silica gel (10%→20%→40% EtOAc in hexanes) to provide **229** (153 mg, 14%) a pale yellow solid; m.p = 48-50 ºC; ^{1}H NMR: δ = 7.37 (s, 1H), 7.23 (d, *J* = 8.8 Hz,2H), 7.15 (d, *J* = 8.8 Hz,2H), 6.83 (d, *J* = 8.8 Hz,2H), 6.75 (d, *J*= 8.8 Hz, 2H), 4.95 (s, 1H), 4.77 (s, 1 H), 3.96-3.91 (m, 2H), 3.78 (s, 3H), 3.70-3.67 (m, 2H), 3.67 (s, 3H), 0.97 (s, 9H), 0.17 (s, 6H); ^{13}C NMR: δ = 158.8, 155.0, 149.4, 139.1, 138.5, 138.1, 129.3, 129.2, 121.5, 119.4, 113.2, 97.3, 77.8, 64.9, 64.9, 55.3, 33.5, 25.8, 18.3, -4.3; IR (KBr, cm-1): = 3227 (br), 3119, 2955, 2932, 2855, 1607, 1508, 1390, 1253, 1167, 1085, 1068, 912, 834; HR-ESIMS (*m/z*): Calcd. for $C_{27}H_{37}N_2O_5$ $[M+H]^+$ 497.2466, found 497.2403; Calcd. for $C_{27}H_{36}N_2NaO_5$ $[M+Na]^+$ 519.2286, found 519.2323.

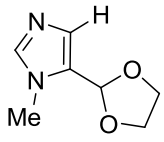
4-*tert*-Butyl-dimethylsilanyloxyphenyl-(5-[1,3]dioxolan-2-yl-1-methyl-1H-imidazol-4-yl)methanol (230):



A 3.0 M solution of EtMgBr in ether (2.98 mL, 8.9 mmol) was instilled into a solution of **179** (2.27 g, 8.1 mmol) in dry THF (20 mL) at rt over 10 min. After stirring for 20 min further, a solution of **146** (4.79 g, 20.3 mmol) in dry THF (10 mL) was added to above Grignard species at rt. After stirring for 24 h, satd. aq. NH_4Cl (5 mL) was added to quench the reaction and the organic layer was extracted with EtOAc (2x15 mL) and washed once with brine. Combined organic layers were dried over anhyd. Na_2SO_4 and concentrated to provide a crude product, which was purified by using a gradient column chromatography (100% EtOAc →10% MeOH in EtOAc) to isolate a mixture of **230** (3.05 g, 96%) and **231** (50 mg, 4%) as a pale yellow oil.

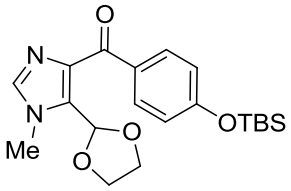
Data for 230: ^{1}H NMR (CD_3OH): δ = 7.53 (s, 1H), 7.24 (d, *J* = 8.3 Hz, 2H), 7.75 (d, *J*= 8.3 Hz, 2H), 6.14 (s, 1 H), 5.82 (s, 1H), 4.11 (m, 2H), 3.95 (m, 2H), 3.69 (s, 3H), 0.97 (s, 9H), 0.15 (s, 6H).

5-[1,3]dioxolan-2-yl-1-methyl-1*H*-imidazole (231):



From above reaction, ^{1}H NMR (CD_3OH): δ = 7.60 (s, 1H), 7.03 (s, 1H), 5.88 (s, 2H), 4.11-4.08 (m, 2H), 3.99-3.97 (m, 2H), 3.69 (s, 3H).

4-*tert*-Butyldimethylsilanyloxyphenyl-(5-[1,3]dioxolan-2-yl-1-methyl-1H-imidazol-4-yl)methanone (232):



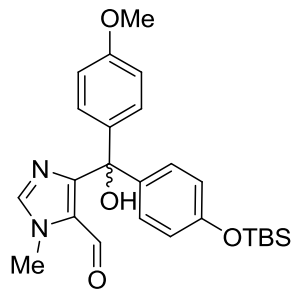
Manganese dioxide (6.90 g, 78.1 mmol) was added to a solution of alcohol **230** (3.05 g, 8.8 mmol) in dichloromethane (30 mL) and the resulting suspension was heated under reflux for 24 h. After cooling to rt, solid particles were filtered through a pad of Celite, and the Celite was washed with dichloromethane (30 mL) and the combined organic layers were concentrated under reduce pressure to provide the ketone **232** (2.54 g, 84%) as pale yellow solid: m.p = 135-136 ºC; ^{1}H NMR: δ = 8.15 (d, *J* = 8.8 Hz, 2H), 7.44 (s, 1H), 7.88 (d, *J*= 8.8 Hz, 2H), 6.54 (s, 1 H), 4.21-4.13 (m, 2H), 4.10-4.04 (m, 2H), 3.79 (s, 3H), 0.98 (s, 9H), 0.22 (s, 6H); ^{13}C NMR: δ = 188.0, 159.9, 141.7, 138.8, 133.0, 131.6, 131.3, 119.6, 96.9, 65.3, 33.6, 25.7, 18.3, -4.3; IR (KBr, cm^{-1}): = 3126, 3057, 2955, 2894, 2855, 1638, 1594, 1565, 1504, 1467, 1377, 1266, 1221, 1162, 1063, 950, 910, 782, 654; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{29}N_2O_4Si$ $[M+H]^+$ 389.1891, found 389.1914; Calcd. for $C_{20}H_{28}N_2NaO_4Si$ $[M+Na]^+$ 411.1711, found 411.1736.

Synthesis of 229 from 232:

A few drops of *p*-bromoanisole (from 3.27 mL, 26.2 mmol) was added drop wise to a two neck round bottom flask containing, freshly crushed oven dried, magnesium turnings (0.63 g, 26.2 mmol) and a small crystal of iodine in THF (25 mL). This mixture was then heated at 45 °C under nitrogen until the iodine color faded. The rest of the *p*-bromoanisole was added drop wise maintaining a gentle reflux. After addition was completed, the mixture was heated to reflux for 1 h and cooled to rt.

Then, a solution of **232** (2.54 g, 6.5 mmol) in THF (10 mL) was added to the above mixture. The resulting mixture was stirred at reflux for overnight and cooled to 0 ° C; 20 mL of satd. aq. NH_4Cl was added and the organic layer was extracted with EtOAc (3x25 mL), washed once with brine, dried over anhyd. Na_2SO_4 and concentrated to give thick brown oil, which was purified by a short plug of silica gel (75% EtOAc in hexanes) to isolate **229** (3.20 g, 99%).

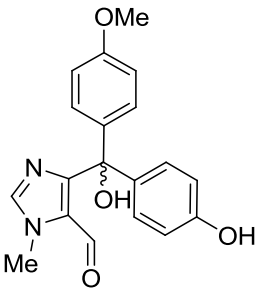
4-[(4-*tert*-Butyldimethylsilanyloxyphenyl)-hadroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole-5-carbaldehyde (233a):



A 10% solution of HCl (10 mL) was added to **229** (6.29 g, 12.7 mmol) in THF (100 mL) and the resulting cloudy reaction was stirred at rt. overnight. Then, the reaction was neutralized with satd. aq. $NaHCO_3$ and the aqueous layer was extracted with EtOAc (2x 50 mL), and combined organic layers were dried over anhyd. Na_2SO_4 and concentrated to provide the crude material, which was purified over silica gel (1:1→3:1 EtOAc in hexanes) to isolate **233a** (3.07 g, 54%) and **233b** (1.71 g, 40%).

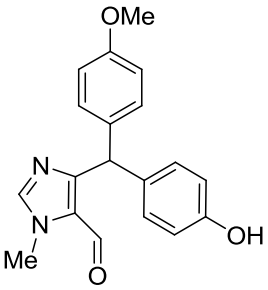
Data for 233a: A pale yellow solid: m.p = 52-55 °C; ^{1}H NMR: δ = 9.12 (s, 1H), 7.45 (s, 1H), 7.22 (d, *J* = 8.7 Hz, 2H), 7.15 (d, *J* = 8.7 Hz, 2H), 6.83 (d, *J* = 8.7 Hz, 2H), 6.76 (d, *J* = 8.7 Hz, 2H), 4.55 (s, 1H), 3.90 (s, 3H), 3.78 (s, 3H), 0.96 (s, 9H), 0.18 (s, 6H); ^{13}C NMR: δ = 182.5, 159.5, 159.1, 155.3, 141.2, 138.4, 137.9, 129.1, 129.1, 127.5, 119.6, 113.5, 78.7, 55.3, 35.1, 25.7, 18.3, -4.3; IR (KBr, cm^{-1}): = 3212 (br), 2955, 2857, 1665, 1606, 1513, 1467, 1389, 1249, 1170, 1036, 914, 841, 780; HR-ESIMS (*m/z*): Calcd. for $C_{25}H_{32}N_2NaO_4Si$ $[M+Na]^+$ 475.2029, found 475.2057.

4-[Hydroxy-(4-hydroxyphenyl)-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole-5-carbaldehyde (233b):



Data for 233a: A pale yellow solid: m.p = 94-96 °C; ^{1}H NMR: δ = 9.16 (s, 1H), 7.51 (s, 1H), 7.23 (d, *J* = 8.7 Hz, 2H), 7.07 (d, *J* = 8.7 Hz, 2H), 6.83 (d, *J* = 8.7 Hz, 2H), 6.61 (d, *J* = 8.7 Hz, 2H), 4.44 (s, 1H), 3.91 (s, 3H), 3.78 (s, 3H); ^{13}C NMR: δ = 182.8, 159.6, 159.2, 155.6, 141.2, 137.6, 137.4, 129.2, 129.0, 127.6, 115.2, 113.5, 78.9, 55.3, 35.2; IR (KBr, cm^{-1}): = 3306 (br), 3008, 1956, 1857, 1660, 1609, 1509, 1349, 1251, 1172, 1013, 915, 832; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{18}N_2NaO_4$ $[M+Na]^+$ 361.1159, found 361.1175.

4-[4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl]methyl-1-methyl-1*H*-imidazole-5-carbaldehyde (234) from **233b:**

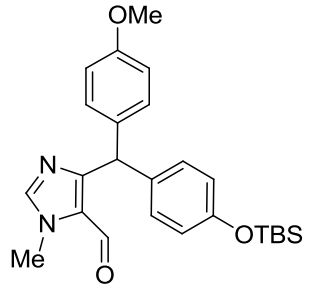


TFA (2.09 mL, 27.2 mmol) was added to a solution of **233b** (1.71 g, 5.1 mmol) in dry CH_2Cl_2 (20 mL), and to the resulting blood-red reaction was added Et_3SiH (4.35 mL, 27.2 mmol) at rt. After stirring for 19 h, reaction mixture was neutralized with satd. aq. $NaHCO_3$ and the aqueous layer was extracted with CH_2Cl_2 (3x15 mL), and combined organic layers were dried over anhyd. Na_2SO_4 and concentrated to provide the crude product, which was purified over silica gel (1:1→3:1 EtOAc in hexanes) to isolate **234** (1.08 g, 61%) as pale yellow solid: m.p = 81-84 ºC; ^{1}H NMR: δ = 9.67 (s, 1H), 7.53 (s, 1H), 7.15 (d, *J* = 8.7 Hz, 2H), 6.92 (d, *J* = 8.7 Hz, 2H), 6.80 (d, *J* = 8.7 Hz, 2H), 6.52 (d, *J* = 8.7 Hz, 2H), 5.74 (s, 1H), 3.85 (s, 3H), 3.73 (s, 3H); ^{13}C NMR: δ =

179.9, 158.4, 157.9, 155.6, 142.7, 134.2, 132.9, 130.0, 129.8, 127.2, 115.8, 114.1, 55.3, 47.9, 34.8; IR (KBr, cm^{-1}): = 3110 (br), 2975, 2836, 1665, 1610, 1511, 1446, 1352, 1249, 1176, 1032, 823; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{19}N_2O_3$ $[M+H]^+$ 323.1390, found 323.1404; Calcd. for $C_{19}H_{18}N_2NaO_3$ $[M+Na]^+$ 345.1210, found 345.1229.

Synthesis of 234 from **233a:** Following the above procedure, TFA (2.04 mL, 26.5 mmol), **233a** (3.00 g, 6.6 mmol) in dry CH_2Cl_2 (20 mL) and Et_3SiH (4.23 mL, 26.5 mmol) were used to synthesize **234** (1.70 g, 80%).

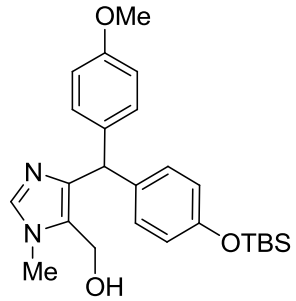
5-[4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl]methyl-1-methyl-1*H*-imidazole-4-carbaldehyde (226):



NaH (60% in mineral oil, 0.19 g, 4.8 mmol) was added to a solution of **234** (1.30 g, 4.0 mmol) in dry THF (25 mL) at rt. After stirring for 25 min, TBSCl (0.70 g, 4.4 mmol) was added to the reaction mixture and stirred overnight. Water (2 mL) was added to the reaction and extracted with EtOAc (3x15 mL). Combined organic layers were dried (Na_2SO_4) and concentrated to provide crude material, which was purified over silica gel (3:7 →7:3 EtOAc in hexanes) to isolate **226** (1.21 g, 70%) as a pale yellow oil: ^1H NMR: δ = 9.76 (s, 1H), 7.54 (s, 1H), 7.17 (d, *J* = 8.7 Hz, 2H), 7.10 (d, *J* = 8.7 Hz, 2H), 6.82 (d, *J* = 8.7 Hz, 2H), 6.75 (d, *J* = 8.7 Hz, 2H), 5.74 (s, 1H), 3.88 (s, 3H), 3.76 (s, 3H), 0.96 (s, 9H), 0.17 (s, 6H); ^{13}C NMR: δ = 179.5, 158.4, 157.9, 154.4, 142.9, 135.0, 134.6, 130.0, 129.9, 127.2, 120.0, 114.0, 55.3, 47.9, 34.6, 25.7, 18.2, -4.3; IR (KBr, cm^{-1}): = 2955, 2930, 2857, 1665, 1508, 1252, 1174, 1035, 915, 839, 782; HR-ESIMS (*m/z*):

Calcd. for $C_{25}H_{33}N_2O_3Si$ $[M+H]^+$ 437.2255, found 437.2269; Calcd. for $C_{25}H_{32}N_2NaO_3Si$ $[M+Na]^+$ 459.2074, found 459.2040.

4-(4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl)methyl-5-hydroxymethyl-1-methyl-1*H*-imidazole (225):



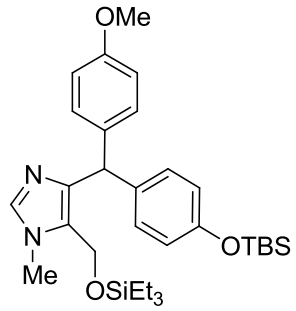
$NaBH_4$ (50 mg, 1.3 mmol) was added to a solution of **226** (1.13 g, 2.6 mmol) in 30:1 mixture of THF-H_2O (15.5 mL). After stirring at rt. for 5 min, water was added to quench the reaction and the organic layer was extracted with EtOAc (2x15 mL). The combined organic layers were dried over Na_2SO_4 and concentrated to provide **225** (1.11 g, quant) as a white solid: m.p = 72-74 ºC; ^{1}H NMR: δ = 7.34 (s, 1H), 7.12 (d, *J* = 8.7 Hz, 2H), 7.05 (d, *J* = 8.7 Hz, 2H), 6.80 (d, *J* = 8.7 Hz, 2H), 6.73 (d, *J* = 8.7 Hz, 2H), 5.40 (s, 1H), 4.39 (s, 2H), 3.75 (s, 3H), 3.62 (s, 3H), 0.95 (s, 9H), 0.15 (s, 6H); ^{13}C NMR: δ = 158.1, 154.2, 142.6, 137.6, 136.3, 135.7, 129.9, 129.8, 127.1, 199.9, 113.8, 55.3, 53.2, 48.1, 31.7, 25.7, 18.2, -4.3; IR (KBr, cm^{-1}): = 2955, 2930, 2857, 1665, 1508, 1252, 1174, 1035, 915, 839, 782; HR-ESIMS (*m/z*): Calcd. for $C_{25}H_{35}N_2O_3Si$ $[M+H]^+$ 439.2411, found 439.2435; Calcd. for $C_{25}H_{34}N_2NaO_3Si$ $[M+Na]^+$ 461.2231, found 461.2258.

Synthesis of 225 from **229:**

A mixture of **229** (3.00 g, 6.0 mmol), $BF_3.OEt_2$ (3.83 mL, 30.2 mmol) and Et_3SiH (9.65 mL, 60.4 mmol) in dichloromethane was stirred at rt. for 24 h under argon atmosphere. After addition of triethylamine (3 mL), the reaction mixture was filtered through a pad of Celite and the filtrate was concentrated. The resulting residue was

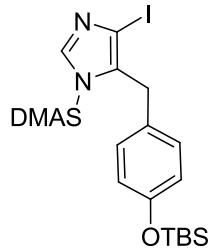
purified by flash column chromatography on silica gel (EtOAc) to isolate **225 (**306 mg, 11%) and **235** (697 mg, 21%).

4-(4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl)methyl-5-triethylsilanyloxymethyl-1-methyl-1*H*-imidazole (235):



From above reaction, **236** light orange oil; ^{1}H NMR: δ = 7.93 (s, 1H), 7.04 (d, *J* = 8.8 Hz, 2H), 6.98 (d, *J* = 8.5 Hz, 2H), 6.82 (d, *J* = 8.8 Hz, 2H), 6.77 (d, *J* = 8.5 Hz, 2H), 5.49 (s, 1H), 4.30 (s, 2H), 3.77 (s, 3H), 3.76 (s, 3H), 3.19 (q, *J* = 7.2 Hz, 6H), 1.35 (t, *J* = 7.2 Hz, 9H), 0.95 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 158.9, 155.1, 136.0, 135.6, 132.9, 132.2, 129.9, 129.8, 120.5, 114.4, 55.4, 52.2, 47.3, 46.1, 33.6, 25.7, 18.2, 8.8, -4.4; IR (neat, cm^{-1}): = 3527, 3157, 2930, 2857, 1608, 1560, 1447, 1254, 1024, 914, 759; HR-ESIMS (*m/z*): Calcd. for $C_{31}H_{49}N_2O_3Si_2$ $[M+H]^+$ 553.3203, found 325.1545, consistence with double desilylated product; $C_{19}H_{21}N_2O_3$ $[M+H]^+$ 325.1552.

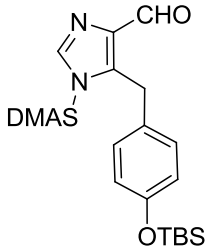
5-(4-*tert*-Butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-4-iodo-1*H*-imidazole (236)



EtMgBr (3.0 M solution in ether, 6.44 mL, 19.3 mmol) was added to a solution of **210** (7.50 g, 17.6 mmol) in dry CH_2Cl_2 (100 mL) at rt. and stirred for 15 min. Then, 1.0 M solution of CuCN.2LiCl in dry THF (20 mL, 2 mmol) was added followed by the bromide **144** (5.7 g, 18.9 mmol). Resulting dark brown solution was stirred at rt. for 48 h and poured into half sat. NH_4Cl containing 2%

concentrated NH_3 (50 mL). After stirring for 20 min, the resulting solid was filtered off and the filtrate was partitioned with CH_2Cl_2 (3x50 mL). The organic layer was dried (Na_2SO_4) , concentrated and purified by chromatography (EtOAc/hexane, 3.5:6.5) to afford **236** (4.00 g, 49%) as a pale yellow oil: ^{1}H NMR: δ = 7.91 (s. 1H), 6.98 (d, *J* = 8.7 Hz, 2H), 6.75 (d, *J* = 8.7 Hz, 2H), 4.13 (s, 2H), 2.49 (s, 3H), 0.95 (s, 9H), 0.15 (s, 6H); ^{13}C NMR: δ = 154.6, 139.7, 132.6, 129.8, 129.1, 120.3, 90.6, 37.5, 30.0, 25.7, 18.3, -4.3; IR (neat,cm^{-1}): = 2961, 2928, 2858, 1690, 1507, 1390, 1251, 1145, 1092, 909, 834, 778, 723; HR-DARTMS (*m/z*): Calcd. for $C_{18}H_{29}IN_3O_3SSi$ $[M+H]^+$ 522.0738, found 522.0759.

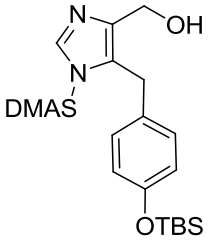
5-(4-*tert*-Butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-4-formyl-1*H*-imidazole (237):



EtMgBr (3.0 M solution in ether, 2.68 mL, 8.0 mmol) was added to a solution of **236** (4.00 g, 7.7 mmol) in dry CH_2Cl_2 (40 mL) at rt. and stirred for 20 min. Then, **165** (1.04 mL, 8.4 mmol) was added to above reaction and was stirred for overnight. After the addition of water (10 mL), organic layer was extracted with CH_2Cl_2 (3x25 mL). The organic layer was dried (Na_2SO_4) , concentrated and purified by chromatography (EtOAc/hexane, 2:8) to isolate **237** (2.39 g, 74%) as a pale yellow oil: ^{1}H NMR: δ = 10.04 (s, 1H), 7.95 (s. 1H), 6.97 (d, *J* = 8.5 Hz, 2H), 6.72 (d, *J* = 8.5 Hz, 2H), 4.52 (s, 2H), 2.56 (s, 3H), 0.94 (s, 9H), 0.14 (s, 6H); ^{13}C NMR: δ = 187.5, 154.7, 139.0, 138.6, 137.0, 129.5, 129.2, 120.4, 37.7, 28.4, 25.7, 18.3, -4.3; IR (neat, cm^{-1}): = 2953, 2929, 2857, 1691, 1507, 1391, 1251,

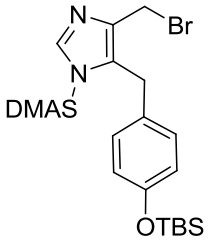
1150, 1124, 1005, 910, 836; HR-DARTMS (*m/z*): Calcd. for $C_{19}H_{30}N_3O_4SSi$ $[M+H]^+$ 424.1721, found 424.1741.

5-(4-*tert*-Butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-4-hydroxymethyl-1*H*-imidazole (238):



$NaBH_4$ (105 mg, 2.8 mmol) was added to a solution of **237** (2.36 g, 5.6 mmol) in 30:1 mixture of THF-H_2O (31 mL). After stirring at rt. for 10 min, water was added to quench the reaction and the organic layer was extracted with EtOAc (2x15 mL). The combined organic layers were dried over Na_2SO_4 and concentrated to provide **238** (2.35 g, quant) as a pale yellow oil: ^{1}H NMR: δ = 7.93 (s. 1H), 6.98 (d, *J* = 8.5 Hz, 2H), 6.75 (d, *J* = 8.5 Hz, 2H), 4.53 (s, 2H), 4.16 (s, 2H), 2.55 (s, 3H), 0.96 (s, 9H), 0.15 (s, 6H); ^{13}C NMR: δ = 154.5, 141.2, 138.2, 130.6, 129.1, 126.7, 120.3, 57.1, 37.5, 28.1, 25.7, 18.3, -4.3; IR (neat, cm^{-1}): = 3200 (br), 2928, 2856, 1690, 1507, 1391, 1251, 1177, 1142, 1000,, 834, 779, 725 ; HR-DARTMS (*m/z*): Calcd. for $C_{19}H_{32}N_3O_4SSi$ $[M+H]^+$ 426.1877, found 426.1867.

4-Bromomethyl-5-(4-*tert*-butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-1*H*-imidazole (239a):



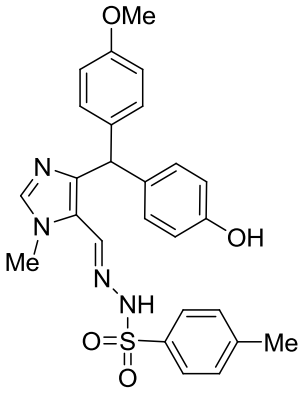
N-Bromosuccinimide (150 mg, 0.8 mmol) was added portionwise over a 5 min to a stirred solution of **238** (300 mg, 0.7 mmol) and PPh_3 (222 mg, 0.9 mmol) in dichloromethane (15 mL) at 0 ºC. The reaction was stirred for 2 h at the same

temperature then quenched with satd. aq. $NaHCO_3$ (10 mL). The aqueous layer was extracted with dichloromethane (2x10 mL), combined organic layers were dried over Na_2SO_4 and concentrated to provide the crude orange oil, which was immediately purified over silica gel (EtOAc) to isolate **239** (277 mg, 80%) as a light orange oil, which was used in the next step without full characterization: ^{1}H NMR: δ = 7.93 (s. 1H), 6.99 (d, *J* = 8.7 Hz, 2H), 6.76 (d, *J* = 8.7 Hz, 2H), 4.36 (s, 2H), 4.17 (s, 2H), 2.57 (s, 3H), 0.96 (s, 9H), 0.16 (s, 6H); ^{13}C NMR: δ = 154.7, 138.3, 138.3, 129.9, 129.2, 128.1, 120.4, 37.6, 28.5, 25.7, 25.2, 18.3, -4.3.

Unknown compound 239b:

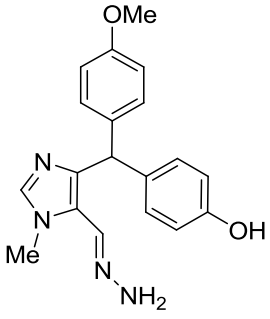
MsCl (0.04 mL, 0.5 mmol) was added to a stirred solution of **238** (115 mg, 0.3 mmol) and Et_3N (0.09 mL, 0.7 mmol) in dichloromethane (5 mL) at 0 ºC. After stirring for 30 min at the same temperature, water (2 mL) was added to the reaction and the reaction mixture was washed with 10% cold HCl (3 mL) and satd. aq.$NaHCO_3$ solutions. The organic layer was dried (Na_2SO_4) and concentrated to provide a crude mixture, which was purified over silica gel (EtOAc) to isolate **239b** (126 mg) as a pale yellow oil: ^{1}H NMR: δ = 7.91 (s. 1H), 6.96 (d, *J* = 8.4 Hz, 2H), 6.73 (d, *J* = 8.4 Hz, 2H), 4.45 (s, 2H), 4.15 (s, 2H), 3.65 (s, 4H), 3.11 (s, 6H), 2.55 (s, 3H), 0.93 (s, 9H), 0.13 (s, 6H); ^{13}C NMR: δ = 154.7, 138.3, 138.1, 130.0, 129.2, 128.4, 120.4, 52.6, 38.5, 37.6, 31.7, 28.3, 25.7, 18.3, -4.3.

5-[4-Hydroxyphenyl-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazol-4-yl-methylene)-4-methyphenylsulfonylhydrazone (241):



Tosylhydrazide (163 mg, 0.9 mmol) was added to a solution of **234** (260 mg, 0.8 mmol) in EtOH at rt., and the reaction was heated to reflux for 3 h. After cooling to rt., solvent was removed under reduce pressure and the crude product was purified over silica (1:1, EtOAc: hexanes→ 1:9, MeOH: EtOAc) to afford **241** (290 mg, 73%) as white solid: ^{1}H NMR (Acetone-d6): δ = 8.09 (s, 1H), 8.00 (s, 1H), 7.78 (d, *J* = 8.3 Hz, 2H), 7.52 (s, 1H), 7.34 (d, *J* = 8.3 Hz, 2H), 7.19 (d, *J* = 8.7 Hz, 2H), 7.09 (d, *J* = 8.7 Hz, 2H), 6.75 (d, *J* = 8.7 Hz, 2H), 6.67 (d, *J* = 8.7 Hz, 2H), 5.49 (s, 1H), 3.71 (s, 3H), 2.37 (s, 3H); ^{13}C NMR: δ = 158.1, 155.7, 148.5, 143.8, 140.7, 138.6, 136.5, 136.3, 134.9, 130.0, 129.6, 127.9, 122.2, 114.7, 113.2, 78.4, 54.6, 46.8, 33.6, 20.6; IR (neat, cm^{-1}): = 3502, 3002, 2915, 1637, 1610, 1500, 1362, 1250, 1177, 772; HR-ESIMS (*m/z*): Calcd. for $C_{26}H_{27}N_4O_4S$ $[M+H]^+$ 491.1748, found 491.1774; Calcd. for $C_{25}H_{26}N_4NaO_4S$ $[M+Na]^+$ 513.1567, found 513.1600.

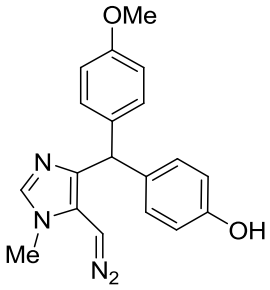
5-(4-Hydroxyphenyl-4-methoxyphenyl)methyl-1-methyl-1*H*-imidazol-4-yl-methylenehydrazone (242):



Hydrazine monohydrate (0.32 mL, 6.5 mmol) was added to a solution of **234** (209 mg, 0.6 mmol) in EtOH at rt., and the reaction was heated to reflux for 2 h. After cooling to rt., solvent was removed under reduce pressure and the crude

product was purified over silica (1:1, EtOAc: hexanes) to afford **242** (213 mg, 98%) as white solid: m.p = 102-105 ºC ; ^{1}H NMR: δ = 7.51 (s, 1H), 7.31 (s, 1H), 7.10 (d, *J* = 8.5 Hz, 2H), 6.85 (d, *J* = 8.5 Hz, 2H), 6.75 (d, *J* = 8.5 Hz, 2H), 6.46 (d, *J* = 8.5 Hz, 2H), 5.44 (s, 1H), 3.67 (s, 3H), 2.65 (s, 3H); ^{13}C NMR: δ = 158.0, 155.6, 145.1, 139.3, 135.3, 134.0, 133.7, 130.1, 129.8, 123.7, 115.8, 113.8, 55.3, 47.5, 34.8;

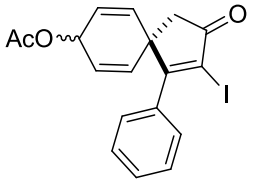
4-(4-Hydroxyphenyl-4-methoxyphenyl)methyl-5-diazomethyl-1-methyl-1*H*-imidazole (243):



MnO_2 (218 mg, 2.5 mmol) was added to a solution of **242** (168 mg, 0.5 mmol) in dichloromethane at rt. and the reaction was stirred for 1h. Solvent was removed under reduced pressure to afford 243 (146 mg, 87%) as a pink solid, which was used in the next step without further purification: ^{1}H NMR: δ = 8.48 (s, 1H), 7.46 (s, 1H), 7.16 (d, *J* = 8.3 Hz, 2H), 6.88 (d, *J* = 8.3 Hz, 2H), 6.76 (d, *J* = 8.5 Hz, 2H), 6.43 (d, *J* = 8.5 Hz, 2H), 5.56 (s, 1H), 3.85 (s, 3H), 3.70 (s, 3H); ^{13}C NMR (DMSO-d6): δ = 157.9, 155.9, 150.2, 148.3, 142.0, 136.7, 134.8, 130.2, 130.1, 123.0, 115.3, 113.9, 55.5, 46.4, 34.8.

254 and 255 were prepared following Larock's protocol.[135]

2-iodo-3-oxo-1-phenyl-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (256):



Alkynone **255** (1.35 g, 6.1 mmol) was dissolved in acetic acid (5 mL) followed by the careful addition of *N*-

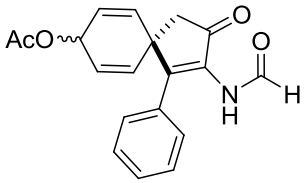
iodosuccinimide (NIS) (1.65 g, 7.3 mmol) to maintain ambient temperature. After stirring the resulting mixture for 30 min, ethyl acetate was added to it followed by aqueous saturated $NaHCO_3$ to neutralize the reaction mixture. The EtOAc layer was separated and the aqueous layer was extracted with EtOAc two more times. Combined organic layers were dried (Na_2SO_4) and concentrated to provide a crude product which was separated chromatographically with 10% ethyl acetate in hexanes as the eluent to provide a 2:1 mixture of **256** (1.46 g, 60%) as a pale brown oil: ^1H NMR (integrated together, underlined are the corresponding peaks from the minor isomer): δ = 7.42-7.35 (m, 3H), 7.24-7.22 (m, 2H), 5.95-5.88 (m, 2H), 5.87-5.81 (m, 2H), 5.55, 5.25 (m, 1H), 2.82, 2.76 (s, 2H), 2.04, 1.94 (s, 3H); ^{13}C NMR (minor isomer in parentheses): δ = 201.1(201.0), 180.1(178.9), 170.5(170.5), 135.2(135.0), 132.5(133.8), 130.0(129.9), 128.3(128.0), 127.3(127.9), 126.3(125.4), 105.4(104.8), 63.6(63.4), 51.8(51.4), 46.7(47.6), 21.1(21.0), ; IR (neat, cm^{-1}): = 3033, 2924, 1721, 1369, 1236, 1016, 928, 739, 698; HR-ESIMS (*m/z*): Calcd. for $C_{18}H_{15}INaO_3$ $[M+Na]^+$ 428.9958, found: 428.9926.

General procedure for cross-coupling reaction of 256:[137]

A 10 mL resealable, thick-walled tube was charged with CuX (X = I, Br, CN: 5%-100%), iodo-ketone **256** (100 mg, 0.3 mmol) and base (Cs_2CO_3, K_2CO_3 or K_3PO_4: 1.5 -2 equiv). The tube was evacuated and back filled with nitrogen. Then, the ligand (10%-200%), formamide (0.01 mL, 0.3 mmol) and THF (3 mL) were added under nitrogen. The pressure tube was sealed with a Teflon cap, immersed in a preheated oil bath at 70 °C and the reaction mixture was stirred following reaction progress by TLC. After the reaction was allowed to reach to rt., EtOAc was added. The reaction

mixture was filtered through a pad of silica eluting with EtOAc. The filtrate was concentrated and the residue was purified by plash chromatography on silica gel provide **259** as brown oil.

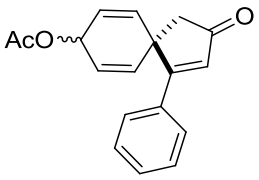
2-formylamino-3-oxo-1-phenyl-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (259):



A 10 mL resealable, thick-walled tube was charged with CuI (20 mg, 0.1 mmol), iodo-ketone **256** (841 mg, 2.1 mmol) and Cs_2CO_3 (1.01 g, 3.1 mmol). The tube was evacuated and back filled with nitrogen and *N,N'*-dimethylethylenediamine (0.02 mL, 0.2 mmol), formamide (0.10 mL, 2.5 mmol) and THF (5 mL) were added under nitrogen. The pressure tube was sealed with a Teflon cap, immersed in a preheated oil bath at 70 °C and the reaction mixture was stirred 48 h following reaction progress by TLC. After the resulting pale blue suspension was allowed to reach to rt., EtOAc was added. The reaction mixture was filtered through a pad of silica eluting with EtOAc. The filtrate was concentrated and the residue was purified by plash chromatography on silica gel with 10% ethyl acetate in hexanes to provide **259** as brown oil (334 mg, 50%): ^{1}H NMR (overlapped signals from both isomers are integrated together, underlined peaks correspond to the minor isomer): δ = 8.88, 8.83 (d, *J* = 11.5 Hz, 1 H), 7.39 (s, 3H), 7.34 (m, 1H), 7.18 (m, 2H), 5.95-5.90 (m, 2H), 5.86-5.80 (m, 2H), 5.59, 5.32 (m, 1H), 2.73, 2.67 (s, 2H), 2.05, 1.95 (s, 3H); ^{13}C NMR (other isomer in parentheses) δ = 199.9 (199.8), 170.5, 161.8 (162.0), 154.1 (153.5), 133.3 (134.5),133.9 (133.6), 132.3 (132.5), 129.6 (129.7), 129.1 (128.8), 127.6 (128.1), 126.1 (125.3), 63.6 (63.4), 47.4 (48.2), 46.2 (45.9), 21.2 (21.0); IR

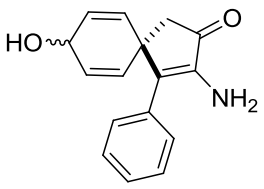
(neat, cm^{-1}): = 3195, 3023, 2917, 2850, 1667, 1494, 1441, 1221, 1097, 748, 698; HR-DARTMS (*m/z*): Calcd. for $C_{19}H_{18}NO_4$ $[M+H]^+$ 324.1120, found 324.1230.

3-Oxo-1-phenyl-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (260):



From above reaction, **260** (87 mg, 15%) as a dark brown oil: ^{1}H NMR (peaks are integrated together, underlined are the peaks from the minor isomer): δ = 7.75-7.72 (m, 1 H), 7.54-7.51 (m, 1H), 7.43-7.32 (m, 3H), 6.58, 7.51 (s, 1H), 6.01-5.93. (m, 4 H), 5.78-5.77 (m, 1 H), 2.67, 2.60 (s, 2H), 2.13, 2.11 (s, 3H); ^{13}C NMR (other isomer in parentheses): δ =205.8 (205.7), 176.9 (175.8), 170.8 (170.6), 135.4 (135.2), 133.9 (133.4), 131.0 (130.8), 129.9 (129.3), 128.7 (128.5), 128.5 (127.8), 124.4 (124.3), 64.2 (64.0), 51.3 (50.7), 48.5 (47.9), 21.3 (21.3); IR (neat, cm^{-1})= 3022, 1726, 1691, 1590, 1568, 1368, 1229, 1012, 966, 863, 754, 647; HR-DARTMS (*m/z*): Calcd. for $C_{18}H_{17}O_3$ $[M+H]^+$ 281.1172, found 281.1168.

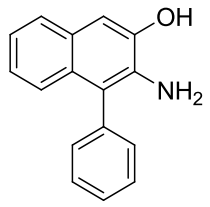
3-Amino-8-hydroxy-4-phenyl-spiro[4.5]deca-3,6,9-trien-2-one (261):



Methylamine hydrochloride (200 mg, 3.0 mmol) and K_2CO_3 (410 mg, 3.0 mmol) were added to a solution of **259** (192 mg, 0.6 mmol) in ethanol (10 mL) and the mixture was heated at reflux temperature overnight. Water was added to the reaction mixture after evaporating the solvent and the aqueous layer was extracted with EtOAc (2x10 mL). combined organic layers were dried (Na_2SO_4) and concentrated to provide a crude product, which was purified over silica gel using 2:3 mixture of EtOAc: hexanes to isolate **261** (30 mg, 20%) as a light brown solid: ^{1}H

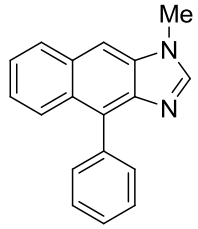
NMR: δ = 7.40-7.38 (m, 2H), 7.35-7.32 (m, 2H), 7.28-7.27 (m, 1H), 5.95 (dd, *J* = 3.2, 10.1 Hz, 2H), 5.72 (dd, *J* = 1.8, 10.1 Hz, 2H), 4.37 (s, 1H), 8.84 (s, 2H), 2.58(s, 2H); ^{13}C NMR δ = 201.6, 140.7, 140.1, 135.0, 134.4, 128.7, 128.6,128.2, 127.5, 61.9, 48.0, 44.9;

3-Amino-4-phenyl-2-naphthol (262):



Aminol **261** (23 mg, 0.1 mmol) was added to a solution of cynamide (90 mg, 2.1 mmol) in water. This mixture was acidified to pH 4.5 by careful addition of 10% HCl and the resulting mixture was heated at 90 °C for 3 h. After cooling, the resulting mixture was made basic to pH 10 with 20% NaOH and the aqueous layer was extracted with EtOAc (2x10 mL). Combined organic layers were dried ((Na_2SO_4), concentrated to provide **262** (60%) as a brown solid: m.p = 162-164 ºC; ^{1}H NMR (Acetone-d_6): δ = 7.58-7.53 (m, 3H), 7.46-7.43 (m, 1H), 7.34-7.32 (m, 2H), 7.17 (s, 1H), 7.10-7.07 (m, 2H), 7.06-7.03 (m, 1H),4.25 (brd, 2H), 2.85 (brd, 2H); ^{13}C NMR (Methanol-d_3): δ = 145.8, 137.5, 133.7, 130.6, 128.9, 128.8, 128.7, 127.2, 125.7, 123.5, 122.7, 122.1, 120.1, 107.6.

1-Methyl-4-phenyl-1*H*-naphtho[2,3-*d*]imidazole (263):

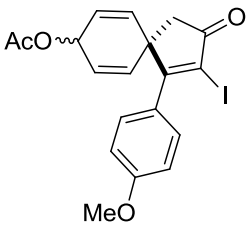


Methylamine hydrochloride (158 mg, 2.4 mmol) and Et_3N (0.13 mL, 0.9 mmol) were added to a solution of **259** (153 mg, 0.5 mmol) in ethanol (5 mL) and the mixture was heated at reflux temperature overnight. Water was added to the reaction mixture after evaporating the solvent and the aqueous layer was extracted with EtOAc (2x 10

mL). The combined organic layers were dried (Na_2SO_4) and concentrated to provide a crude product, which was purified over silica gel using 3:2 mixture of EtOAc: hexanes to produce **263** (43 mg, 35%) as a green solid: m. p = 185-188 ºC; ^{1}H NMR: δ = 8.70 (d, *J* = 10.1 Hz, 1H), 8.27 (d, *J* = 9.2 Hz, 1H), 8.01 (s, 1H), 7.97 (d, *J* = 7.8 Hz 2H), 7.54 (t, *J* = 7.8 Hz, 2H), 7.47 (t, *J* = 10.1 Hz, 1H), 7.34 (t, *J* = 7.8 Hz, 1H), 7.09 (t, *J* = 9.2 Hz, 1H), 7.01 (t, *J* = 10.1 Hz, 1H), 4.21 9s, 3H); ^{13}C NMR: δ = 155.7, 147.1, 135.7, 135.4, 135.2, 133.9, 129.8, 128.8, 128.0, 126.8, 126.5, 122.7, 121.9, 121.4, 117.1, 33.8; IR (neat, cm^{-1}): = 3039, 2918, 2844, 1607, 1592, 1572, 1538, 1495, 1456, 1372, 1261, 1171, 1044, 896, 740; HR-ESIMS (*m/z*): Calcd. for $C_{18}H_{14}N_2$ $[M+H]^+$: 259.1230, found 259.1227; Calcd. for $C_{36}H_{29}N_4$ $[2M+H]^+$ 517.2387, found 517.2390.

270 and **252** were prepared following the literature peocedures.[135, 157]

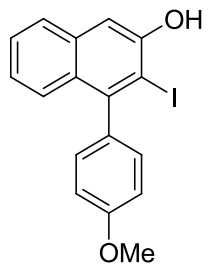
2-iodo-1-(4-methoxy-phenyl)-3-oxo-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (251):



Alkynone **252** (530 mg, 2.1 mmol) was dissolved in acetic acid (5 mL) followed by careful addition of NIS (577 mg, 5.5 mmol) at rt. After stirring the resulting mixture for 15 min, ethyl acetate was added to it followed by satd. aq. $NaHCO_3$ to neutralize the reaction mixture. The organic layer was separated and the aqueous layer was extracted with ethyl acetate two more times. Combined organic layers were dried (Na_2SO_4) and concentrated to provide a crude product which was separated chromatographically with 20% ethyl acetate in hexanes as the eluent to provide a 5:4 mixture of **251** (747 mg, 81%) as a reddish brown oil: ^{1}H NMR (corresponding peaks

from the minor isomer are underlined): δ = 7.57, 7.32 (d, *J* = 8.7 Hz, 2H)), 6.88, 6.87 (d, *J* = 8.7 Hz, 2H), 5.95-5.92, 5.94-5.91 (m, 2H), 5.86-5.82, 5.85-5.80 (m, 2H), 5.59, 5.39 (m, 1H), 3.82, 3.82 (s, 3H), 2.78, 2.71 (s, 2H), 2.05, 2.01 (s, 3H); ^{13}C NMR (peaks from minor isomer are in parentheses): δ = 201.1 (200.9), 179.1 (177.6), 170.5 (170.5), 161.1 (160.8), 133.3 (134.3), 129.2 (130.2), 127.3 (127.0), 125.8 (125.0), 113.7 (113.4), 104.2 (103.0), 63.8 (63.5), 55.3, 51.7 (51.3), 47.0 (47.9), 21.2 (21.1); IR (neat, cm^{-1})= 2930, 2837, 1728, 1704, 1603, 1504, 1330, 1226, 1175, 1020, 900, 835, 760; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{18}IO_4$ $[M+H]^+$ 437.0244, found 437.0243.

3-Iodo-4-(4-methoxy-phenyl)-naphthalen-2-ol (271):

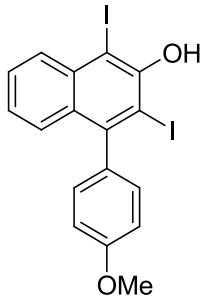


Alkynone **252** (530 mg, 2.1 mmol) was dissolved in acetic acid (5 mL) and NIS (577 mg, 5.5 mmol) was added at once to the reaction at rt. (reaction mixture was warmed up due to reactivity of NIS). After stirring the resulting mixture for 15 min, usual workup provided the crude material, which was purified over silica gel (1:9 EtOAc: hexanes) to isolate **271** (3.50 g, 31%) and **272** (4.46 g, 17%)

271, reddish brown solid: m.p = 139-141 ºC; ^{1}H NMR: δ = 7.72 (d, *J* = 7.8 Hz, 1H), 7.43 (t, *J* = 7.8 Hz, 2H), 7.34 (d, *J* = 8.7 Hz, 1H), 7.19 (d, *J* = 7.8 Hz, 1H), 7.18 (d, *J* = 8.7 Hz, 2H), 7.06 (d, *J* = 8.7 Hz, 2H), 5.66 (s, 1H), 3.92 (s,3H); ^{13}C NMR (DEPT 135º): δ = 159.4 (C), 151.1 (C), 146.2 ©, 138.9 (C), 134.7 (C), 131.0 (CH), 129.1 (C), 127.5 (CH), 127.2 (CH), 126.7 (CH), 124.4 (CH), 114.0 (CH), 108.9 (CH), 97.7 (C), 55.4 (CH_3); IR (neat, cm^{-1}): = 3478 (br), 2954, 2833, 1604, 1583, 1510, 1328, 1242, 1213, 1170, 1026, 872, 843, 795, 773, 753; HR-ESIMS (*m/z*): Calcd. for $C_{17}H_{14}IO_2$

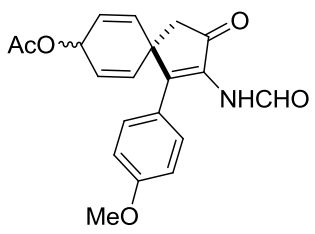
$[M+H]^+$ 377.0033, found 377.0046; Calcd. for $C_{17}H_{13}INaO_2$ $[M+Na]^+$ 398.9853, found 398.9867.

1,3-Diiodo-4-(4-methoxy-phenyl)-naphthalen-2-ol (272):



272 (4.46 g, 17%), reddish brown solid: m.p = 104-106 ºC; ^{1}H NMR: δ = 8.06 (d, *J* = 8.7 Hz, 1H), 7.53 (tt, *J* = 1.4, 7.8 Hz, 1H), 7.36 (d, *J* = 8.2 Hz, 1H), 7.22 (tt, *J* = 1.4, 7.8 Hz, 1H), 7.14 (d, *J* = 8.7 Hz, 2H), 7.08 (d, *J* = 8.7 Hz, 2H), 6.36 (s, 1H), 3.92 (s,3H); ^{13}C NMR (DEPT 135º): δ = 159.5 (C), 151.1 (C), 147.5 (C), 135.5 (C), 134.9 (C), 131.1 (CH), 131.0 (CH), 129.6 (C), 128.9 (CH), 128.5 (CH), 125.1 (CH), 114.1 (CH), 93.5 (C), 83.2 (C), 55.6 (CH_3); IR (neat, cm^{-1}): = 3391 (br), 3062, 2953, 2840, 1605, 1511, 1486, 1371, 1234, 1171, 1027, 749; HR-ESIMS (*m/z*): Calcd. for $C_{17}H_{13}I_2O_2$ $[M+H]^+$ 502.9005, found 341.1512; Calcd. for $C_{17}H_{12}I_2NaO_2$ $[M+Na]^+$ 524.8824, found 363.1328. observed ion consistent with MW of 340 Da. This structure was confirmed by X-ray crystallography.

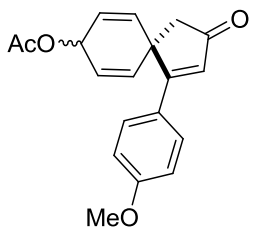
2-formylamino-1-(4-methoxy-phenyl)-3-oxo-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (250):



Following the optimized procedure as for **259**; CuI (2 mg, 0.01 mmol), iodo-ketone **251** (102 mg, 0.23 mmol) and Cs_2CO_3 (152 mg, 0.50 mmol), *N, N'*-dimethylethylenediamine (0.01 mL, 0.02 mmol), formamide (0.02 mL, 0.50 mmol) and THF (2 mL) were heated at reflux temperature for 12 h. The crude product was purified with 1:1 mixture of EtOAc: hexanes to isolate 1:1 mixture of **250** (58 mg, 70%) as light brown oil; ^{1}H NMR (Some peaks are

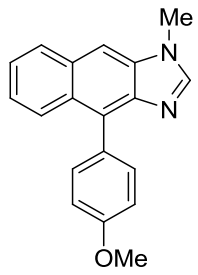
overlapping and the underlined signals correspond to the other product and the relevant signals are integrated together with the major product): δ = 8.87, 8.83 (two overlapped doublets, 1 H), 7.38, 7.38 (d, *J* = 8.5 Hz, 1H), 7.19, 7.19 (d, *J* = 8.5 Hz, 2H), 6.89, 6.89 (d, *J* = 8.5 Hz, 2H), 5.98-5.90 (m, 2H), 5.90-5.78 (m, 2H), 5.64, 5.44 (s, 1H), 3.81, 3.81 (s, 3H), 2.69, 2.62 (s, 2H), 2.06, 2.01(s, 3H); ^{13}C NMR (other isomer in parentheses): δ = 199.8 (199.7), 170.6 (170.5), 162.0 (162.3), 160.6 (160.8), 154.4 (153.9), 133.9 (134.8), 133.3 (132.7), 129.1 (129.8), 125.8 (125.7), 124.4 (124.5), 114.6 (114.3), 63.8 (63.6), 55.4 (55.4), 47.6 (48.5), 46.1 (45.7), 21.2 (21.1); IR (neat, cm^{-1}): = 3317, 3011, 2965, 2828, 1699, 1662, 1604, 1507, 1384, 1283, 1245, 1175, 1030, 804, 734; HR-ESIMS (*m/z*): Calcd. for $C_{20}H_{20}NO_5$ $[M+H]^+$ 354.1341, found 354.1311; Calcd. for $C_{20}H_{19}NNaO_5$ $[M+Na]^+$ 376.1161, found 376.1129.

1-(4-Methoxy-phenyl)-3-oxo-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (276):



From above reaction, **276** (15 mg, 20%) was isolated as a 1;1 mixture of dark brown soilid: m.p = 92-95 ºC; ^{1}H NMR: δ = 7.72, 7.42 (d, *J* = 8.8 Hz, 2H), 6.75 (d, *J* = 8.3 Hz, 4 H), 6.37, 6.34 (s, 1H), 5.89-5.81 (m, 4H), 5.74, 5.64 (m, 1H), 3.70, 3.68 (s, 3H), 2.49,2.42 (s, 2H), 2.04,2.00(s, 3H); ^{13}C NMR(other isomer in parentheses): δ = 205.6 (205.5), 176.0 (175.2), 170.9 (170.6), 161.9 (161.8), 136.1 (135.5), 130.5 (129.8), 127.7 (127.1), 126.2 (125.9), 124.1 (123.9), 114.2 (114.0), 64.3 (64.1), 55.5, 51.3 (50.8), 48.2 (47.7), 21.3; IR (neat, cm-1): = 2841, 1728, 1680, 1600, 1585, 1508, 1238, 1176, 1022, 869, 806, 757; HR-DARTMS (*m/z*): Calcd. for $C_{19}H_{19}O_4$ $[M+H]^+$ 311.1278, found 311.1285.

1-Methyl-4-(4-methoxyphenyl)-1*H*-naphtho[2,3-*d*]imidazole (278):



A solution of methylamine in methanol (8.03 M, 0.05 mL,0.4 mmol) was added to amide **250** (120 mg, 0.4 mmol) in pre-absorbed silica (1g). After stirring at rt. for overnight, $Zn(BH_4)$ in THF (4.0 M, 0.10 mL, 0.4 mmol) was added to above reaction, and stirred for 2 h. After removing the solvent, crude product was purified over silica gel (1:1 EtOAc: hexanes) to isolate **278** (30 mg, 30%) as a light brown solid: m. p = 158-160 ºC ; ^{1}H NMR: δ = 8.07 (d, *J* = 8.7 Hz, 1H), 8.00 (d, *J* = 8.7 Hz, 2H), 7.79 (s, 1H), 7.57 (d, *J* = 8.7, 2H), 7.45 (tt, *J* = 1.4, 7.3 Hz, 1H), 7.35 (tt, *J* = 1.4, 7.3 Hz, 1H), 7.12 (d, *J* = 8.7 Hz, 2H), 3.92 (s, 3H), 3.91 (s, 1H); ^{13}C NMR δ = 159.1, 147.5, 142.5, 134.9, 132.3, 131.1, 129.6, 128.9, 128.3, 127.8, 126.5 124.4, 123.5, 114.0, 104.6, 55.4, 31.2; IR (neat, cm^{-1}): = 3054, 2920, 2844, 1667, 1513, 1241, 1174, 1024, 828, 746; HR-ESIMS (*m/z*): Calcd. for $C_{19}H_{17}N_2O$ $[M+H]^+$ 289.1335, found 289.1335.

APPENDIX 1

^{1}H and ^{13}C NMR Spectra of

1-Trimethylsilylethoxymethyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (**91d**)

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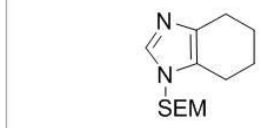
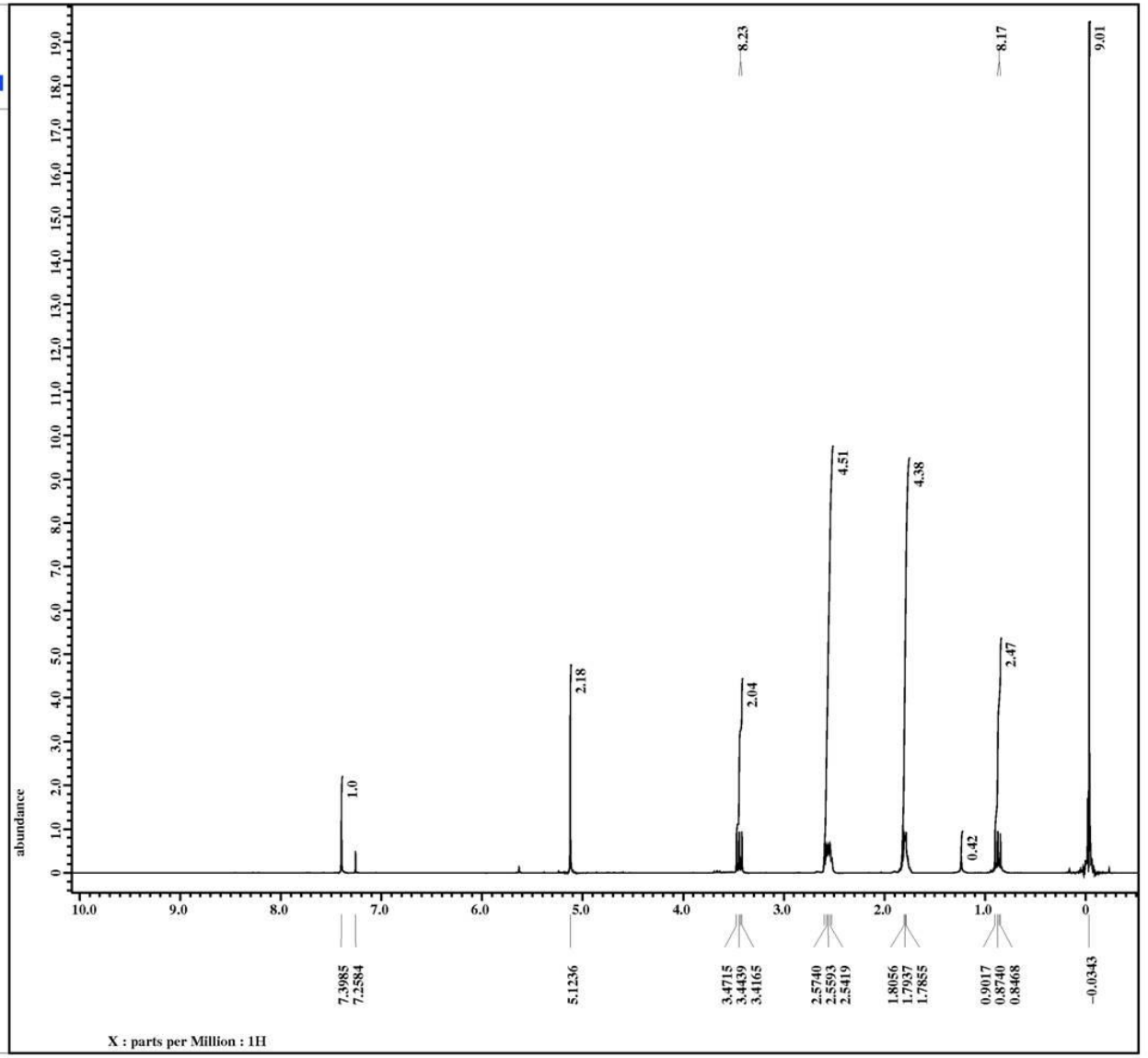
Filename          = SEMTHB-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#441228
Solvent           = CHLOROFORM-D
Creation_time     = 23-MAR-2006 13:15:28
Revision_time     = 17-MAR-2010 13:34:45
Current_time      = 17-MAR-2010 13:35:41

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

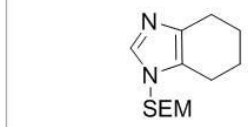
Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

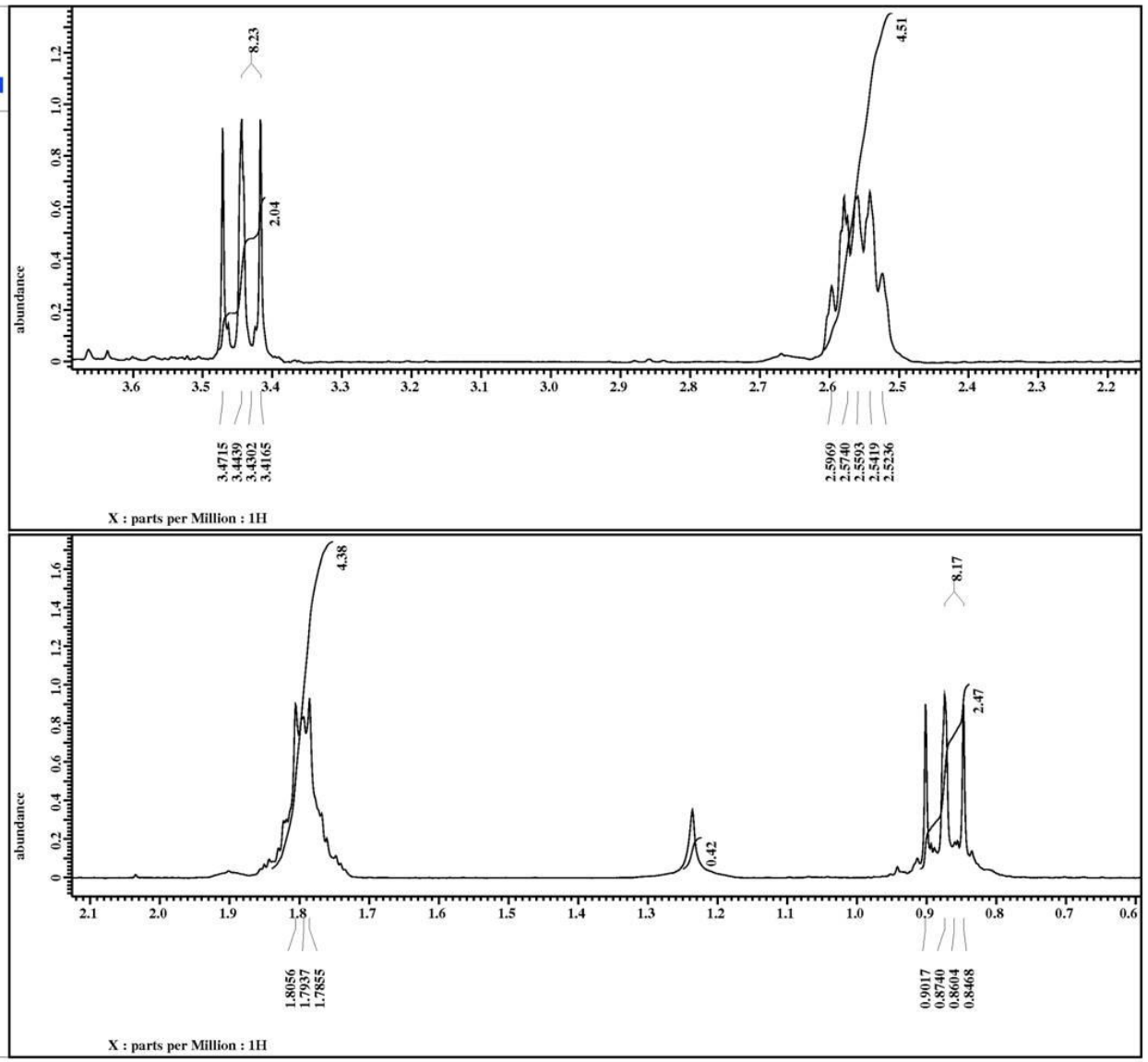
X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 38
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22[dC]
```





Filename	= SEMTHB-4.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#441228
Solvent	= CHLOROFORM-D
Creation_time	= 23-MAR-2006 13:15:28
Revision_time	= 17-MAR-2010 13:34:45
Current_time	= 17-MAR-2010 13:36:13
Comment	= single_pulse
Data_format	= 1D REAL
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 38
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22[dC]





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Filename          = II_p_SEMTHB-7.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#836421
Solvent           = CHLOROFORM-D
Creation_time     =  3-OCT-2006 09:49:27
Revision_time     = 17-MAR-2010 13:36:48
Current_time      = 17-MAR-2010 13:37:08

Comment           = single pulse decouple
Data_format       = 1D REAL
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7449
Total_scans       = 7449

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.9[dC]
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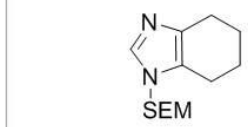

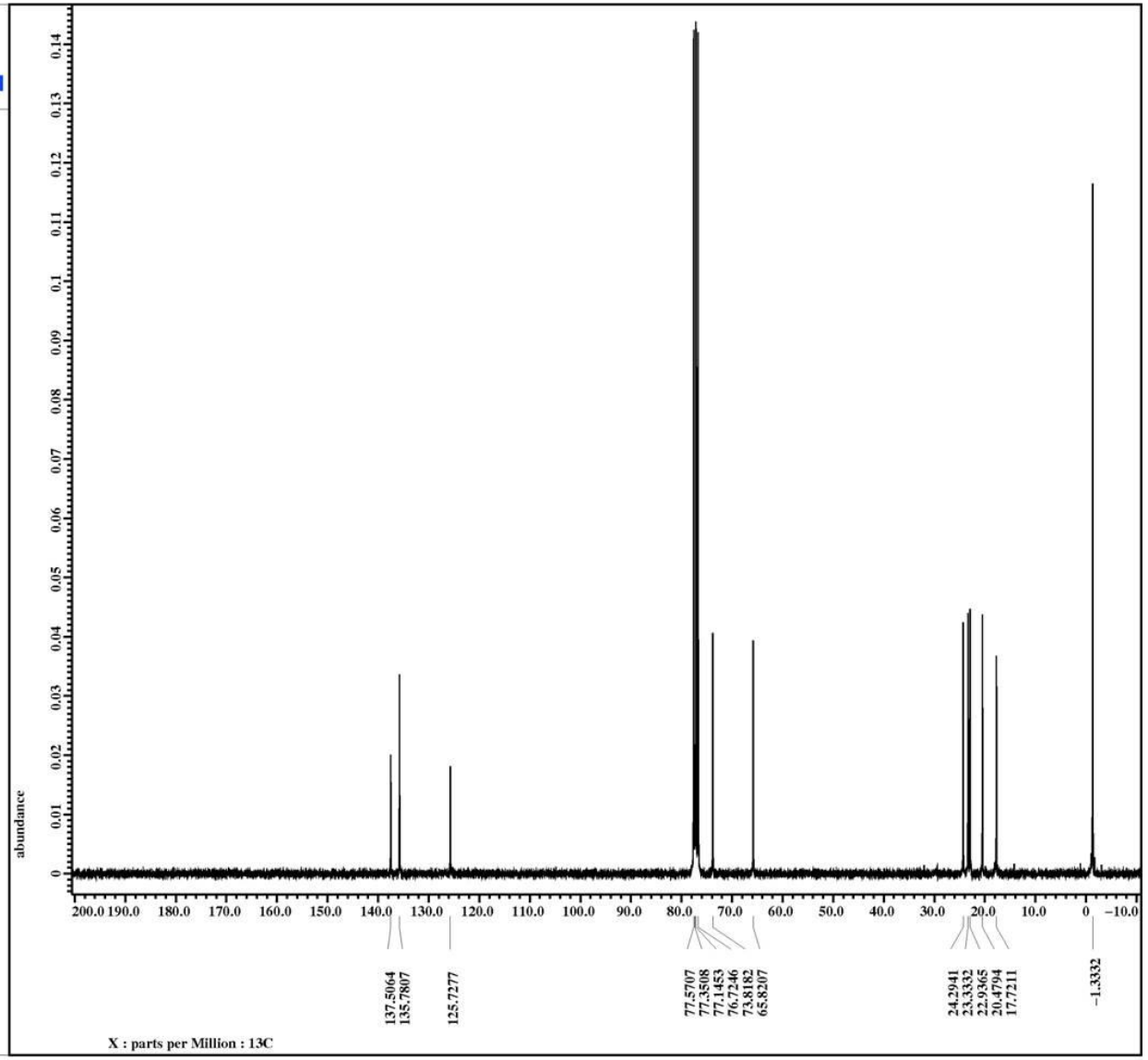

APPENDIX 2

^{1}H and ^{13}C NMR Spectra of

3-Trimethylsilylethoxymethyl-1,3-diazaspiro[4.4]non-2-ene-4-one (**92d**)

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Filename          = II_p_066_product-5.jd
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#9263
Solvent           = CHLOROFORM-D
Creation_time     = 12-AUG-2006 00:55:05
Revision_time     = 17-MAR-2010 13:55:14
Current_time      = 17-MAR-2010 13:55:39

Comment           = single_pulse
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Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 42
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
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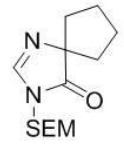

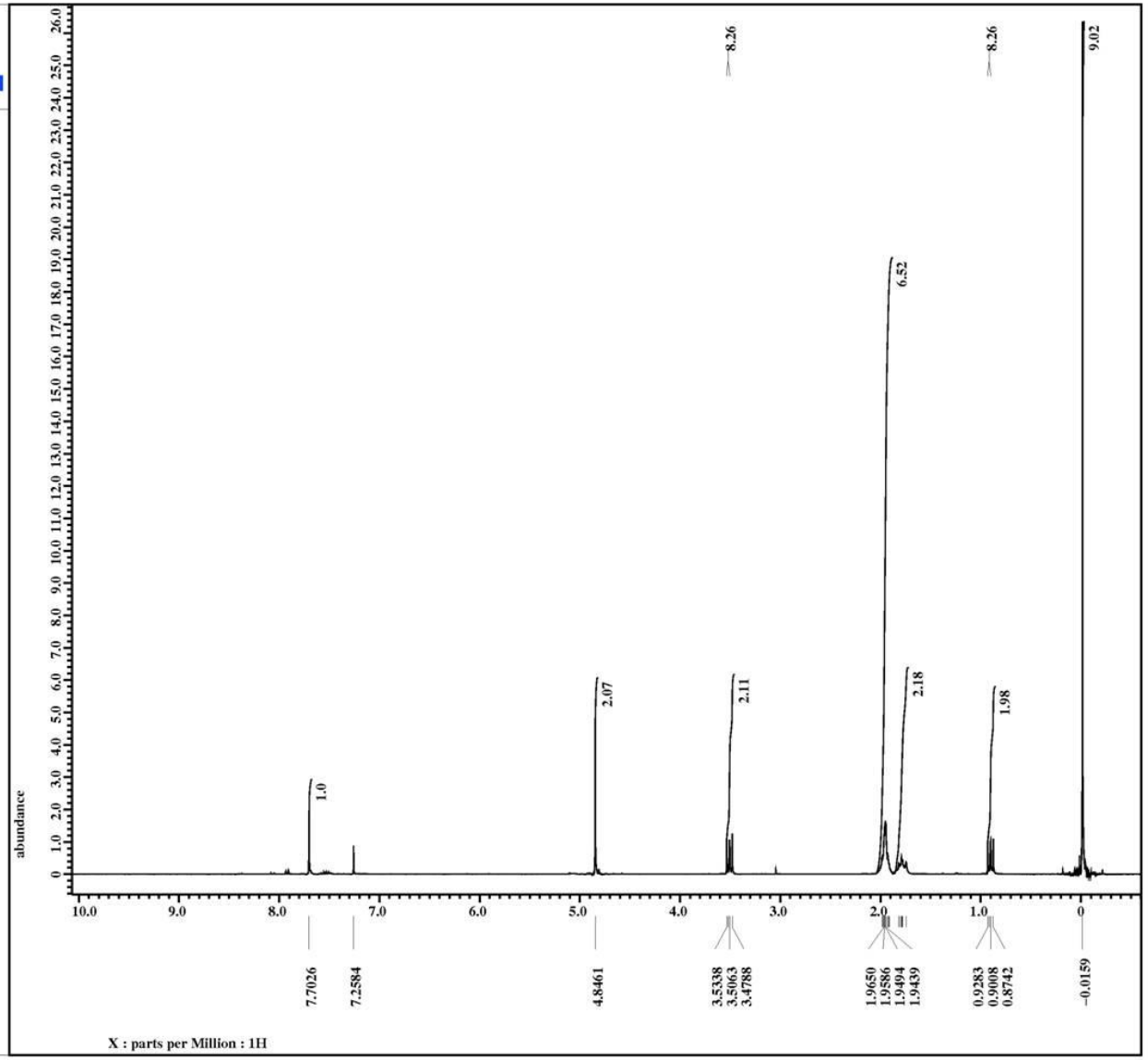

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Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#9263
Solvent           = CHLOROFORM-D
Creation_time     = 12-AUG-2006 00:55:05
Revision_time     = 17-MAR-2010 13:55:14
Current_time      = 17-MAR-2010 13:57:25

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 42
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```
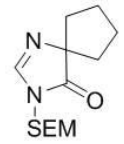
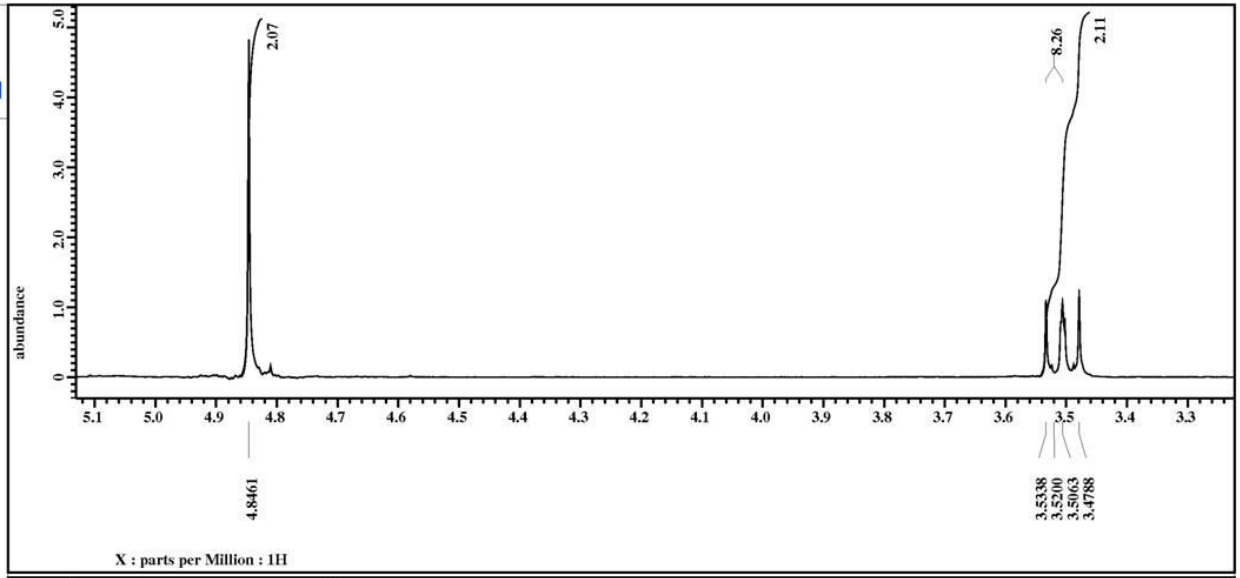
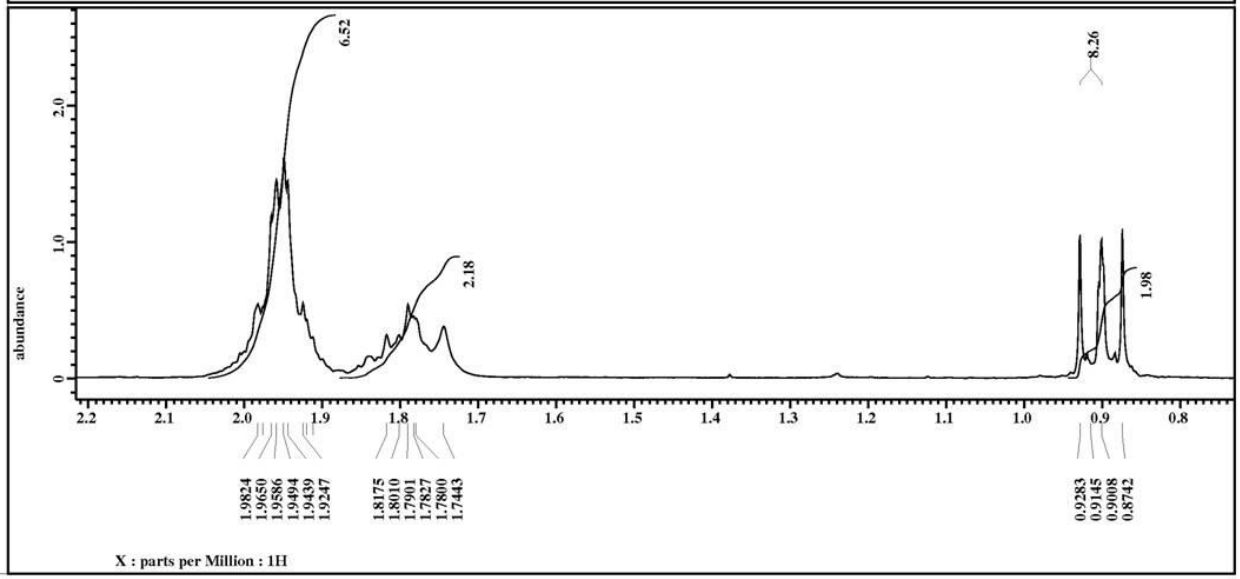
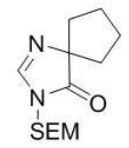
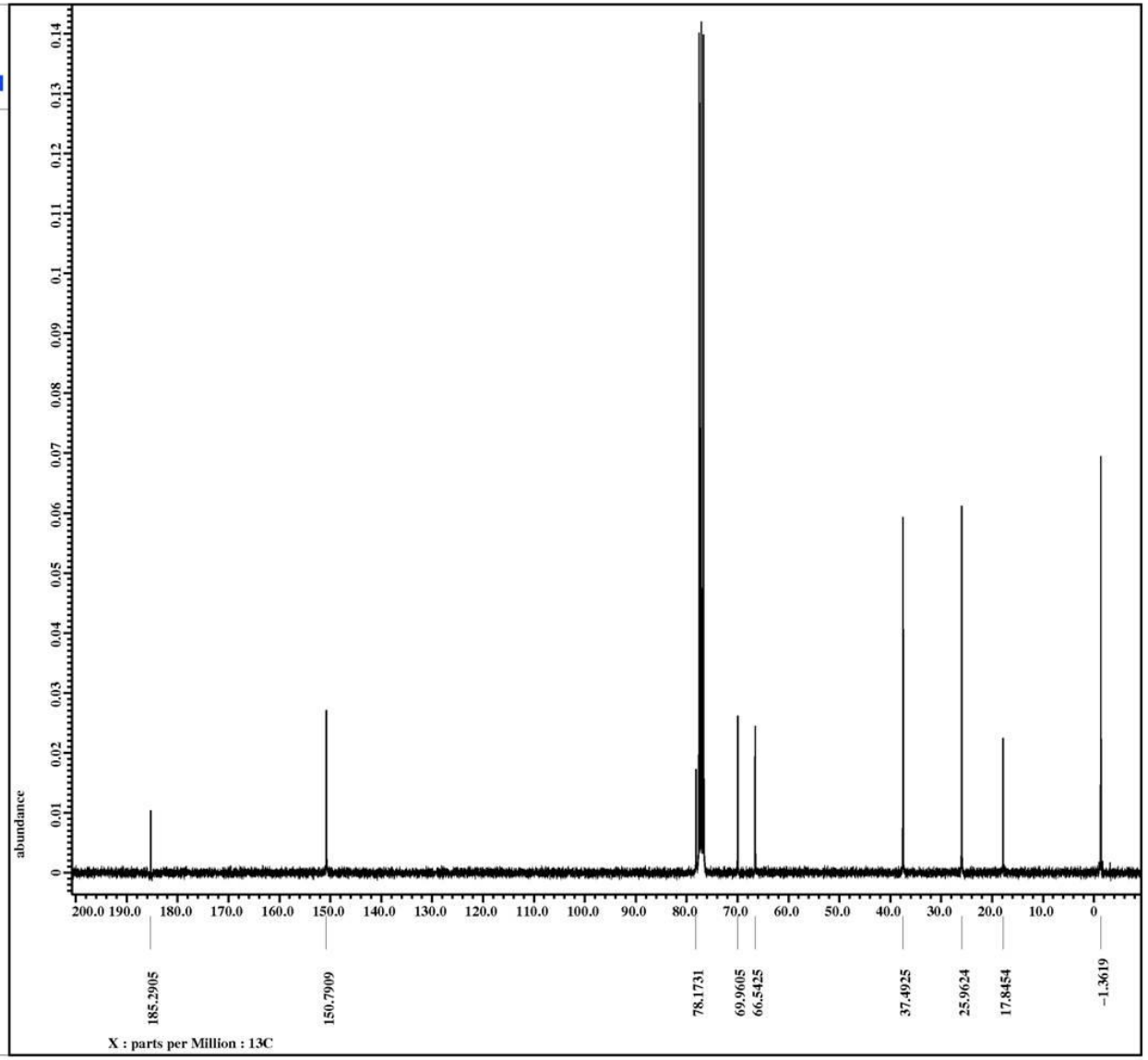







```
Filename          = II_p_066_product-4.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#10359
Solvent           = CHLOROFORM-D
Creation_time     = 12-AUG-2006 10:13:30
Revision_time     = 17-MAR-2010 14:02:35
Current_time      = 17-MAR-2010 14:03:04

Comment           = single pulse decouple
Data_format       = 1D REAL
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7000
Total_scans       = 7000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```



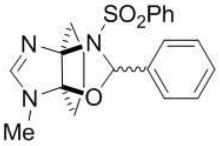


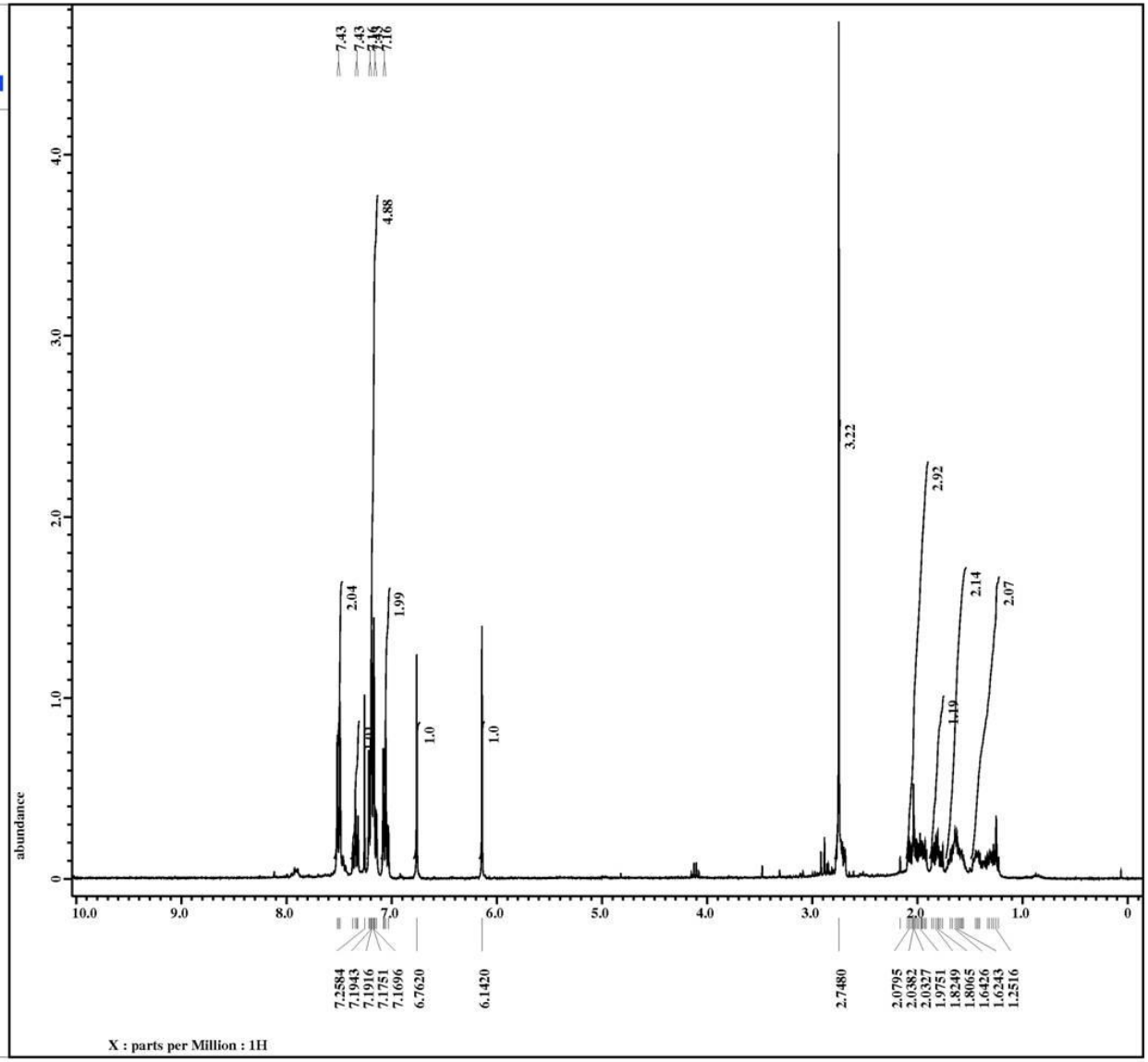
APPENDIX 3

^{1}H, ^{13}C and DEPT NMR Spectra of

(1*R**,6*S**,8*R**/8*S**)-9-benzenesulfonyl-12-methyl-7-oxa-8-phenyl-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (**94a**)

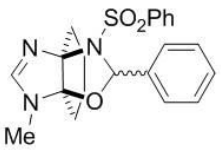
```
Filename          = p_61_II-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#403501
Solvent           = CHLOROFORM-D
Creation_time     =  6-JAN-2006 12:03:59
Revision_time     = 17-MAR-2010 14:25:57
Current_time      = 17-MAR-2010 14:26:16

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.2[dC]
```

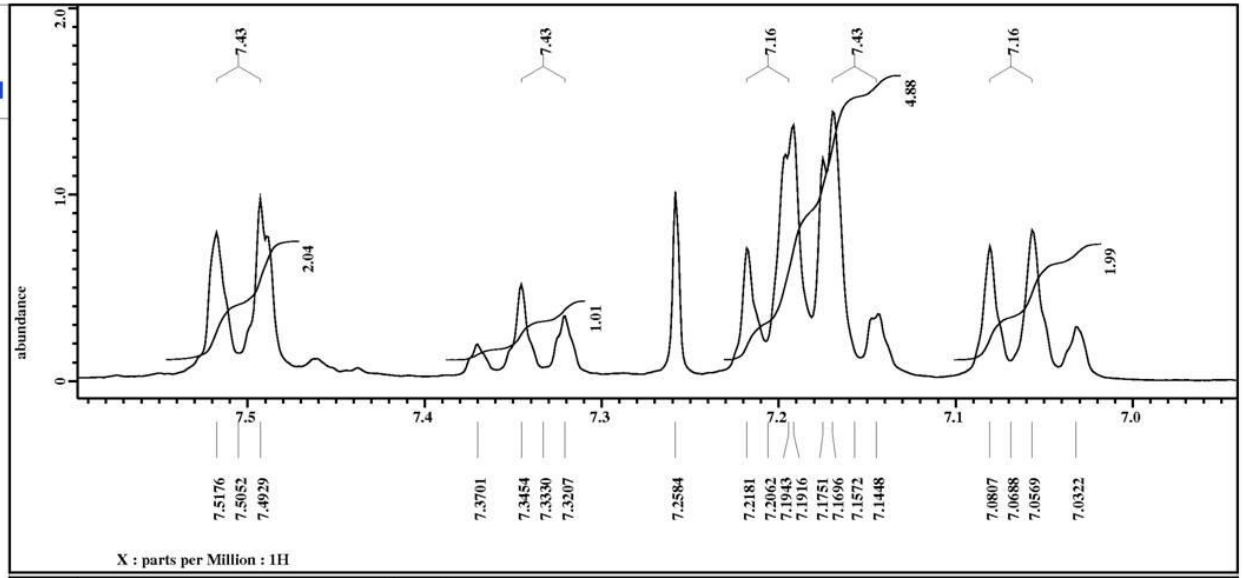


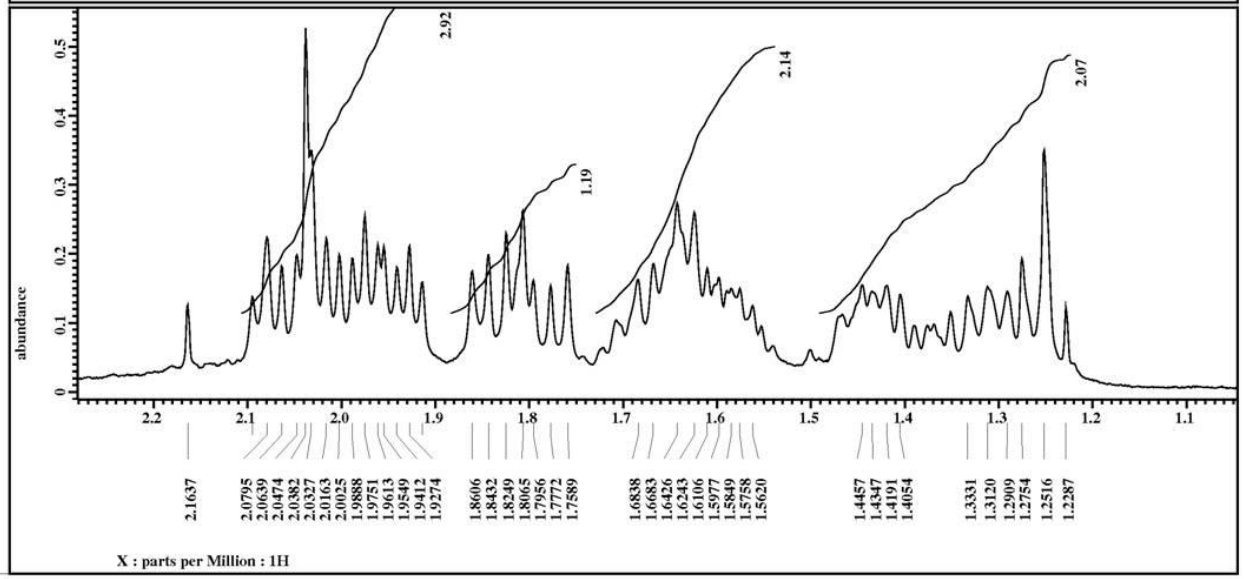


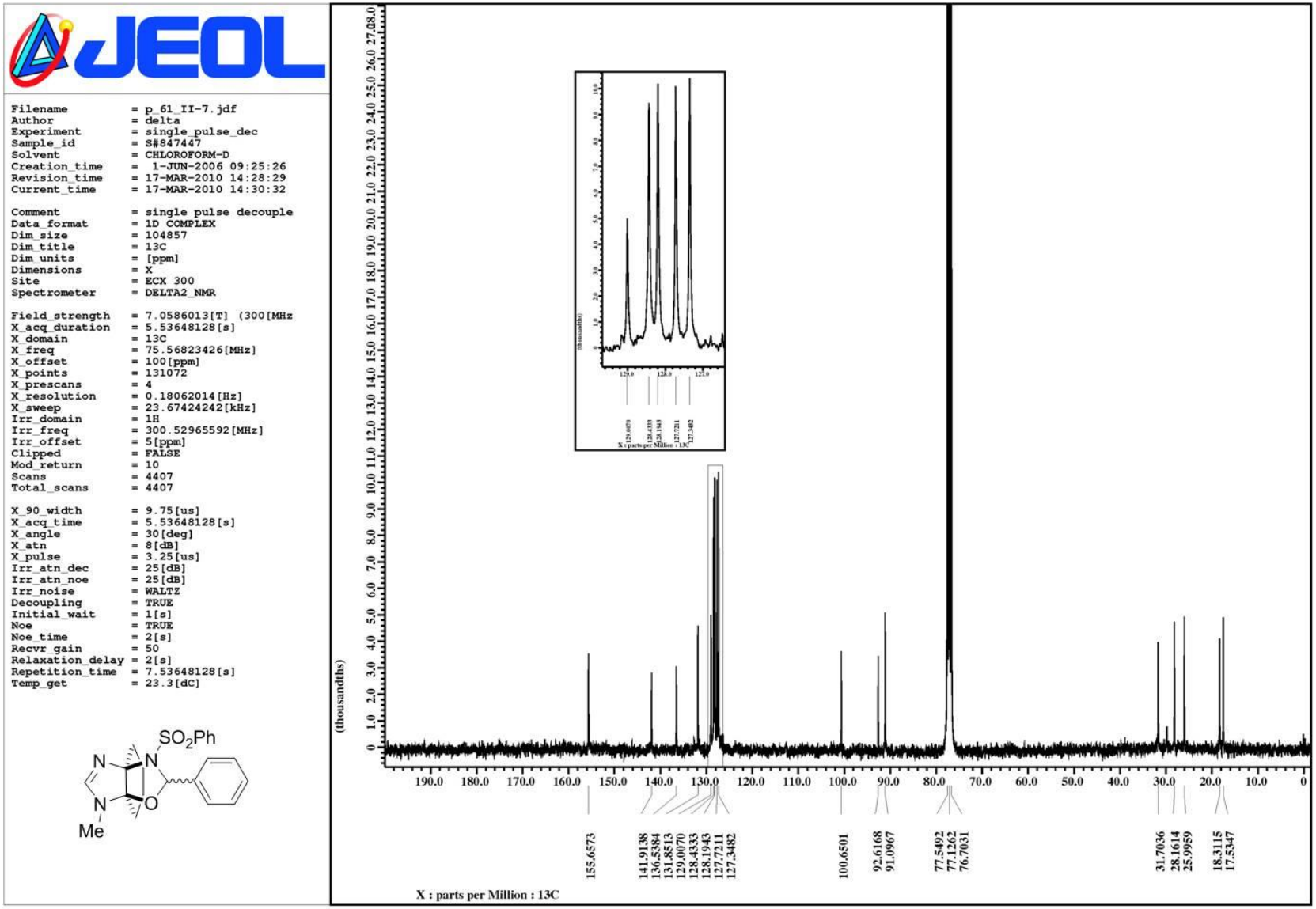
JEOL

Filename	= p_61_II-4.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#403501
Solvent	= CHLOROFORM-D
Creation_time	= 6-JAN-2006 12:03:59
Revision_time	= 17-MAR-2010 14:25:57
Current_time	= 17-MAR-2010 14:26:50
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.2[dC]



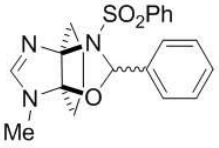


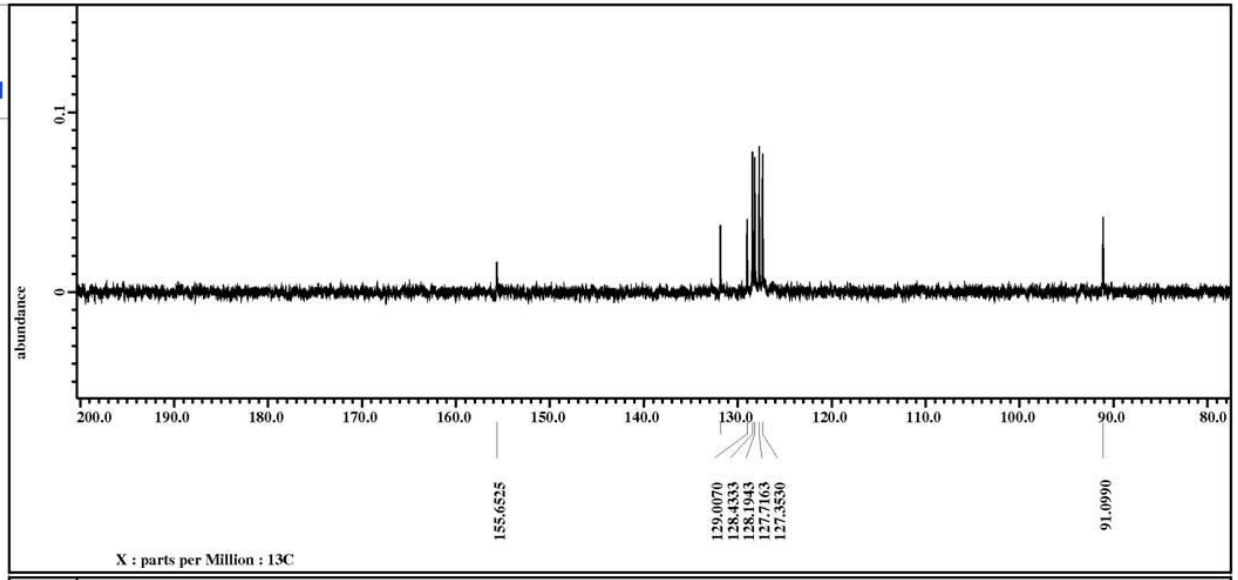


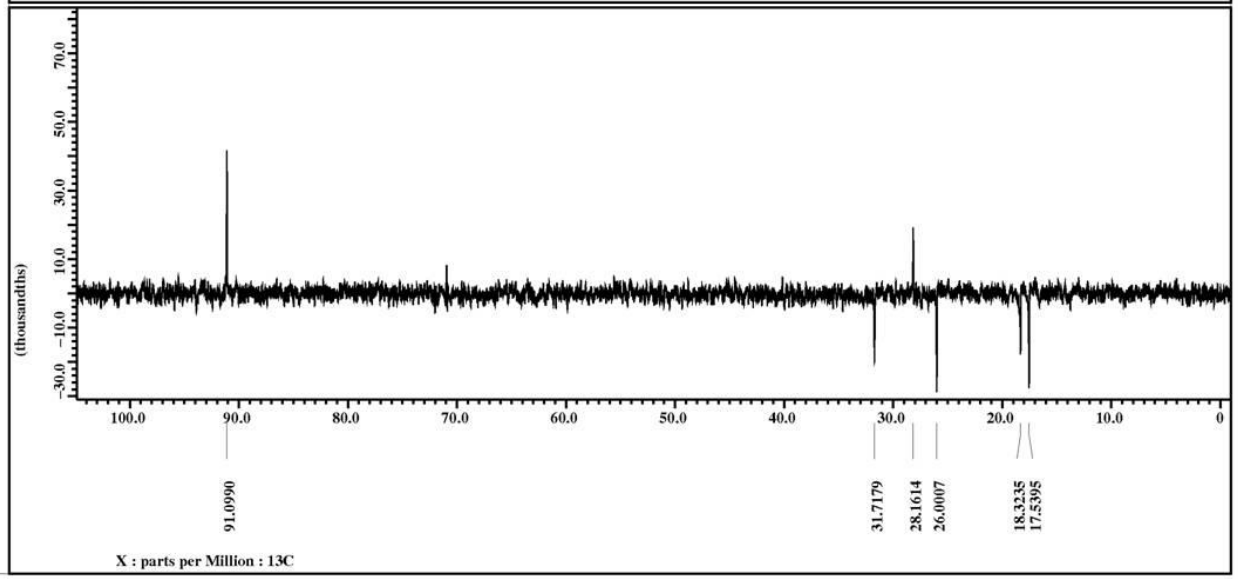


```
Filename          = p_61_II-7.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#847447
Solvent           = CHLOROFORM-D
Creation_time     =  1-JUN-2006 09:25:26
Revision_time     = 17-MAR-2010 14:28:29
Current_time      = 17-MAR-2010 14:30:32

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 104857
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 5.53648128[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 131072
X_prescans        = 4
X_resolution      = 0.18062014[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 4407
Total_scans       = 4407

X_90_width        = 9.75[us]
X_acq_time        = 5.53648128[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 7.53648128[s]
Temp_get          = 23.3[dC]
```

```
Filename          = p_61_II-5.jdf
Author            = delta
Experiment        = dept.ex2
Sample_id         = S#390834
Solvent           = CHLOROFORM-D
Creation_time     =  1-JUN-2006 11:49:34
Revision_time     = 17-MAR-2010 14:32:04
Current_time      = 17-MAR-2010 14:33:07

Comment           = DEPT with decoupling
Data_format       = 1D COMPLEX
Dim_size          = 26214
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 1.38412032[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 32768
X_prescans        = 4
X_resolution      = 0.72248054[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 338
Total_scans       = 338

X_acq_time        = 1.38412032[s]
X_atn             = 8[dB]
X_pulse           = 9.75[us]
Irr_atn           = 4[dB]
Irr_atn_dec       = 25[dB]
Irr_noise         = WALTZ
Irr_pulse         = 13.01[us]
Decoupling        = TRUE
Initial_wait      = 1[s]
J_constant        = 140[Hz]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Selection_angle   = 135[deg]
Selection_pulse   = 19.515[us]
Temp_get          = 23.8[dC]
```





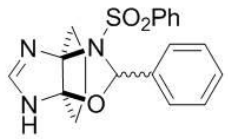


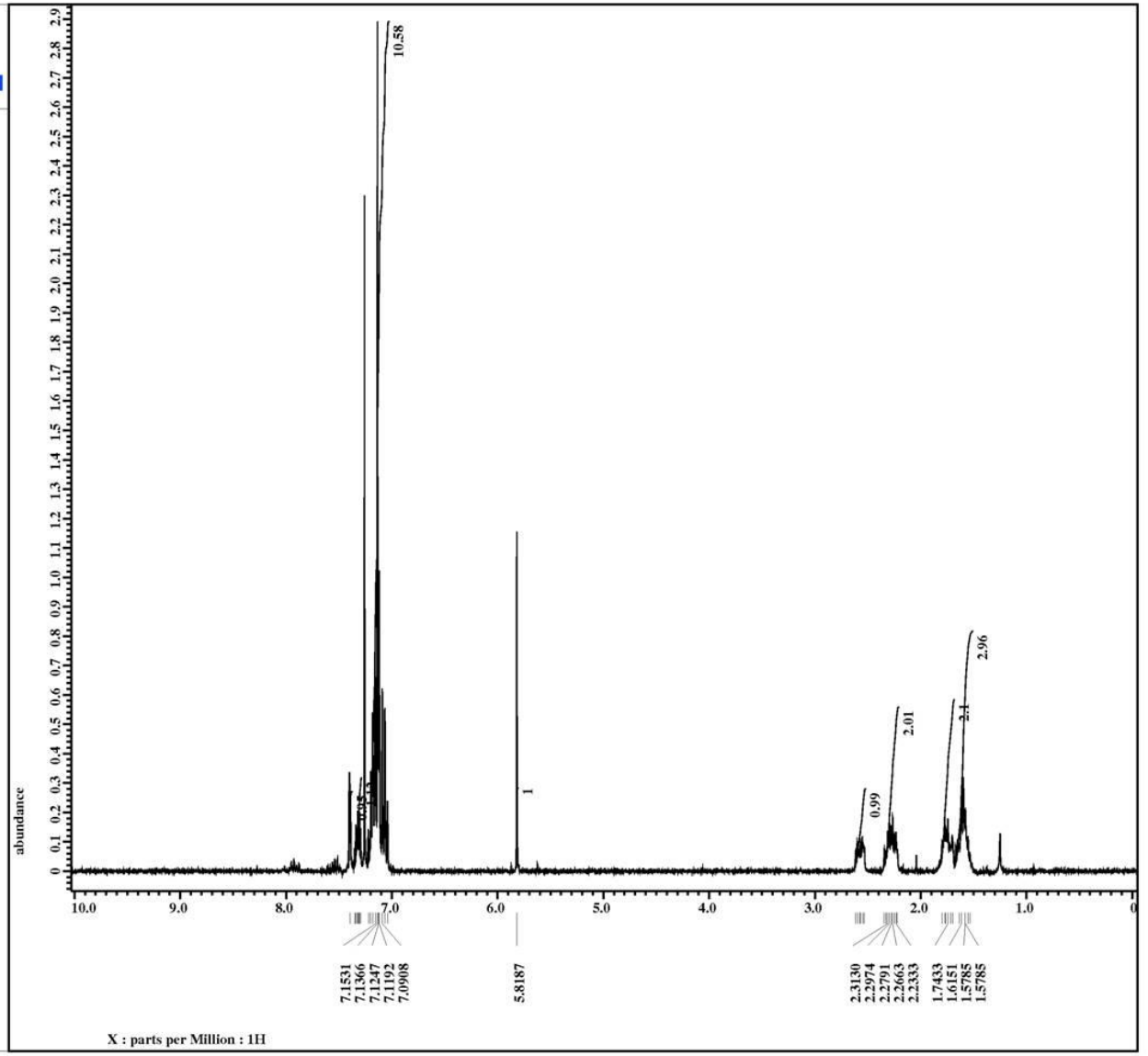
APPENDIX 4

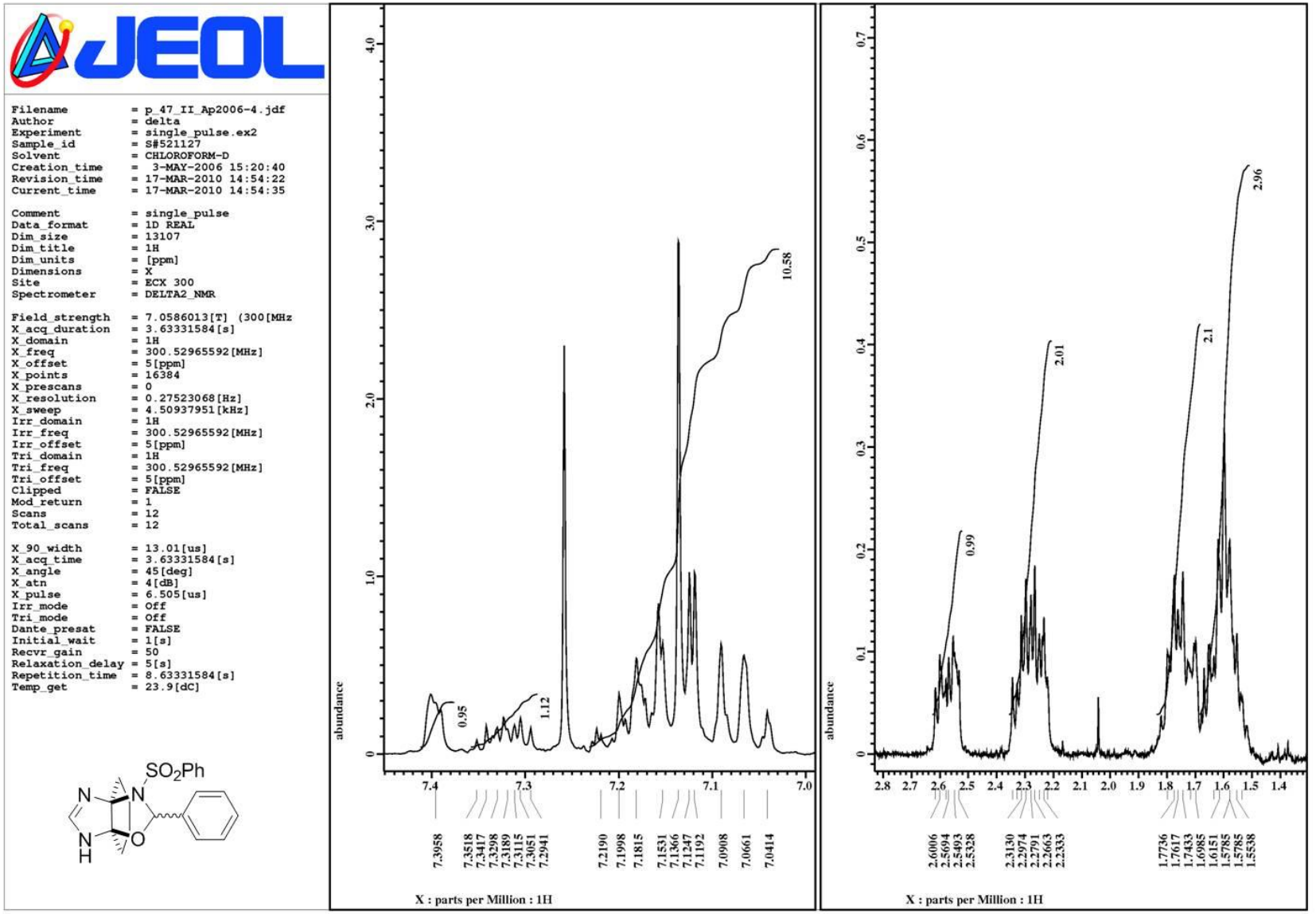
^{1}H and ^{13}C NMR Spectra of

(1*R**,6*S**,8*R**/8*S**)-9-Benzenesulfonyl-7-oxa-8-phenyl-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (**94c-i**)

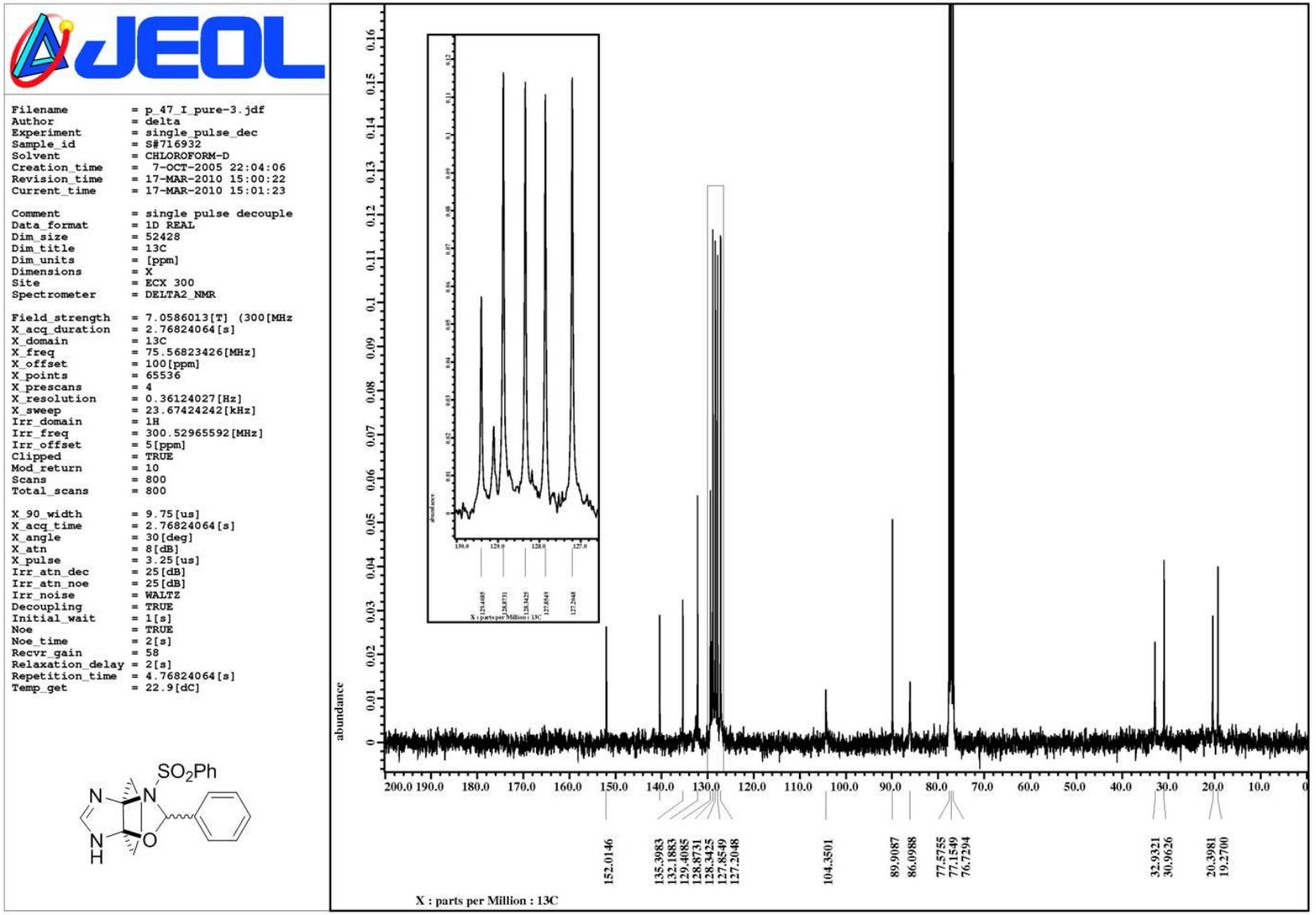
```
Filename          = p_47_II_Ap2006-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#521127
Solvent           = CHLOROFORM-D
Creation_time     =  3-MAY-2006 15:20:40
Revision_time     =  3-MAY-2006 14:35:41
Current_time      = 17-MAR-2010 14:53:21

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.9[dC]
```







```
Filename          = p_47_II_Ap2006-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#521127
Solvent           = CHLOROFORM-D
Creation_time     =  3-MAY-2006 15:20:40
Revision_time     = 17-MAR-2010 14:54:22
Current_time      = 17-MAR-2010 14:54:35

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.9[dC]
```

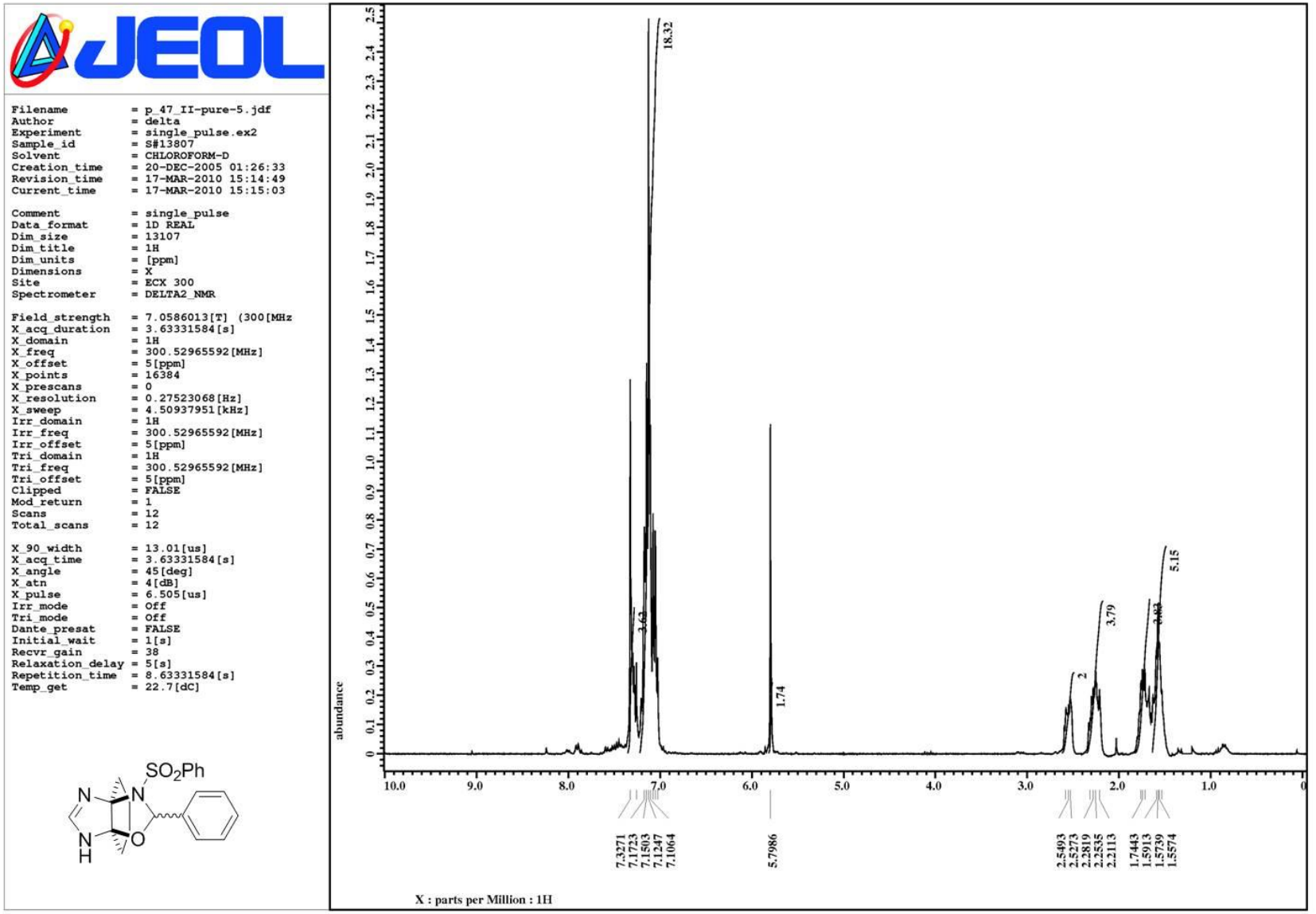


```
Filename          = p_47_I_pure-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#716932
Solvent           = CHLOROFORM-D
Creation_time     =  7-OCT-2005 22:04:06
Revision_time     = 17-MAR-2010 15:00:22
Current_time      = 17-MAR-2010 15:01:23

Comment           = single pulse decouple
Data_format       = 1D REAL
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 10
Scans             = 800
Total_scans       = 800

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 58
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.9[dC]
```

APPENDIX 5

^{1}H, ^{13}C and DEPT NMR Spectra of

(1*R**,6*S**,8*R**/8*S**)-9-Benzenesulfonyl-7-oxa-8-phenyl-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (**94c-ii**)



Filename = p_47_II-pure-5.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#13807
Solvent = CHLOROFORM-D
Creation_time = 20-DEC-2005 01:26:33
Revision_time = 17-MAR-2010 15:14:49
Current_time = 17-MAR-2010 15:15:03

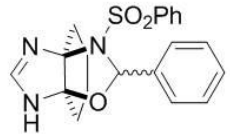
Comment = single_pulse
Data_format = 1D REAL
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR

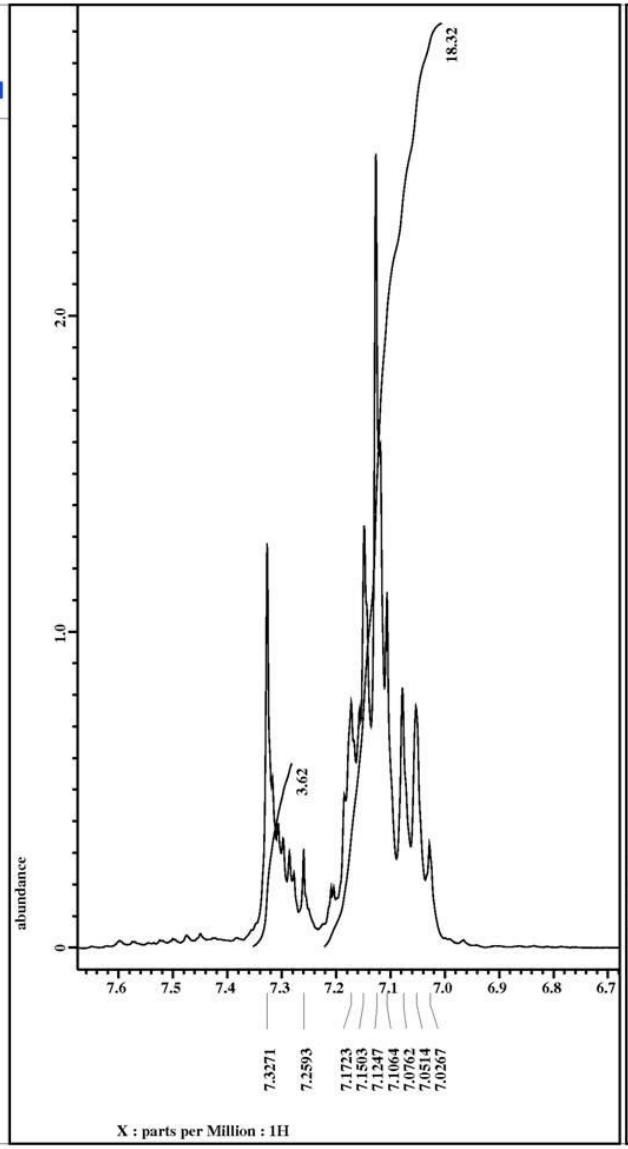
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12

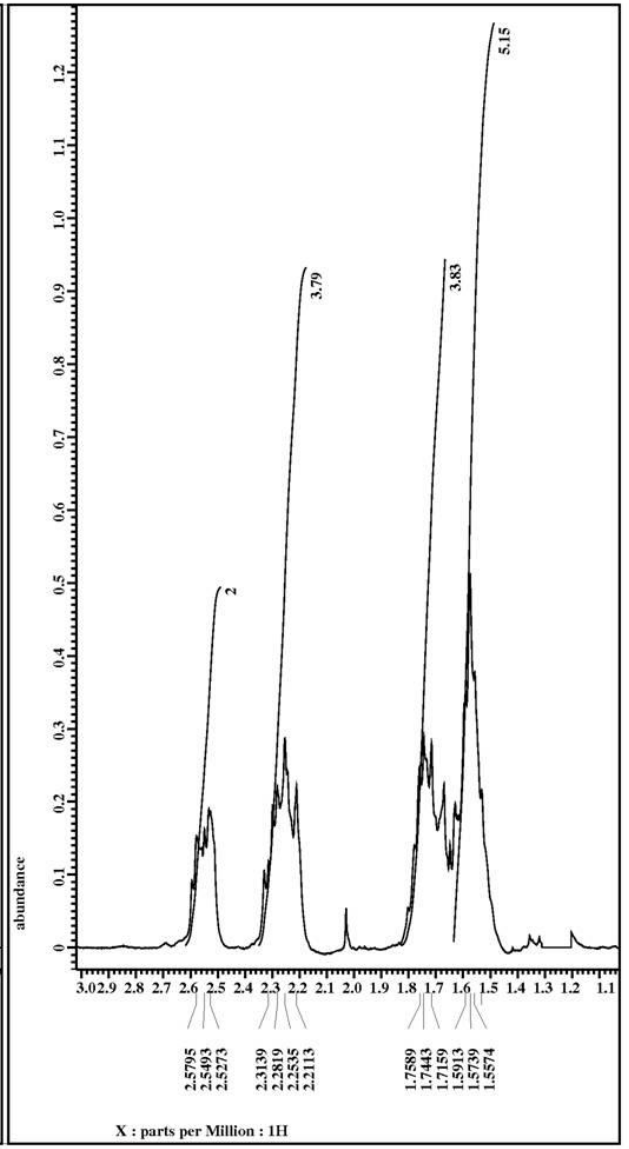
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 38
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 22.7[dC]

JEOL

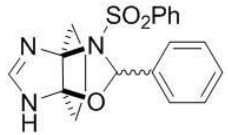
Filename	= p_47_II-pure-5.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#13807
Solvent	= CHLOROFORM-D
Creation_time	= 20-DEC-2005 01:26:33
Revision_time	= 17-MAR-2010 15:14:49
Current_time	= 17-MAR-2010 15:15:32
Comment	= single_pulse
Data_format	= 1D REAL
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 38
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.7[dC]

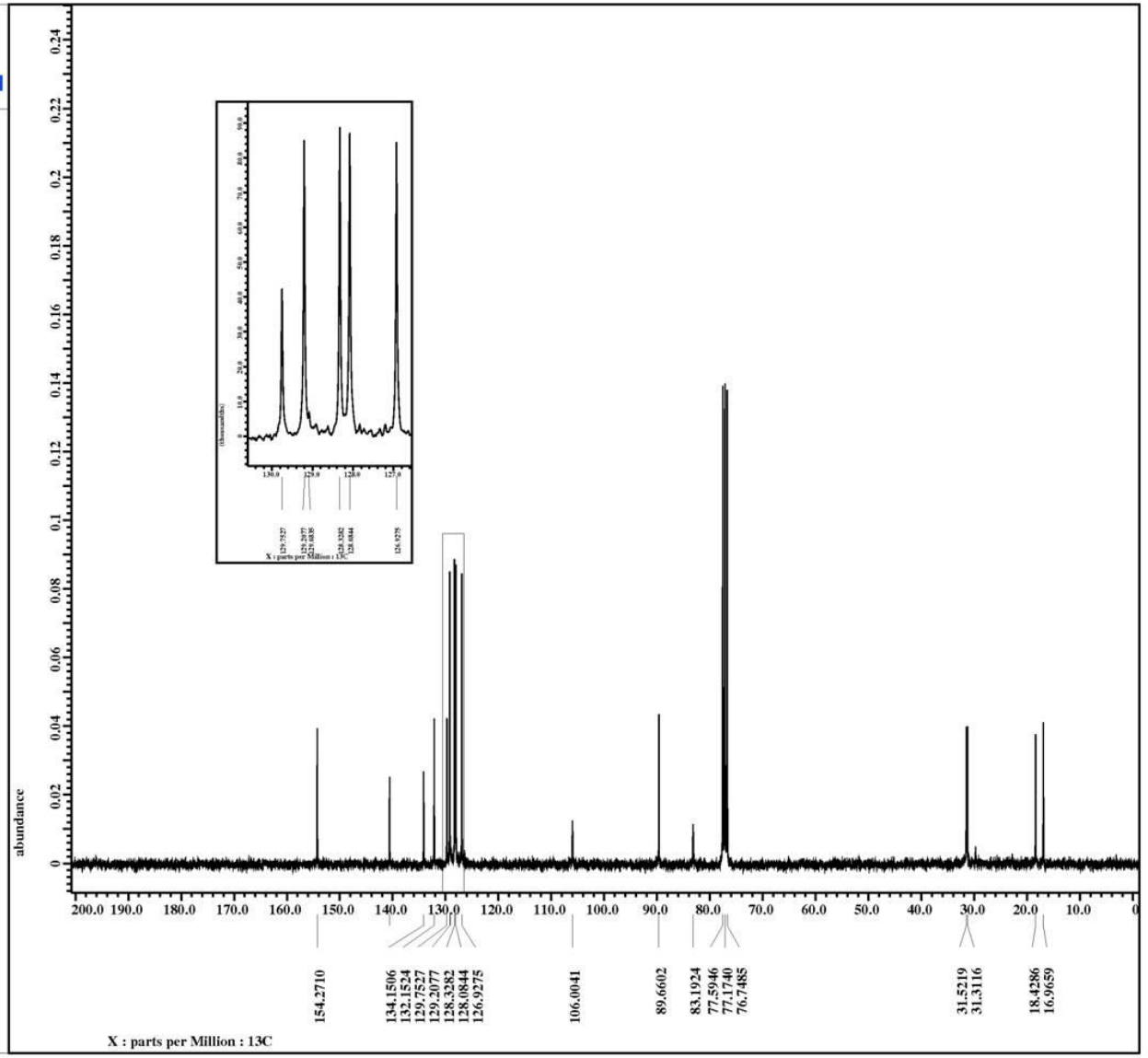


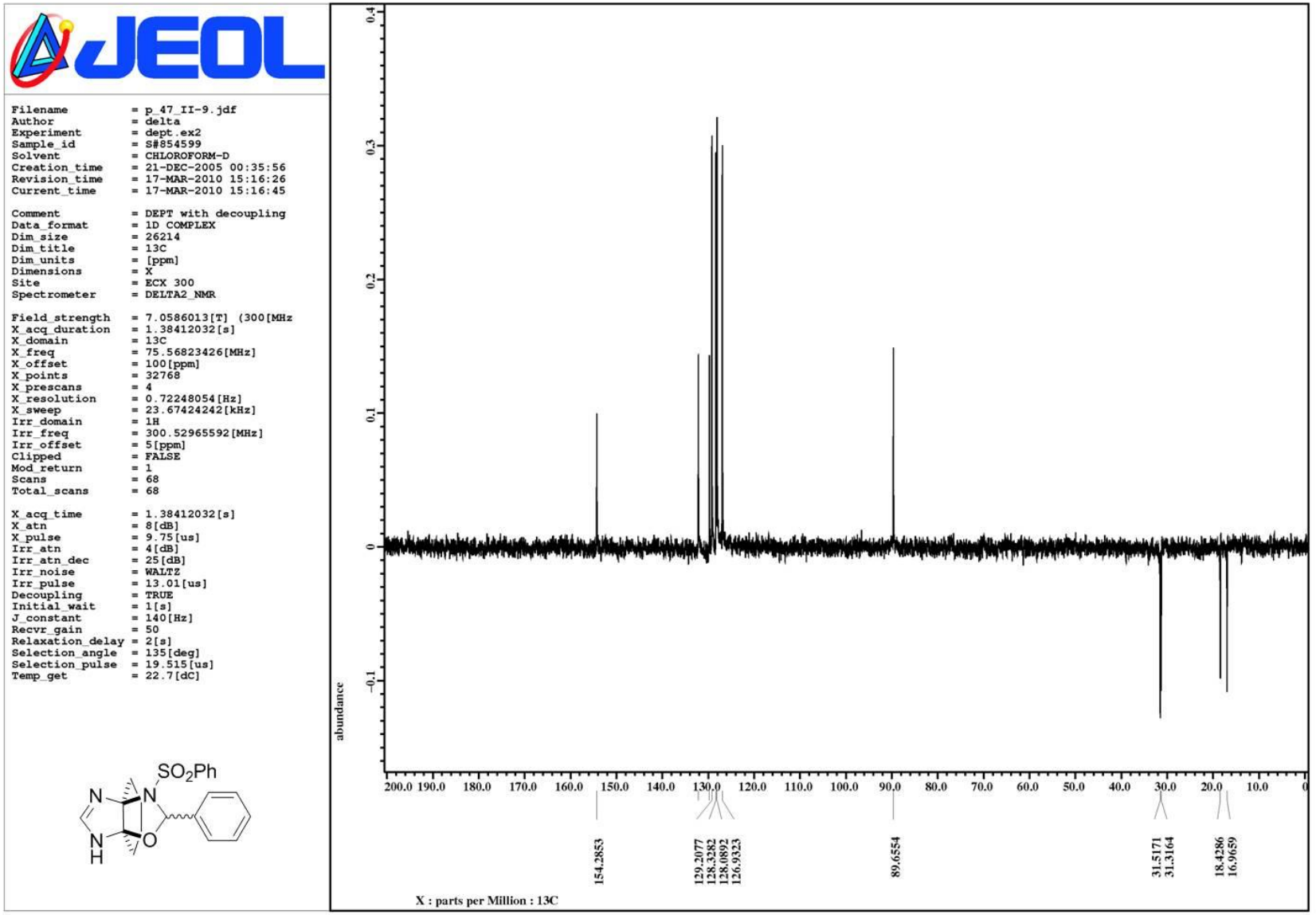




Filename	= p_47_II-4.jdf
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#43904
Solvent	= CHLOROFORM-D
Creation_time	= 20-DEC-2005 03:17:08
Revision_time	= 17-MAR-2010 15:13:14
Current_time	= 17-MAR-2010 15:13:55
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 800
Total_scans	= 800
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 22.7[dC]







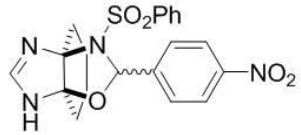
```
Filename          = p_47_II-9.jdf
Author            = delta
Experiment        = dept.ex2
Sample_id         = S#854599
Solvent           = CHLOROFORM-D
Creation_time     = 21-DEC-2005 00:35:56
Revision_time     = 17-MAR-2010 15:16:26
Current_time      = 17-MAR-2010 15:16:45

Comment           = DEPT with decoupling
Data_format       = 1D COMPLEX
Dim_size          = 26214
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 1.38412032[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 32768
X_prescans        = 4
X_resolution      = 0.72248054[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 68
Total_scans       = 68

X_acq_time        = 1.38412032[s]
X_atn             = 8[dB]
X_pulse           = 9.75[us]
Irr_atn           = 4[dB]
Irr_atn_dec       = 25[dB]
Irr_noise         = WALTZ
Irr_pulse         = 13.01[us]
Decoupling        = TRUE
Initial_wait      = 1[s]
J_constant        = 140[Hz]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Selection_angle   = 135[deg]
Selection_pulse   = 19.515[us]
Temp_get          = 22.7[dC]
```

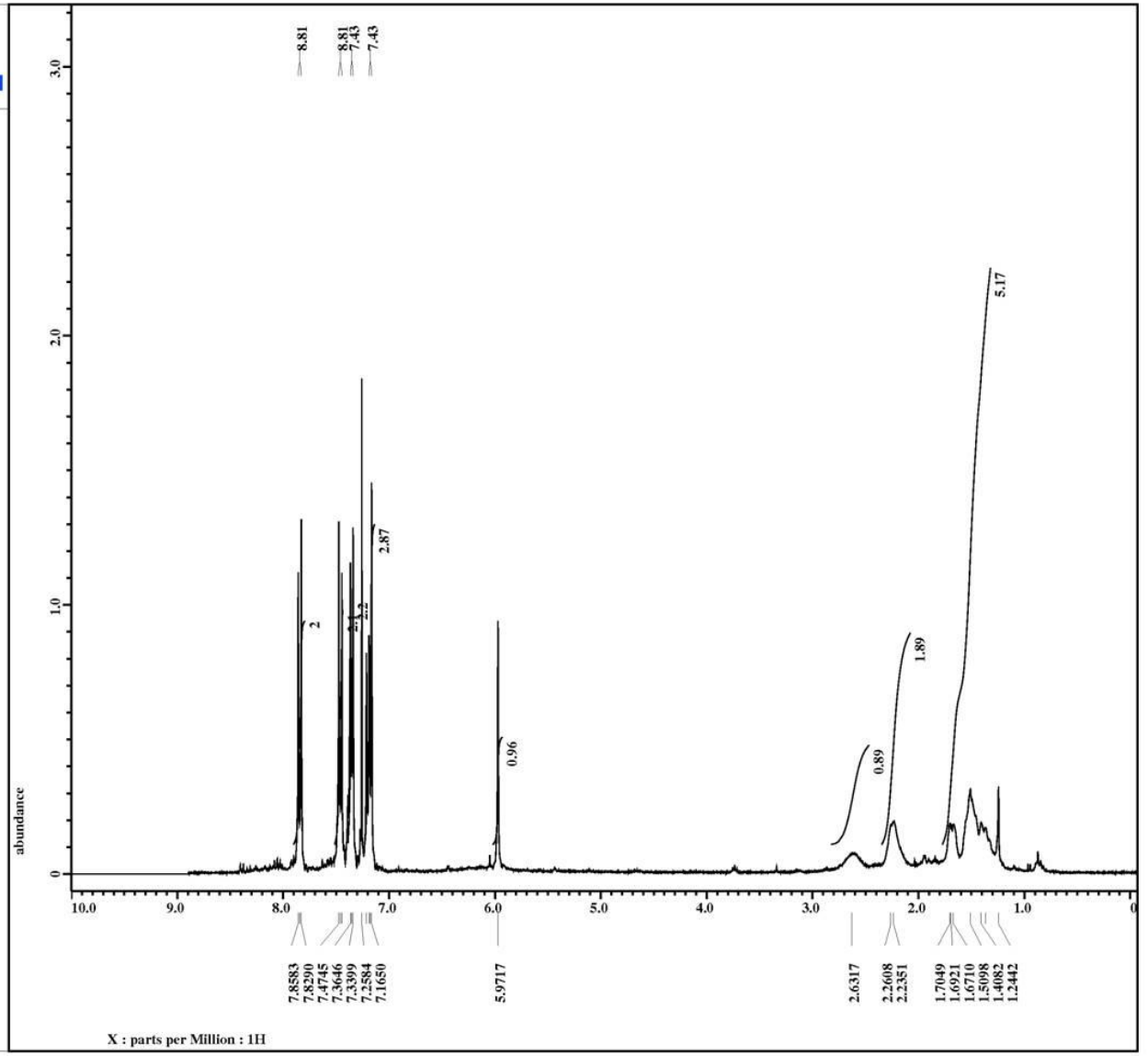
APPENDIX 6

^{1}H and ^{13}C NMR Spectra of

(1*R**,6*S**,8*R**/8*S**)- 9-Benzenesulfonyl-8-(4-nitrophenyl)-7-oxa-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (**94d-i**)

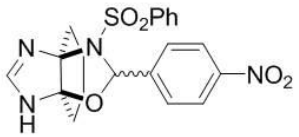
```
Filename          = II_p_122_I-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#370964
Solvent           = CHLOROFORM-D
Creation_time     =  8-DEC-2006 11:06:36
Revision_time     = 16-MAR-2010 21:51:12
Current_time      = 16-MAR-2010 21:55:42

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 21.7[dC]
```

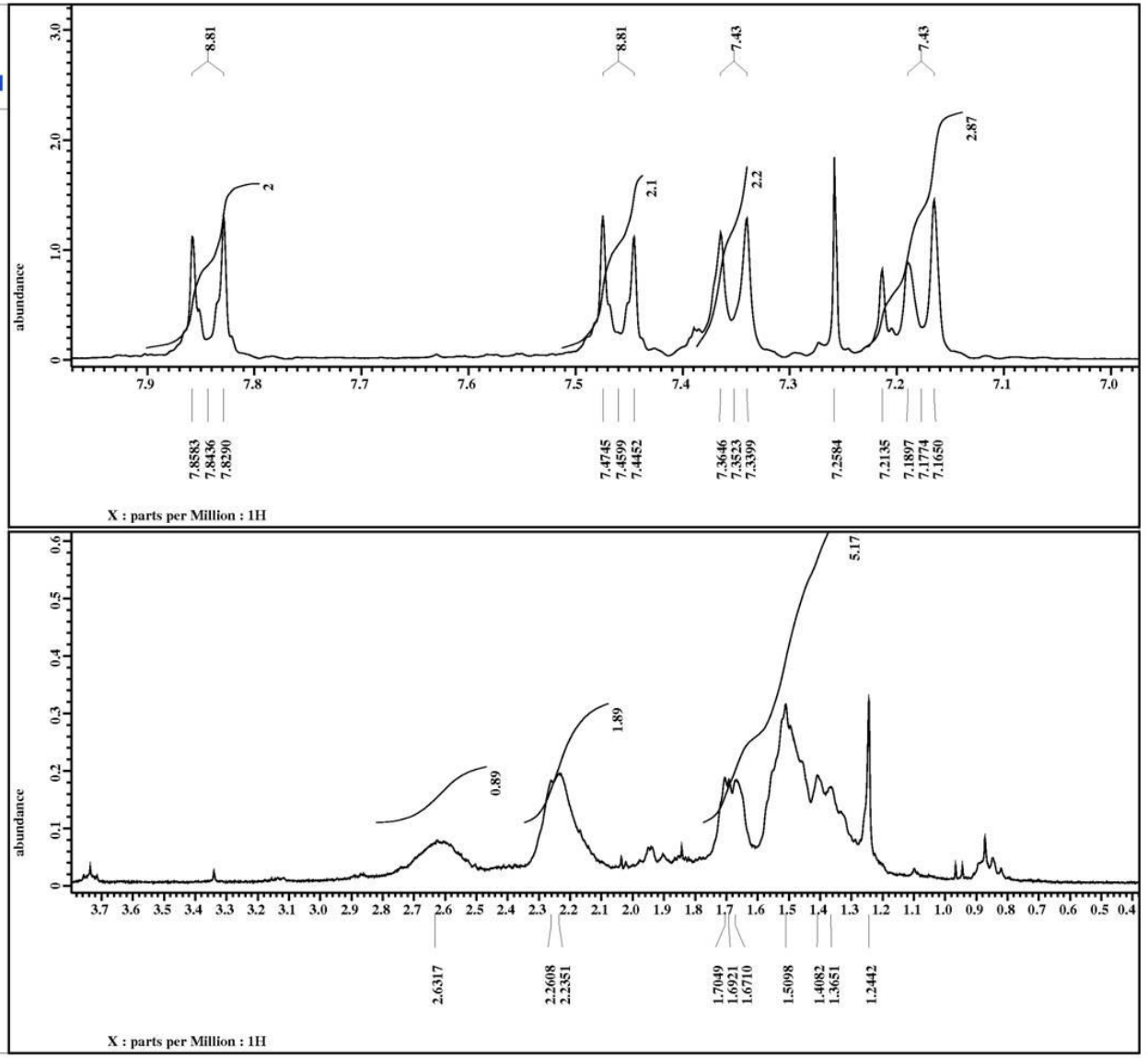




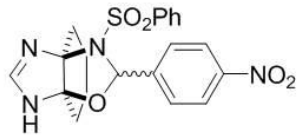
JEOL

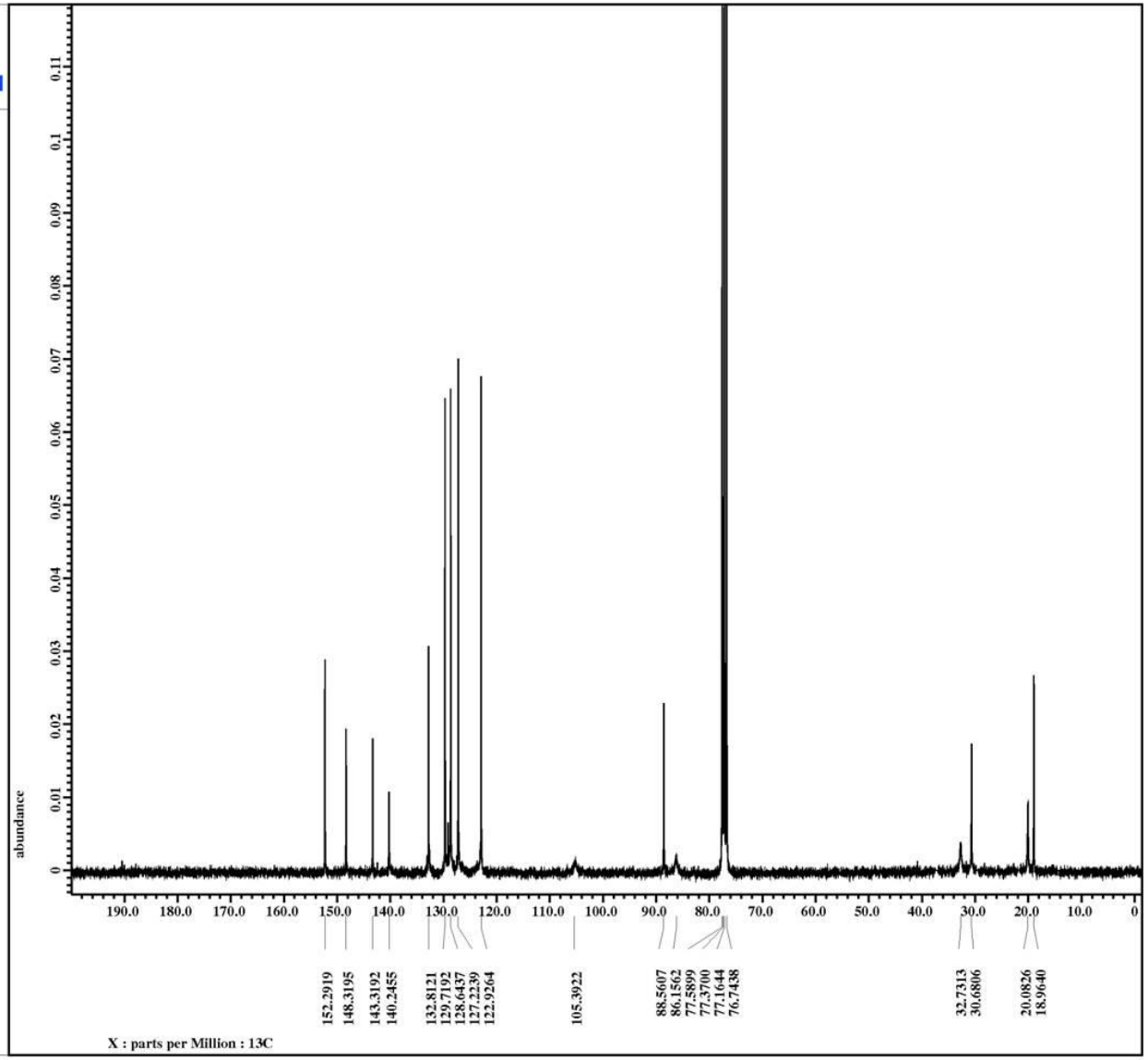
Filename	= II_p_122_I-2.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#370964
Solvent	= CHLOROFORM-D
Creation_time	= 8-DEC-2006 11:06:36
Revision_time	= 16-MAR-2010 21:51:12
Current_time	= 16-MAR-2010 21:56:23
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 21.7[dC]





```
Filename          = II_p_122_I-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#29518
Solvent           = CHLOROFORM-D
Creation_time     = 12-DEC-2006 09:55:08
Revision_time     = 16-MAR-2010 22:01:09
Current_time      = 16-MAR-2010 22:02:13

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6280
Total_scans       = 6280

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.6[dC]
```





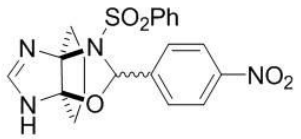
APPENDIX 7

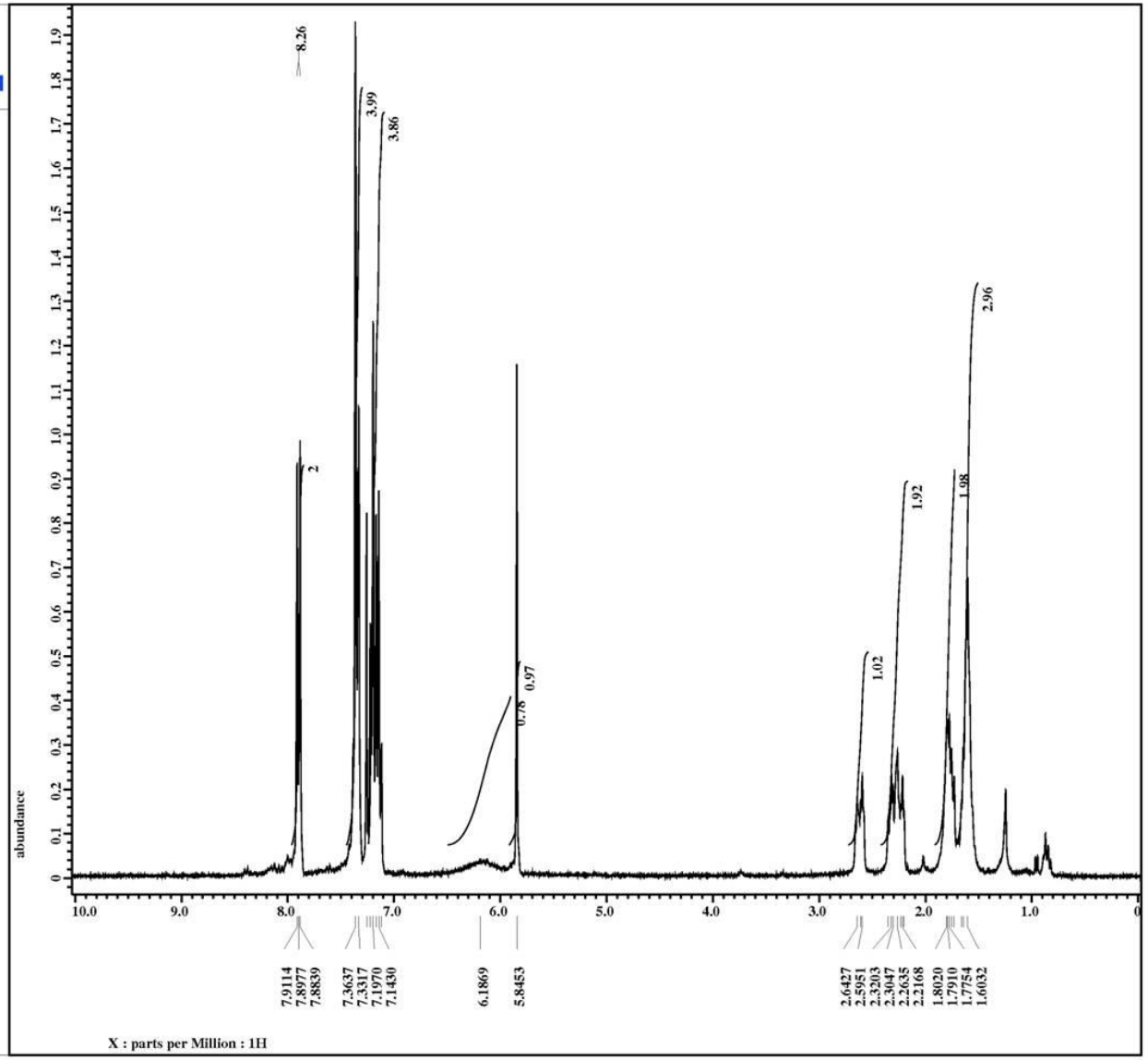
^{1}H and ^{13}C NMR Spectra of

(1*R**,6*S**,8*R**/8*S**)- 9-Benzenesulfonyl-8-(4-nitrophenyl)-7-oxa-9,10,12-triazatricyclo[4.3.3.0]dodec-10-ene (**94d-ii**)

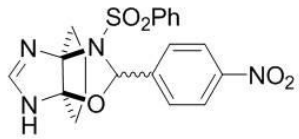
JEOL

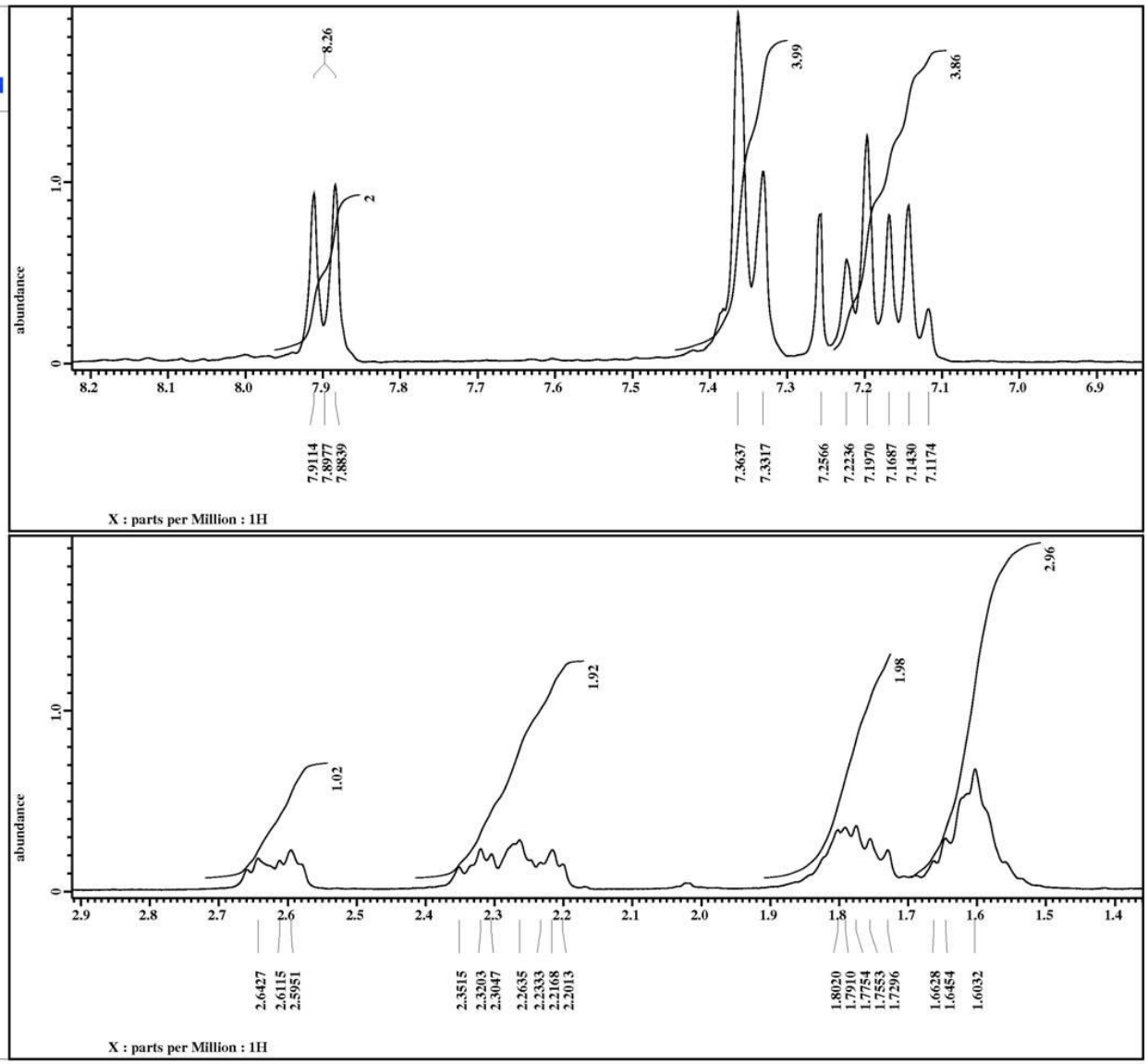
```
Filename          = II_p_122_II-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#378564
Solvent           = CHLOROFORM-D
Creation_time     =  8-DEC-2006 11:19:16
Revision_time     = 16-MAR-2010 22:16:21
Current_time      = 16-MAR-2010 22:16:34

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.4[dC]
```



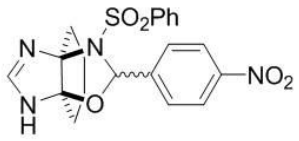


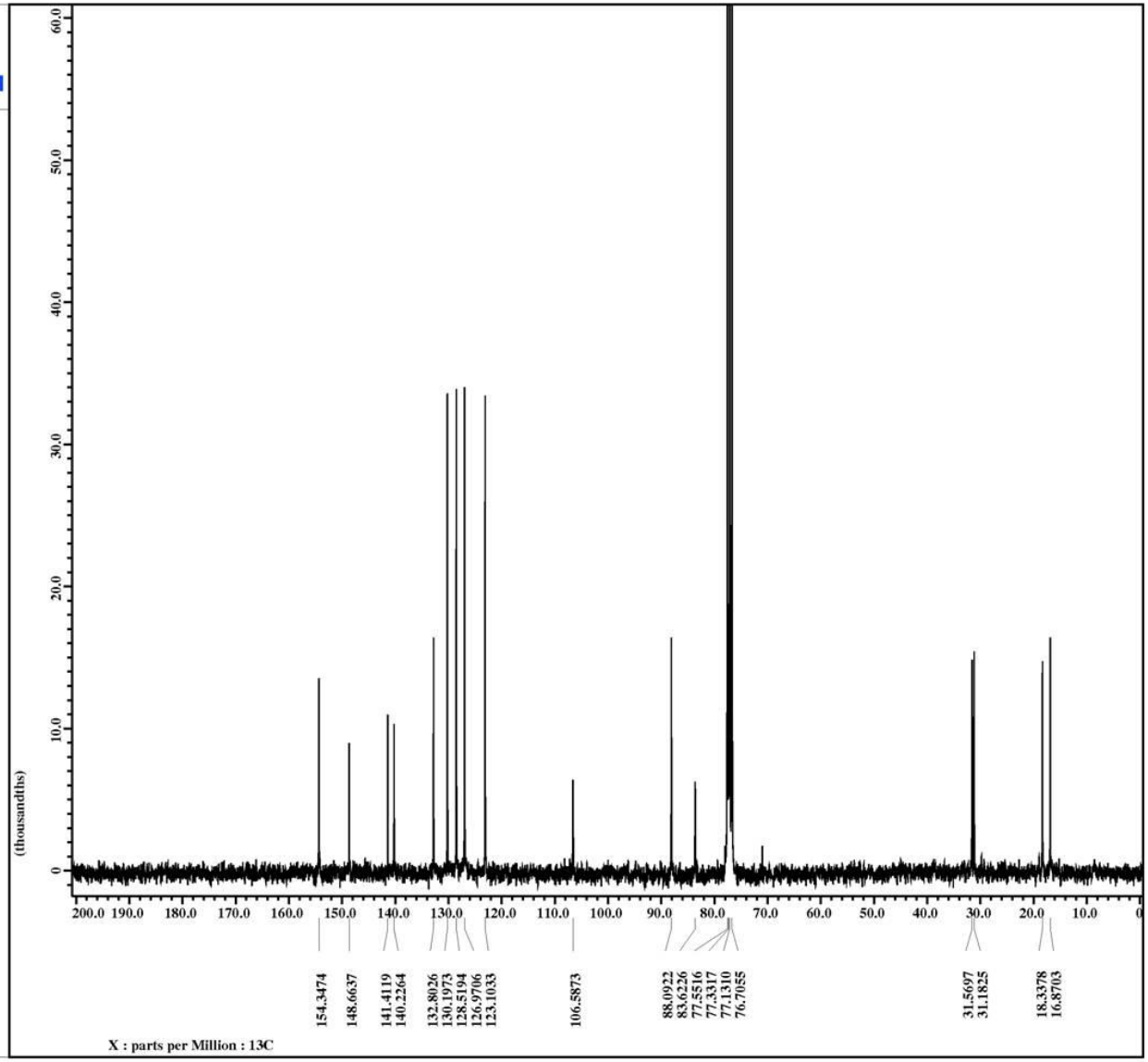
Filename	= II_p_122_II-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#378564
Solvent	= CHLOROFORM-D
Creation_time	= 8-DEC-2006 11:19:16
Revision_time	= 16-MAR-2010 22:16:21
Current_time	= 16-MAR-2010 22:16:58
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.4[dC]





```
Filename          = II_p_122_II-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#625777
Solvent           = CHLOROFORM-D
Creation_time     =  9-DEC-2006 20:45:26
Revision_time     = 16-MAR-2010 22:11:10
Current_time      = 16-MAR-2010 22:11:39

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1960
Total_scans       = 1960

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 24.1[dC]
```



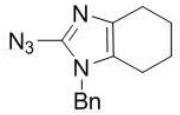


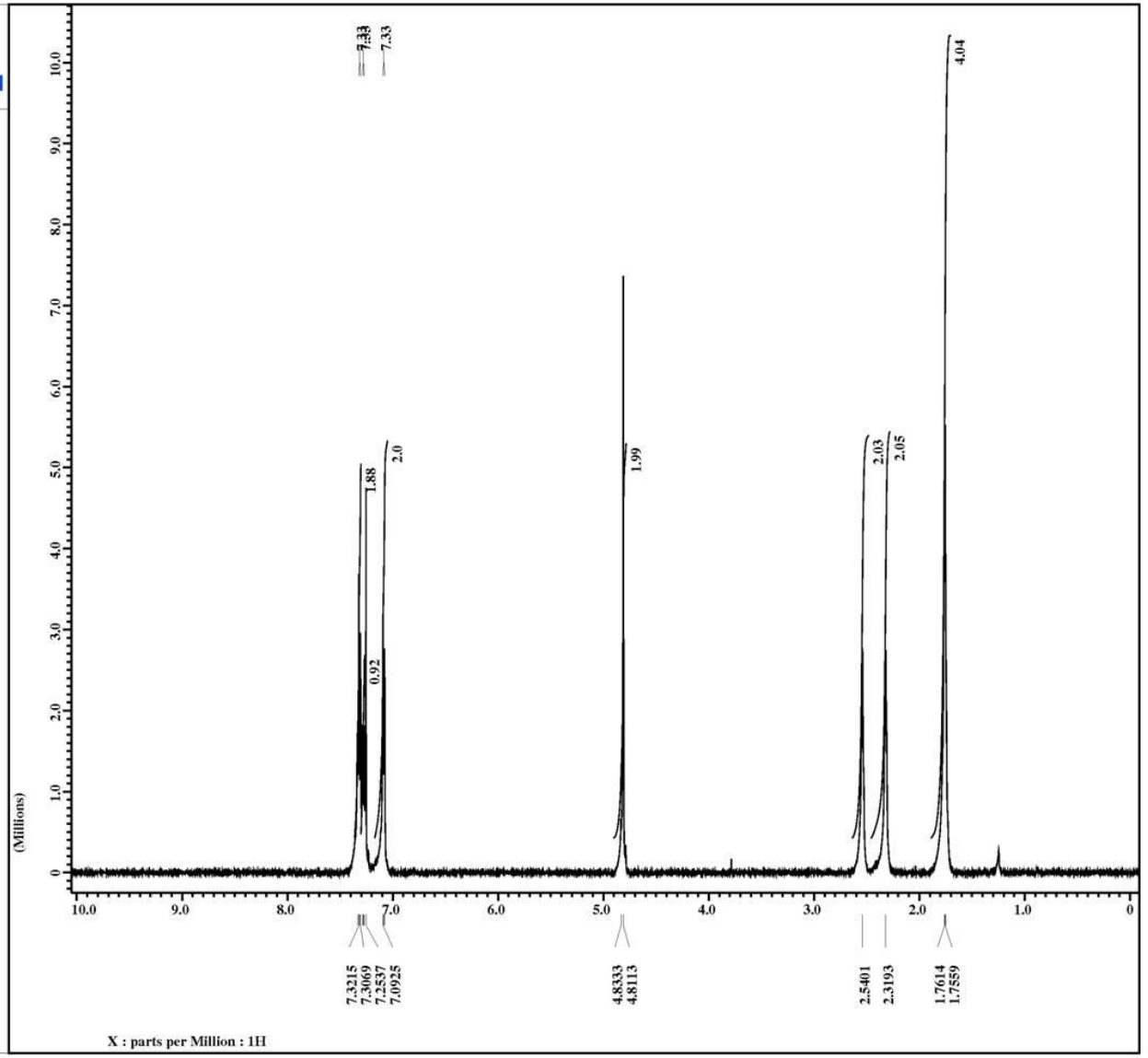
APPENDIX 8

^{1}H and ^{13}C NMR Spectra of

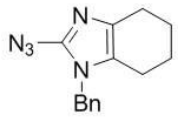
2-Azido-1-benzyl-4,5,6,7-tetrahydro-1*H* -benzimidazole (**97**)

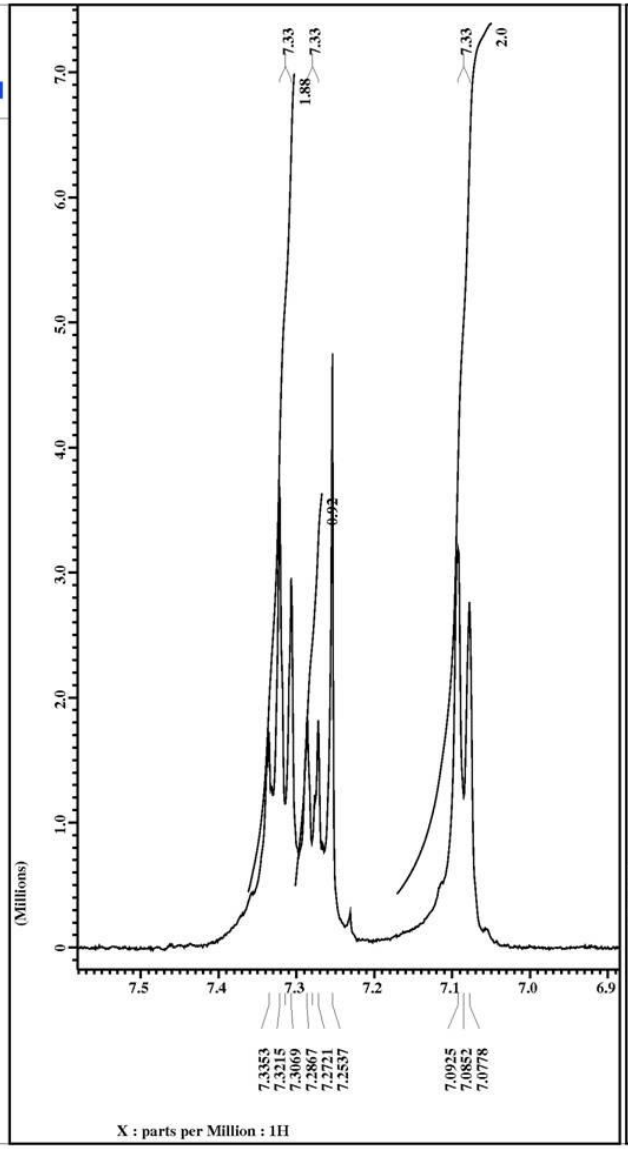
```
Filename          = II_P_135_AzidoBnTHB-4
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#804284
Solvent           = CHLOROFORM-D
Creation_time     =  4-JAN-2007 00:32:58
Revision_time     = 16-MAR-2010 22:26:22
Current_time      = 16-MAR-2010 22:29:08

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 8
Total_scans       = 8

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 23.8[dC]
Unblank_time      = 2[us]
```

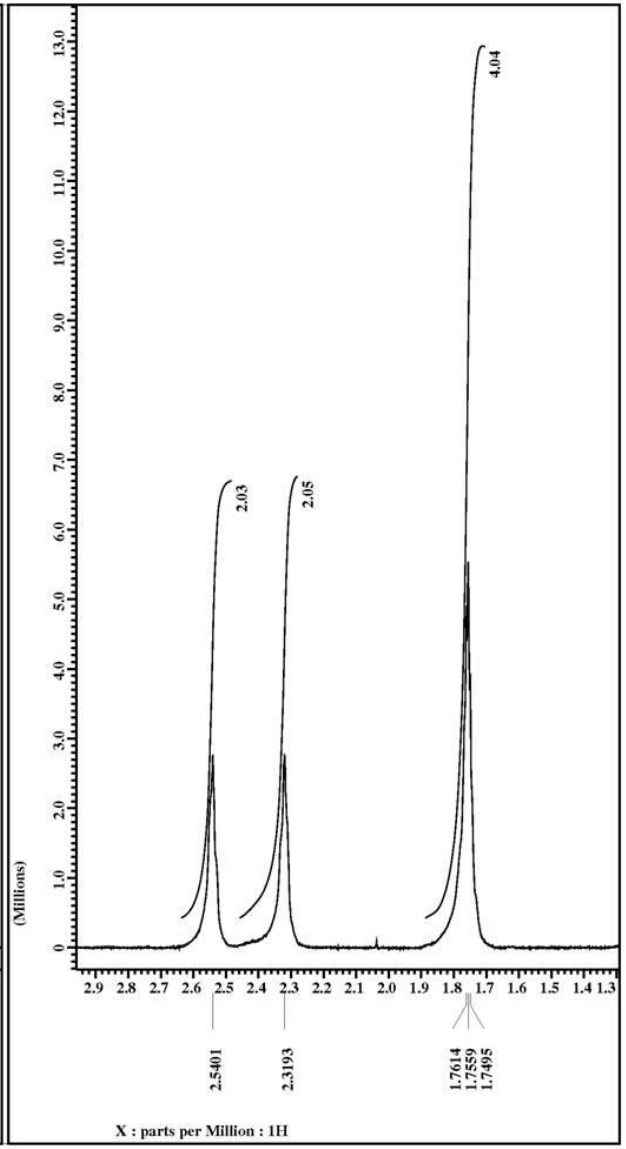




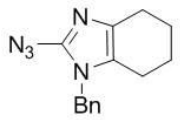
Filename	= II_P_135_AzidoBnTHB-4
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#804284
Solvent	= CHLOROFORM-D
Creation_time	= 4-JAN-2007 00:32:58
Revision_time	= 16-MAR-2010 22:26:22
Current_time	= 16-MAR-2010 22:29:47
Comment	= Single Pulse Experime
Data_format	= 1D REAL
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 8
Total_scans	= 8
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 20
Relaxation_delay	= 4[s]
Temp_get	= 23.8[dC]
Unblank_time	= 2[us]

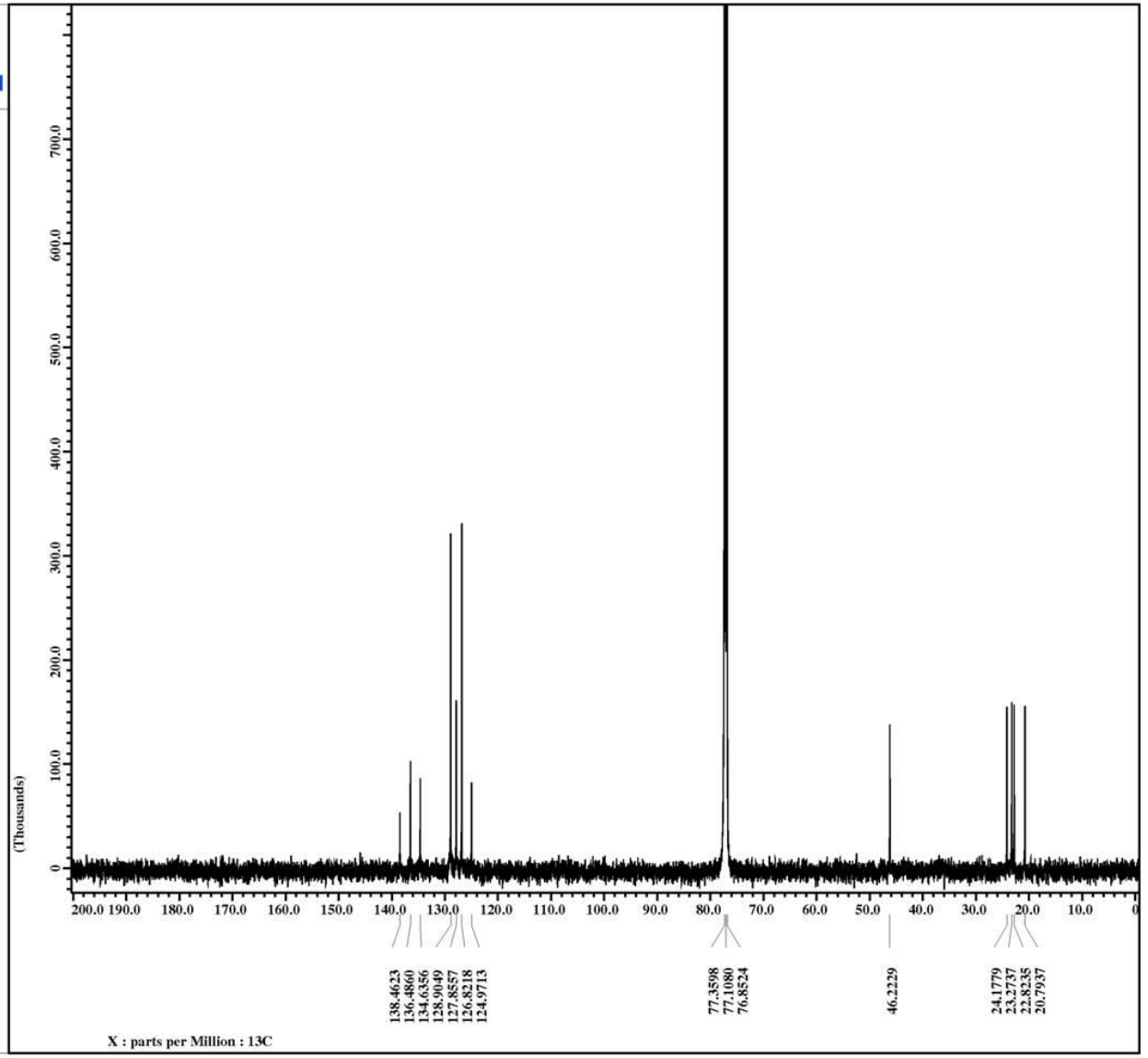






```
Filename          = II_135_AzidoBnTHB-4.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#813896
Solvent           = CHLOROFORM-D
Creation_time     =  4-JAN-2007 05:03:51
Revision_time     = 16-MAR-2010 22:30:50
Current_time      = 16-MAR-2010 22:31:37

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 10
Scans             = 3013
Total_scans       = 3013

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 25.1[dC]
Unblank_time      = 2[us]
```



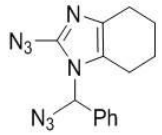


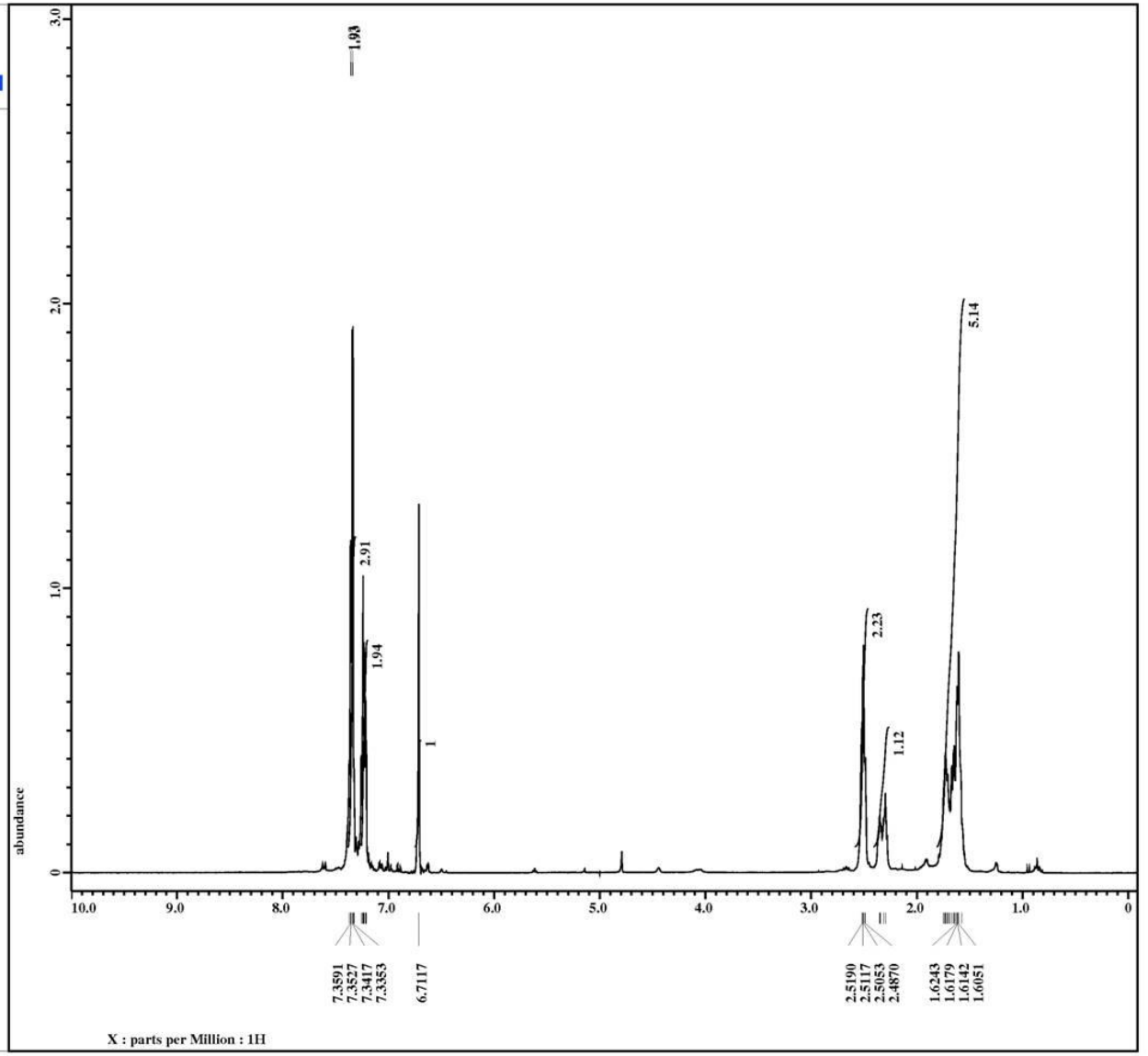
APPENDIX 9

^{1}H, ^{13}C and DEPT NMR Spectra of

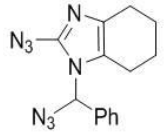
2-Azido-1-(azidophenylmethyl)-4,5,6,7-tetrahydro-1*H*-benzimidazole (**98**)

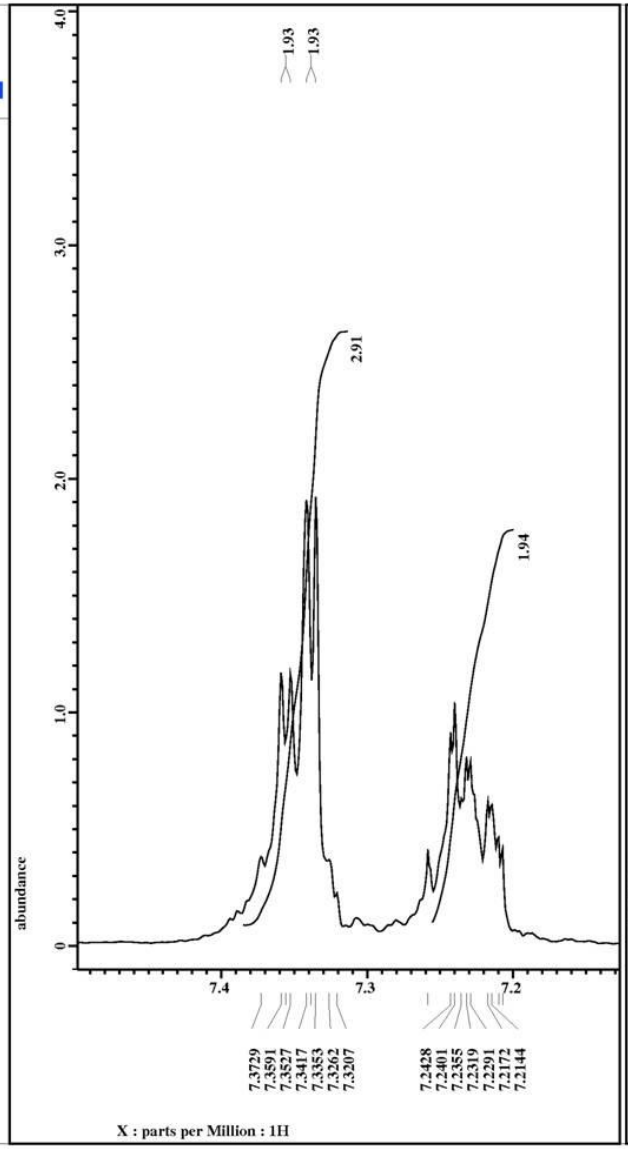
Filename	= II_p_125_I-2.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#796581
Solvent	= CHLOROFORM-D
Creation_time	= 11-DEC-2006 22:56:21
Revision_time	= 16-MAR-2010 22:40:30
Current_time	= 16-MAR-2010 22:40:47
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22[dC]

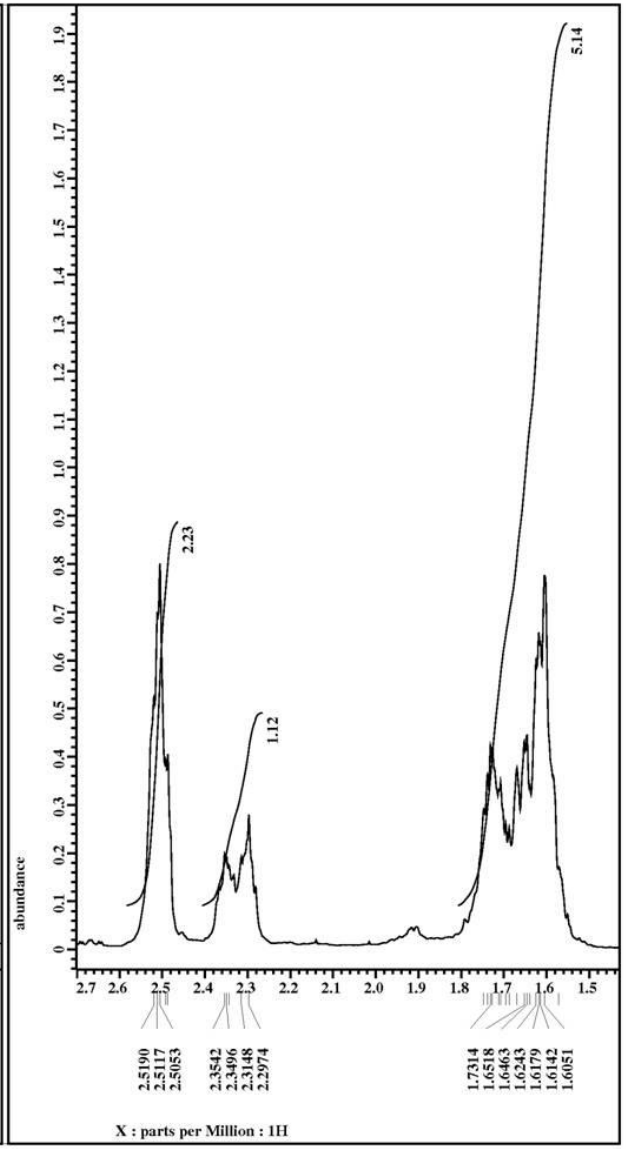




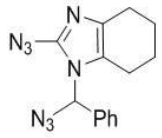
Filename	= II_p_125_I-2.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#796581
Solvent	= CHLOROFORM-D
Creation_time	= 11-DEC-2006 22:56:21
Revision_time	= 16-MAR-2010 22:40:30
Current_time	= 16-MAR-2010 22:41:21
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22[dC]

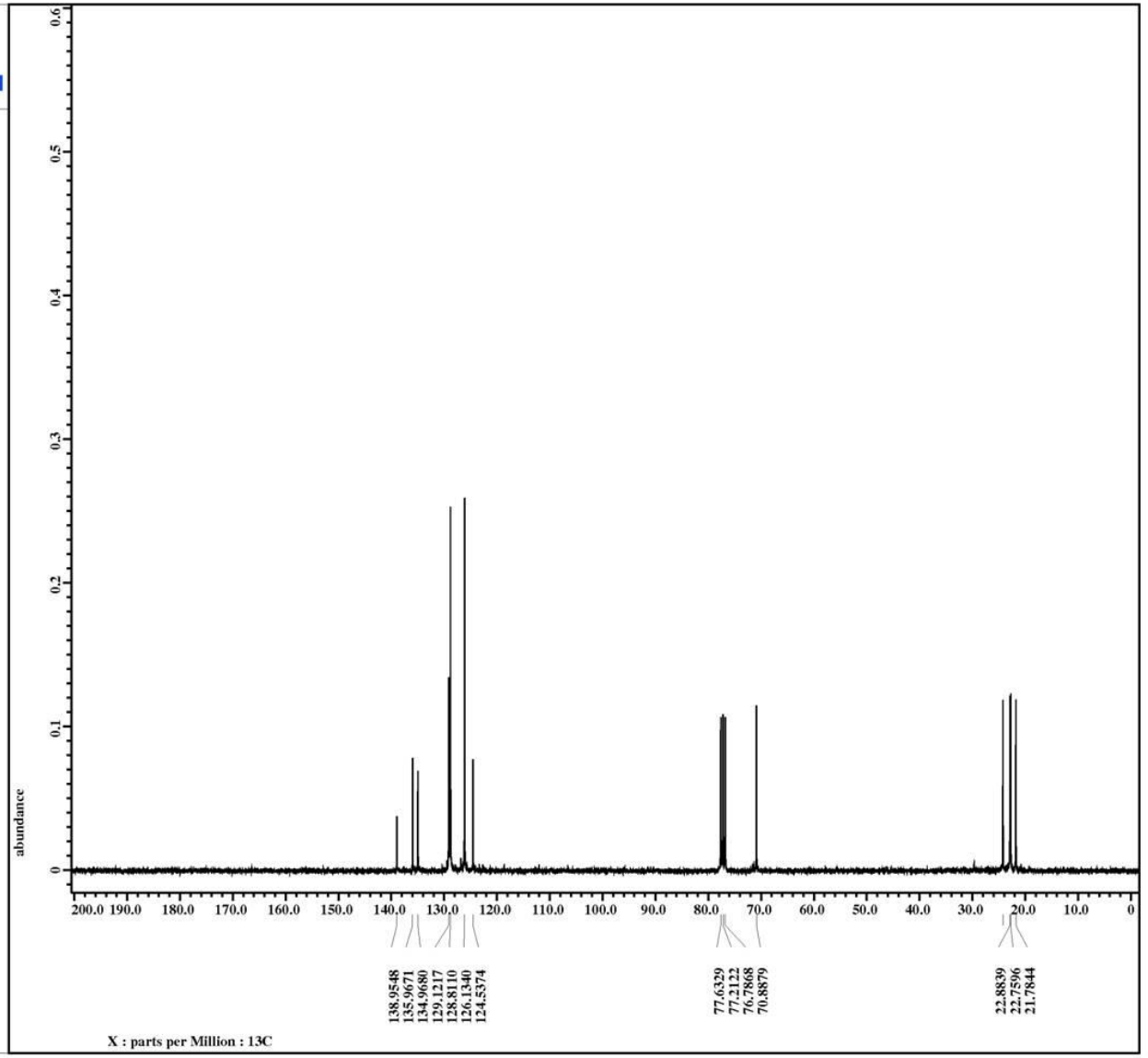




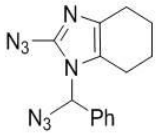


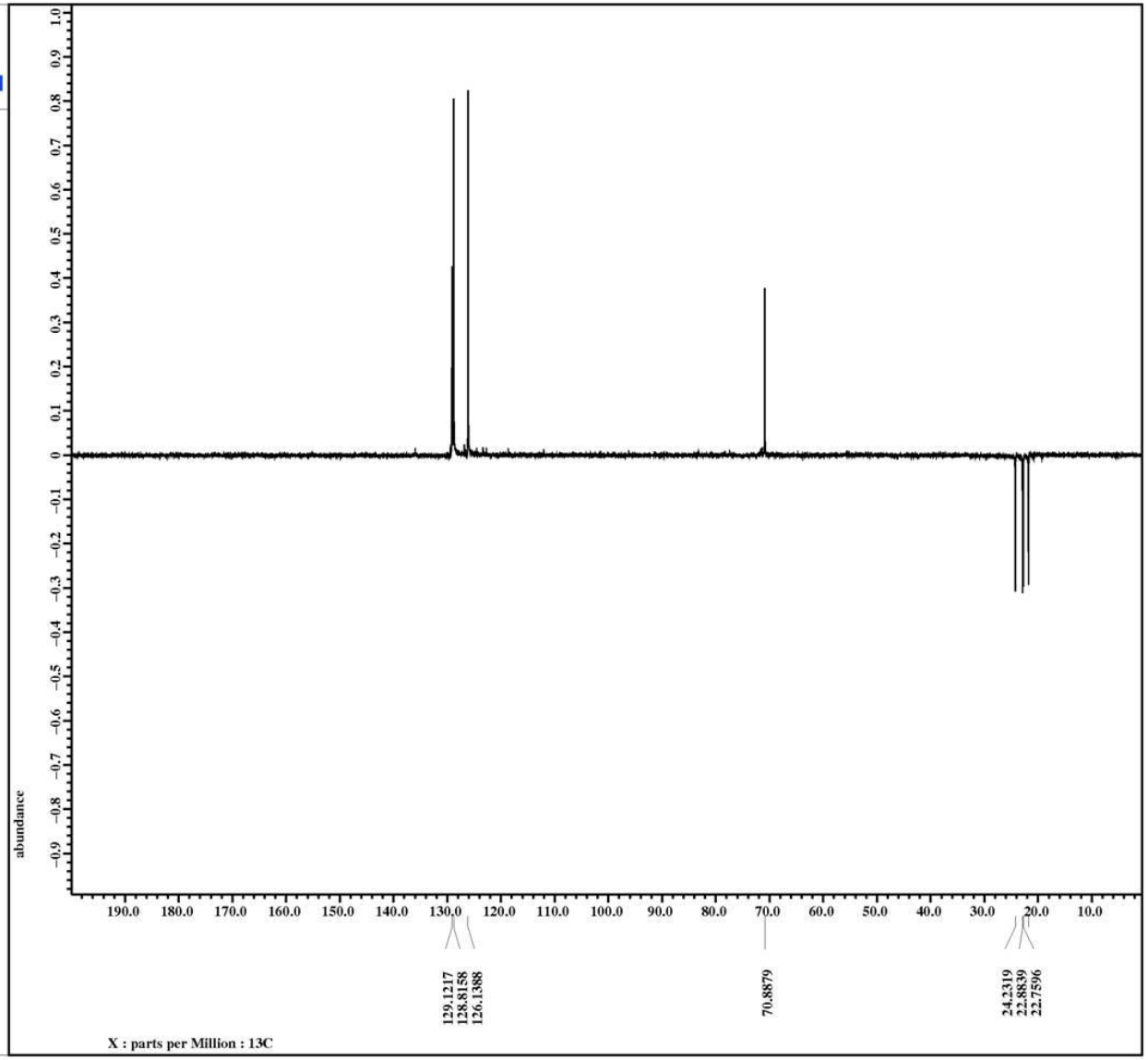
```
Filename          = II_p_125_I-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#798437
Solvent           = CHLOROFORM-D
Creation_time     = 12-DEC-2006 01:04:43
Revision_time     = 16-MAR-2010 22:42:08
Current_time      = 16-MAR-2010 22:42:31

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1599
Total_scans       = 1599

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.2[dC]
```





Filename	= II_p_125_I-4.jdf
Author	= delta
Experiment	= dept.ex2
Sample_id	= S#674226
Solvent	= CHLOROFORM-D
Creation_time	= 13-DEC-2006 20:03:16
Revision_time	= 16-MAR-2010 22:45:57
Current_time	= 16-MAR-2010 22:47:54
Comment	= DEPT with decoupling
Data_format	= 1D COMPLEX
Dim_size	= 26214
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 1.38412032[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 32768
X_prescans	= 4
X_resolution	= 0.72248054[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 574
Total_scans	= 574
X_acq_time	= 1.38412032[s]
X_atn	= 8[dB]
X_pulse	= 9.75[us]
Irr_atn	= 4[dB]
Irr_atn_dec	= 25[dB]
Irr_noise	= WALTZ
Irr_pulse	= 13.01[us]
Decoupling	= TRUE
Initial_wait	= 1[s]
J_constant	= 140[Hz]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Selection_angle	= 135[deg]
Selection_pulse	= 19.515[us]
Temp_get	= 22.2[dC]



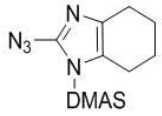


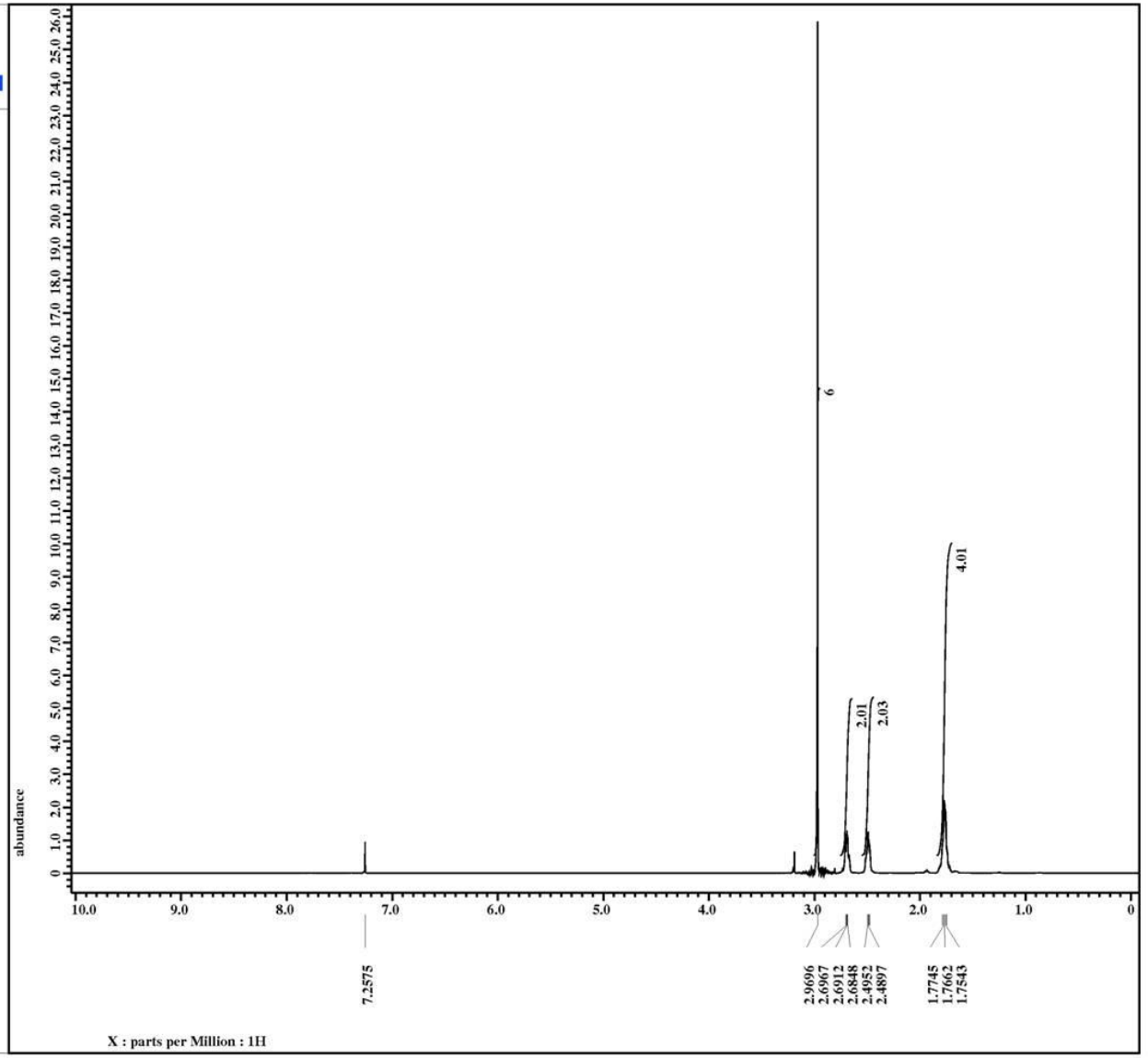
APPENDIX 10

^{1}H and ^{13}C NMR Spectra of

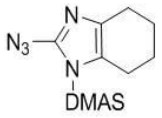
2-Azido-1-dimethylaminosulfonyl-4,5,6,7-tetrahydrobenzimidazole (**99**)

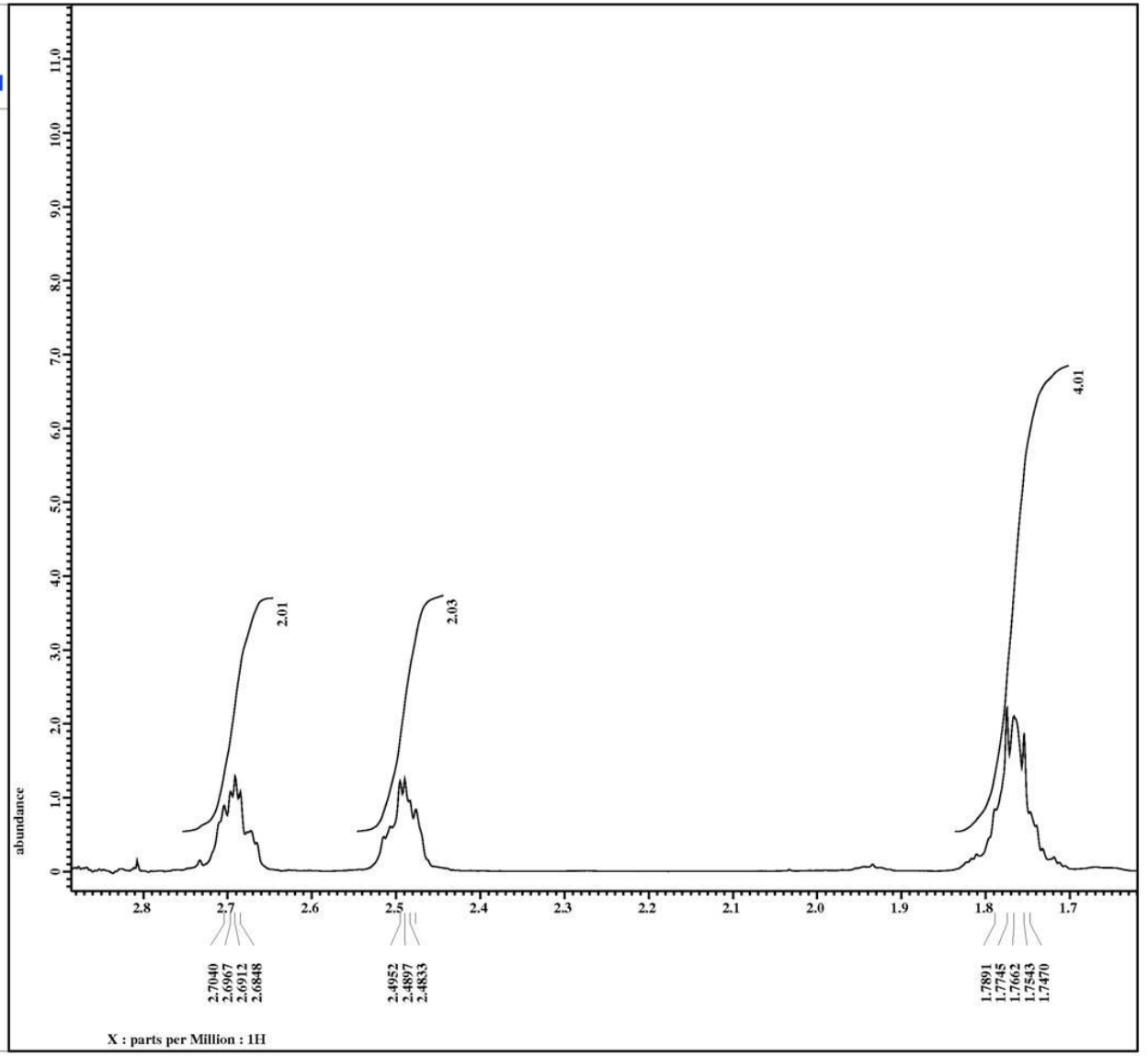
Filename	= II_p_2_azidoDMASTHB-3
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#390094
Solvent	= CHLOROFORM-D
Creation_time	= 11-JAN-2007 11:42:34
Revision_time	= 16-MAR-2010 23:14:45
Current_time	= 16-MAR-2010 23:15:00
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 44
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.1[dC]



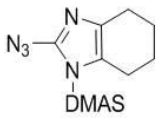


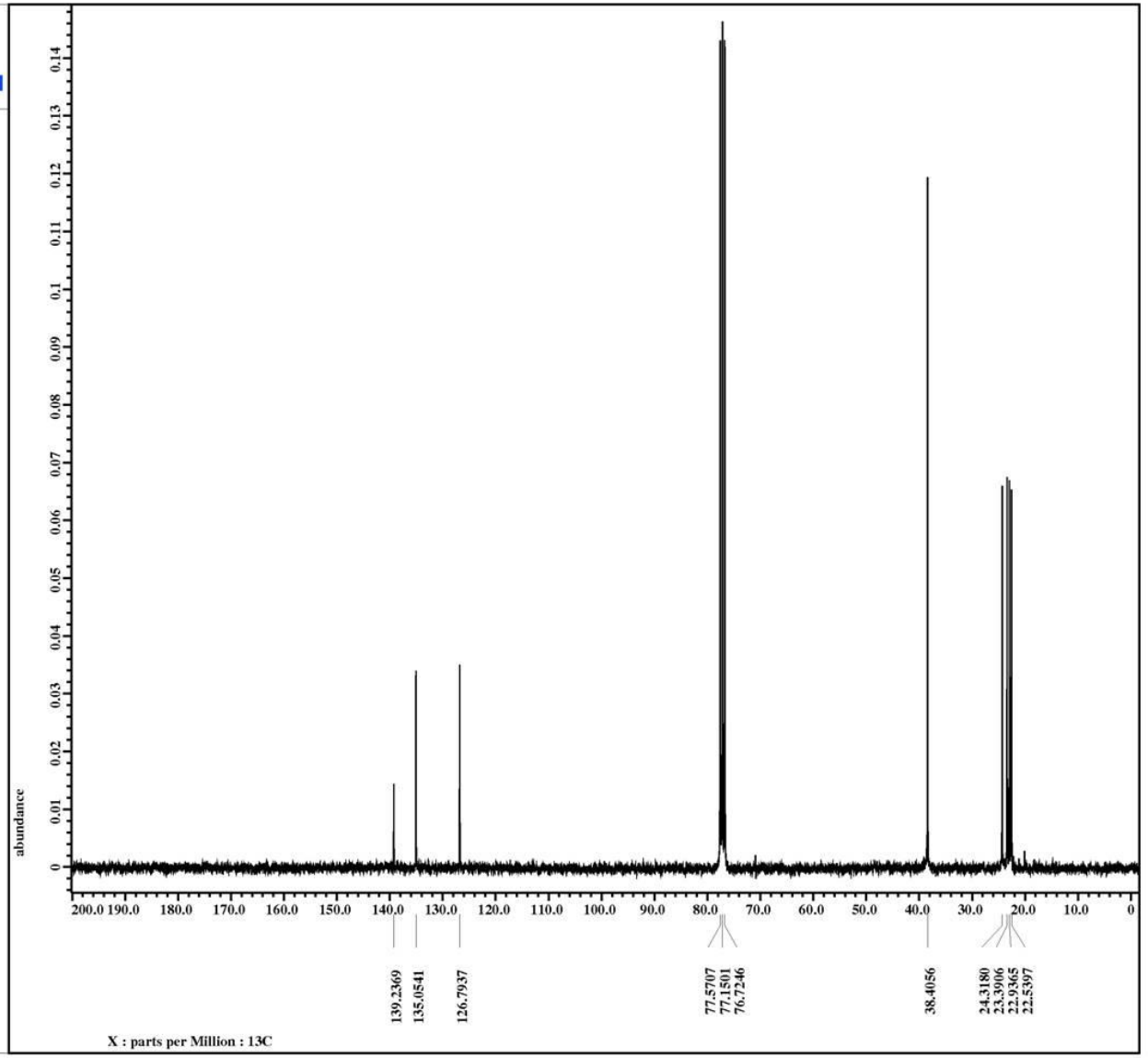
```
Filename          = II_p_2_azidoDMASTHB-3
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#390094
Solvent           = CHLOROFORM-D
Creation_time     = 11-JAN-2007 11:42:34
Revision_time     = 16-MAR-2010 23:14:45
Current_time      = 16-MAR-2010 23:15:24

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 44
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.1[dC]
```





```
Filename          = II_p_2-AzidoDMASTHB-3
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#755021
Solvent           = CHLOROFORM-D
Creation_time     = 11-JAN-2007 23:49:11
Revision_time     = 16-MAR-2010 23:16:10
Current_time      = 16-MAR-2010 23:16:20

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1516
Total_scans       = 1516

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.3[dC]
```



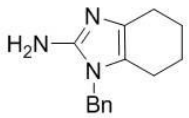


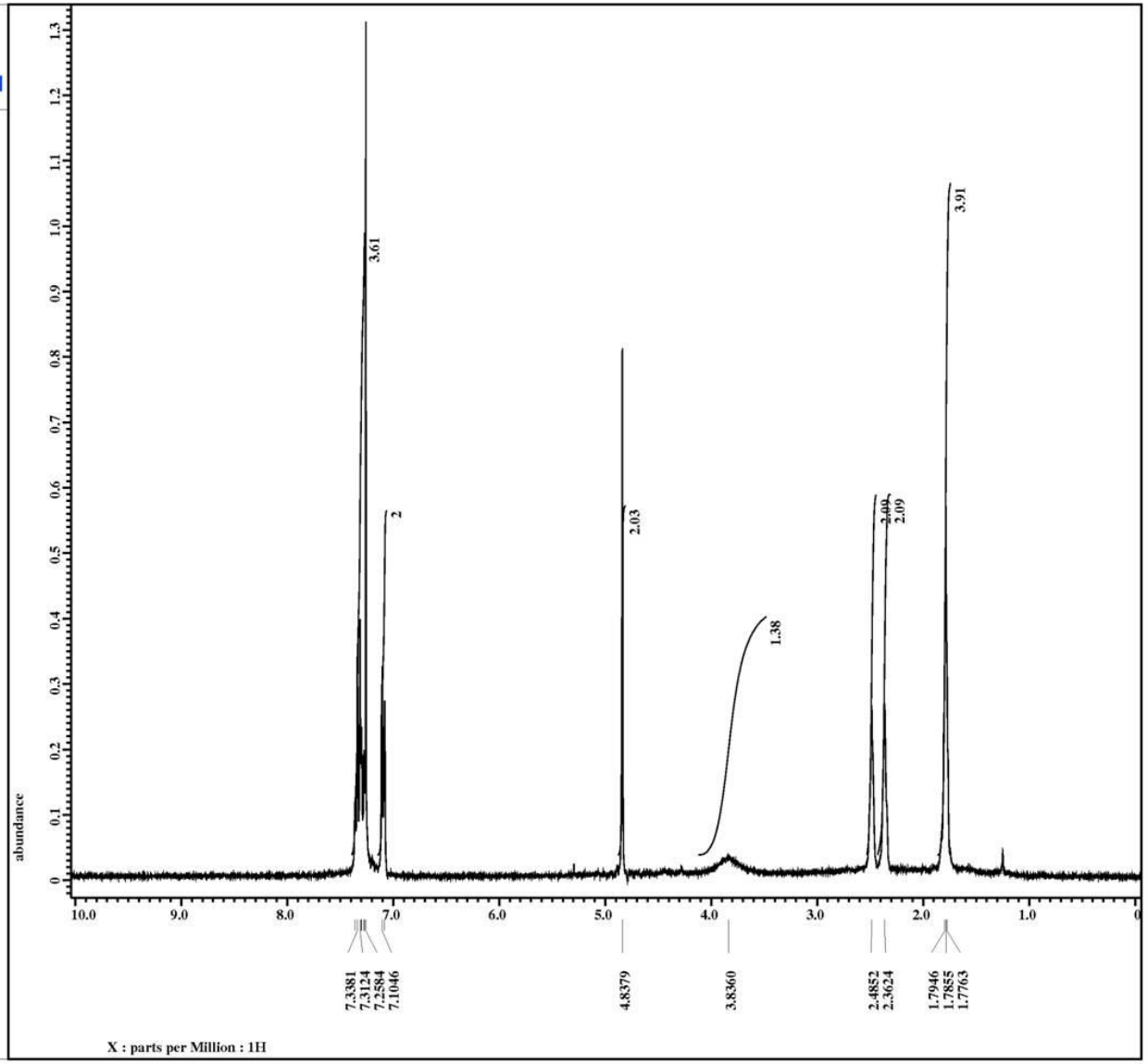
APPENDIX 11

^{1}H and ^{13}C NMR Spectra of

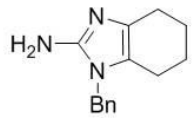
2-Amino-1-benzyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (**100**)

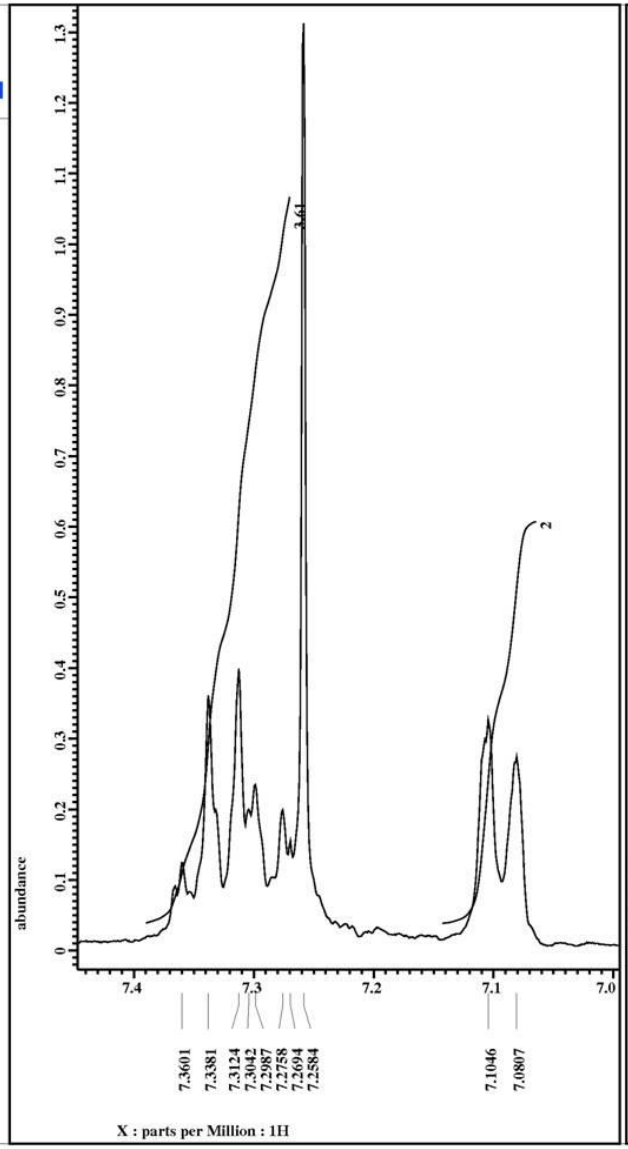
```
Filename          = II_p_unkn-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#573355
Solvent           = CHLOROFORM-D
Creation_time     =  3-APR-2008 15:09:22
Revision_time     = 17-MAR-2010 15:44:45
Current_time      = 17-MAR-2010 15:45:05

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 21.7[dC]
```

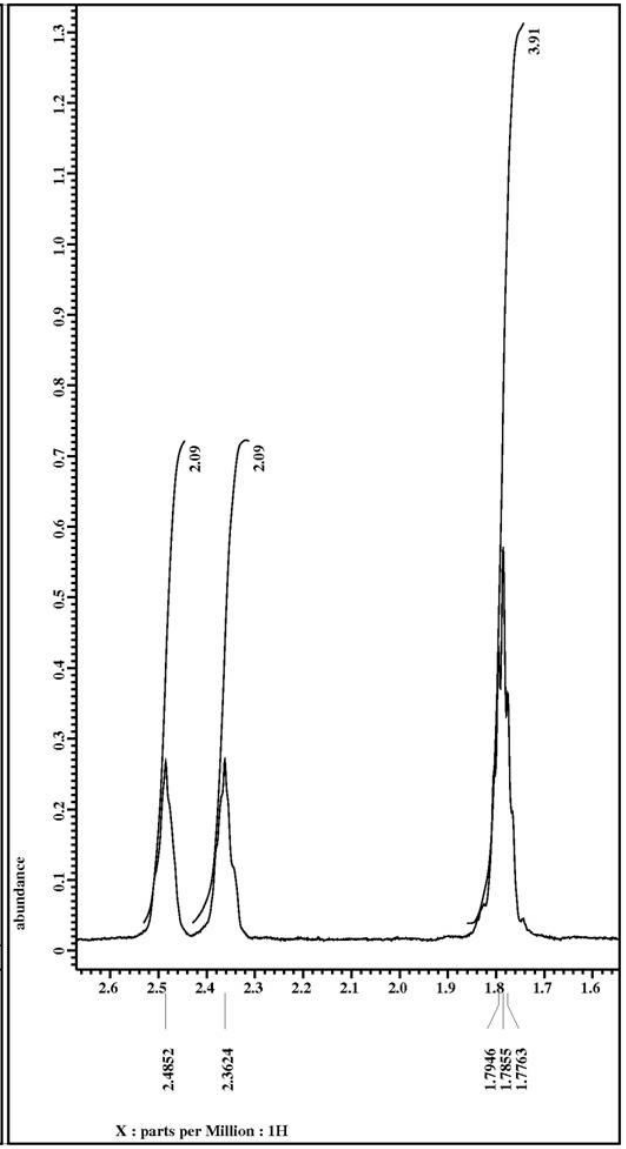




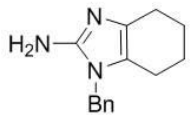
Filename	= II_p_unkn-2.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#573355
Solvent	= CHLOROFORM-D
Creation_time	= 3-APR-2008 15:09:22
Revision_time	= 17-MAR-2010 15:44:45
Current_time	= 17-MAR-2010 15:45:52
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 21.7[dC]

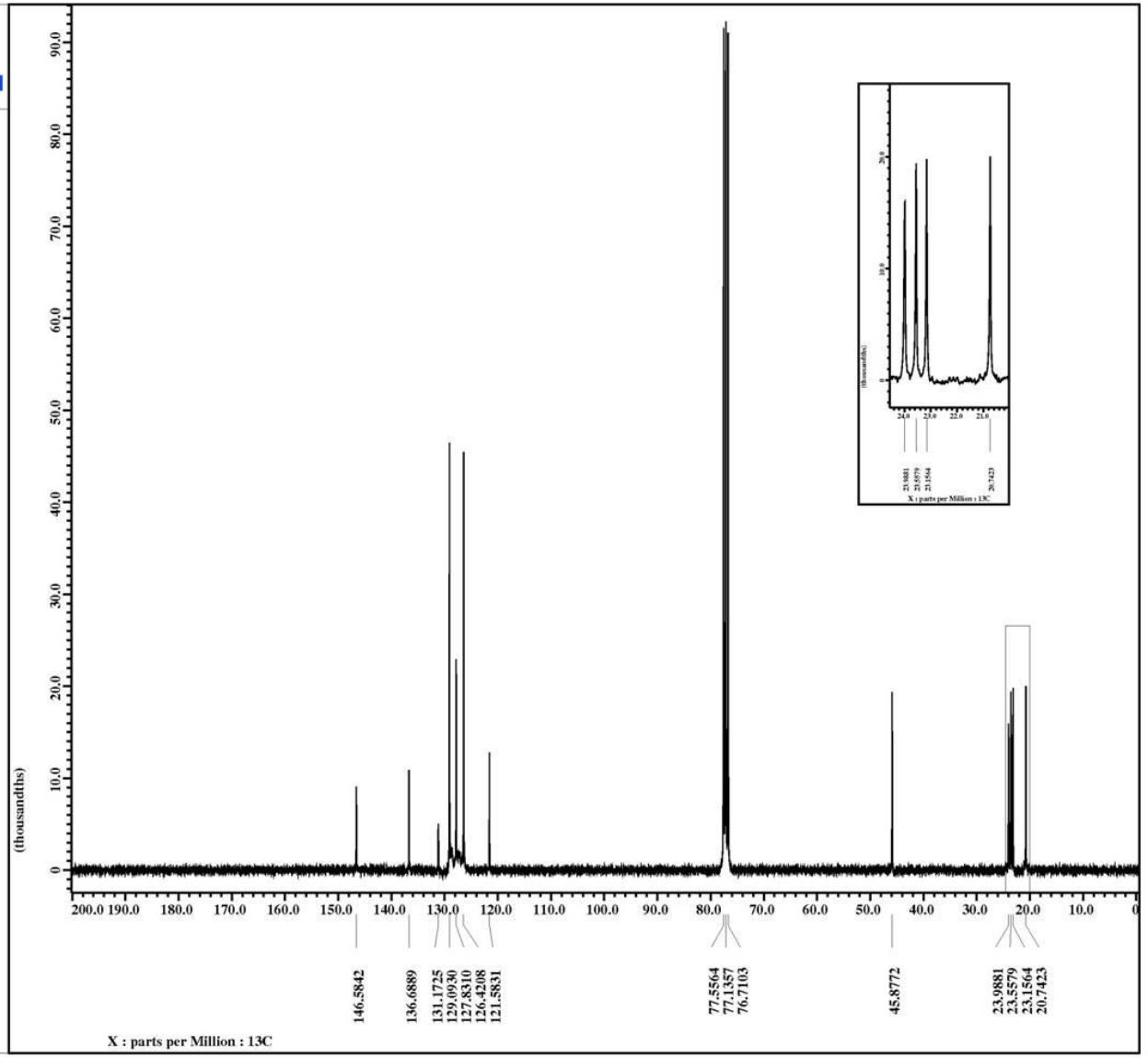






Filename	= II_p_072_AminoBnTHB-3
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#805699
Solvent	= CHLOROFORM-D
Creation_time	= 22-SEP-2006 10:20:32
Revision_time	= 22-SEP-2006 09:40:21
Current_time	= 17-MAR-2010 15:47:40
Comment	= single pulse decouple
Data_format	= 1D REAL
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 8500
Total_scans	= 8500
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 23.4[dC]



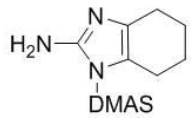


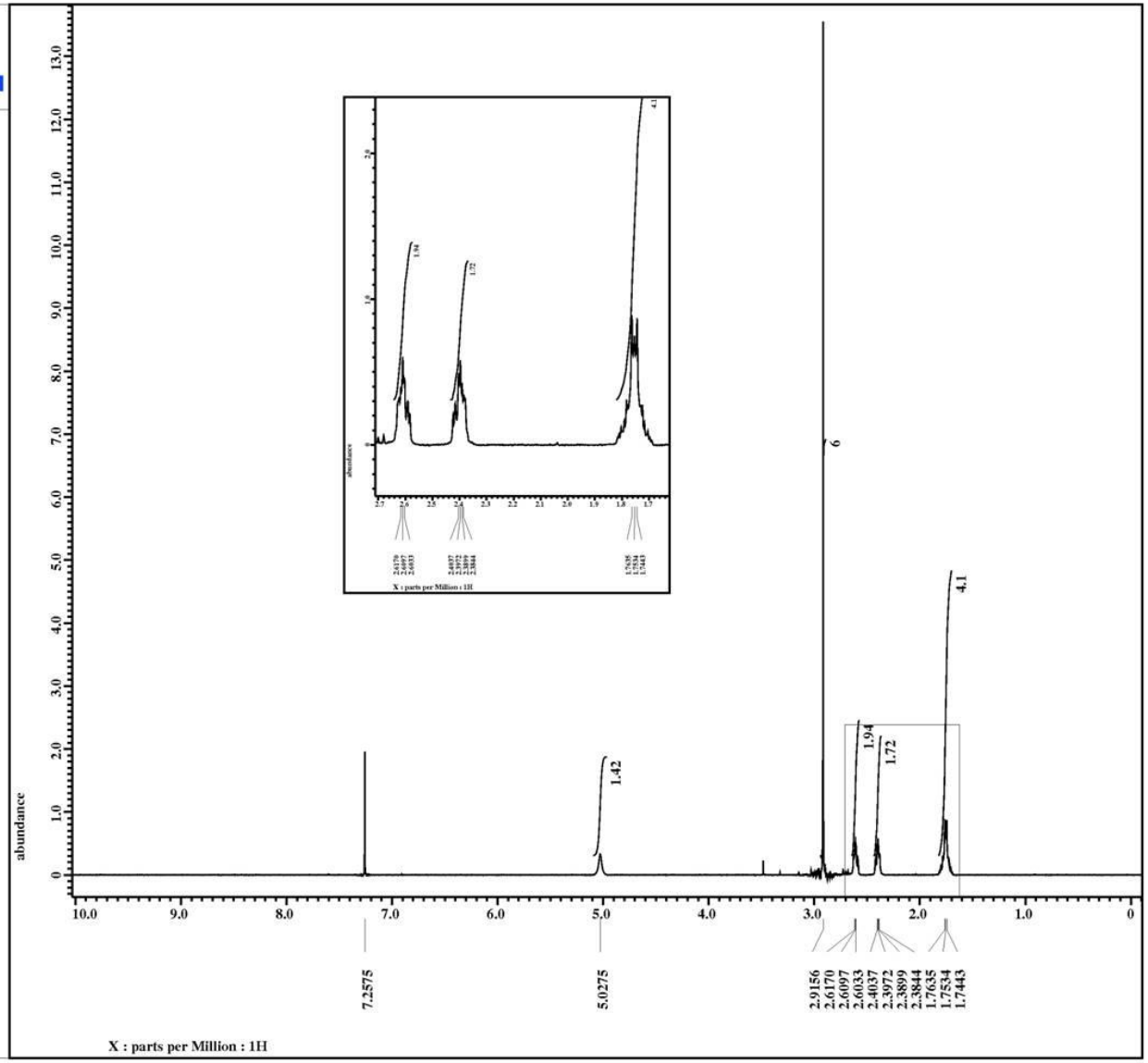
APPENDIX 12

^{1}H and ^{13}C NMR Spectra of

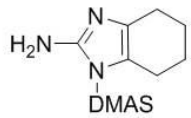
2-Amino-1-dimethylaminosulfonyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (**101**)

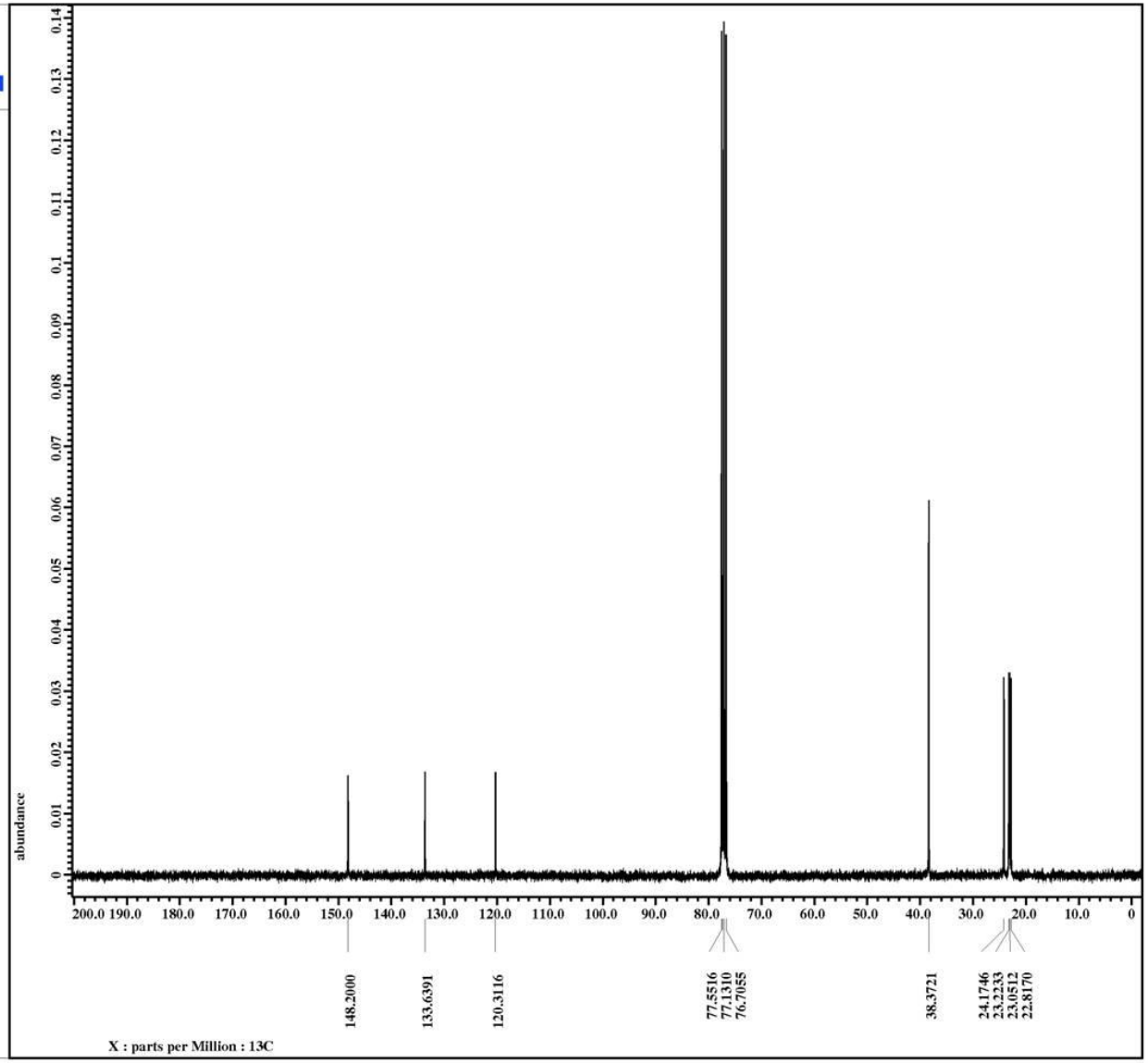
```
Filename          = p_31_2_amino_1_THB-4.
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#545668
Solvent           = CHLOROFORM-D
Creation_time     = 27-OCT-2005 16:05:32
Revision_time     = 17-MAR-2010 16:12:01
Current_time      = 17-MAR-2010 16:12:57

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.6[dC]
```





```
Filename          = p_21_product-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#812168
Solvent           = CHLOROFORM-D
Creation_time     = 20-SEP-2006 09:47:44
Revision_time     = 17-MAR-2010 16:14:30
Current_time      = 17-MAR-2010 16:14:43

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7940
Total_scans       = 7940

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.6[dC]
```



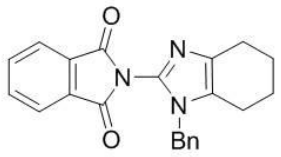


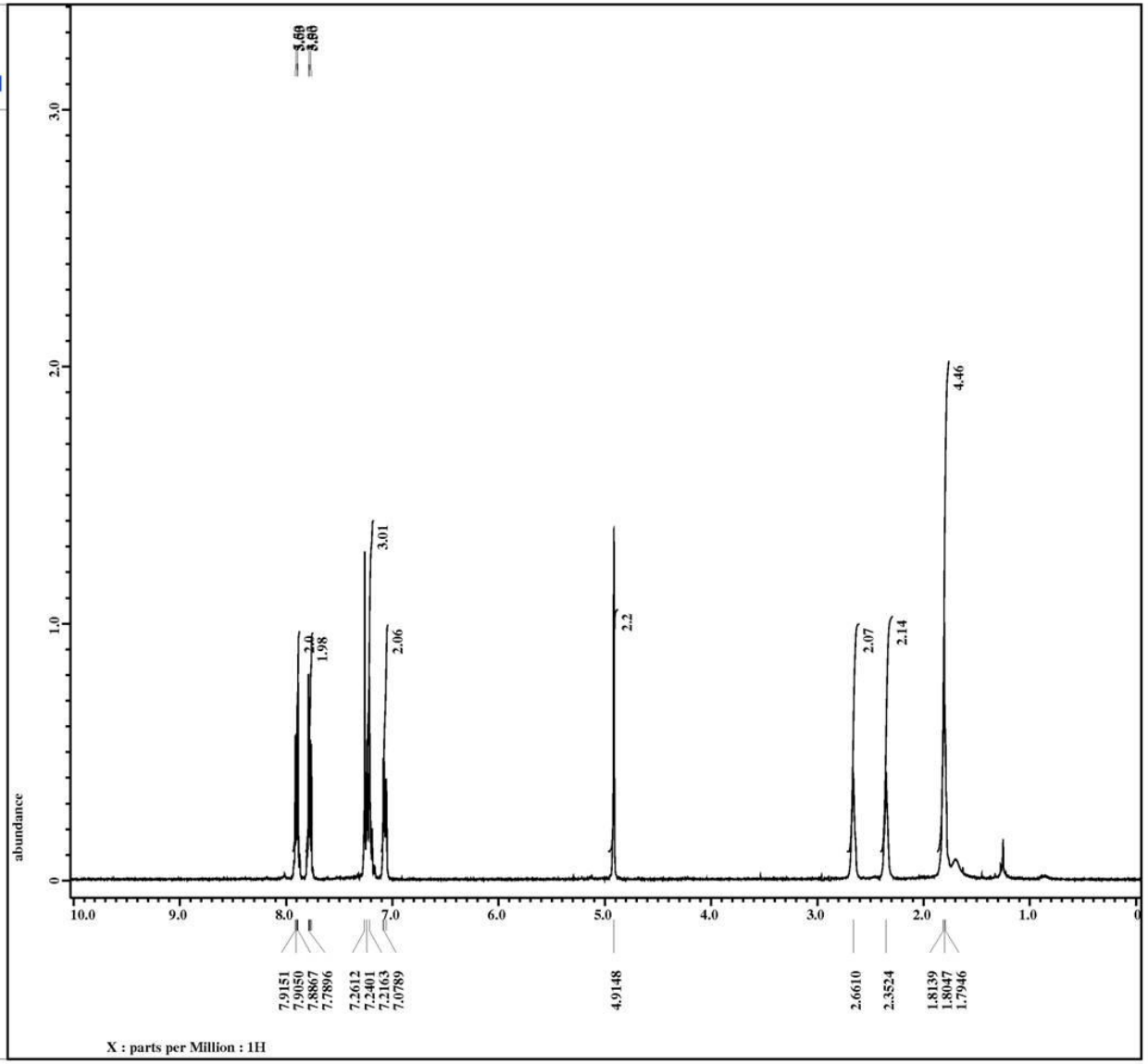
APPENDIX 13

^{1}H and ^{13}C NMR Spectra of

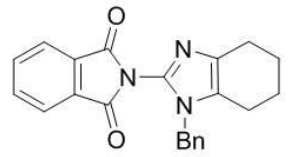
1-Benzyl-2-phthalimidoyl-4,5,6,7-tetrahydro-1*H*-benzimidazole (**103**)

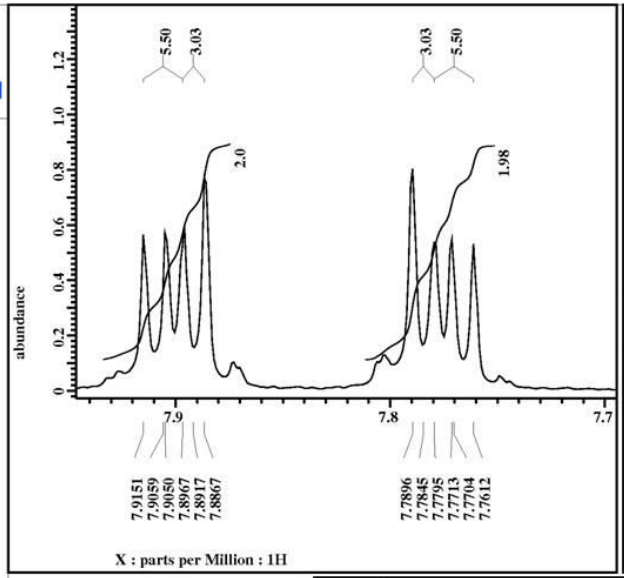
```
Filename          = II_p_074_I-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#472072
Solvent           = CHLOROFORM-D
Creation_time     =  4-SEP-2006 13:48:57
Revision_time     = 17-MAR-2010 16:35:15
Current_time      = 17-MAR-2010 16:35:30

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

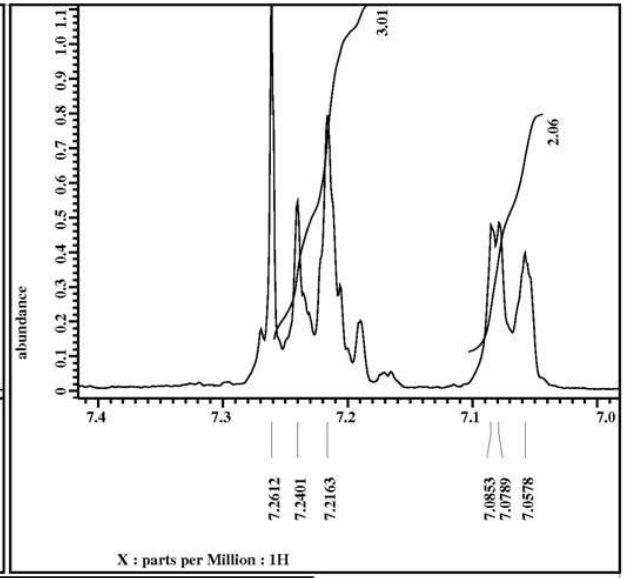


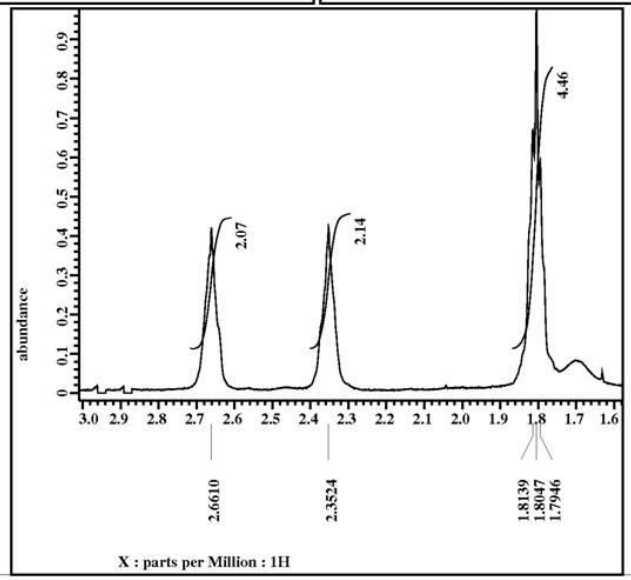


```
Filename          = II_p_074_I-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#472072
Solvent           = CHLOROFORM-D
Creation_time     =  4-SEP-2006 13:48:57
Revision_time     = 17-MAR-2010 16:35:15
Current_time      = 17-MAR-2010 16:36:23

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

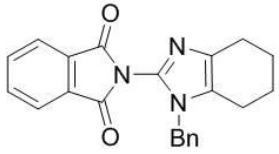


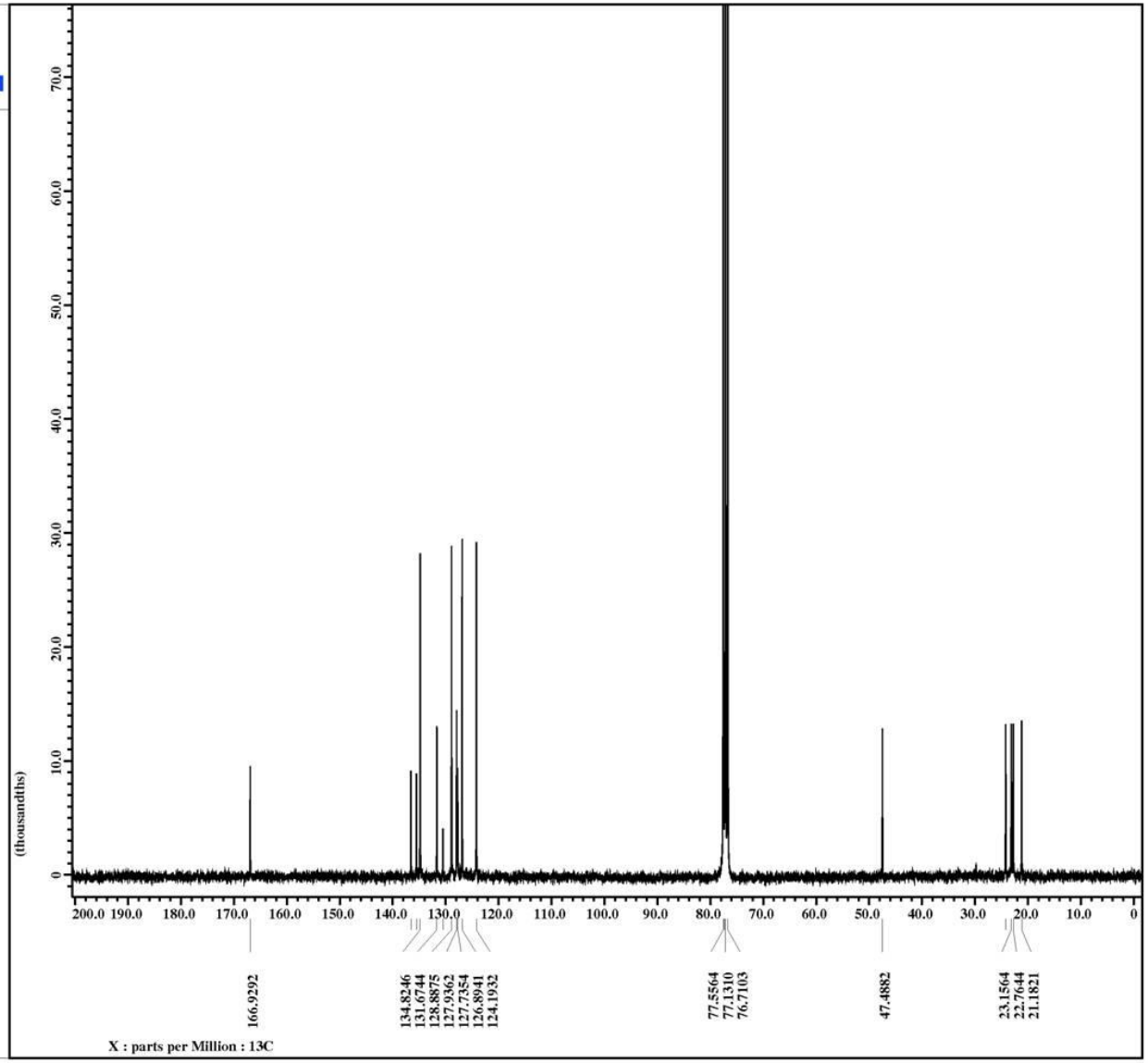




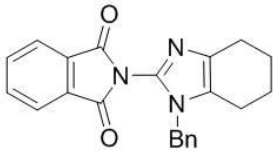


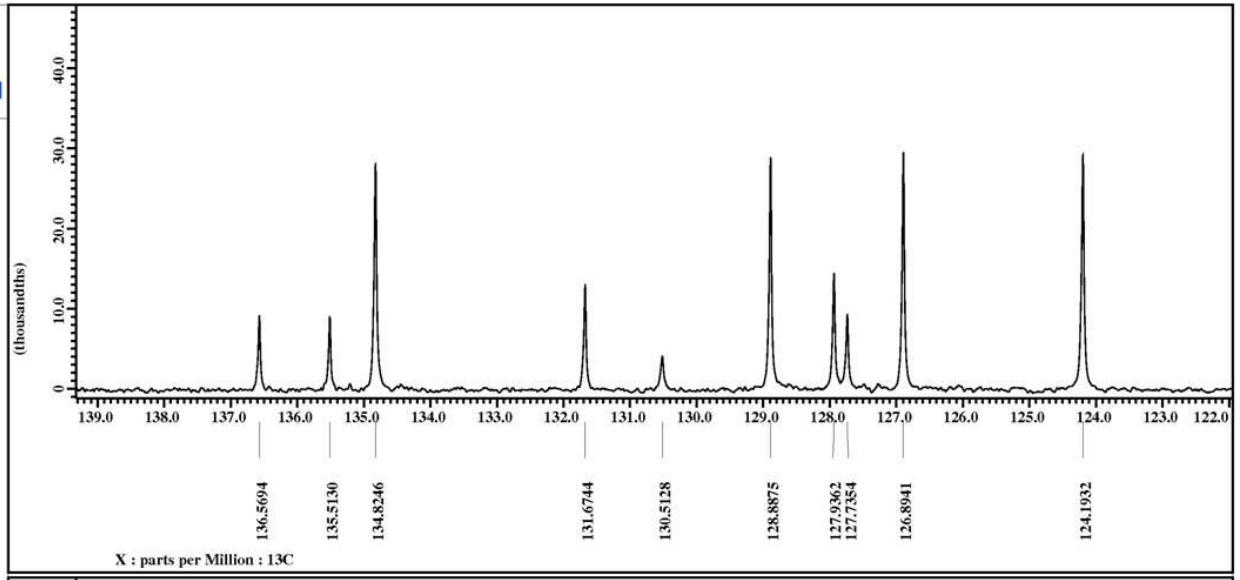
Filename	= II_p_074-3.jdf
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#782553
Solvent	= CHLOROFORM-D
Creation_time	= 28-SEP-2006 09:42:37
Revision_time	= 17-MAR-2010 16:21:15
Current_time	= 17-MAR-2010 16:21:28
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 8500
Total_scans	= 8500
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 23.6[dC]

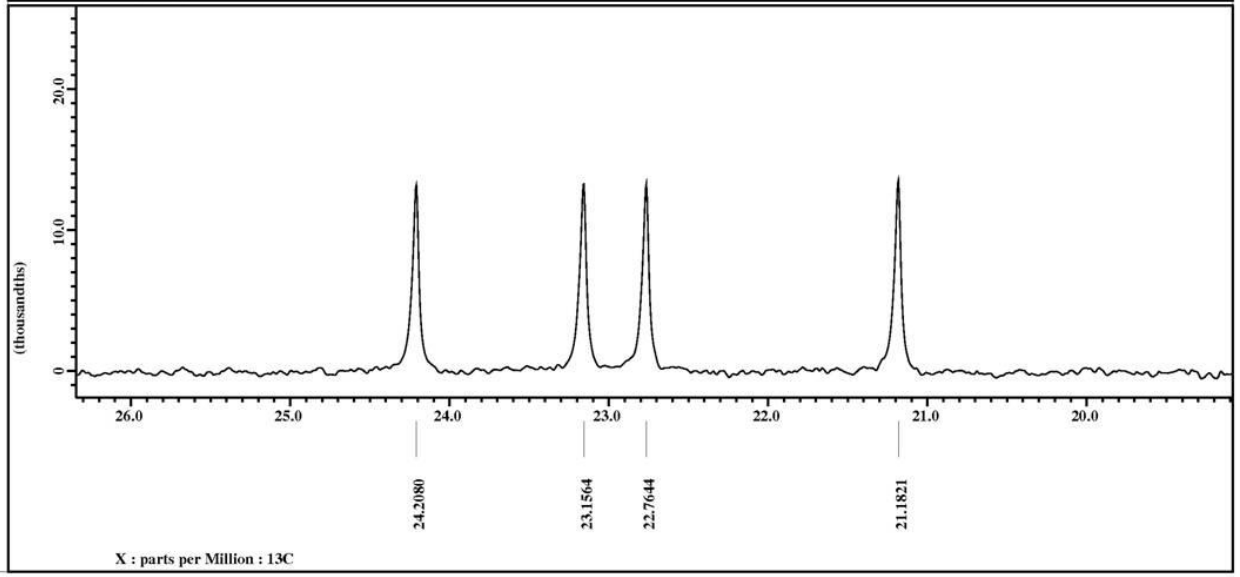




```
Filename          = II_p_074-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#782553
Solvent           = CHLOROFORM-D
Creation_time     = 28-SEP-2006 09:42:37
Revision_time     = 17-MAR-2010 16:21:15
Current_time      = 17-MAR-2010 16:22:10

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8500
Total_scans       = 8500

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.6[dC]
```







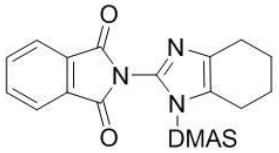
APPENDIX 14

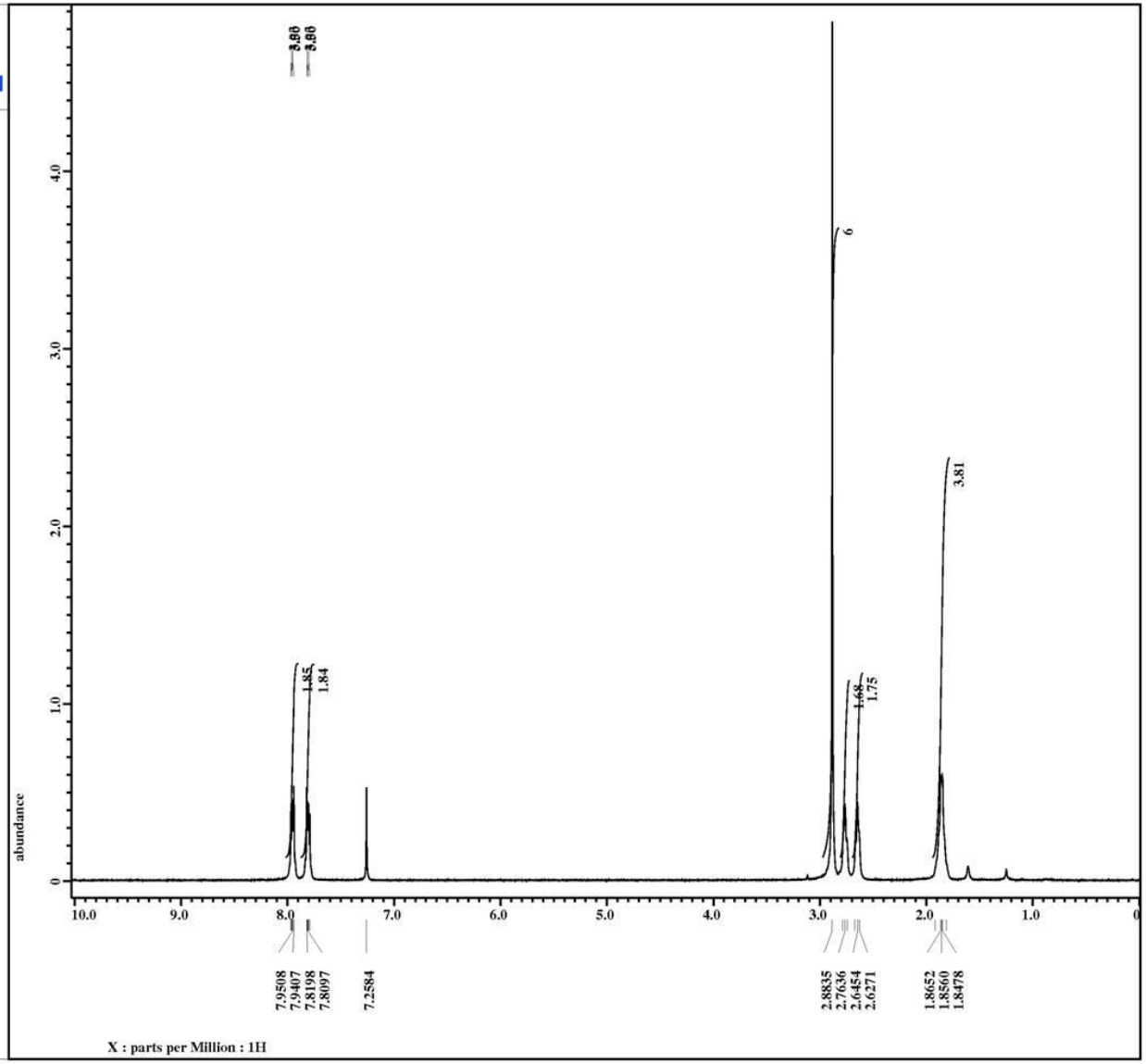
^{1}H and ^{13}C NMR Spectra of

1-Dimethylaminosulfonyl-2-phthalimidoyl 4,5,6,-7-tetrahydro-1*H*-benzlimidazole

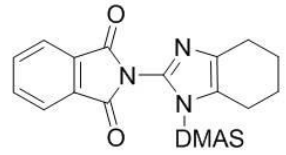
(**104**)

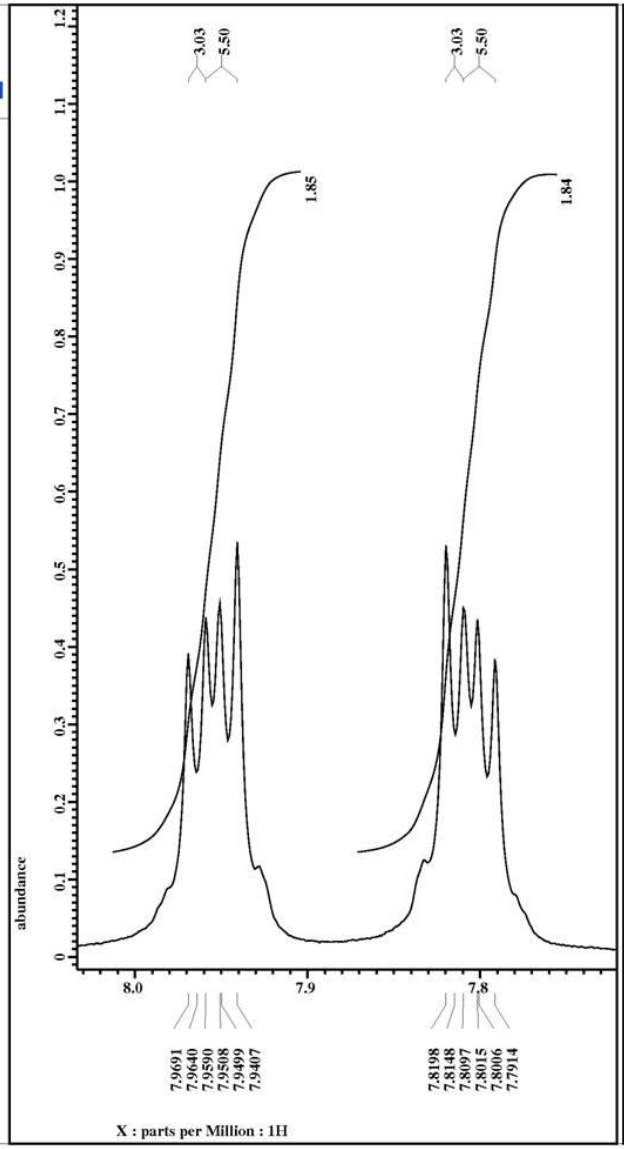
```
Filename          = p_38_nefkens-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#805304
Solvent           = CHLOROFORM-D
Creation_time     = 21-NOV-2005 23:21:16
Revision_time     = 17-MAR-2010 17:32:45
Current_time      = 17-MAR-2010 17:32:57

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

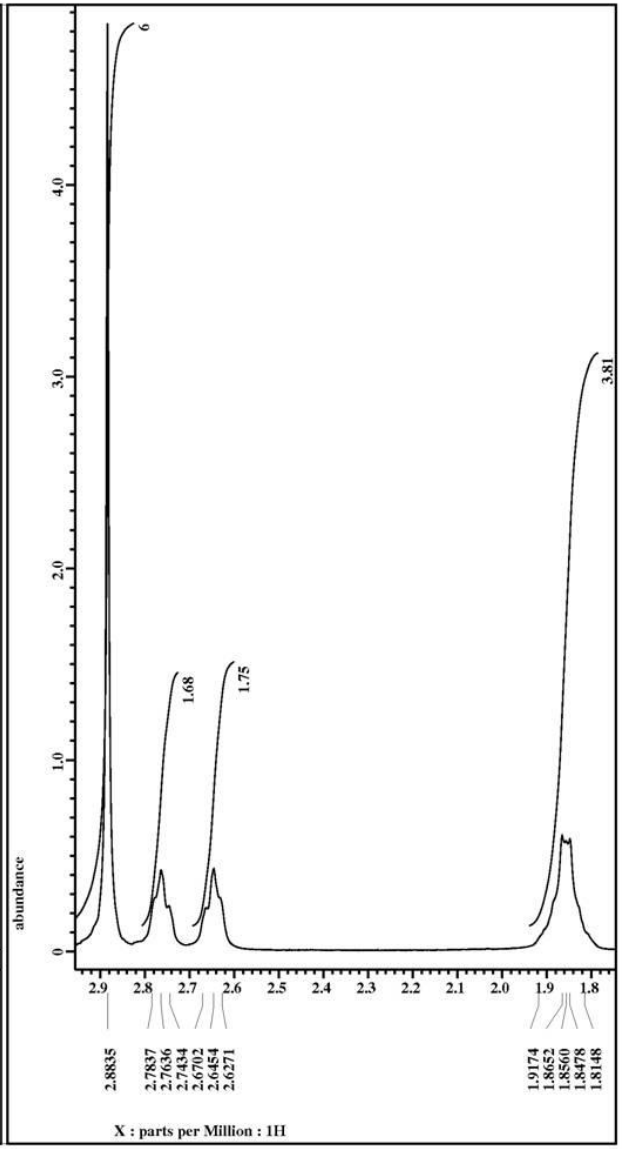




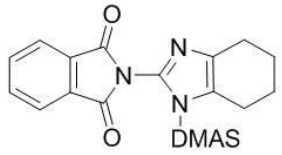
Filename	= p_38_nefkens-4.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#805304
Solvent	= CHLOROFORM-D
Creation_time	= 21-NOV-2005 23:21:16
Revision_time	= 17-MAR-2010 17:32:45
Current_time	= 17-MAR-2010 17:33:30
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.9[dC]

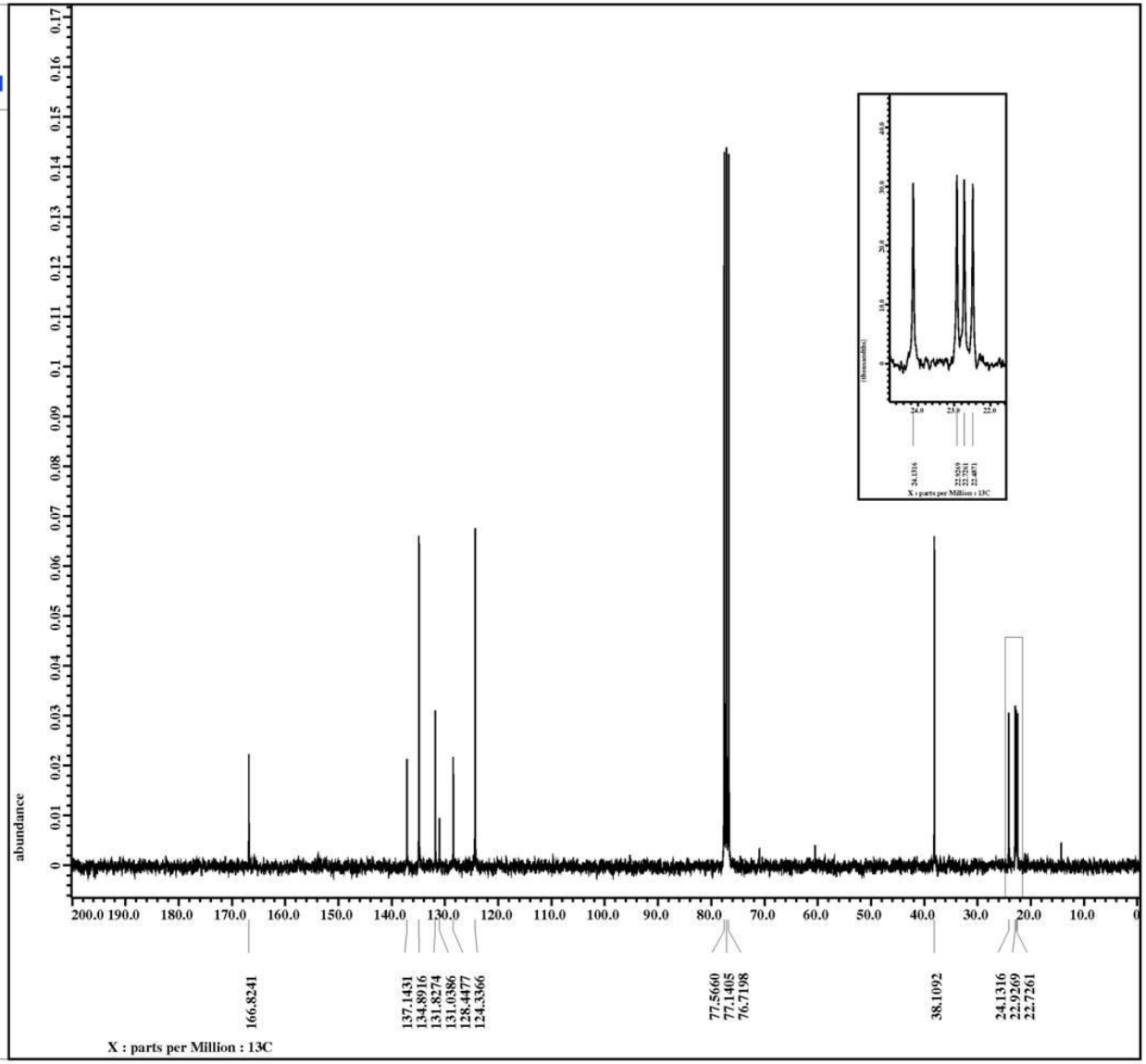






Filename	= p_38-5.jdf
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#640385
Solvent	= CHLOROFORM-D
Creation_time	= 15-NOV-2005 19:21:14
Revision_time	= 17-MAR-2010 17:34:11
Current_time	= 17-MAR-2010 17:35:03
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 470
Total_scans	= 470
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 23[dC]



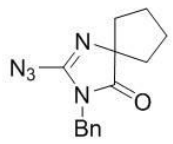


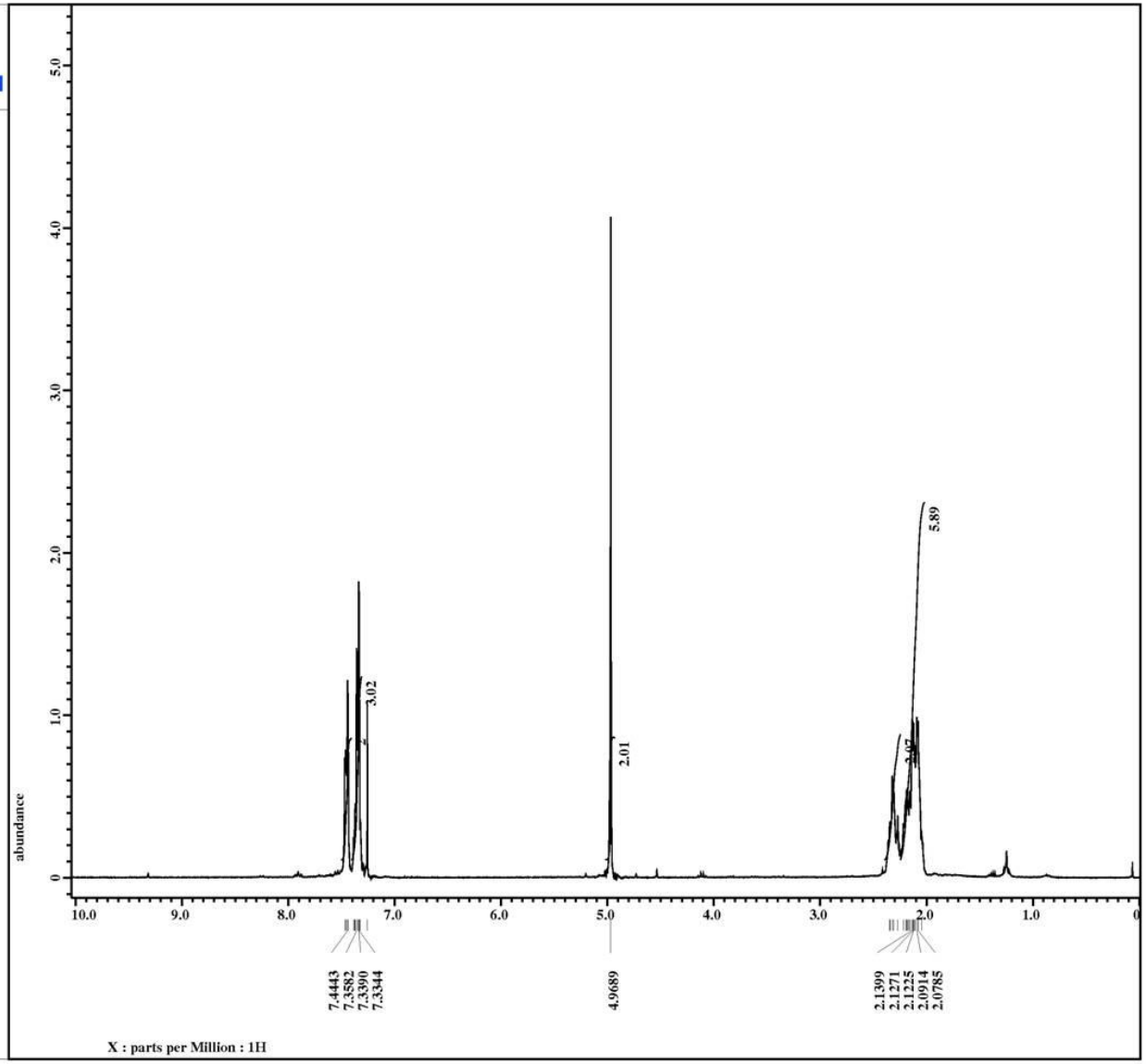
APPENDIX 15

^{1}H and ^{13}C NMR Spectra of

2-Azido-3-benzyl-1,3-diazaspiro[4.4]non-2-ene-4-one (**105**)

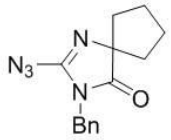
```
Filename          = II_p_140-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#387406
Solvent           = CHLOROFORM-D
Creation_time     = 11-JAN-2007 11:38:06
Revision_time     = 17-MAR-2010 17:45:28
Current_time      = 17-MAR-2010 17:45:51

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 42
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 21.8[dC]
```

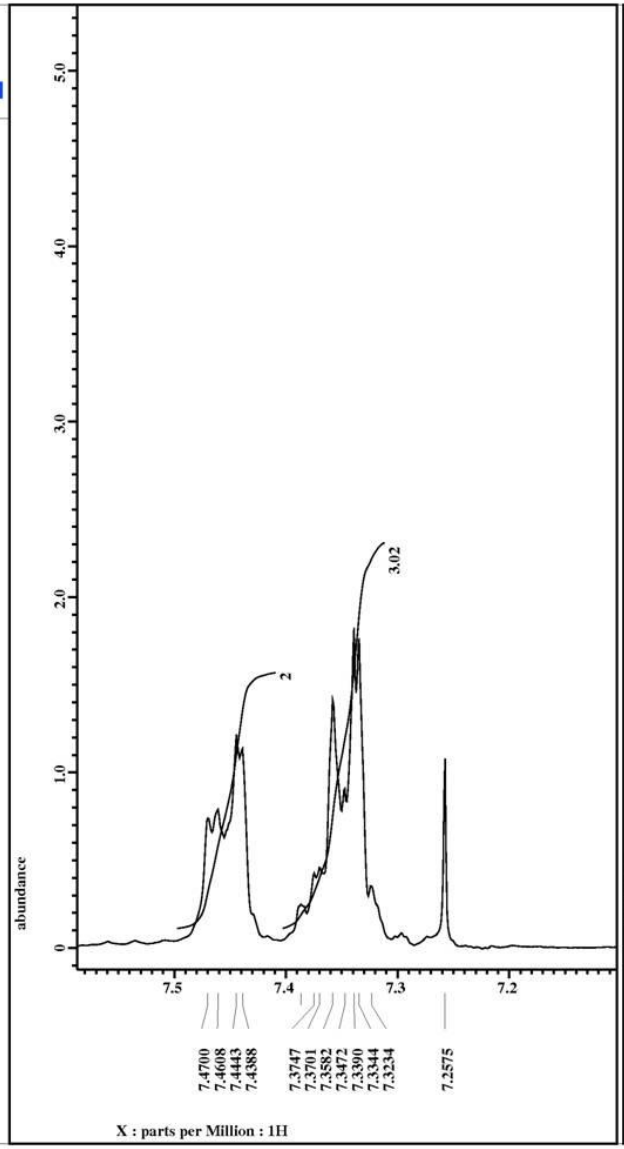


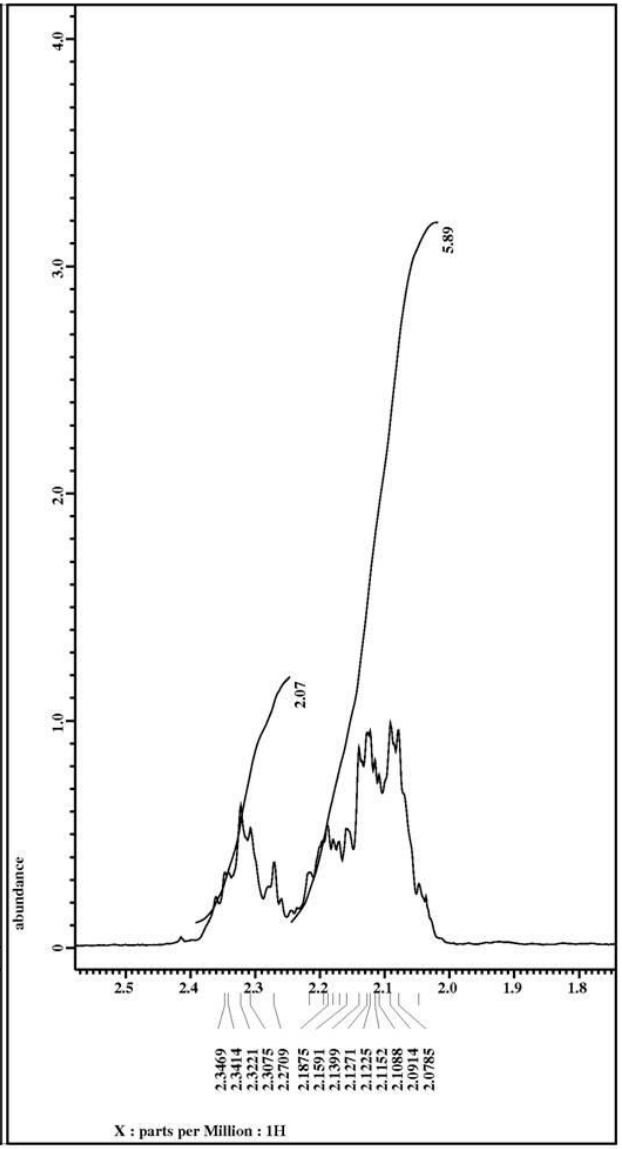


JEOL

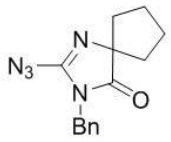
```
Filename          = II_p_140-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#387406
Solvent           = CHLOROFORM-D
Creation_time     = 11-JAN-2007 11:38:06
Revision_time     = 17-MAR-2010 17:45:28
Current_time      = 17-MAR-2010 17:46:42

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 42
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 21.8[dC]
```

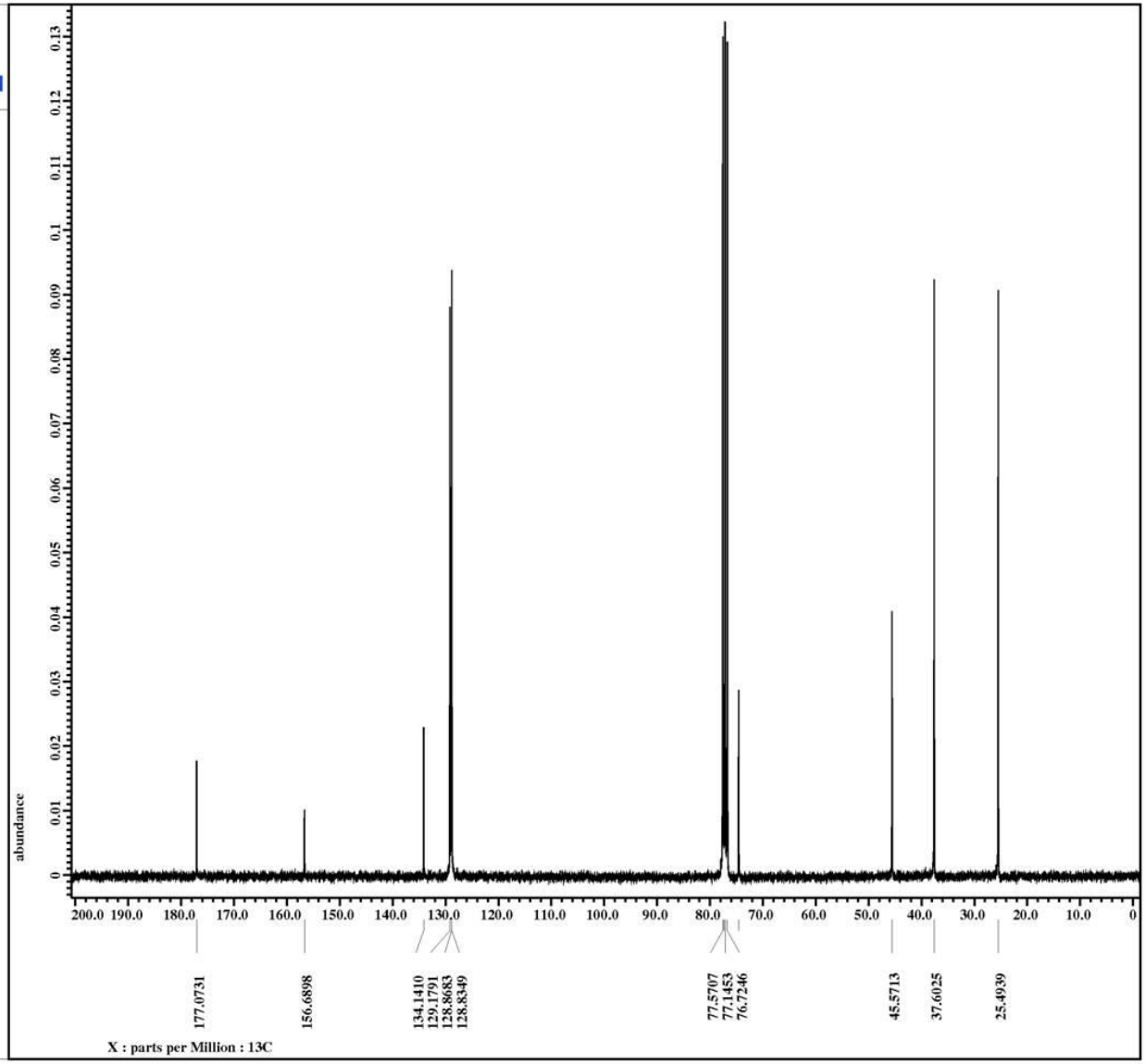






```
Filename          = II_p_140-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#833564
Solvent           = CHLOROFORM-D
Creation_time     = 12-JAN-2007 09:43:43
Revision_time     = 17-MAR-2010 17:43:44
Current_time      = 17-MAR-2010 17:44:05

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7350
Total_scans       = 7350

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.4[dC]
```



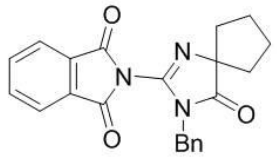


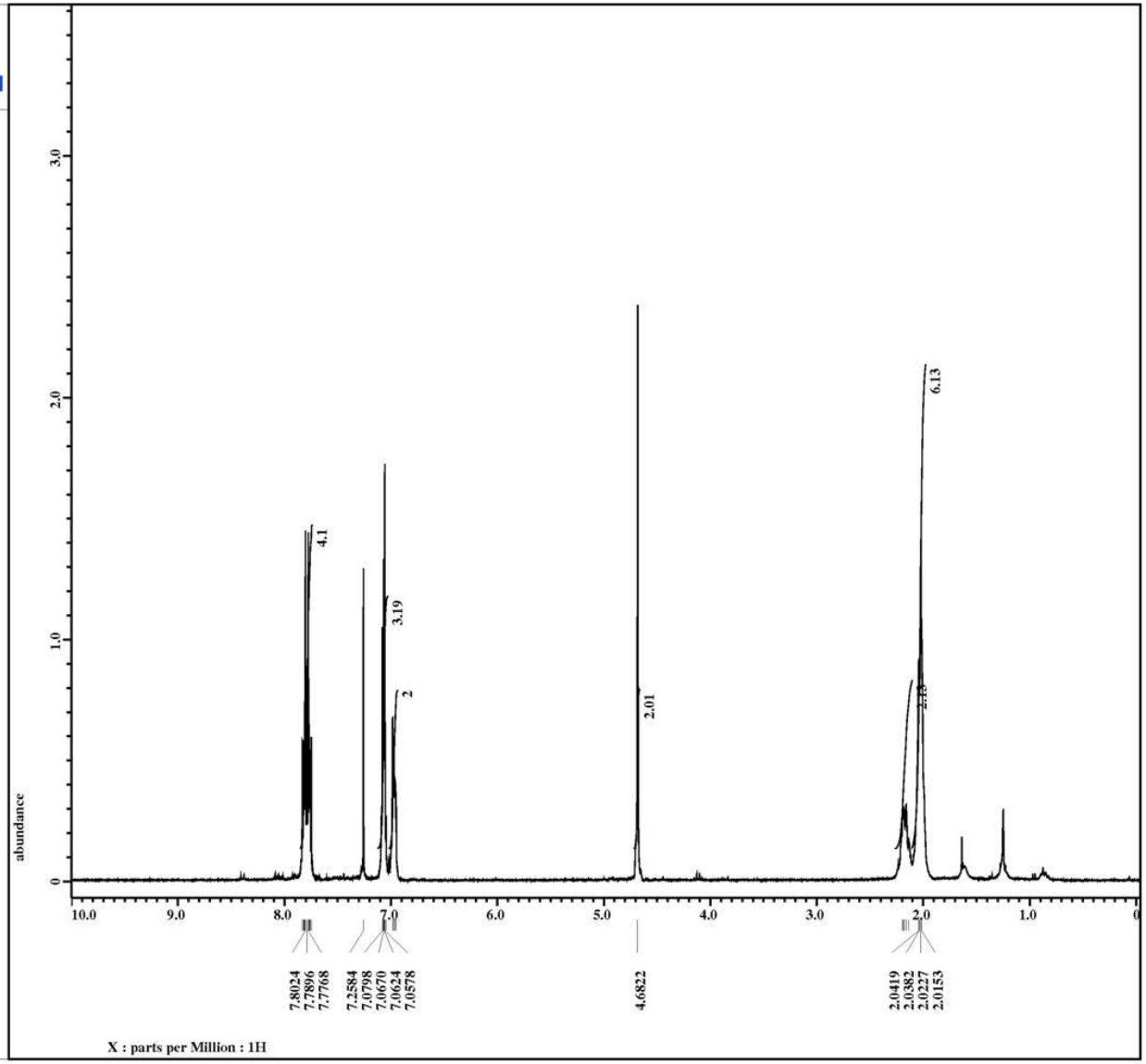
APPENDIX 16

^{1}H and ^{13}C NMR Spectra of

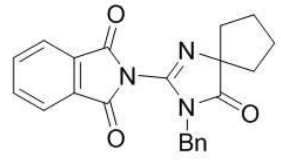
3-benzyl-2-phthalimidoyl-1,3-diazaspiro[4.4]non-2-ene-4-one (**106a**)

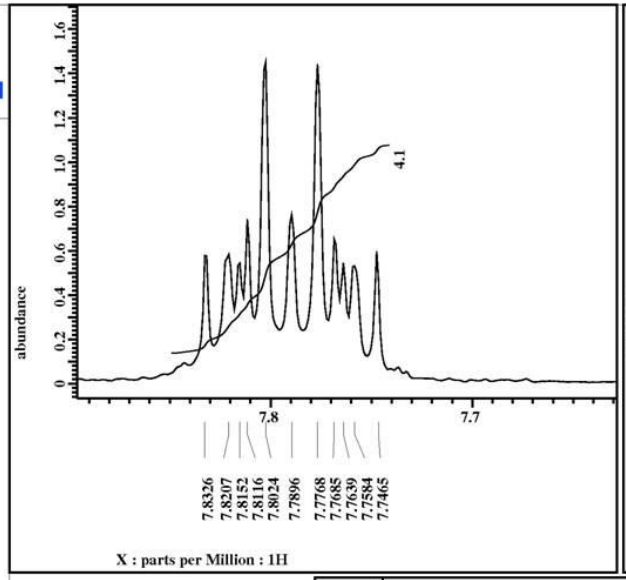
```
Filename          = II_p_129_I-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#793447
Solvent           = CHLOROFORM-D
Creation_time     = 18-DEC-2006 22:52:17
Revision_time     = 17-MAR-2010 17:54:04
Current_time      = 17-MAR-2010 17:54:20

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.6[dC]
```

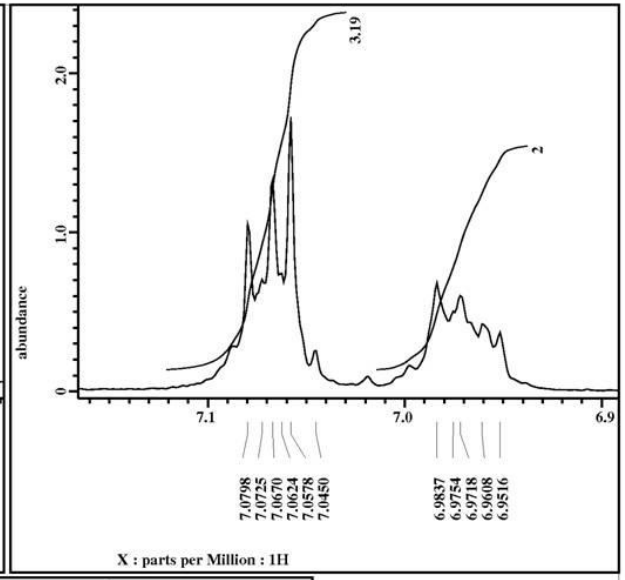


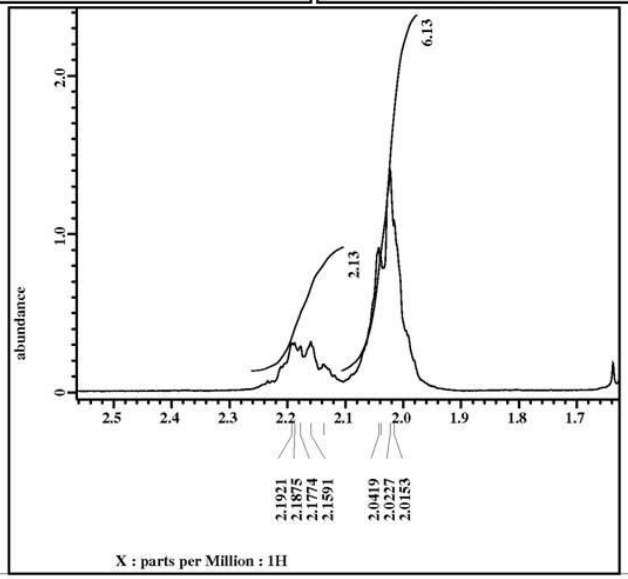


```
Filename          = II_p_129_I-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#793447
Solvent           = CHLOROFORM-D
Creation_time     = 18-DEC-2006 22:52:17
Revision_time     = 17-MAR-2010 17:54:04
Current_time      = 17-MAR-2010 17:54:49

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.6[dC]
```

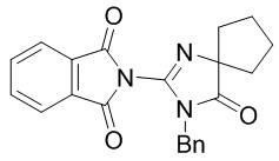


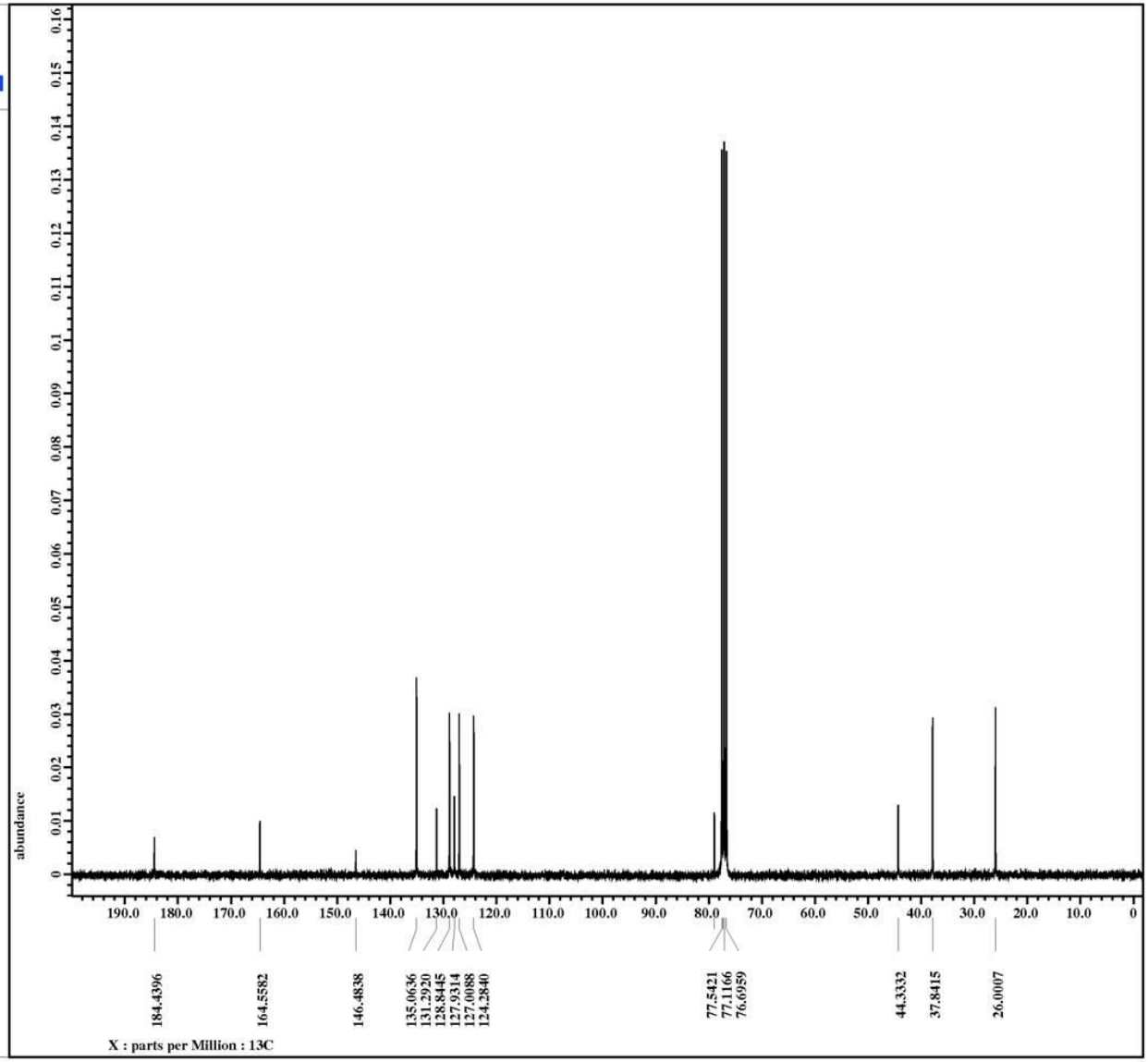






```
Filename          = II_p_129_I-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#795240
Solvent           = CHLOROFORM-D
Creation_time     = 19-DEC-2006 08:49:09
Revision_time     = 17-MAR-2010 17:55:20
Current_time      = 17-MAR-2010 17:55:39

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7500
Total_scans       = 7500

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```





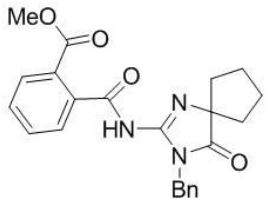
APPENDIX 17

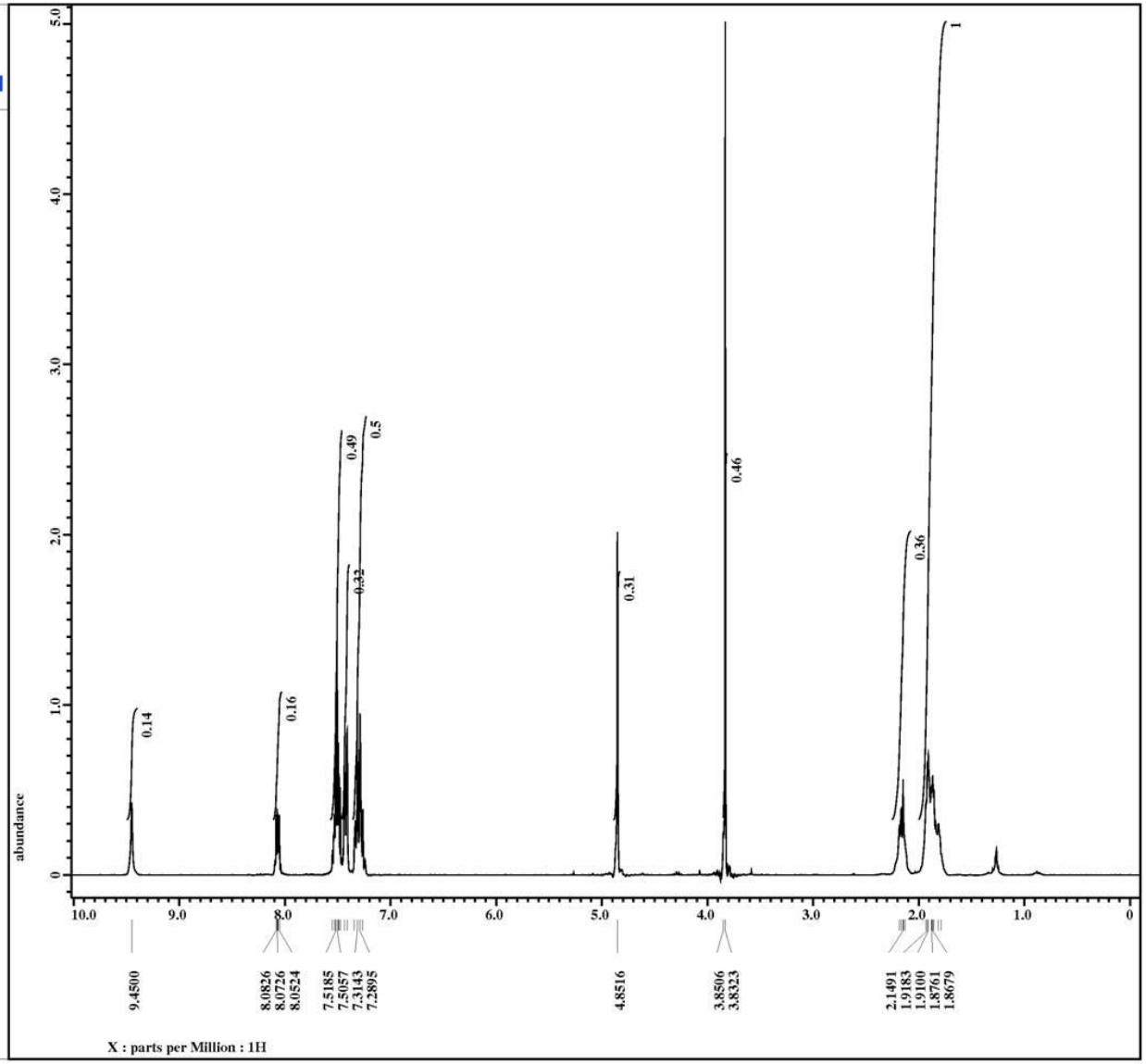
^{1}H and ^{13}C NMR Spectra of

N-(3-Benzyl-4-oxo-1,3-diaza-spiro[4.4]non-1-en-2-yl)-phthalamic acid methyl ester

(**106b**)

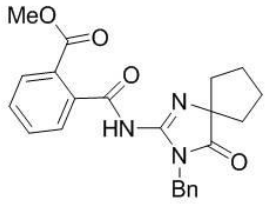
```
Filename          = II_p_070_product-5.jd
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#839724
Solvent           = CHLOROFORM-D
Creation_time     = 29-AUG-2006 00:01:01
Revision_time     = 17-MAR-2010 18:08:09
Current_time      = 17-MAR-2010 18:11:41

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

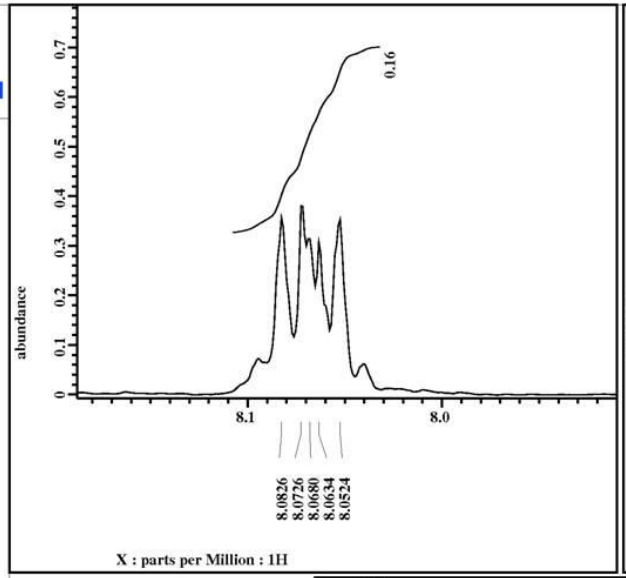


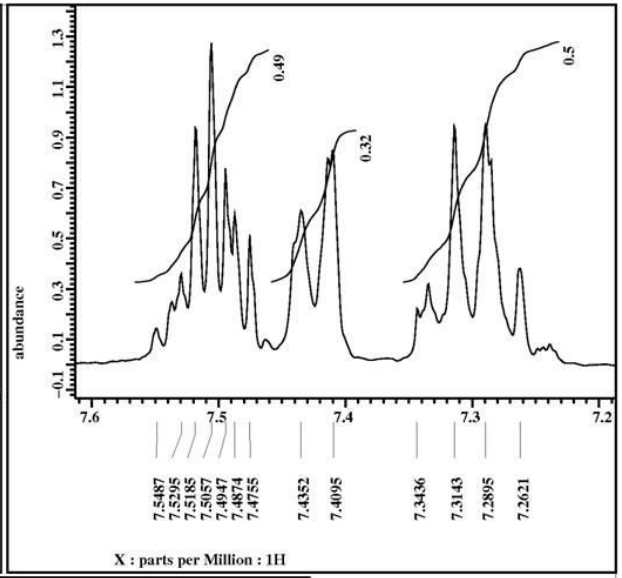


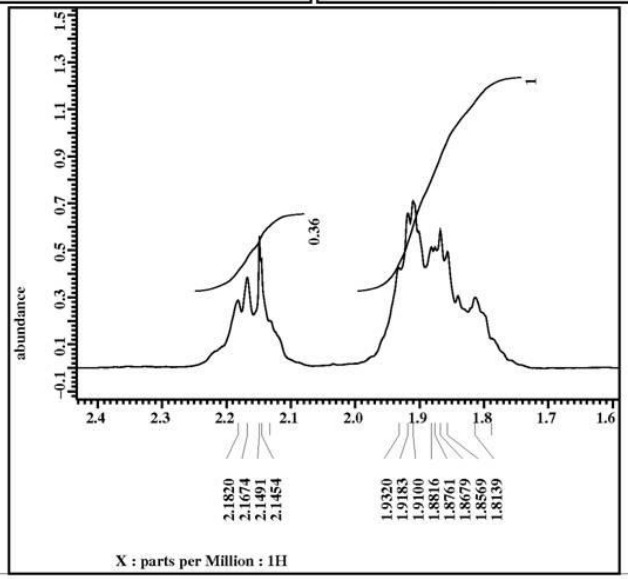
JEOL

Filename	= II_p_070_product-5.jd
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#839724
Solvent	= CHLOROFORM-D
Creation_time	= 29-AUG-2006 00:01:01
Revision_time	= 17-MAR-2010 18:08:09
Current_time	= 17-MAR-2010 18:12:32
Comment	= single_pulse
Data_format	= 1D REAL
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.9[dC]

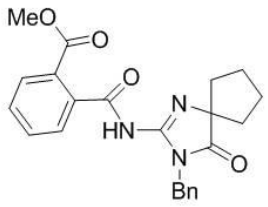


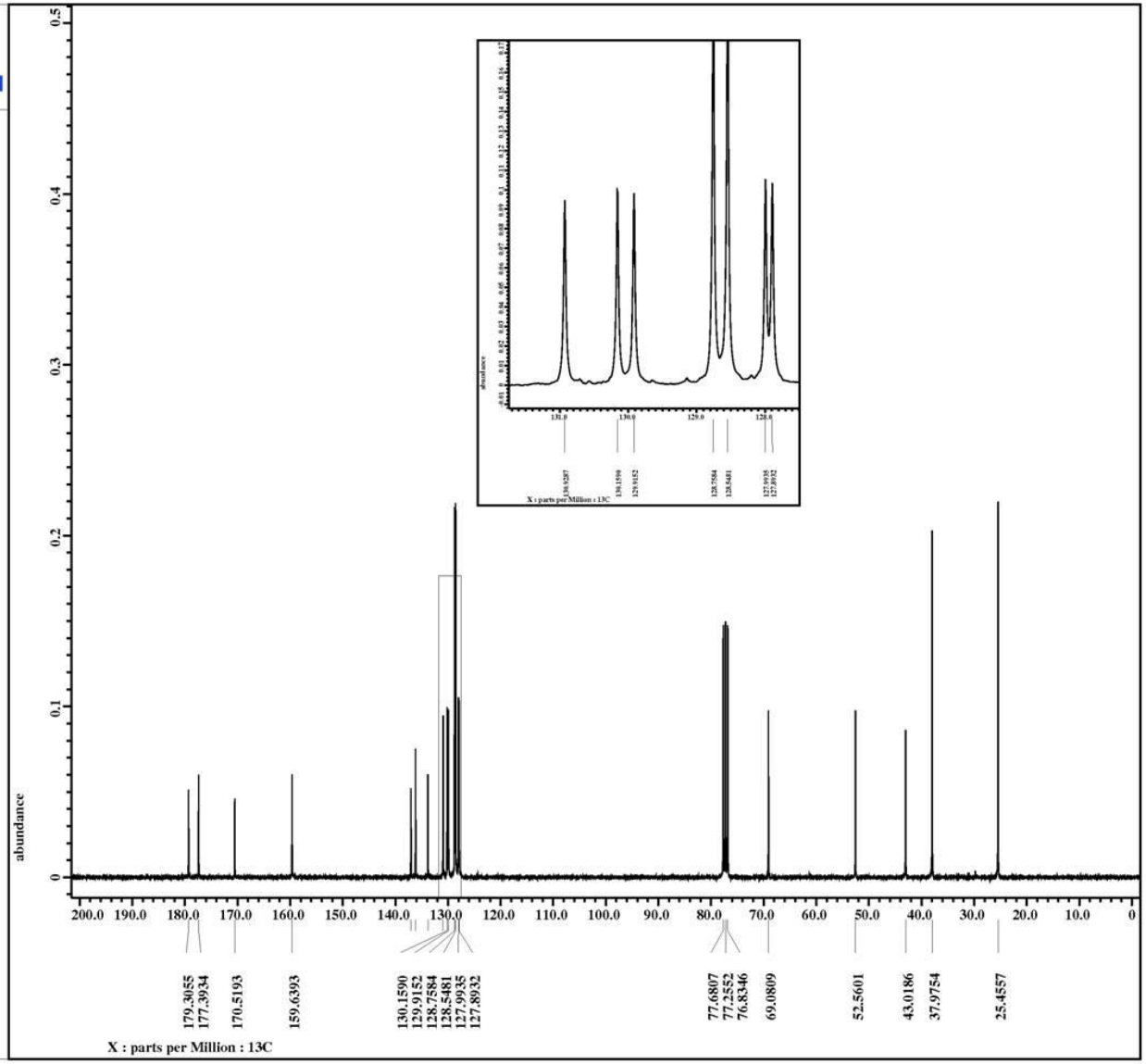






```
Filename          = II_p_070_product-4.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#840011
Solvent           = CHLOROFORM-D
Creation_time     = 29-AUG-2006 03:40:46
Revision_time     = 17-MAR-2010 18:15:48
Current_time      = 17-MAR-2010 18:18:19

Comment           = single pulse decouple
Data_format       = 1D REAL
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 2760
Total_scans       = 2760

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.8[dC]
```



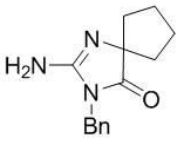


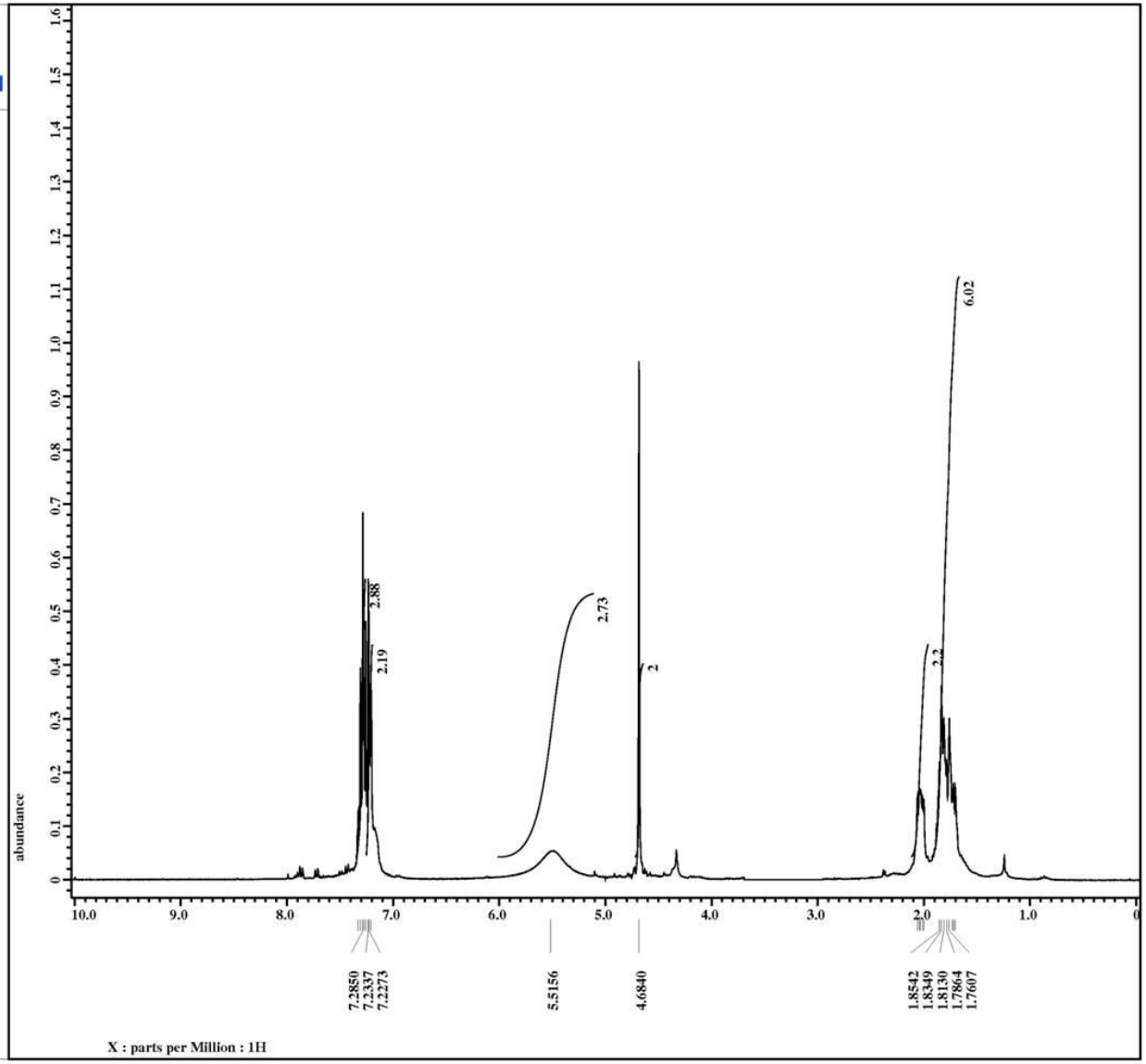
APPENDIX 18

^{1}H and ^{13}C NMR Spectra of

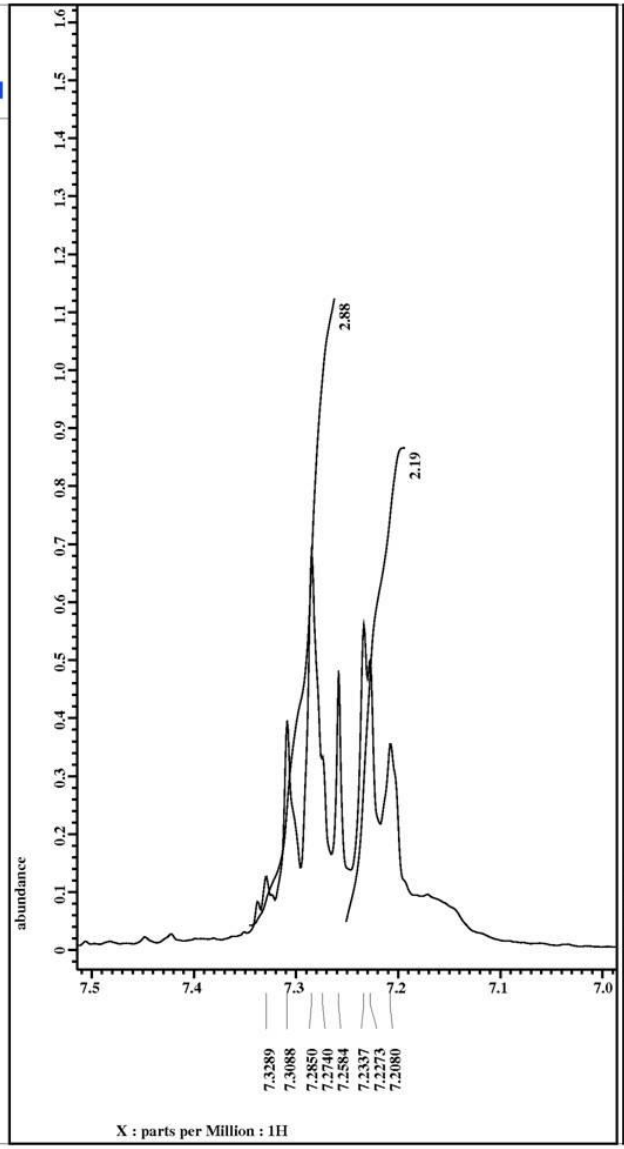
2-Amino-3-benzyl-1,3-diazaspiro[4.4]non-2-ene-4-one (**106c**)

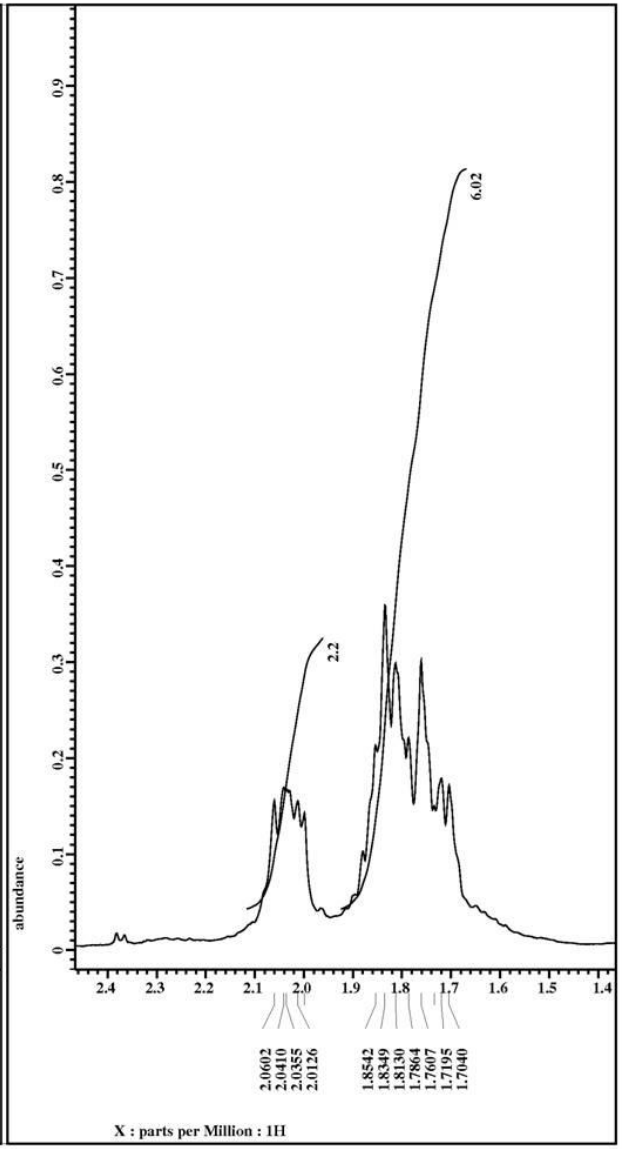
```
Filename          = II_p_055_product-3.jd
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#495180
Solvent           = CHLOROFORM-D
Creation_time     = 27-JUL-2006 14:25:16
Revision_time     = 17-MAR-2010 18:31:50
Current_time      = 17-MAR-2010 18:32:11

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23[dC]
```



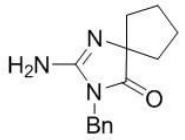


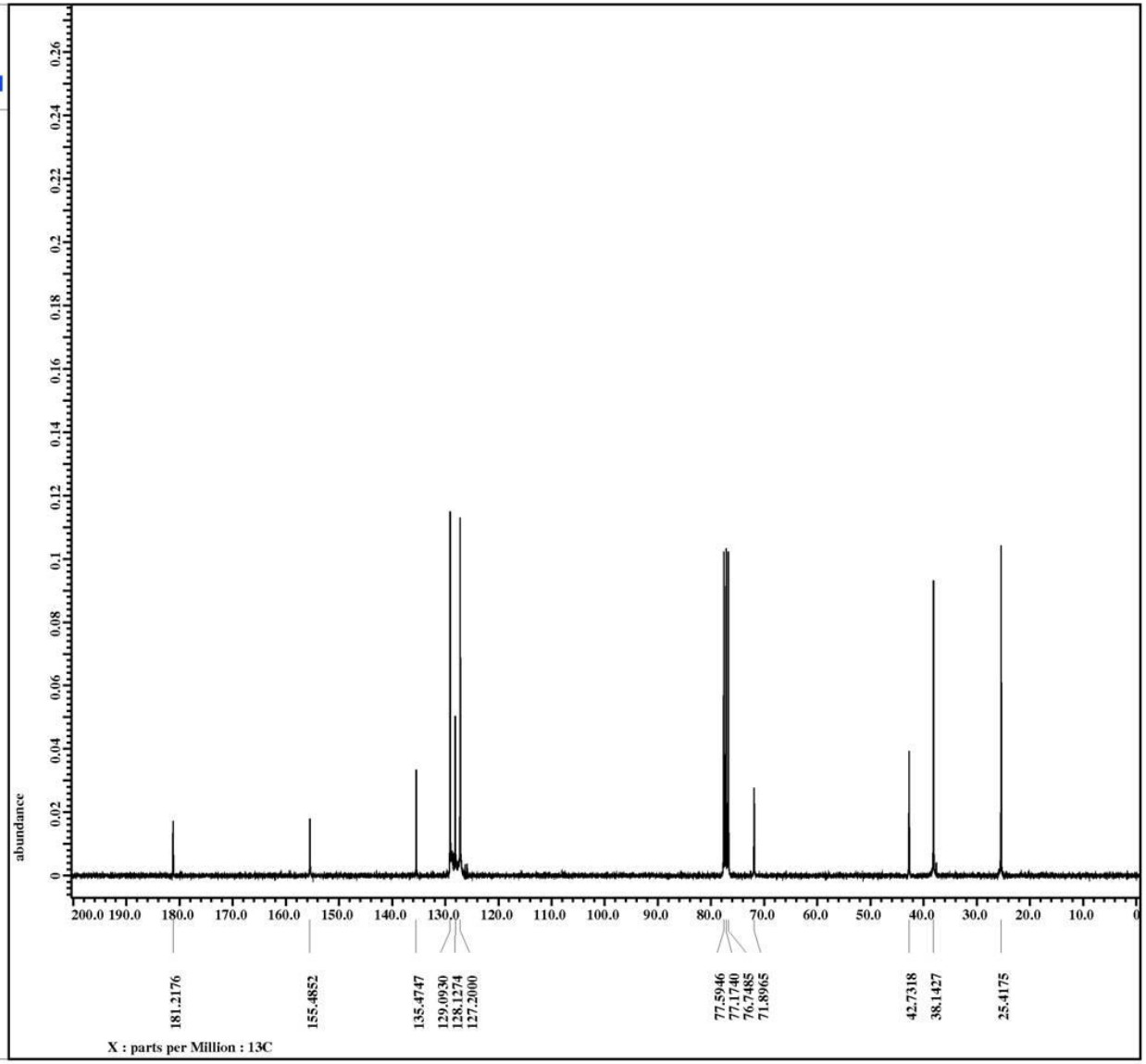
Filename	= II_p_055_product-3.jd
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#495180
Solvent	= CHLOROFORM-D
Creation_time	= 27-JUL-2006 14:25:16
Revision_time	= 17-MAR-2010 18:31:50
Current_time	= 17-MAR-2010 18:32:55
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23[dC]





```
Filename          = II_p_055_product-4.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#795401
Solvent           = CHLOROFORM-D
Creation_time     = 28-JUL-2006 09:19:36
Revision_time     = 17-MAR-2010 18:33:29
Current_time      = 17-MAR-2010 18:33:42

Comment           = single pulse decouple
Data_format       = 1D REAL
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8000
Total_scans       = 8000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.8[dC]
```

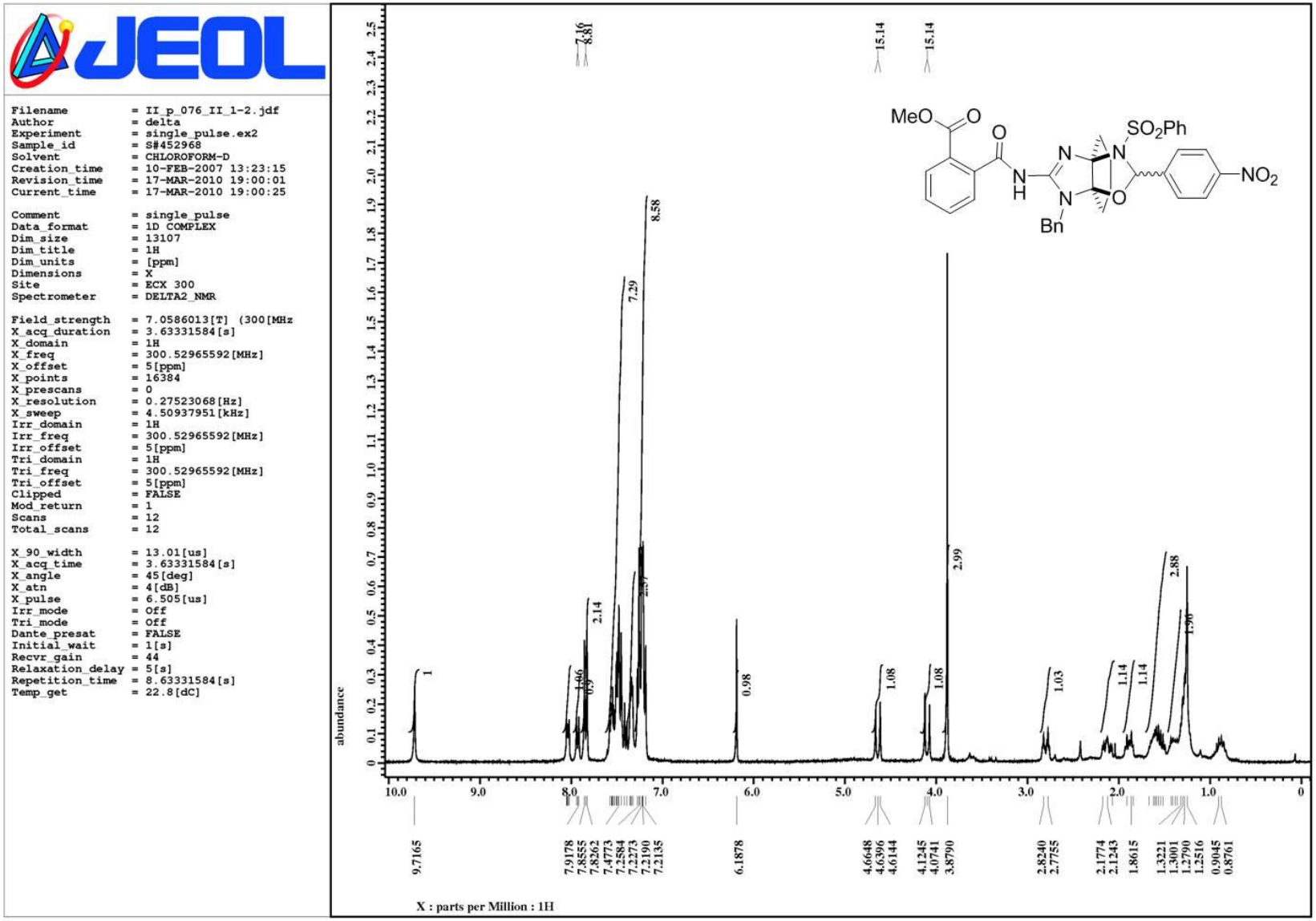




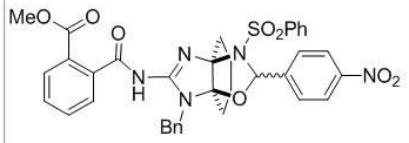
APPENDIX 19

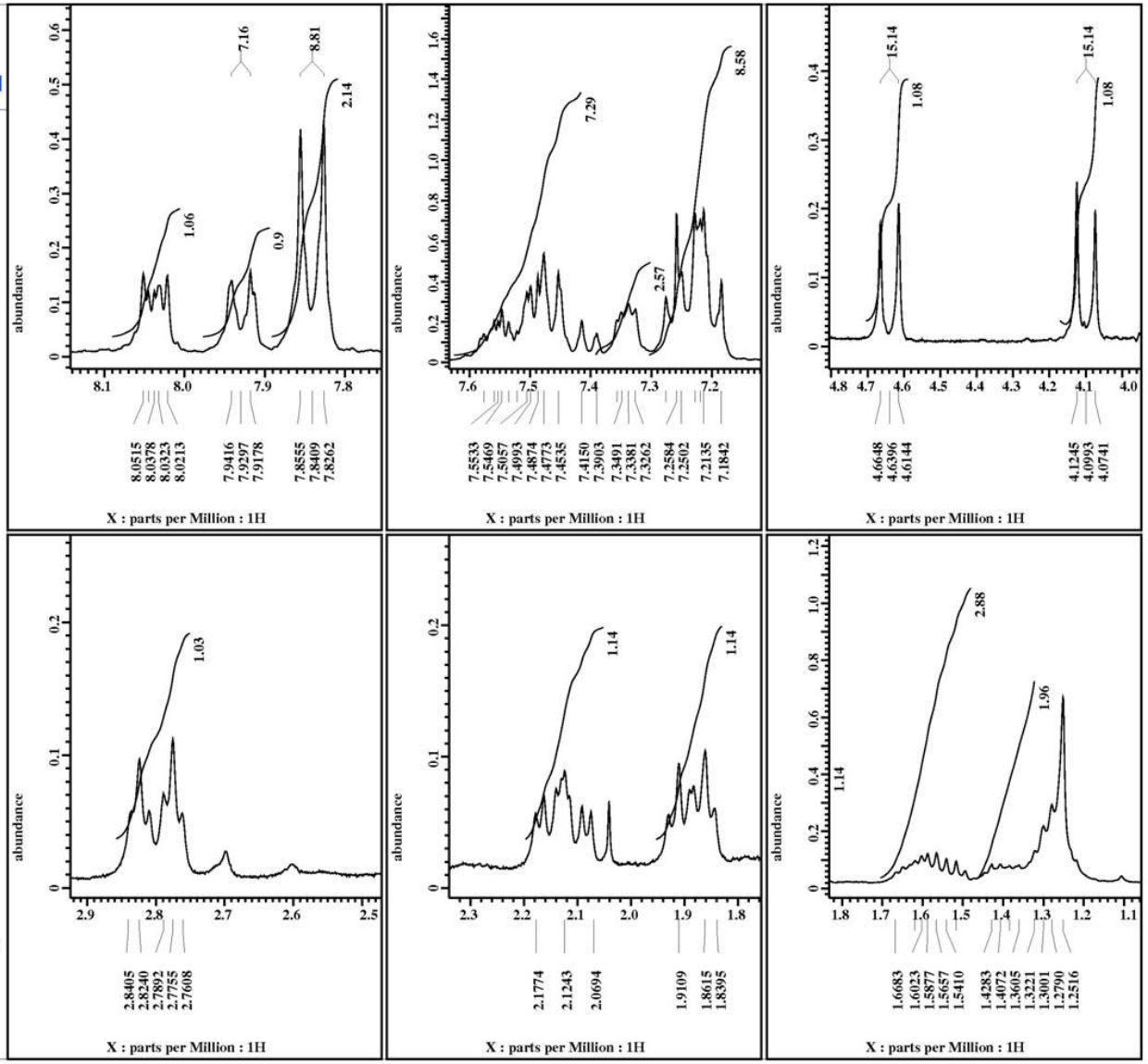
^{1}H and ^{13}C NMR Spectra of

(1R,6S*,8R*/8S*)-N*-[9-Benzenesulfonyl-12-benzyl-8-(4-nitrophenyl)-7-oxa-9,10,12-triaza-tricyclo[4.3.3.0]dodec-10-en-11-yl]-phthalamic acid methyl ester (**107a**)

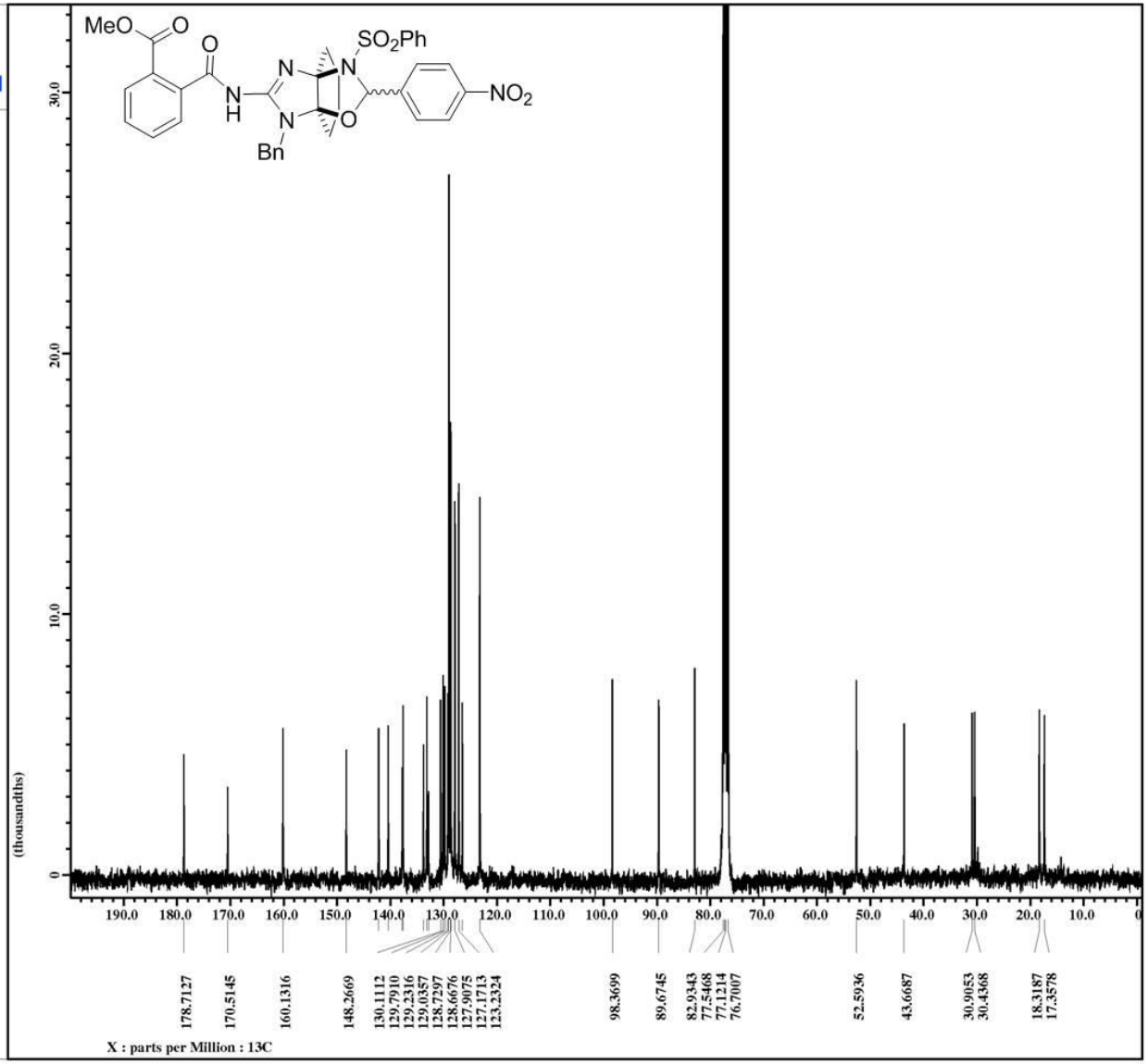

Filename = II_p_076_II_1-2.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#452968
Solvent = CHLOROFORM-D
Creation_time = 10-FEB-2007 13:23:15
Revision_time = 17-MAR-2010 19:00:01
Current_time = 17-MAR-2010 19:00:25
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 44
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 22.8[dC]
MeO
O
O
N
H
N
N
N
Bn
SO2Ph
NO2
abundance
X : parts per Million : 1H

Filename	= II_p_076_II_1-2.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#452968
Solvent	= CHLOROFORM-D
Creation_time	= 10-FEB-2007 13:23:15
Revision_time	= 17-MAR-2010 19:01:50
Current_time	= 17-MAR-2010 19:02:07
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 44
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.8[dC]

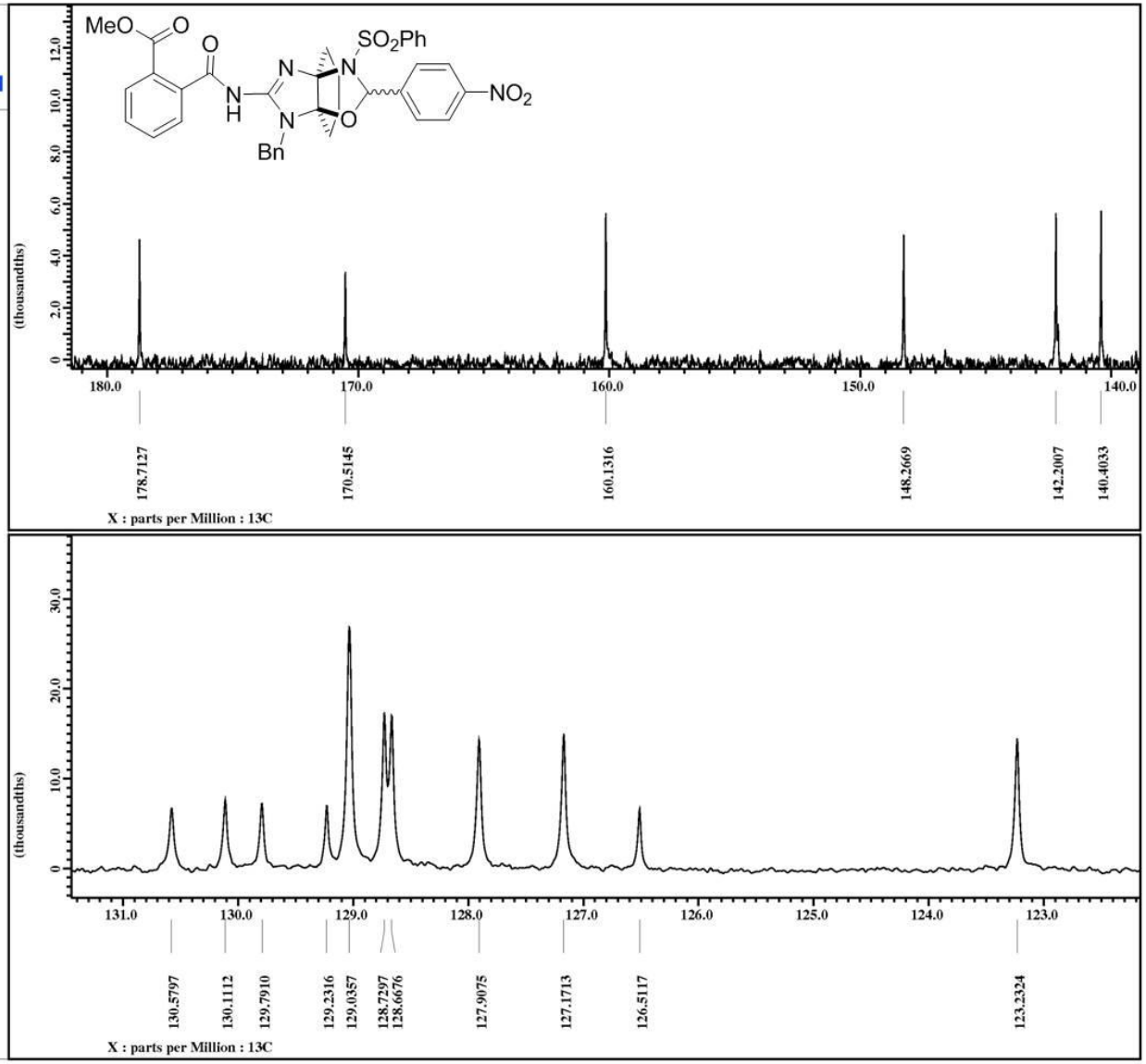




```
Filename          = II_p_076_II_1-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 11-FEB-2007 08:33:03
Revision_time     = 17-MAR-2010 19:03:40
Current_time      = 17-MAR-2010 19:04:12

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 47
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7000
Total_scans       = 7000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.8[dC]
```



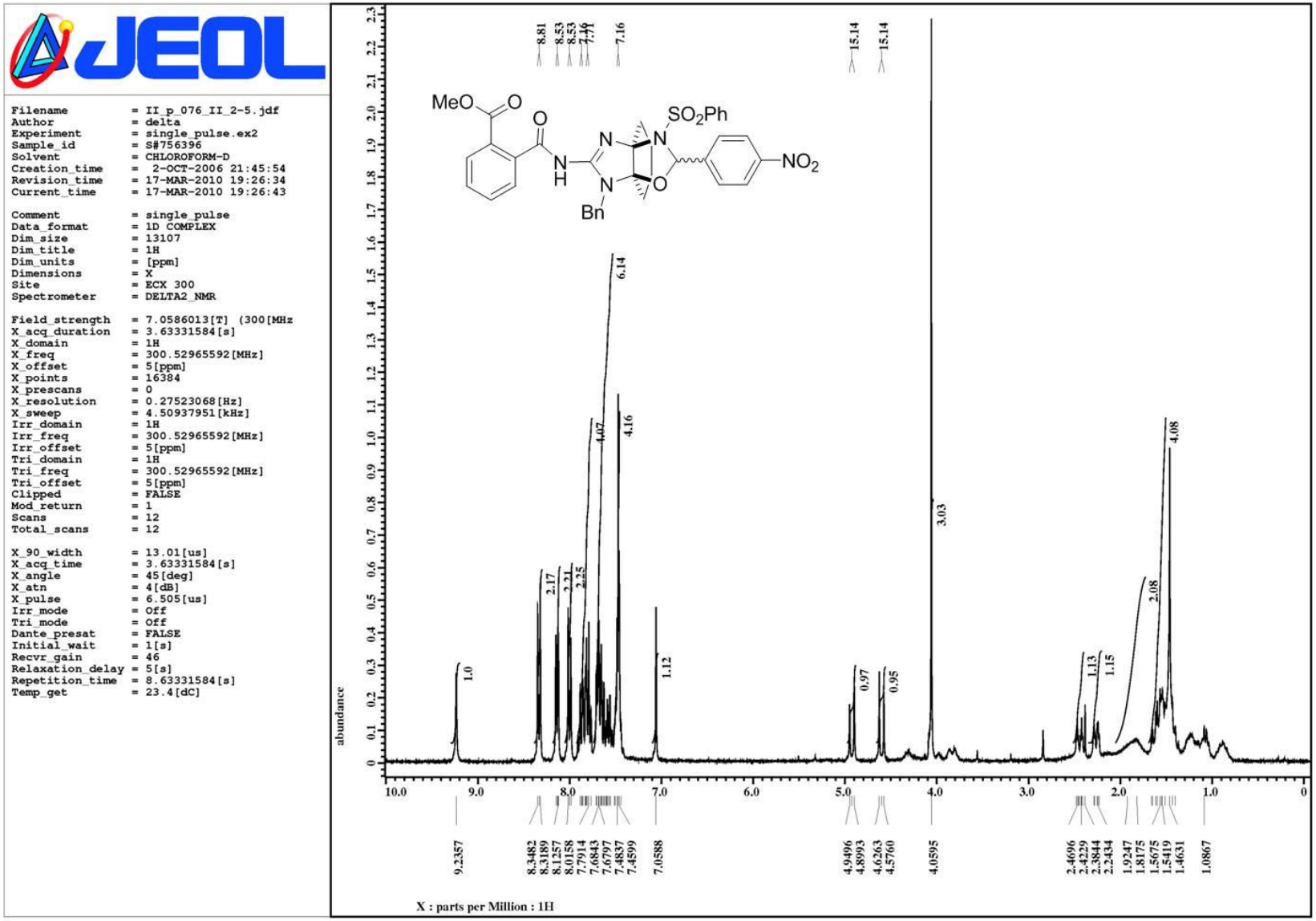
```
Filename          = II_p_076_II_1-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 11-FEB-2007 08:33:03
Revision_time     = 17-MAR-2010 19:03:40
Current_time      = 17-MAR-2010 19:05:05

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 47
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7000
Total_scans       = 7000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.8[dC]
```



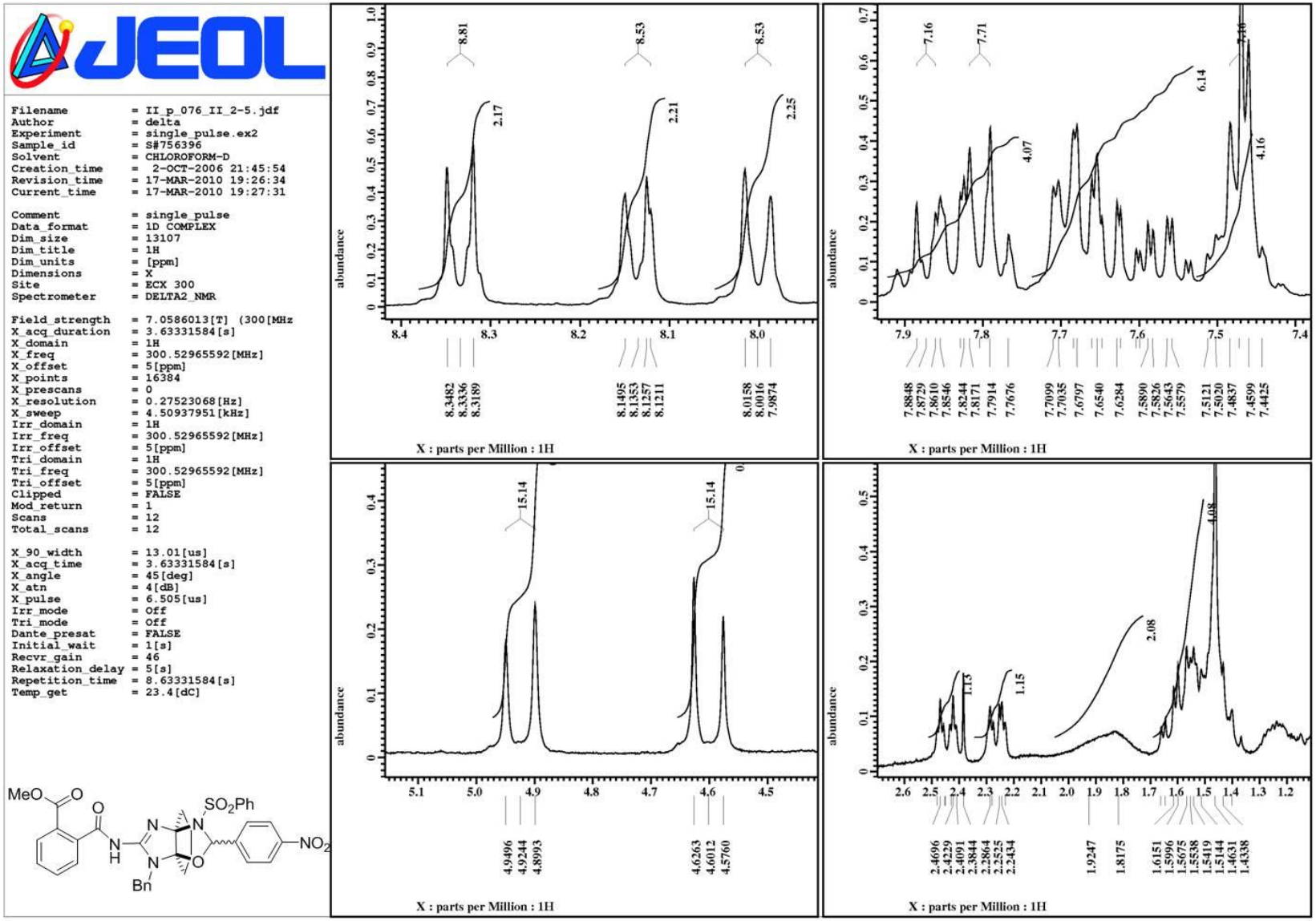
APPENDIX 20

^{1}H and ^{13}C NMR Spectra of

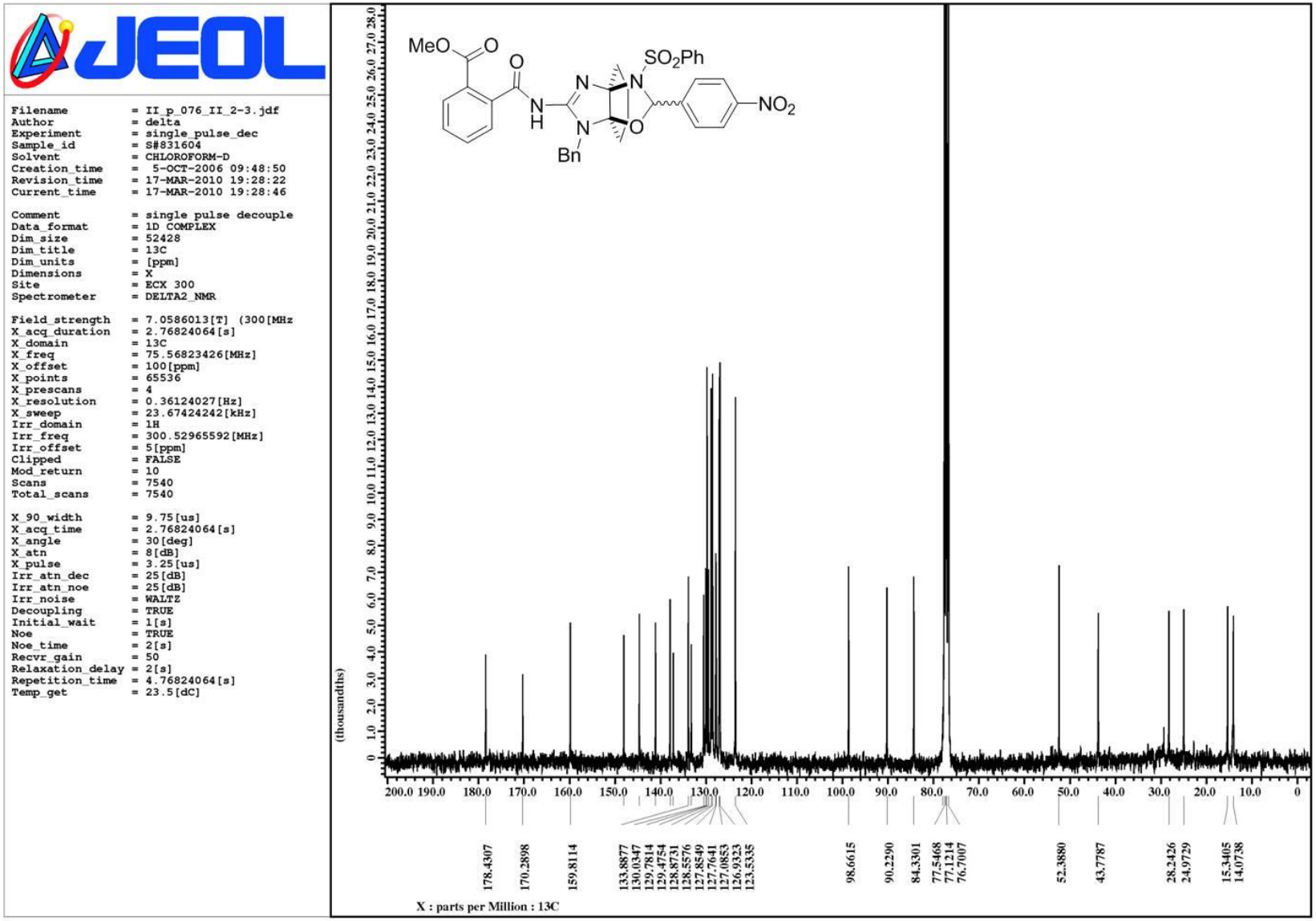
(1R,6S*,8R*/8S*)-N*-[9-Benzenesulfonyl-12-benzyl-8-(4-nitrophenyl)-7-oxa-9,10,12-triaza-tricyclo[4.3.3.0]dodec-10-en-11-yl]-phthalamic acid methyl ester (**107b**)



```
Filename          = II_p_076_II_2-5.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#756396
Solvent           = CHLOROFORM-D
Creation_time     =  2-OCT-2006 21:45:54
Revision_time     = 17-MAR-2010 19:26:34
Current_time      = 17-MAR-2010 19:26:43

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.4[dC]
```

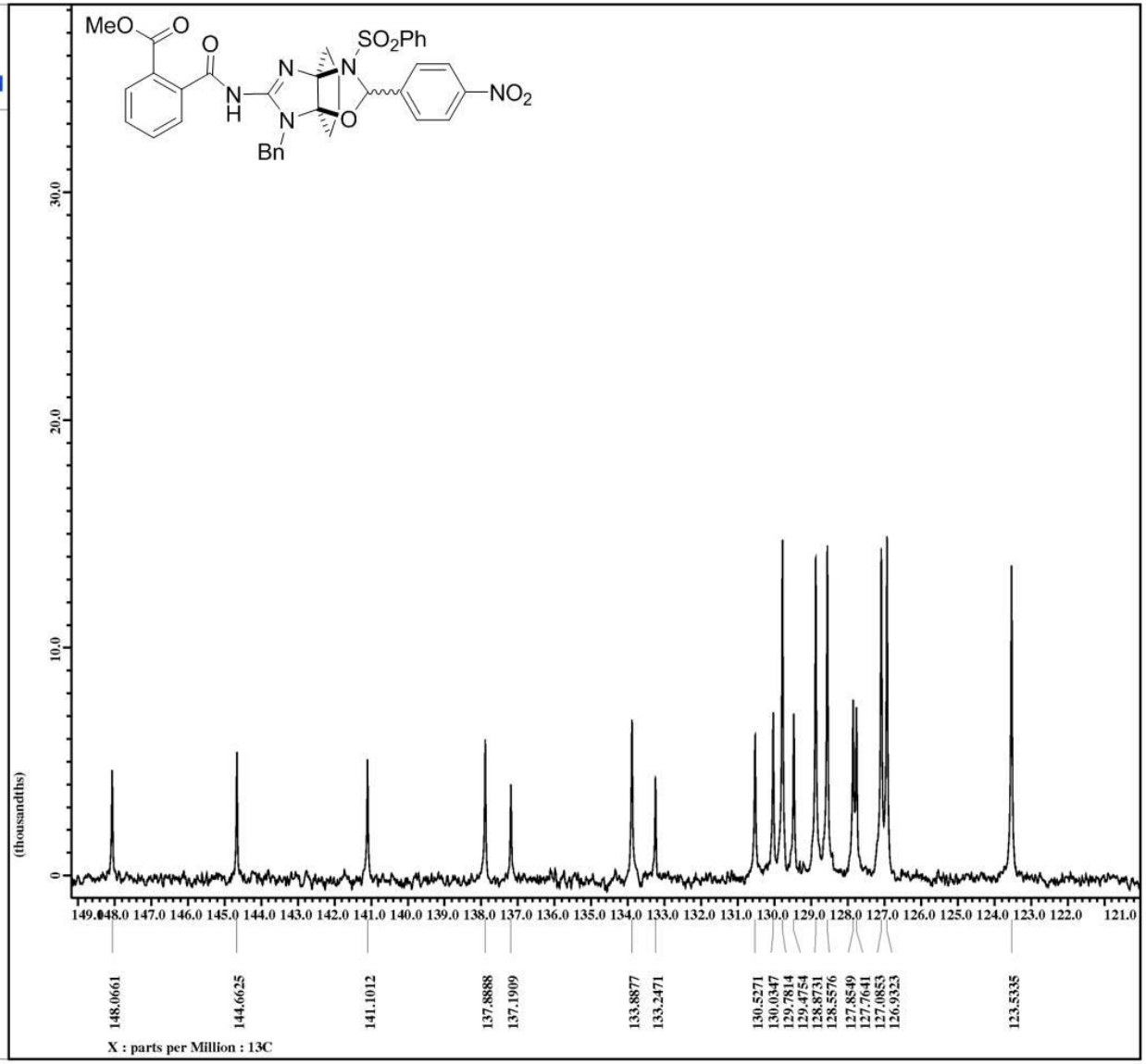


```
Filename          = II_p_076_II_2-5.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#756396
Solvent           = CHLOROFORM-D
Creation_time     =  2-OCT-2006 21:45:54
Revision_time     = 17-MAR-2010 19:26:34
Current_time      = 17-MAR-2010 19:27:31

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.4[dC]
```


Filename = II_p_076_II_2-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#831604
Solvent = CHLOROFORM-D
Creation_time = 5-OCT-2006 09:48:50
Revision_time = 17-MAR-2010 19:28:22
Current_time = 17-MAR-2010 19:28:46
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 7540
Total_scans = 7540
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.5[dC]
MeO
O
O
N
H
N
N
Bn
N
O
SO2Ph
NO2
(thousandths)
178.4307
170.2898
159.8114
133.8877
130.0347
129.7814
129.4754
128.8731
128.5576
127.8549
127.7641
127.0853
126.9323
123.5335
98.6615
90.2290
84.3301
77.5468
77.1214
76.7007
52.3880
43.7787
28.2426
24.9729
15.3405
14.0738
X : parts per Million : 13C

JEOL

Filename	= II_p_076_II_2-2.jdf
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#831604
Solvent	= CHLOROFORM-D
Creation_time	= 5-OCT-2006 09:48:50
Revision_time	= 5-OCT-2006 09:21:48
Current_time	= 17-MAR-2010 19:35:49
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 7540
Total_scans	= 7540
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 23.5[dC]



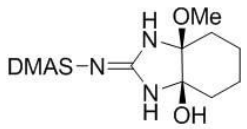
APPENDIX 21

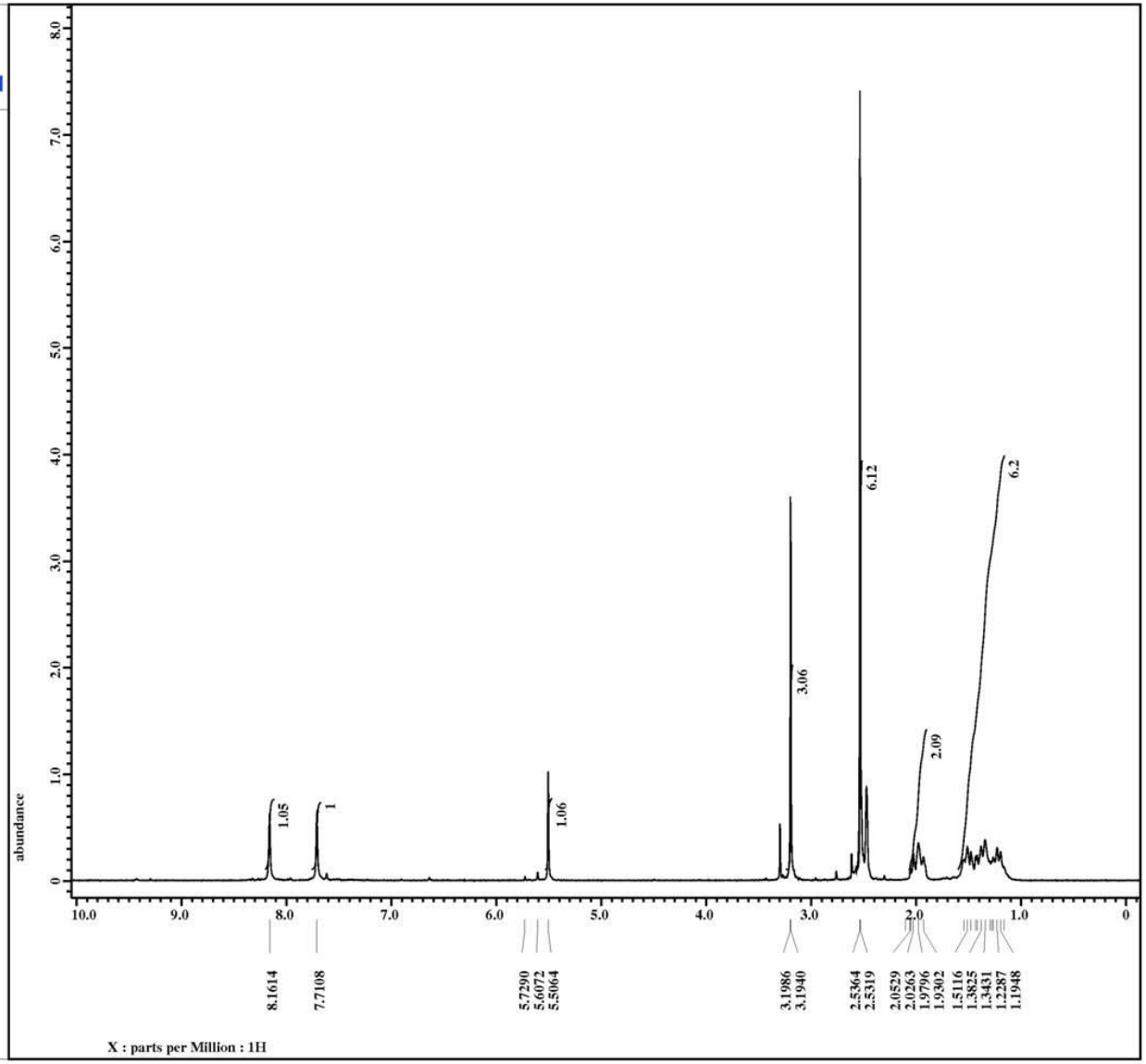
^{1}H and ^{13}C NMR Spectra of

2-Dimethylaminosulfonylimino-3a-hydroxy-7a-methoxyoctahydrobenzimidazole

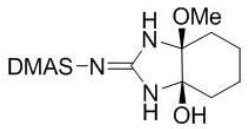
(**108a**)

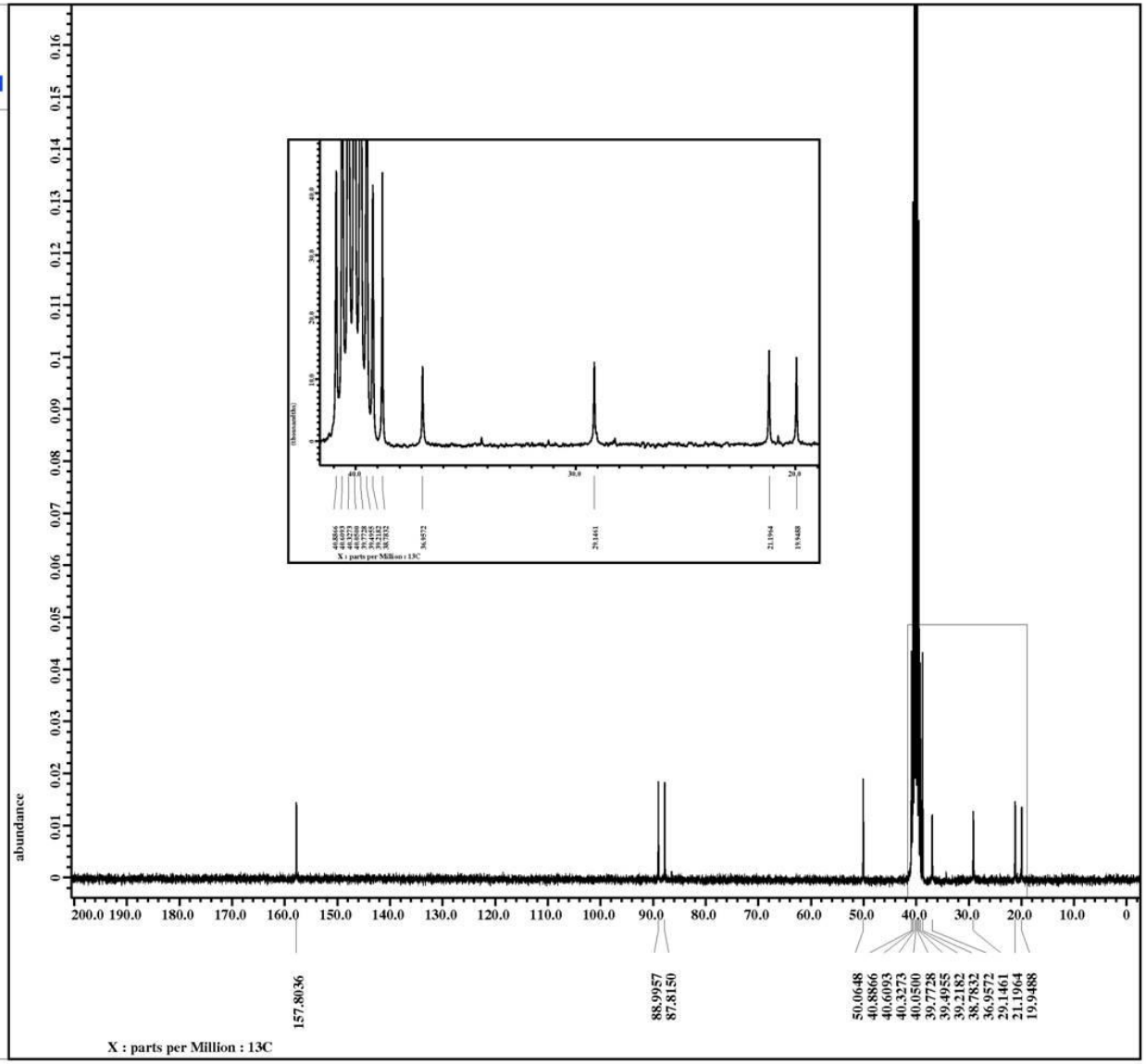
```
Filename          = II_p_097_IIIcrope_DMS
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#592263
Solvent           = DMSO-D6
Creation_time     =  3-NOV-2006 17:11:30
Revision_time     = 17-MAR-2010 19:55:42
Current_time      = 17-MAR-2010 19:55:59

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23[dC]
```





```
Filename          = II_p_097_IIIcrope-2.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#786798
Solvent           = DMSO-D6
Creation_time     =  4-NOV-2006 09:50:34
Revision_time     = 17-MAR-2010 19:53:15
Current_time      = 17-MAR-2010 19:53:28

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8500
Total_scans       = 8500

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```



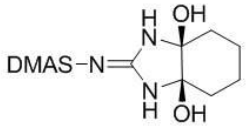


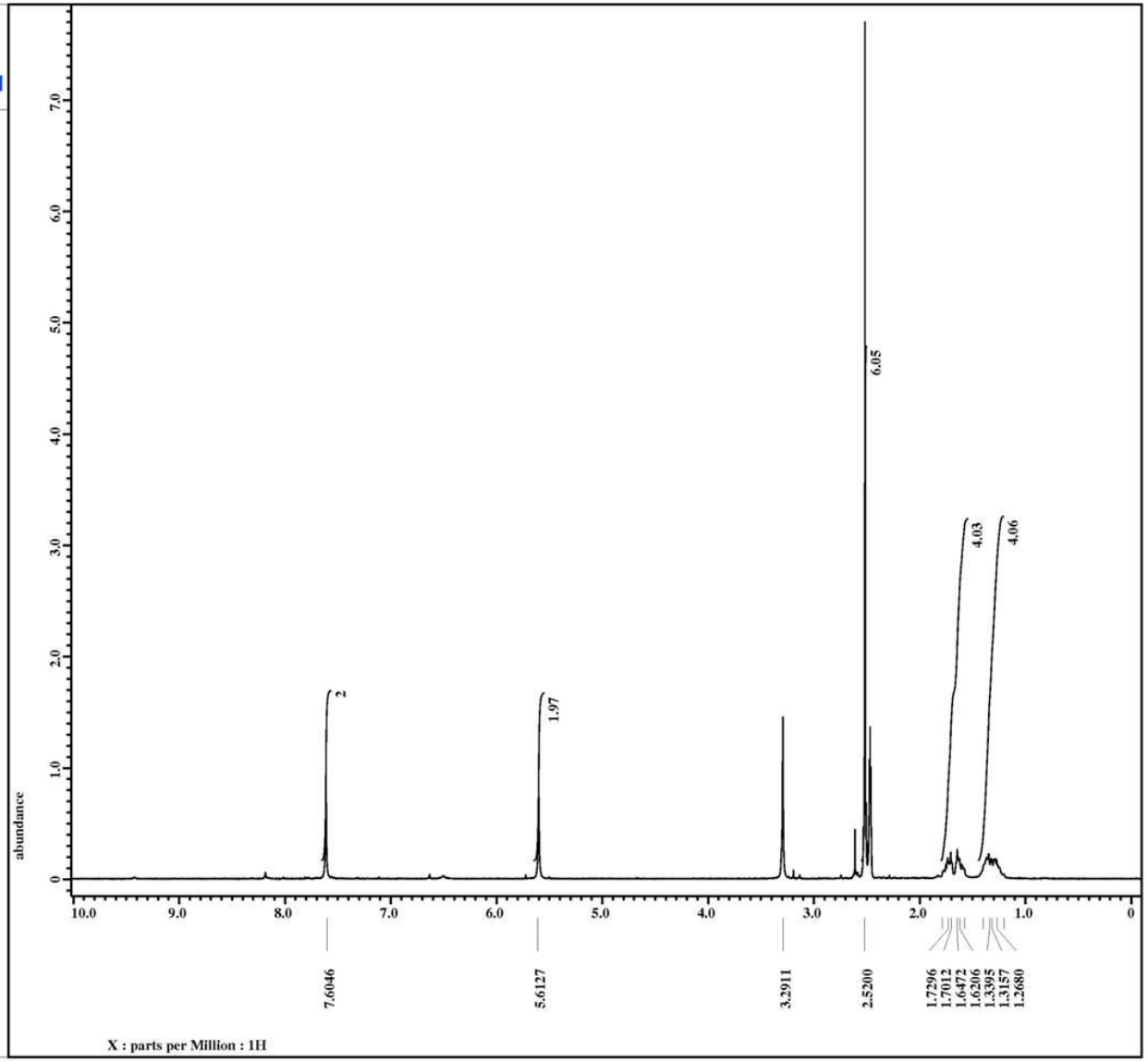
APPENDIX 22

^{1}H and ^{13}C NMR Spectra of

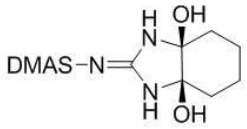
2-Dimethylaminosulfonylimino-3a,7a-dihydroxyhexahydrobenzimidazole (**108b**)

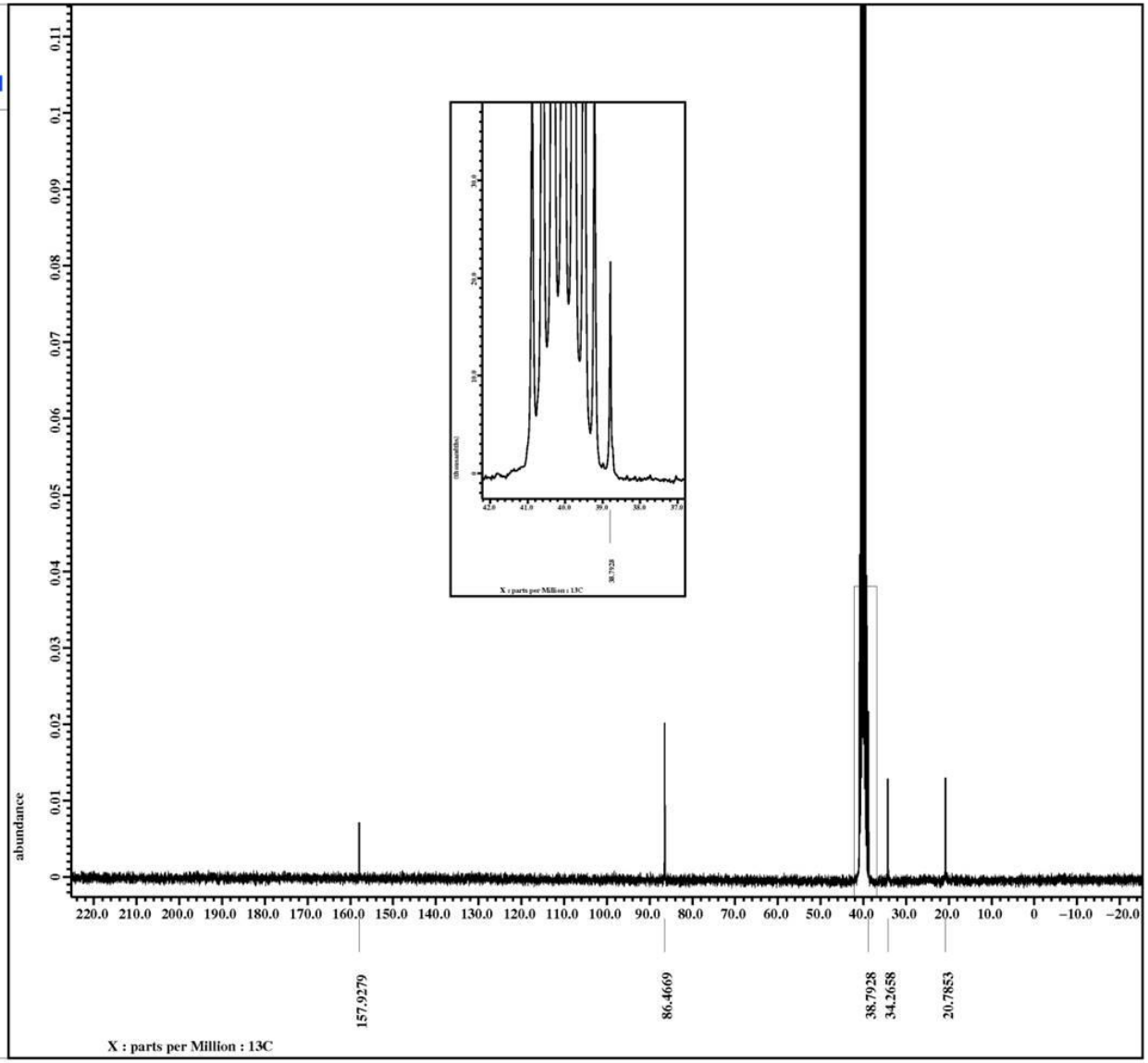
```
Filename          = II_p_154_III_DMSO-3.j
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#758370
Solvent           = DMSO-D6
Creation_time     = 22-FEB-2007 21:41:01
Revision_time     = 17-MAR-2010 20:01:24
Current_time      = 17-MAR-2010 20:01:43

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.7[dC]
```





```
Filename          = II_p_154_III_DMSO-2.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#760281
Solvent           = DMSO-D6
Creation_time     = 23-FEB-2007 08:17:51
Revision_time     = 23-FEB-2007 10:24:35
Current_time      = 17-MAR-2010 20:02:52

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8000
Total_scans       = 8000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.9[dC]
```





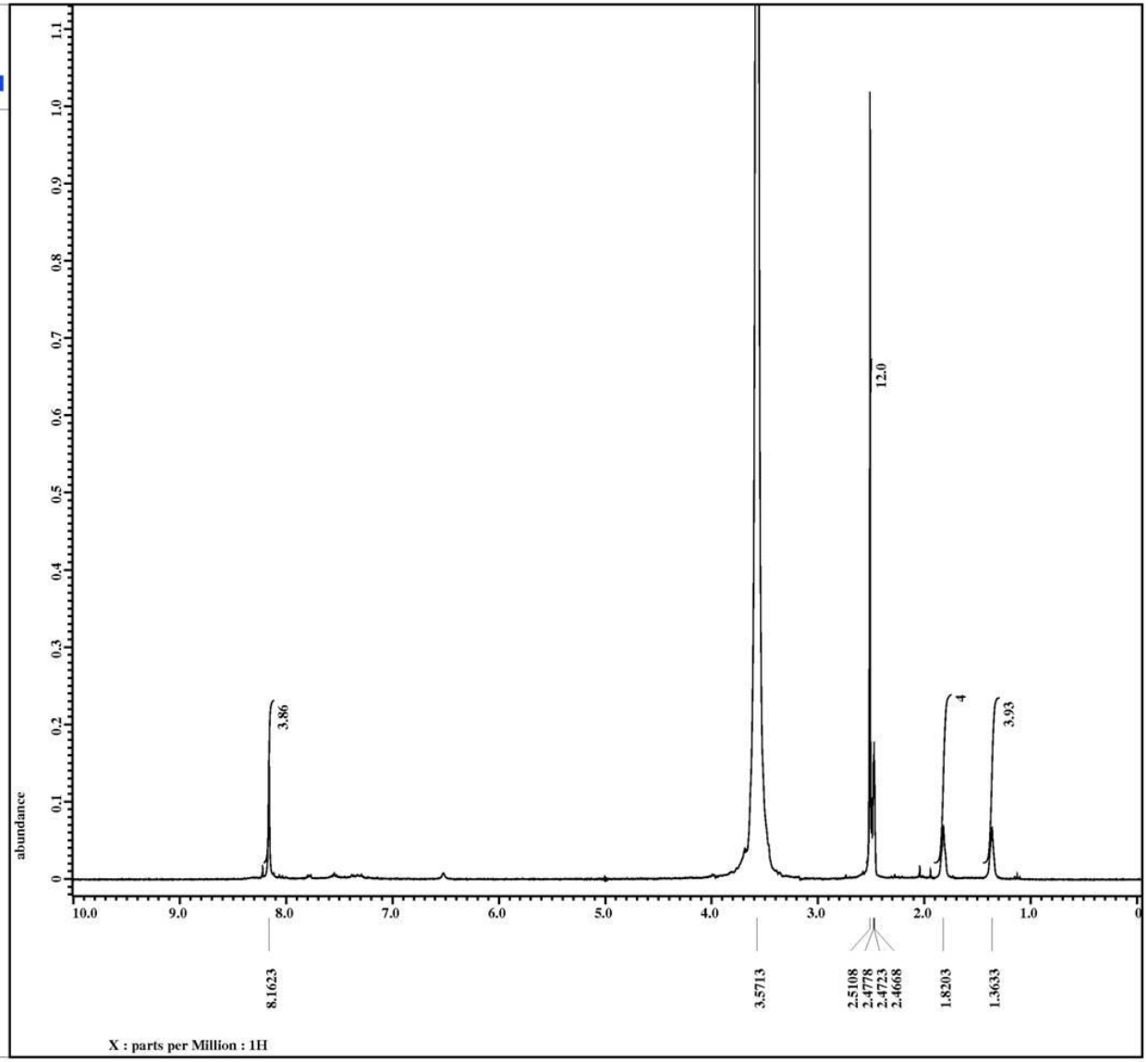
APPENDIX 23

^{1}H and ^{13}C NMR Spectra of

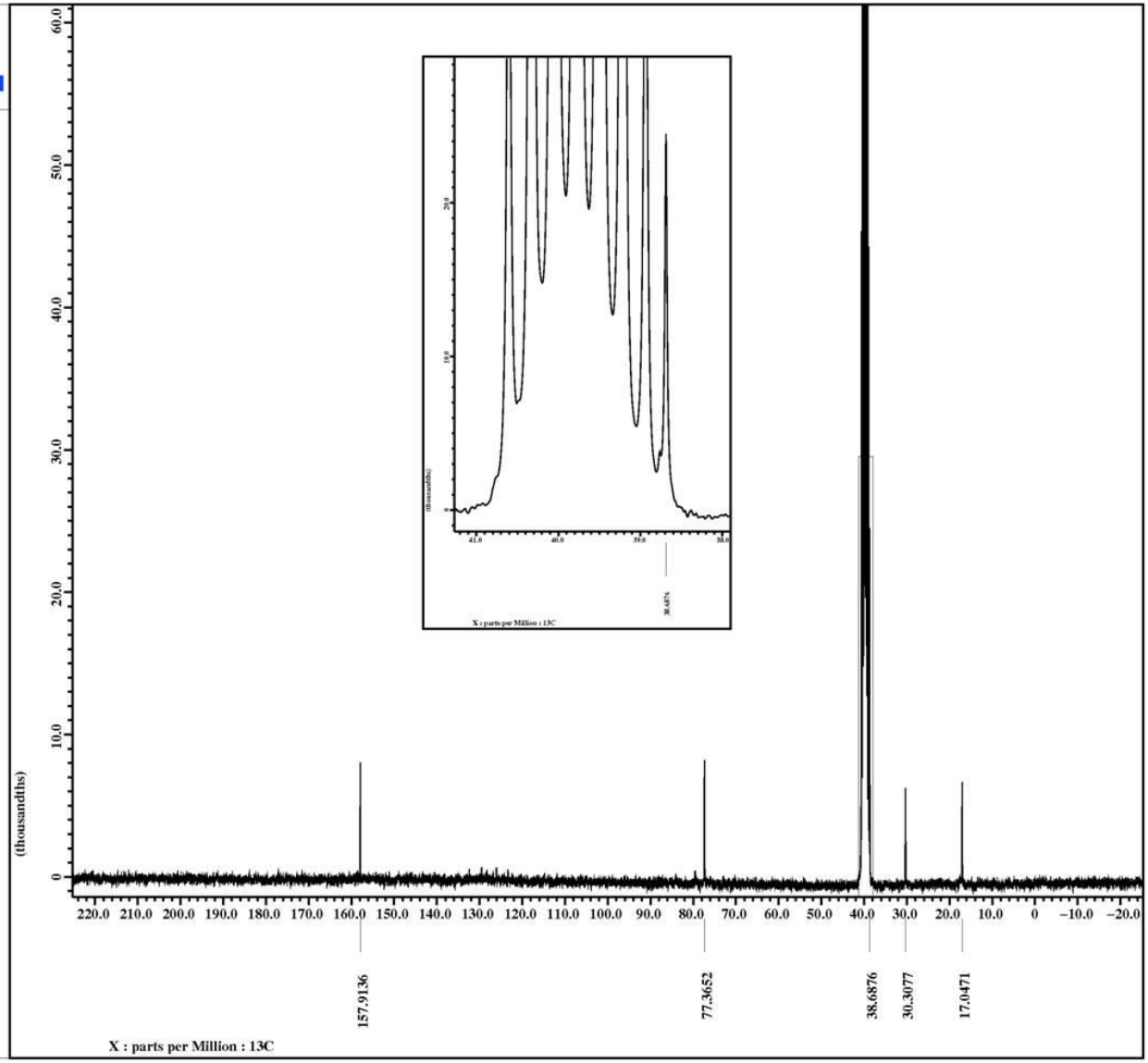
8,11-Bis(*N*,*N*-dimethylsulfonylimino)-7,9,10,12-tetraazatricyclo[4.3.3.0]dodecane

(**120**)

```
Filename          = III_p_290_product-3.j
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#748997
Solvent           = DMSO-D6
Creation_time     = 22-APR-2008 21:01:35
Revision_time     = 17-MAR-2010 20:08:40
Current_time      = 17-MAR-2010 20:08:59

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```



```
Filename          = III_p_290_product-2.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#751917
Solvent           = DMSO-D6
Creation_time     = 23-APR-2008 08:59:50
Revision_time     = 23-APR-2008 09:51:44
Current_time      = 17-MAR-2010 20:10:15

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 9000
Total_scans       = 9000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.4[dC]
```

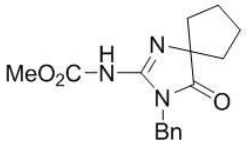


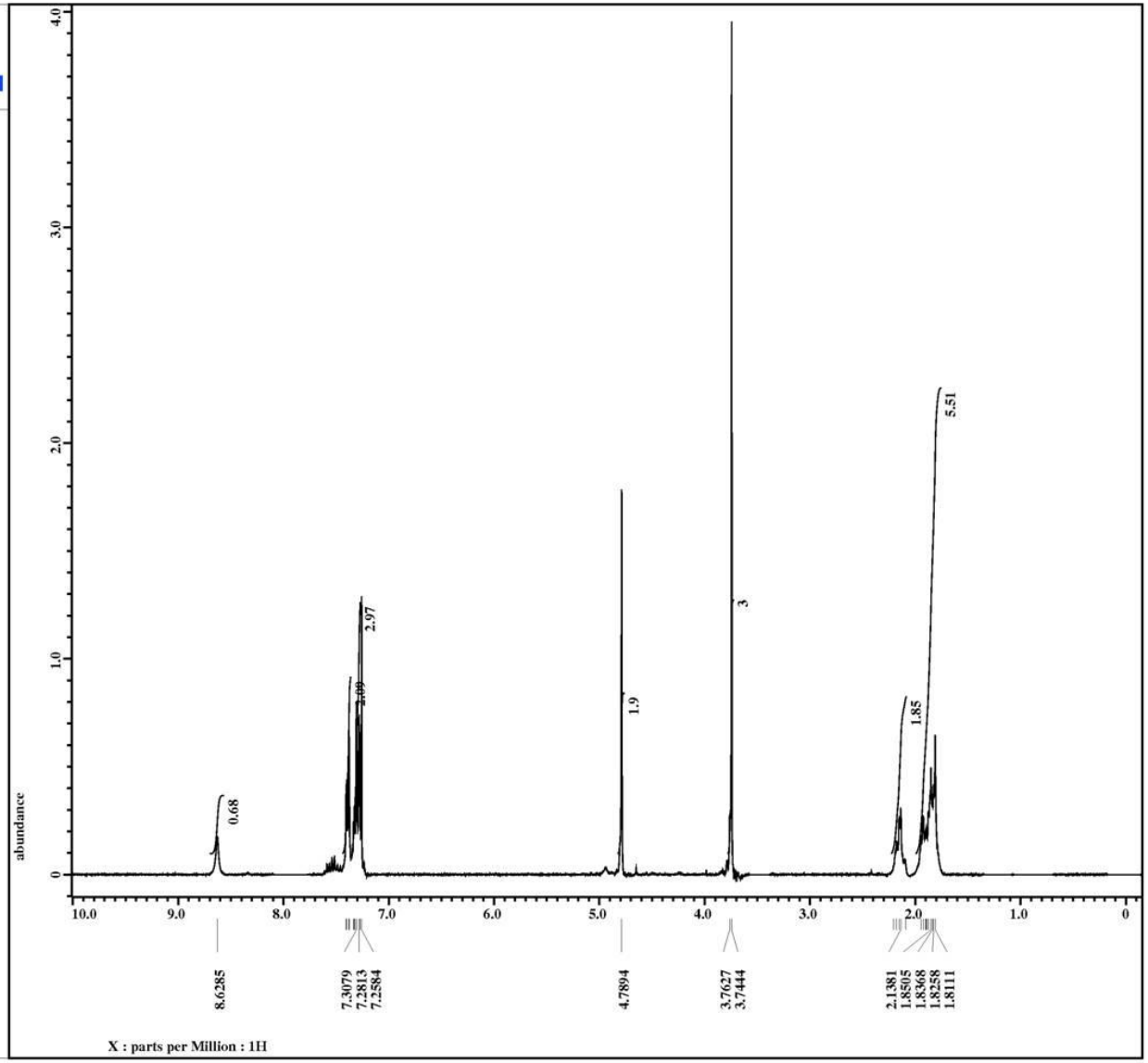
APPENDIX 24

^{1}H and ^{13}C NMR Spectra of

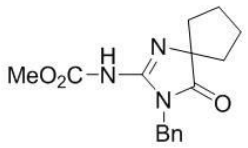
1-Benzyl-2-methylcarbamato-1,3-diazaspiro[4,4]non-1-en-4-one (**106d**)

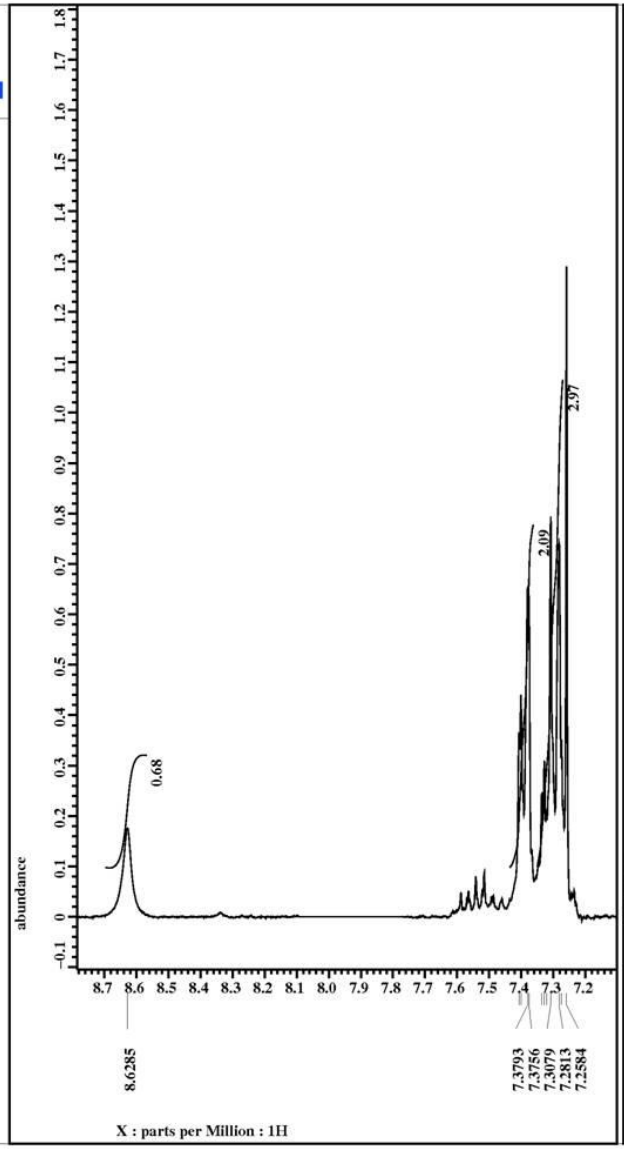
Filename	= III_p_300_spiro-4.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#734931
Solvent	= CHLOROFORM-D
Creation_time	= 26-APR-2008 20:38:36
Revision_time	= 16-MAR-2010 00:24:56
Current_time	= 16-MAR-2010 00:26:23
Comment	= single_pulse
Data_format	= 1D REAL
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.2[dC]

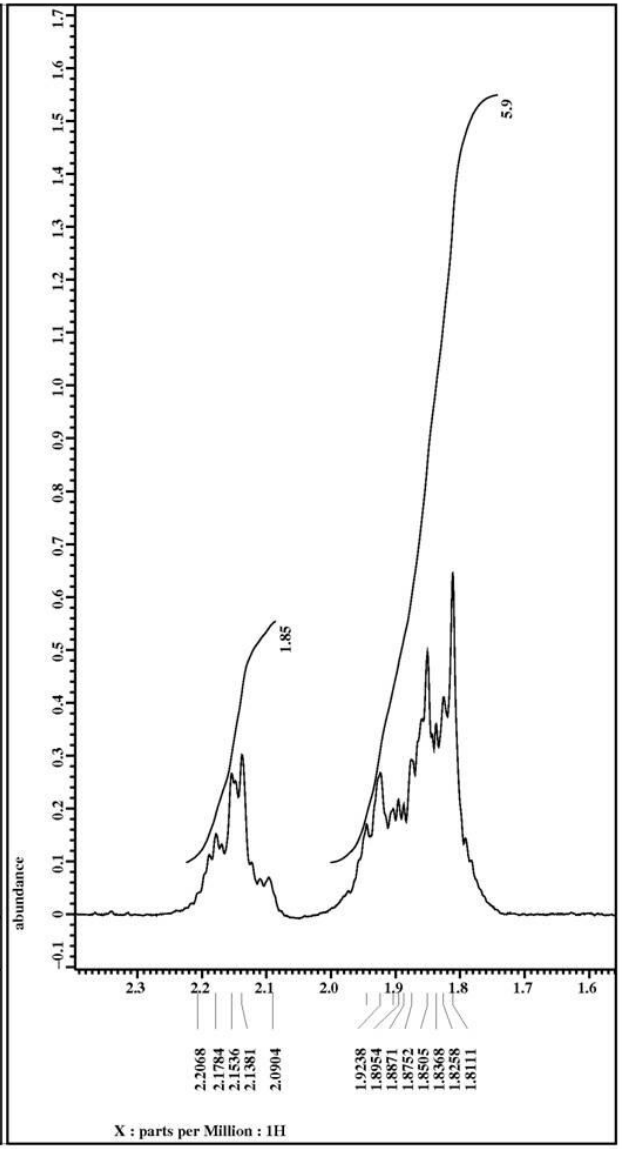


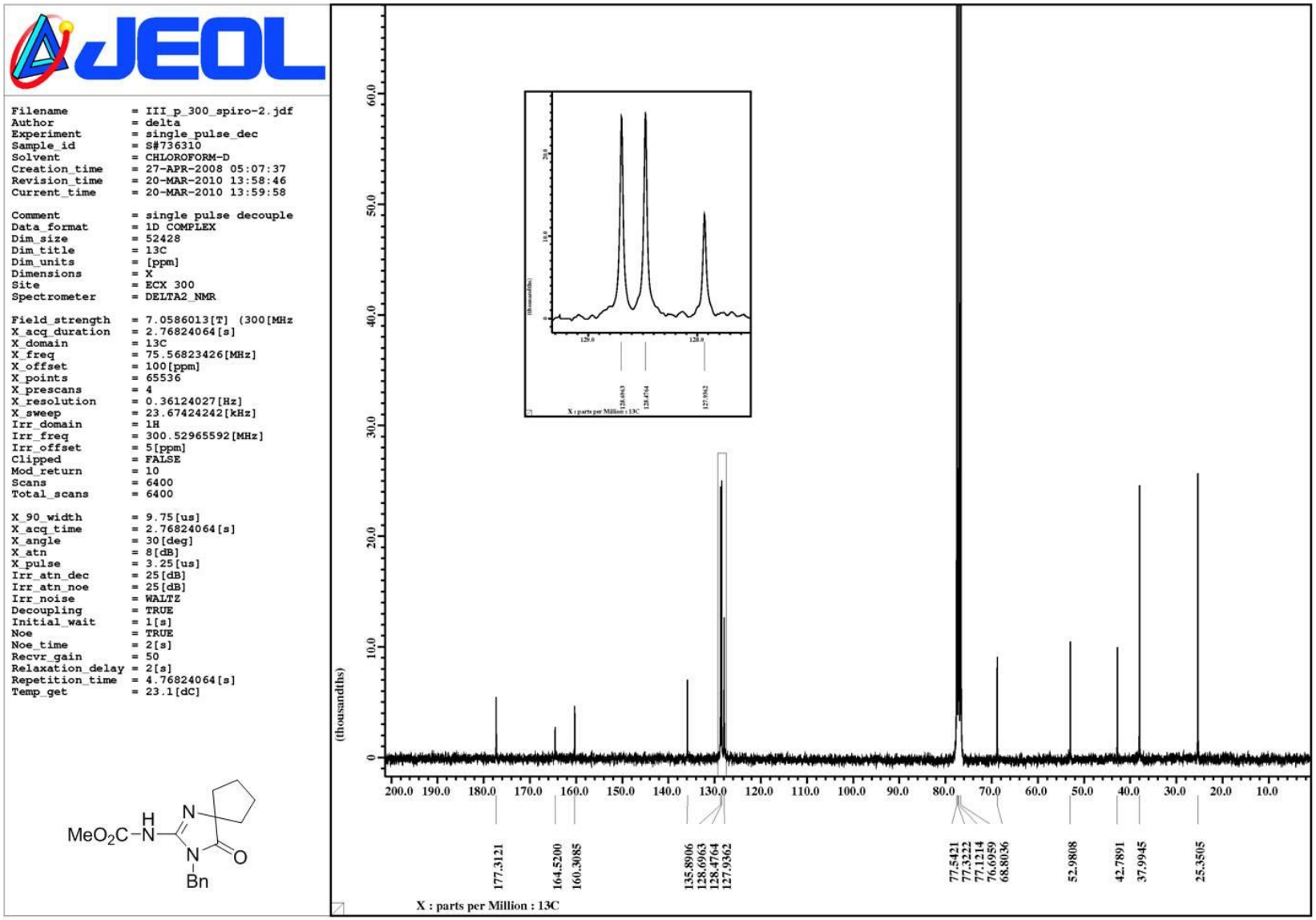


```
Filename          = III_p_300_spiro-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#734931
Solvent           = CHLOROFORM-D
Creation_time     = 26-APR-2008 20:38:36
Revision_time     = 16-MAR-2010 00:27:04
Current_time      = 16-MAR-2010 00:27:21

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.2[dC]
```









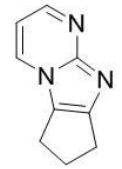
```
Filename          = III_p_300_spiro-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#736310
Solvent           = CHLOROFORM-D
Creation_time     = 27-APR-2008 05:07:37
Revision_time     = 20-MAR-2010 13:58:46
Current_time      = 20-MAR-2010 13:59:58

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.1[dC]
```

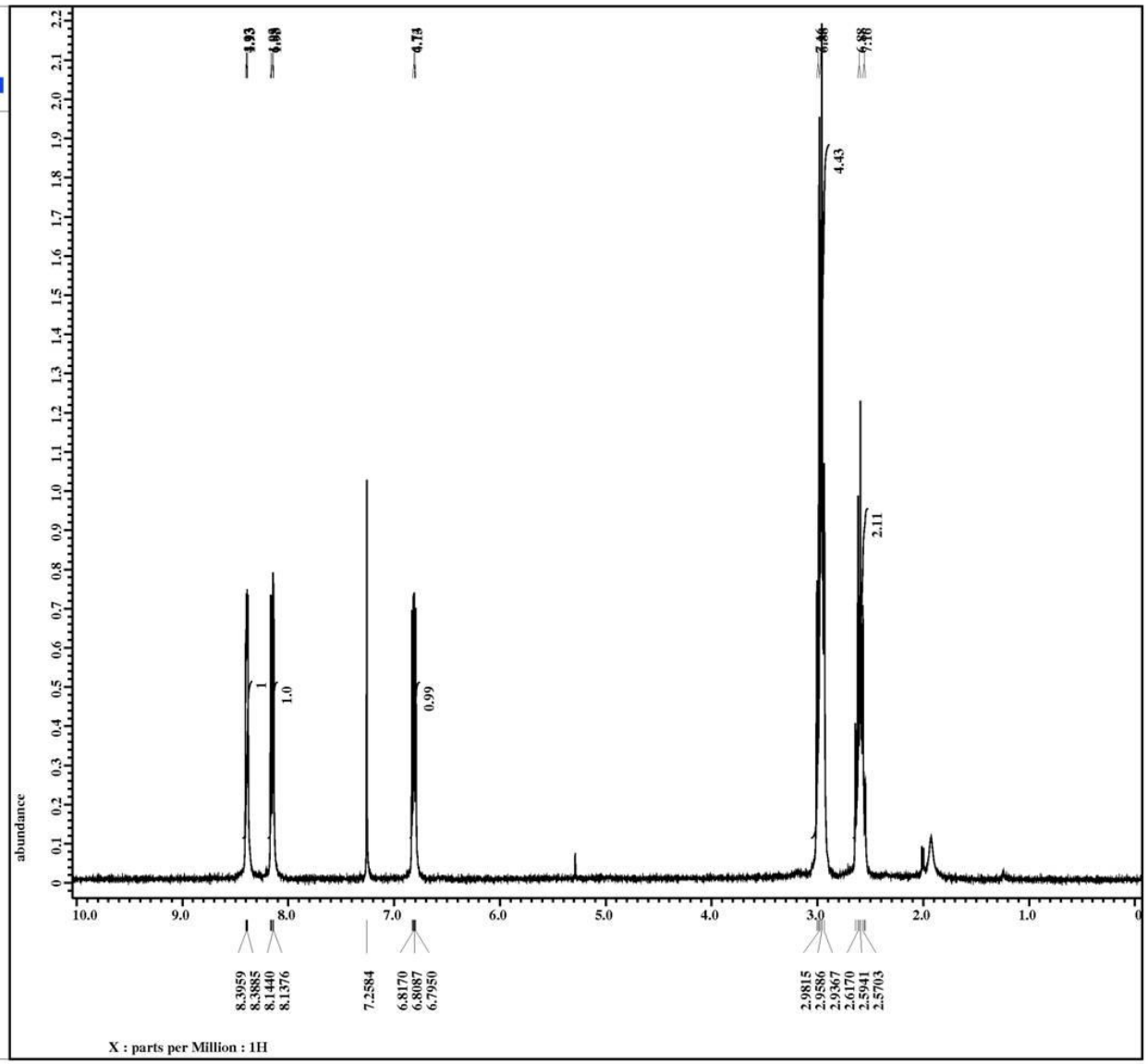
APPENDIX 25

^{1}H and ^{13}C NMR Spectra of

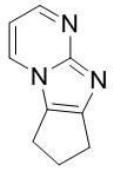
4,5,6-trihydrocyclopentaimidazo[1,2-a]pyrimidine (**134**)

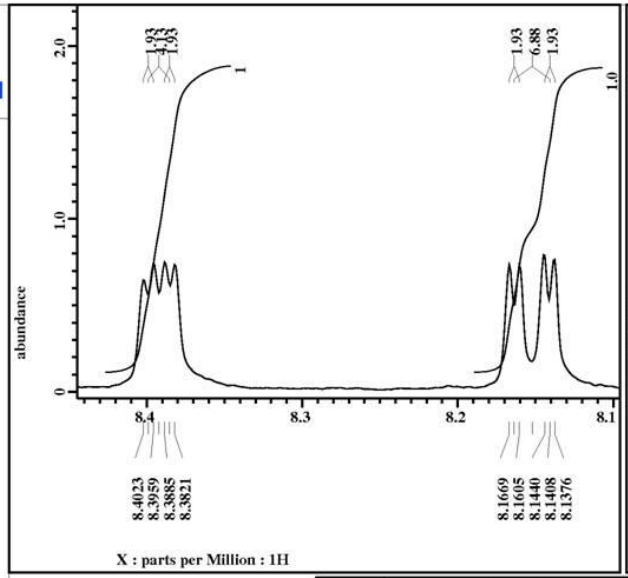
```
Filename          = II_p_149_seconstep-2.
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#385799
Solvent           = CHLOROFORM-D
Creation_time     = 24-JAN-2007 11:28:50
Revision_time     = 15-MAR-2010 13:15:00
Current_time      = 15-MAR-2010 13:15:13

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.8[dC]
```

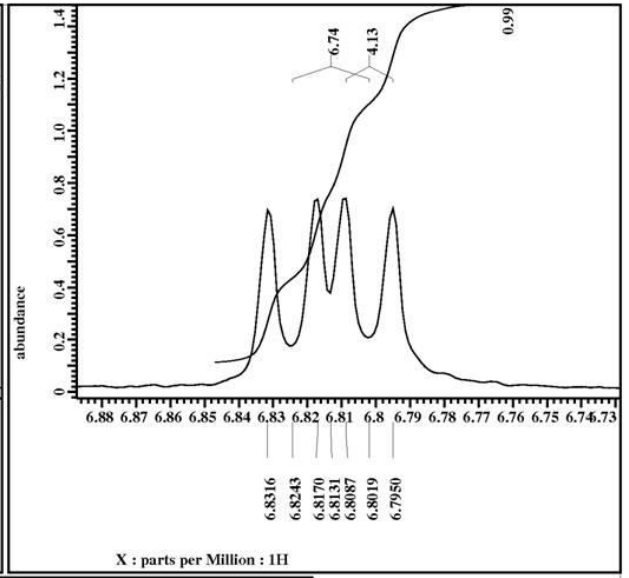


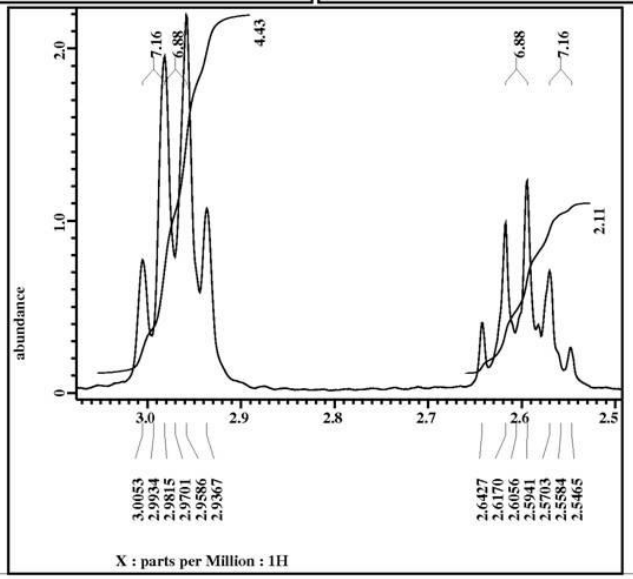


Filename	= II_p_149_seconstep-2.
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#385799
Solvent	= CHLOROFORM-D
Creation_time	= 24-JAN-2007 11:28:50
Revision_time	= 15-MAR-2010 13:15:15
Current_time	= 15-MAR-2010 13:16:08
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 50
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.8[dC]

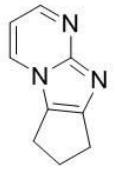


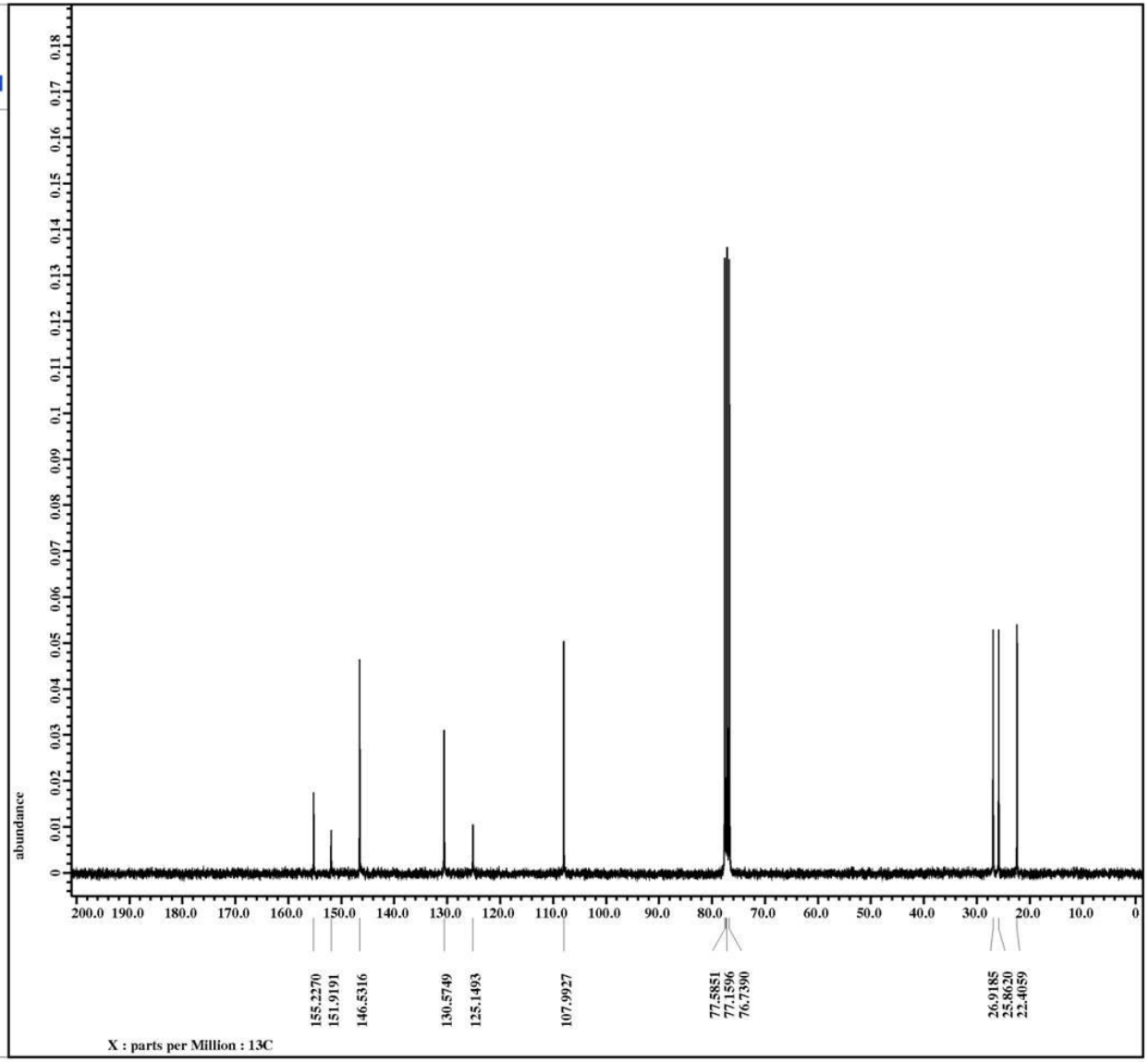






Filename	= II_p_149_product-2.jd
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#87712
Solvent	= CHLOROFORM-D
Creation_time	= 25-JAN-2007 09:26:20
Revision_time	= 16-MAR-2010 13:56:44
Current_time	= 16-MAR-2010 13:57:06
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 4729
Total_scans	= 4729
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 23.5[dC]



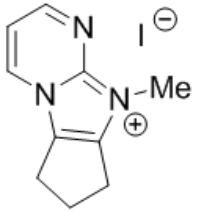


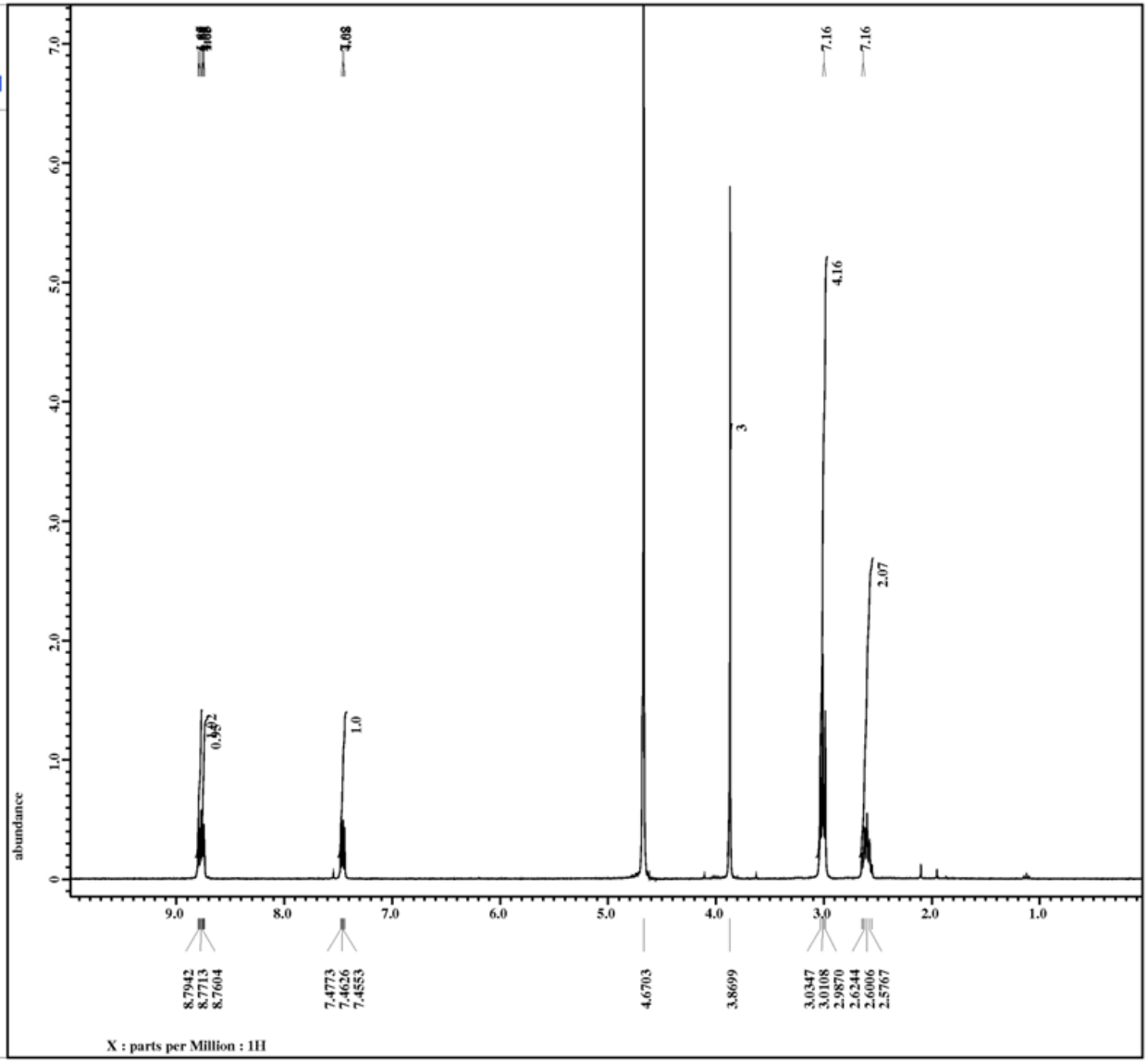
APPENDIX 26

^{1}H and ^{13}C NMR Spectra of

1-Methyl-4,5,6-trihydrocyclopentaimidazolium[1,2-*a*]pyrimidine (**135**)

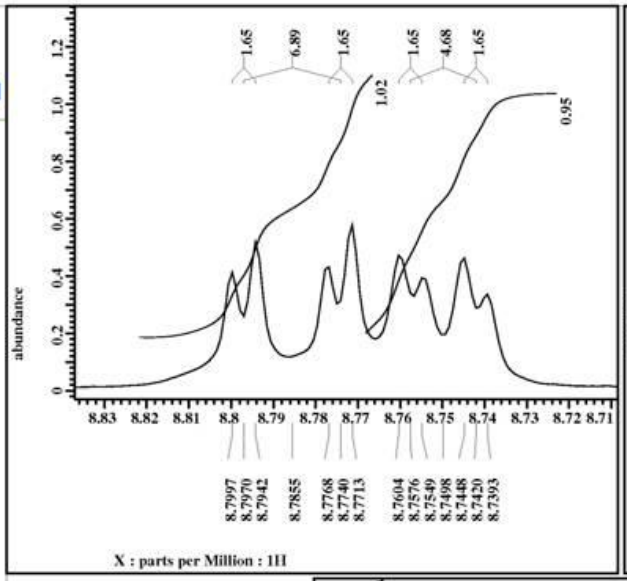
```
Filename          = II_p_162_D2O-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#513685
Solvent           = D2O
Creation_time     =  7-MAR-2007 14:43:31
Revision_time     = 15-MAR-2010 14:16:15
Current_time      = 15-MAR-2010 14:16:26

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

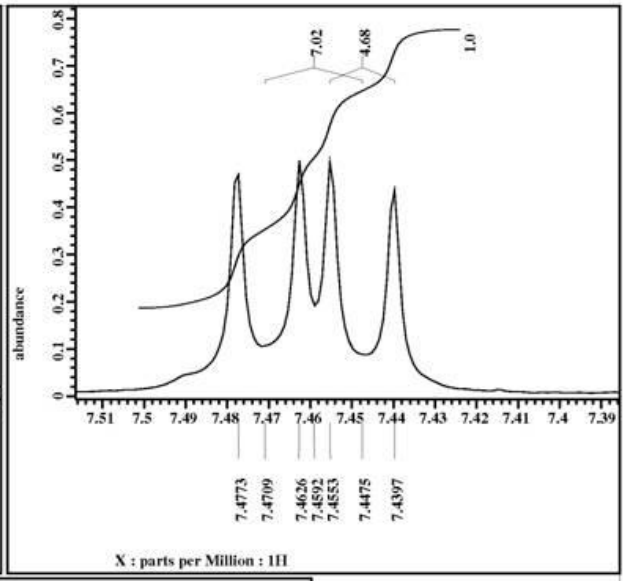


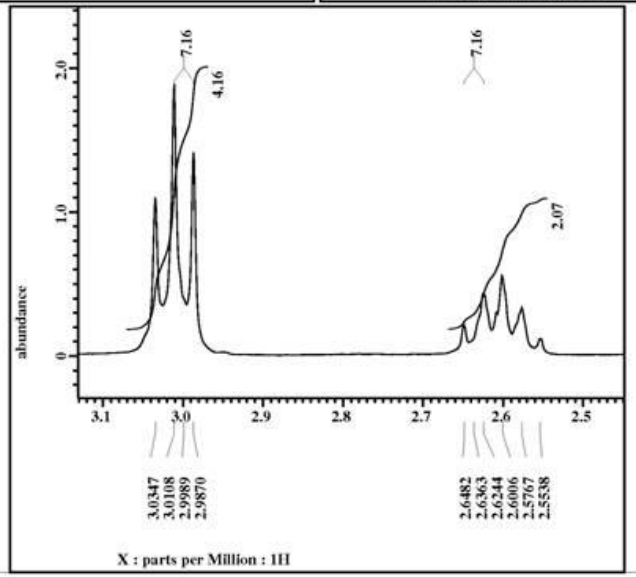


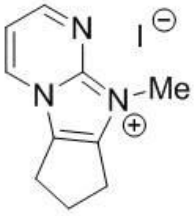
JEOL

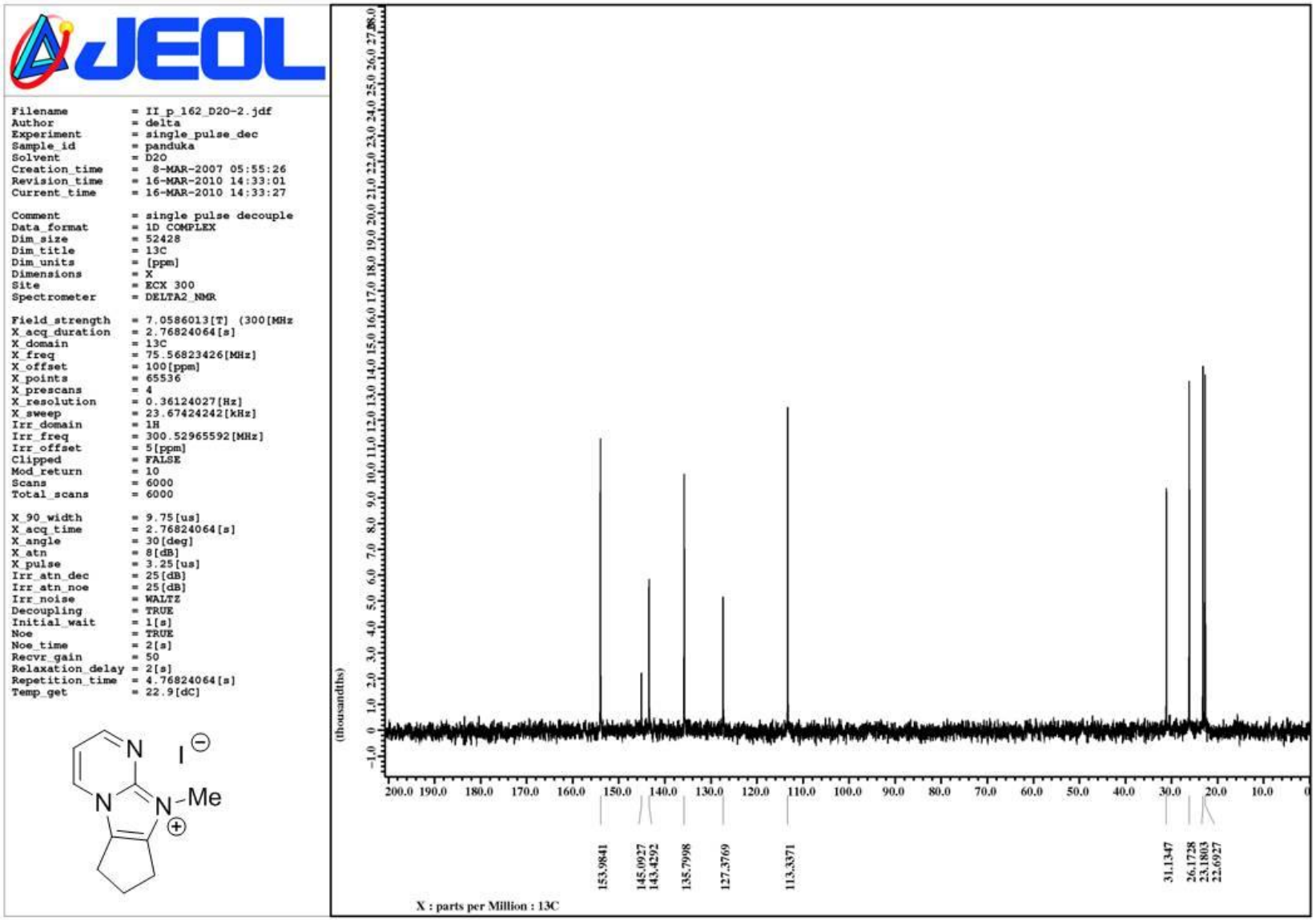
```
Filename          = II_p_162_D2O-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#513685
Solvent           = D2O
Creation_time     =  7-MAR-2007 14:43:31
Revision_time     = 15-MAR-2010 14:16:49
Current_time      = 15-MAR-2010 14:17:36

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```











```
Filename          = II_p_162_D2O-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = D2O
Creation_time     =  8-MAR-2007 05:55:26
Revision_time     = 16-MAR-2010 14:33:01
Current_time      = 16-MAR-2010 14:33:27

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6000
Total_scans       = 6000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.9[dC]
```

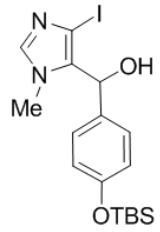
APPENDIX 27

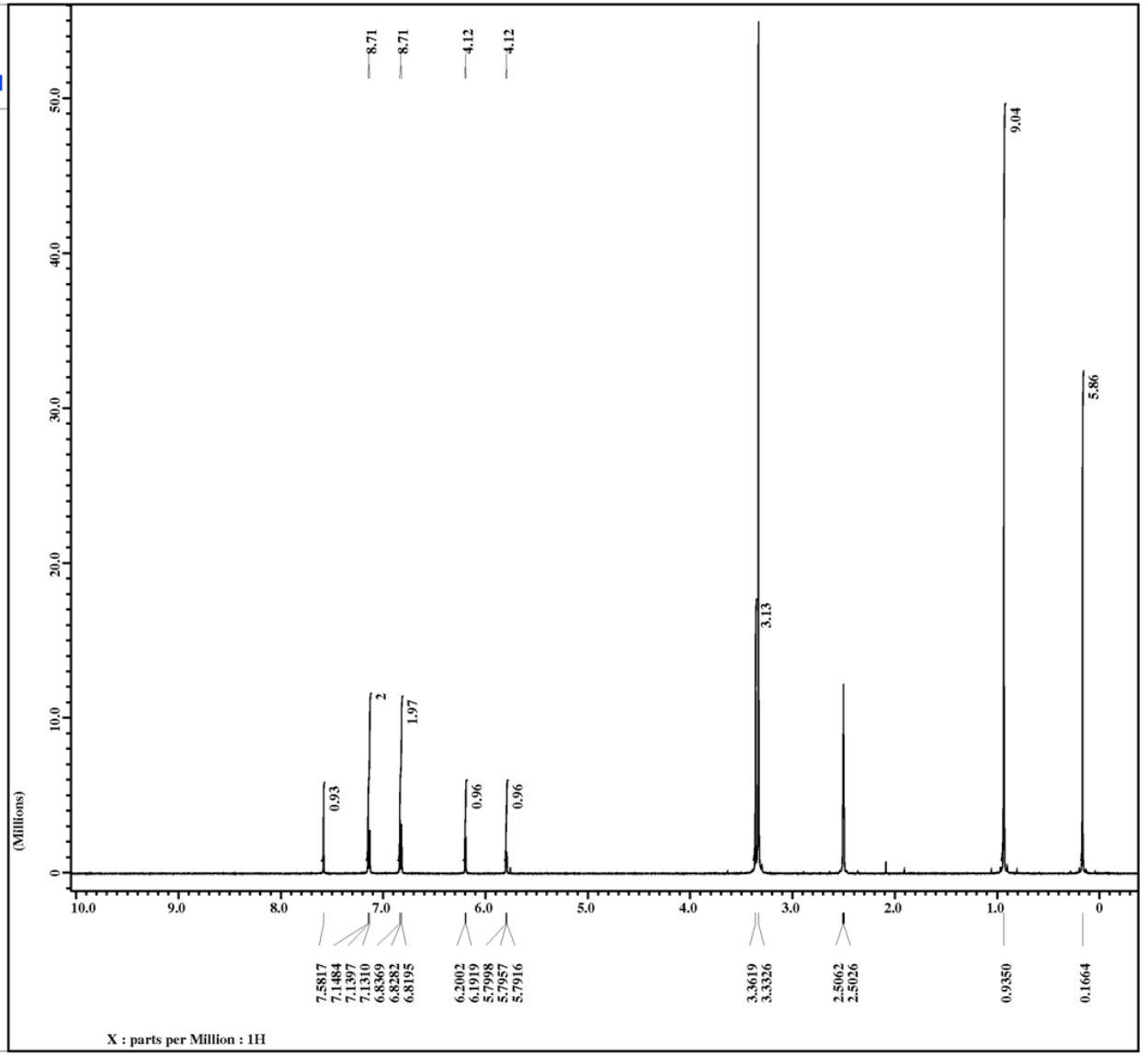
^{1}H and ^{13}C NMR Spectra of

5-(4-*t*-Butyldimethylsiloxyphenyl)hydroxymethyl-4-iodo-1-methyl-*1H*-imidazole

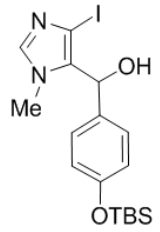
(**147**)

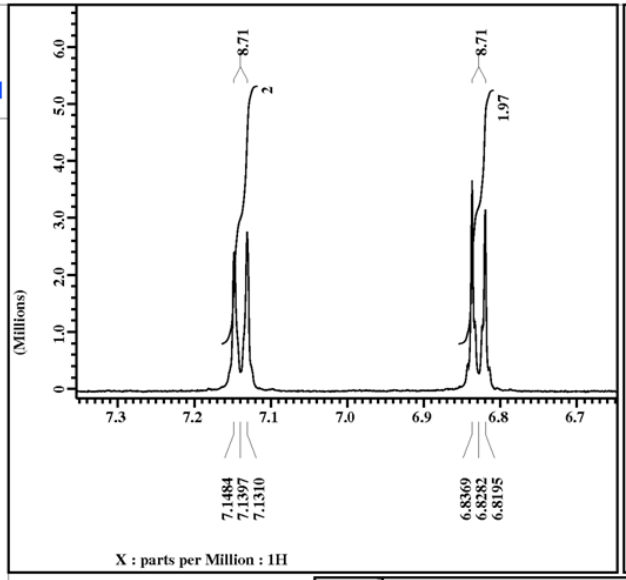
Filename	= II_p_177_I-2.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#644182
Solvent	= DMSO-D6
Creation_time	= 27-MAR-2007 19:53:09
Revision_time	= 16-MAR-2010 14:59:36
Current_time	= 16-MAR-2010 14:59:49
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 20
Relaxation_delay	= 4[s]
Temp_get	= 24.7[dC]
Unblank_time	= 2[us]

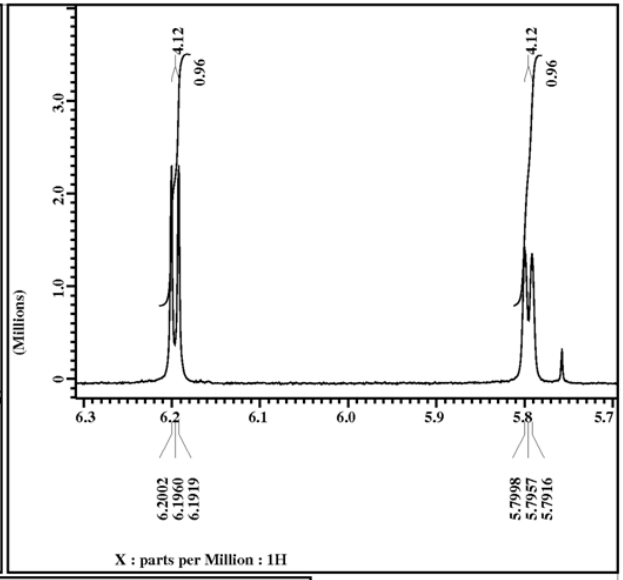


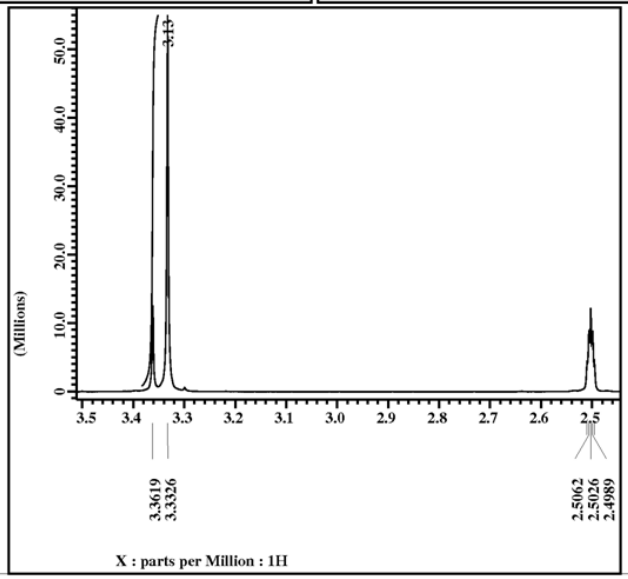


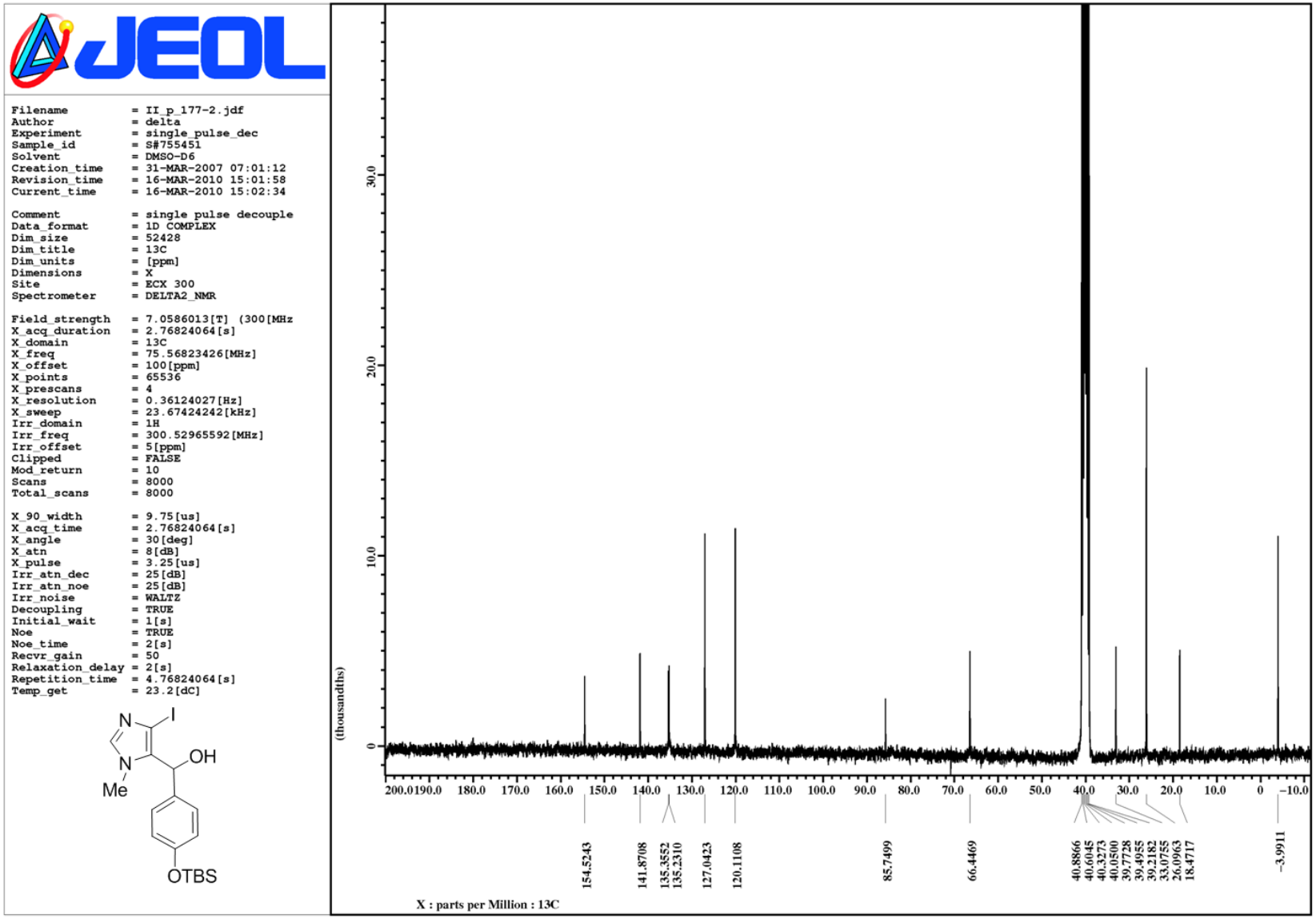
```
Filename          = II_p_177_I-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#644182
Solvent           = DMSO-D6
Creation_time     = 27-MAR-2007 19:53:09
Revision_time     = 16-MAR-2010 14:59:36
Current_time      = 16-MAR-2010 15:00:50

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 24.7[dC]
Unblank_time      = 2[us]
```











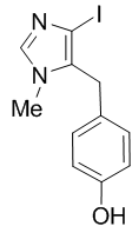
```
Filename          = II_p_177-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#755451
Solvent           = DMSO-D6
Creation_time     = 31-MAR-2007 07:01:12
Revision_time     = 16-MAR-2010 15:01:58
Current_time      = 16-MAR-2010 15:02:34

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8000
Total_scans       = 8000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.2[dC]
```

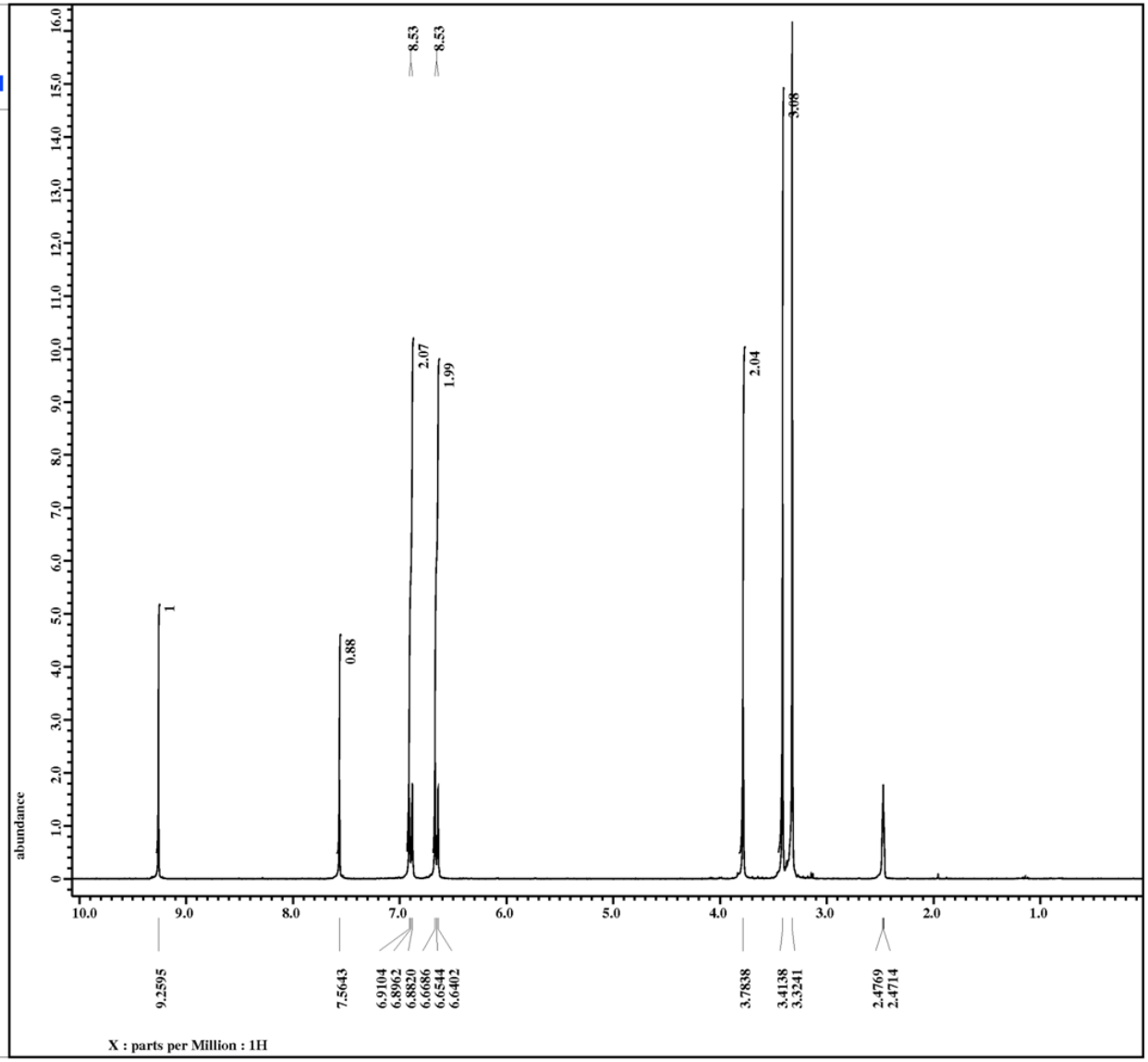
APPENDIX 28

^{1}H and ^{13}C NMR Spectra of

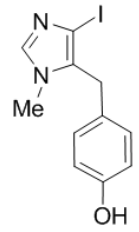
5-(4-hydroxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (**148**)

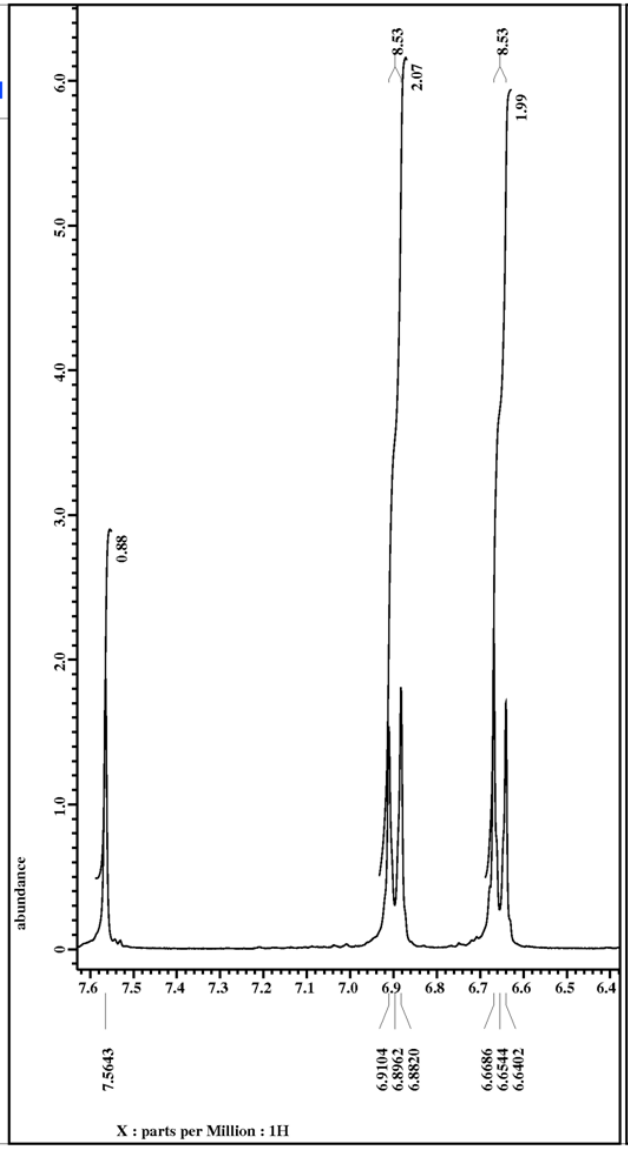
```
Filename          = II_p_225_pure_DMSO-3.
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#793137
Solvent           = DMSO-D6
Creation_time     =  5-AUG-2007 22:30:57
Revision_time     = 16-MAR-2010 15:18:02
Current_time      = 16-MAR-2010 15:18:35

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.8[dC]
```

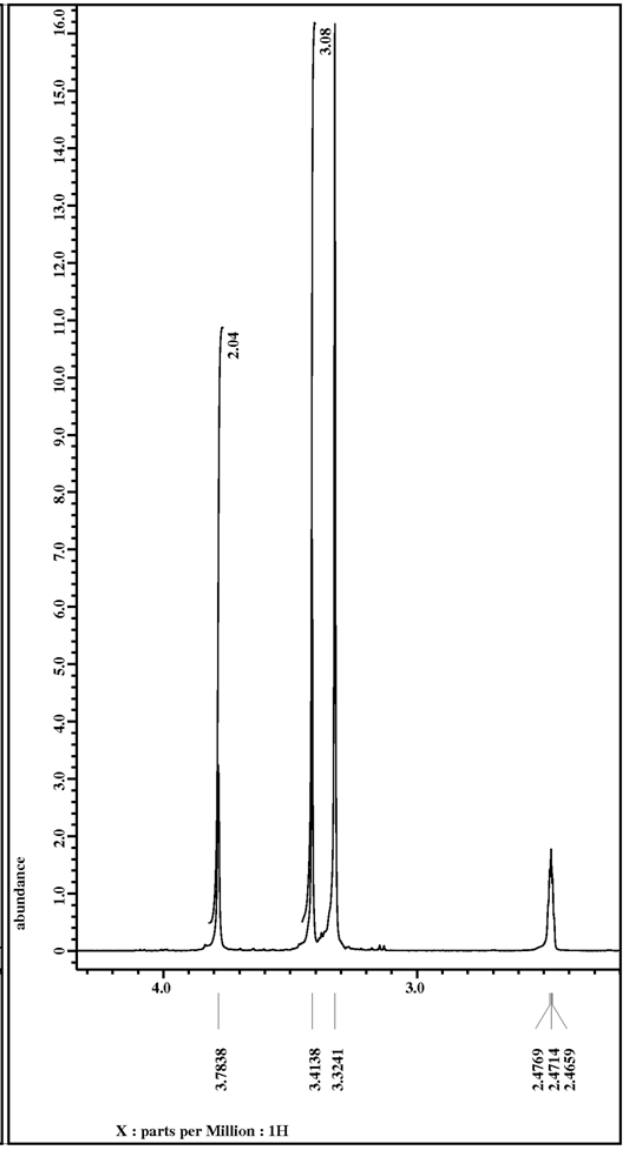




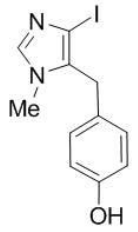
```
Filename          = II_p_225_pure_DMSO-3.
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#793137
Solvent           = DMSO-D6
Creation_time     =  5-AUG-2007 22:30:57
Revision_time     = 16-MAR-2010 15:18:02
Current_time      = 16-MAR-2010 15:19:09

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.8[dC]
```

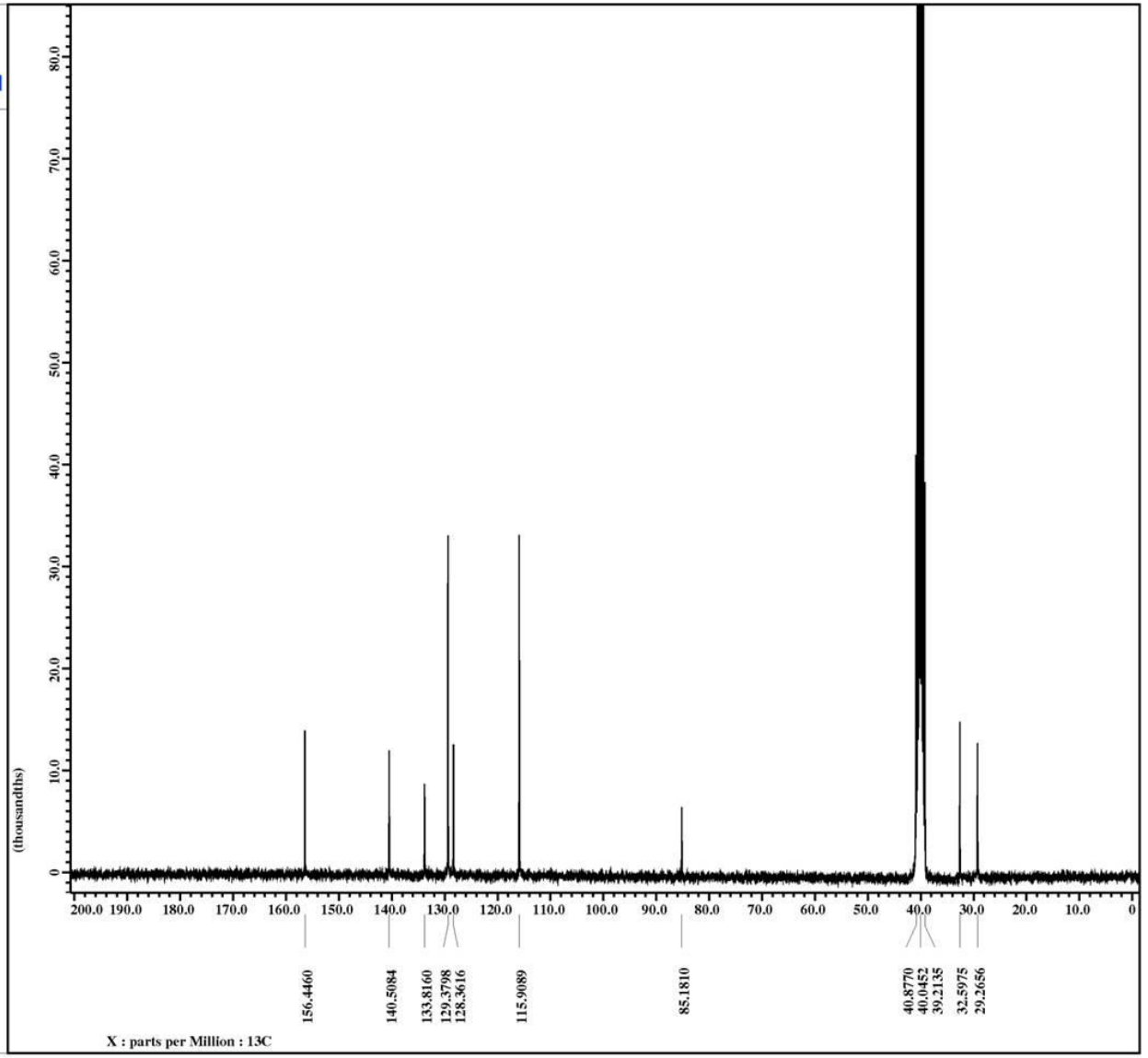






```
Filename          = II_p_225_pure_DMSO-3.
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka_225
Solvent           = DMSO-D6
Creation_time     =  6-AUG-2007 08:29:24
Revision_time     = 16-MAR-2010 15:20:52
Current_time      = 16-MAR-2010 15:21:12

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7500
Total_scans       = 7500

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.9[dC]
```



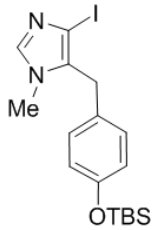


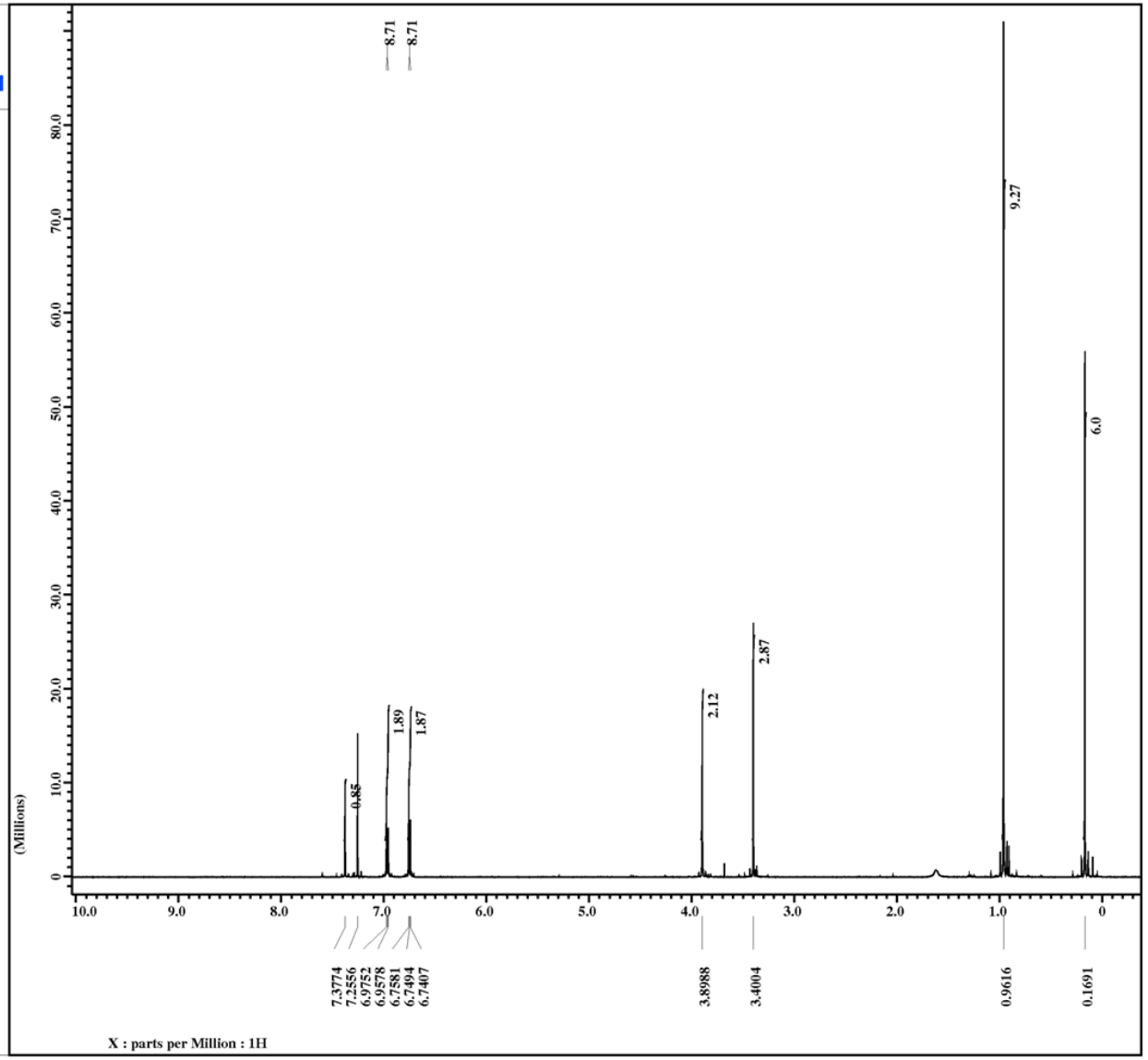
APPENDIX 29

^{1}H and ^{13}C NMR Spectra of

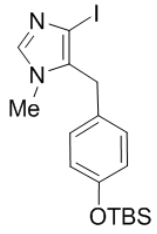
5-(4-*t*-Butyldimethylsilyloxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (**80**)

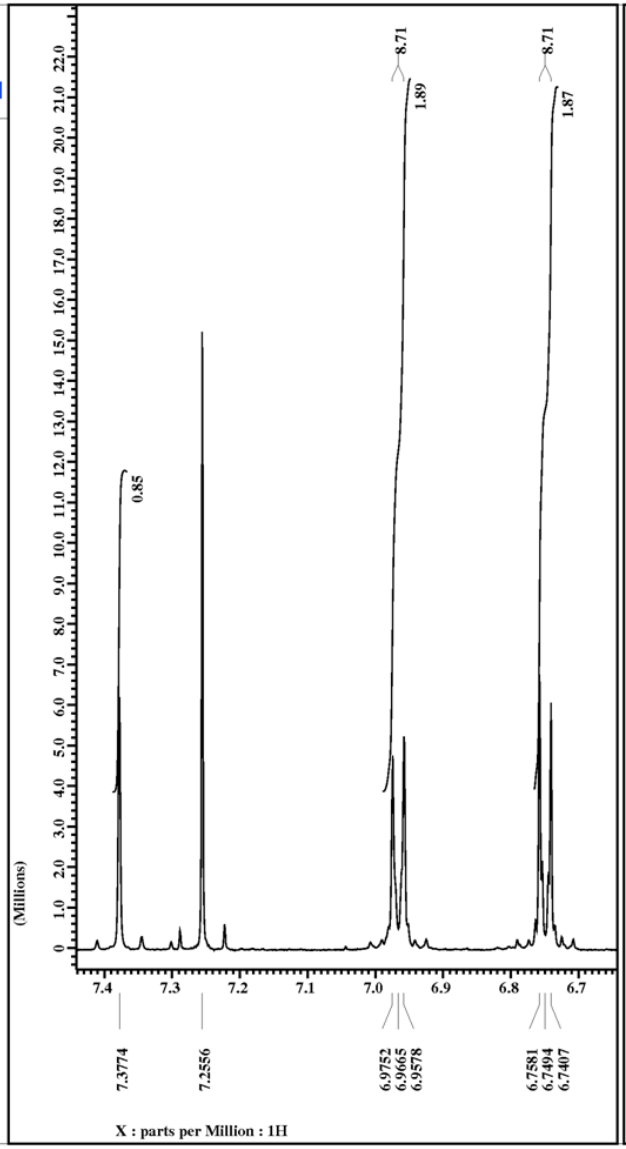
```
Filename          = III_p_079_product-3.j
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#446594
Solvent           = CHLOROFORM-D
Creation_time     =  1-DEC-2007 17:29:15
Revision_time     = 16-MAR-2010 15:38:36
Current_time      = 16-MAR-2010 15:39:13

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 22
Relaxation_delay  = 4[s]
Temp_get          = 25[dC]
Unblank_time      = 2[us]
```

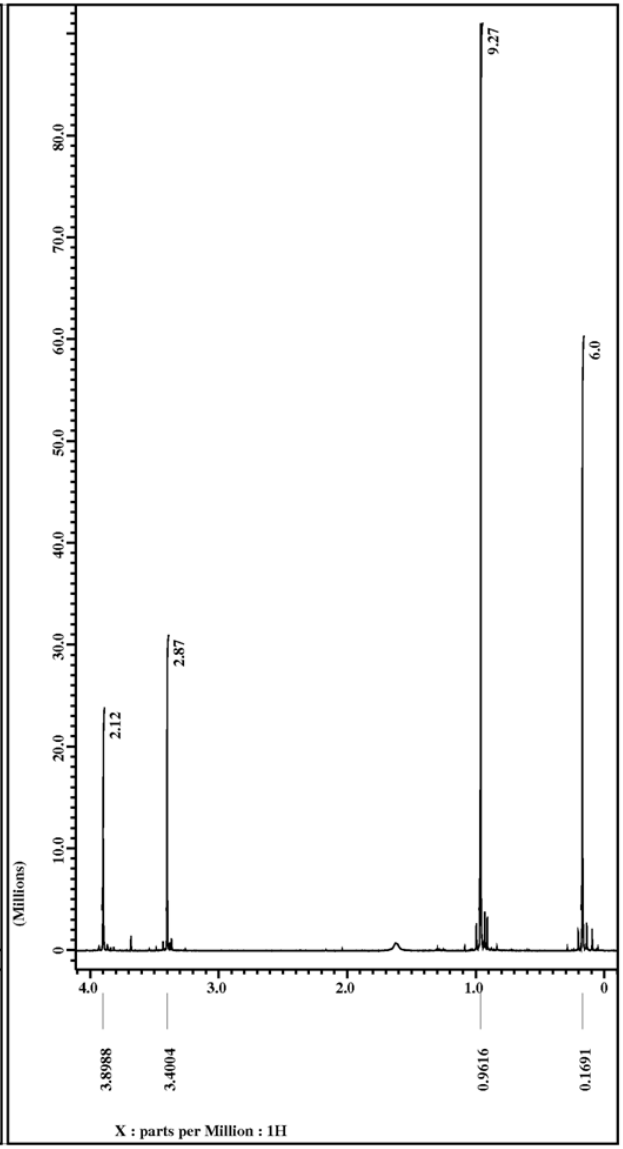




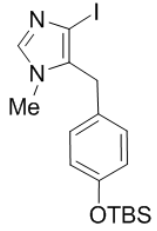
Filename	= III_p_079_product-3.j
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#446594
Solvent	= CHLOROFORM-D
Creation_time	= 1-DEC-2007 17:29:15
Revision_time	= 16-MAR-2010 15:38:36
Current_time	= 16-MAR-2010 15:39:47
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 22
Relaxation_delay	= 4[s]
Temp_get	= 25[dC]
Unblank_time	= 2[us]

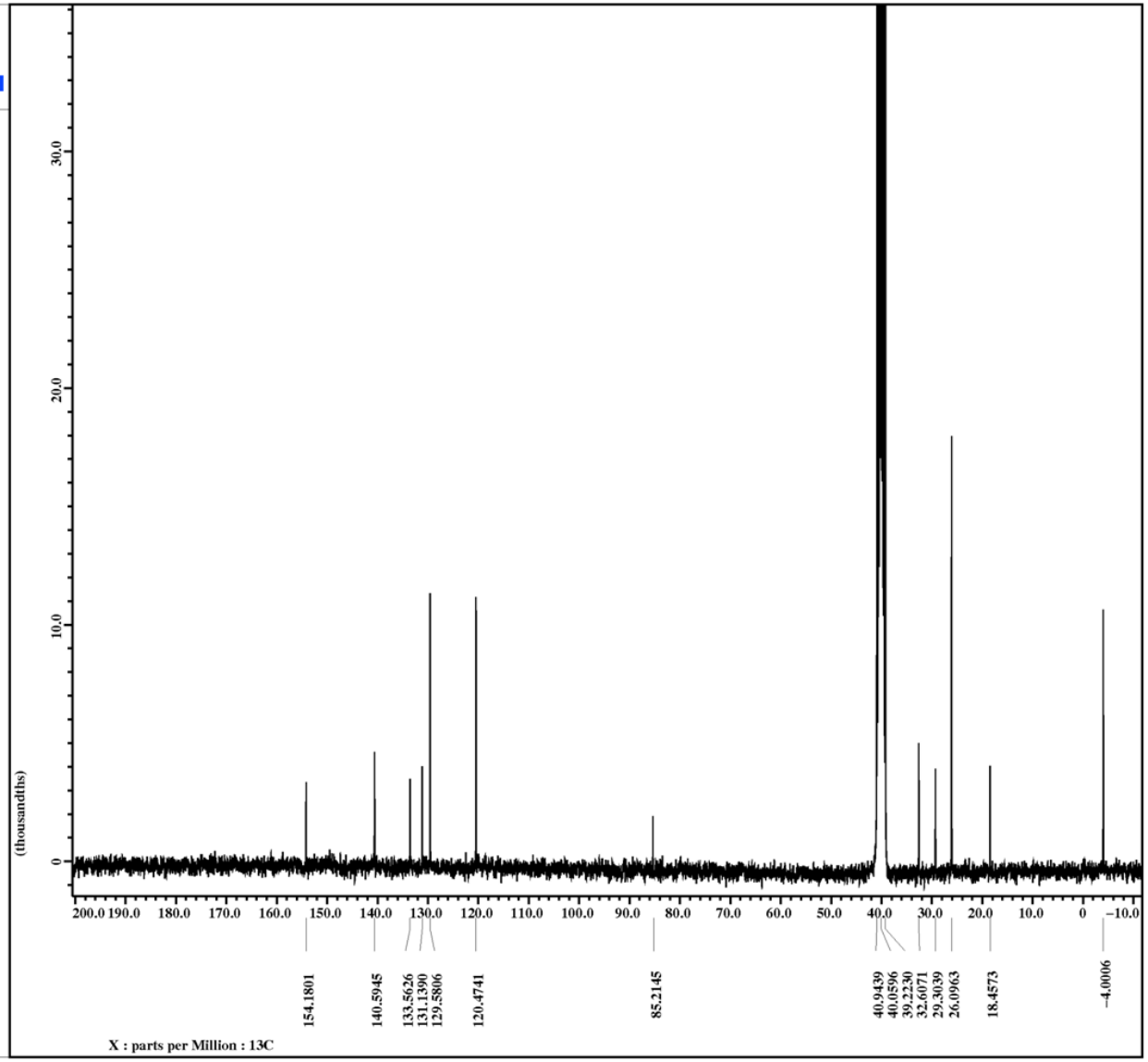






```
Filename          = II_p_218_product-3.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = DMSO-D6
Creation_time     = 14-MAY-2007 06:01:43
Revision_time     = 16-MAR-2010 15:40:48
Current_time      = 16-MAR-2010 15:41:11

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 5000
Total_scans       = 5000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.7[dC]
```



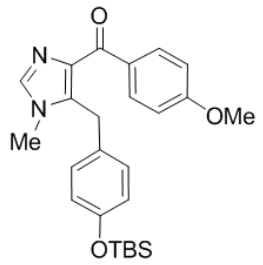


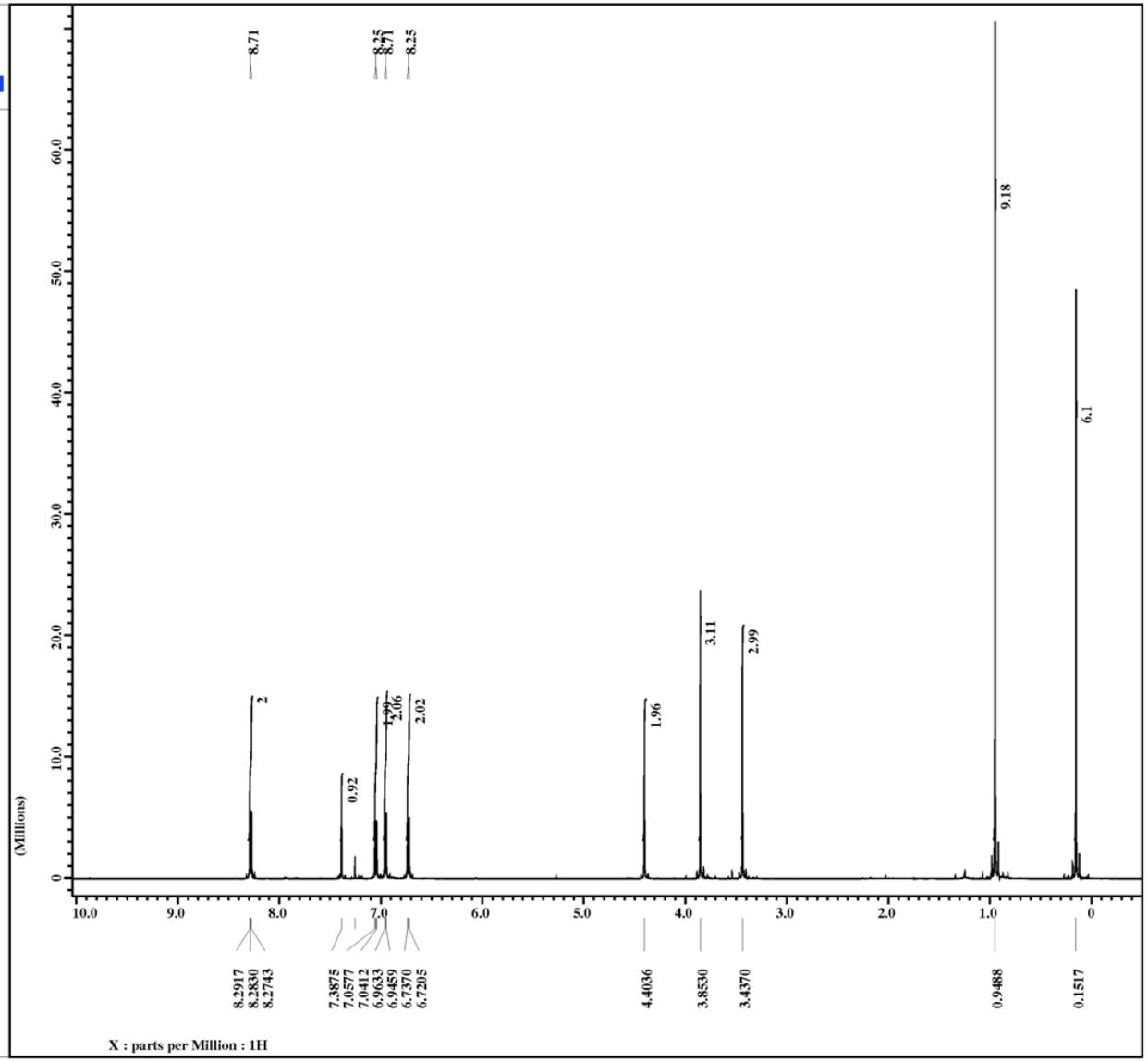
APPENDIX 30

^{1}H and ^{13}C NMR Spectra of

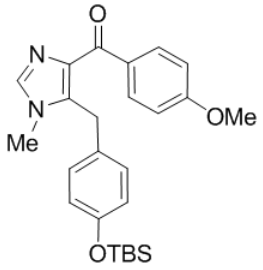
{5-(4-*t*-Butyldimethylsilyloxybenzyl)-1-methyl-1*H*-imidazol-4-yl}-(4-methoxy)phenylmethanone (**149**)

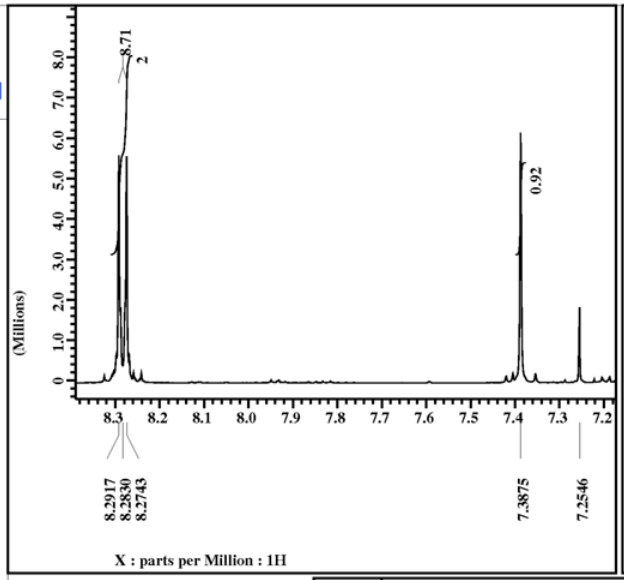
```
Filename          = II_p_310_II-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#579841
Solvent           = CHLOROFORM-D
Creation_time     = 12-OCT-2007 20:41:53
Revision_time     = 16-MAR-2010 15:55:30
Current_time      = 16-MAR-2010 15:55:44

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```

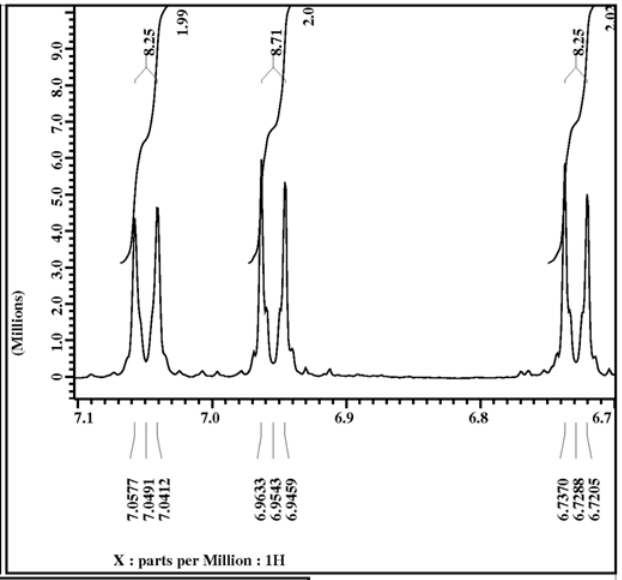


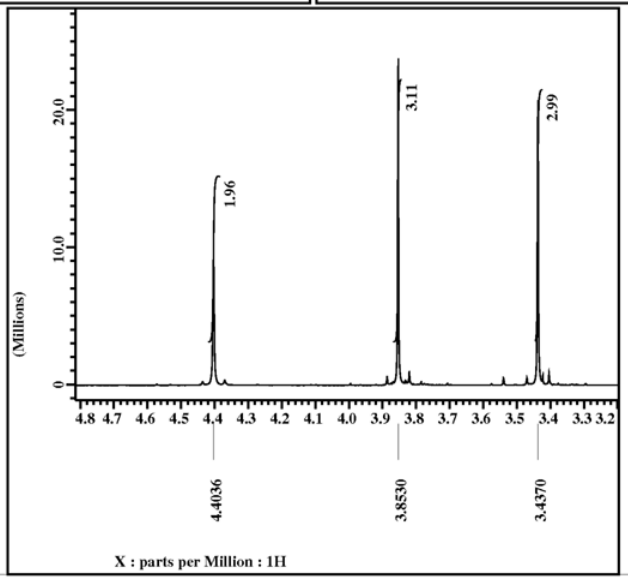


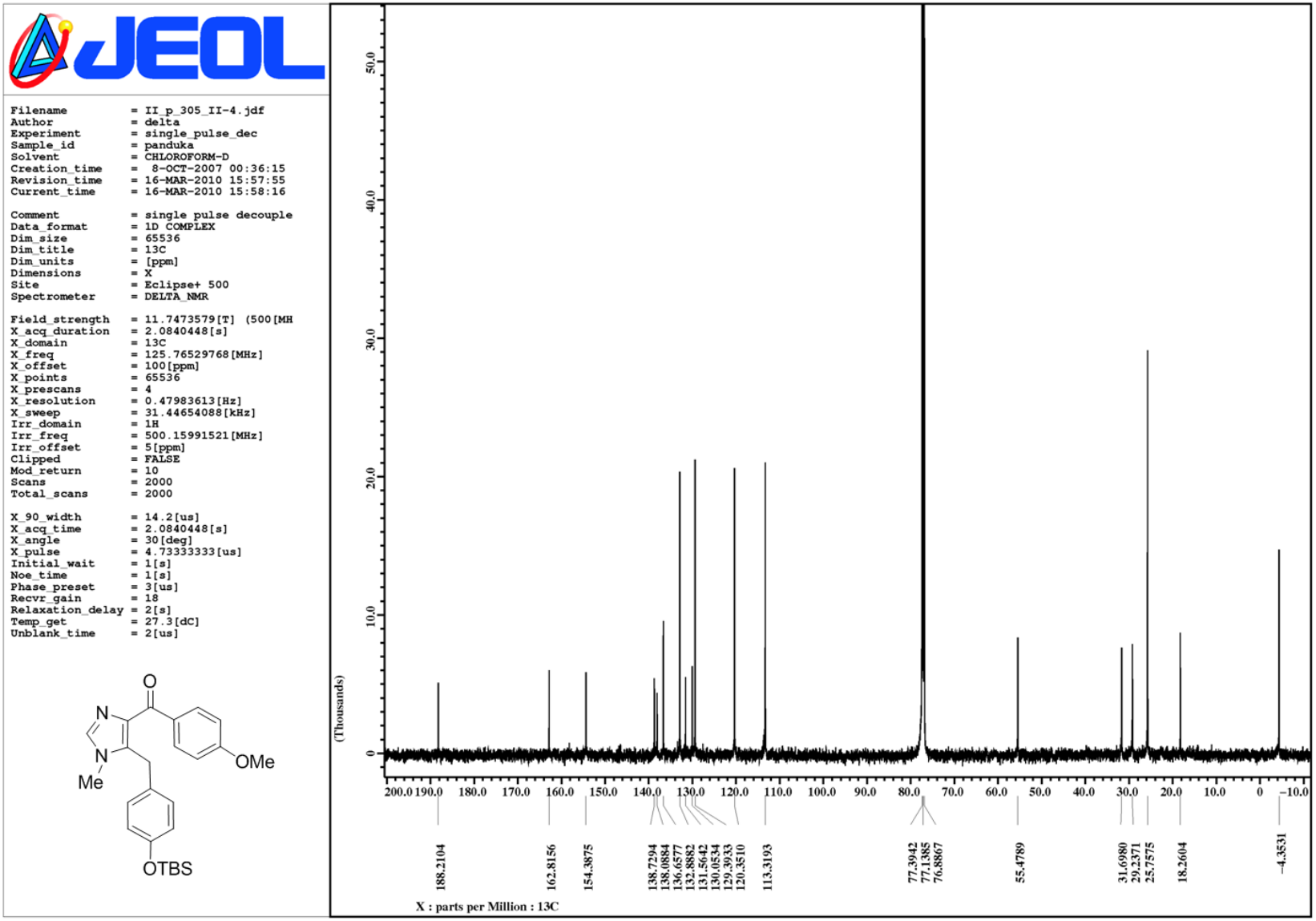
```
Filename          = II_p_310_II-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#579841
Solvent           = CHLOROFORM-D
Creation_time     = 12-OCT-2007 20:41:53
Revision_time     = 16-MAR-2010 15:55:30
Current_time      = 16-MAR-2010 15:57:10

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```











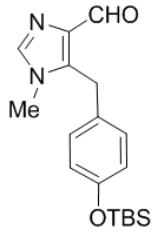
```
Filename          = II_p_305_II-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     =  8-OCT-2007 00:36:15
Revision_time     = 16-MAR-2010 15:57:55
Current_time      = 16-MAR-2010 15:58:16

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 2000
Total_scans       = 2000

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 2[s]
Temp_get          = 27.3[dC]
Unblank_time      = 2[us]
```

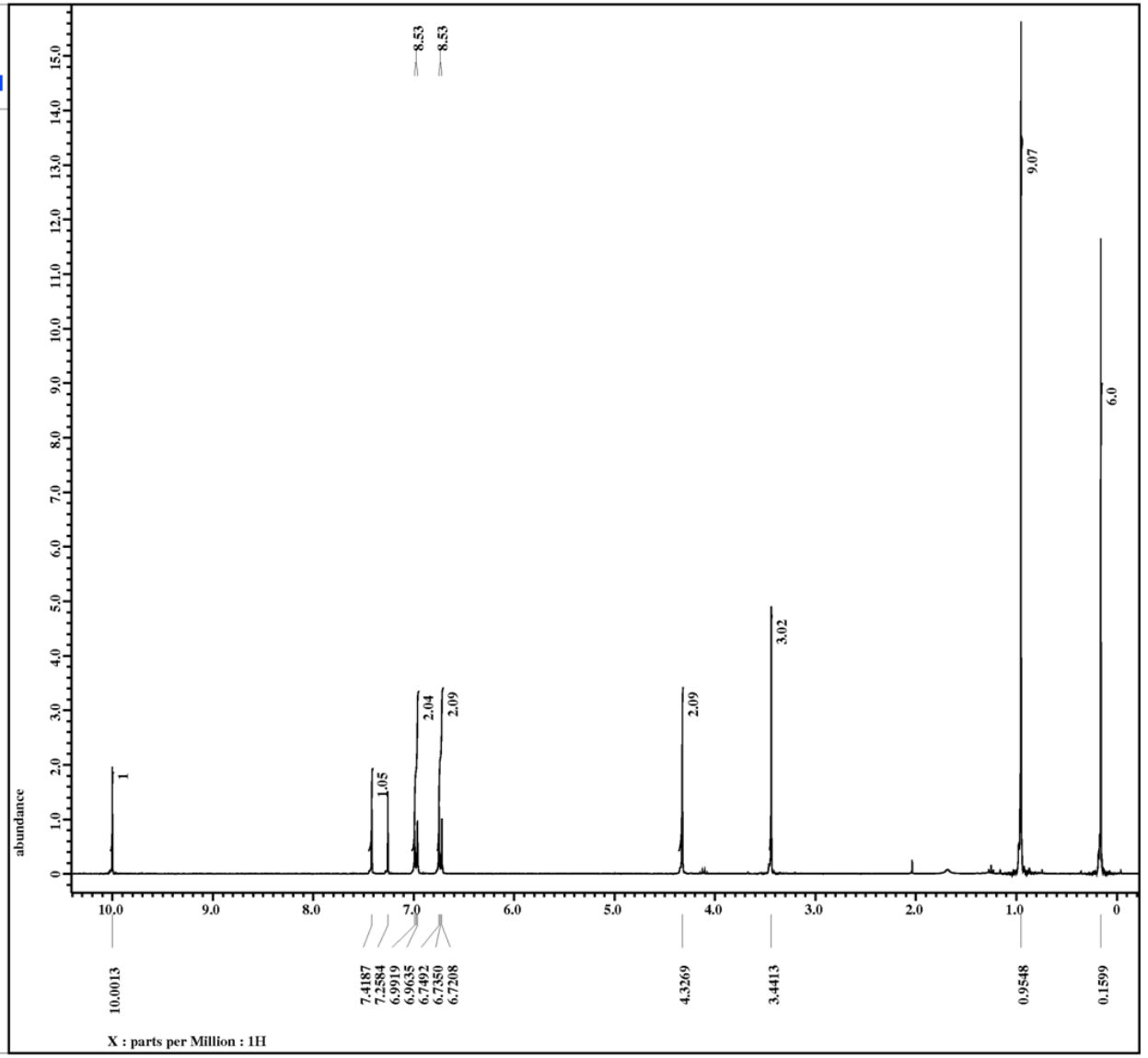
APPENDIX 31

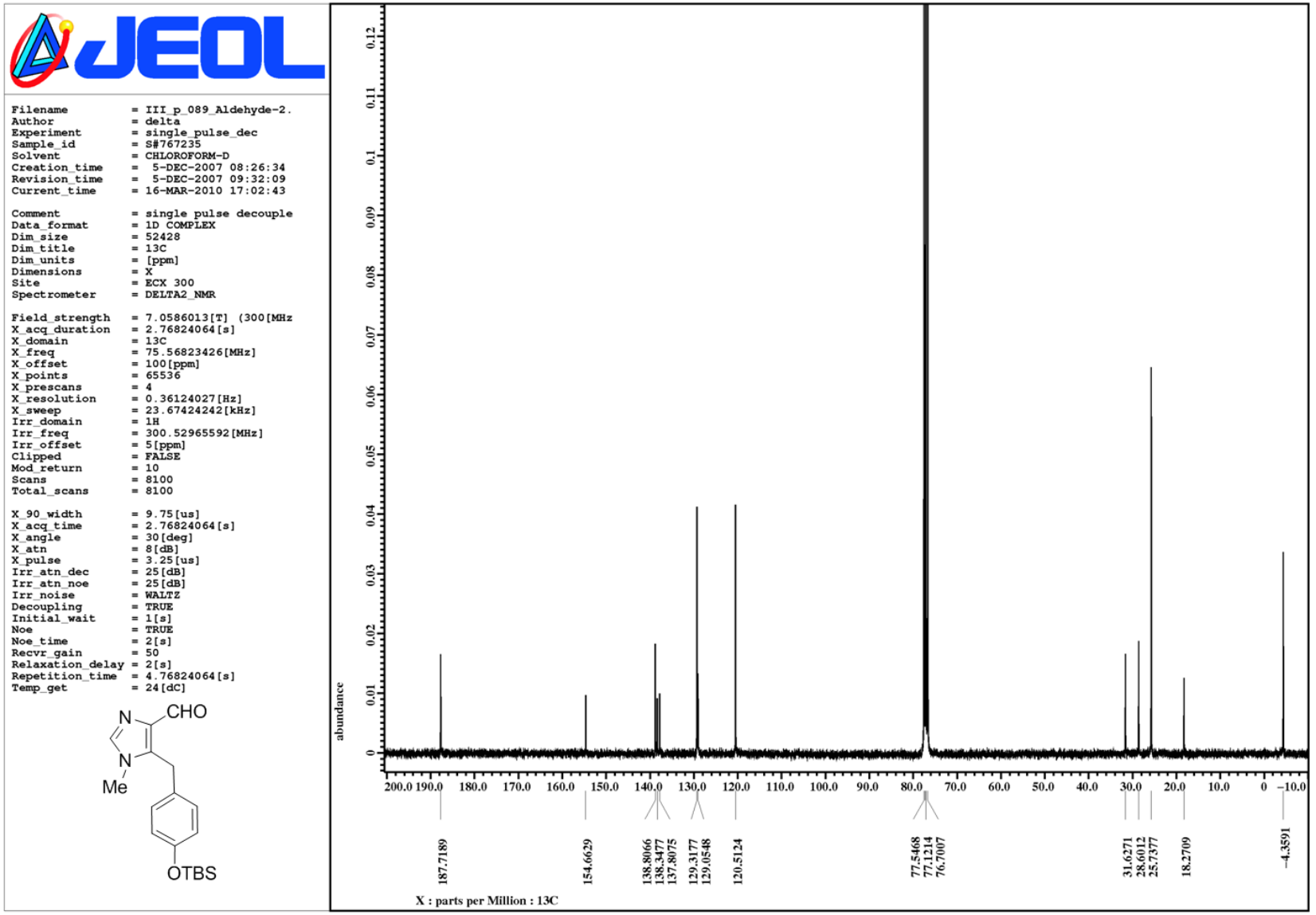
^{1}H and ^{13}C NMR Spectra of

5-(4-*t*-Butyldimethylsilyloxybenzyl)-1-methyl-1*H*-imidazole-4-carbaldehyde (**152**)

```
Filename          = III_p_089_Aldehyde-3.
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#748432
Solvent           = CHLOROFORM-D
Creation_time     =  4-DEC-2007 21:12:14
Revision_time     = 16-MAR-2010 17:01:17
Current_time      = 16-MAR-2010 17:01:33

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.5[dC]
```

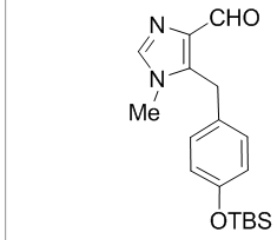


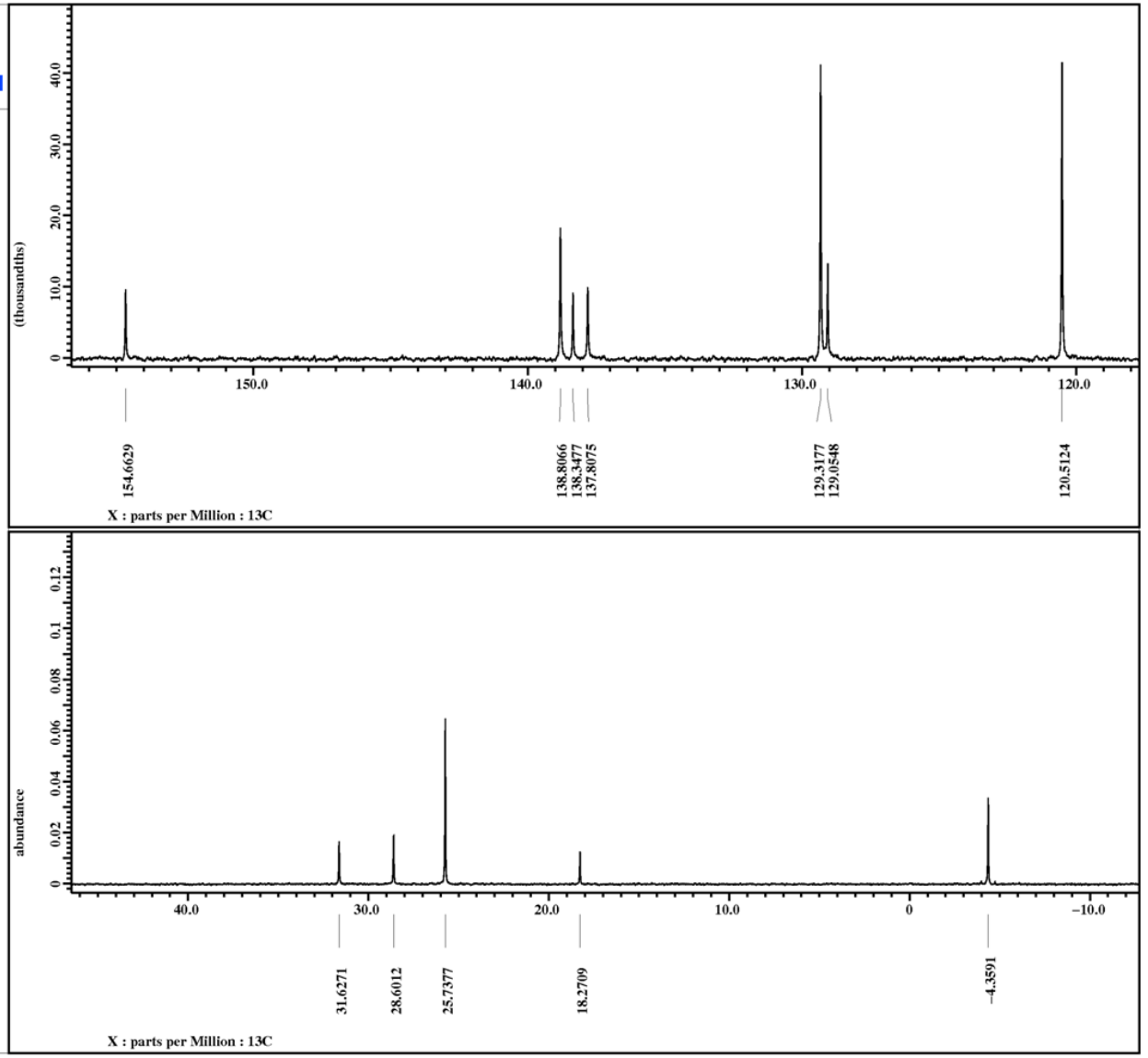




```
Filename          = III_p_089_Aldehyde-2.
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#767235
Solvent           = CHLOROFORM-D
Creation_time     =  5-DEC-2007 08:26:34
Revision_time     =  5-DEC-2007 09:32:09
Current_time      = 16-MAR-2010 17:02:43

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8100
Total_scans       = 8100

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 24[dC]
```

```
Filename          = III_p_089_Aldehyde-2.
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#767235
Solvent           = CHLOROFORM-D
Creation_time     =  5-DEC-2007 08:26:34
Revision_time     =  5-DEC-2007 09:32:09
Current_time      = 16-MAR-2010 17:04:19

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 8100
Total_scans       = 8100

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 24[dC]
```



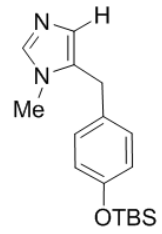


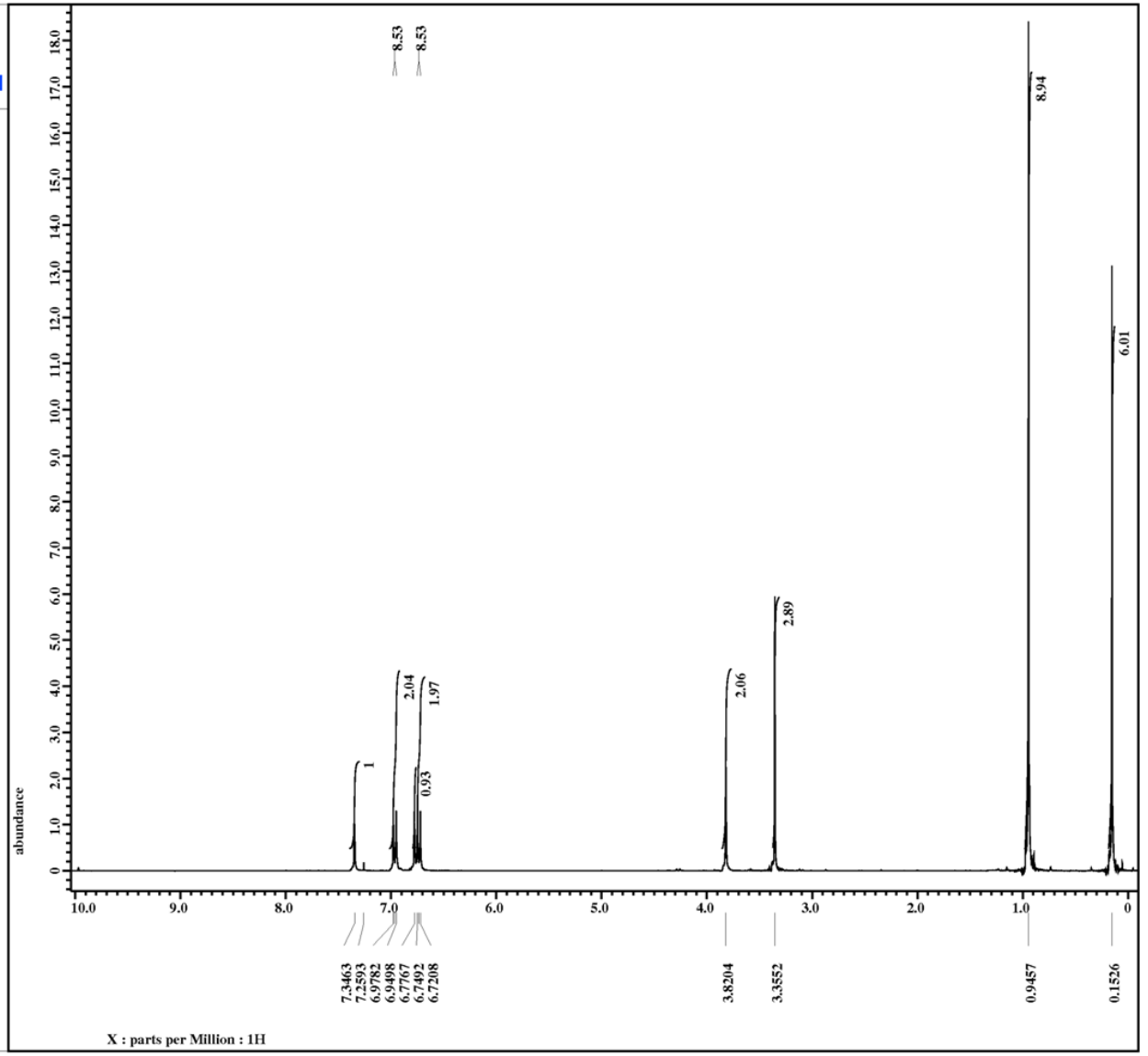
APPENDIX 32

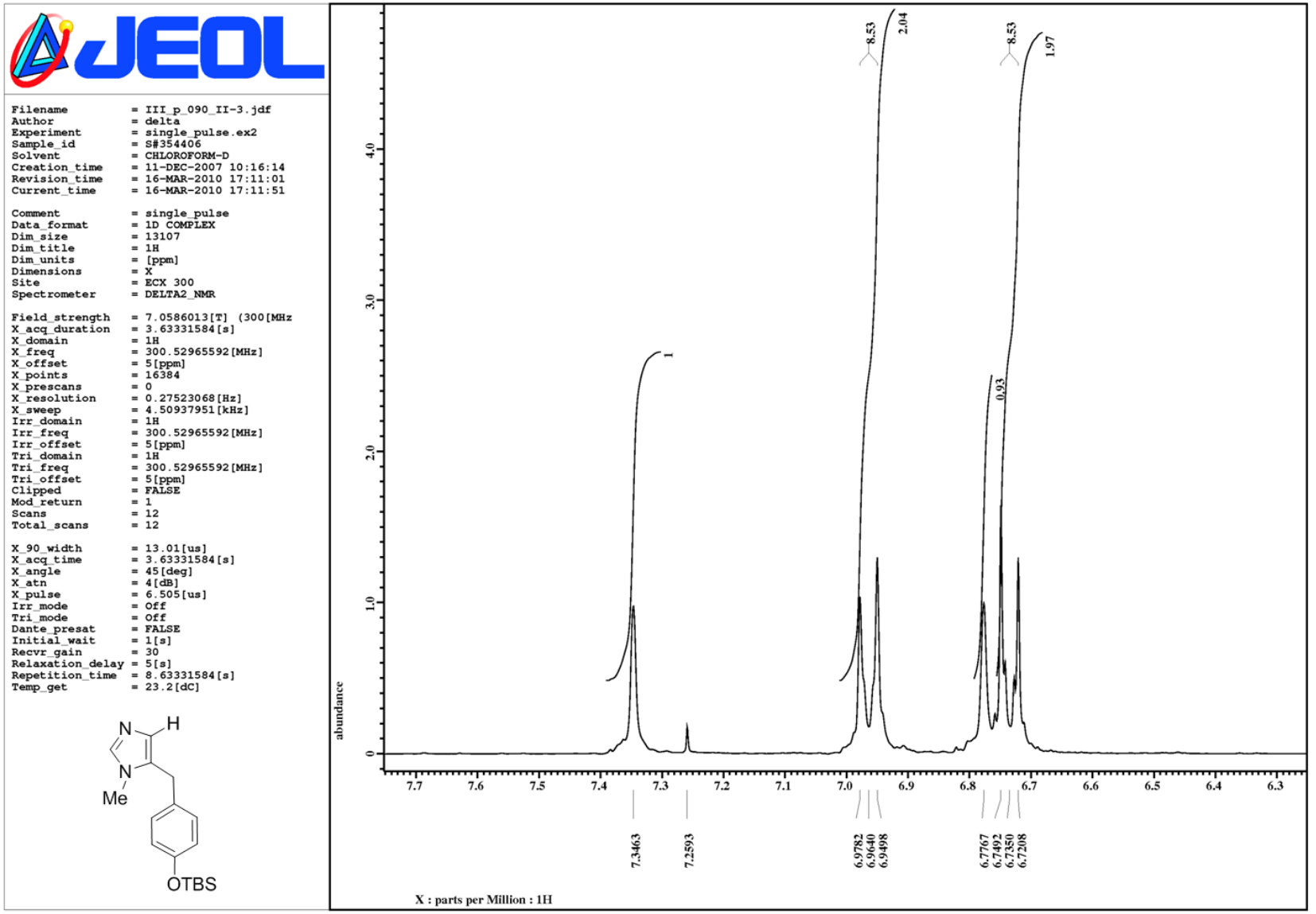
^{1}H and ^{13}C NMR Spectra of

5-(4-*t*-Butyldimethylsilyloxybenzyl)-1-methyl-1*H*-imidazole (**153**)

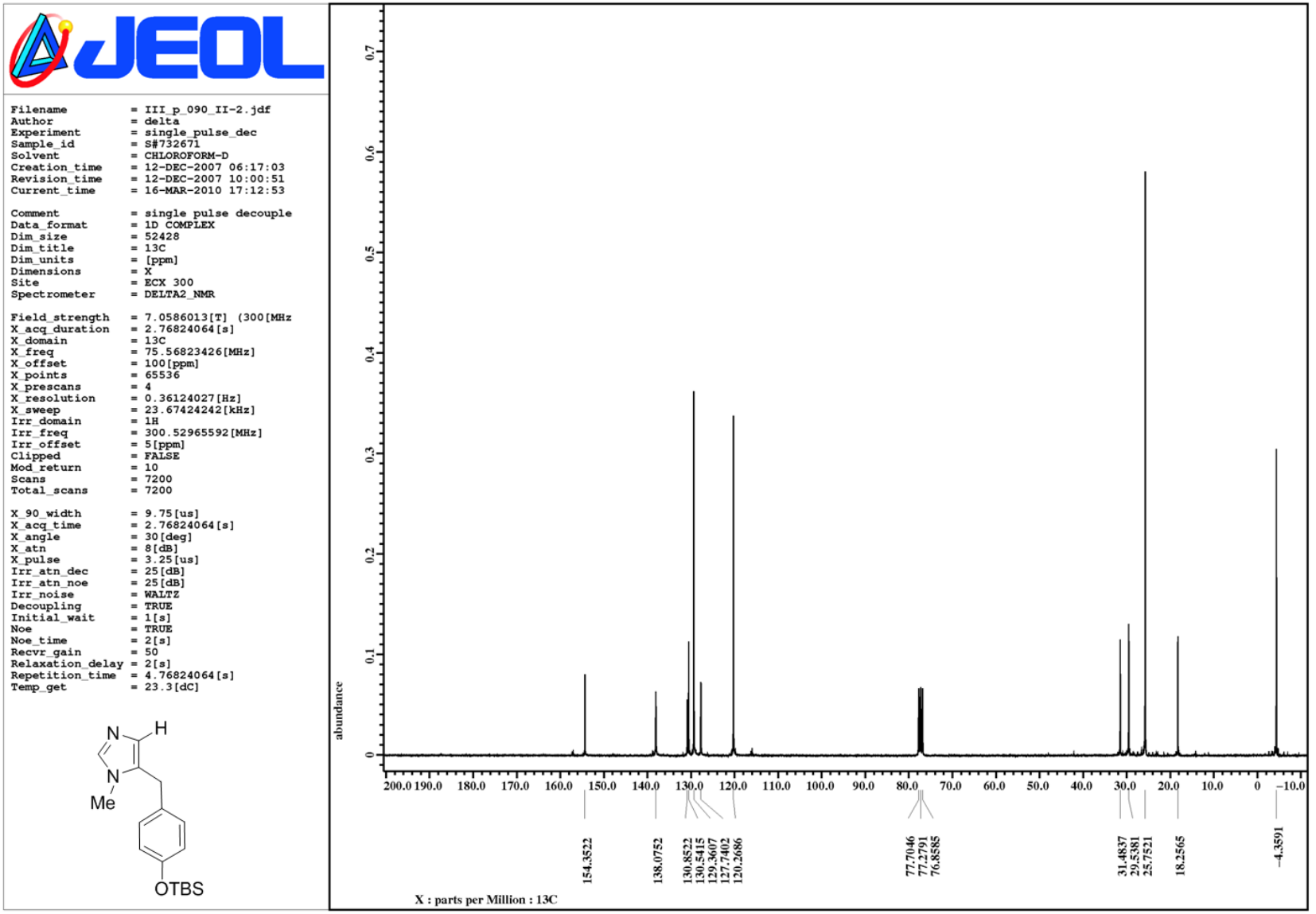
Filename	= III_p_090_II-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#354406
Solvent	= CHLOROFORM-D
Creation_time	= 11-DEC-2007 10:16:14
Revision_time	= 16-MAR-2010 17:11:01
Current_time	= 16-MAR-2010 17:11:26
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.2[dC]







```
Filename          = III_p_090_II-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#354406
Solvent           = CHLOROFORM-D
Creation_time     = 11-DEC-2007 10:16:14
Revision_time     = 16-MAR-2010 17:11:01
Current_time      = 16-MAR-2010 17:11:51

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.2[dC]
```

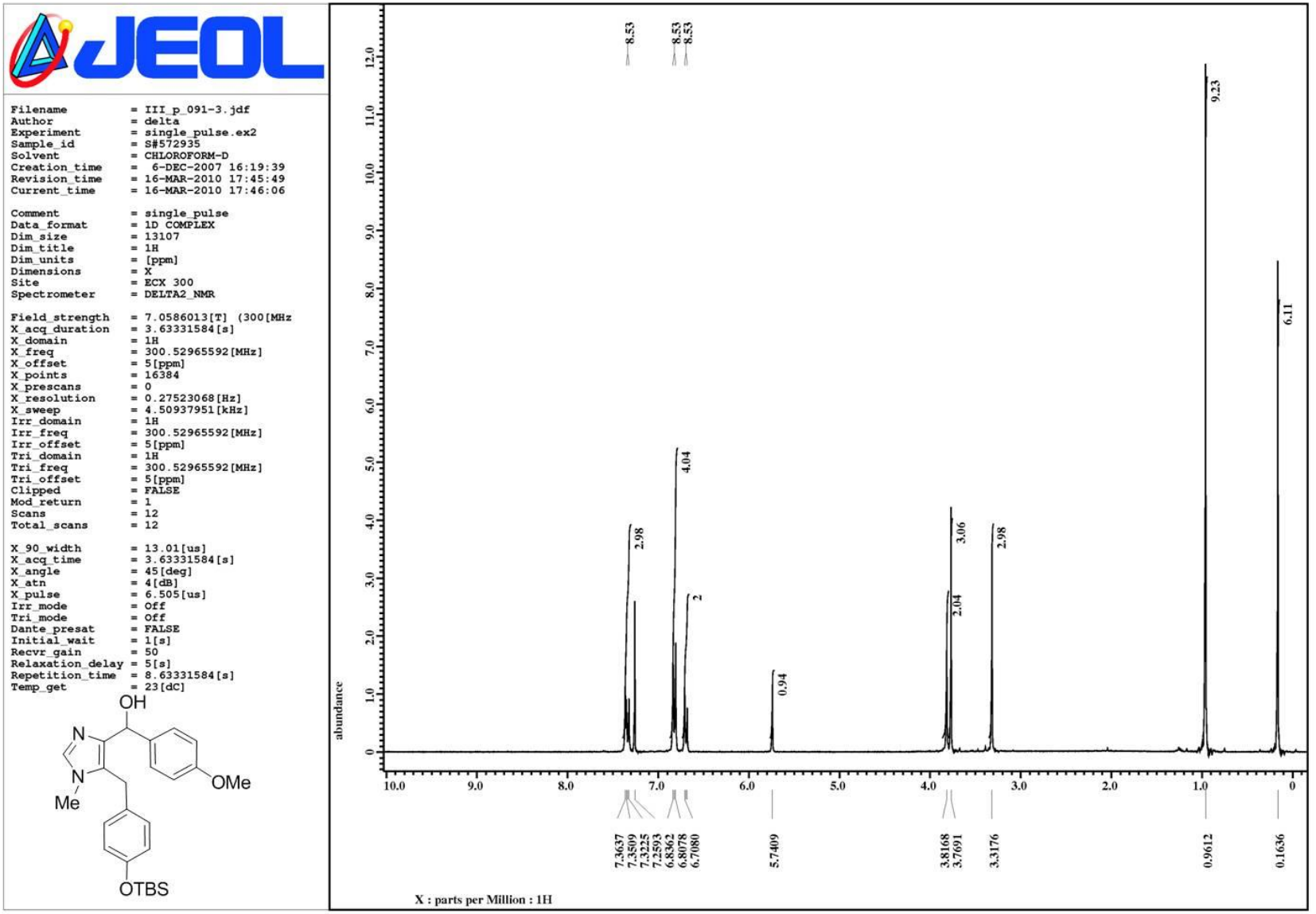


```
Filename          = III_p_090_II-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#732671
Solvent           = CHLOROFORM-D
Creation_time     = 12-DEC-2007 06:17:03
Revision_time     = 12-DEC-2007 10:00:51
Current_time      = 16-MAR-2010 17:12:53

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 7200
Total_scans       = 7200

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```

APPENDIX 33

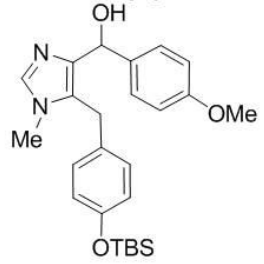
^{1}H and ^{13}C NMR Spectra of

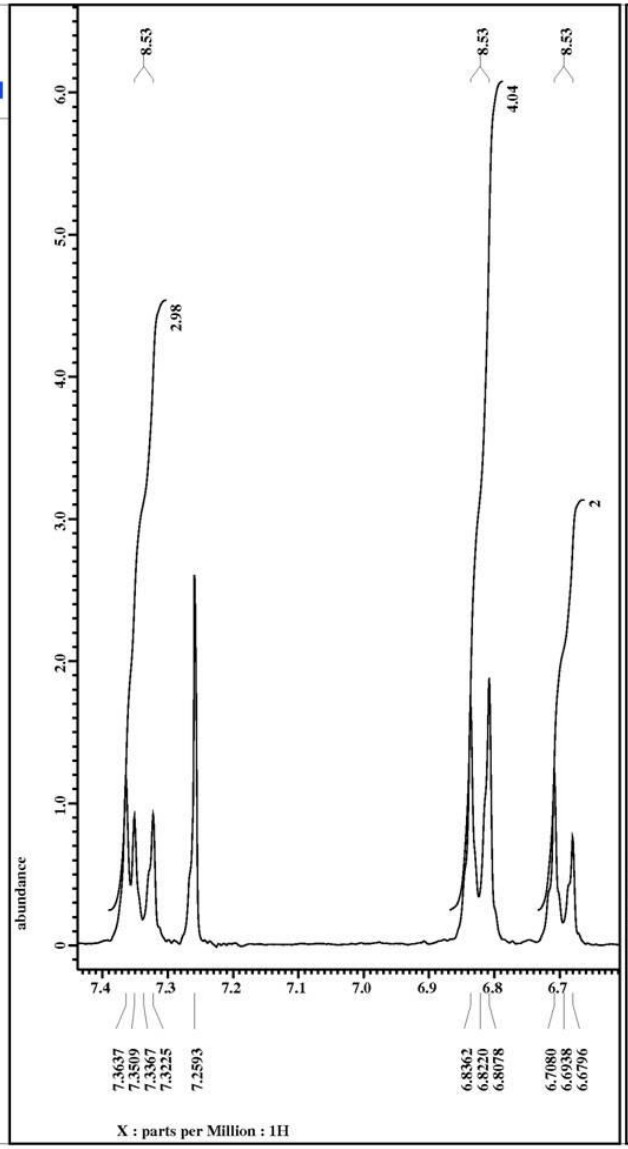
5-(4-*t*-Butyldimethylsilyloxybenzyl)-4-[hydroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**77**)

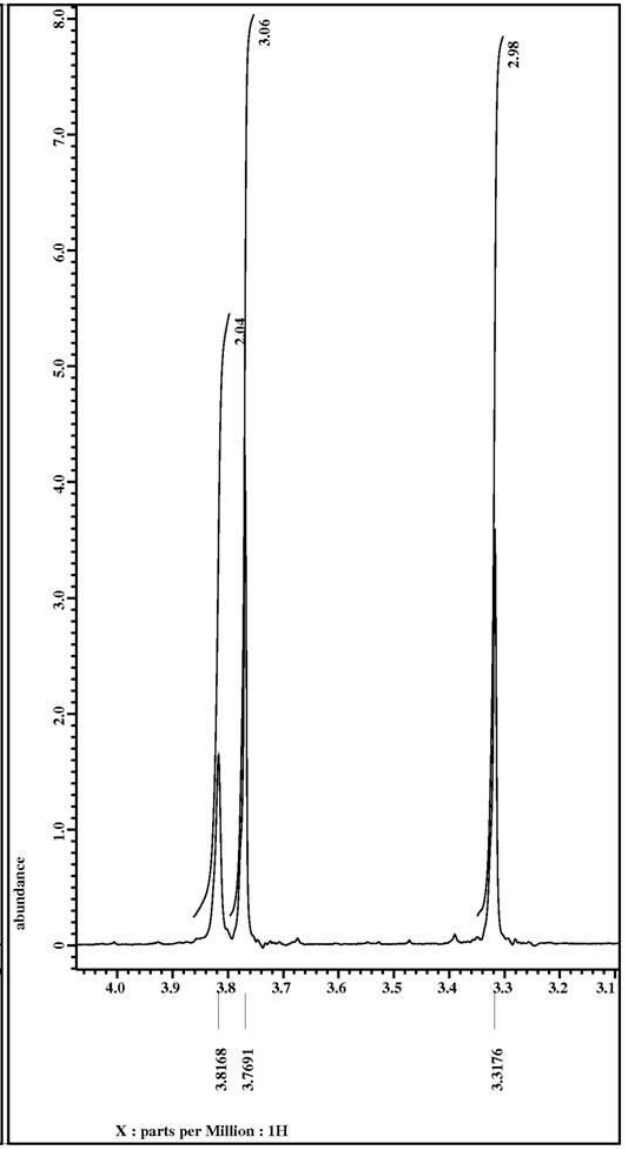


```
Filename          = III_p_091-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#572935
Solvent           = CHLOROFORM-D
Creation_time     =  6-DEC-2007 16:19:39
Revision_time     = 16-MAR-2010 17:45:49
Current_time      = 16-MAR-2010 17:46:06

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23[dC]
```

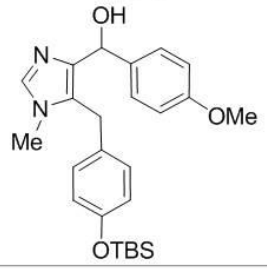
```
Filename          = III_p_091-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#572935
Solvent           = CHLOROFORM-D
Creation_time     =  6-DEC-2007 16:19:39
Revision_time     = 16-MAR-2010 17:45:49
Current_time      = 16-MAR-2010 17:46:38

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23[dC]
```

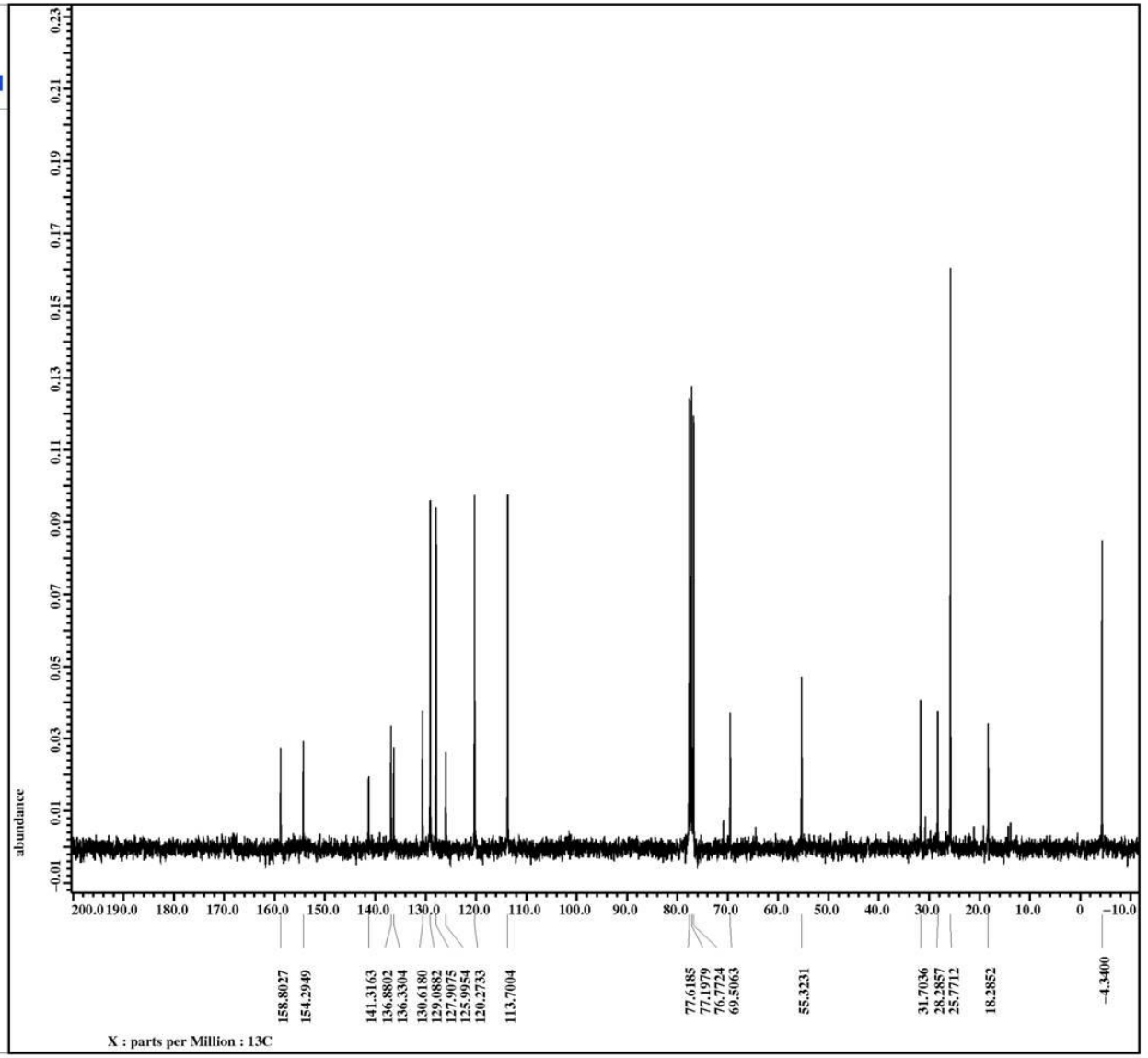






```
Filename          = III_p_010_II-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#726253
Solvent           = CHLOROFORM-D
Creation_time     = 18-OCT-2007 20:41:38
Revision_time     = 18-OCT-2007 20:21:29
Current_time      = 16-MAR-2010 17:41:35

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 106
Total_scans       = 106

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.7[dC]
```





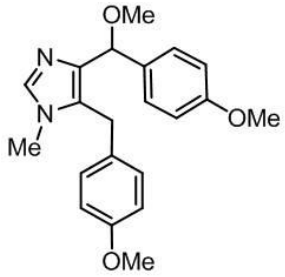
APPENDIX 34

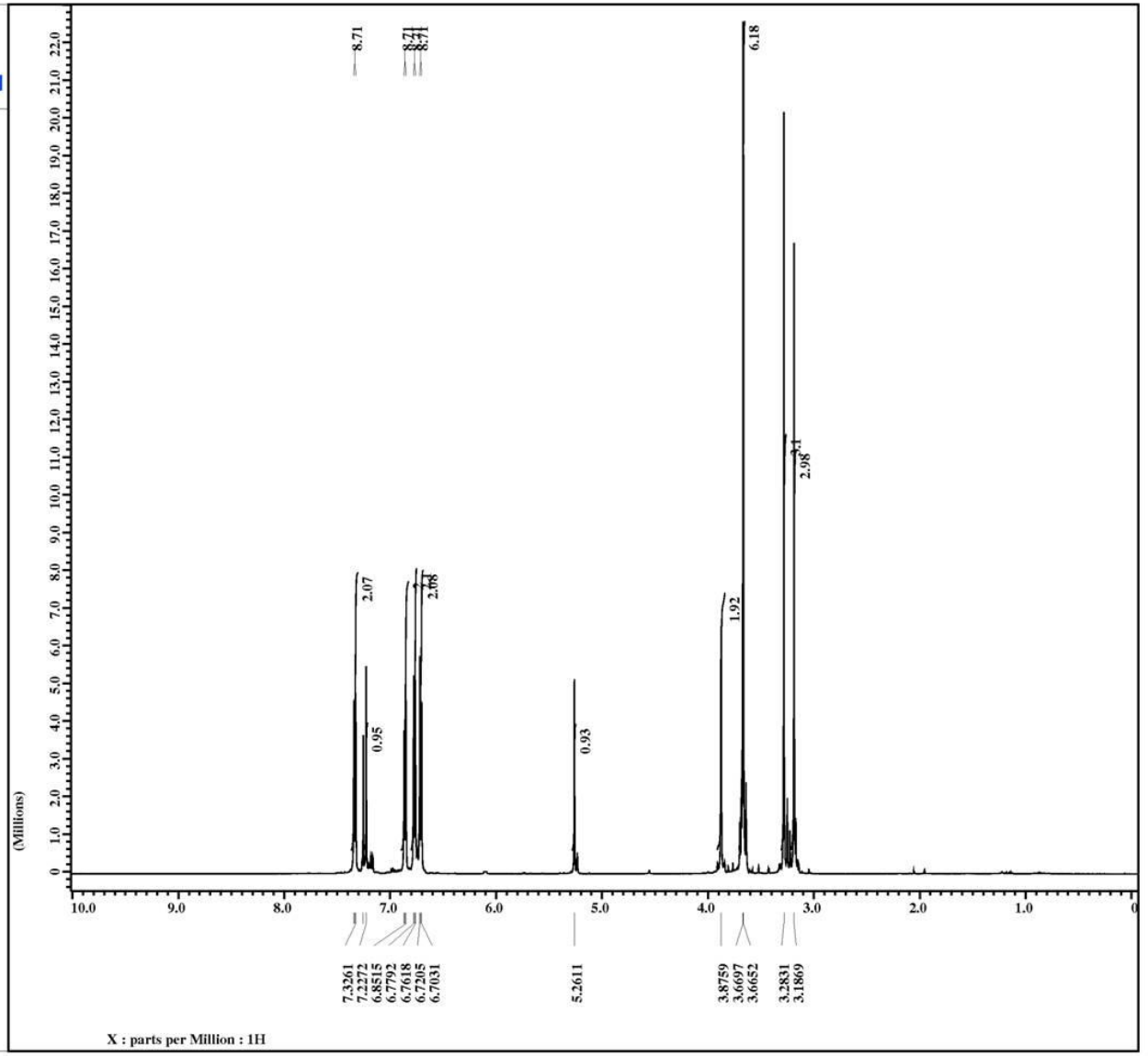
^{1}H and ^{13}C NMR Spectra of

5-(4-Methoxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole

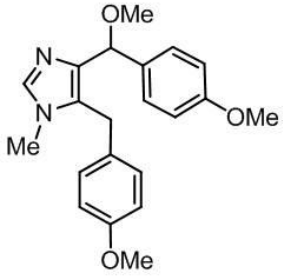
(**155**)

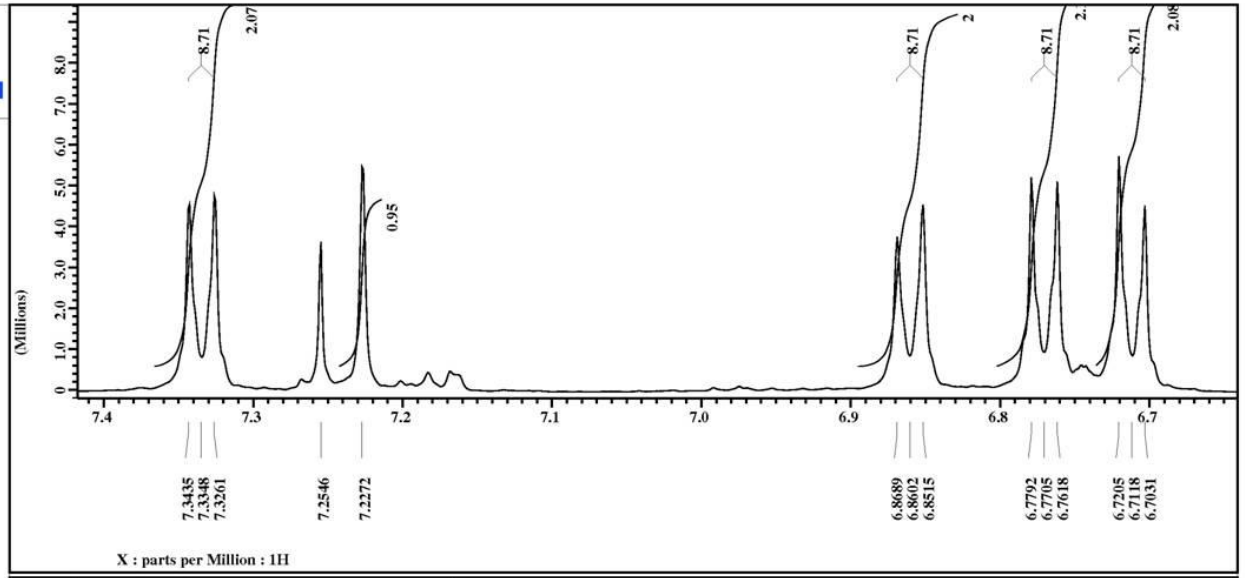
Filename	= V_p_024_MeO-5.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#481334
Solvent	= CHLOROFORM-D
Creation_time	= 21-NOV-2008 21:14:18
Revision_time	= 16-MAR-2010 18:11:38
Current_time	= 16-MAR-2010 18:12:19
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 10
Relaxation_delay	= 4[s]
Temp_get	= 25.8[dC]
Unblank_time	= 2[us]

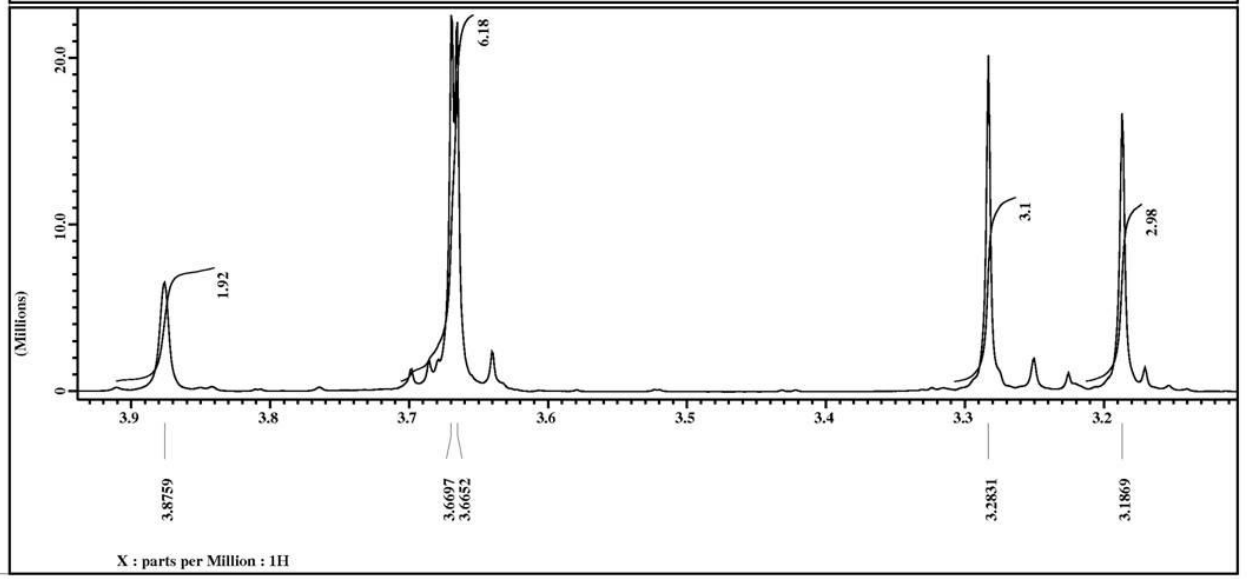




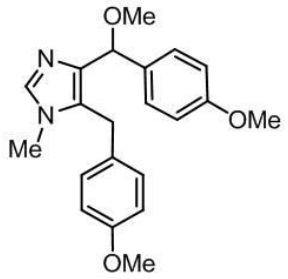
Filename	= V_p_024_MeO-5.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#481334
Solvent	= CHLOROFORM-D
Creation_time	= 21-NOV-2008 21:14:18
Revision_time	= 16-MAR-2010 18:11:38
Current_time	= 16-MAR-2010 18:12:55
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 10
Relaxation_delay	= 4[s]
Temp_get	= 25.8[dC]
Unblank_time	= 2[us]

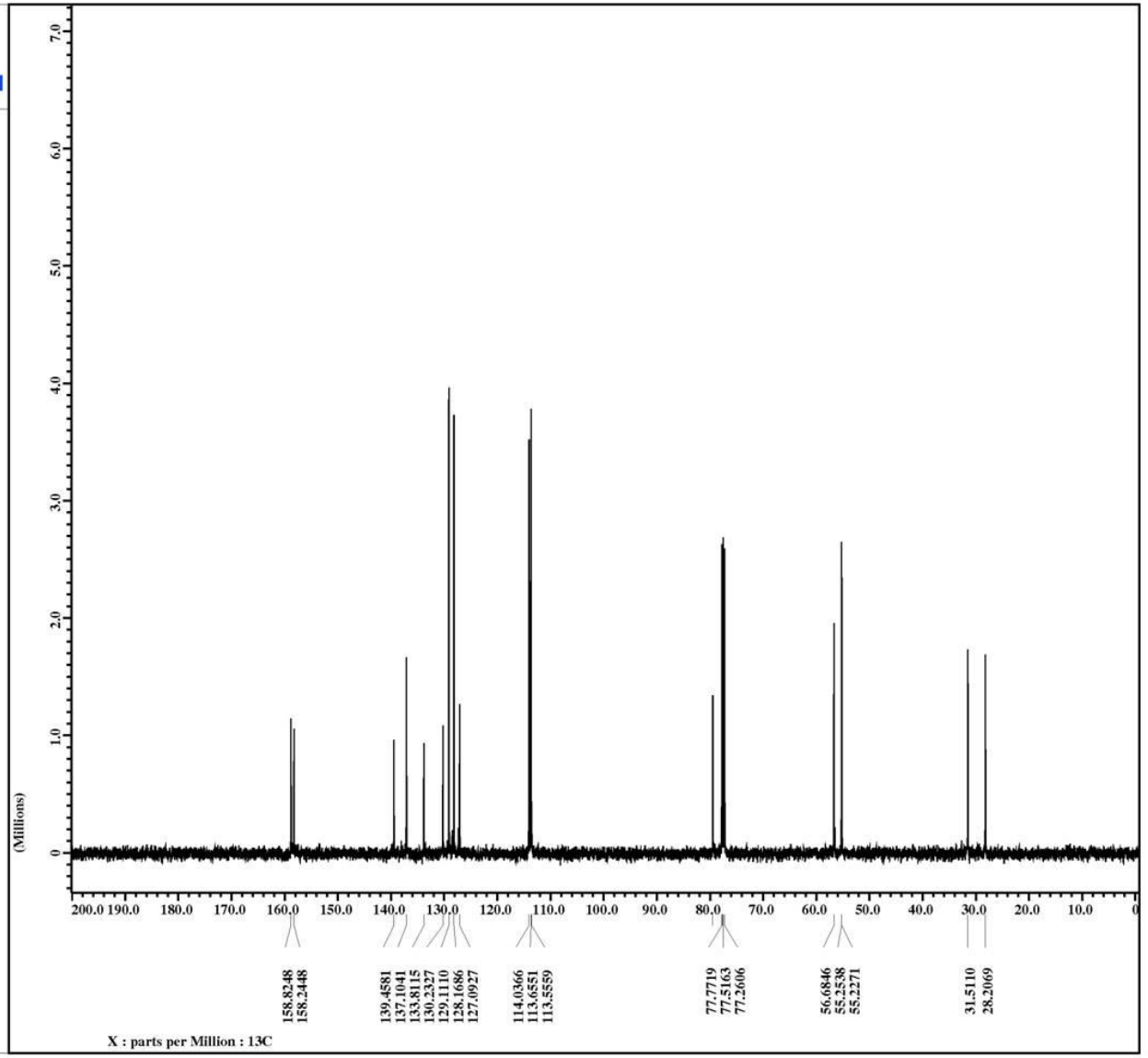




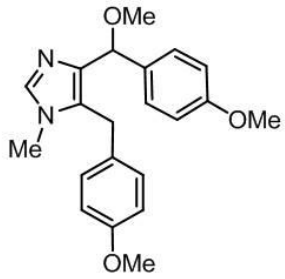


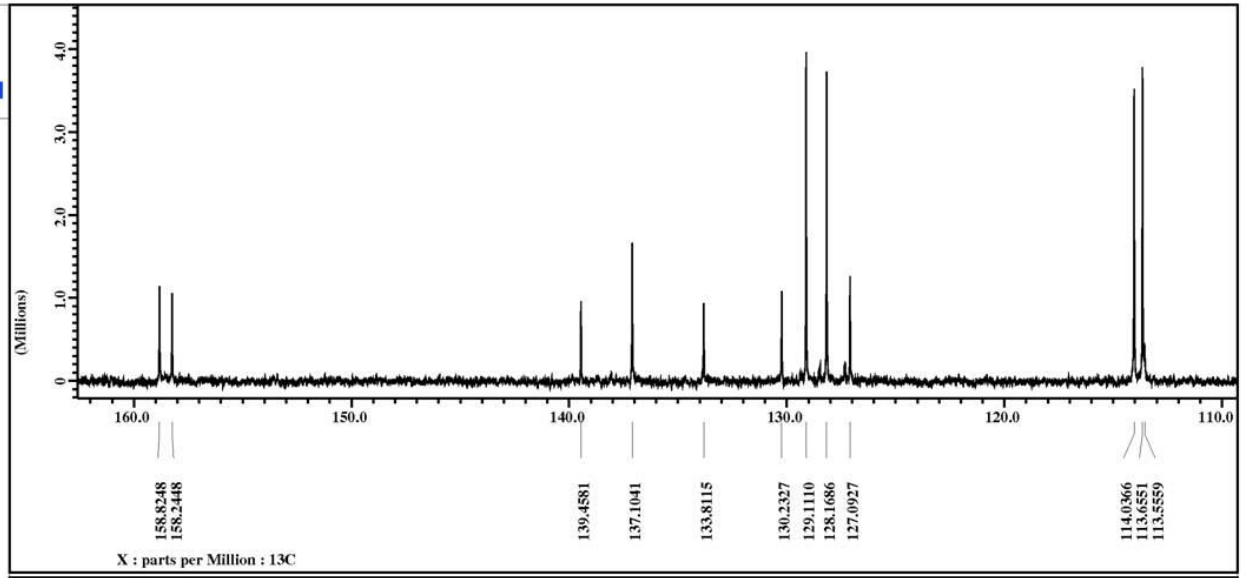
Filename	= V_p_024_MeO-3.jdf
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#482433
Solvent	= CHLOROFORM-D
Creation_time	= 21-NOV-2008 21:22:22
Revision_time	= 16-MAR-2010 18:13:26
Current_time	= 16-MAR-2010 18:14:21
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 65536
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.0840448[s]
X_domain	= 13C
X_freq	= 125.76529768[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.47983613[Hz]
X_sweep	= 31.44654088[kHz]
Irr_domain	= 1H
Irr_freq	= 500.15991521[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 71
Total_scans	= 71
X_90_width	= 14.2[us]
X_acq_time	= 2.0840448[s]
X_angle	= 30[deg]
X_pulse	= 4.73333333[us]
Initial_wait	= 1[s]
Noe_time	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 29
Relaxation_delay	= 2[s]
Temp_get	= 27.5[dC]
Unblank_time	= 2[us]

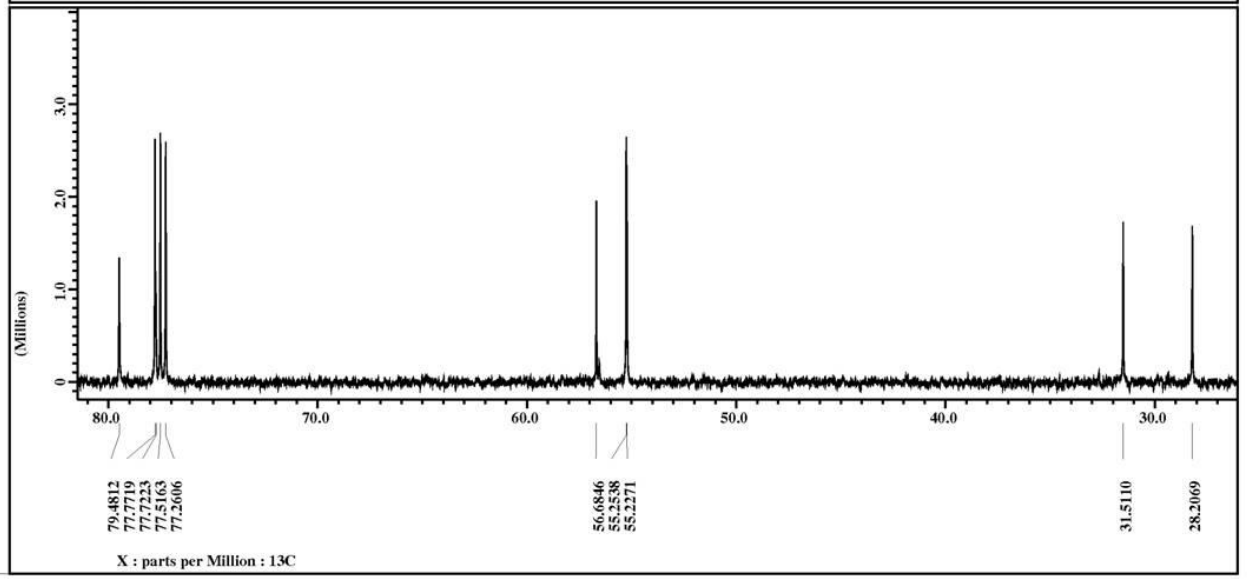




```
Filename          = V_p_024_MeO-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#482433
Solvent           = CHLOROFORM-D
Creation_time     = 21-NOV-2008 21:22:22
Revision_time     = 16-MAR-2010 18:13:26
Current_time      = 16-MAR-2010 18:16:09

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 71
Total_scans       = 71

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 29
Relaxation_delay  = 2[s]
Temp_get          = 27.5[dC]
Unblank_time      = 2[us]
```





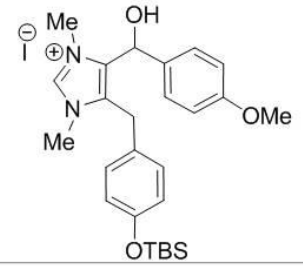


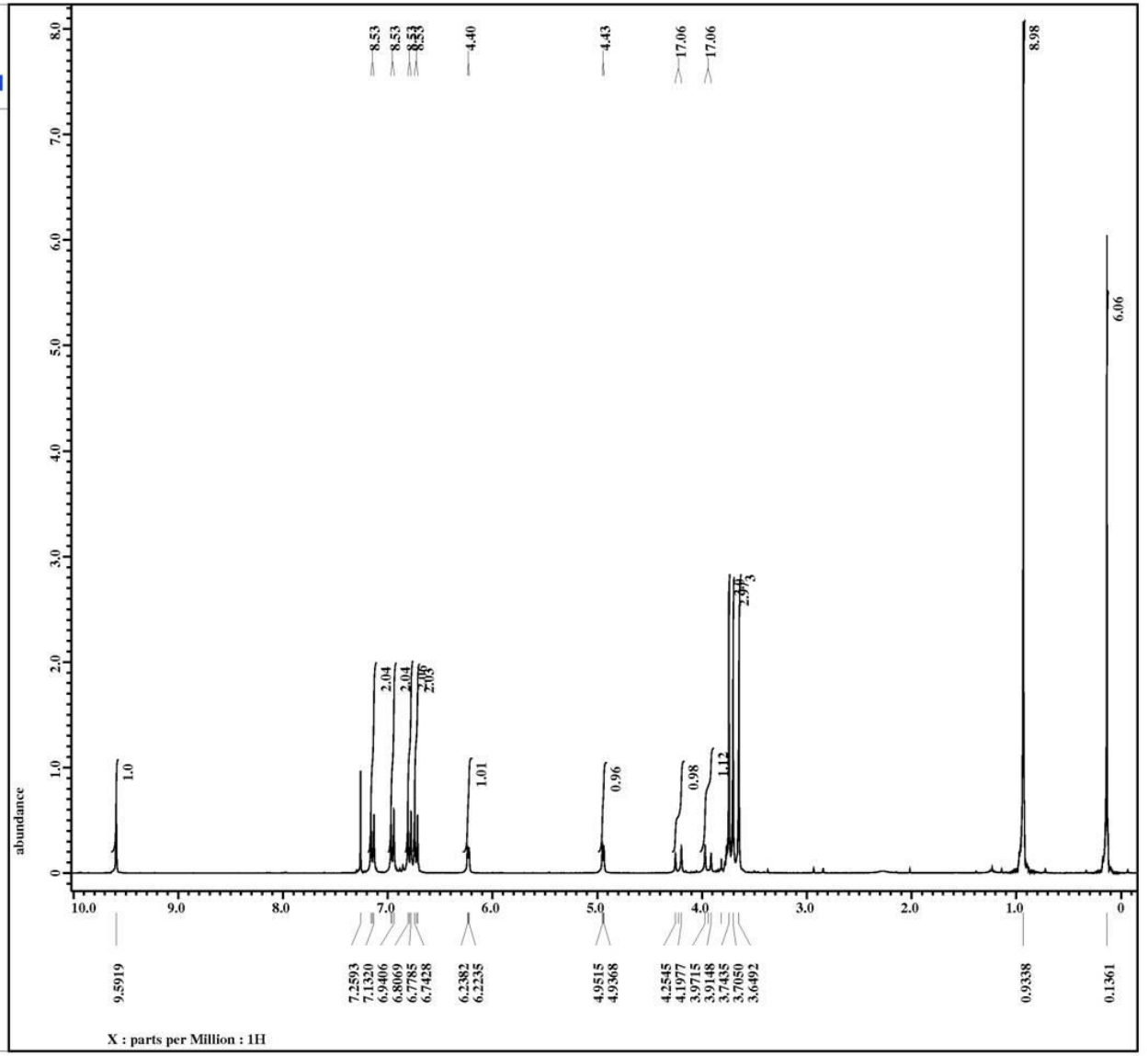
APPENDIX 35

^{1}H and ^{13}C NMR Spectra of

4-(4-*t*-Butyldimethylsilanyloxybenzyl)-1,3-dimethyl-5-[hydroxy-(4-methoxyphenyl)]methyl- 3*H*-imidazol-1-ium iodide (**156**)

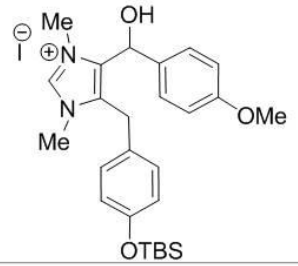
```
Filename          = III_p_014_product-2.j
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#686916
Solvent           = CHLOROFORM-D
Creation_time     = 20-OCT-2007 19:28:54
Revision_time     = 20-OCT-2007 19:14:25
Current_time      = 16-MAR-2010 18:29:10

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 36
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```

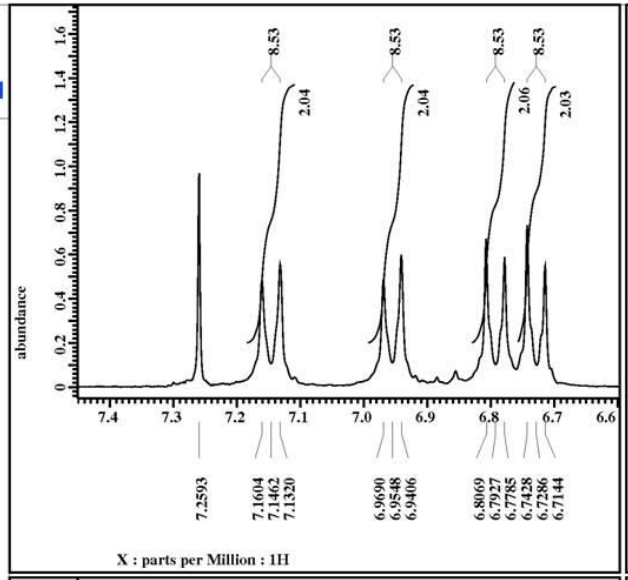


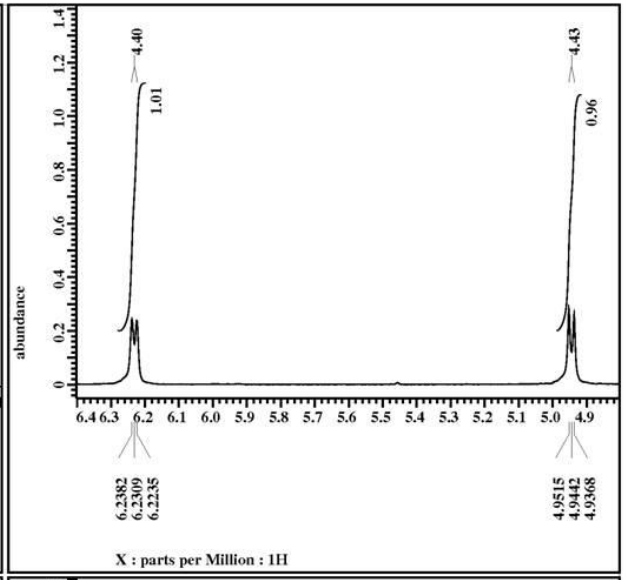


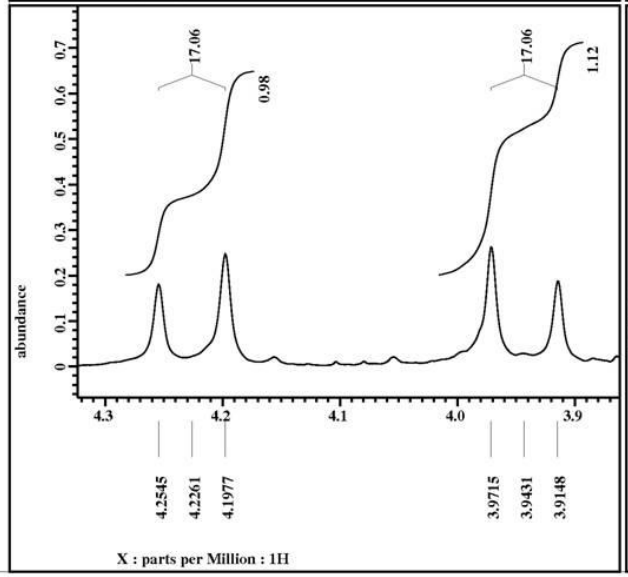
JEOL

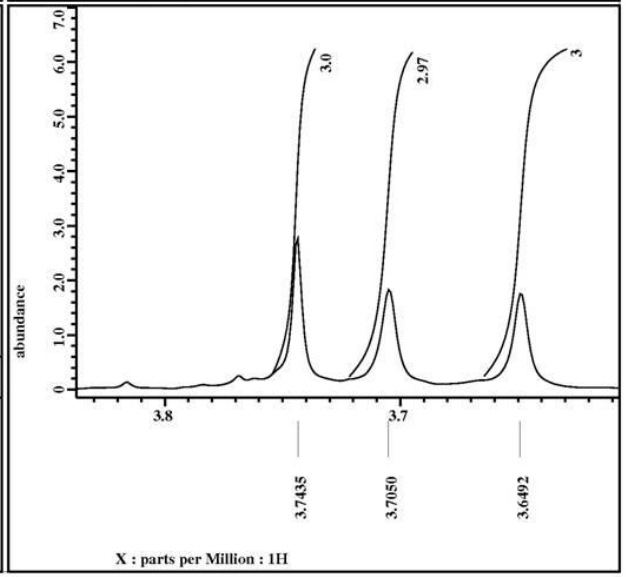
Filename	= III_p_014_product-3.j
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#686916
Solvent	= CHLOROFORM-D
Creation_time	= 20-OCT-2007 19:28:54
Revision_time	= 16-MAR-2010 18:31:33
Current_time	= 16-MAR-2010 18:32:04
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 36
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.1[dC]

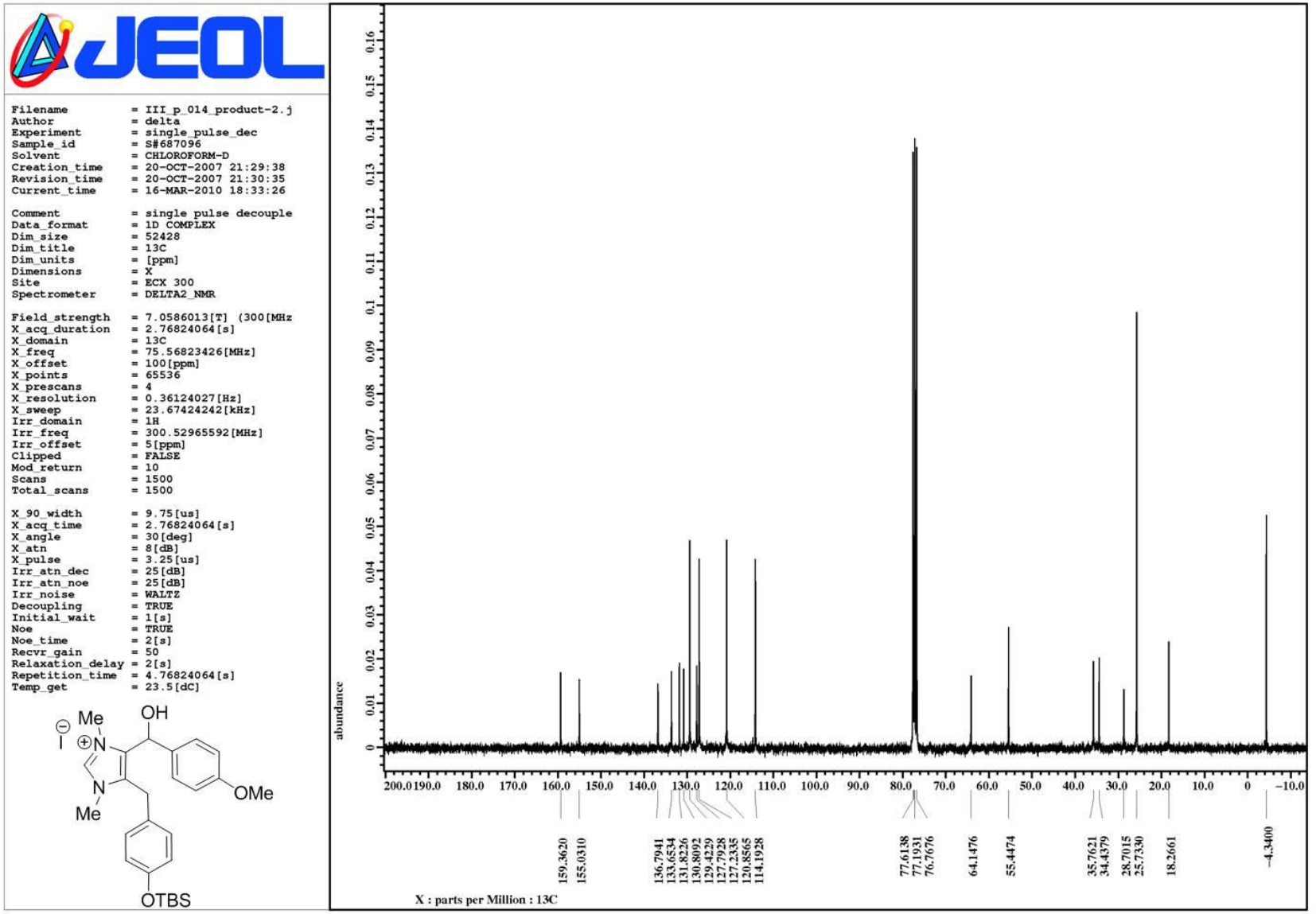












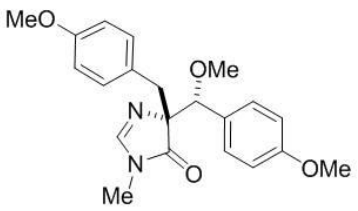
```
Filename          = III_p_014_product-2.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#687096
Solvent           = CHLOROFORM-D
Creation_time     = 20-OCT-2007 21:29:38
Revision_time     = 20-OCT-2007 21:30:35
Current_time      = 16-MAR-2010 18:33:26

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1500
Total_scans       = 1500

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```

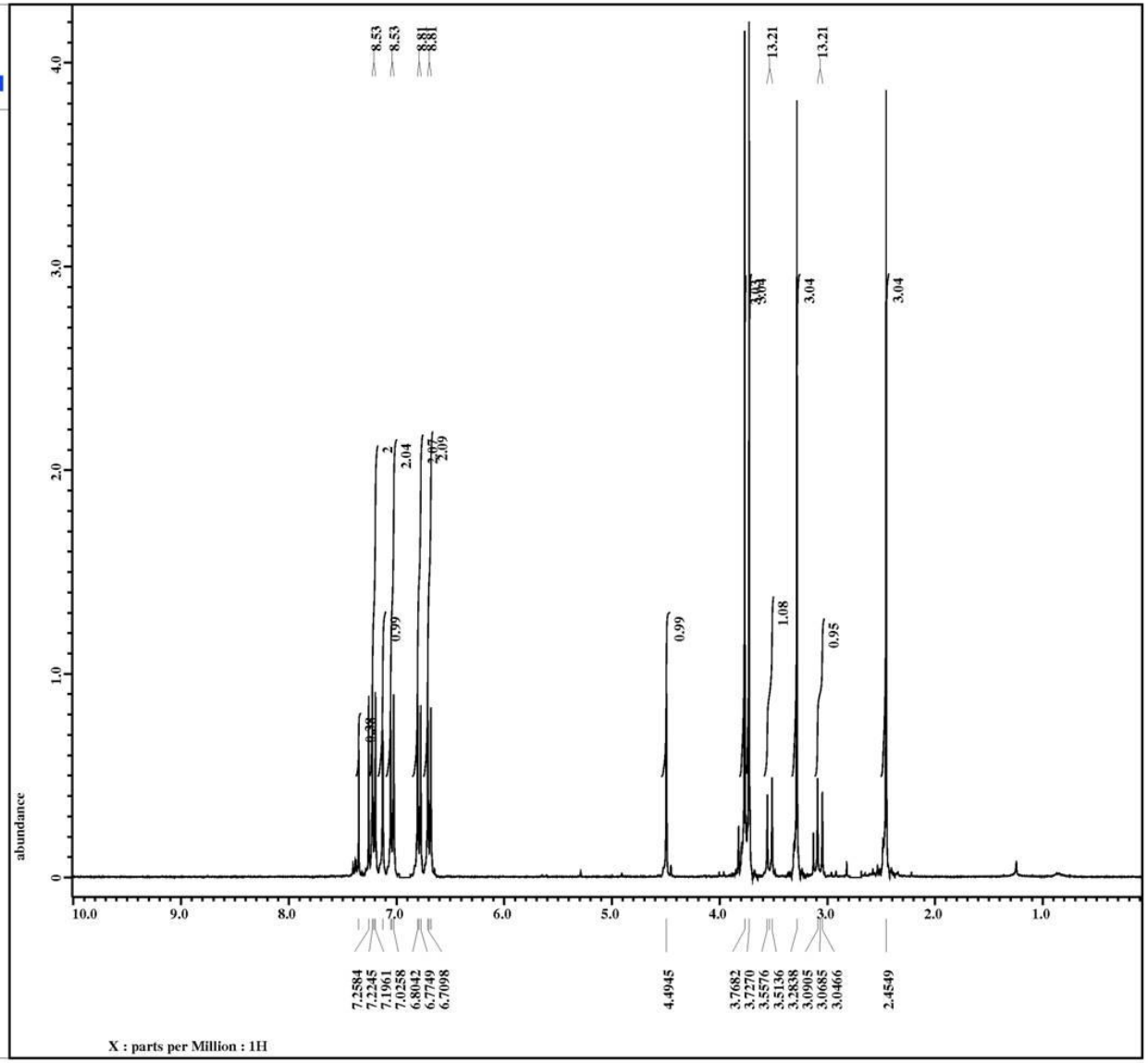
APPENDIX 36

^{1}H and ^{13}C NMR Spectra of

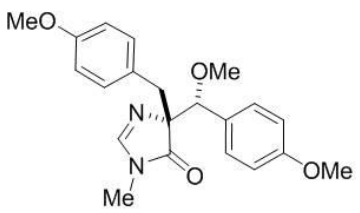
5-(4-Methoxy-benzyl)-5-[methoxy-(4-methoxy-phenyl)-methyl]-3-methyl-3,5-dihydro-imidazol-4-one (**157**)

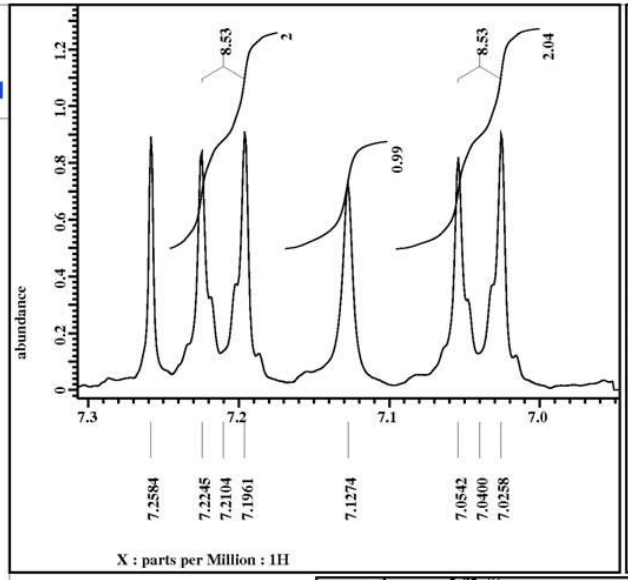
```
Filename          = II_p_296_recrystalliz
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#635500
Solvent           = CHLOROFORM-D
Creation_time     = 21-APR-2008 17:52:41
Revision_time     = 16-MAR-2010 18:44:17
Current_time      = 16-MAR-2010 18:44:39

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 44
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

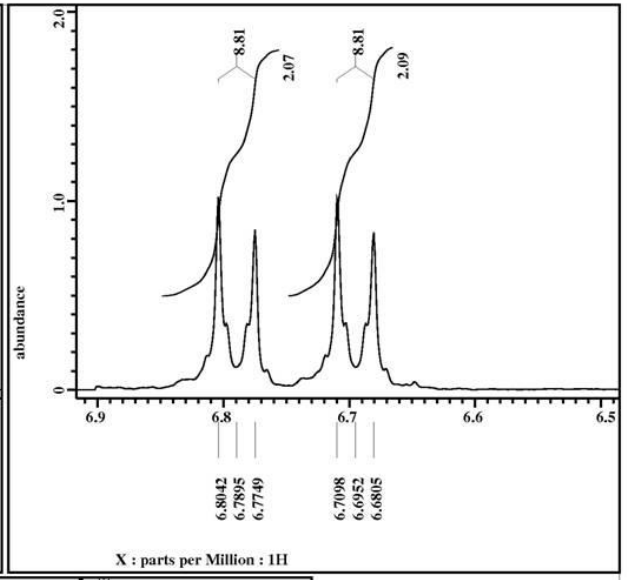


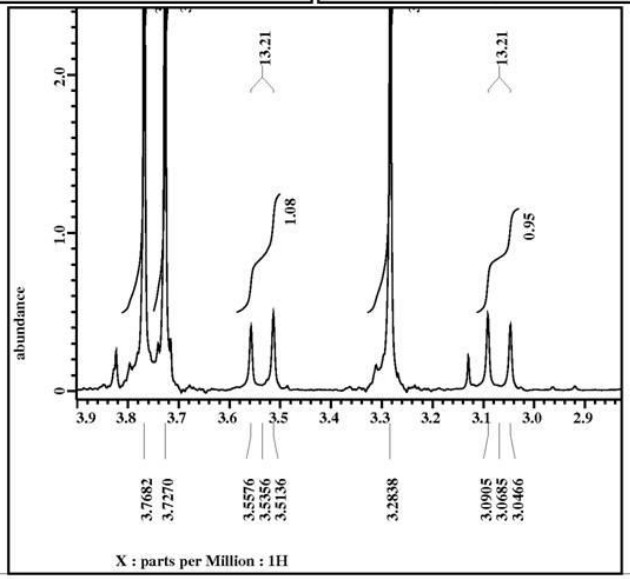


```
Filename          = II_p_296_recrystalliz
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#635500
Solvent           = CHLOROFORM-D
Creation_time     = 21-APR-2008 17:52:41
Revision_time     = 15-MAR-2010 15:33:37
Current_time      = 15-MAR-2010 15:36:45

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 44
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```

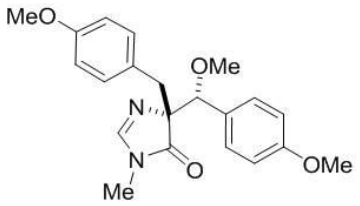


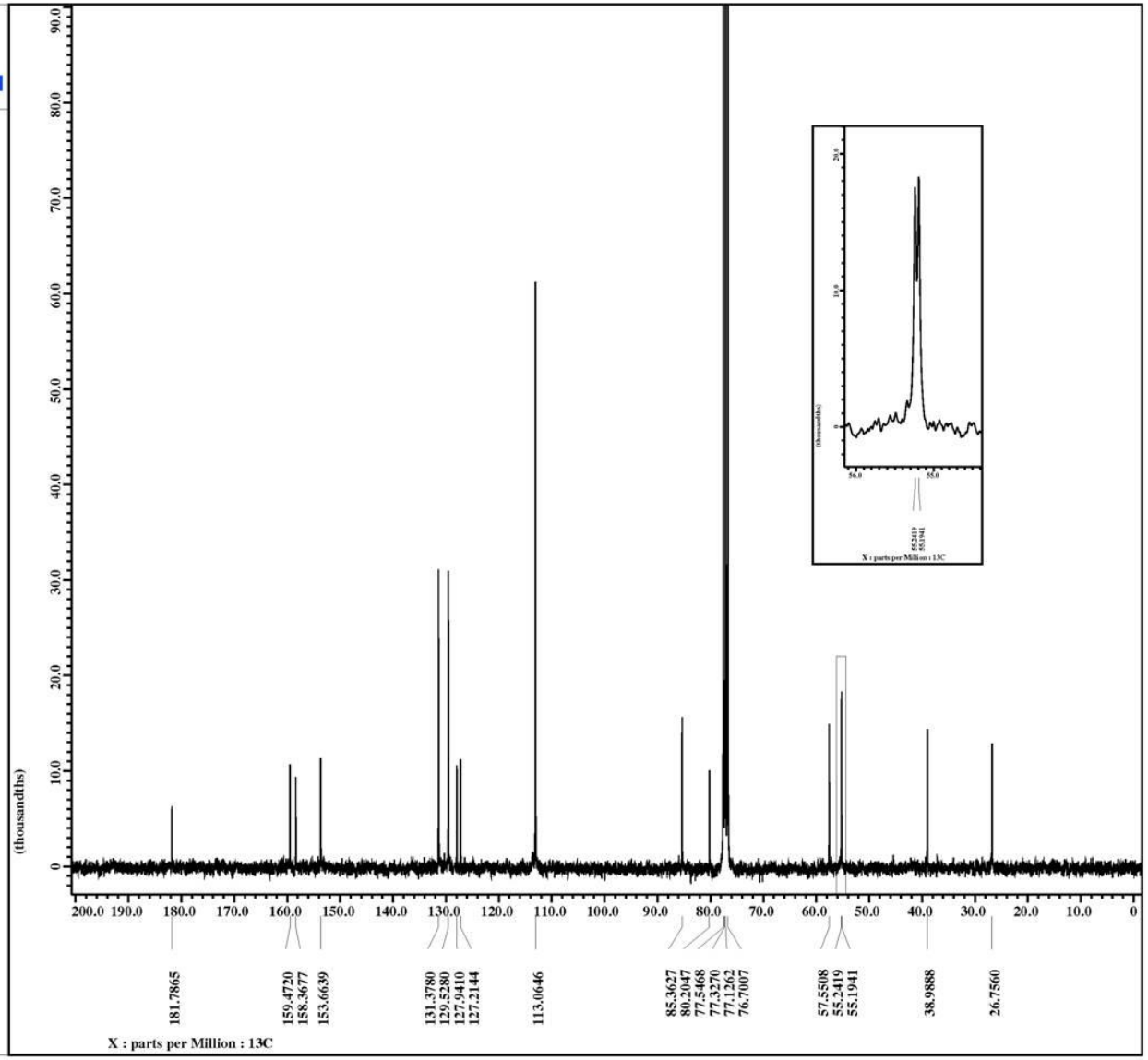






```
Filename          = II_p_296_recrystalliz
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#637284
Solvent           = CHLOROFORM-D
Creation_time     = 21-APR-2008 20:00:35
Revision_time     = 16-MAR-2010 18:48:25
Current_time      = 16-MAR-2010 18:49:59

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1600
Total_scans       = 1600

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.2[dC]
```

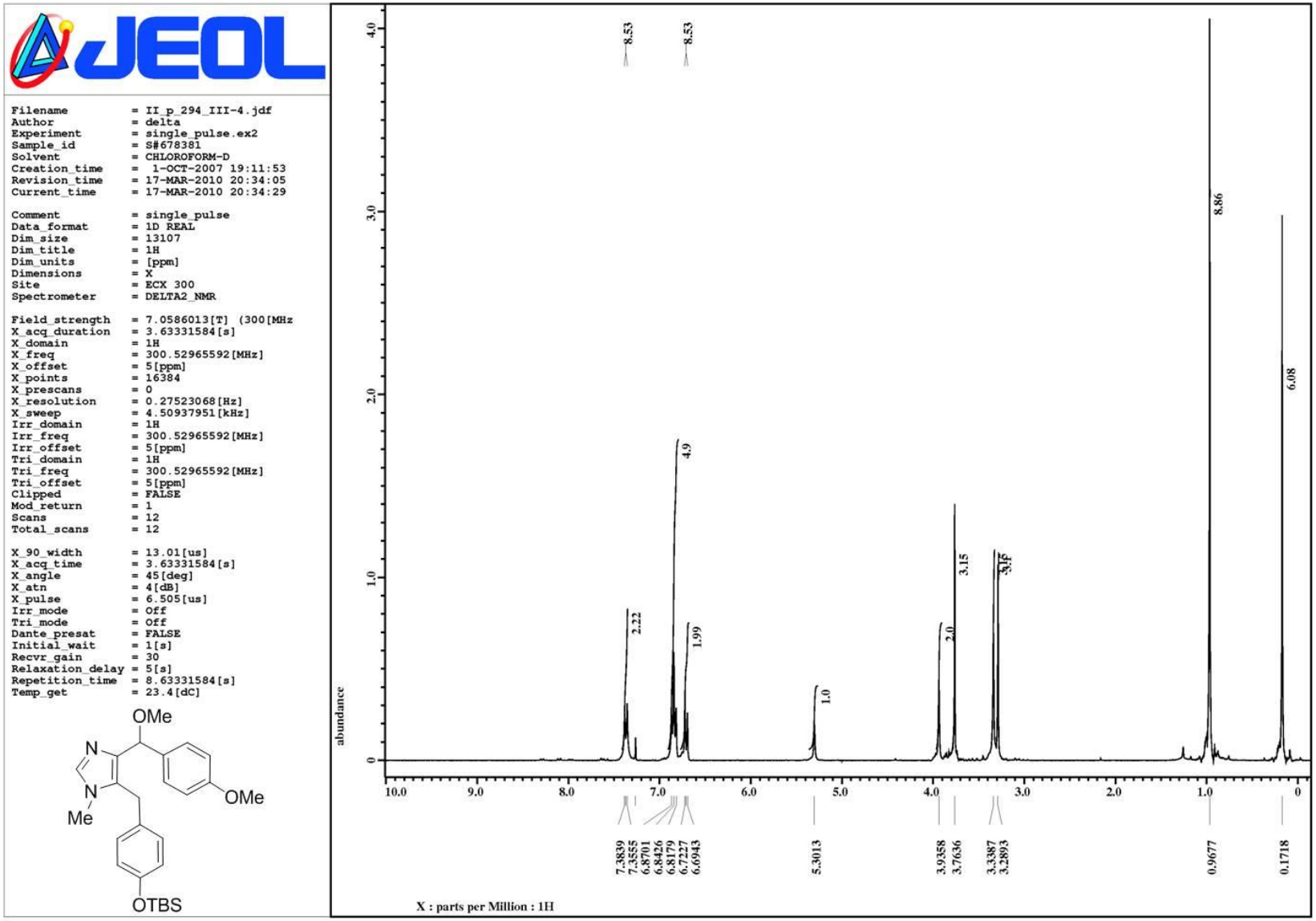




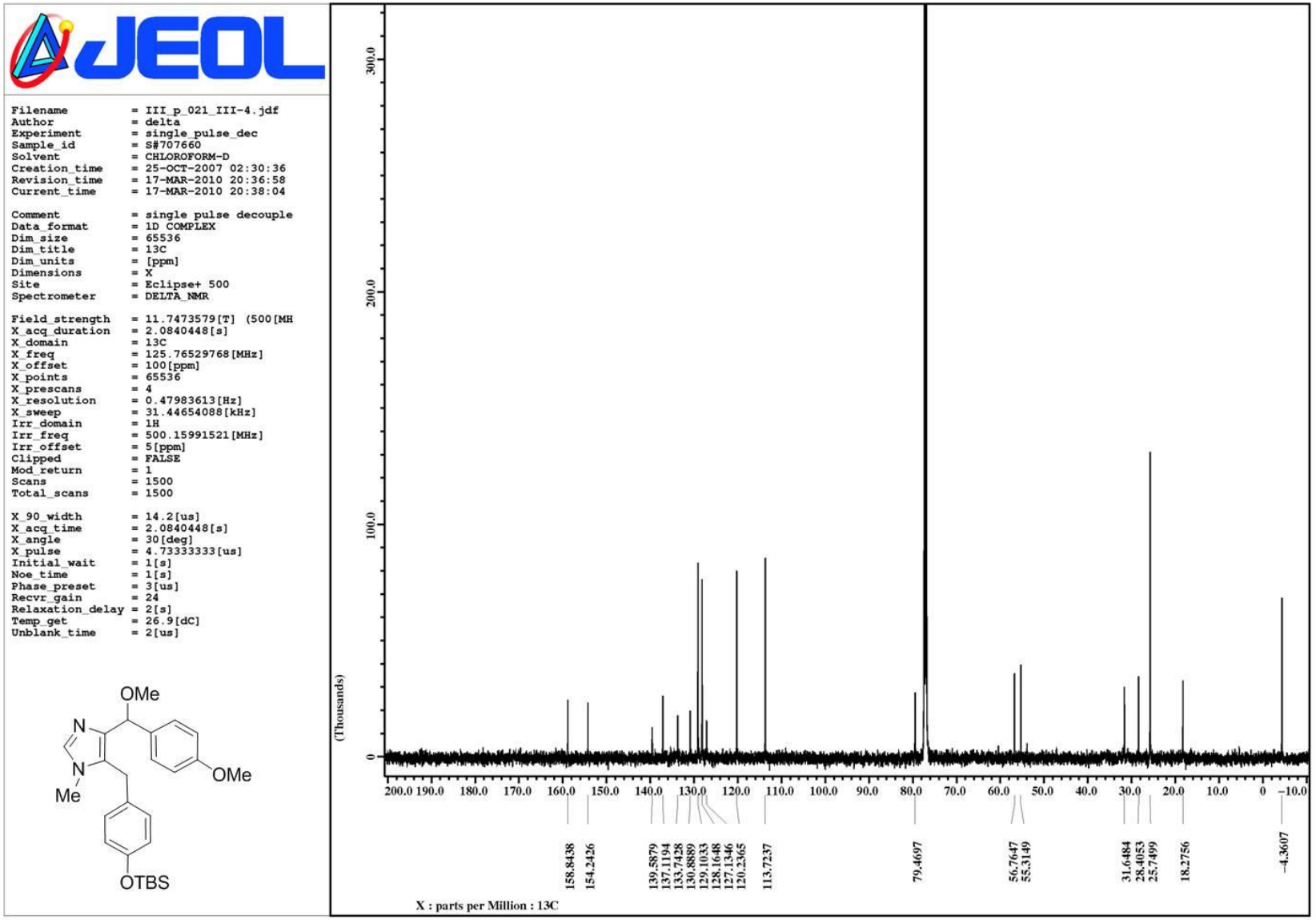
APPENDIX 37

^{1}H and ^{13}C NMR Spectra of

5-(4-*t*-Butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**76**)



```
Filename          = II_p_294_III-4.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#678381
Solvent           = CHLOROFORM-D
Creation_time     =  1-OCT-2007 19:11:53
Revision_time     = 17-MAR-2010 20:34:05
Current_time      = 17-MAR-2010 20:34:29

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.4[dC]
```



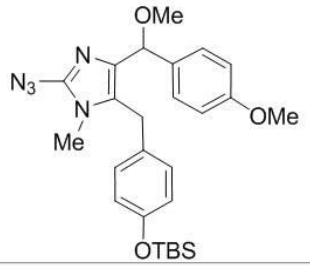
```
Filename          = III_p_021_III-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#707660
Solvent           = CHLOROFORM-D
Creation_time     = 25-OCT-2007 02:30:36
Revision_time     = 17-MAR-2010 20:36:58
Current_time      = 17-MAR-2010 20:38:04

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 1500
Total_scans       = 1500

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 24
Relaxation_delay  = 2[s]
Temp_get          = 26.9[dC]
Unblank_time      = 2[us]
```

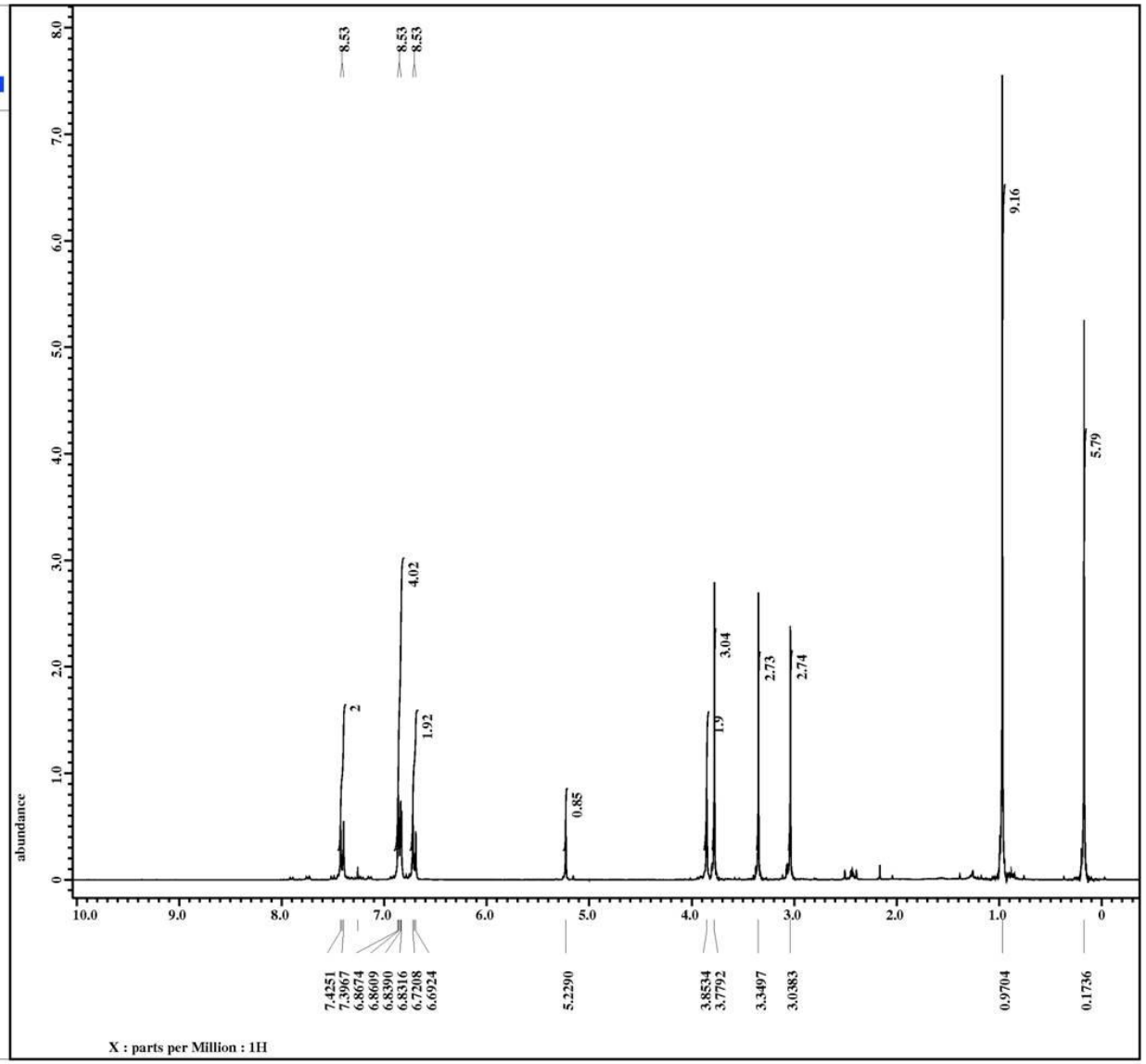
APPENDIX 38

^{1}H and ^{13}C NMR Spectra of

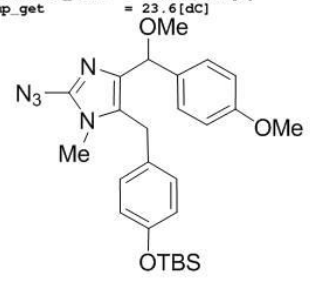
2-Azido-5-(4-*t*-butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**158**)

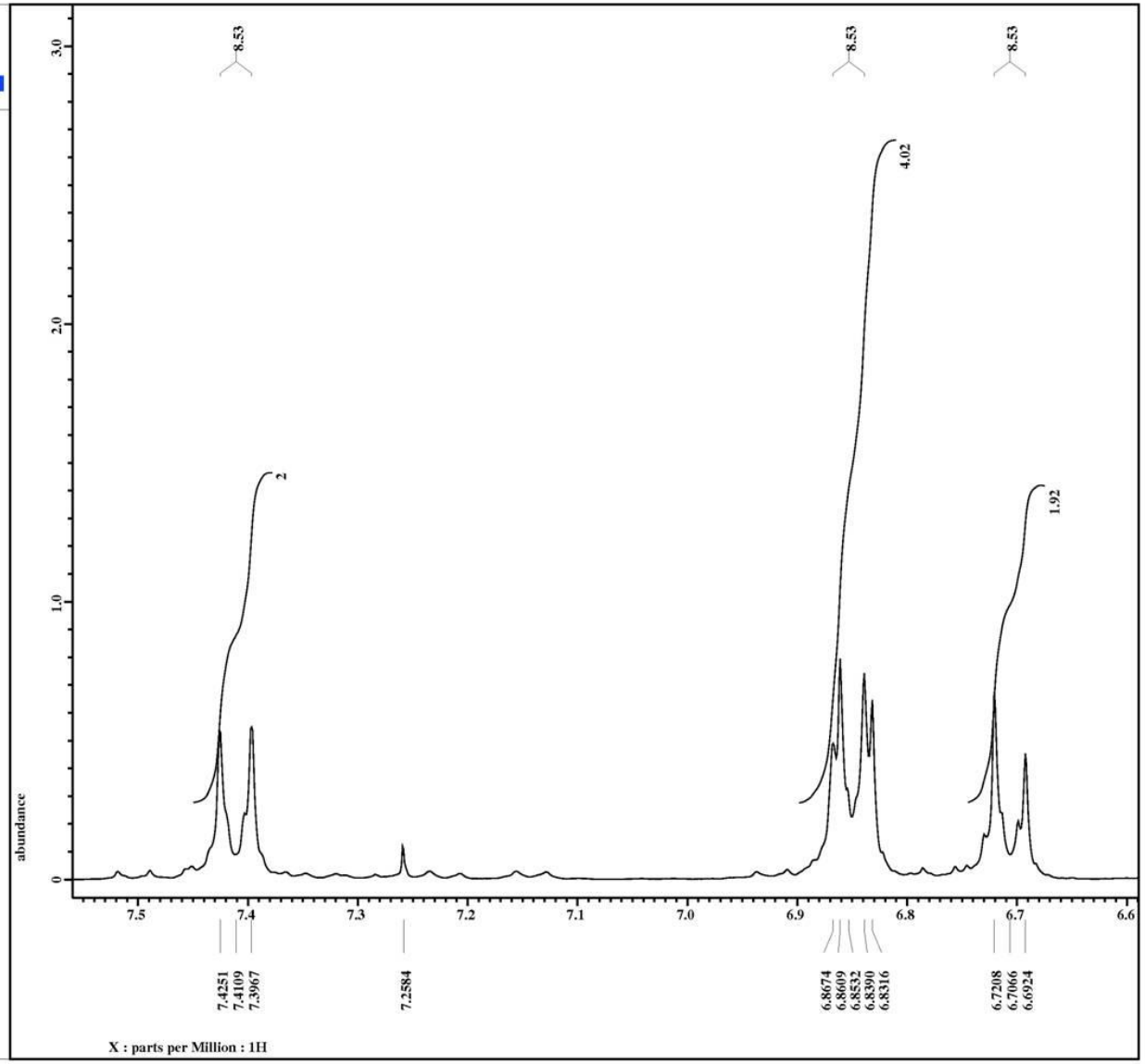
```
Filename          = III_p_008_azide-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#30817
Solvent           = CHLOROFORM-D
Creation_time     = 18-OCT-2007 01:15:46
Revision_time     = 17-MAR-2010 21:00:33
Current_time      = 17-MAR-2010 21:01:23

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.6[dC]
```



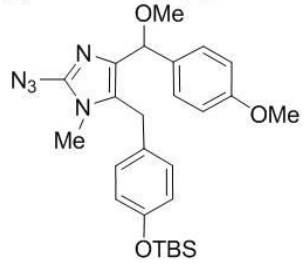


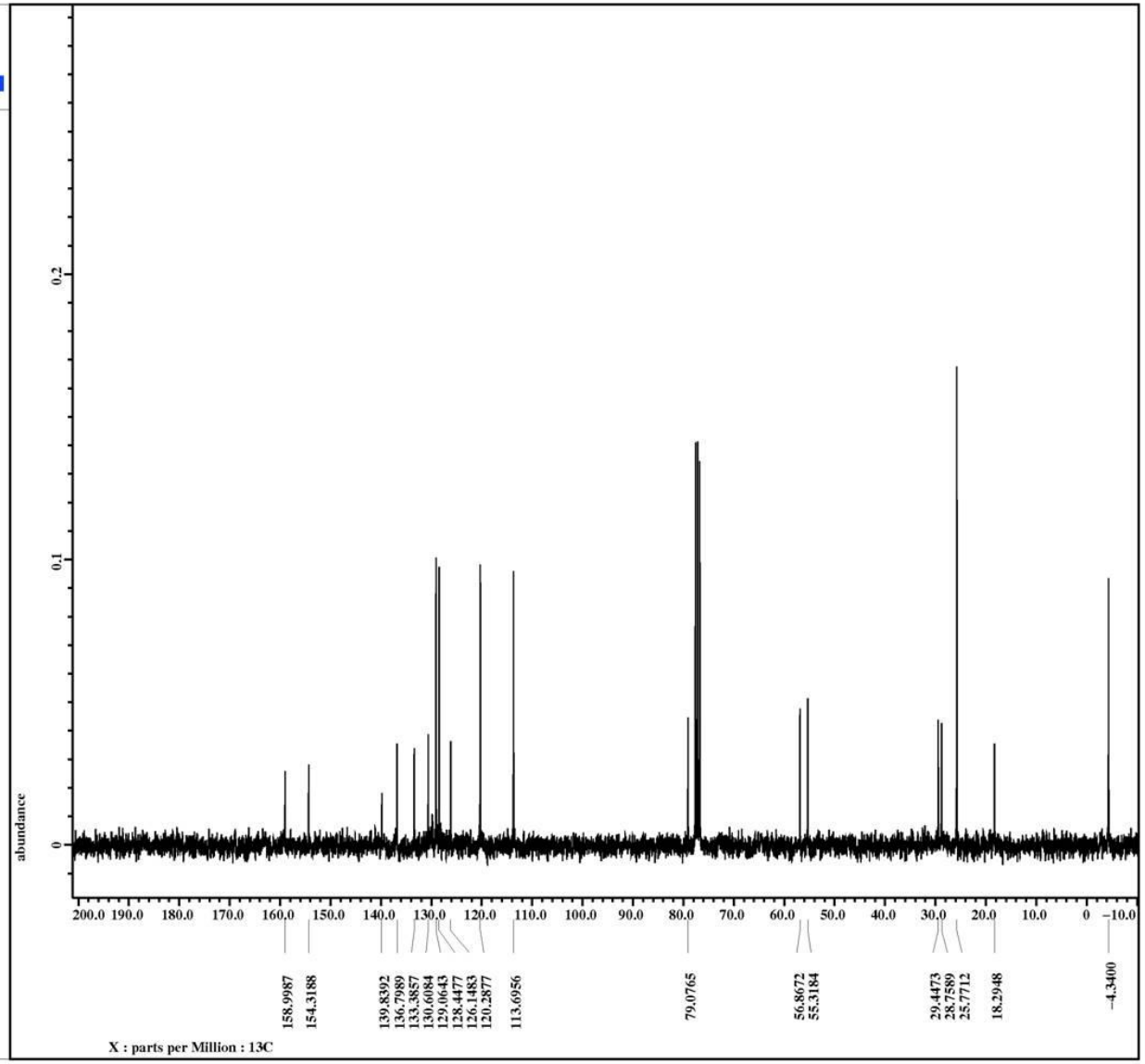
```
Filename          = III_p_008_azide-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#30817
Solvent           = CHLOROFORM-D
Creation_time     = 18-OCT-2007 01:15:46
Revision_time     = 17-MAR-2010 21:00:33
Current_time      = 17-MAR-2010 21:02:10

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.6[dC]
```





```
Filename          = III_p_008_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#32770
Solvent           = CHLOROFORM-D
Creation_time     = 18-OCT-2007 01:21:00
Revision_time     = 18-OCT-2007 01:03:08
Current_time      = 17-MAR-2010 21:02:43

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 46
Total_scans       = 46

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.6[dC]
```



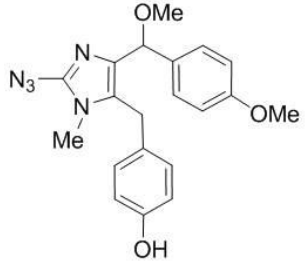


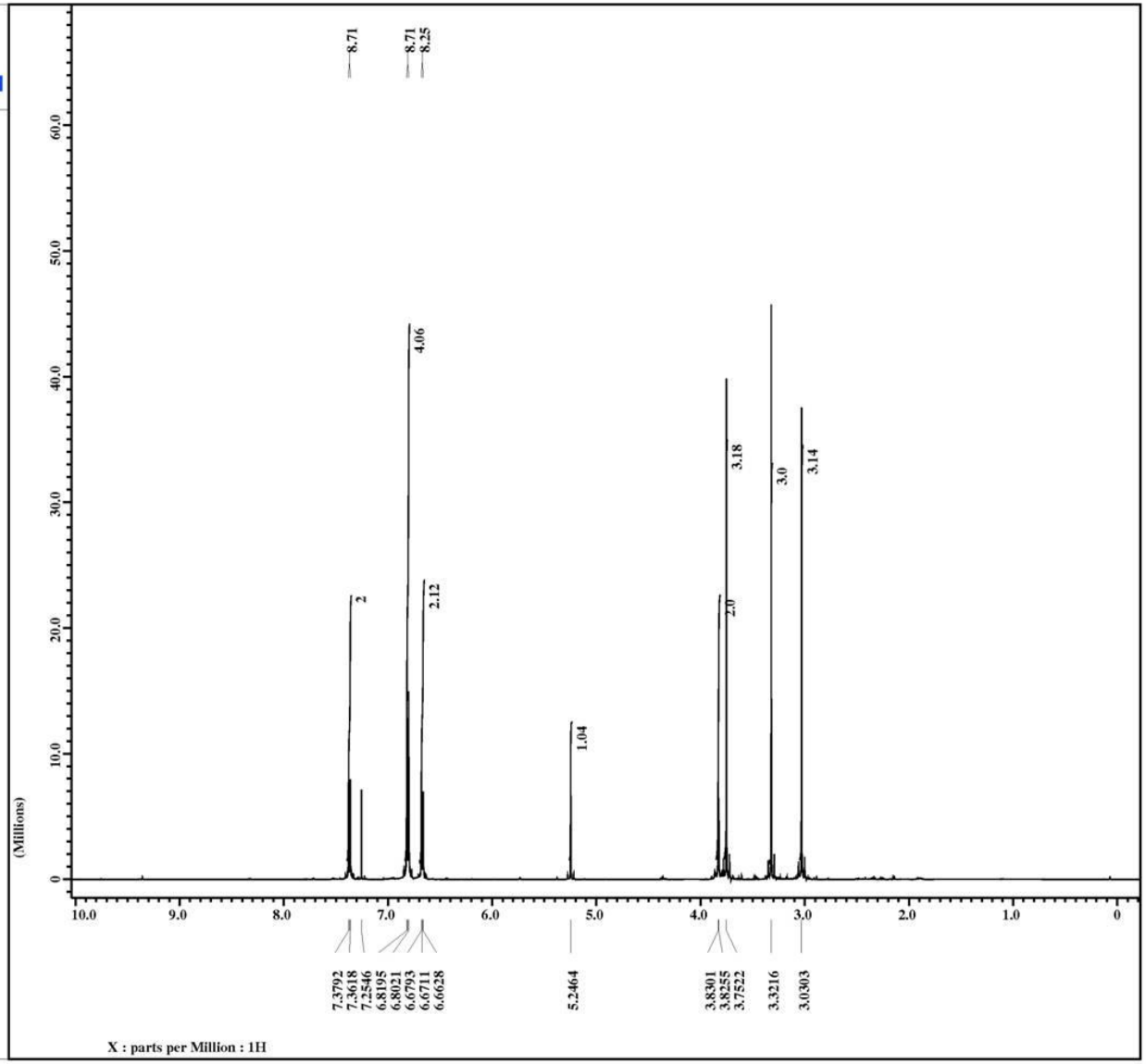
APPENDIX 39

^{1}H and ^{13}C NMR Spectra of

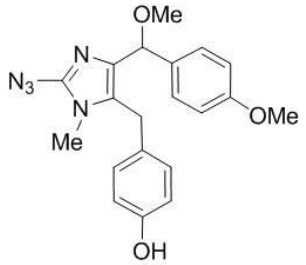
2-Azio-5-(4-hydroxybenzyl)-4-[methoxy-(4-methoxy)phenyl]methyl-1-methyl-1*H*-imidazole (**159**)

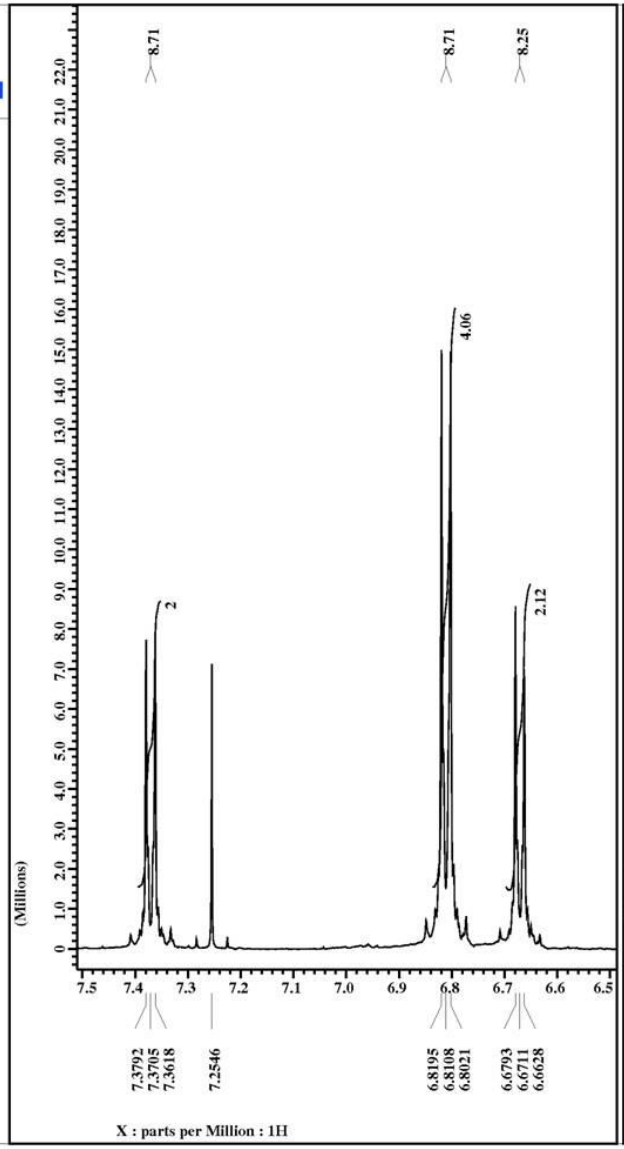
Filename	= III_p_031_pure-5.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#719303
Solvent	= CHLOROFORM-D
Creation_time	= 30-OCT-2007 00:44:49
Revision_time	= 17-MAR-2010 21:12:38
Current_time	= 17-MAR-2010 21:12:56
Comment	= Single Pulse Experime
Data_format	= 1D REAL
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 18
Relaxation_delay	= 4[s]
Temp_get	= 25[dC]
Unblank_time	= 2[us]

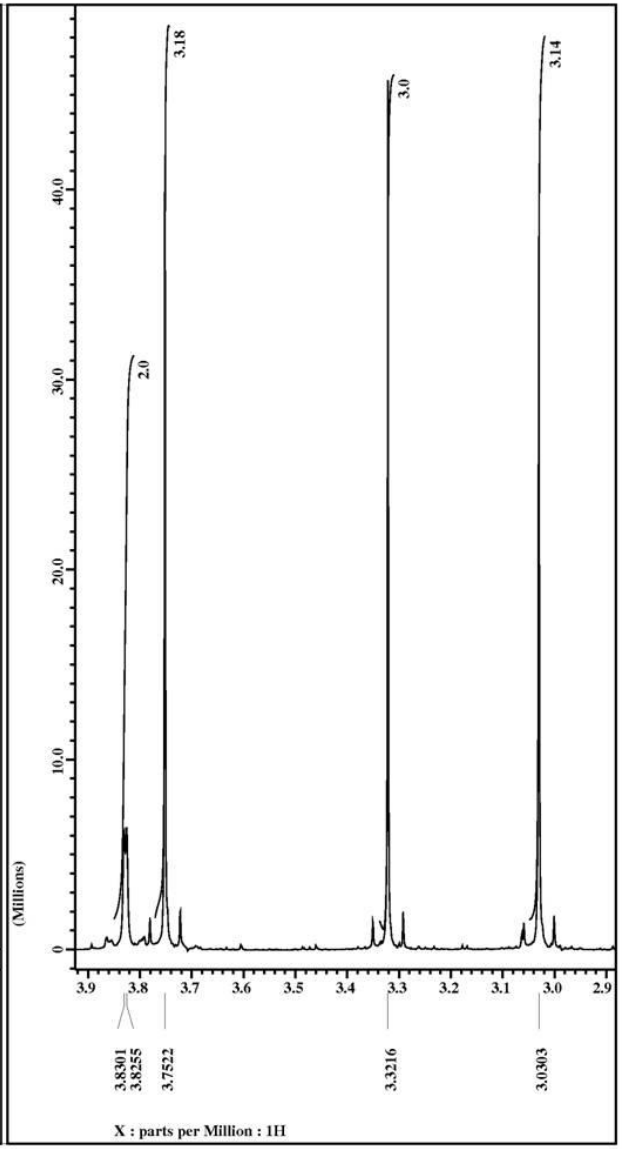


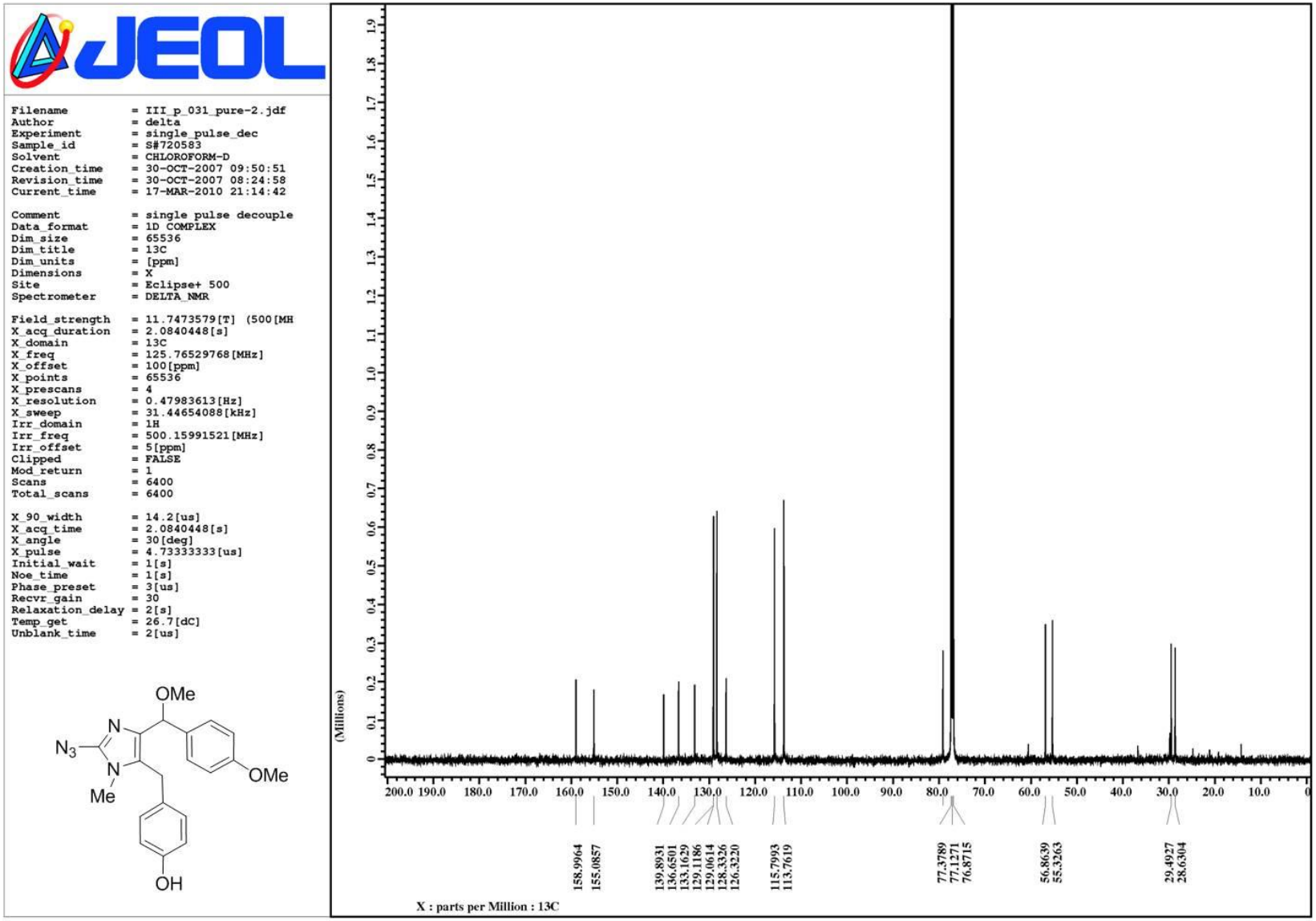


Filename	= III_p_031_pure-5.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#719303
Solvent	= CHLOROFORM-D
Creation_time	= 30-OCT-2007 00:44:49
Revision_time	= 17-MAR-2010 21:12:38
Current_time	= 17-MAR-2010 21:14:11
Comment	= Single Pulse Experime
Data_format	= 1D REAL
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 18
Relaxation_delay	= 4[s]
Temp_get	= 25[dC]
Unblank_time	= 2[us]









```
Filename          = III_p_031_pure-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#720583
Solvent           = CHLOROFORM-D
Creation_time     = 30-OCT-2007 09:50:51
Revision_time     = 30-OCT-2007 08:24:58
Current_time      = 17-MAR-2010 21:14:42

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 6400
Total_scans       = 6400

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.7[dC]
Unblank_time      = 2[us]
```

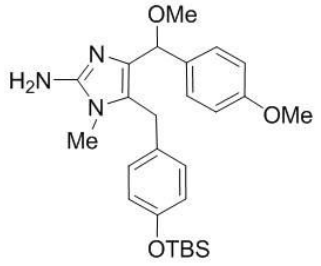
APPENDIX 40

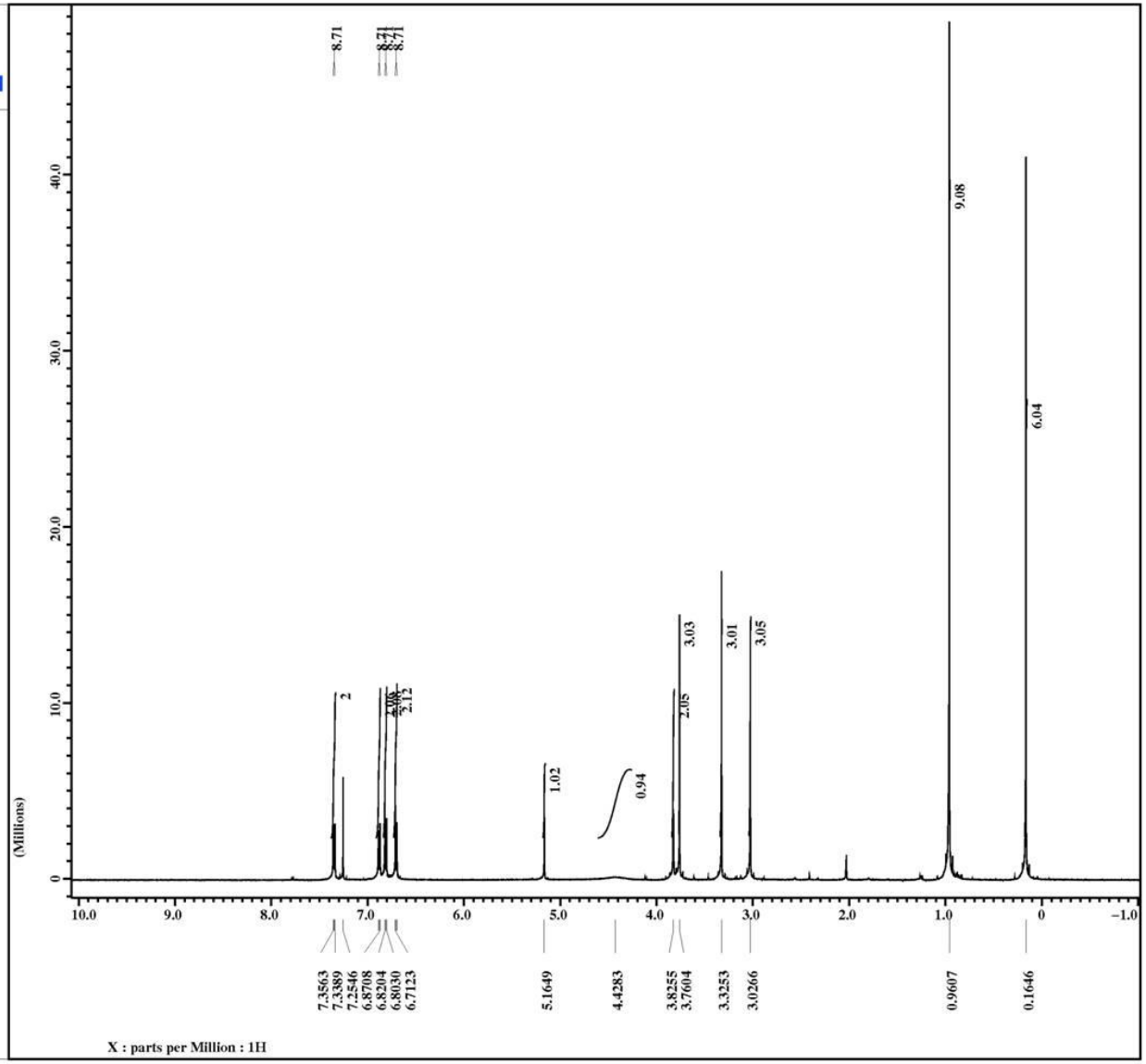
^{1}H and ^{13}C NMR Spectra of

2-Amino-5-(4-*t*-butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]

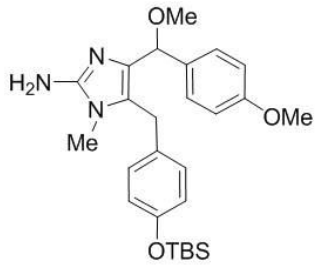
methyl-1-methyl-1*H*-imidazole (**160**)

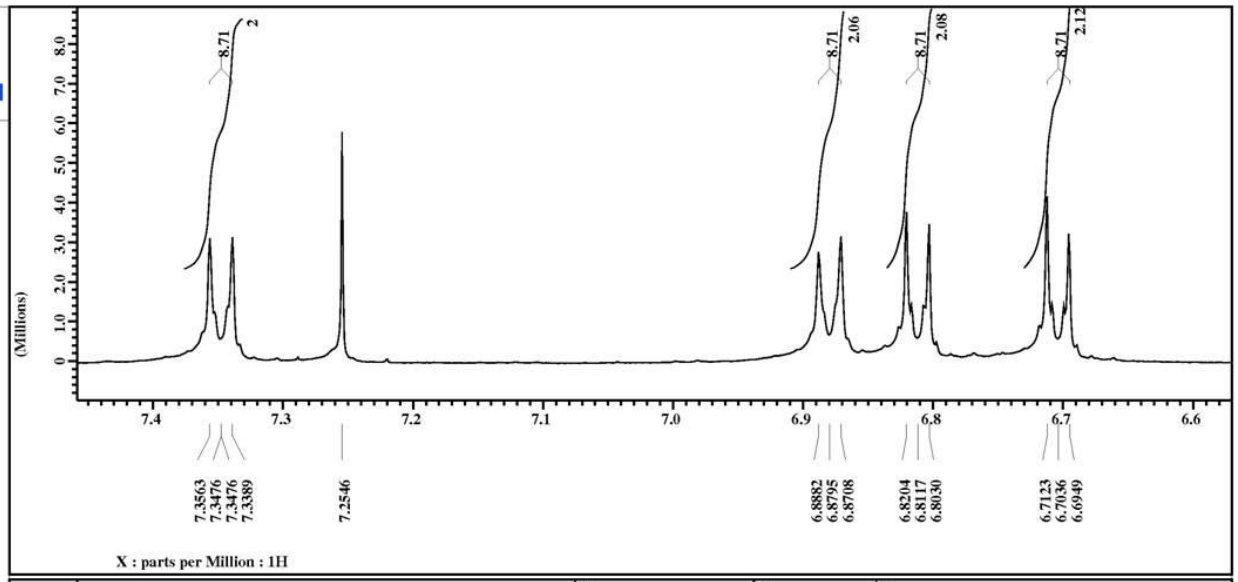
Filename	= III_p_024_amine-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#26360
Solvent	= CHLOROFORM-D
Creation_time	= 26-OCT-2007 05:27:53
Revision_time	= 17-MAR-2010 21:21:12
Current_time	= 17-MAR-2010 21:21:37
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 19
Relaxation_delay	= 4[s]
Temp_get	= 25.4[dC]
Unblank_time	= 2[us]

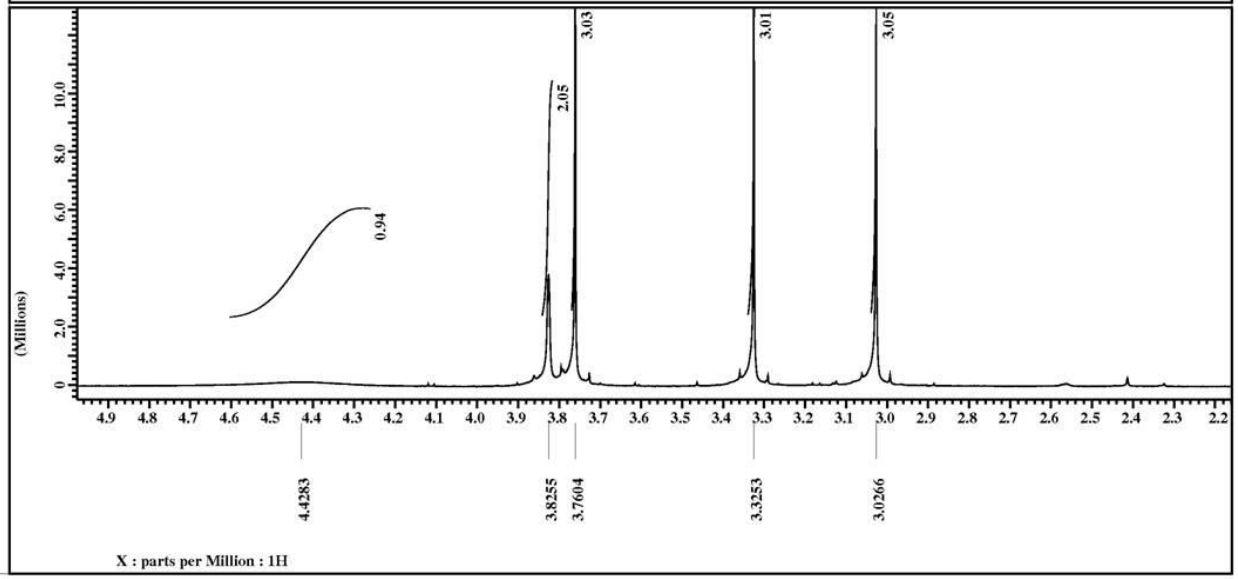




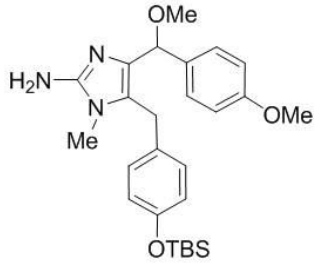
```
Filename          = III_p_024_amine-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#26360
Solvent           = CHLOROFORM-D
Creation_time     = 26-OCT-2007 05:27:53
Revision_time     = 17-MAR-2010 21:21:12
Current_time      = 17-MAR-2010 21:22:27

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 19
Relaxation_delay  = 4[s]
Temp_get          = 25.4[dC]
Unblank_time      = 2[us]
```

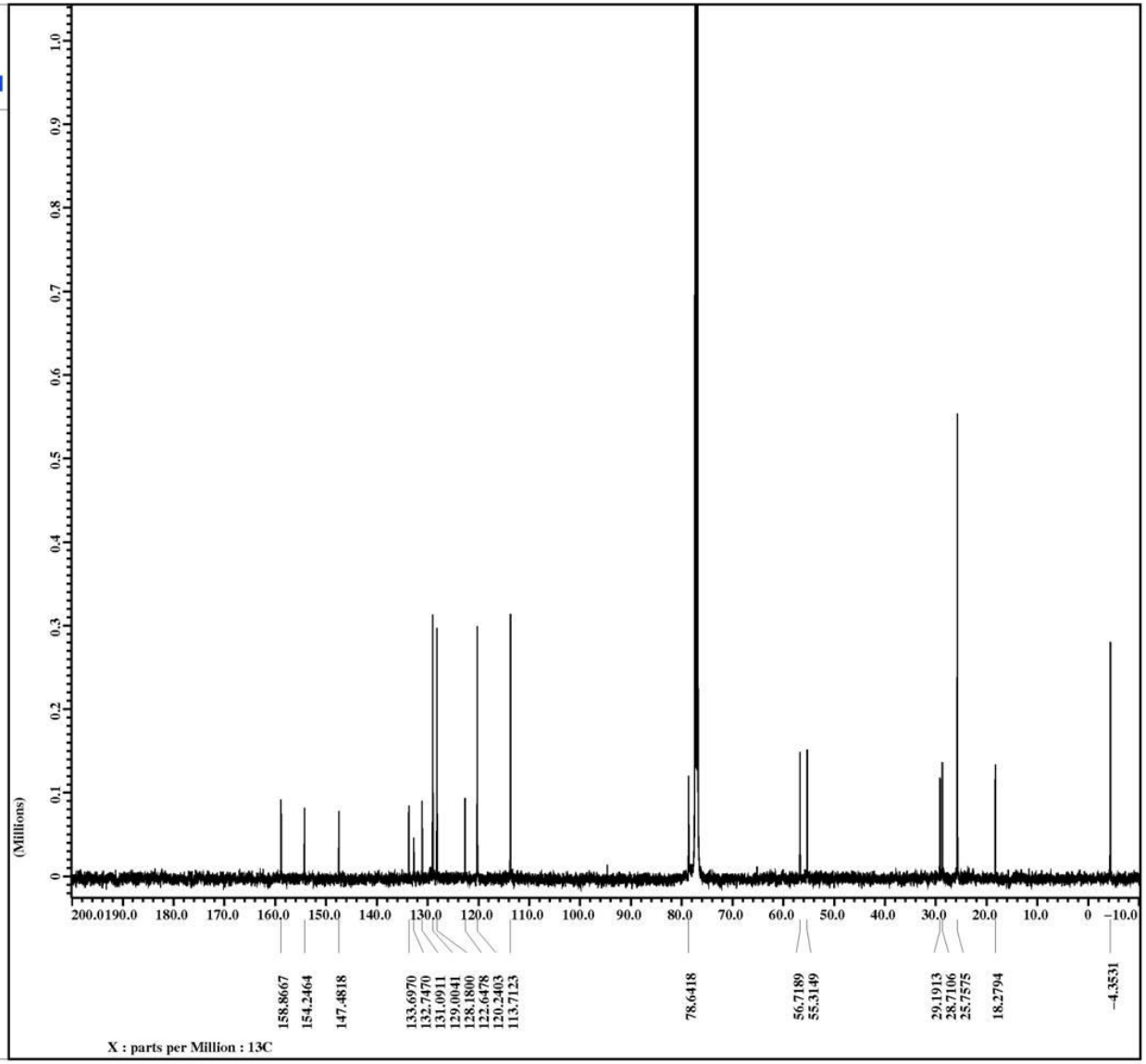






```
Filename          = III_p_024_amine-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#29120
Solvent           = CHLOROFORM-D
Creation_time     = 26-OCT-2007 13:26:15
Revision_time     = 26-OCT-2007 10:19:32
Current_time      = 17-MAR-2010 21:20:06

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 5600
Total_scans       = 5600

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.6[dC]
Unblank_time      = 2[us]
```



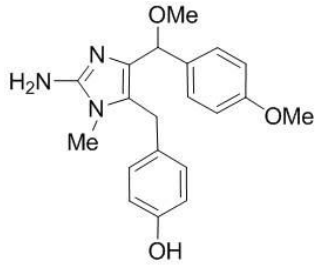


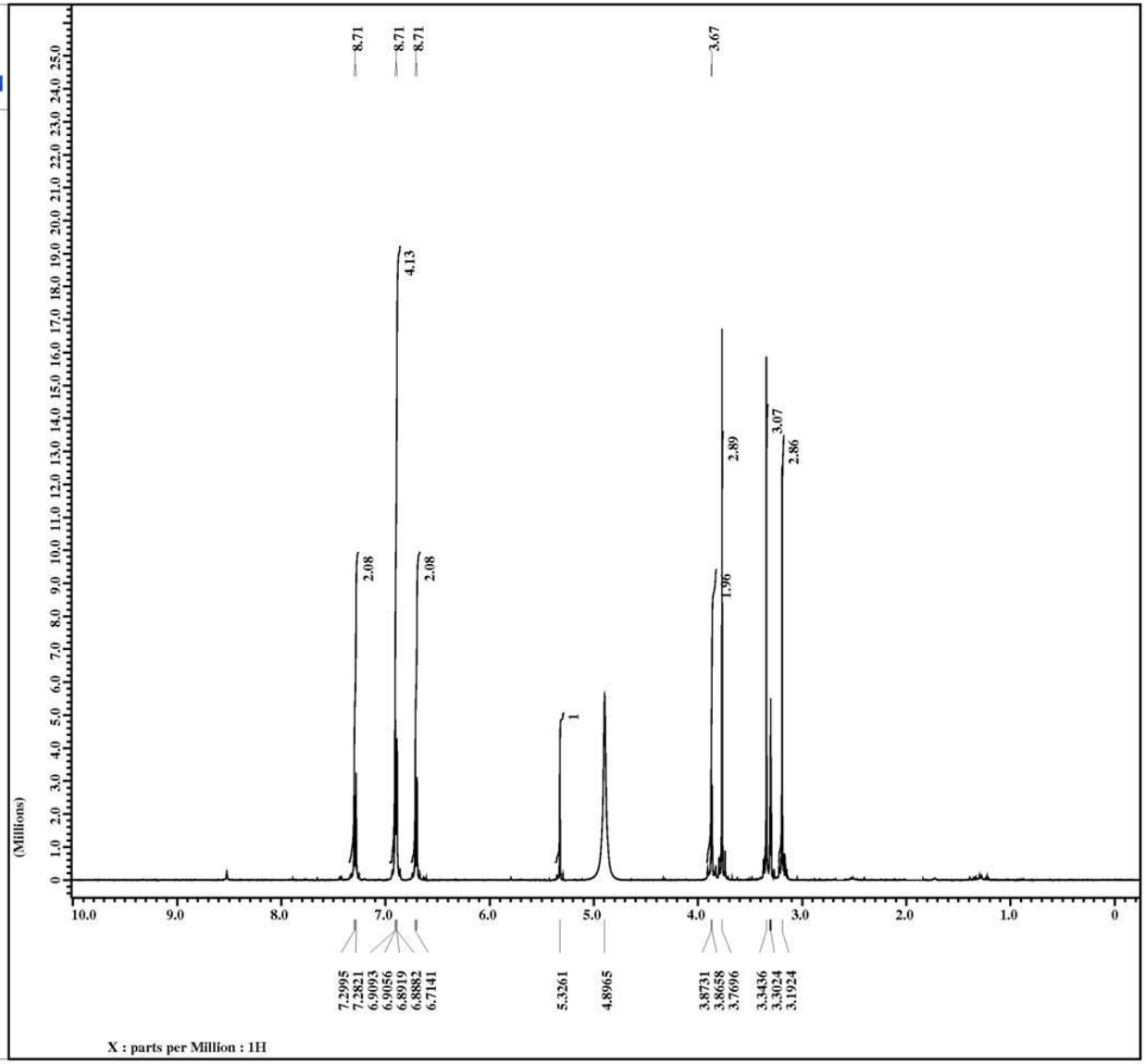
APPENDIX 41

^{1}H and ^{13}C NMR Spectra of

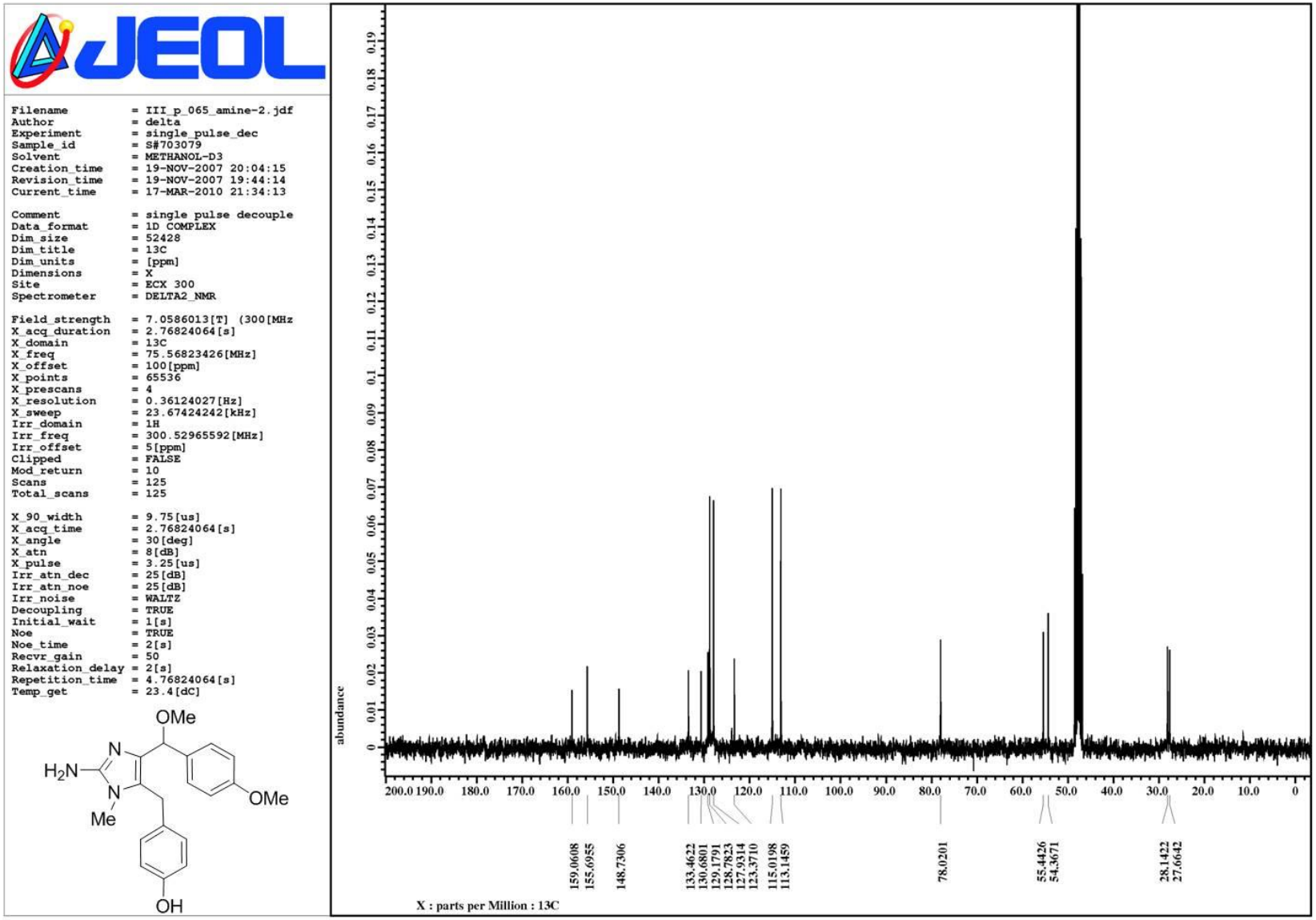
2-Amino-5-(4-hydroxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (14-methoxynaamine A) (**75**)

```
Filename          = III_p_033_amine-5.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#434686
Solvent           = METHANOL-D3
Creation_time     = 30-OCT-2007 16:50:41
Revision_time     = 17-MAR-2010 21:32:23
Current_time      = 17-MAR-2010 21:32:35

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 17
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```





JEOL
Filename = III_p_033_amine-5.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#434686
Solvent = METHANOL-D3
Creation_time = 30-OCT-2007 16:50:41
Revision_time = 17-MAR-2010 21:32:23
Current_time = 17-MAR-2010 21:32:57
Comment = Single Pulse Experime
Data_format = 1D REAL
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.74735791[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 17
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
OMe
OMe
OH
N
N
Me
H_2N
(Millions)
X : parts per Million : 1H
8.71
8.71
8.71
4.13
2.08
2.08
7.2995
7.2908
7.2821
6.9056
6.8919
6.8882
6.7141
6.7054
6.6967
(Millions)
X : parts per Million : 1H
3.67
2.89
1.96
3.07
2.86
3.8731
3.8695
3.8658
3.7696
3.4436
3.3060
3.3024
3.2996
3.1924



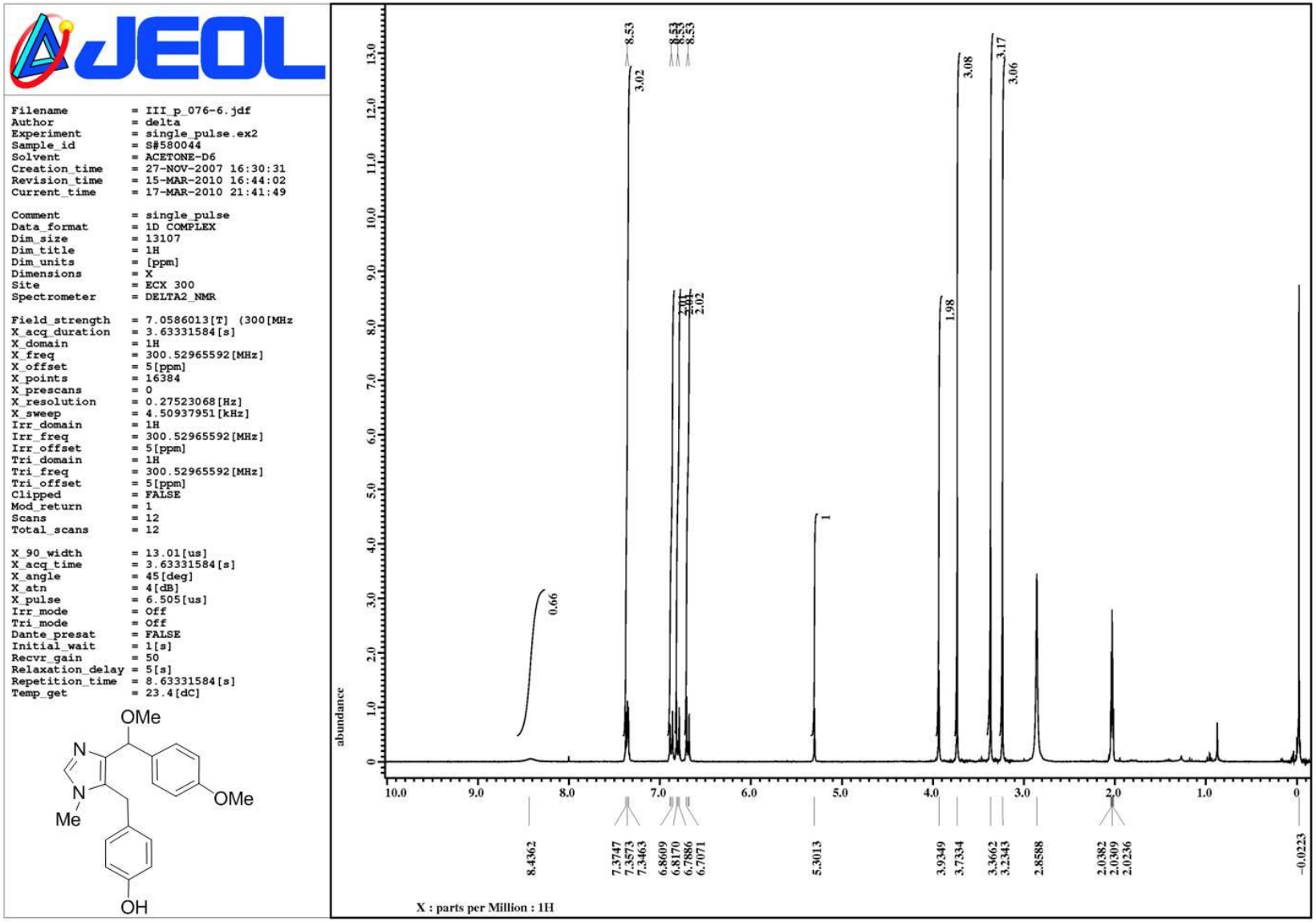
```
Filename          = III_p_065_amine-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#703079
Solvent           = METHANOL-D3
Creation_time     = 19-NOV-2007 20:04:15
Revision_time     = 19-NOV-2007 19:44:14
Current_time      = 17-MAR-2010 21:34:13

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 125
Total_scans       = 125

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.4[dC]
```

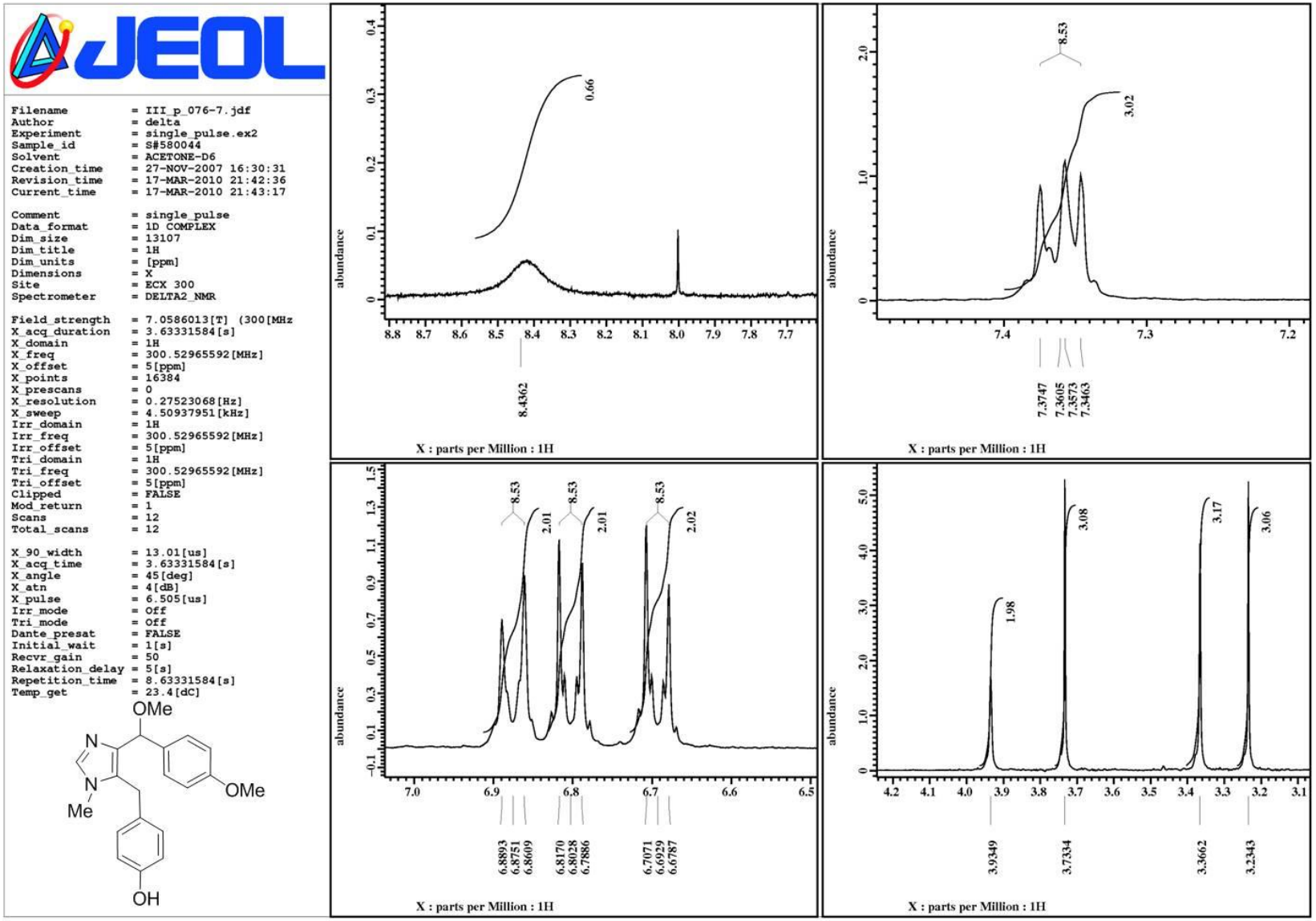
APPENDIX 42

^{1}H and ^{13}C NMR Spectra of

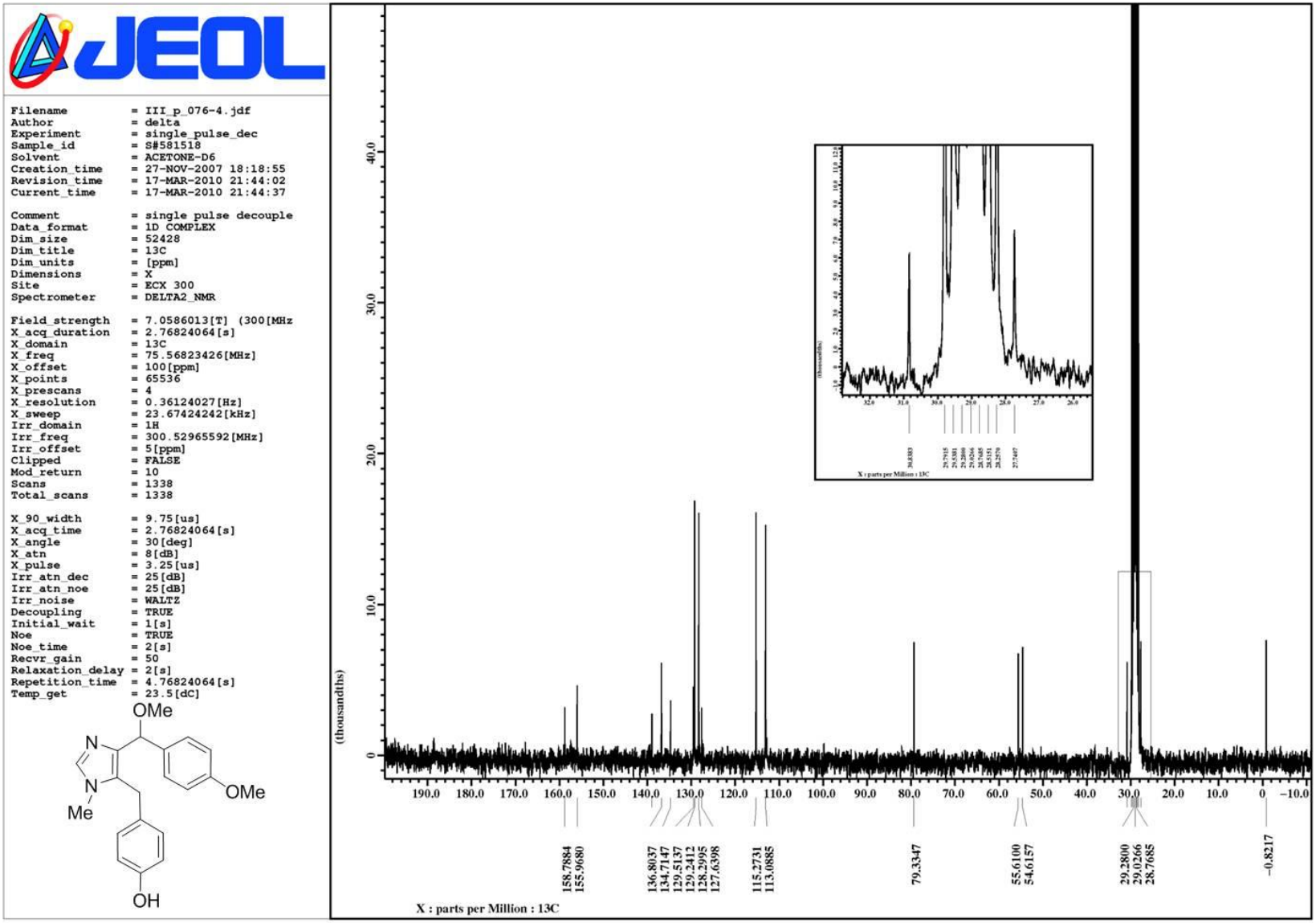
5-(4-Hydroxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole

(**161**)


Filename = III_p_076-6.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#580044
Solvent = ACETONE-D6
Creation_time = 27-NOV-2007 16:30:31
Revision_time = 15-MAR-2010 16:44:02
Current_time = 17-MAR-2010 21:41:49
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 50
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.4[dC]
OMe
N
N
Me
OMe
OH
abundance
8.53
3.02
0.66
1
1.98
3.08
3.17
3.06
2.02
8.4362
7.3747
7.3573
7.3463
6.8609
6.8170
6.7886
6.7071
5.3013
3.9349
3.7334
3.3662
3.2343
2.8588
2.0382
2.0309
2.0236
-0.0223
X : parts per Million : 1H



```
Filename          = III_p_076-7.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#580044
Solvent           = ACETONE-D6
Creation_time     = 27-NOV-2007 16:30:31
Revision_time     = 17-MAR-2010 21:42:36
Current_time      = 17-MAR-2010 21:43:17

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.4[dC]
```

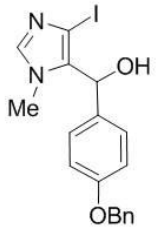

Filename = III_p_076-4.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#581518
Solvent = ACETONE-D6
Creation_time = 27-NOV-2007 18:18:55
Revision_time = 17-MAR-2010 21:44:02
Current_time = 17-MAR-2010 21:44:37
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 1338
Total_scans = 1338
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.5[dC]
OMe
N
N
Me
OMe
OH
(thousandths)
40.0
30.0
20.0
10.0
0
190.0 180.0 170.0 160.0 150.0 140.0 130.0 120.0 110.0 100.0 90.0 80.0 70.0 60.0 50.0 40.0 30.0 20.0 10.0 0 −10.0
158.7884
155.9680
136.8037
134.7147
129.5137
129.2412
128.2995
127.6398
115.2731
113.0885
79.3347
55.6100
54.6157
29.2800
29.0266
28.7685
−0.8217
X : parts per Million : 13C

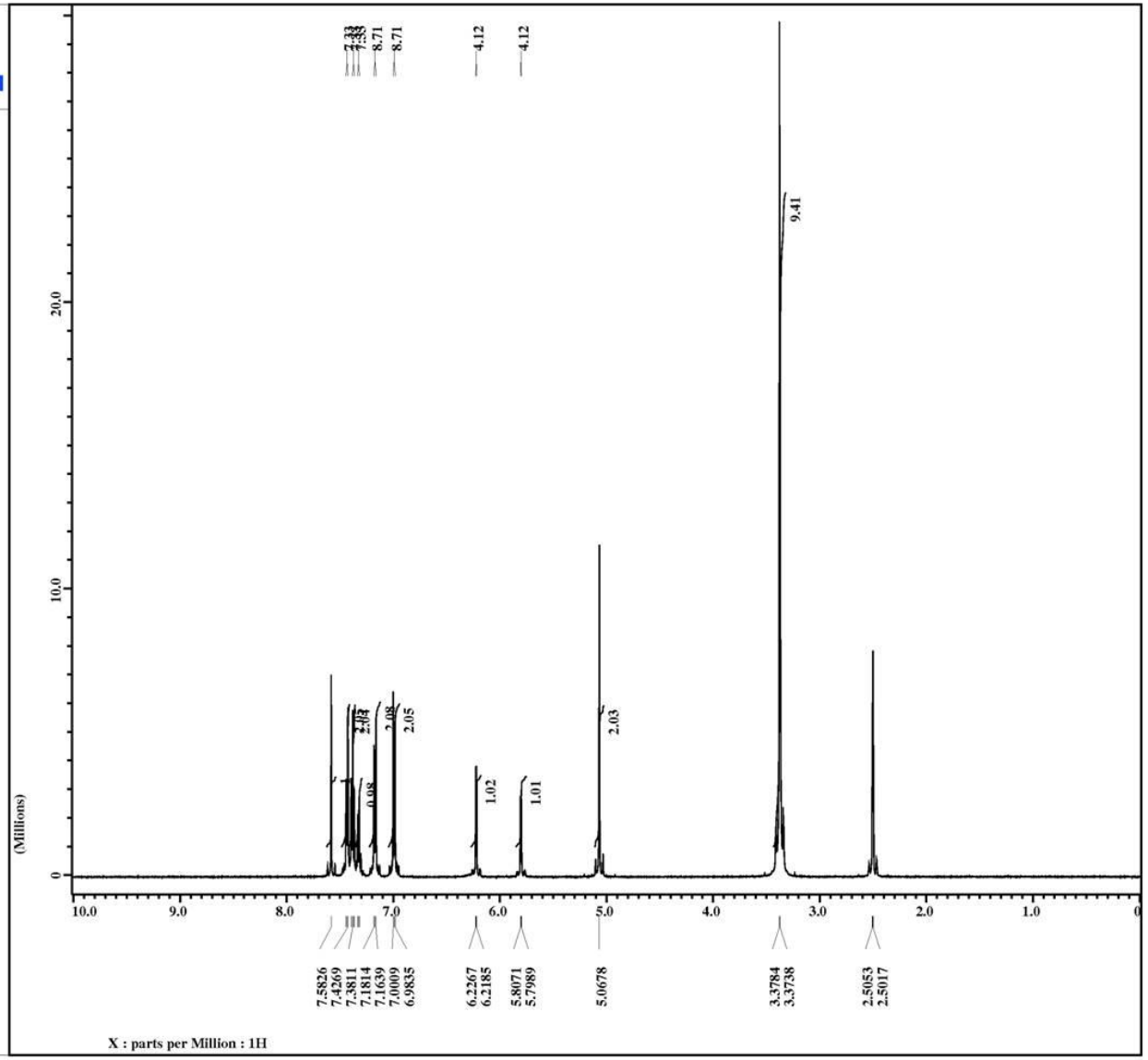
APPENDIX 43

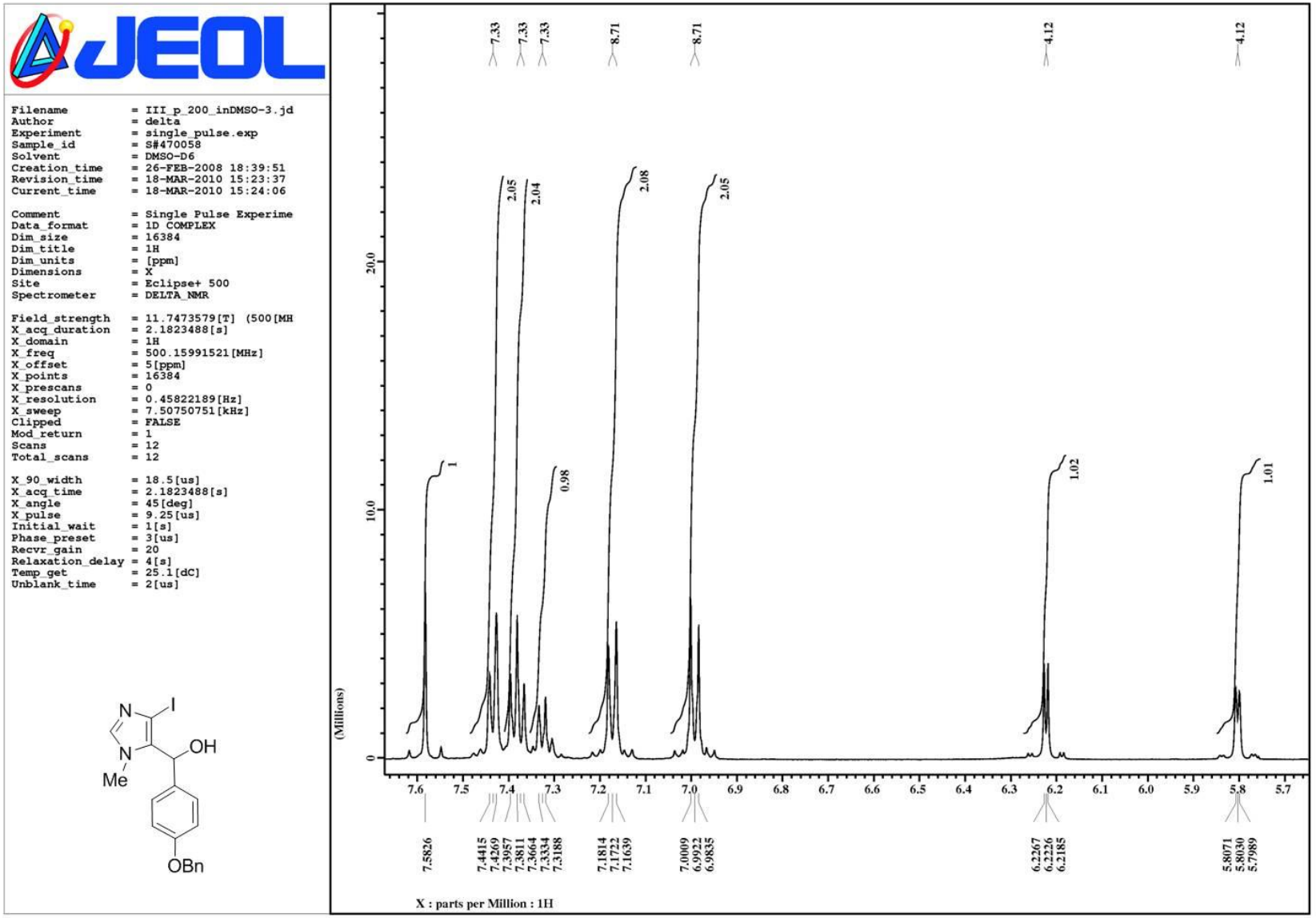
^{1}H and ^{13}C NMR Spectra of

5-[(4-Benzyloxyphenyl)-hydroxy]methyl-4-iodo-1-methyl-1*H*-imidazole (**163**)

Filename	= III_p_200_inDMSO-3.jd
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#470058
Solvent	= DMSO-D6
Creation_time	= 26-FEB-2008 18:39:51
Revision_time	= 18-MAR-2010 15:23:37
Current_time	= 18-MAR-2010 15:23:51
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 20
Relaxation_delay	= 4[s]
Temp_get	= 25.1[dC]
Unblank_time	= 2[us]







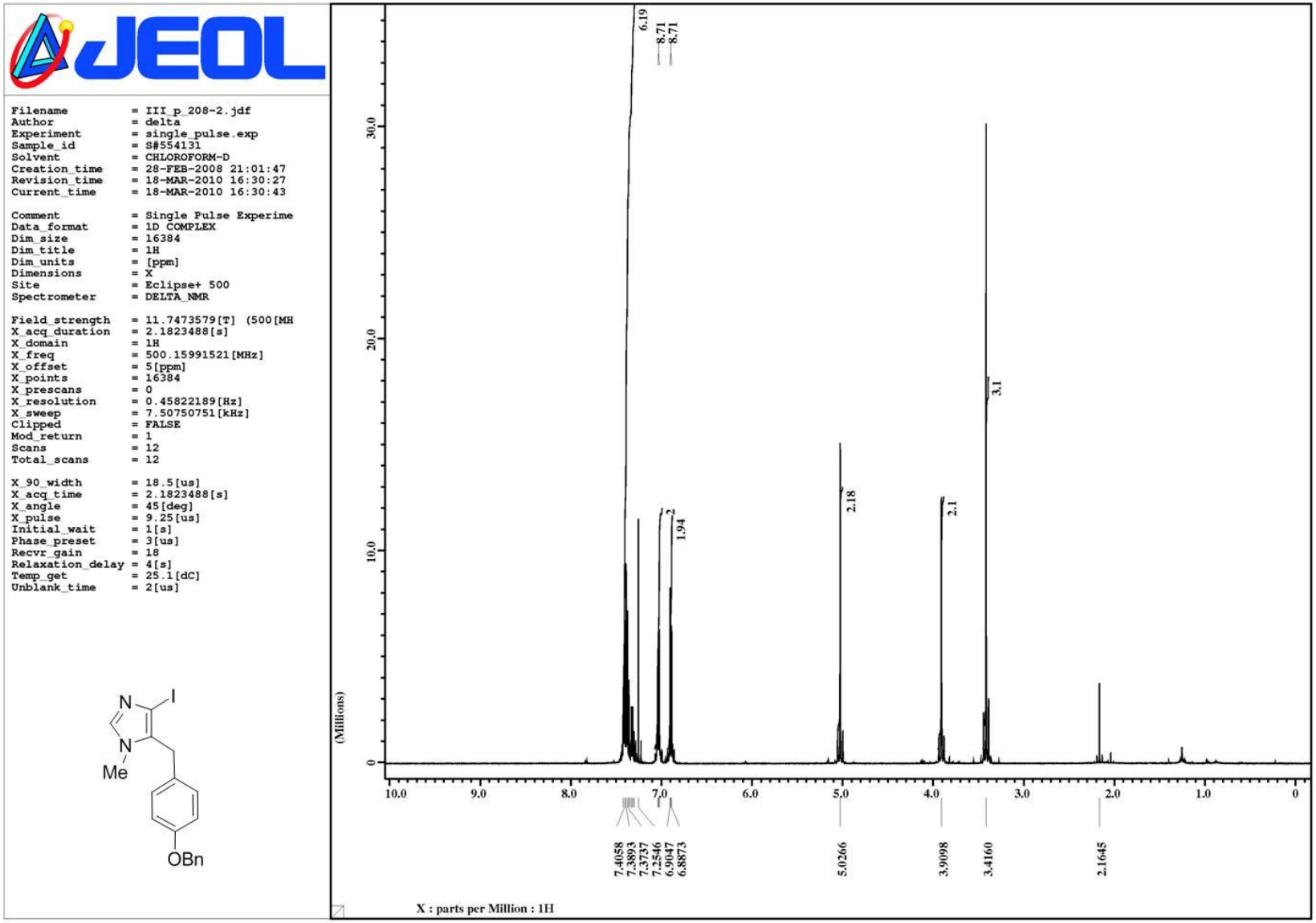
```
Filename          = III_p_200_inDMSO-3.jd
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#470058
Solvent           = DMSO-D6
Creation_time     = 26-FEB-2008 18:39:51
Revision_time     = 18-MAR-2010 15:23:37
Current_time      = 18-MAR-2010 15:24:06

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 25.1[dC]
Unblank_time      = 2[us]
```

JEOL
Filename = III_p_200_inDMSO-3.jd
Author = delta
Experiment = single_pulse_dec
Sample_id = S#741288
Solvent = DMSO-D6
Creation_time = 27-FEB-2008 11:14:26
Revision_time = 18-MAR-2010 15:25:30
Current_time = 18-MAR-2010 15:25:51
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 6400
Total_scans = 6400
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 29
Relaxation_delay = 2[s]
Temp_get = 26.4[dC]
Unblank_time = 2[us]
(Thousands)
600.0
500.0
400.0
300.0
200.0
100.0
0
190.0
180.0
170.0
160.0
150.0
140.0
130.0
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0
157.9014
141.8885
137.6344
135.3796
134.6585
128.9812
128.3822
128.2678
126.9744
115.1087
85.7269
69.7902
66.4976
40.5877
40.0879
39.5842
33.1745
X : parts per Million : 13C
I
OH
N
N
Me
OBn

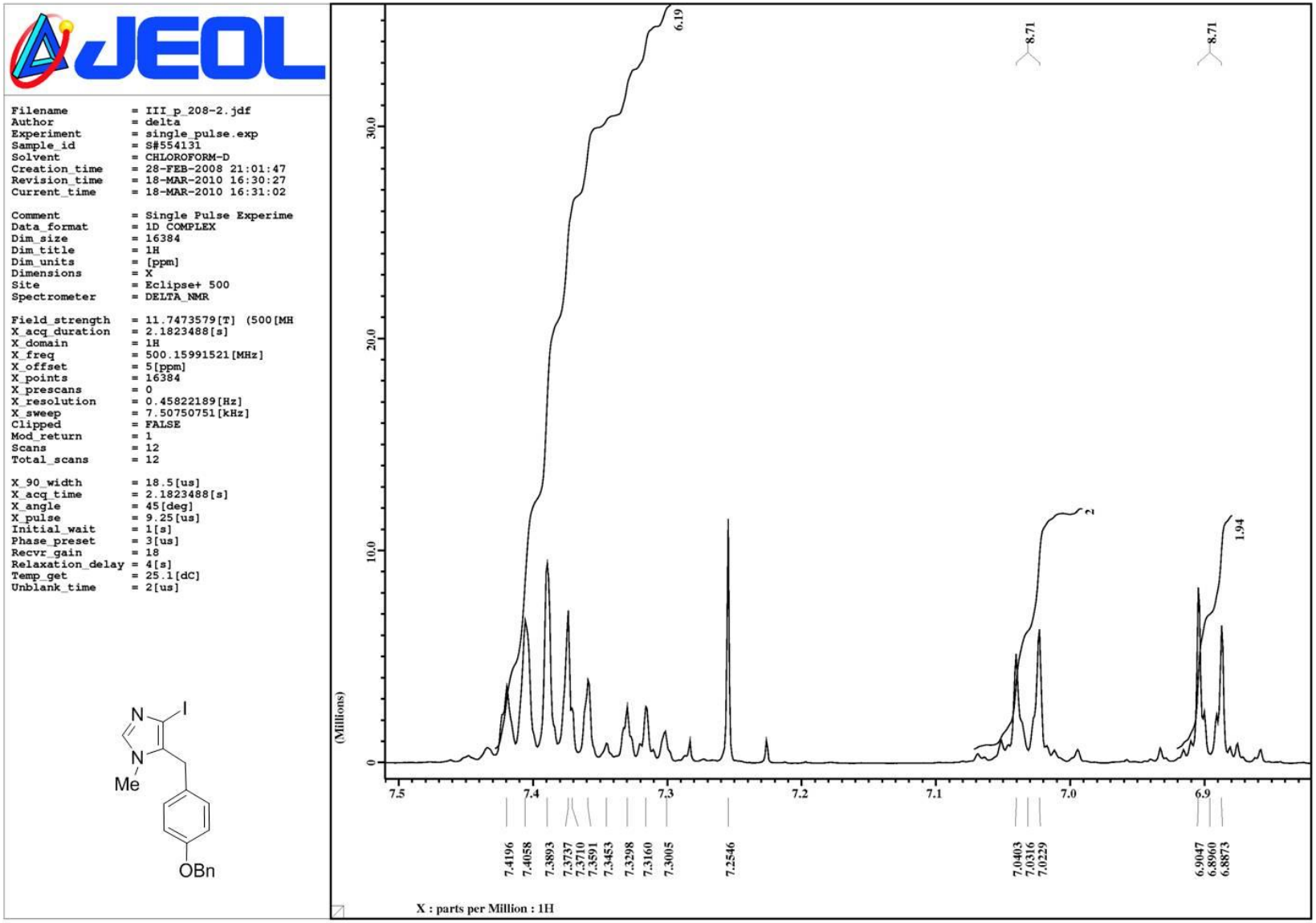
APPENDIX 44

^{1}H and ^{13}C NMR Spectra of

5-(4-Benzoyloxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (**164**)

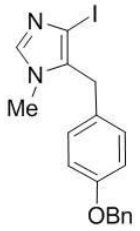


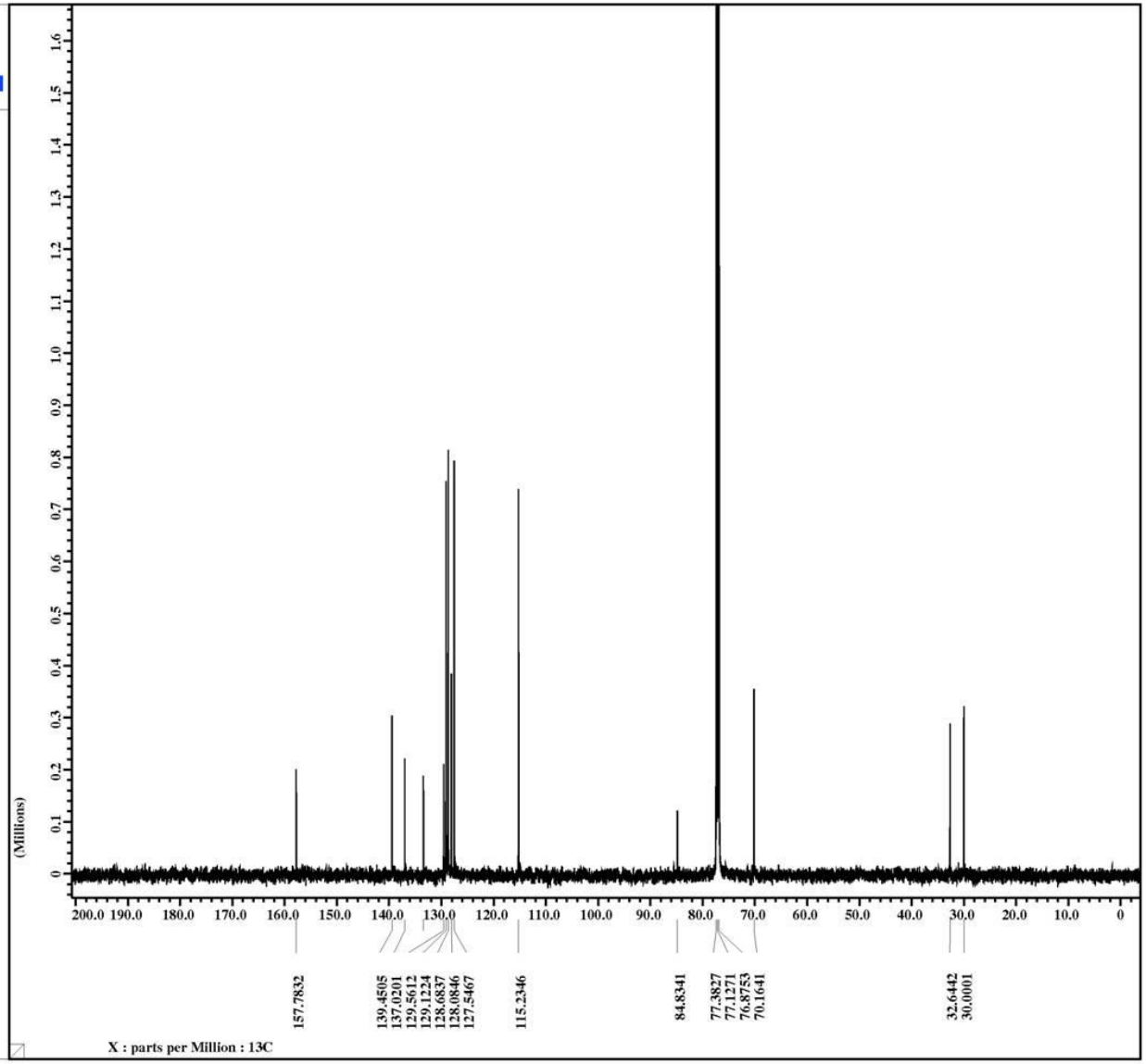
```
Filename          = III_p_208-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#554131
Solvent           = CHLOROFORM-D
Creation_time     = 28-FEB-2008 21:01:47
Revision_time     = 18-MAR-2010 16:30:27
Current_time      = 18-MAR-2010 16:30:43

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 4[s]
Temp_get          = 25.1[dC]
Unblank_time      = 2[us]
```



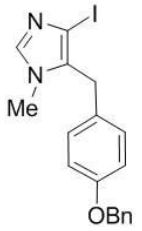
```
Filename          = III_p_208-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#554131
Solvent           = CHLOROFORM-D
Creation_time     = 28-FEB-2008 21:01:47
Revision_time     = 18-MAR-2010 16:30:27
Current_time      = 18-MAR-2010 16:31:02

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 4[s]
Temp_get          = 25.1[dC]
Unblank_time      = 2[us]
```

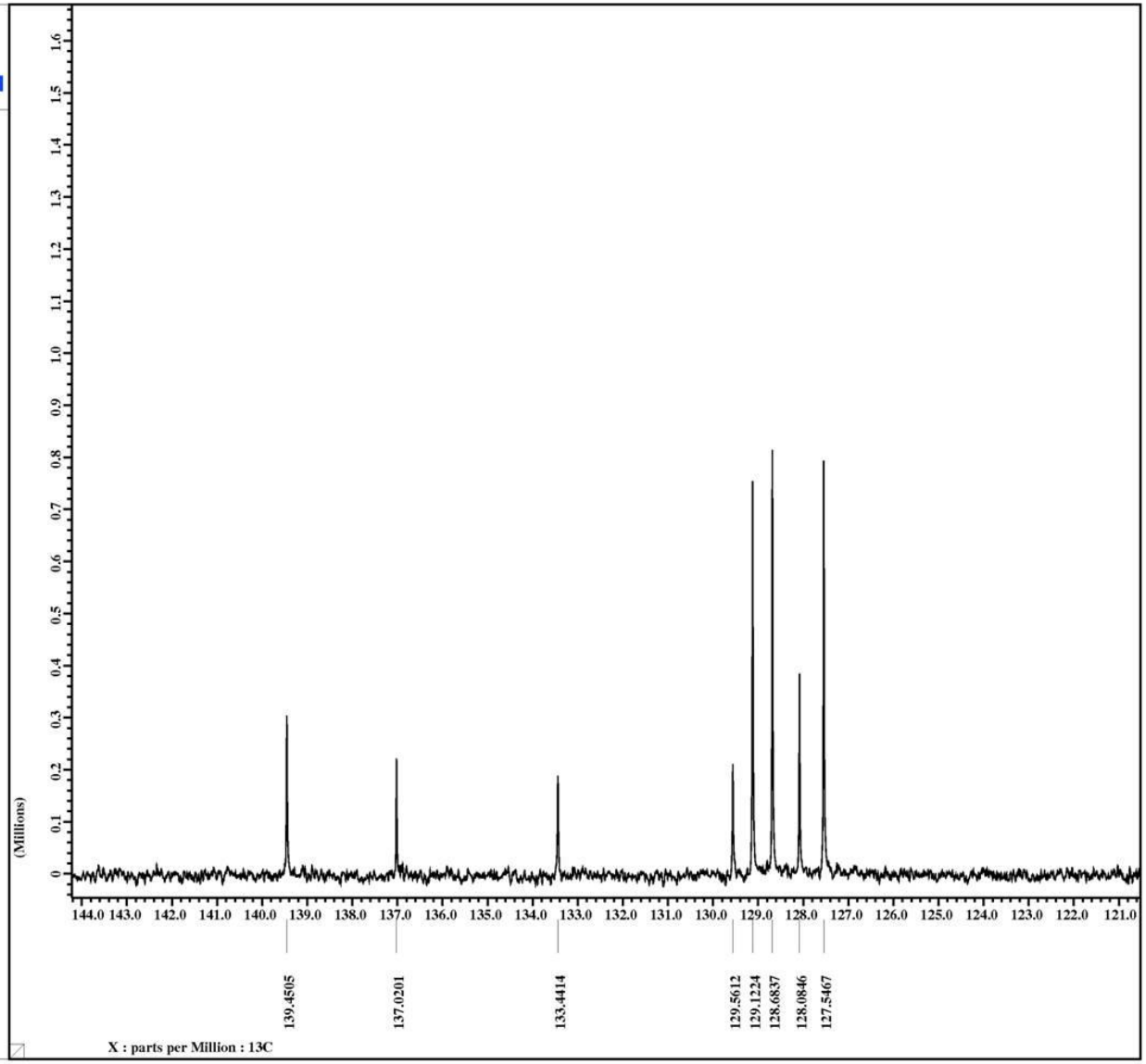
```
Filename          = III_p_208-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#555285
Solvent           = CHLOROFORM-D
Creation_time     = 28-FEB-2008 23:10:49
Revision_time     = 28-FEB-2008 17:35:20
Current_time      = 18-MAR-2010 16:26:09

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T]  (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 1500
Total_scans       = 1500

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.4[dC]
Unblank_time      = 2[us]
```





```
Filename          = III_p_208-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#555285
Solvent           = CHLOROFORM-D
Creation_time     = 28-FEB-2008 23:10:49
Revision_time     = 28-FEB-2008 17:35:20
Current_time      = 18-MAR-2010 16:26:55

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 1500
Total_scans       = 1500

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.4[dC]
Unblank_time      = 2[us]
```



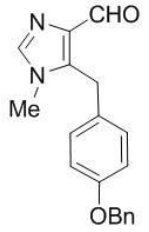


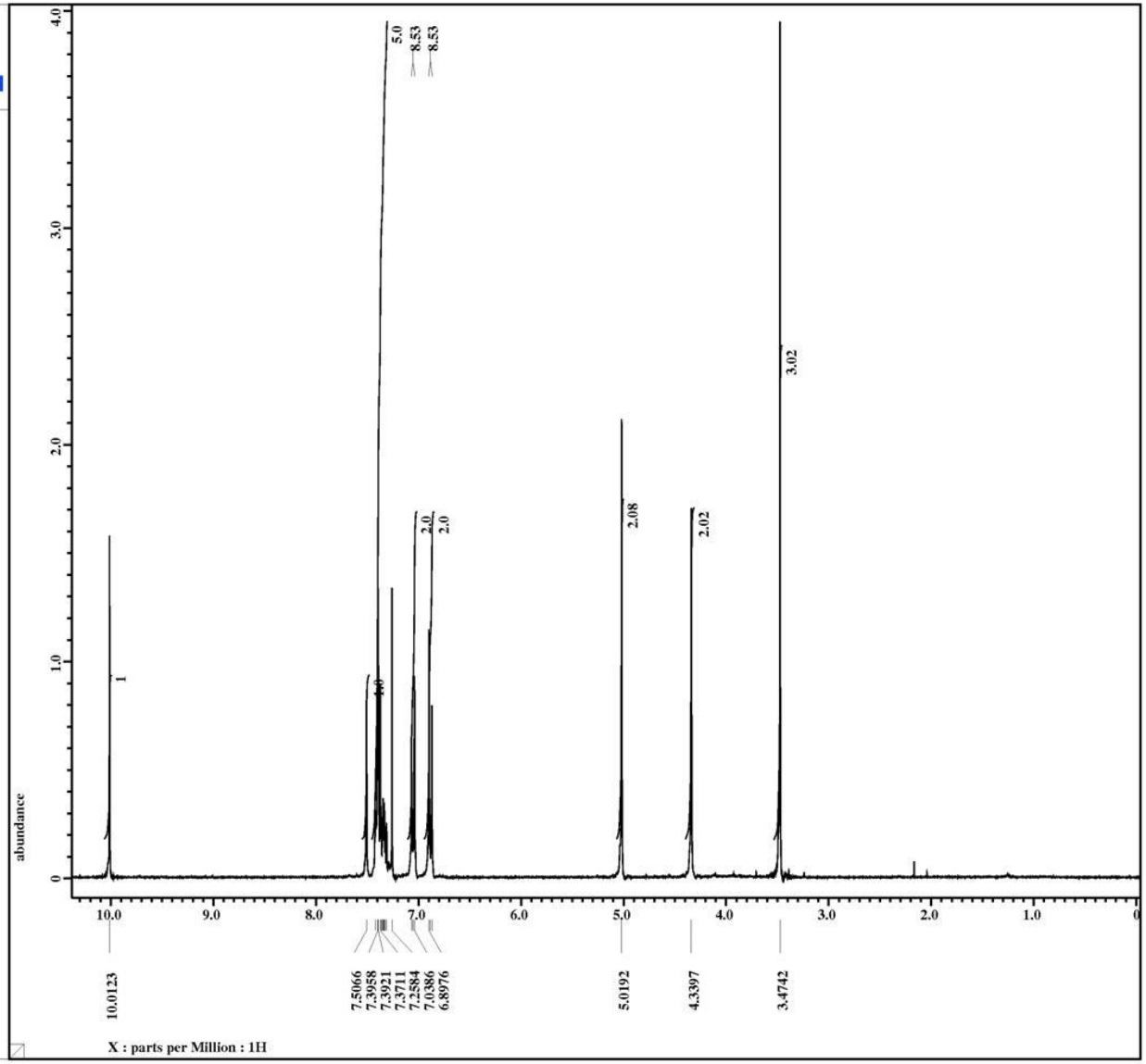
APPENDIX 45

^{1}H and ^{13}C NMR Spectra of

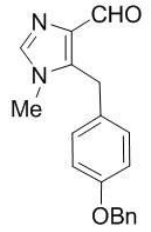
5-(4-Benzoyloxybenzyl)-1-methyl-1*H*-imidazole-4-carboxaldehyde (**166**)

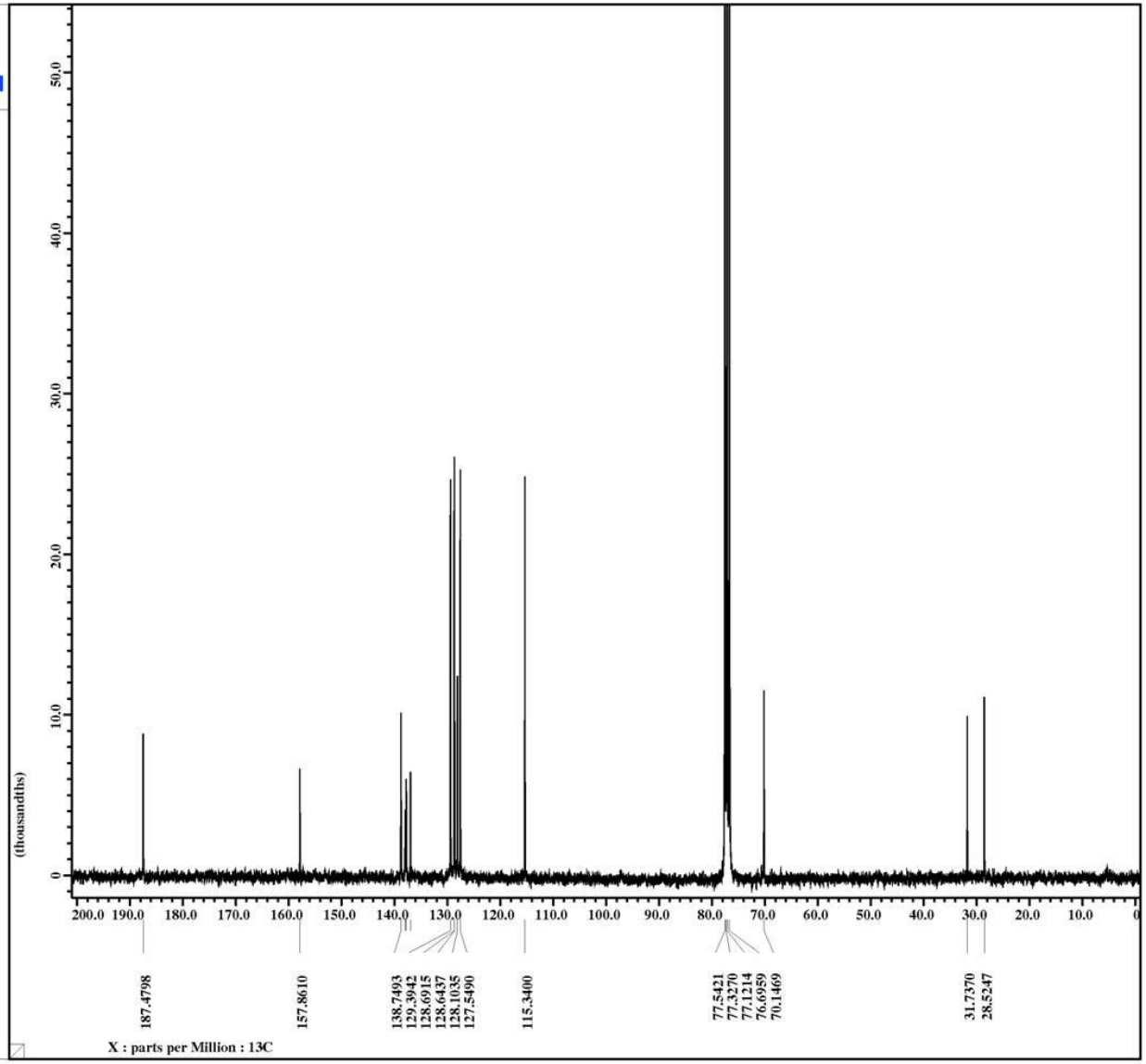
```
Filename          = III_p_272_BnCHO-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#766860
Solvent           = CHLOROFORM-D
Creation_time     = 13-APR-2008 21:30:23
Revision_time     = 18-MAR-2010 16:40:31
Current_time      = 18-MAR-2010 16:40:43

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.3[dC]
```





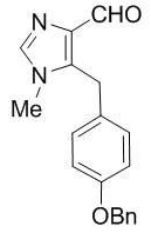
```
Filename          = III_p_272_BnCHO-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#768189
Solvent           = CHLOROFORM-D
Creation_time     = 14-APR-2008 05:59:26
Revision_time     = 14-APR-2008 09:56:23
Current_time      = 18-MAR-2010 16:42:57

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```

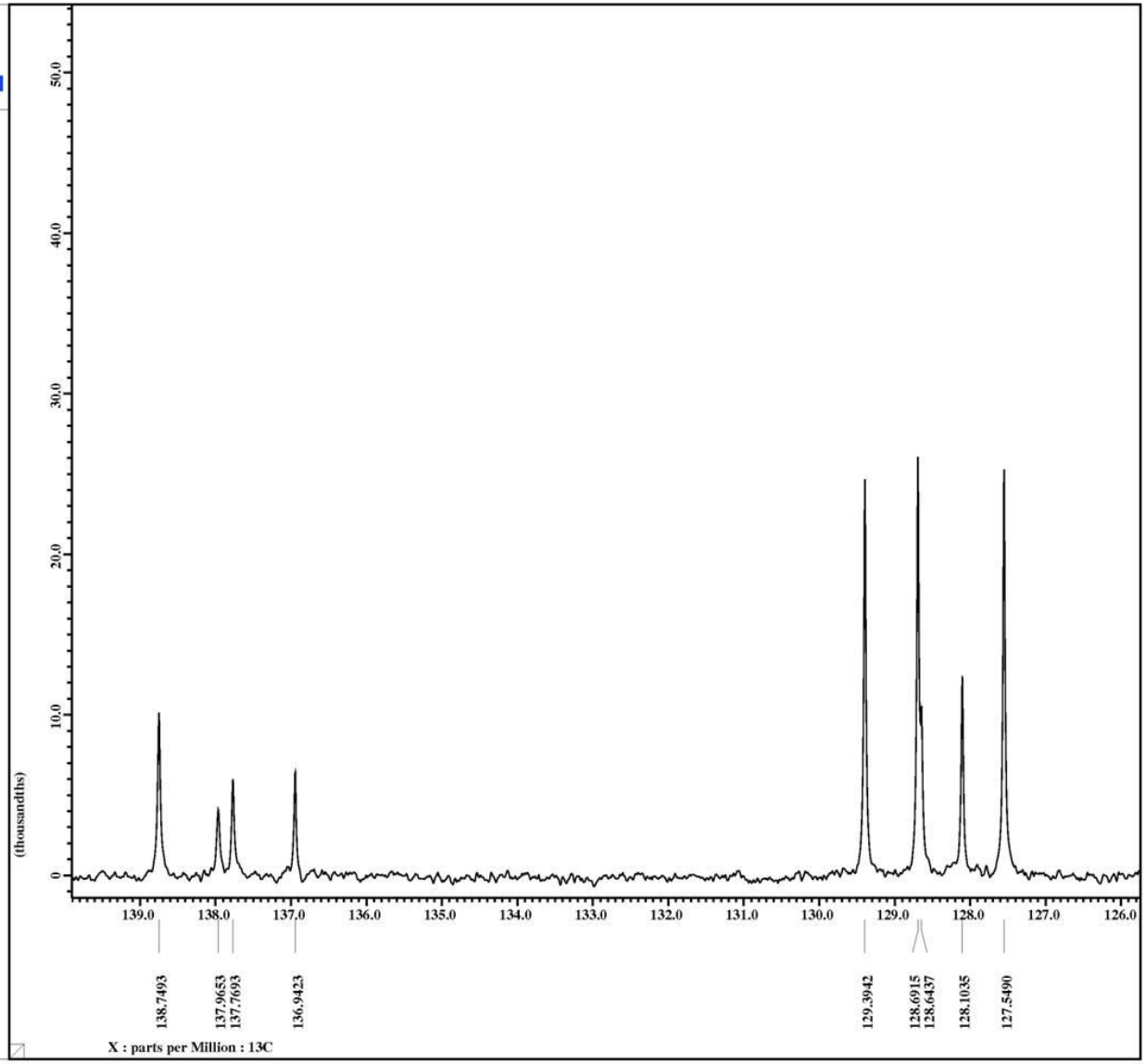




JEOL
Filename = III_p_272_BnCHO-3.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#766860
Solvent = CHLOROFORM-D
Creation_time = 13-APR-2008 21:30:23
Revision_time = 18-MAR-2010 16:40:31
Current_time = 18-MAR-2010 16:40:56
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.509377951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 46
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.3[dC]
CHO
N
N
Me
OBn
abundance
X : parts per Million : 1H
7.5066
7.4196
7.3958
7.3921
7.3711
7.3646
7.3518
7.3454
7.3399
7.3326
7.3334
7.3106
7.2584
7.0670
7.0528
7.0386
6.8976
6.8834
6.8692
8.53
8.53
5.0
1.0
2.0
2.0

```
Filename          = III_p_272_BnCHO-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#768189
Solvent           = CHLOROFORM-D
Creation_time     = 14-APR-2008 05:59:26
Revision_time     = 14-APR-2008 09:56:23
Current_time      = 18-MAR-2010 16:43:09

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```

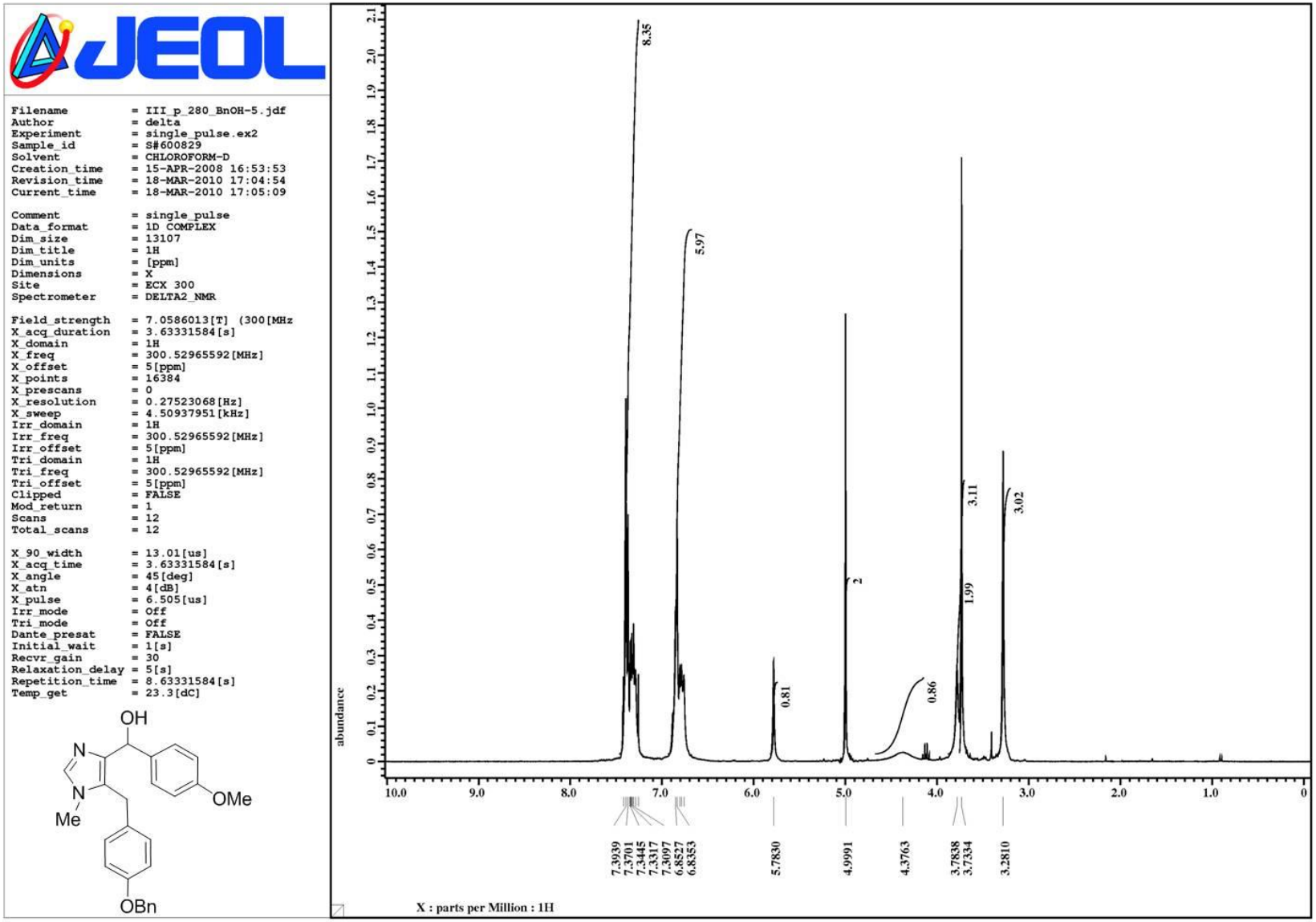




APPENDIX 46

^{1}H and ^{13}C NMR Spectra of

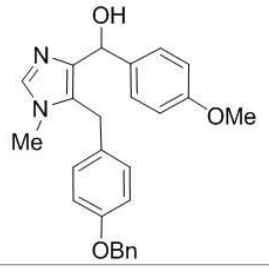
5-(4-Benzoyloxybenzyl)-4-[hydroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**167**)

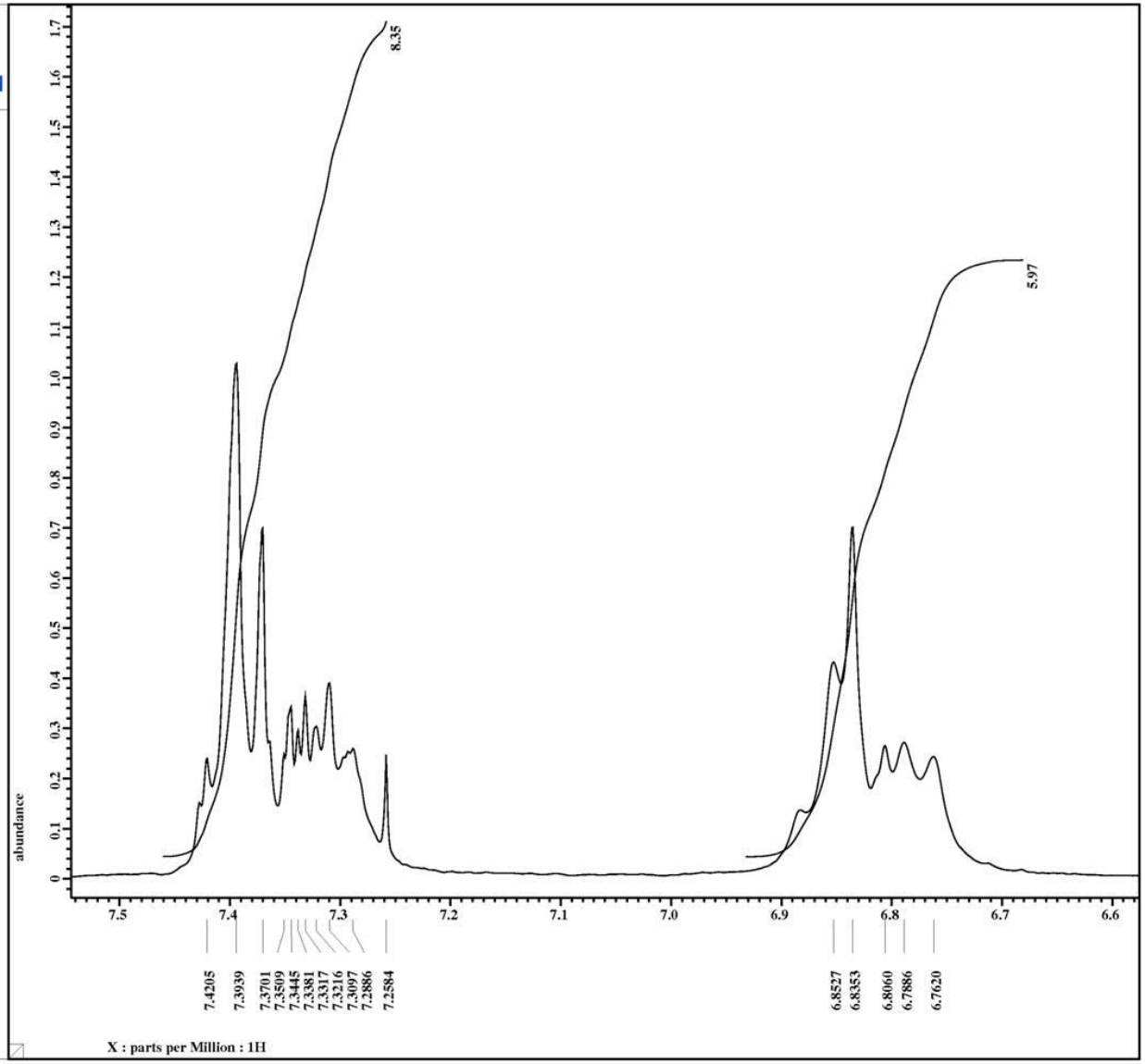


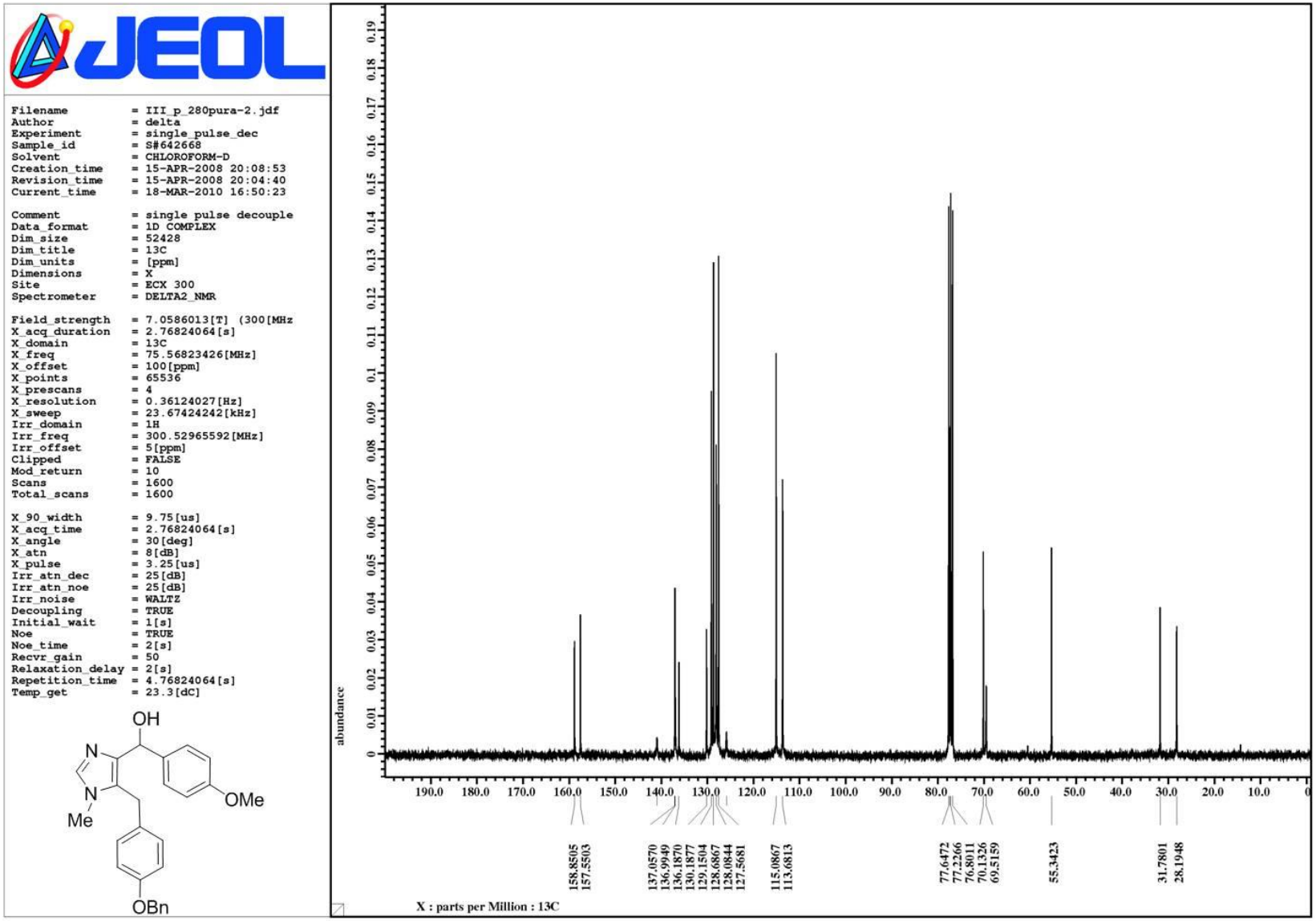
```
Filename          = III_p_280_BnOH-5.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#600829
Solvent           = CHLOROFORM-D
Creation_time     = 15-APR-2008 16:53:53
Revision_time     = 18-MAR-2010 17:04:54
Current_time      = 18-MAR-2010 17:05:09

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.3[dC]
```

JEOL

Filename	= III_p_280_BnOH-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#600829
Solvent	= CHLOROFORM-D
Creation_time	= 15-APR-2008 16:53:53
Revision_time	= 18-MAR-2010 16:54:18
Current_time	= 18-MAR-2010 16:55:08
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.3[dC]

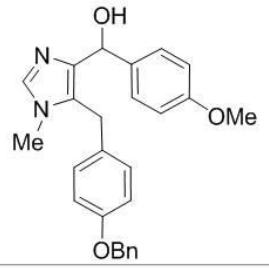


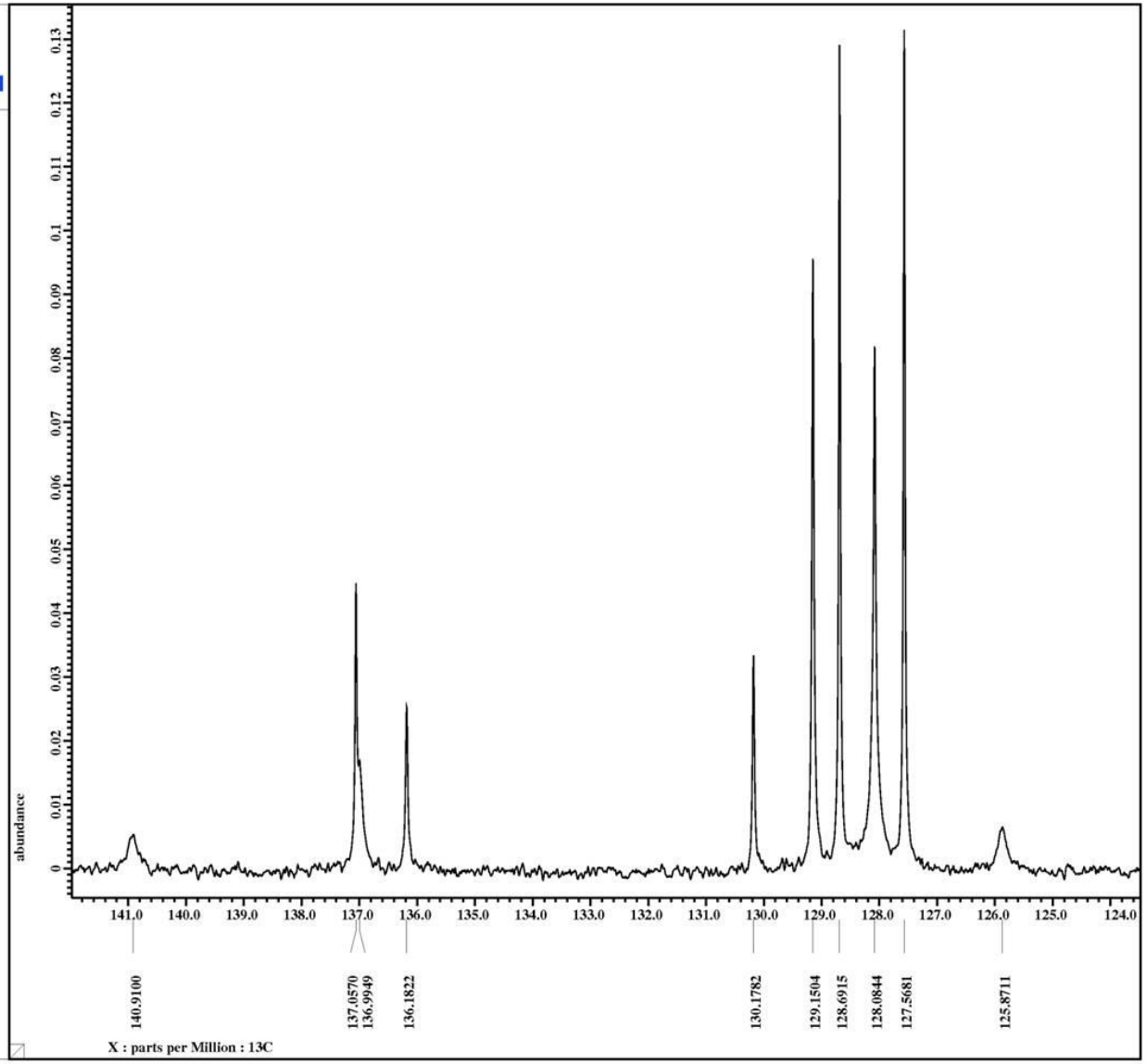




```
Filename          = III_p_280pura-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#642668
Solvent           = CHLOROFORM-D
Creation_time     = 15-APR-2008 20:08:53
Revision_time     = 15-APR-2008 20:04:40
Current_time      = 18-MAR-2010 16:50:23

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1600
Total_scans       = 1600

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```

```
Filename          = III_p_280_BnOH-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#600984
Solvent           = CHLOROFORM-D
Creation_time     = 15-APR-2008 17:58:15
Revision_time     = 18-MAR-2010 16:56:02
Current_time      = 18-MAR-2010 16:56:56

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 804.0
Total_scans       = 804.0

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```





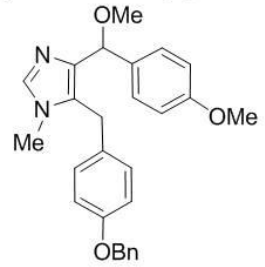
APPENDIX 47

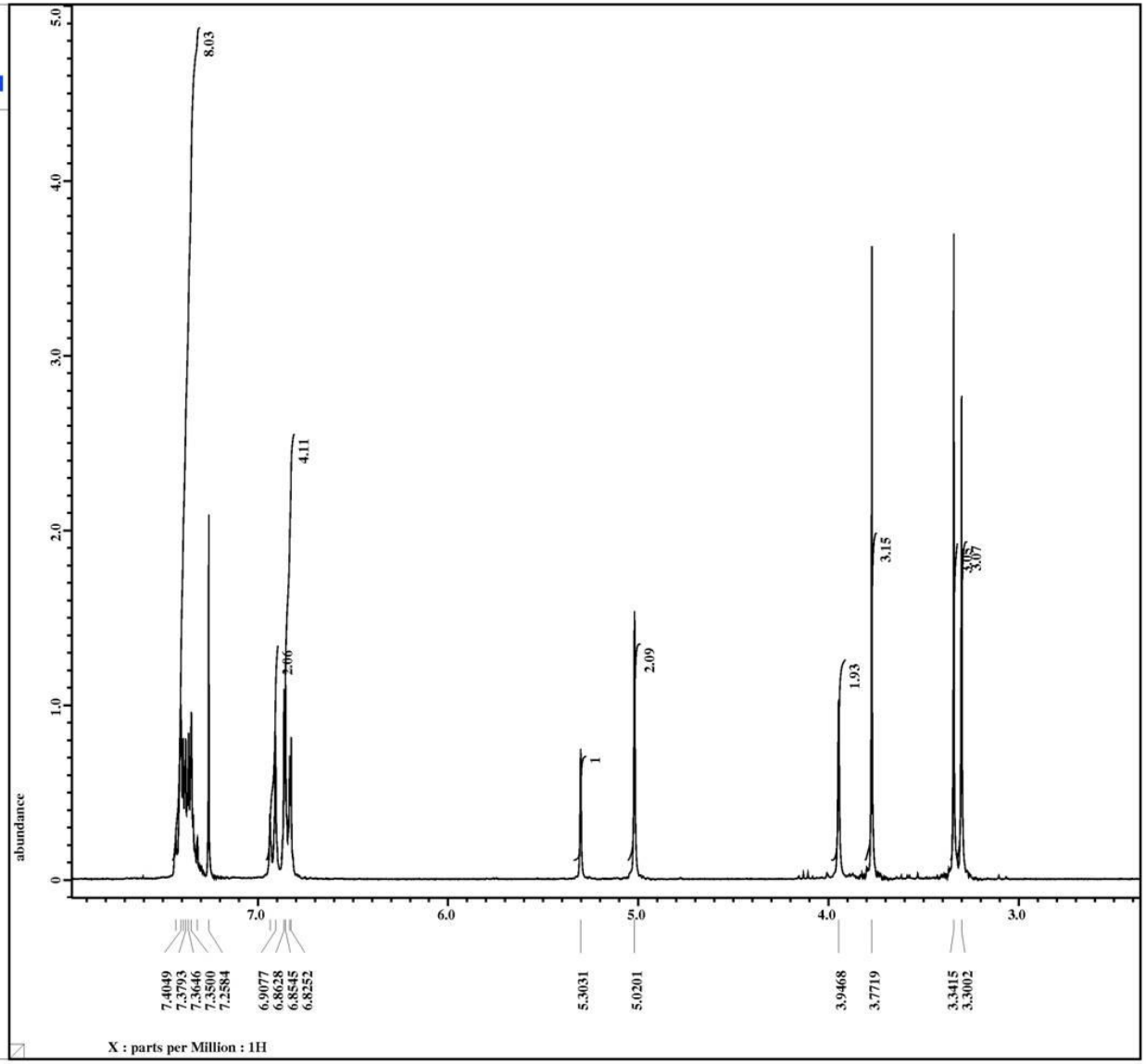
^{1}H and ^{13}C NMR Spectra of

5-(4-Benzoyloxybenzyl)-1-methyl-4-[methoxy-(4-methoxyphenyl)]methyl-1*H*-imidazole (**168**)

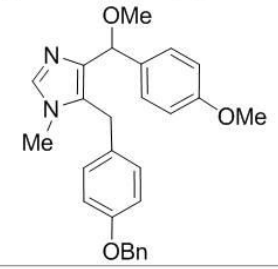
JEOL

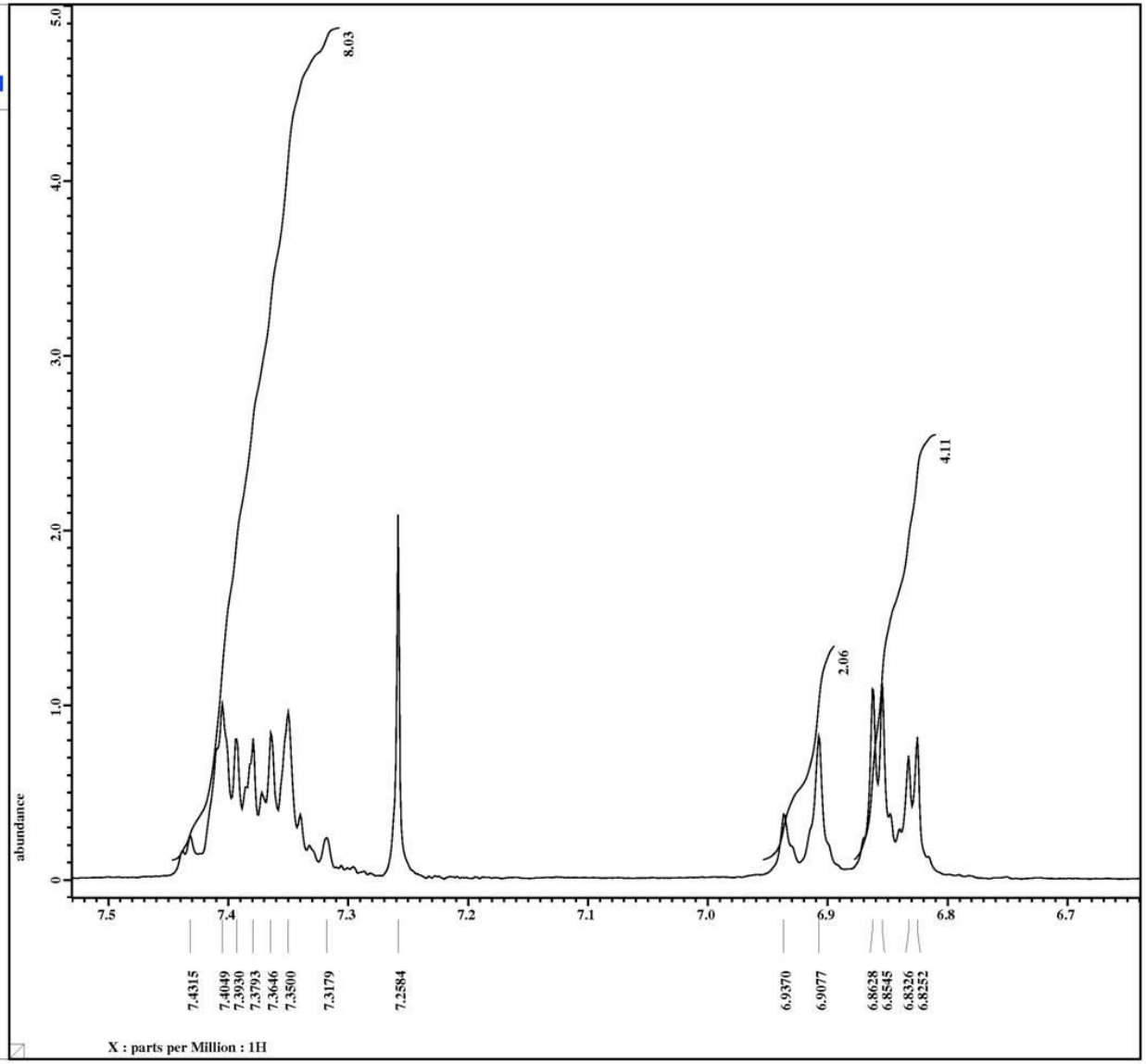
Filename	= IV_p_137_OMe-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#439323
Solvent	= CHLOROFORM-D
Creation_time	= 25-JUL-2008 12:35:06
Revision_time	= 18-MAR-2010 17:31:10
Current_time	= 18-MAR-2010 17:39:12
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 21.6[dC]





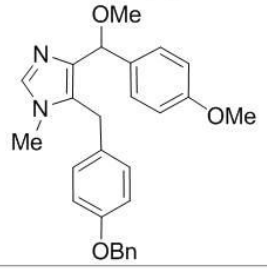
```
Filename          = IV_p_137_OMe-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#439323
Solvent           = CHLOROFORM-D
Creation_time     = 25-JUL-2008 12:35:06
Revision_time     = 18-MAR-2010 17:31:10
Current_time      = 18-MAR-2010 17:39:28

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 21.6[dC]
```

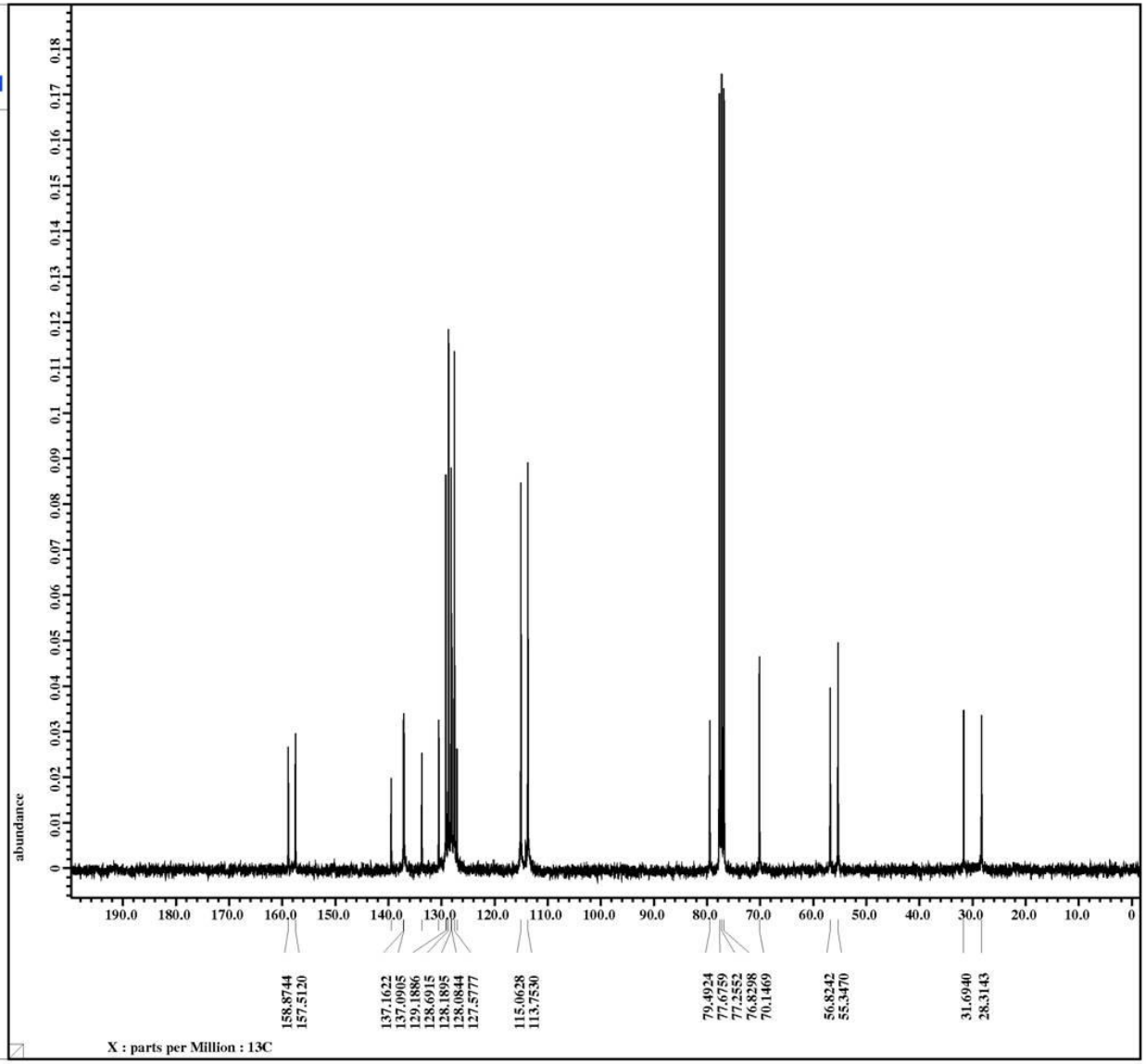




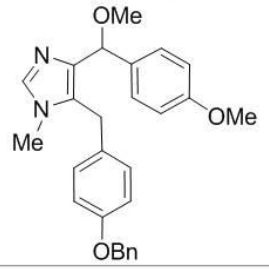
JEOL

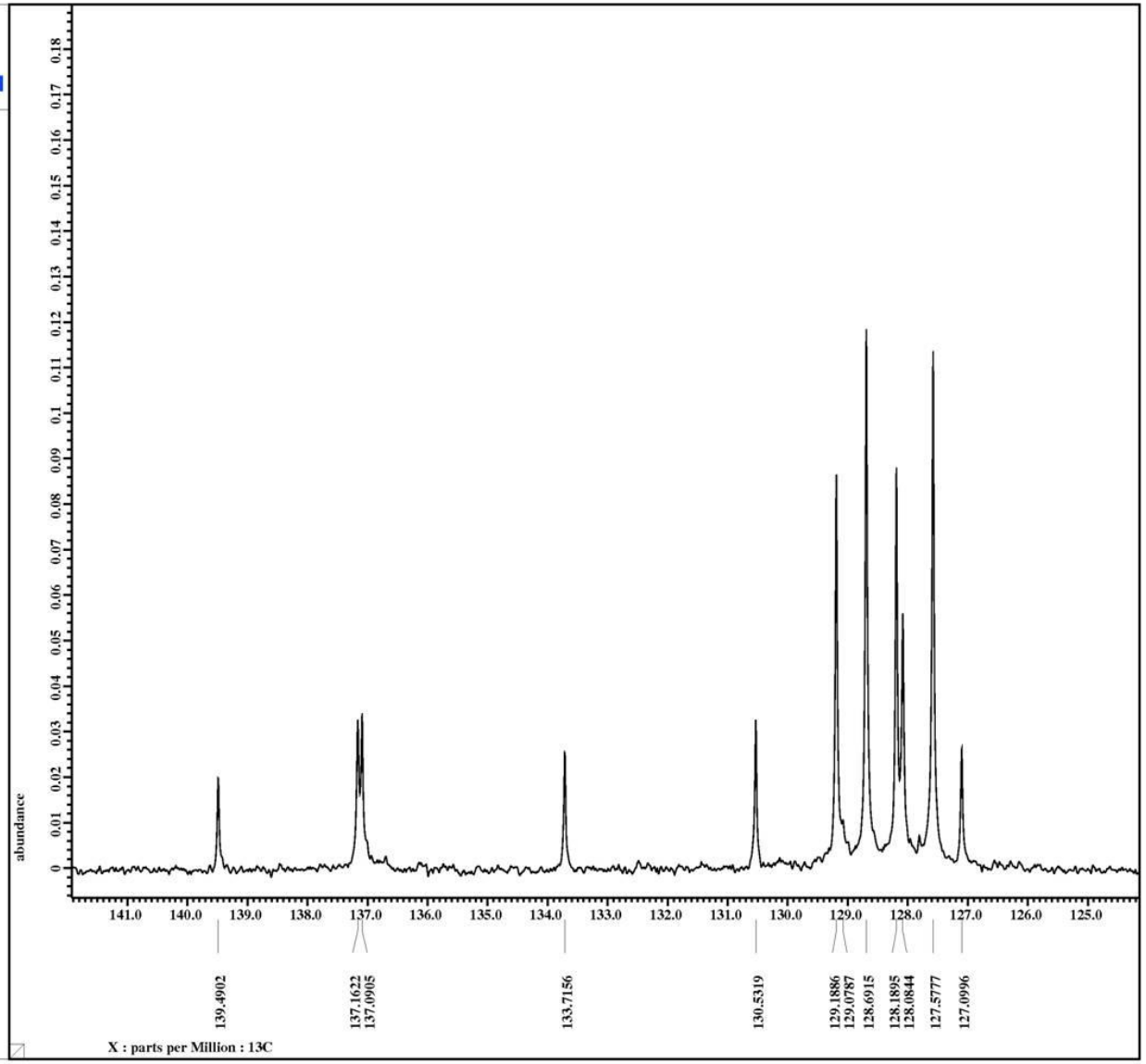
```
Filename          = III_p_228_sm-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#797179
Solvent           = CHLOROFORM-D
Creation_time     = 15-MAR-2008 21:12:27
Revision_time     = 15-MAR-2008 23:05:46
Current_time      = 18-MAR-2010 17:40:13

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 680
Total_scans       = 680

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.7[dC]
```





```
Filename          = III_p_228_sm-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#797179
Solvent           = CHLOROFORM-D
Creation_time     = 15-MAR-2008 21:12:27
Revision_time     = 15-MAR-2008 23:05:46
Current_time      = 18-MAR-2010 17:40:54

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 680
Total_scans       = 680

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.7[dC]
```

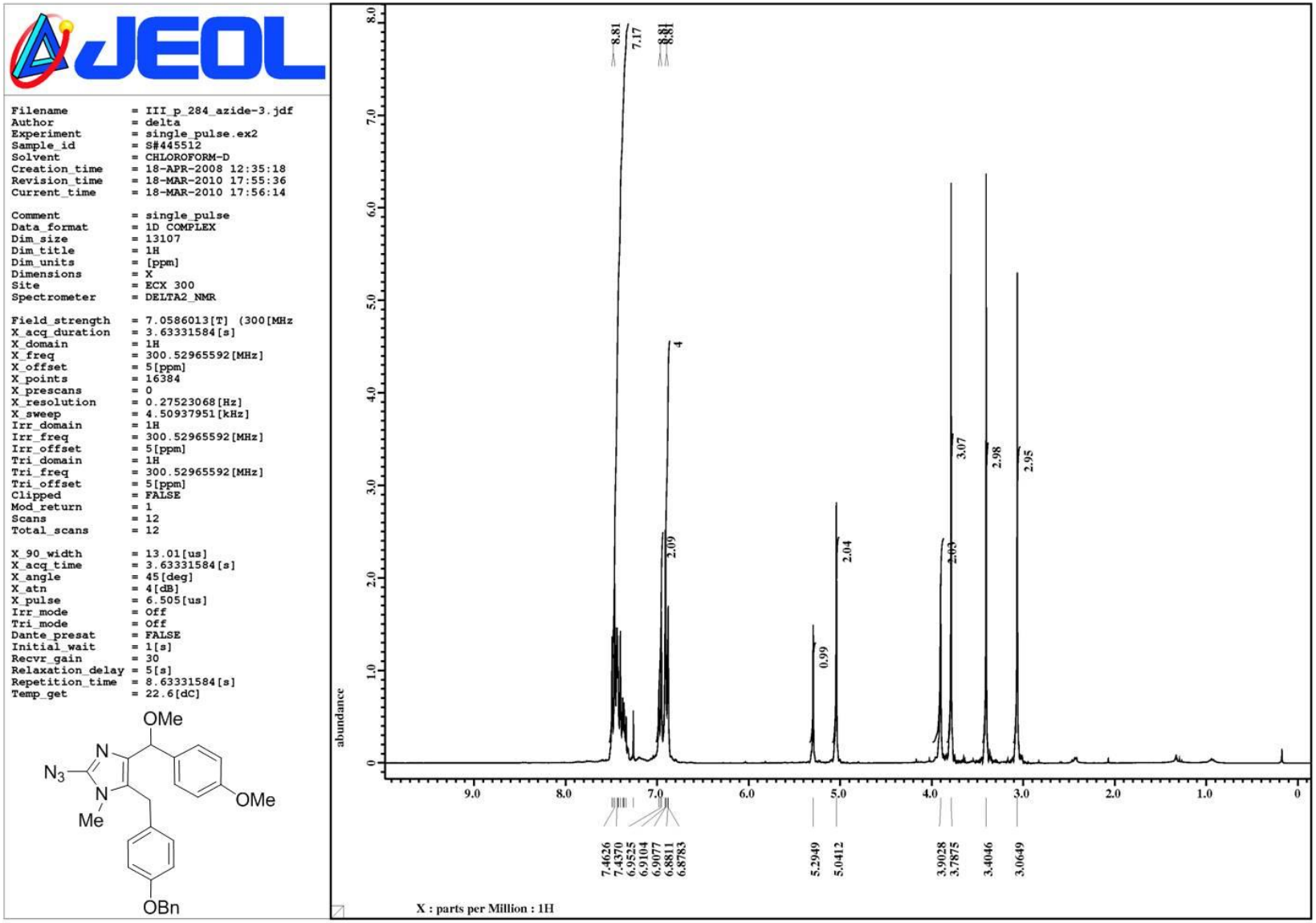




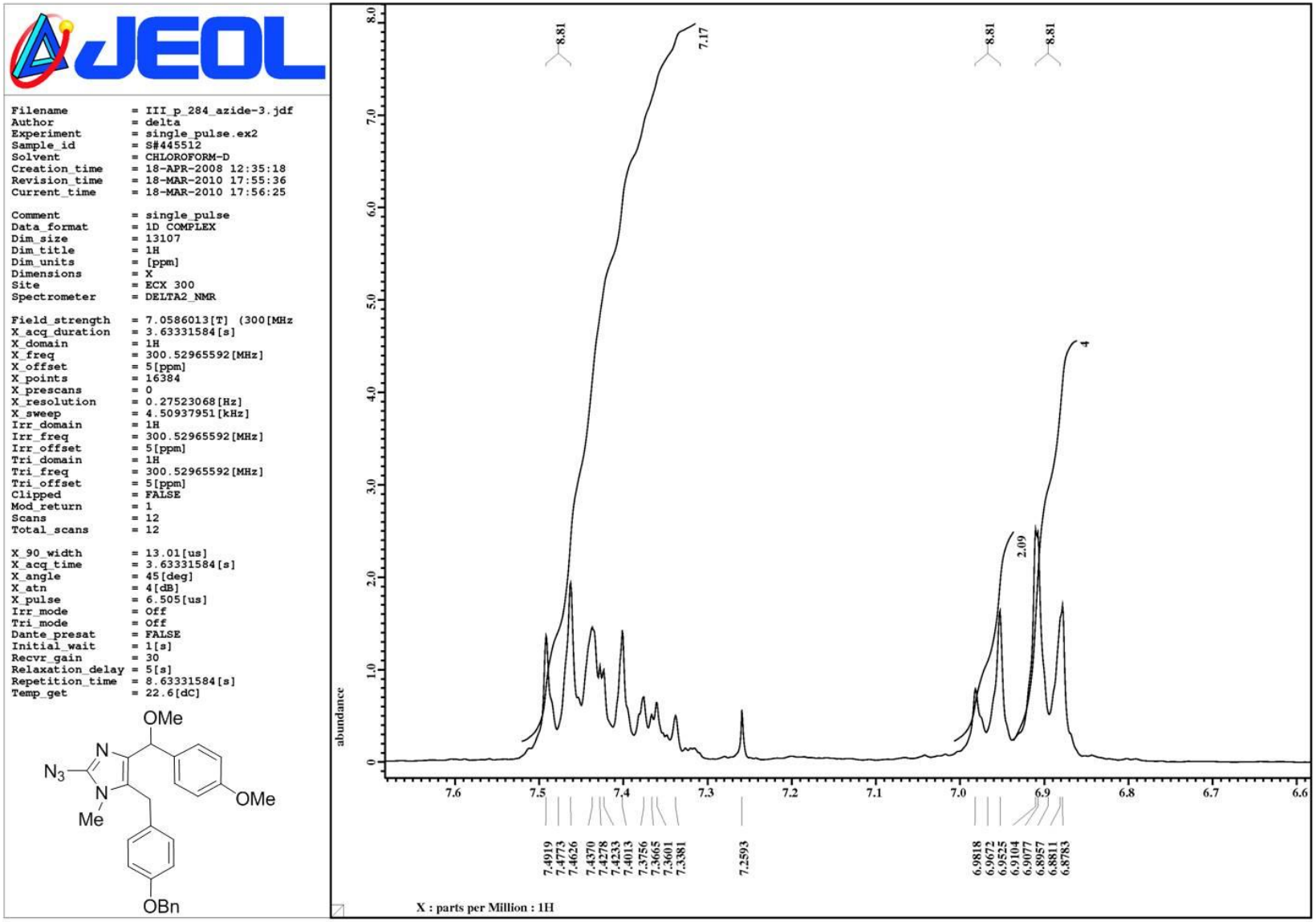
APPENDIX 48

^{1}H and ^{13}C NMR Spectra of

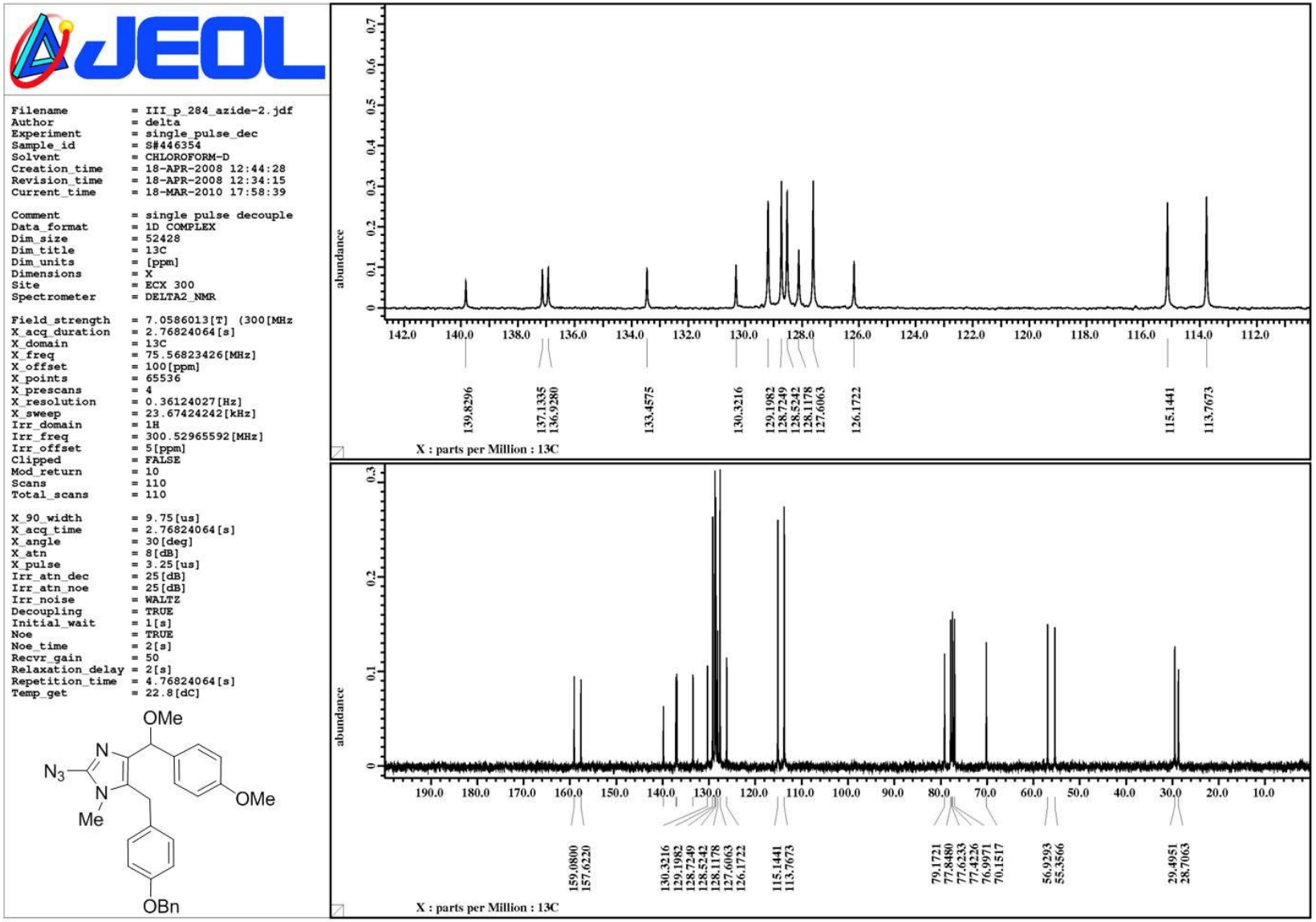
2-Azido-5-(4-benzoyloxybenzyl)-1-methyl-4-[methoxy-(4-methoxyphenyl)]methyl-1*H*-imidazole (169)



Filename	= III_p_284_azide-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#445512
Solvent	= CHLOROFORM-D
Creation_time	= 18-APR-2008 12:35:18
Revision_time	= 18-MAR-2010 17:55:36
Current_time	= 18-MAR-2010 17:56:14
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.6[dC]



```
Filename          = III_p_284_azide-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#445512
Solvent           = CHLOROFORM-D
Creation_time     = 18-APR-2008 12:35:18
Revision_time     = 18-MAR-2010 17:55:36
Current_time      = 18-MAR-2010 17:56:25

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 30
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.6[dC]
```

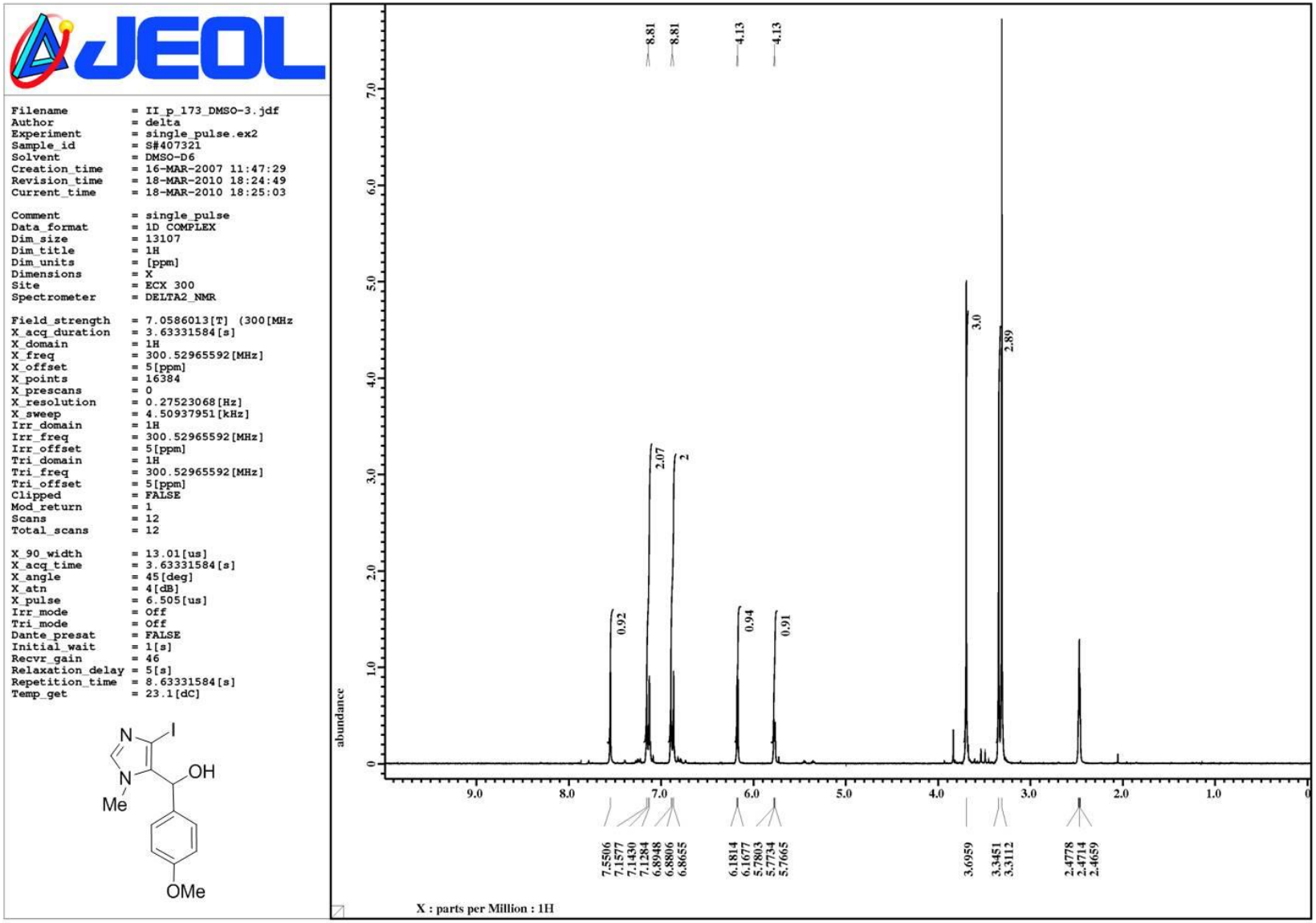


```
Filename          = III_p_284_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#446354
Solvent           = CHLOROFORM-D
Creation_time     = 18-APR-2008 12:44:28
Revision_time     = 18-APR-2008 12:34:15
Current_time      = 18-MAR-2010 17:58:39

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 110
Total_scans       = 110

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.8[dC]
```

APPENDIX 49

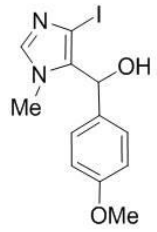
^{1}H and ^{13}C NMR Spectra of

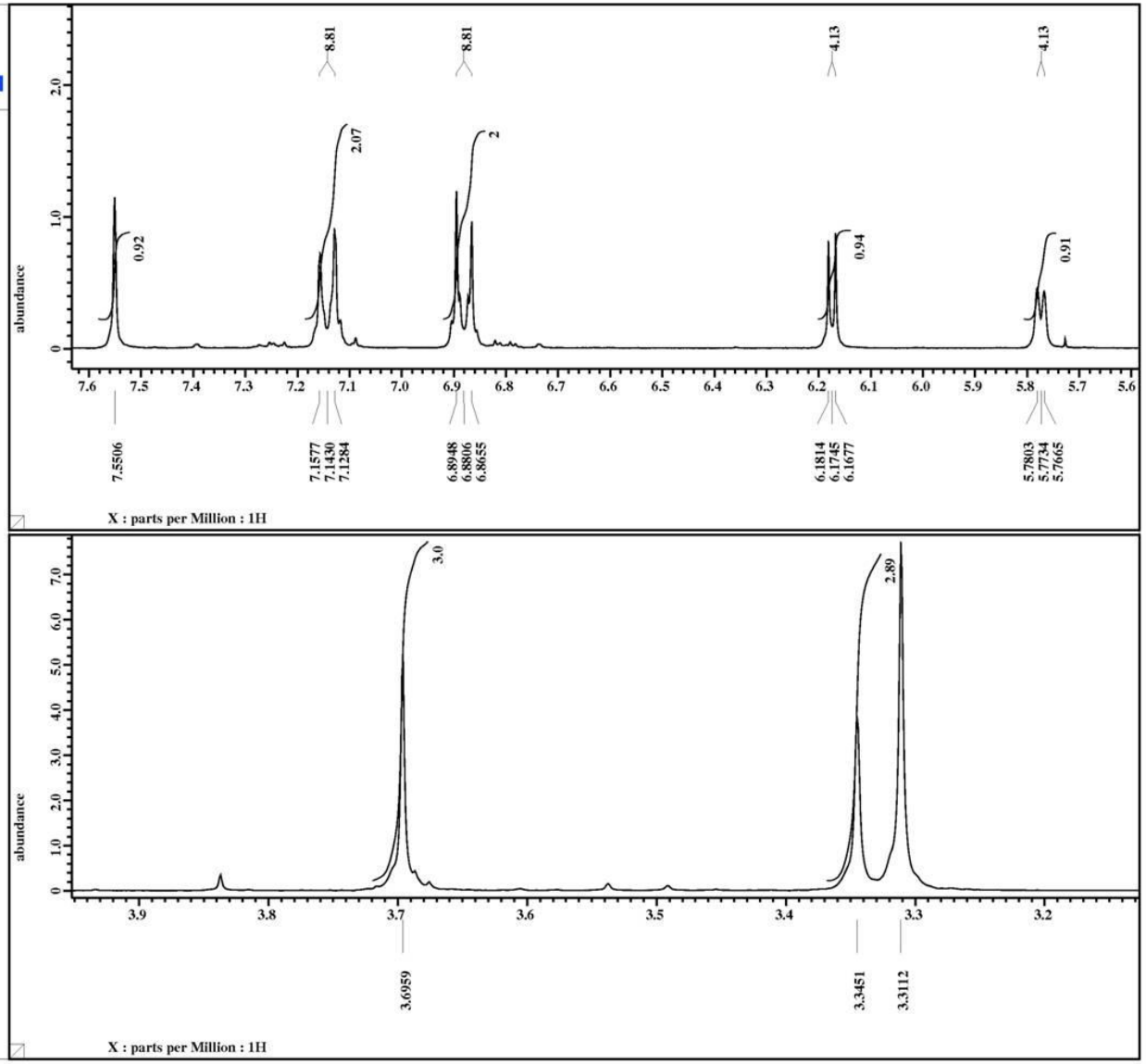
4-[Hydroxy-(4-methoxyphenyl)]methyl-4-iodo-1-methyl-1*H*-imidazole (170)



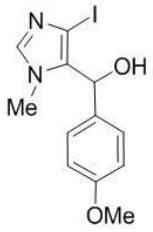
```
Filename          = II_p_173_DMSO-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#407321
Solvent           = DMSO-D6
Creation_time     = 16-MAR-2007 11:47:29
Revision_time     = 18-MAR-2010 18:24:49
Current_time      = 18-MAR-2010 18:25:03

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```

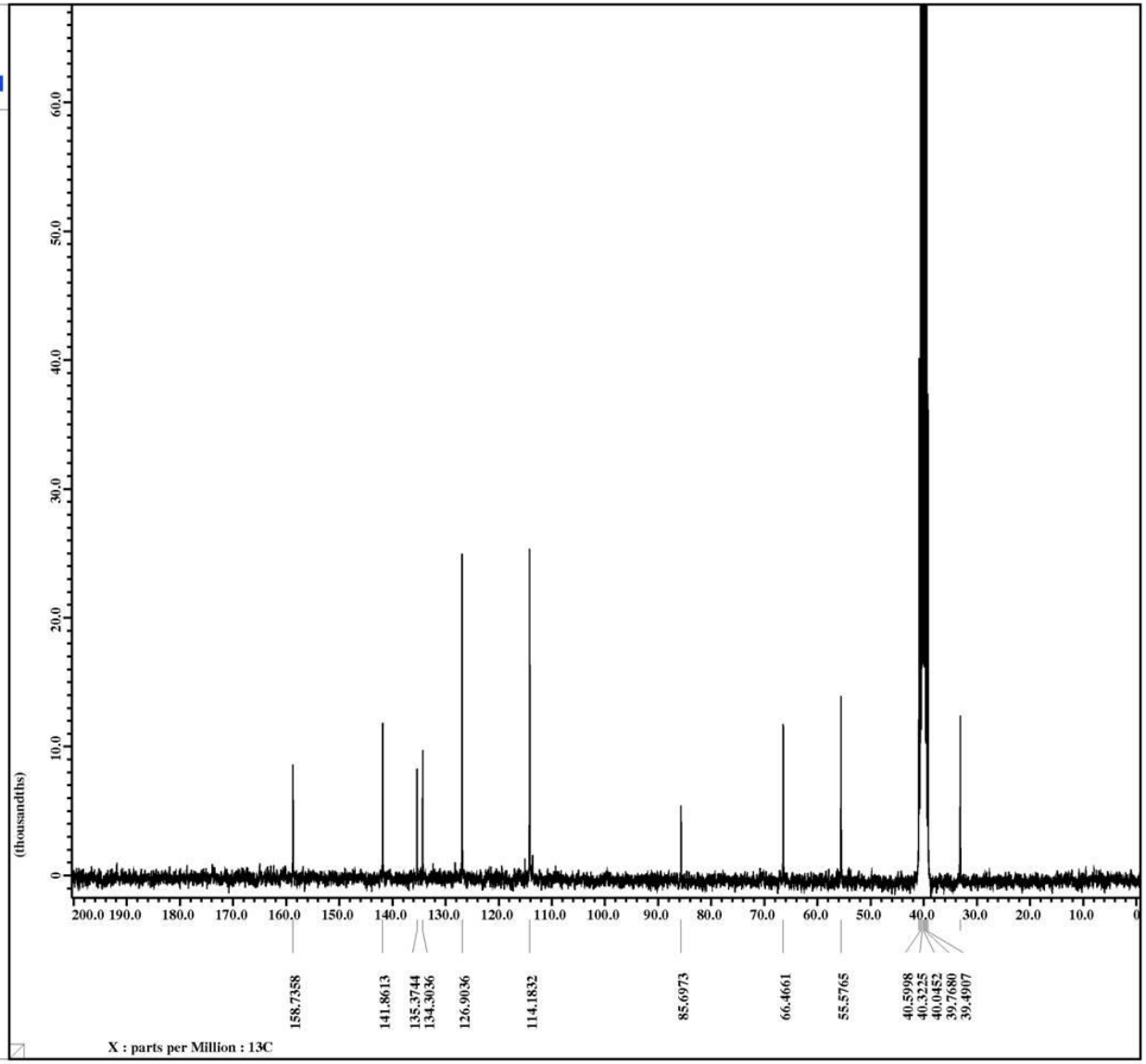
```
Filename          = II_p_173_DMSO-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#407321
Solvent           = DMSO-D6
Creation_time     = 16-MAR-2007 11:47:29
Revision_time     = 18-MAR-2010 18:24:49
Current_time      = 18-MAR-2010 18:25:24

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```





```
Filename          = II_p_173_DMSO-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#796498
Solvent           = DMSO-D6
Creation_time     = 21-MAR-2007 01:11:00
Revision_time     = 18-MAR-2010 18:26:23
Current_time      = 18-MAR-2010 18:26:31

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1970
Total_scans       = 1970

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.6[dC]
```



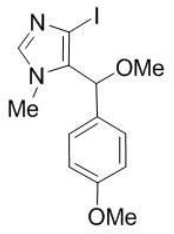


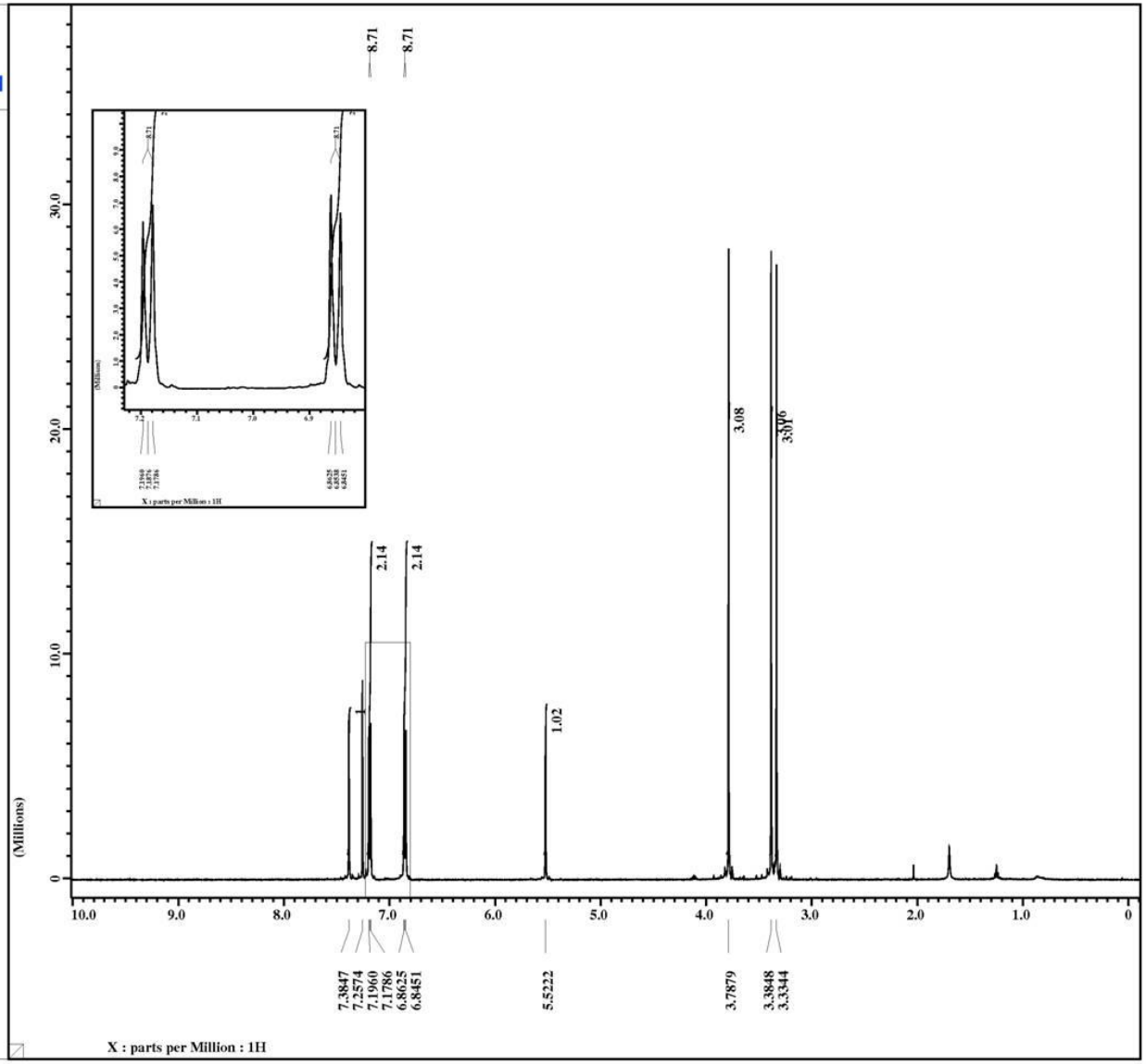
APPENDIX 50

^{1}H and ^{13}C NMR Spectra of

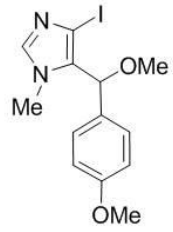
4-Iodo-1-methyl-5-[methoxy-(4-methoxyphenyl)]methyl-1*H*-imidazole (**171**)

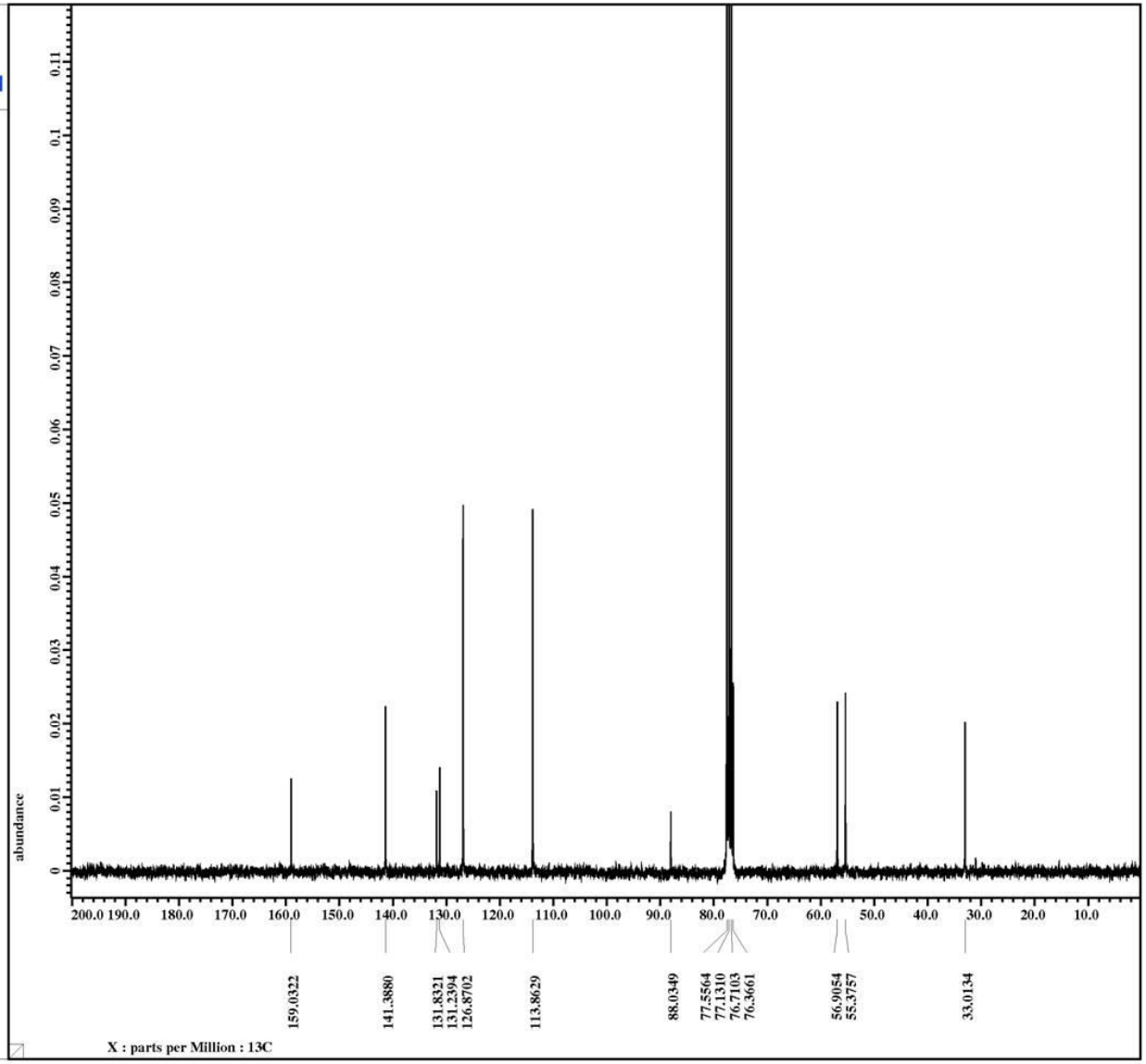
```
Filename          = II_p_268_Pure-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#652951
Solvent           = CHLOROFORM-D
Creation_time     =  8-SEP-2007 22:23:47
Revision_time     = 18-MAR-2010 18:33:04
Current_time      = 18-MAR-2010 18:34:08

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```





```
Filename          = II_p_164-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 11-MAR-2007 00:52:09
Revision_time     = 11-MAR-2007 11:33:25
Current_time      = 18-MAR-2010 18:35:21

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 1630
Total_scans       = 1630

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.1[dC]
```



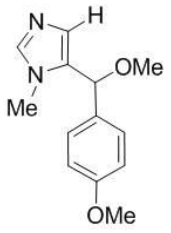


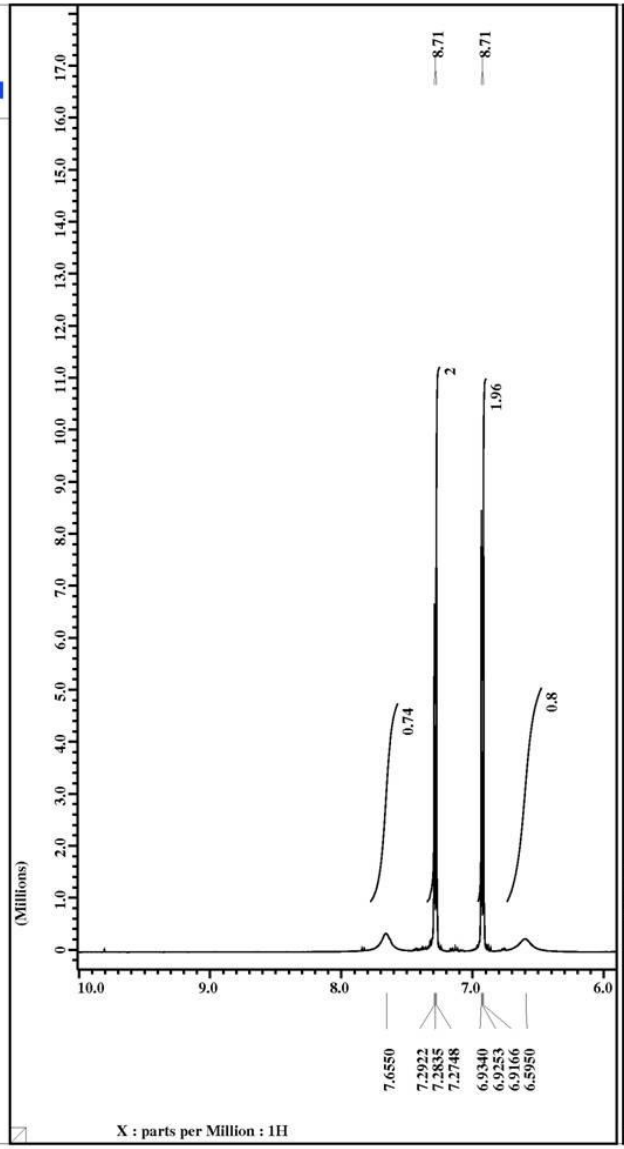
APPENDIX 51

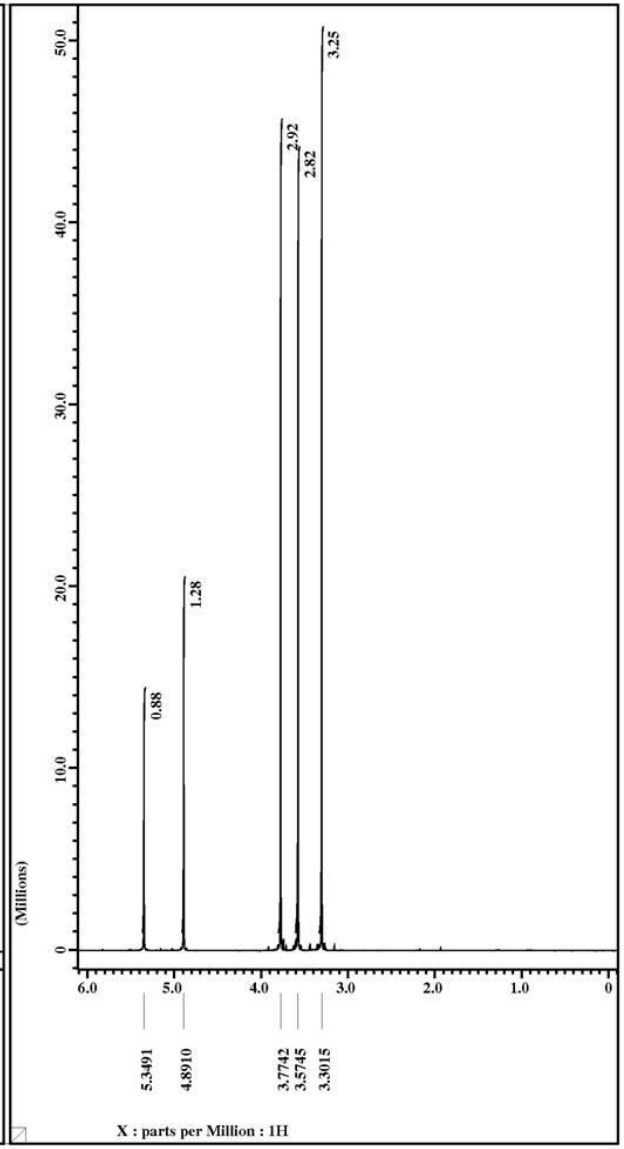
^{1}H and ^{13}C NMR Spectra of

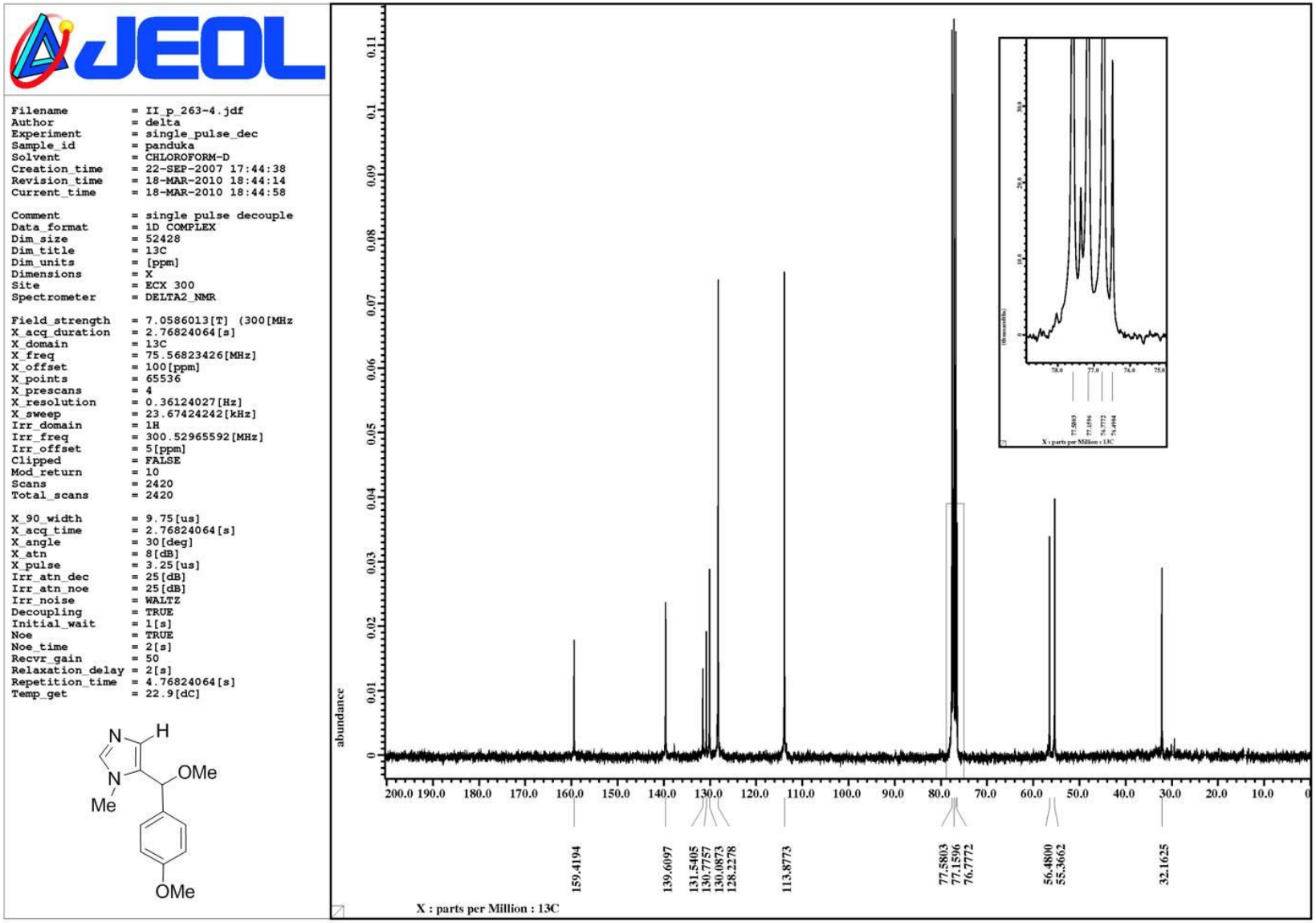
5-[Methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**172**)

```
Filename          = IV_p_283-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#407146
Solvent           = METHANOL-D3
Creation_time     = 21-OCT-2008 18:50:36
Revision_time     = 18-MAR-2010 18:42:16
Current_time      = 18-MAR-2010 18:43:16

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 12
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```







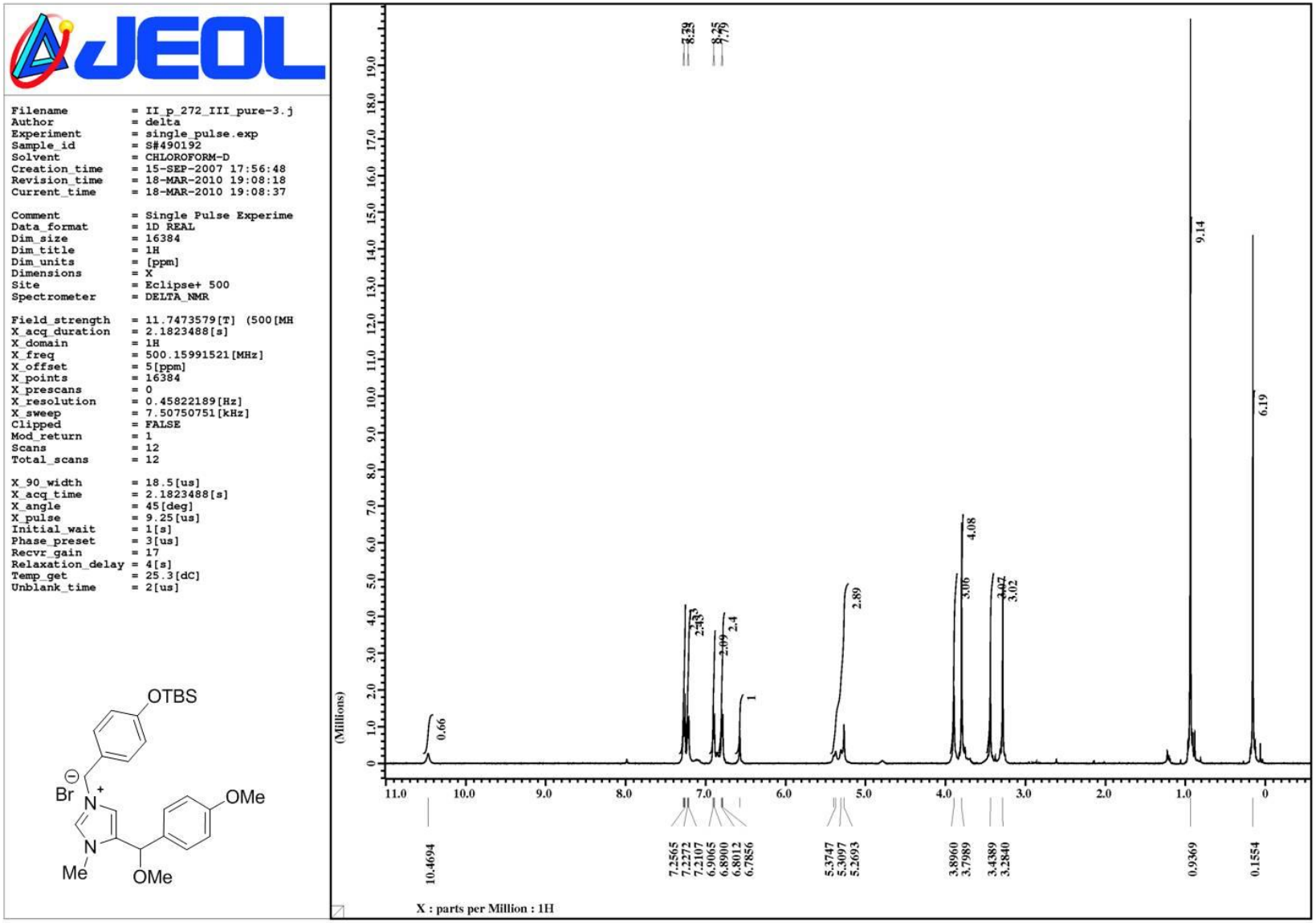


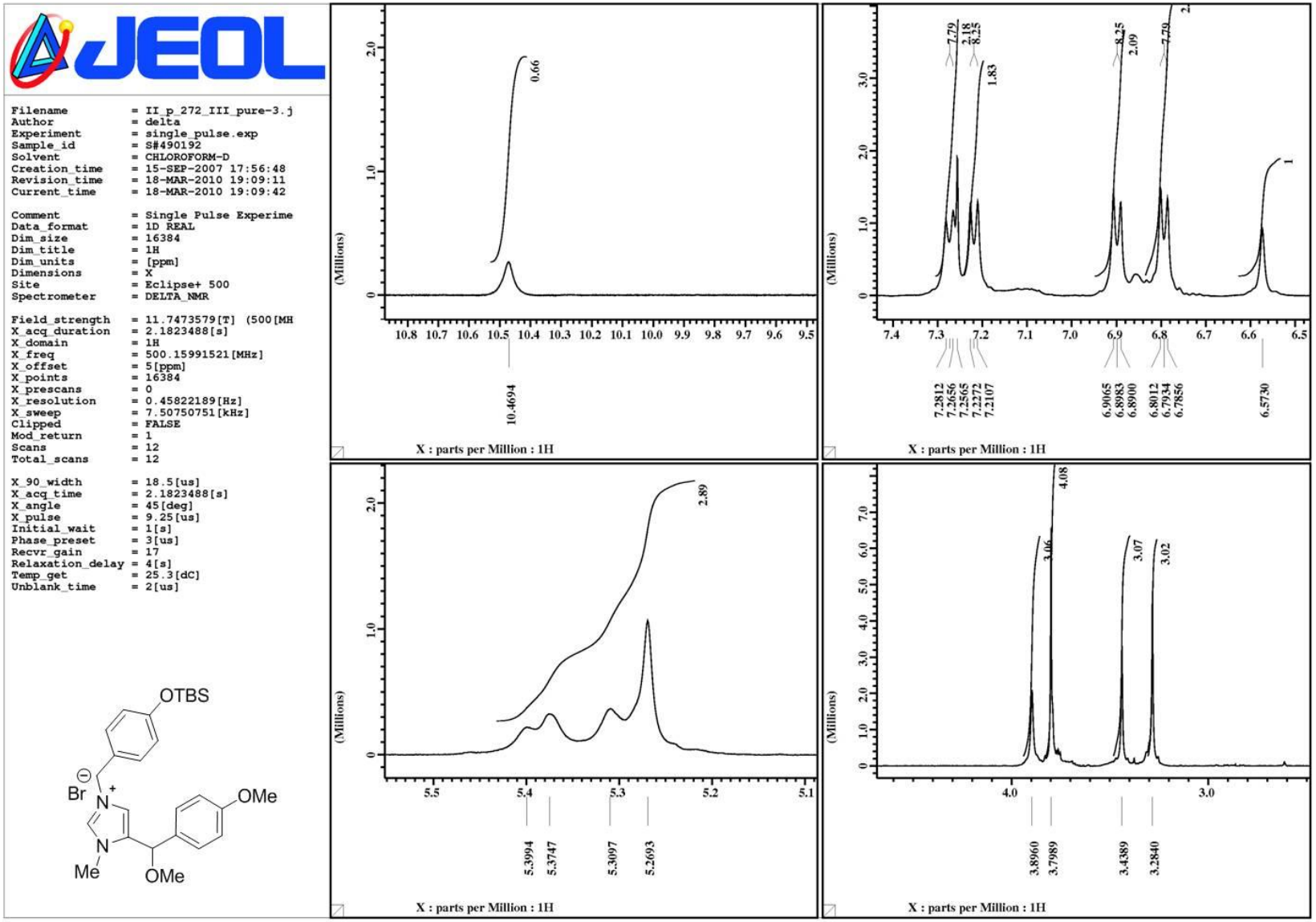
```
Filename          = II_p_263-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 22-SEP-2007 17:44:38
Revision_time     = 18-MAR-2010 18:44:14
Current_time      = 18-MAR-2010 18:44:58

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 2420
Total_scans       = 2420

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.9[dC]
```

APPENDIX 52

^{1}H and ^{13}C NMR Spectra of

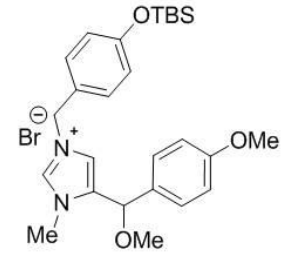
1-(4-*t*-Butyldimethylsilyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-3-methyl-3*H*-imidazol-1-ium bromide (**173**)

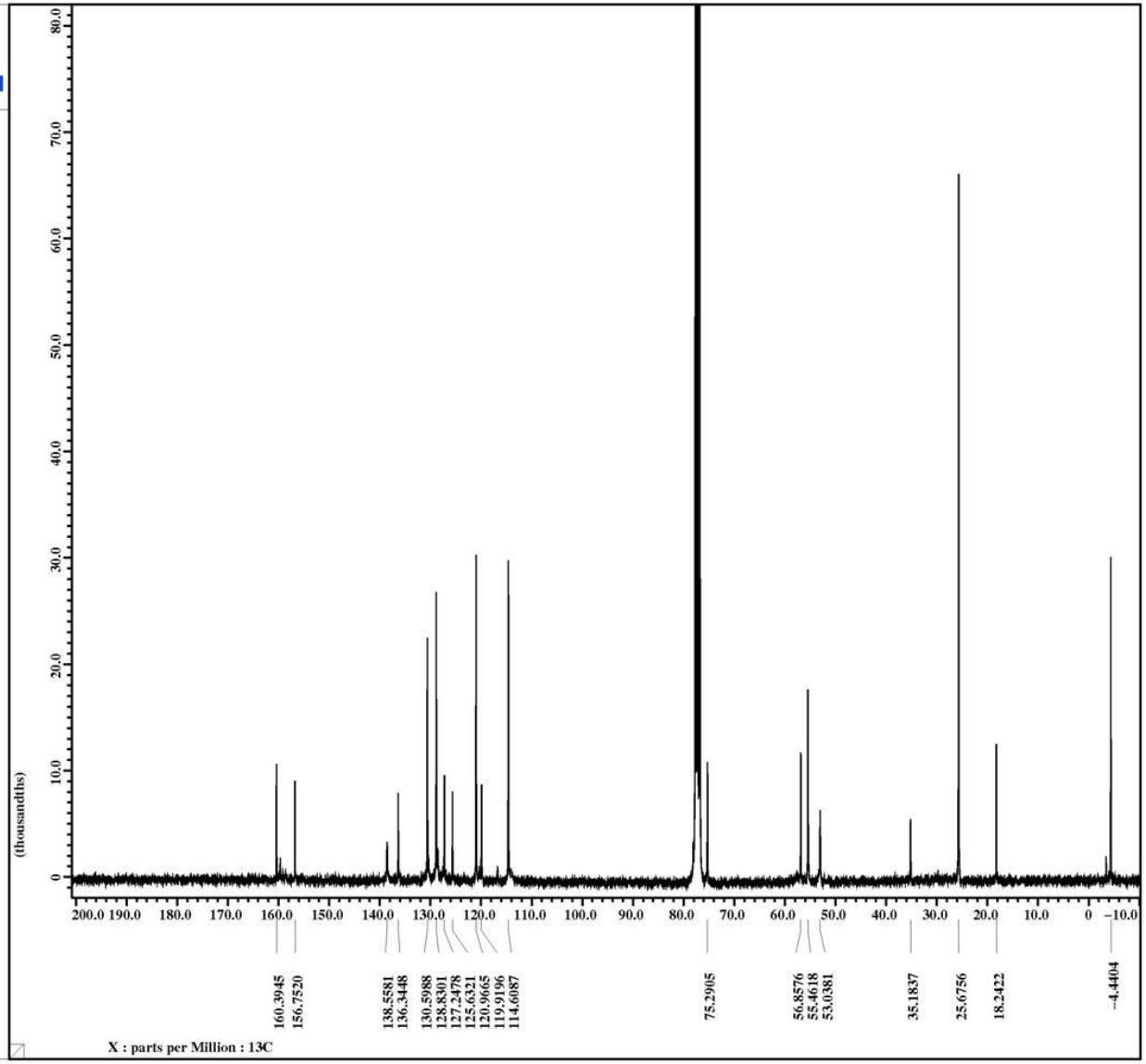

JEOL
Filename = II_p_272_III_pure-3.j
Author = delta
Experiment = single_pulse.exp
Sample_id = S#490192
Solvent = CHLOROFORM-D
Creation_time = 15-SEP-2007 17:56:48
Revision_time = 18-MAR-2010 19:08:18
Current_time = 18-MAR-2010 19:08:37
Comment = Single Pulse Experime
Data_format = 1D REAL
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 17
Relaxation_delay = 4[s]
Temp_get = 25.3[dC]
Unblank_time = 2[us]
OTBS
Br
N
N
Me
OMe
OMe
(Millions)
9.14
6.19
4.08
3.06
3.02
2.89
2.4
1
0.66
10.4694
7.2565
7.2272
7.2107
6.9065
6.8900
6.8012
6.7856
5.3747
5.3097
5.2693
3.8960
3.7989
3.4389
3.2840
0.9369
0.1554
X : parts per Million : 1H



```
Filename          = II_p_272_III_pure-3.j
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#490192
Solvent           = CHLOROFORM-D
Creation_time     = 15-SEP-2007 17:56:48
Revision_time     = 18-MAR-2010 19:09:11
Current_time      = 18-MAR-2010 19:09:42

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 17
Relaxation_delay  = 4[s]
Temp_get          = 25.3[dC]
Unblank_time      = 2[us]
```

```
Filename          = II_p_272-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#743291
Solvent           = CHLOROFORM-D
Creation_time     = 15-DEC-2007 09:47:11
Revision_time     = 18-MAR-2010 19:13:01
Current_time      = 18-MAR-2010 19:13:16

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 9600
Total_scans       = 9600

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.7[dC]
```



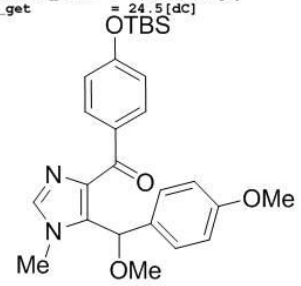


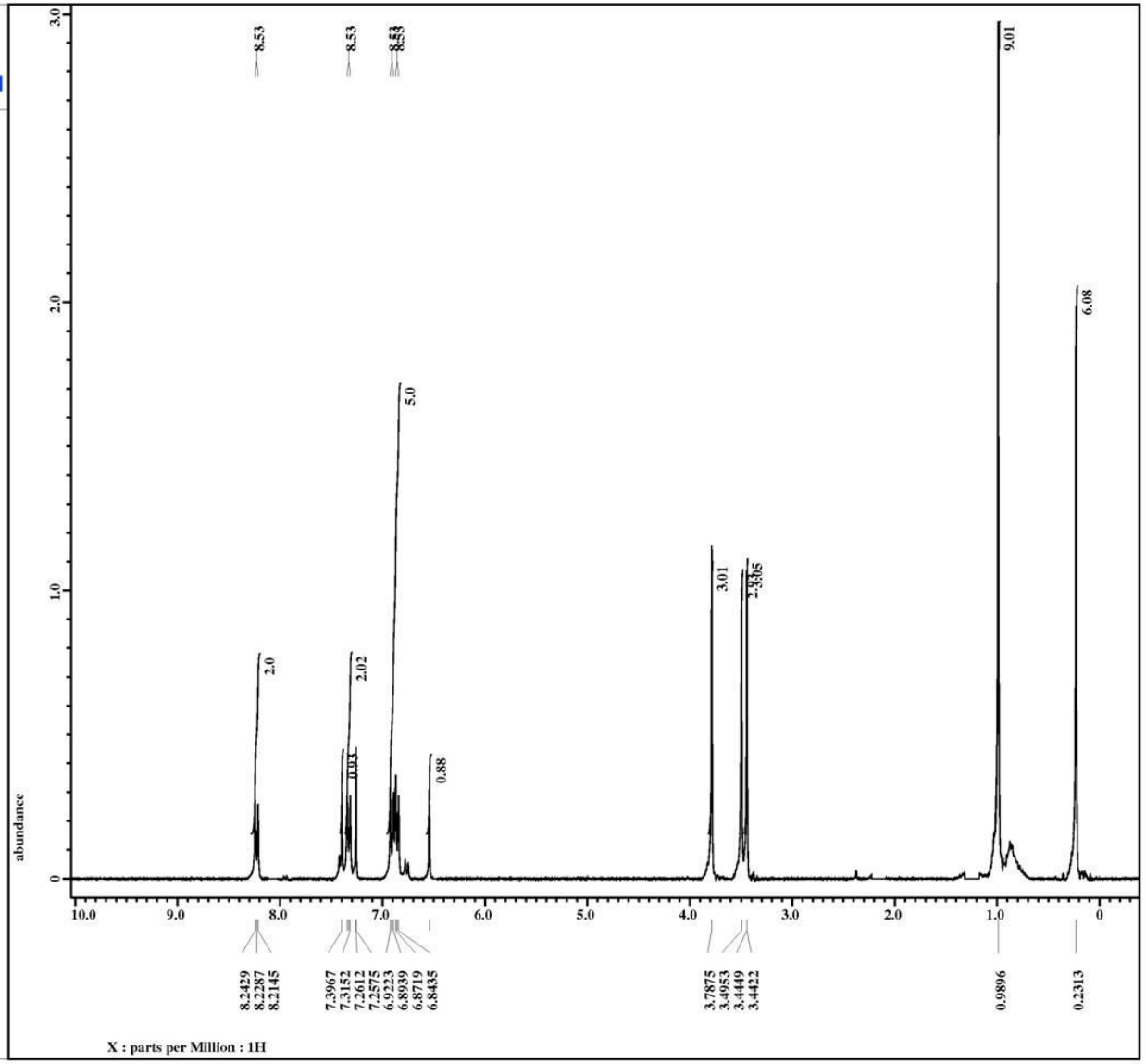
APPENDIX 53

^{1}H and ^{13}C NMR Spectra of

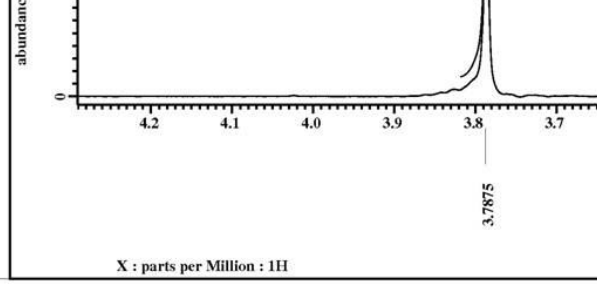
4-(*t*-Butyldimethylsilyloxyphenyl)-{5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazol-4-yl}methanone (**174**)

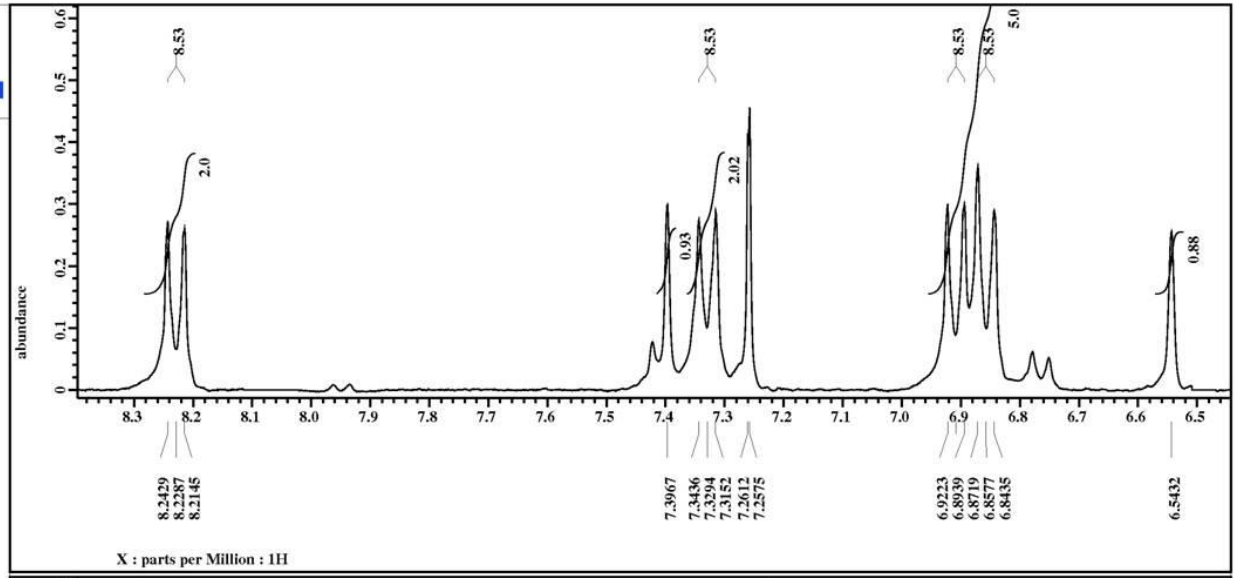
```
Filename          = II_p_257_II-5.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#30292
Solvent           = CHLOROFORM-D
Creation_time     =  2-SEP-2007 01:17:18
Revision_time     = 15-MAR-2010 18:56:31
Current_time      = 15-MAR-2010 18:57:10

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 44
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 24.5[dC]
```



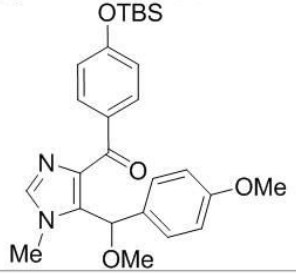


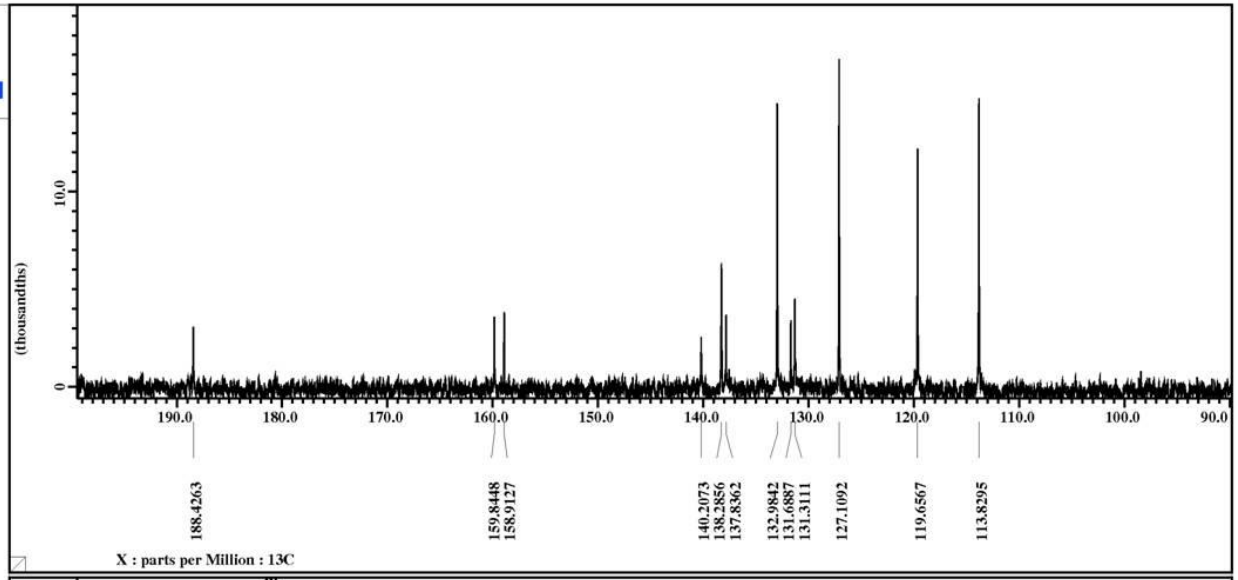
```
Filename          = II_p_257_II-5.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#30292
Solvent           = CHLOROFORM-D
Creation_time     =  2-SEP-2007 01:17:18
Revision_time     = 15-MAR-2010 18:56:31
Current_time      = 15-MAR-2010 18:58:27

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 44
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 24.5[dC]
```

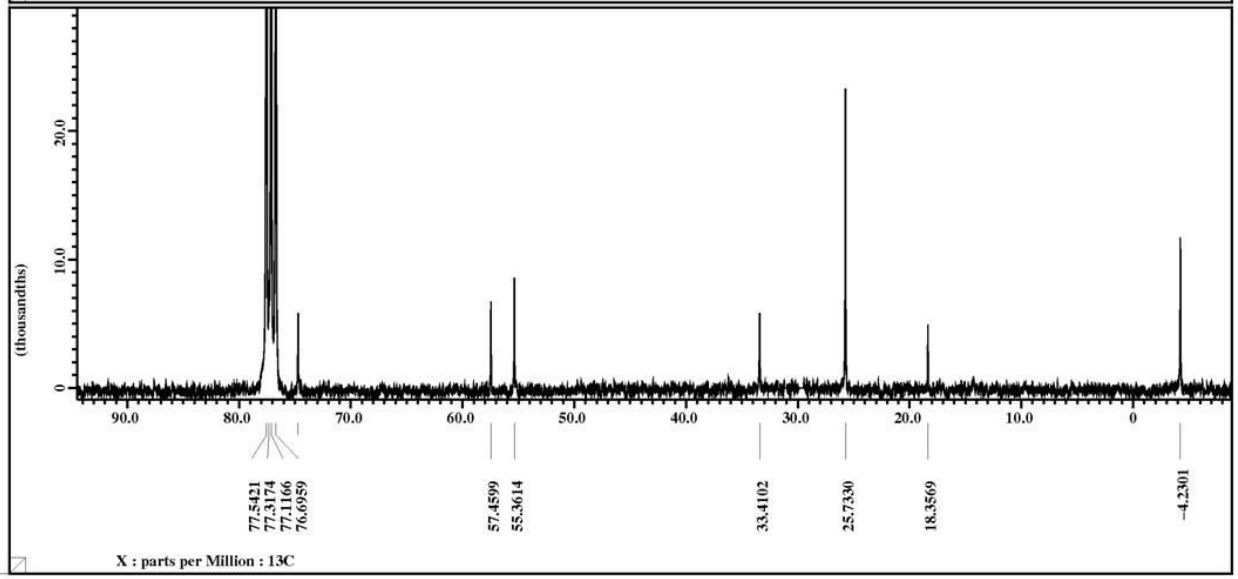




```
Filename          = II_p_257_II-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     =  2-SEP-2007 05:17:48
Revision_time     = 18-MAR-2010 19:24:09
Current_time      = 18-MAR-2010 19:25:51

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 3000
Total_scans       = 3000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```



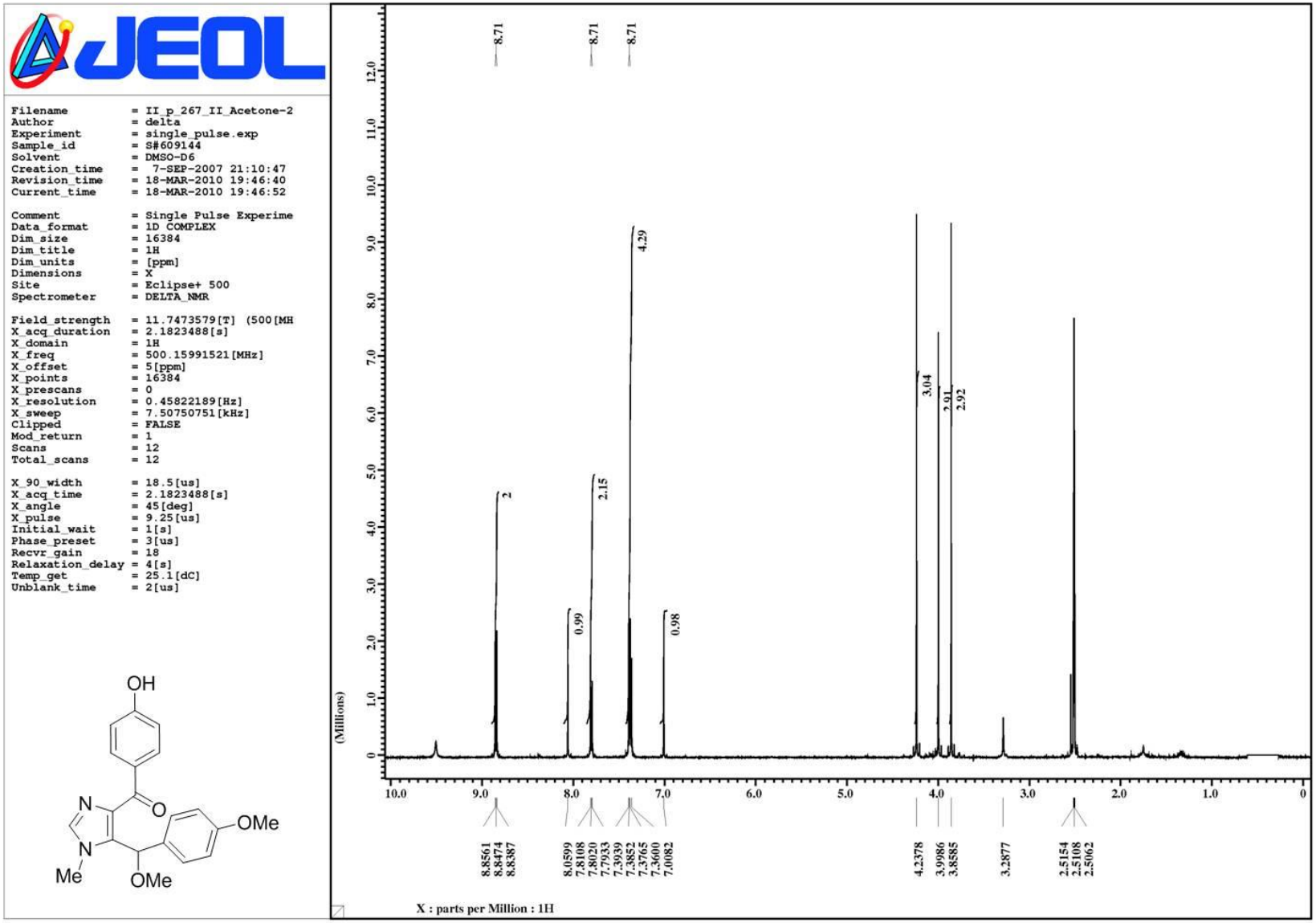




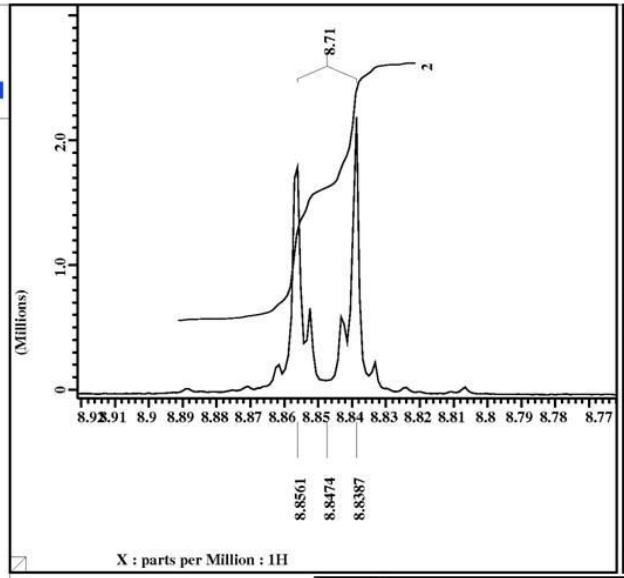
APPENDIX 54

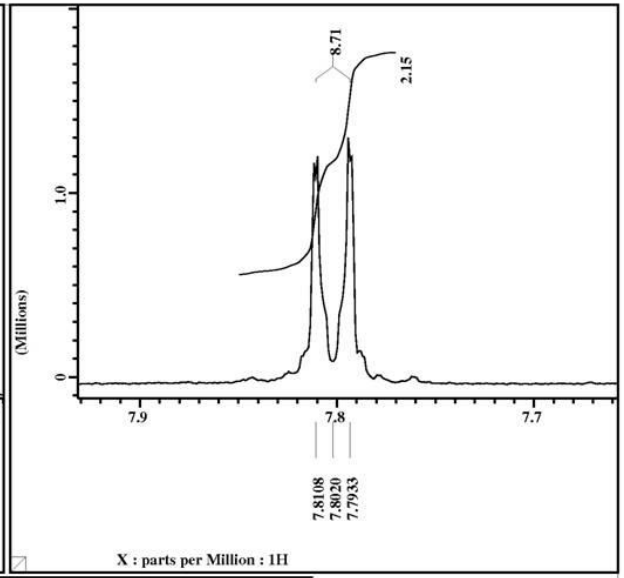
^{1}H and ^{13}C NMR Spectra of

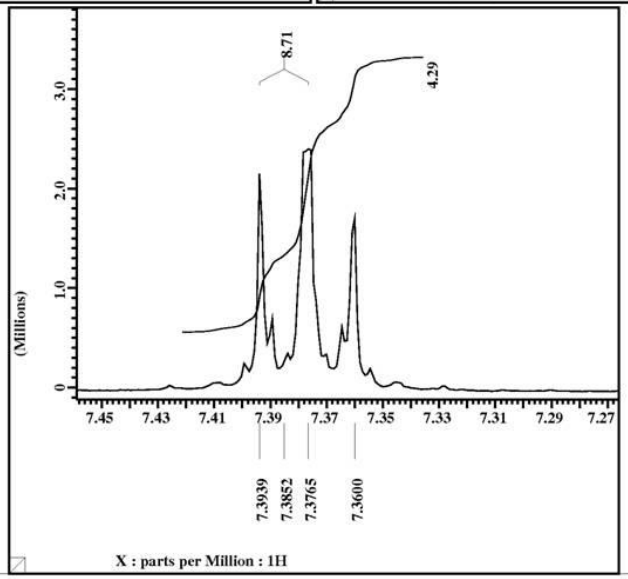
4-Hydroxyphenyl-{5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazol-4-yl}methanone (**175**)

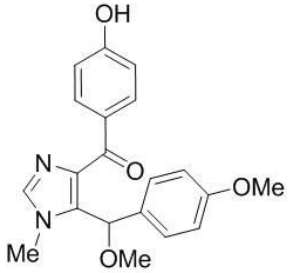

Filename = II_p_267_II_Acetone-2
Author = delta
Experiment = single_pulse.exp
Sample_id = S#609144
Solvent = DMSO-D6
Creation_time = 7-SEP-2007 21:10:47
Revision_time = 18-MAR-2010 19:46:40
Current_time = 18-MAR-2010 19:46:52
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 18
Relaxation_delay = 4[s]
Temp_get = 25.1[dC]
Unblank_time = 2[us]
OH
N
O
OMe
N
Me
OMe
8.71
8.71
8.71
4.29
3.04
2.91
2.92
2
2.15
0.99
0.98
(Millions)
8.8561
8.8474
8.8387
8.0599
7.8108
7.8020
7.7933
7.3939
7.3852
7.3765
7.3600
7.0082
4.2378
3.9986
3.8585
3.2877
2.5154
2.5108
2.5062
X : parts per Million : 1H

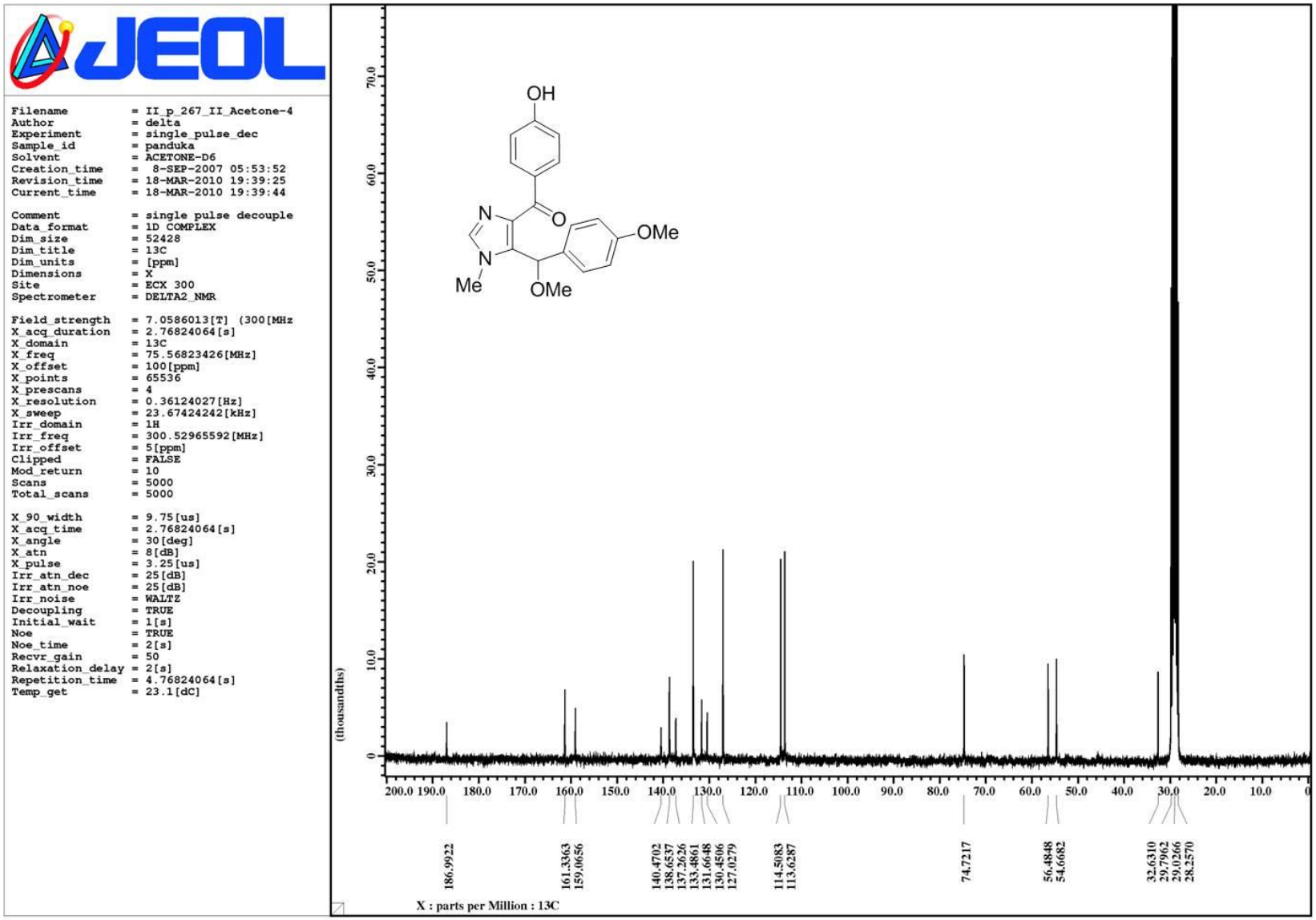
```
Filename          = II_p_267_II_Acetone-2
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#609144
Solvent           = DMSO-D6
Creation_time     =  7-SEP-2007 21:10:47
Revision_time     = 18-MAR-2010 19:46:40
Current_time      = 18-MAR-2010 19:47:53

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 4[s]
Temp_get          = 25.1[dC]
Unblank_time      = 2[us]
```









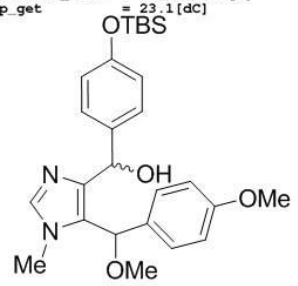

Filename = II_p_267_II_Acetone-4
Author = delta
Experiment = single_pulse_dec
Sample_id = panduka
Solvent = ACETONE-D6
Creation_time = 8-SEP-2007 05:53:52
Revision_time = 18-MAR-2010 19:39:25
Current_time = 18-MAR-2010 19:39:44
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 5000
Total_scans = 5000
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.1[dC]
OH
N
O
OMe
N
Me
OMe
(thousandths)
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0
200.0 190.0 180.0 170.0 160.0 150.0 140.0 130.0 120.0 110.0 100.0 90.0 80.0 70.0 60.0 50.0 40.0 30.0 20.0 10.0
186.9922
161.3363
159.0656
140.4702
138.6537
137.2626
133.4861
131.6648
130.4506
127.0279
114.5083
113.6287
74.7217
56.4848
54.6682
32.6310
29.7962
29.0266
28.2570
X : parts per Million : 13C

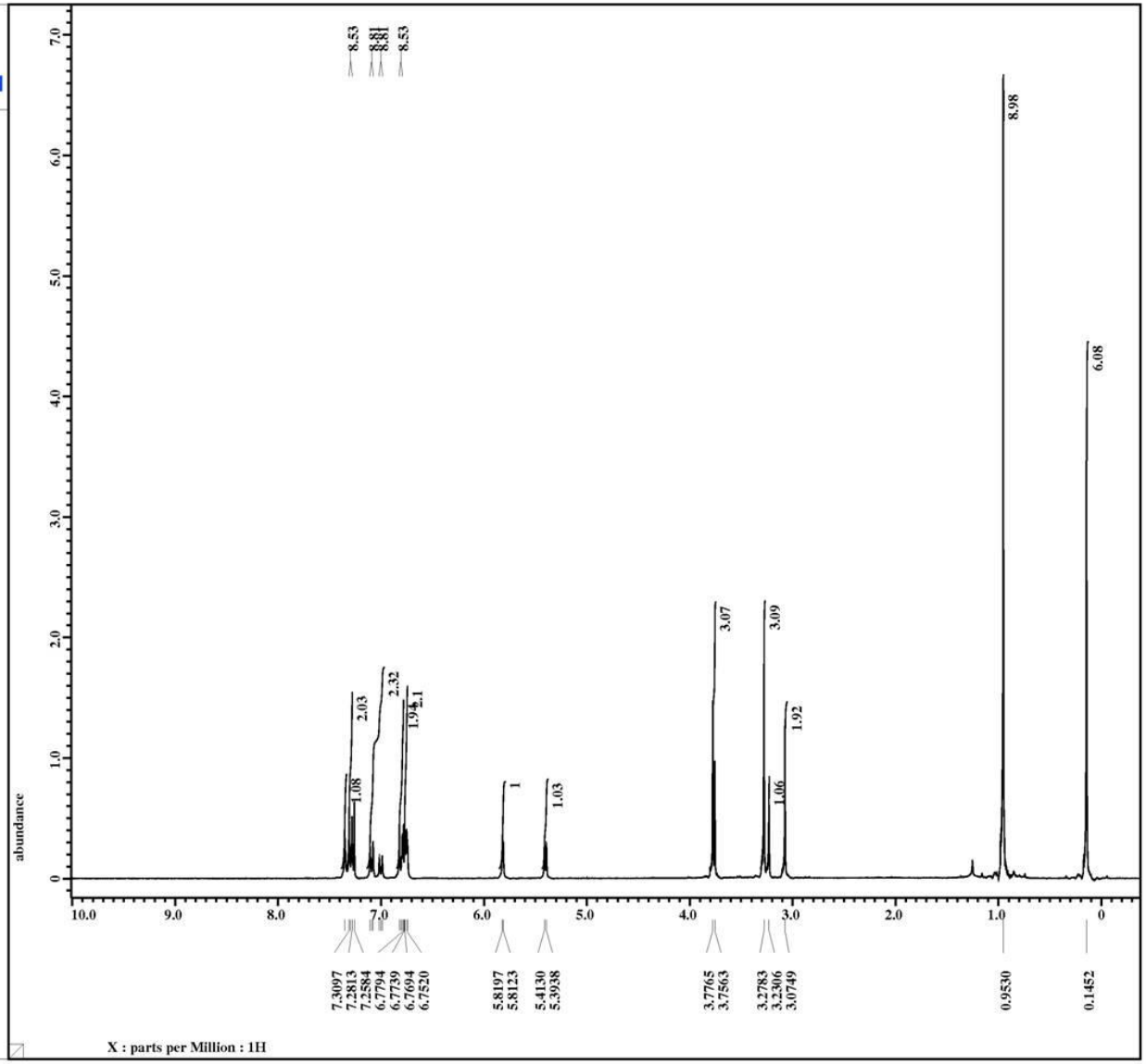
APPENDIX 55

^{1}H and ^{13}C NMR Spectra of

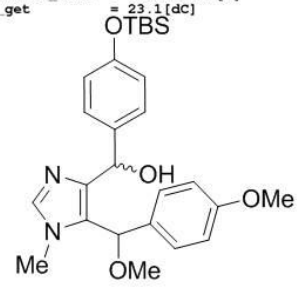
4-(4-*t*-Butyldimethylsilyloxyphenyl)hydroxymethyl-5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**176**)

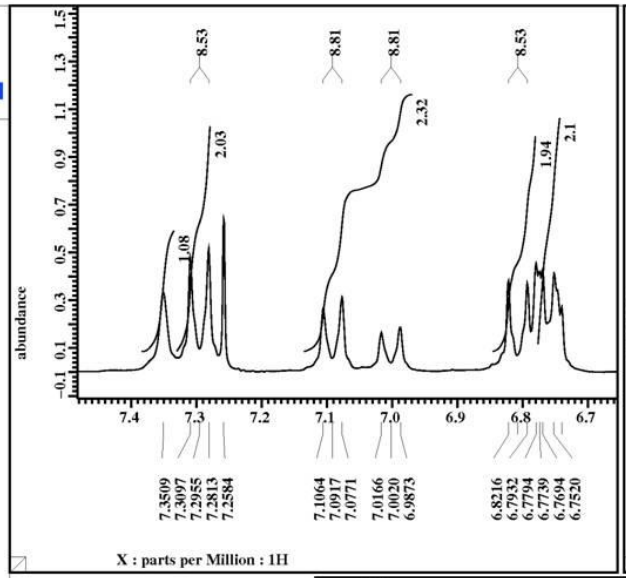
```
Filename          = II_p_282_I-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#644869
Solvent           = CHLOROFORM-D
Creation_time     = 22-SEP-2007 18:14:31
Revision_time     = 18-MAR-2010 20:05:36
Current_time      = 18-MAR-2010 20:07:00

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 40
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```

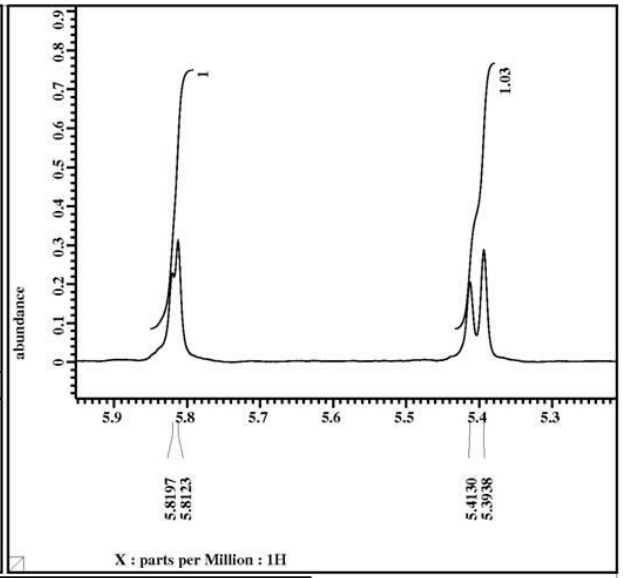


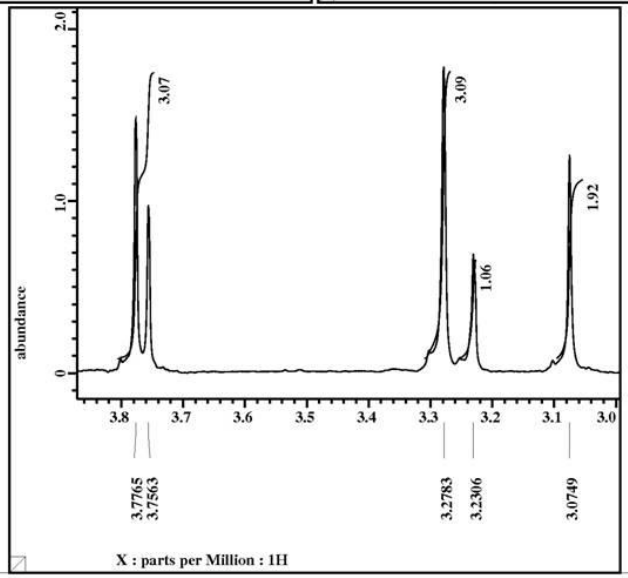


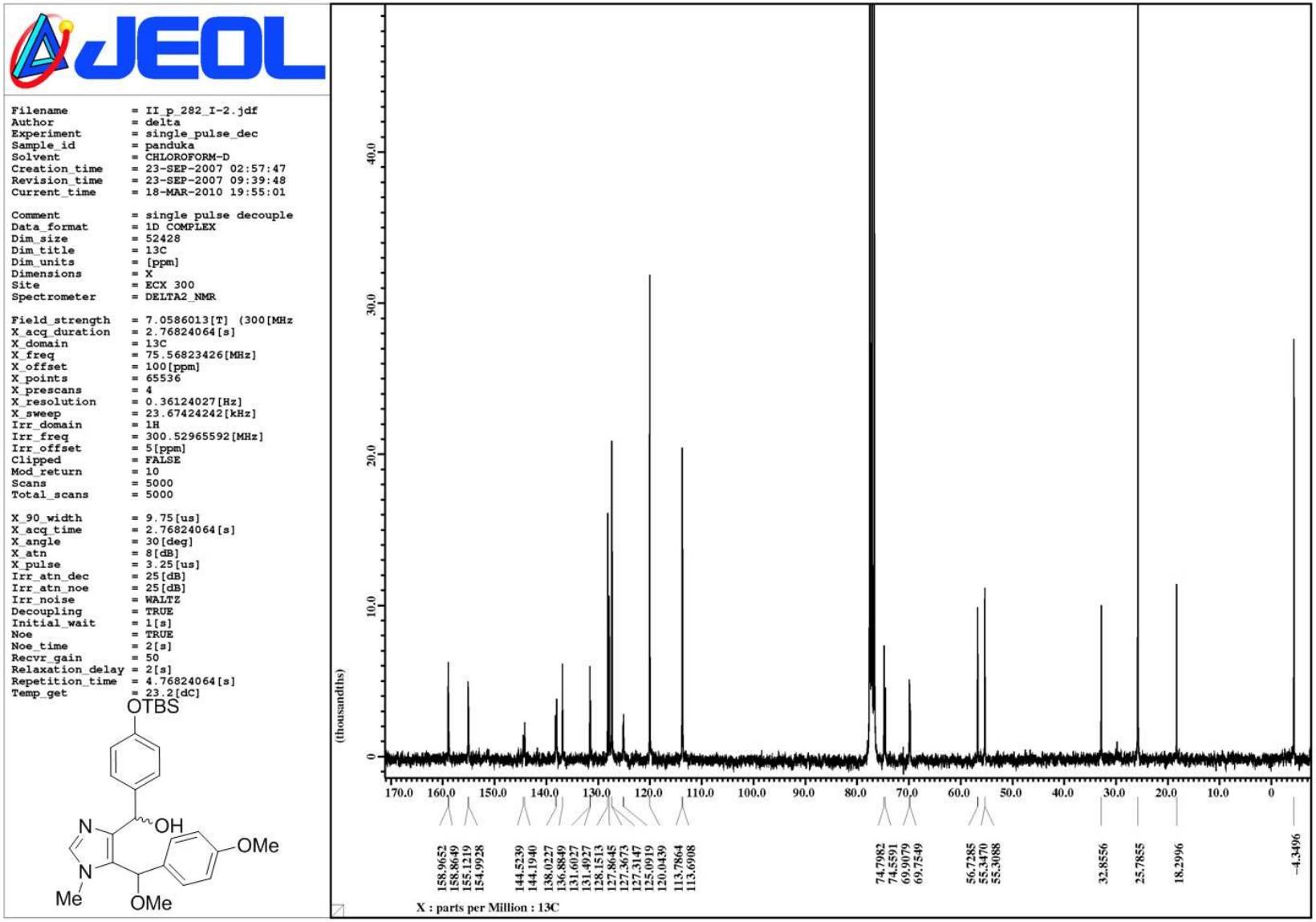
```
Filename          = II_p_282_I-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#644869
Solvent           = CHLOROFORM-D
Creation_time     = 22-SEP-2007 18:14:31
Revision_time     = 18-MAR-2010 20:05:36
Current_time      = 18-MAR-2010 20:07:33

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 40
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```



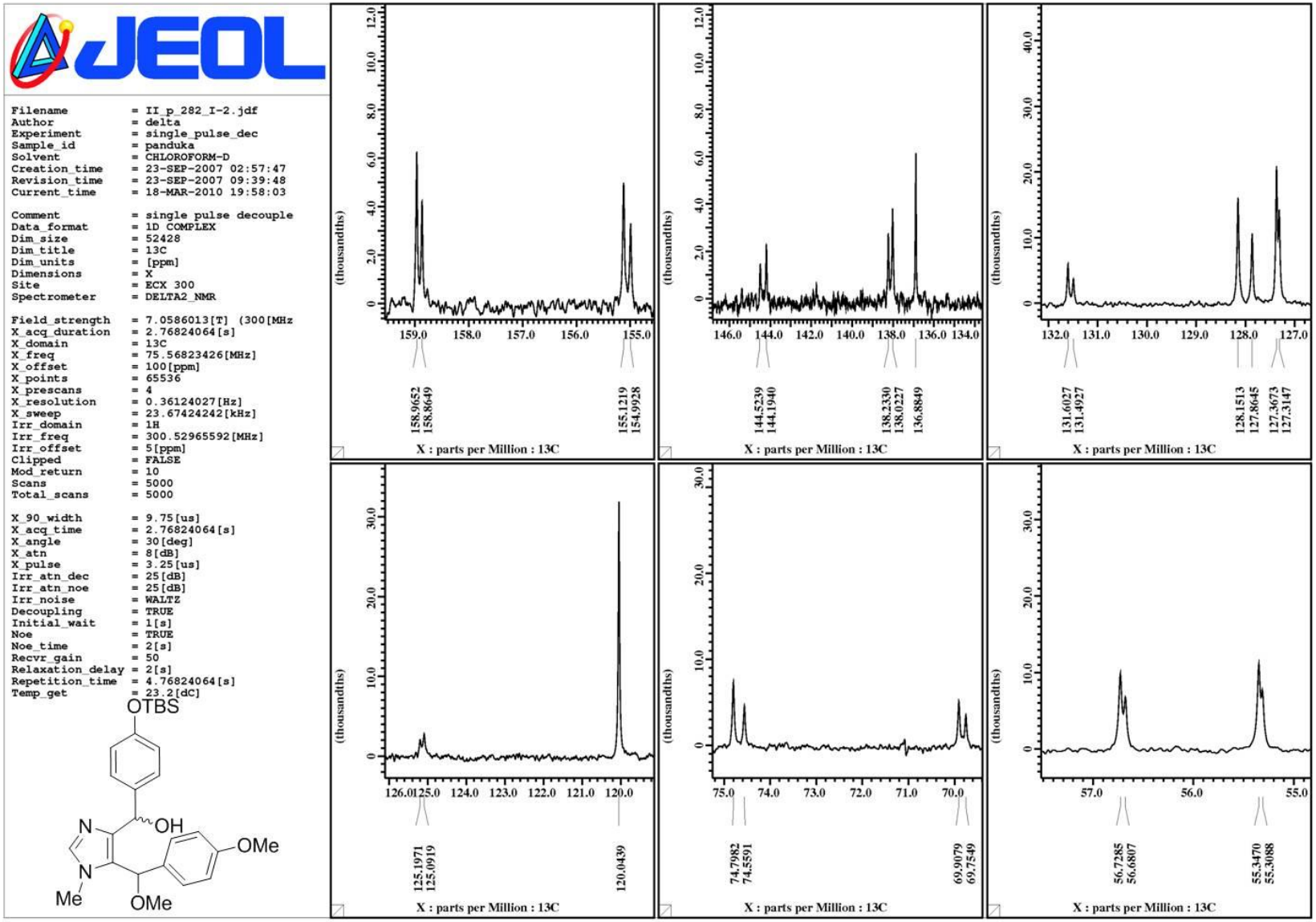








```
Filename          = II_p_282_I-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 23-SEP-2007 02:57:47
Revision_time     = 23-SEP-2007 09:39:48
Current_time      = 18-MAR-2010 19:55:01

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 5000
Total_scans       = 5000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.2[dC]
```



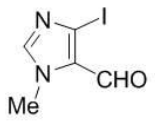
```
Filename          = II_p_282_I-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 23-SEP-2007 02:57:47
Revision_time     = 23-SEP-2007 09:39:48
Current_time      = 18-MAR-2010 19:58:03

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 5000
Total_scans       = 5000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.2[dC]
```

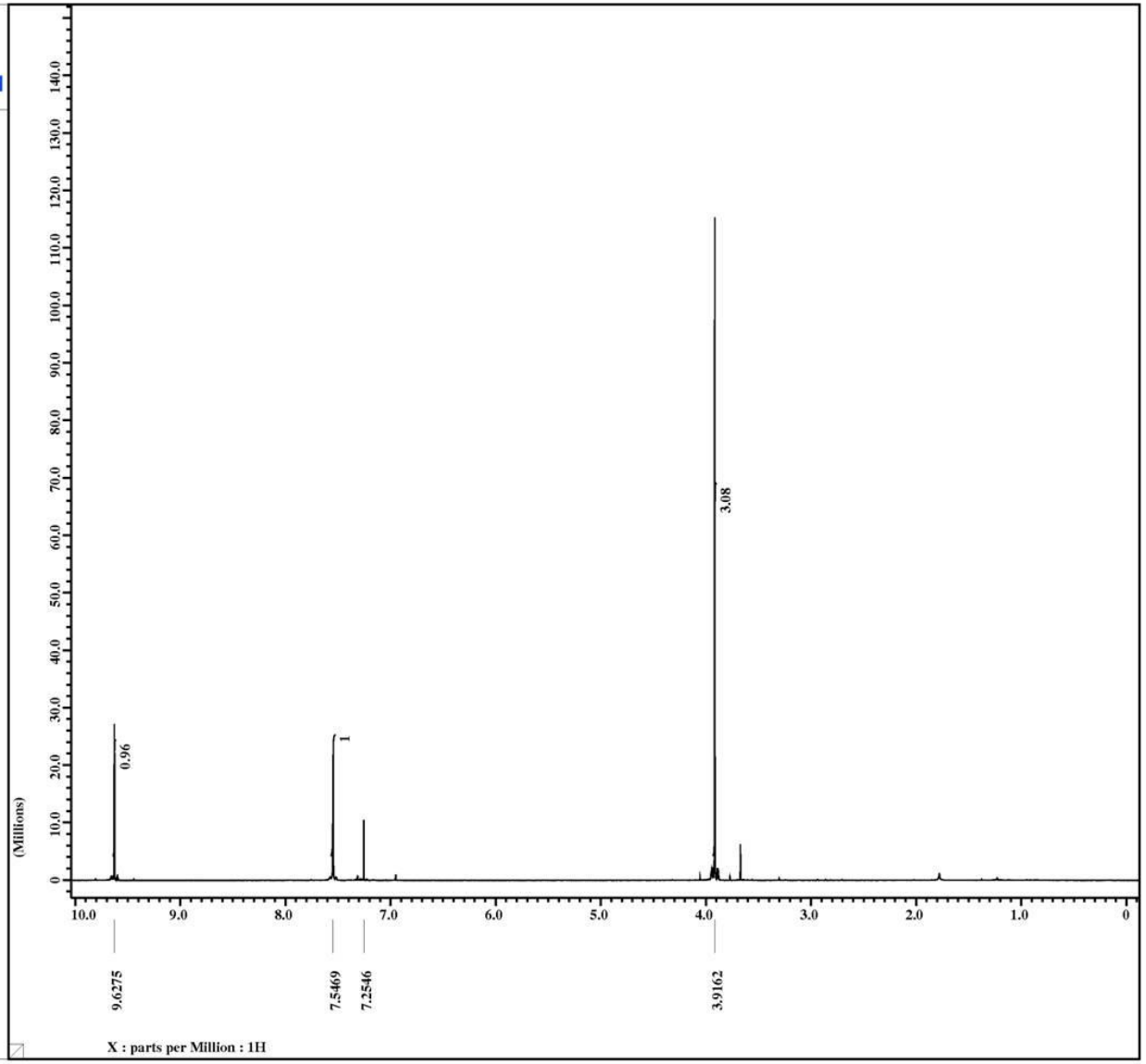
APPENDIX 56

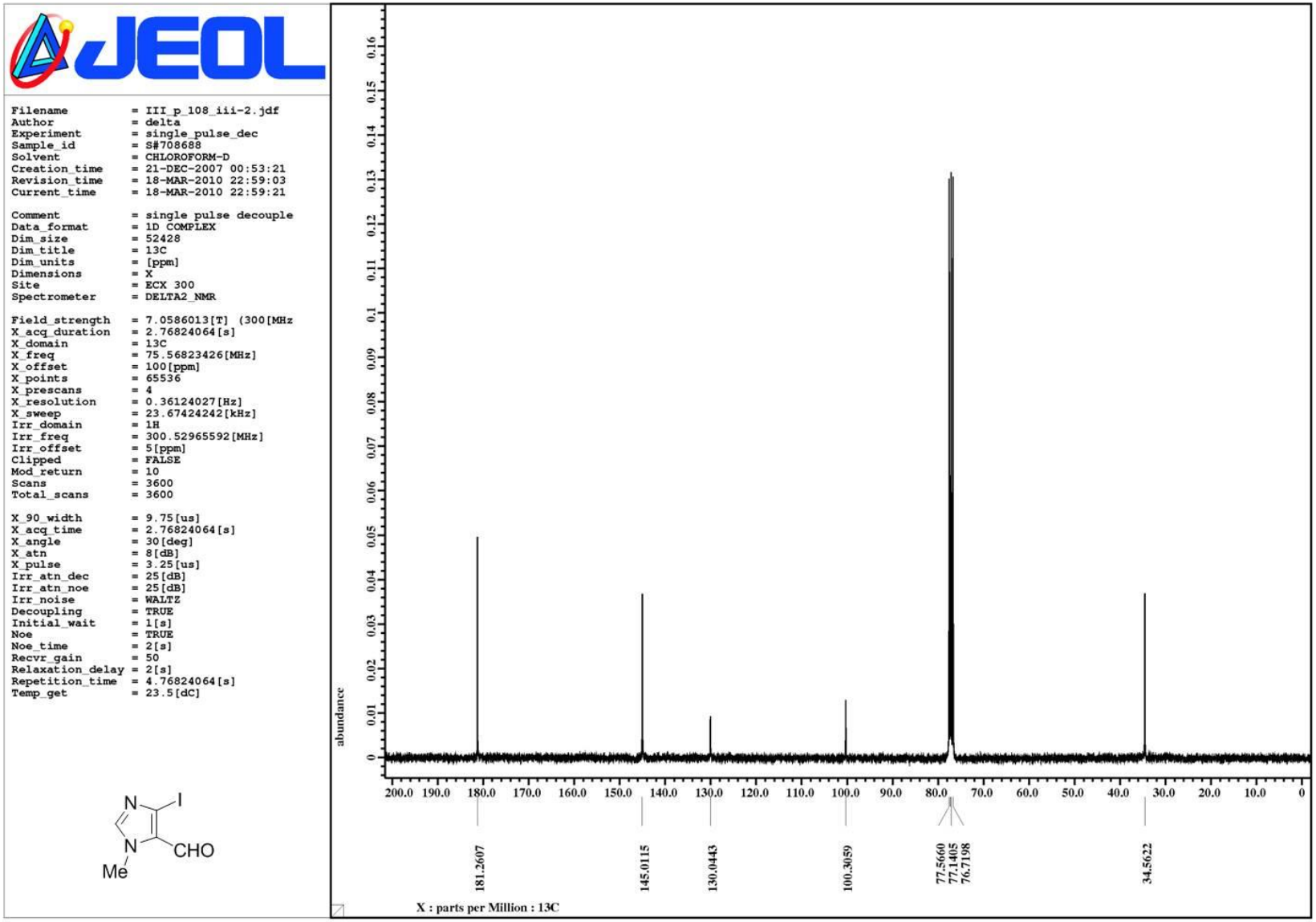
^{1}H and ^{13}C NMR Spectra of

4-Iodo-1-methyl-1*H*-imidazole-5-carboxaldehyde (**178**)

```
Filename          = III_p_108_iii-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#695176
Solvent           = CHLOROFORM-D
Creation_time     = 21-DEC-2007 00:32:21
Revision_time     = 18-MAR-2010 22:57:24
Current_time      = 18-MAR-2010 22:57:54

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 21
Relaxation_delay  = 4[s]
Temp_get          = 24.9[dC]
Unblank_time      = 2[us]
```







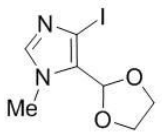
```
Filename          = III_p_108_iii-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#708688
Solvent           = CHLOROFORM-D
Creation_time     = 21-DEC-2007 00:53:21
Revision_time     = 18-MAR-2010 22:59:03
Current_time      = 18-MAR-2010 22:59:21

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 3600
Total_scans       = 3600

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```

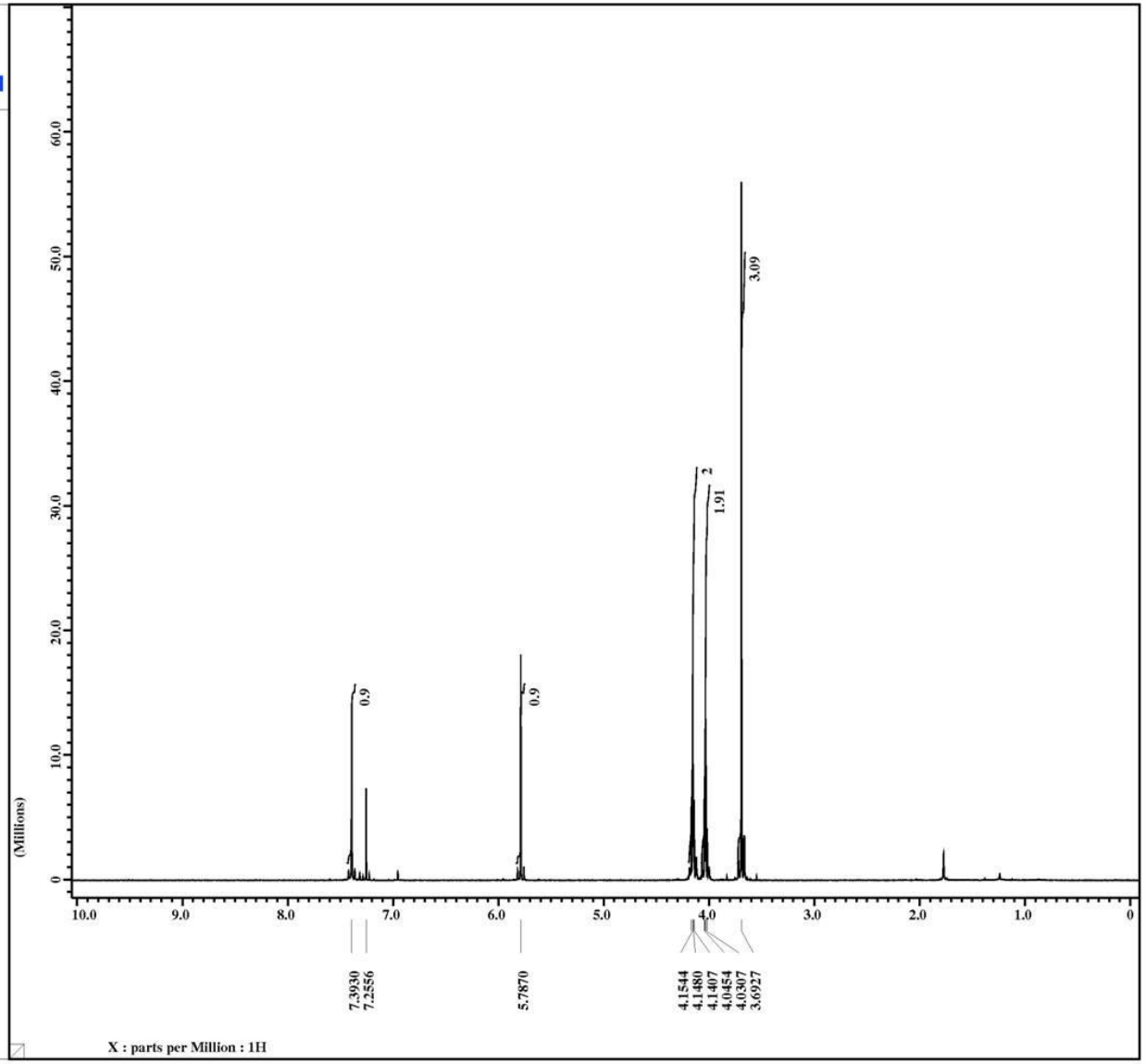
APPENDIX 57

^{1}H and ^{13}C NMR Spectra of

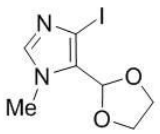
5-[1,3]Dioxolan-2-yl-4-iodo-1-methyl-1*H*-imidazole (**179**)

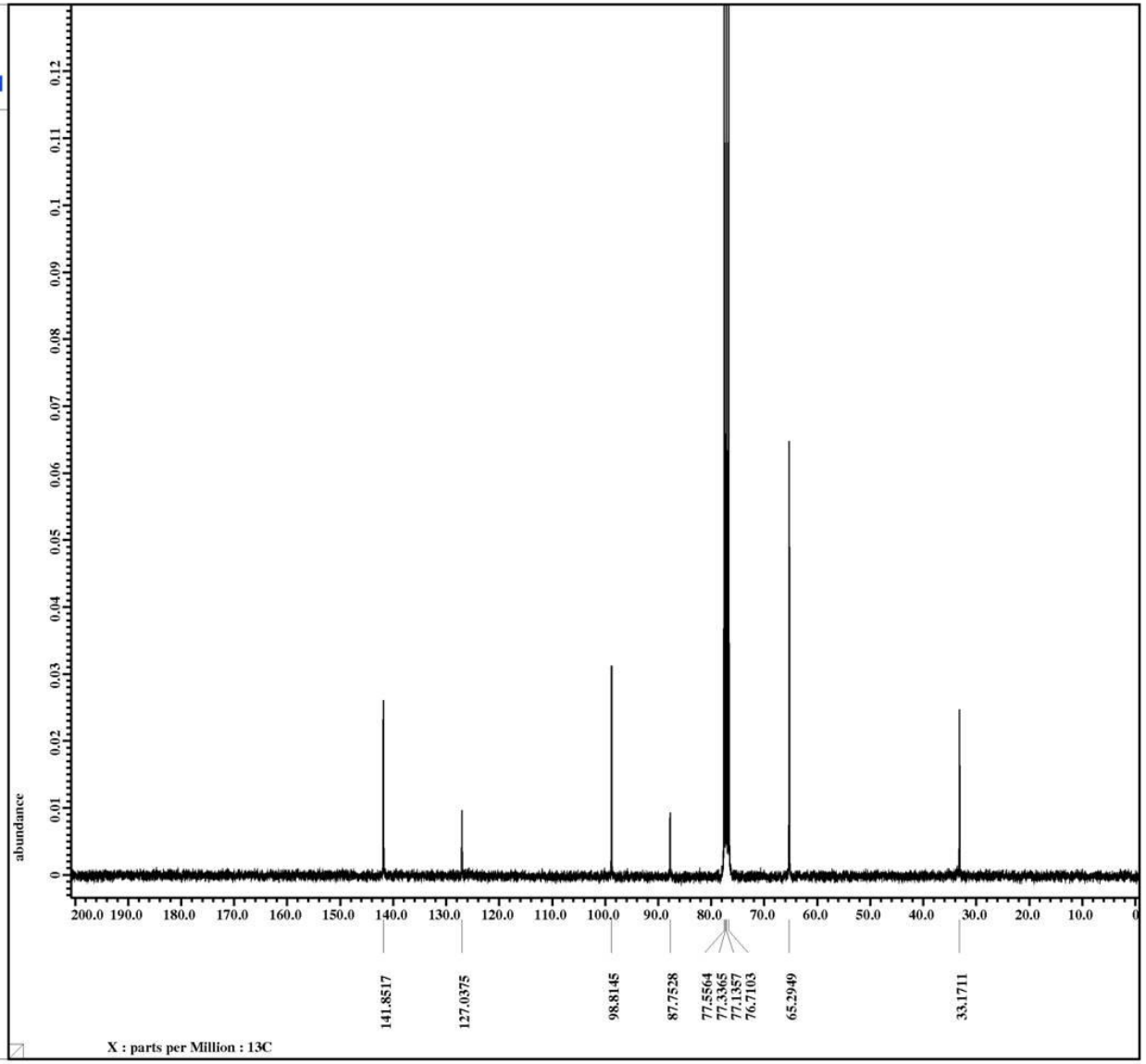
```
Filename          = III_p_109_ii-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#359898
Solvent           = CHLOROFORM-D
Creation_time     = 28-DEC-2007 15:17:22
Revision_time     = 18-MAR-2010 23:06:17
Current_time      = 18-MAR-2010 23:06:42

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 25[dC]
Unblank_time      = 2[us]
```





```
Filename          = III_p_109_ii-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#714454
Solvent           = CHLOROFORM-D
Creation_time     = 29-DEC-2007 04:45:17
Revision_time     = 29-DEC-2007 09:17:21
Current_time      = 18-MAR-2010 23:11:17

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```





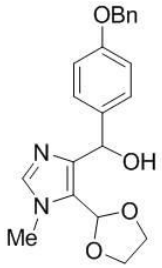
APPENDIX 58

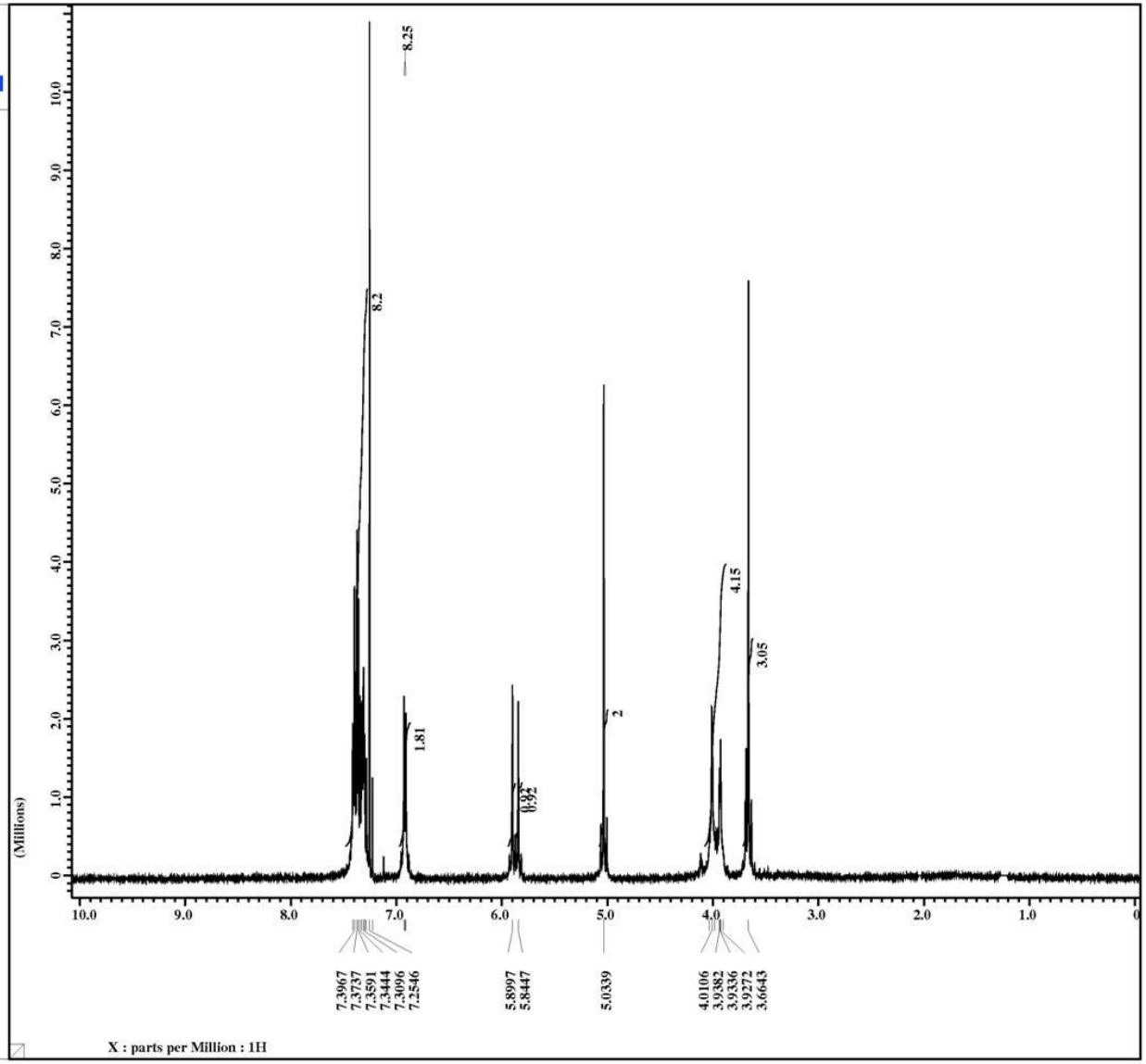
^{1}H and ^{13}C NMR Spectra of

4-(4-Benzyloxyphenyl)hydroxymethyl-5-([1,3]dioxolan-2-yl)-1-methyl-1*H*-imidazole

(**180**)

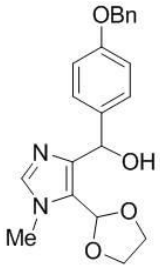
Filename	= IV_p_001-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#623247
Solvent	= CHLOROFORM-D
Creation_time	= 30-APR-2008 23:29:23
Revision_time	= 18-MAR-2010 23:26:52
Current_time	= 18-MAR-2010 23:27:17
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 23
Relaxation_delay	= 4[s]
Temp_get	= 25.3[dC]
Unblank_time	= 2[us]

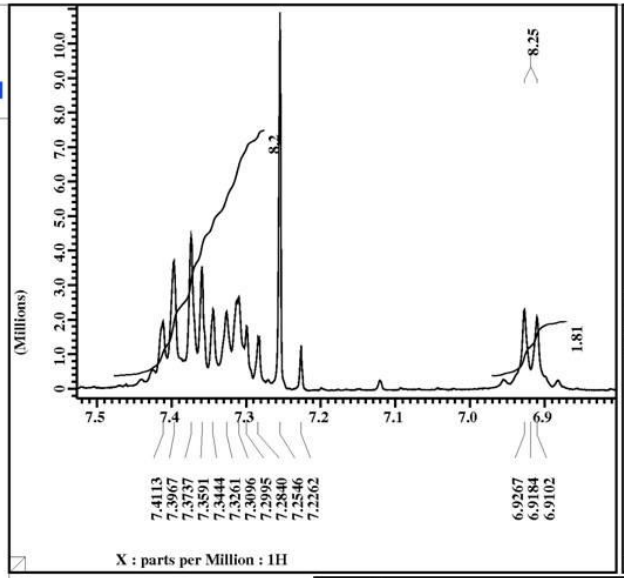


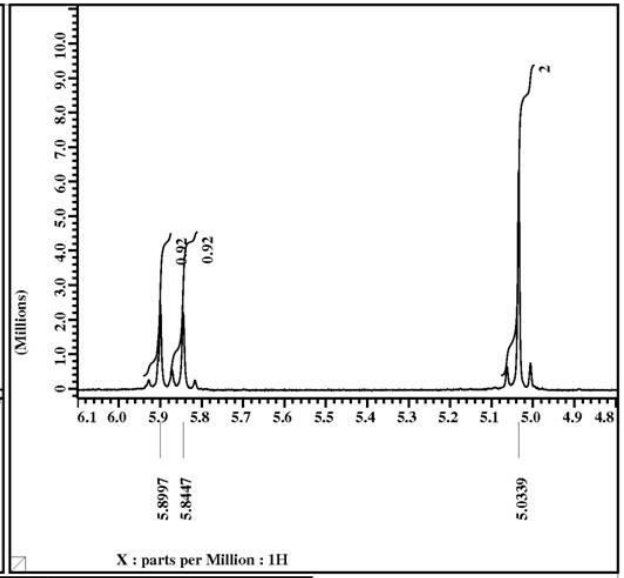


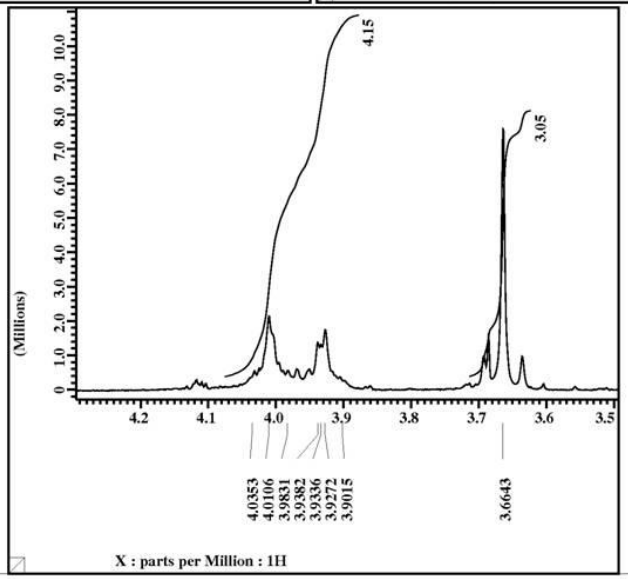
JEOL

Filename	= IV_p_001-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#623247
Solvent	= CHLOROFORM-D
Creation_time	= 30-APR-2008 23:29:23
Revision_time	= 18-MAR-2010 23:26:52
Current_time	= 18-MAR-2010 23:28:03
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 23
Relaxation_delay	= 4[s]
Temp_get	= 25.3[dC]
Unblank_time	= 2[us]

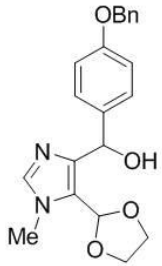


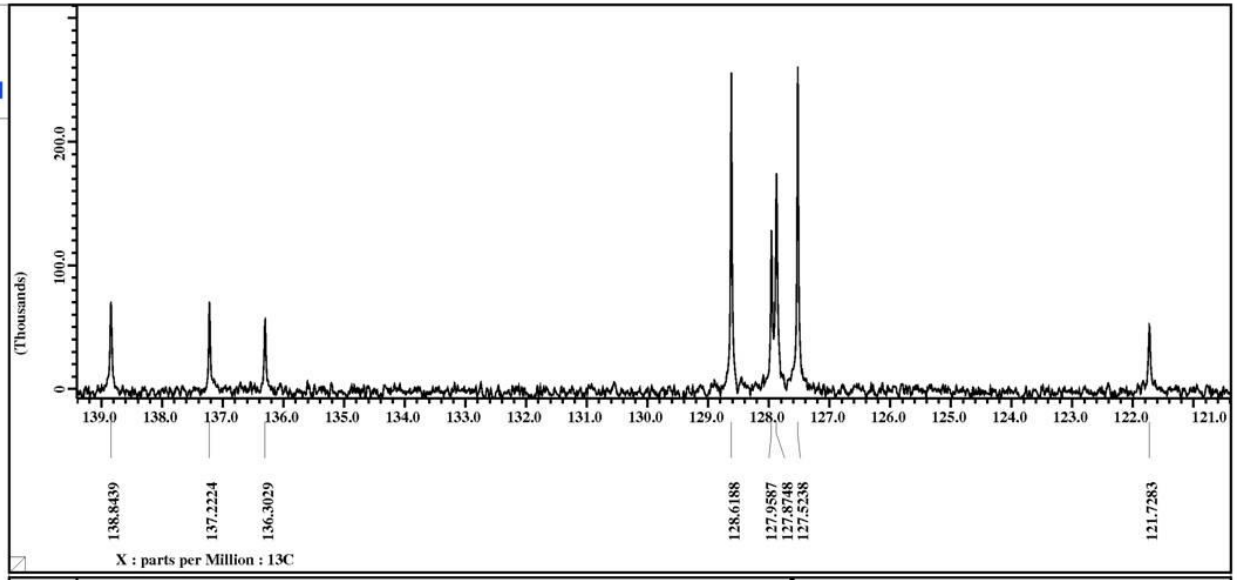


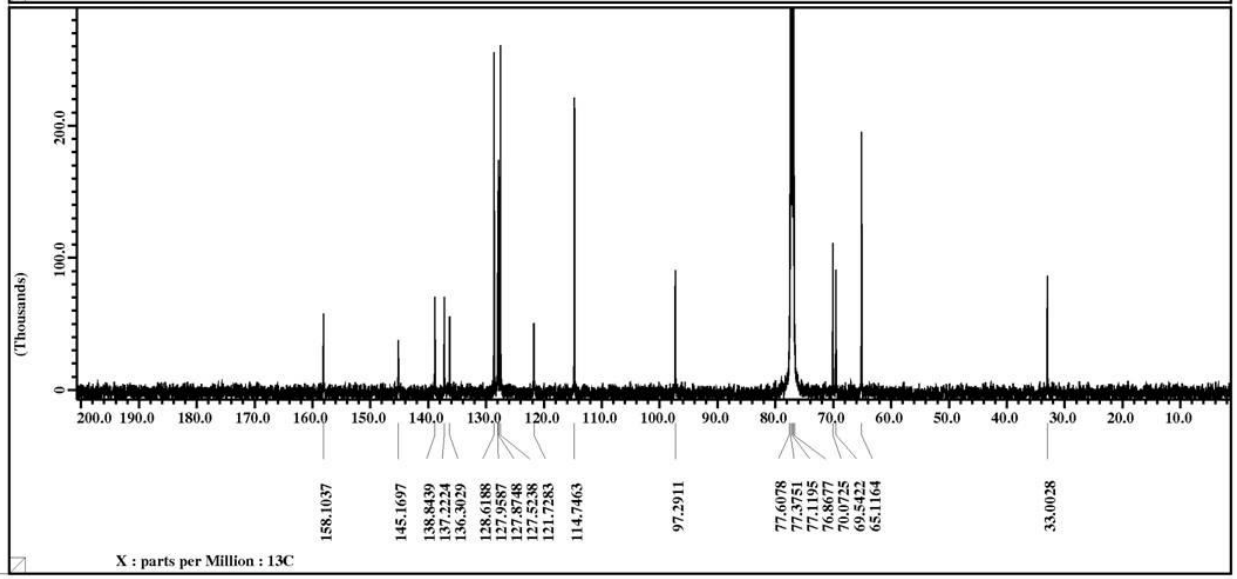




```
Filename          = IV_p_017_Acetal-pure-
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#857226
Solvent           = CHLOROFORM-D
Creation_time     = 16-JUN-2008 15:27:09
Revision_time     = 18-MAR-2010 23:21:47
Current_time      = 18-MAR-2010 23:23:53

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 6400
Total_scans       = 6400

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.2[dC]
Unblank_time      = 2[us]
```





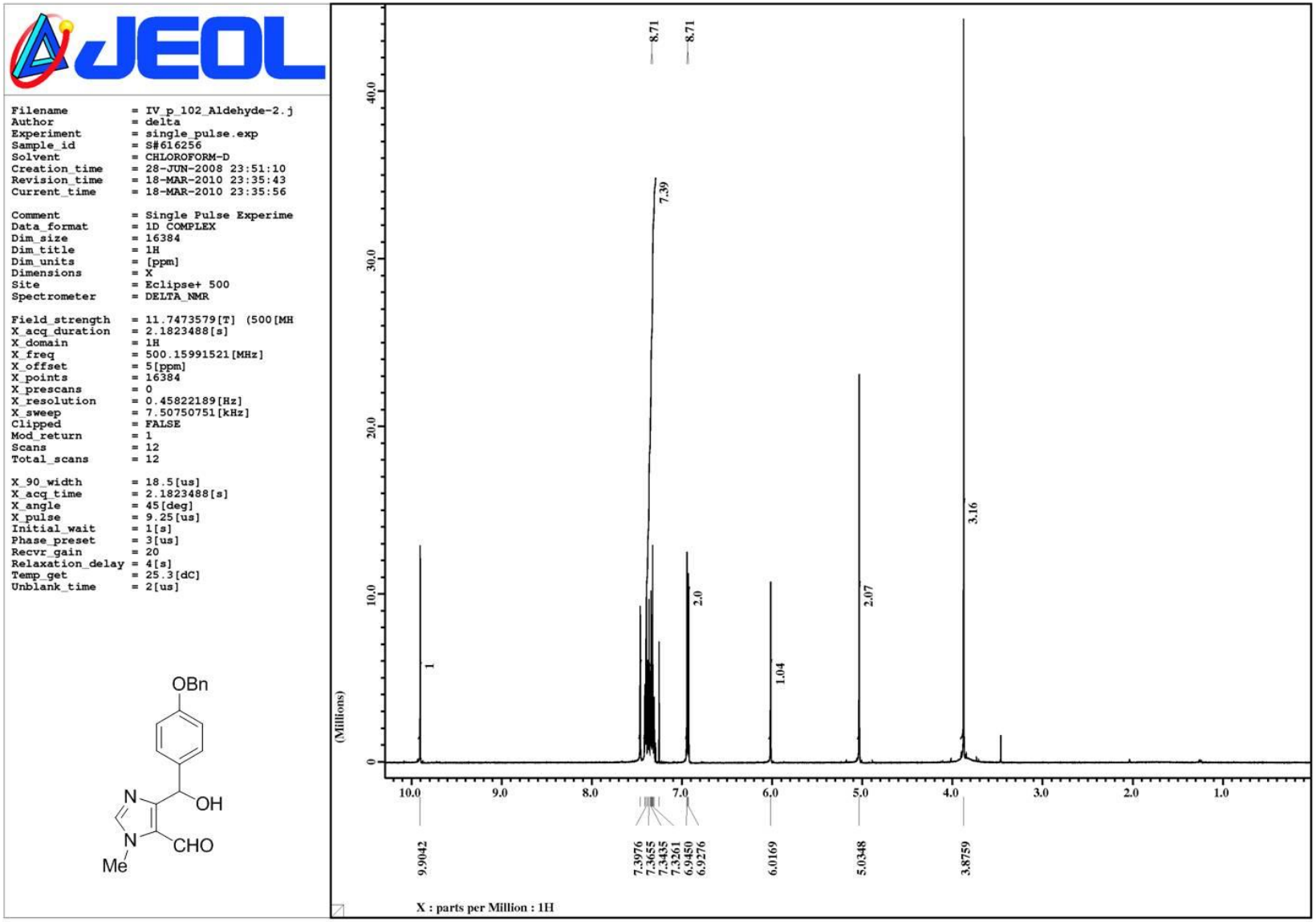


APPENDIX 59

^{1}H and ^{13}C NMR Spectra of

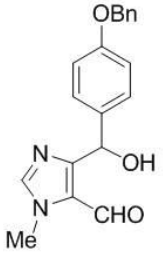
4-(4-benzyloxyphenyl)hydroxymethyl-1-methyl-1*H*-imidazole-5-carboxaldehyde

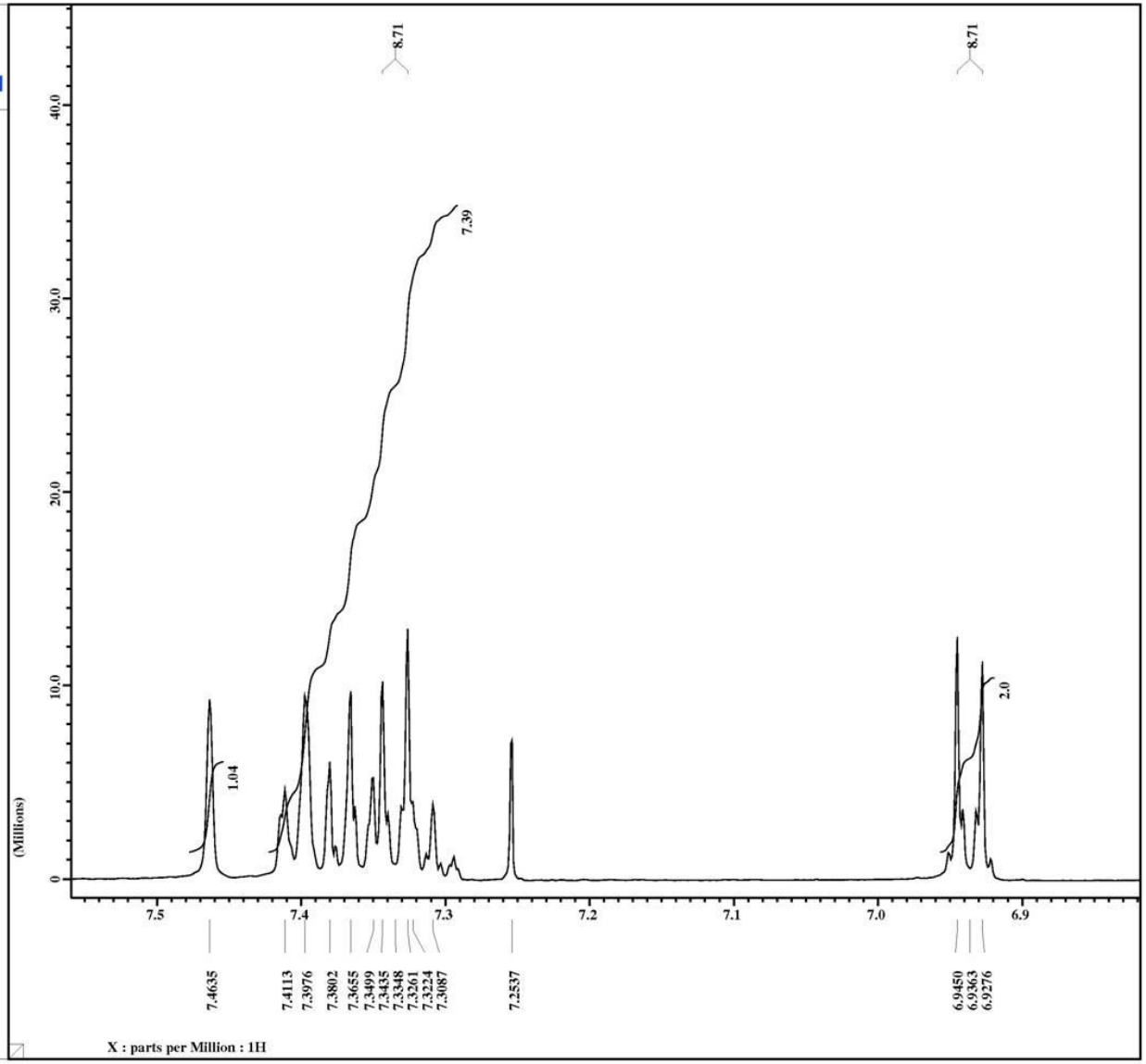
(**181a**)



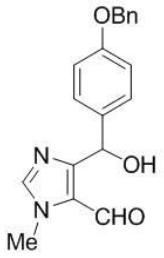
```
Filename          = IV_p_102_Aldehyde-2.j
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#616256
Solvent           = CHLOROFORM-D
Creation_time     = 28-JUN-2008 23:51:10
Revision_time     = 18-MAR-2010 23:35:43
Current_time      = 18-MAR-2010 23:35:56

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 25.3[dC]
Unblank_time      = 2[us]
```

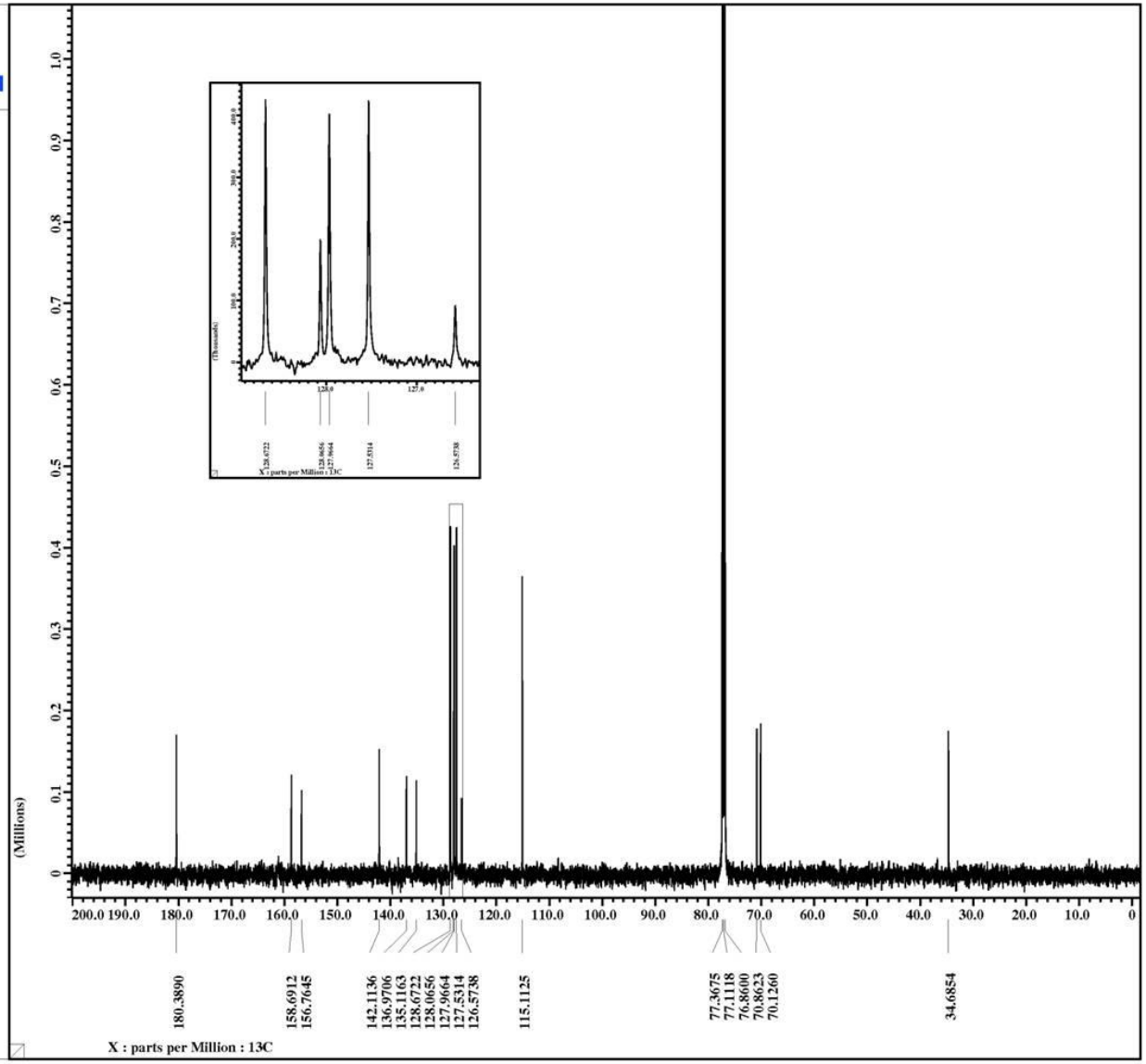
Filename	= IV_p_102_Aldehyde-2.j
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#616256
Solvent	= CHLOROFORM-D
Creation_time	= 28-JUN-2008 23:51:10
Revision_time	= 18-MAR-2010 23:35:43
Current_time	= 18-MAR-2010 23:36:15
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 20
Relaxation_delay	= 4[s]
Temp_get	= 25.3[dC]
Unblank_time	= 2[us]





```
Filename          = IV_p_102_Aldehyde-3.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#616348
Solvent           = CHLOROFORM-D
Creation_time     = 29-JUN-2008 02:10:29
Revision_time     = 18-MAR-2010 23:36:37
Current_time      = 18-MAR-2010 23:37:33

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 1600
Total_scans       = 1600

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.4[dC]
Unblank_time      = 2[us]
```



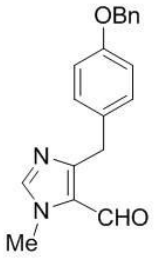


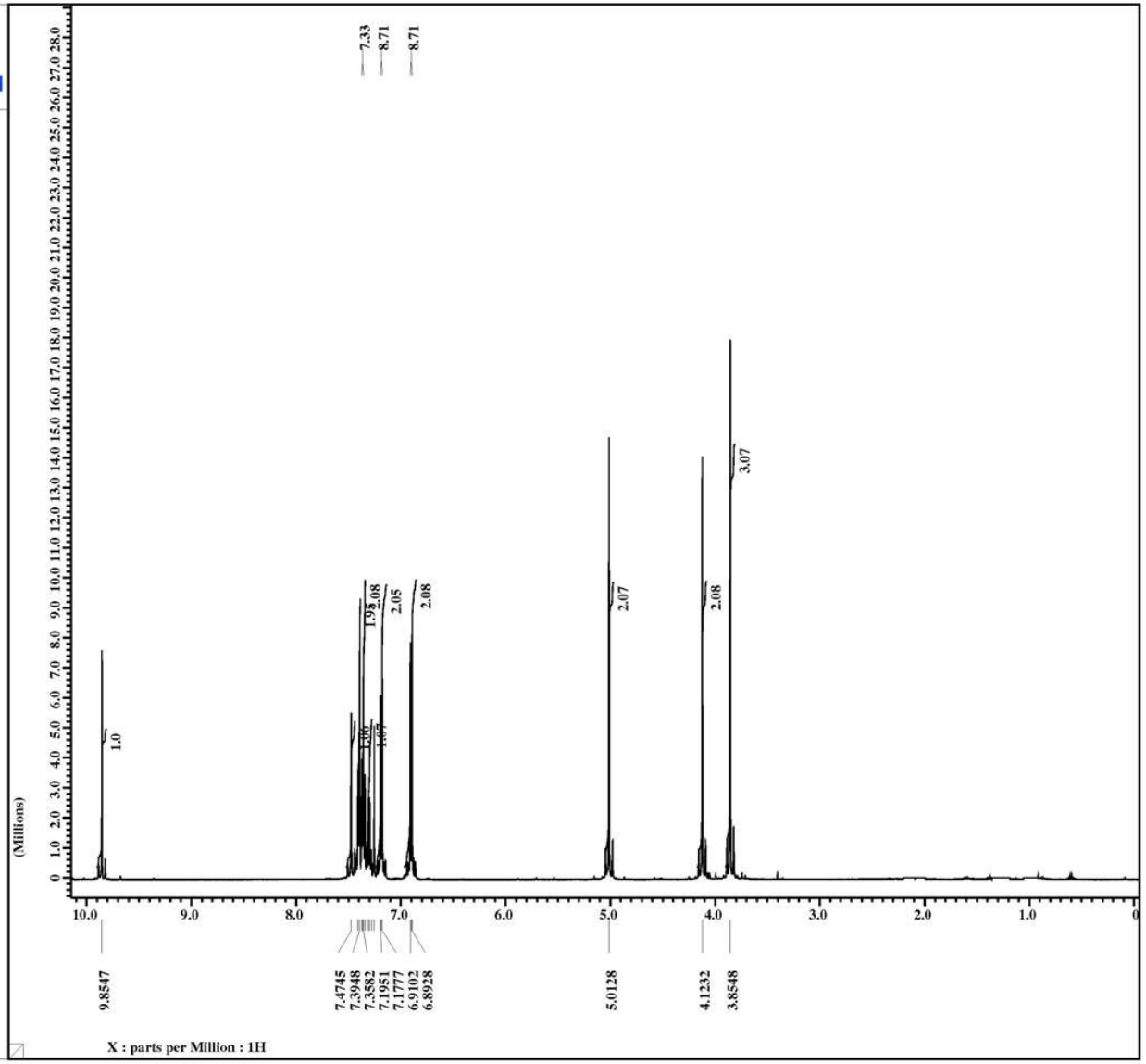
APPENDIX 60

^{1}H and ^{13}C NMR Spectra of

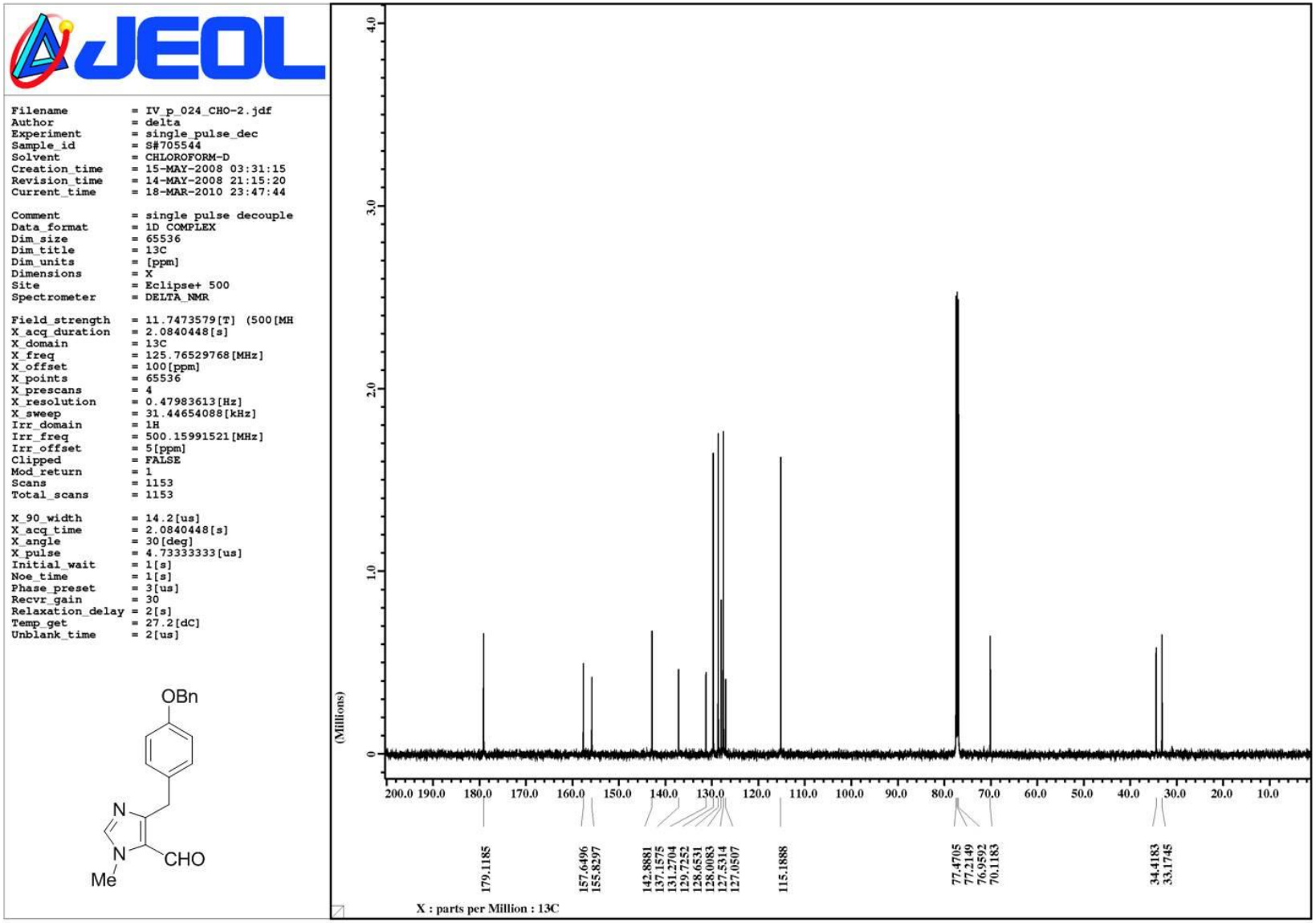
4-(4-Benzyloxybenzyl)-1-methyl-1*H*-imidazole-5-carbaldehyde (**181b**)

```
Filename          = IV_p_024_CHO-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#702782
Solvent           = CHLOROFORM-D
Creation_time     = 15-MAY-2008 01:49:35
Revision_time     = 18-MAR-2010 23:46:13
Current_time      = 18-MAR-2010 23:46:40

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```





Filename = IV_p_024_CHO-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#702782
Solvent = CHLOROFORM-D
Creation_time = 15-MAY-2008 01:49:35
Revision_time = 18-MAR-2010 23:47:01
Current_time = 18-MAR-2010 23:47:14
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 15
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
OBn
N
N
Me
CHO
(Millions)
X : parts per Million : 1H
8.71
8.71
7.33
7.33
6.87
2.08
2.05
2.08
1.95
1.07
1.06
7.4745
7.4086
7.4017
7.3948
7.3728
7.3655
7.3582
7.3426
7.3124
7.3050
7.2977
7.2830
7.2546
7.1951
7.1864
7.1777
6.9102
6.9015
6.8928

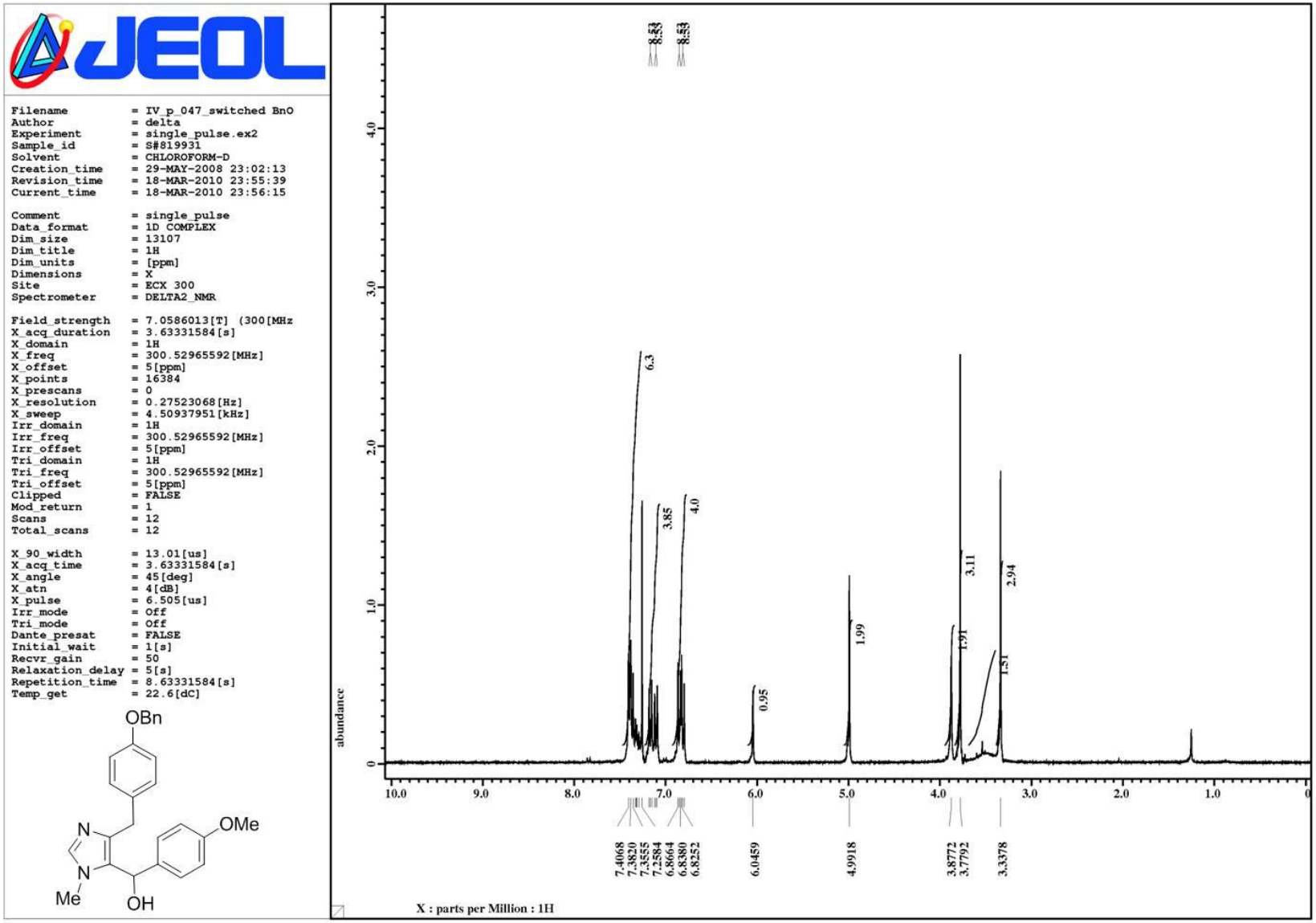


```
Filename          = IV_p_024_CHO-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#705544
Solvent           = CHLOROFORM-D
Creation_time     = 15-MAY-2008 03:31:15
Revision_time     = 14-MAY-2008 21:15:20
Current_time      = 18-MAR-2010 23:47:44

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 1153
Total_scans       = 1153

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.2[dC]
Unblank_time      = 2[us]
```

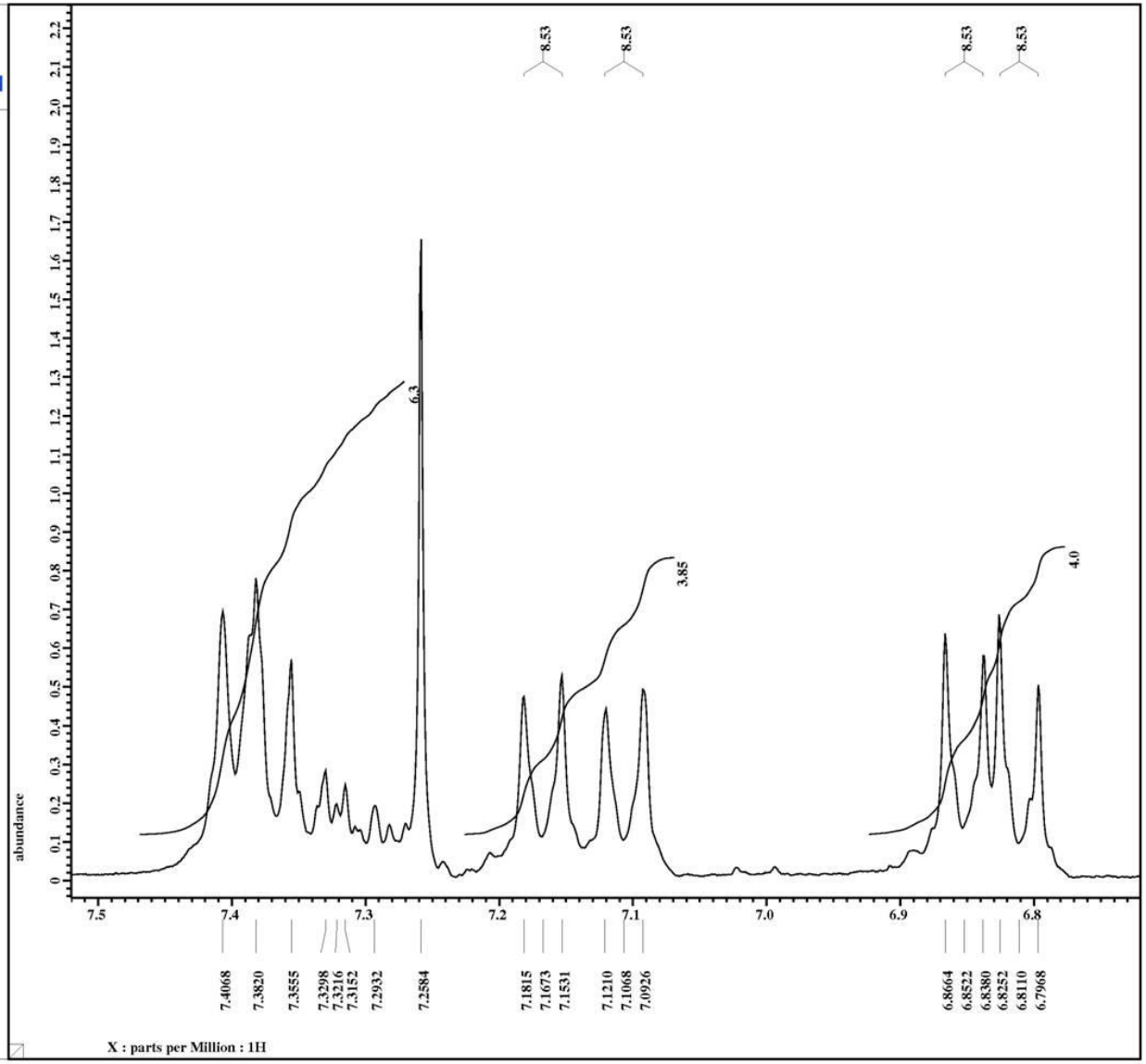
APPENDIX 61

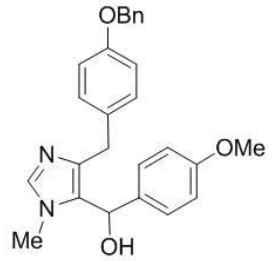
^{1}H and ^{13}C NMR Spectra of

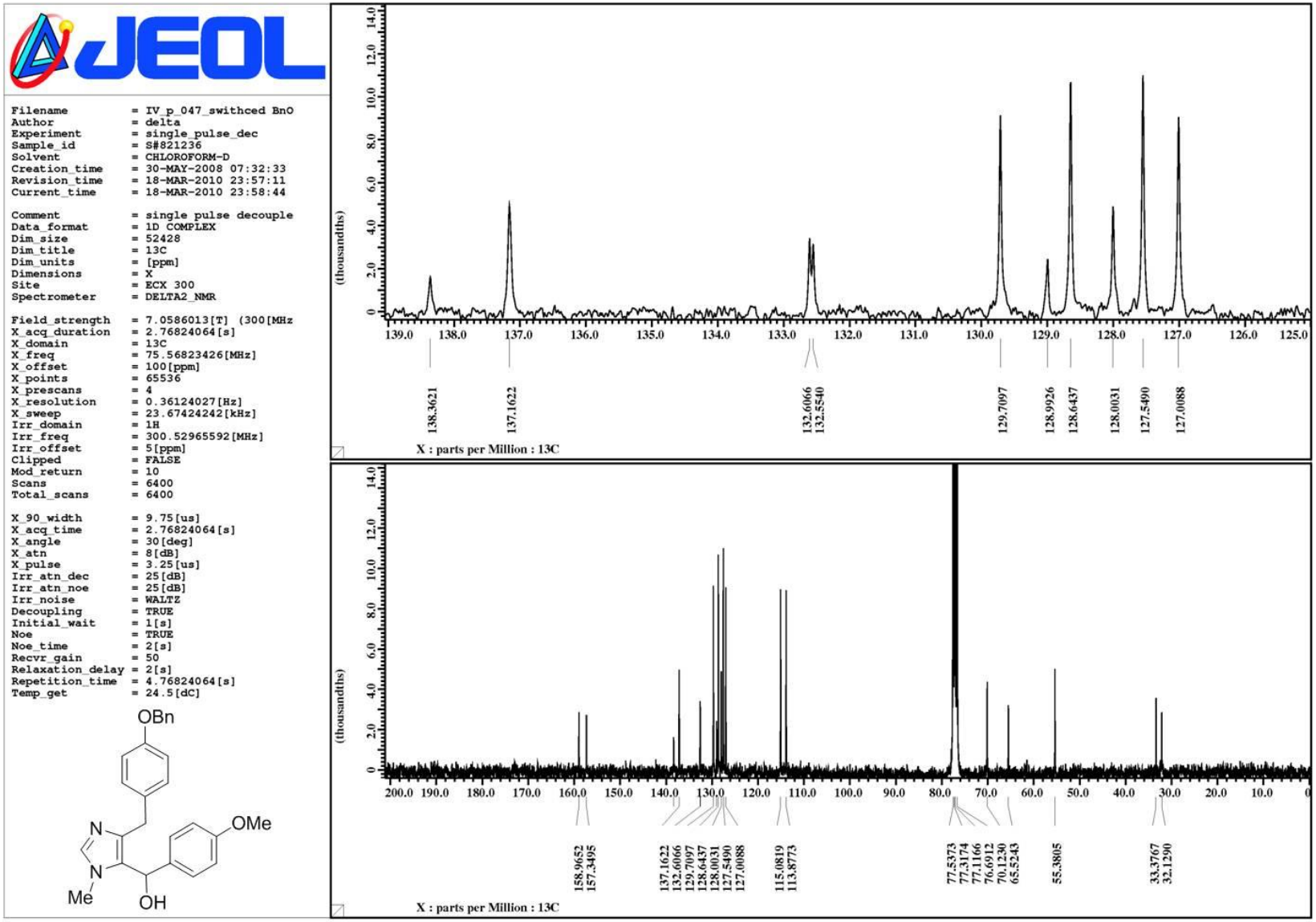
4-(4-Benzyloxybenzyl)-5-[hydroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**182**)


Filename = IV_p_047_switched BnO
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#819931
Solvent = CHLOROFORM-D
Creation_time = 29-MAY-2008 23:02:13
Revision_time = 18-MAR-2010 23:55:39
Current_time = 18-MAR-2010 23:56:15
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 50
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 22.6[dC]
OBn
OMe
N
N
Me
OH
abundance
8.53
8.53
8.53
6.3
3.85
4.0
0.95
1.99
1.91
3.11
2.94
1.51
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
7.4068
7.3820
7.3555
7.2584
6.8664
6.8380
6.8252
6.0459
4.9918
3.8772
3.7792
3.3378
X : parts per Million : 1H

Filename	= IV_p_047_switched BnO
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#819931
Solvent	= CHLOROFORM-D
Creation_time	= 29-MAY-2008 23:02:13
Revision_time	= 18-MAR-2010 23:55:39
Current_time	= 18-MAR-2010 23:56:32
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 50
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.6[dC]







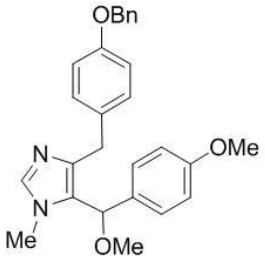
```
Filename          = IV_p_047_swithced BnO
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#821236
Solvent           = CHLOROFORM-D
Creation_time     = 30-MAY-2008 07:32:33
Revision_time     = 18-MAR-2010 23:57:11
Current_time      = 18-MAR-2010 23:58:44

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 24.5[dC]
```

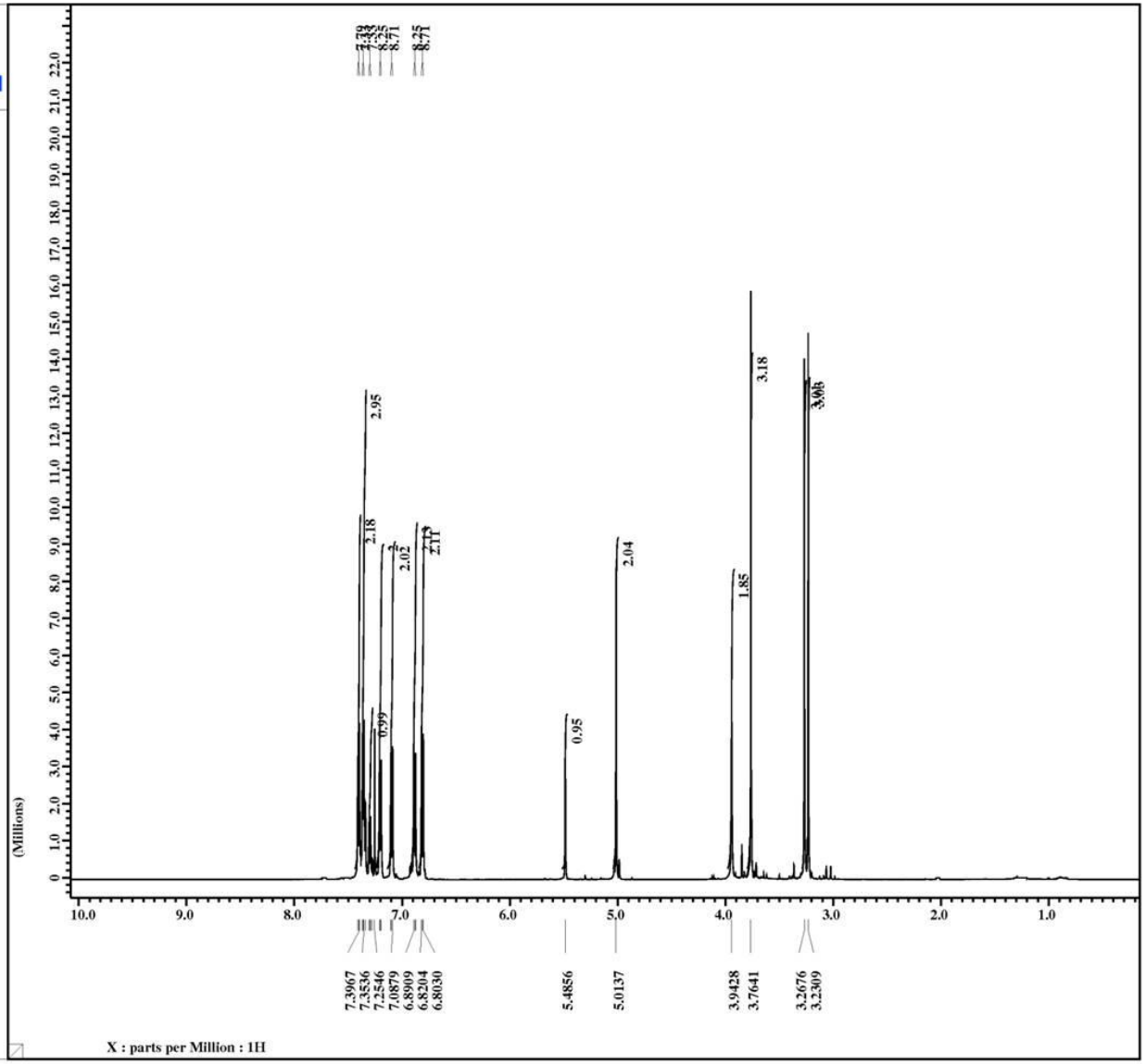
APPENDIX 62

^{1}H and ^{13}C NMR Spectra of

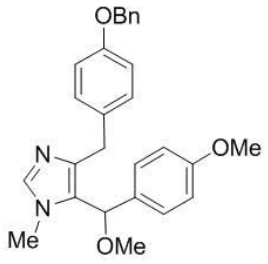
4-(4-Benzyloxybenzyl)-5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**183**)

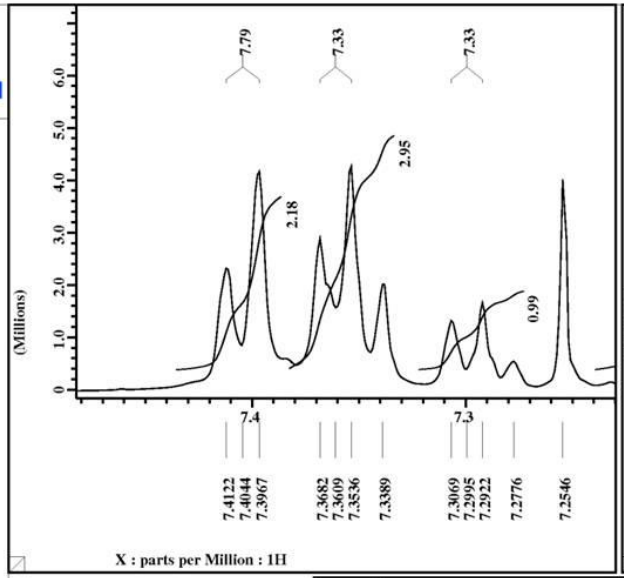
Filename	= IV_p_084_BnOMe-4.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#828456
Solvent	= CHLOROFORM-D
Creation_time	= 17-JUN-2008 05:40:06
Revision_time	= 19-MAR-2010 00:08:32
Current_time	= 19-MAR-2010 00:08:58
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 12
Relaxation_delay	= 4[s]
Temp_get	= 24.9[dC]
Unblank_time	= 2[us]

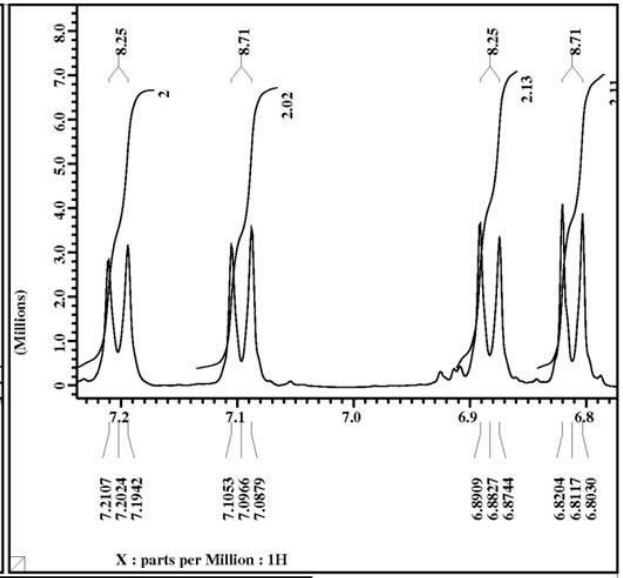


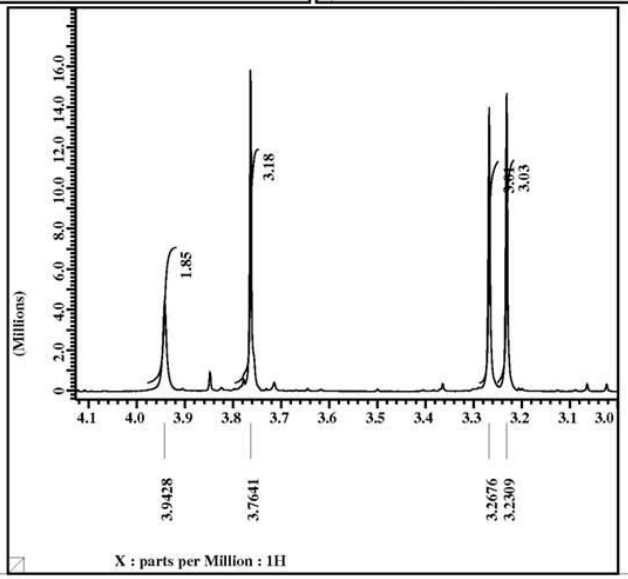


Filename	= IV_p_084_BnOMe-4.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#828456
Solvent	= CHLOROFORM-D
Creation_time	= 17-JUN-2008 05:40:06
Revision_time	= 19-MAR-2010 00:09:47
Current_time	= 19-MAR-2010 00:09:59
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 12
Relaxation_delay	= 4[s]
Temp_get	= 24.9[dC]
Unblank_time	= 2[us]

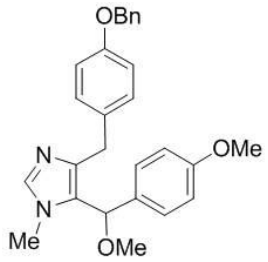


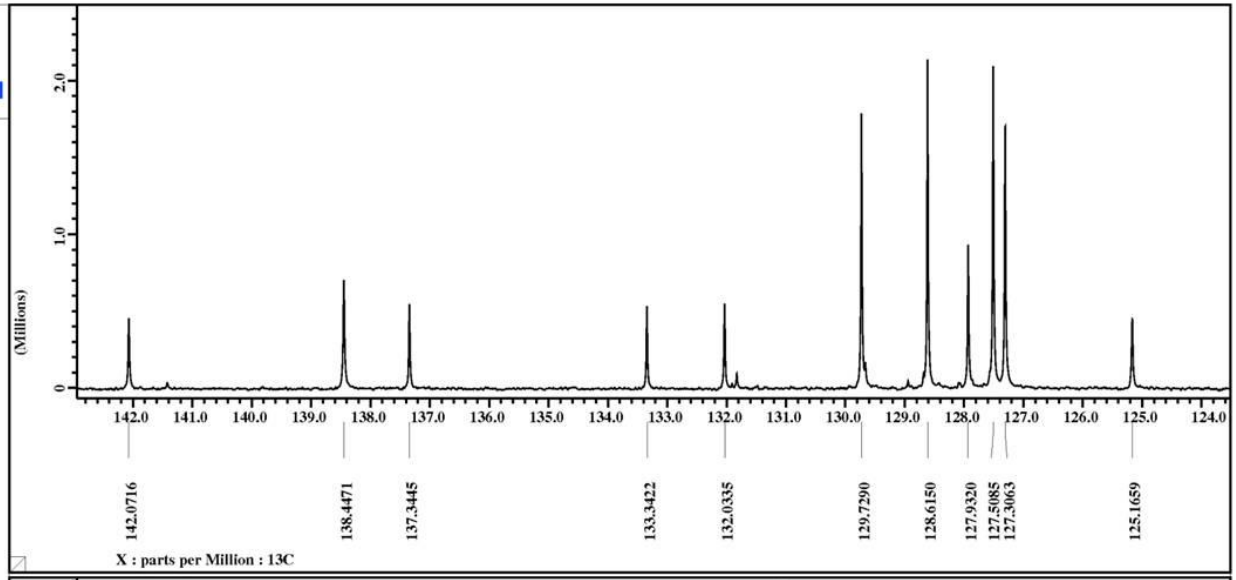


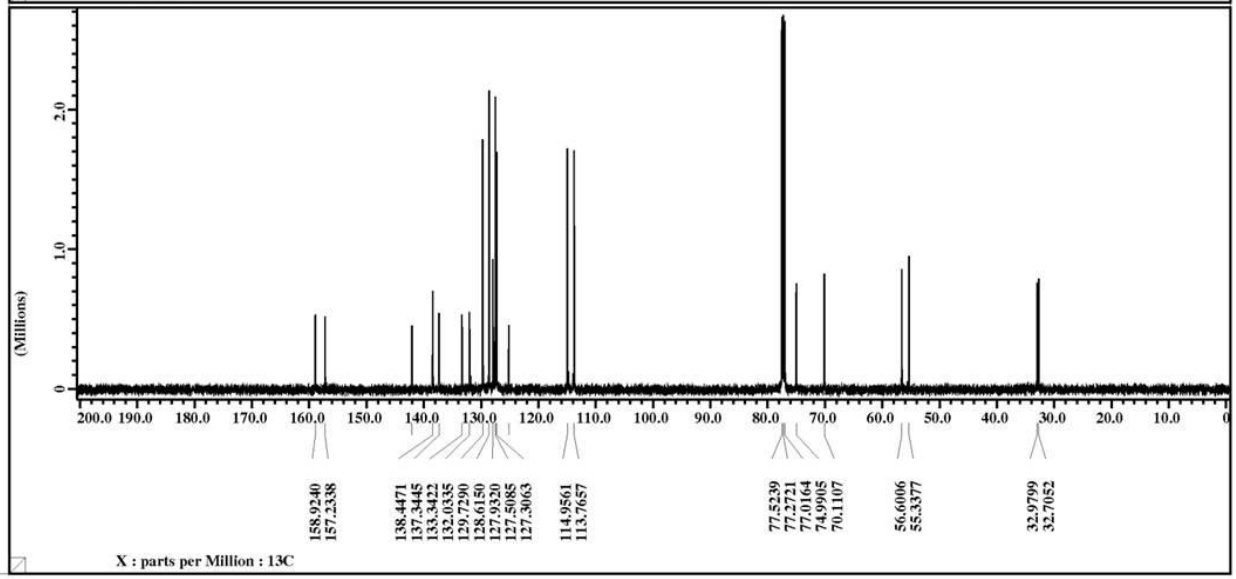




```
Filename          = IV_p_084_BnOMe-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 17-JUN-2008 10:32:47
Revision_time     = 17-JUN-2008 11:38:31
Current_time      = 19-MAR-2010 00:10:55

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 3000
Total_scans       = 3000

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.5[dC]
Unblank_time      = 2[us]
```





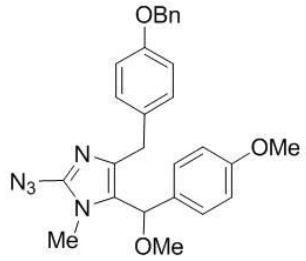


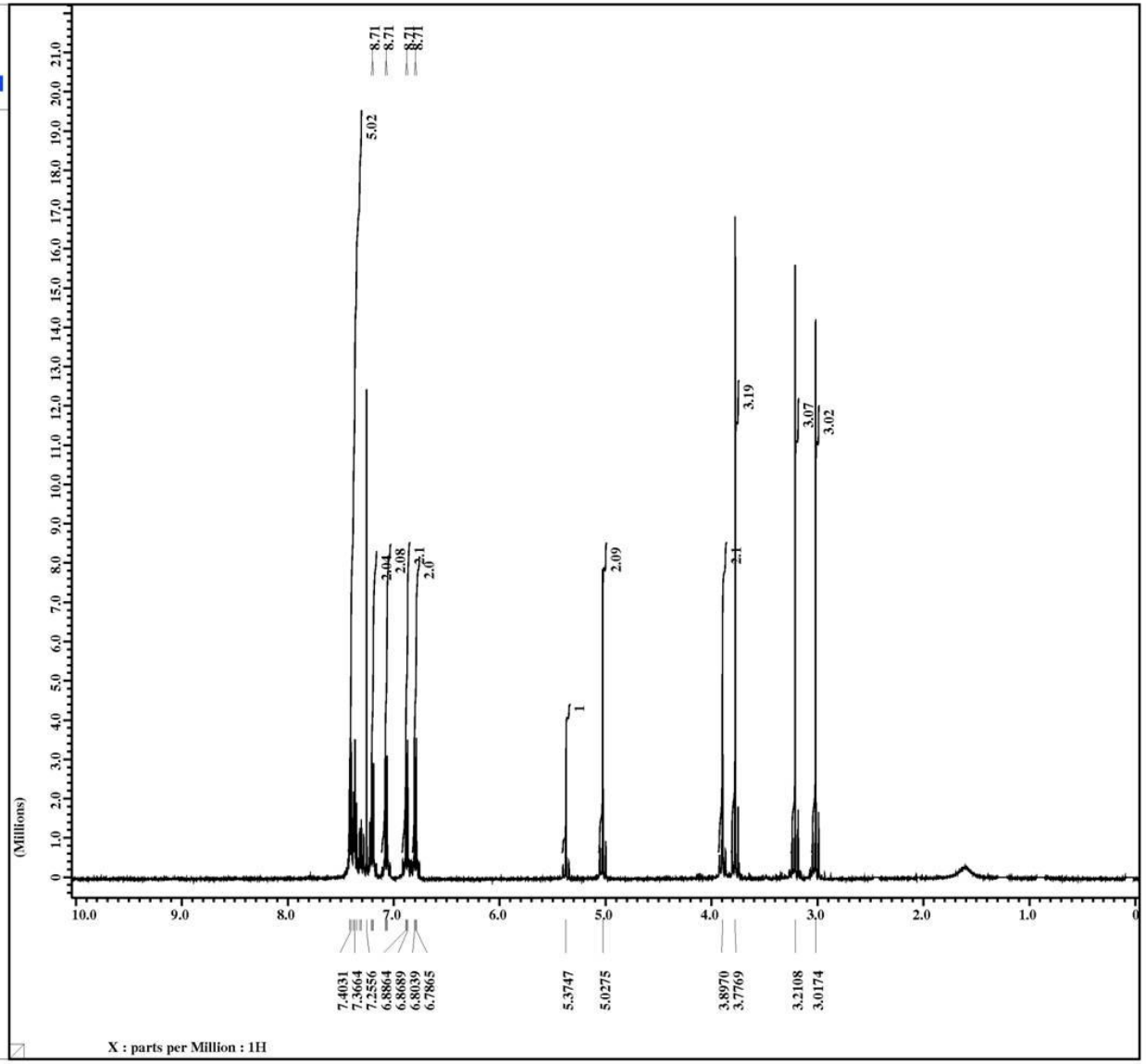
APPENDIX 63

^{1}H and ^{13}C NMR Spectra of

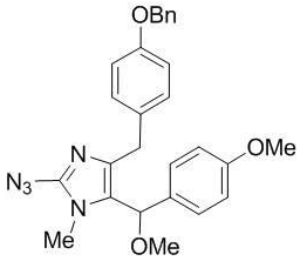
2-Azido-4-(4-benzyloxybenzyl)-5-[methoxy-(4-methoxy)phenyl]methyl-1-methyl-1*H*-imidazole (**184**)

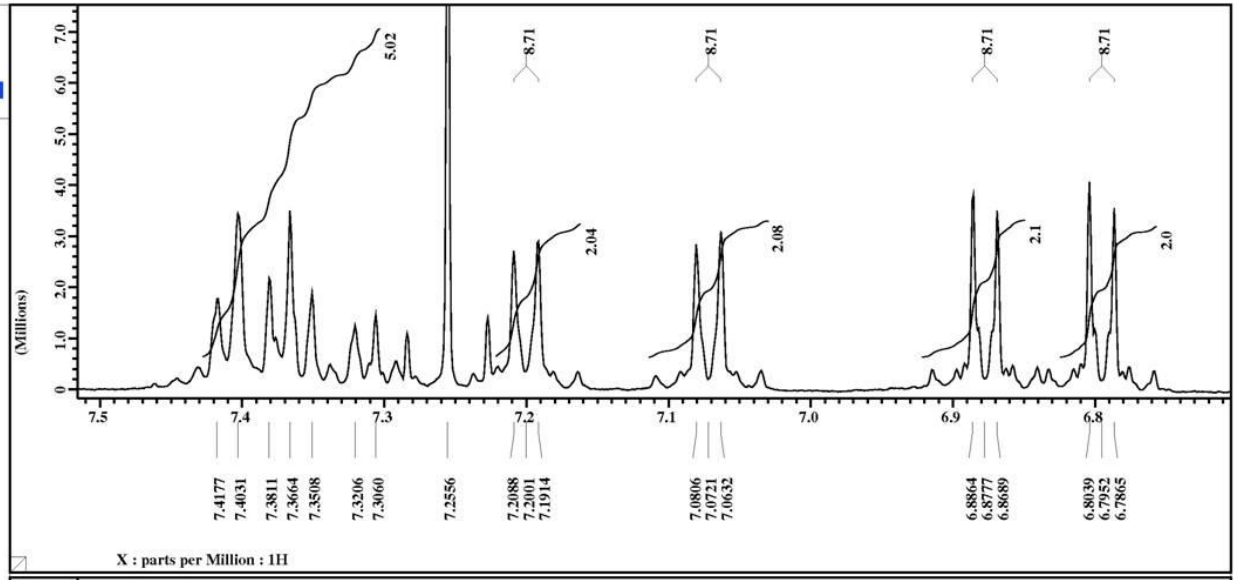
Filename	= IV_p_027_azide-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#687650
Solvent	= CHLOROFORM-D
Creation_time	= 14-MAY-2008 01:24:13
Revision_time	= 19-MAR-2010 00:27:50
Current_time	= 19-MAR-2010 00:28:42
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 23
Relaxation_delay	= 4[s]
Temp_get	= 25.2[dC]
Unblank_time	= 2[us]

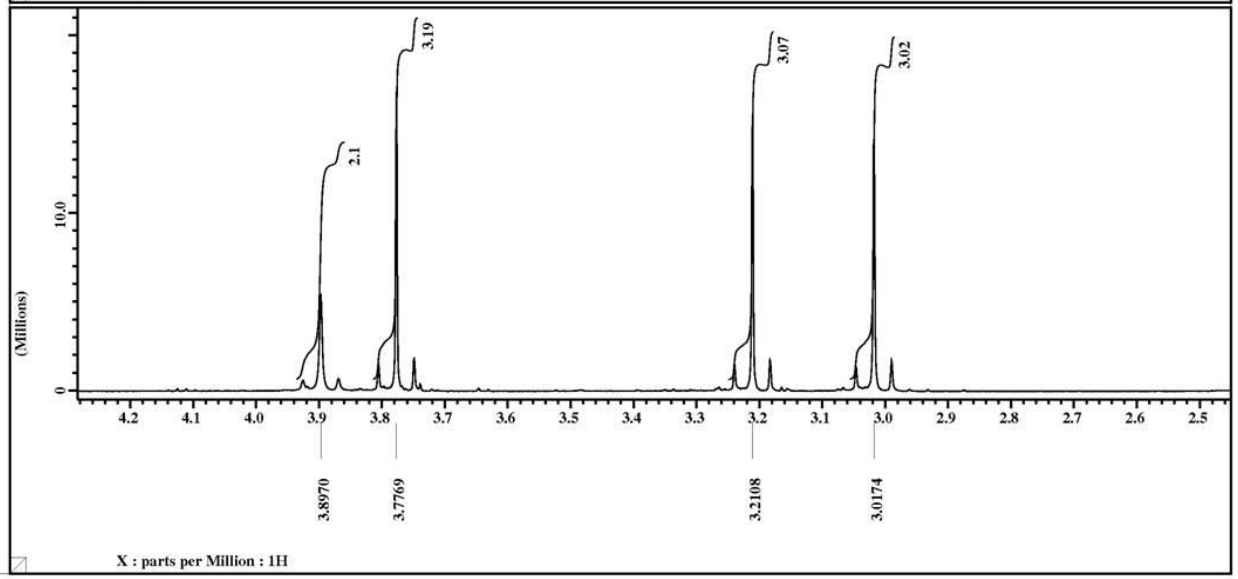




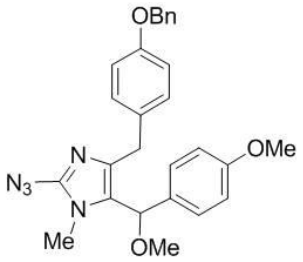
Filename	= IV_p_027_azide-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#687650
Solvent	= CHLOROFORM-D
Creation_time	= 14-MAY-2008 01:24:13
Revision_time	= 19-MAR-2010 00:27:50
Current_time	= 19-MAR-2010 00:29:15
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 23
Relaxation_delay	= 4[s]
Temp_get	= 25.2[dC]
Unblank_time	= 2[us]

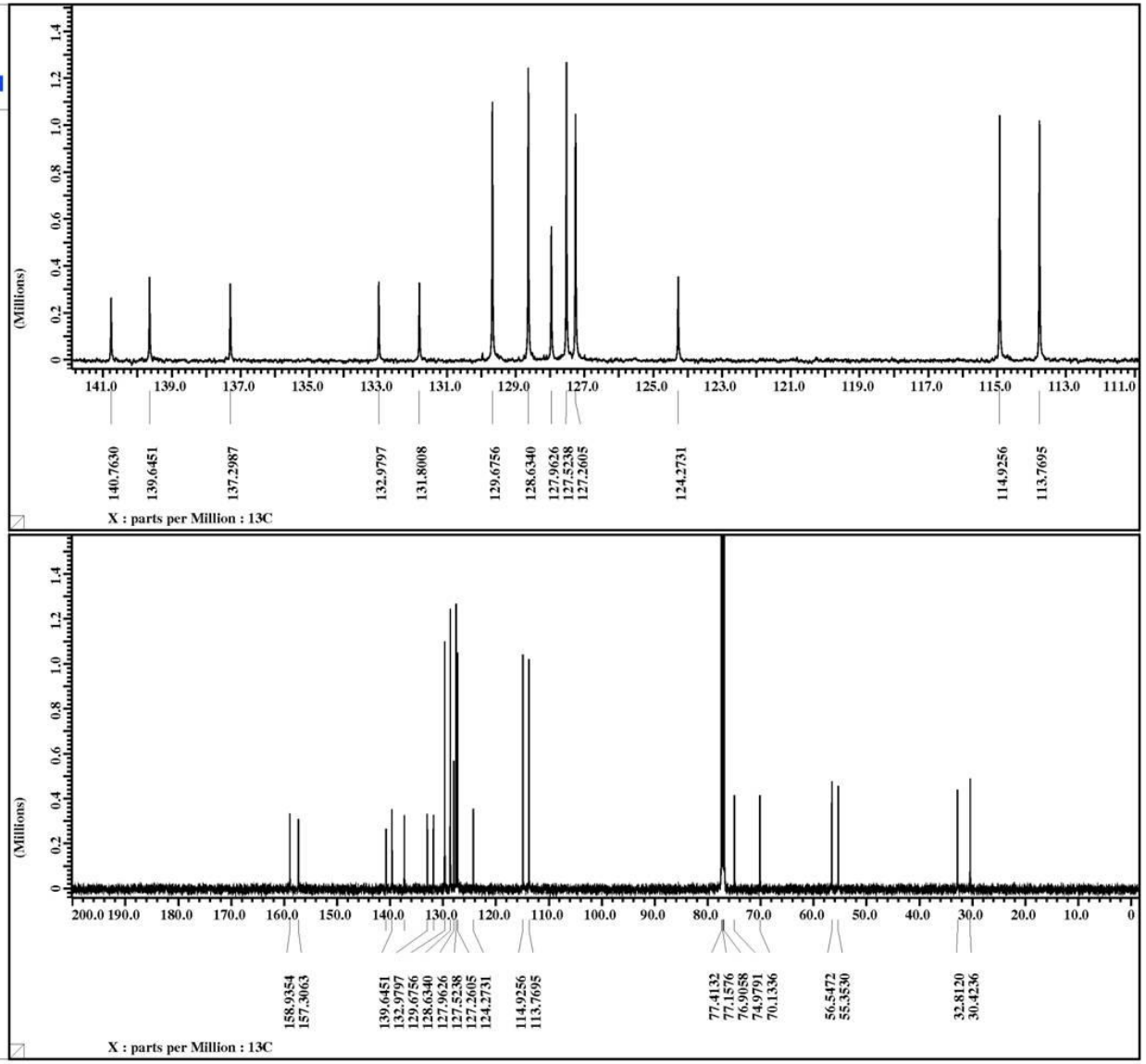






```
Filename          = IV_p_027_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#773866
Solvent           = CHLOROFORM-D
Creation_time     = 14-MAY-2008 08:54:16
Revision_time     = 14-MAY-2008 10:00:30
Current_time      = 19-MAR-2010 00:30:14

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 3600
Total_scans       = 3600

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.2[dC]
Unblank_time      = 2[us]
```

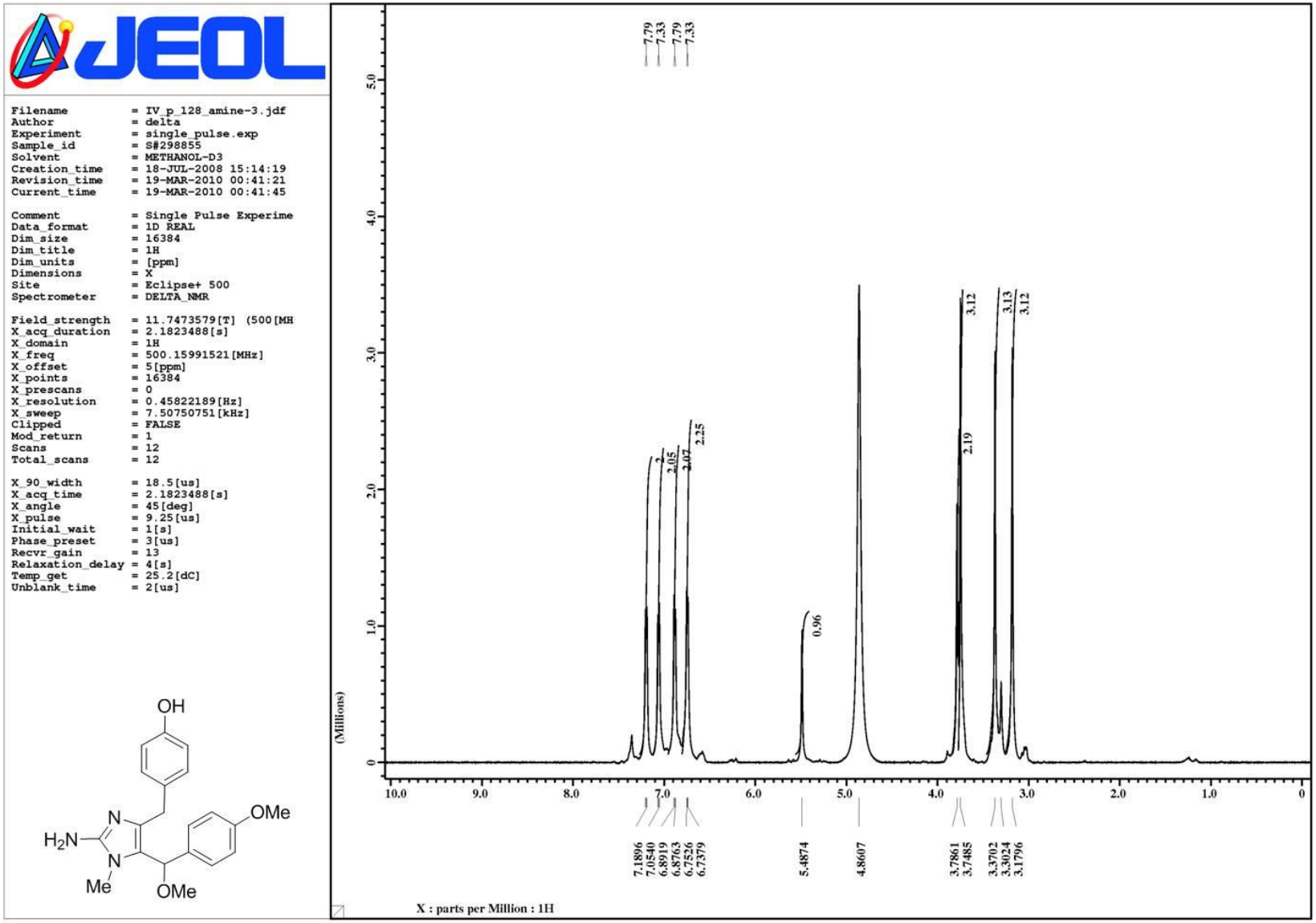




APPENDIX 64

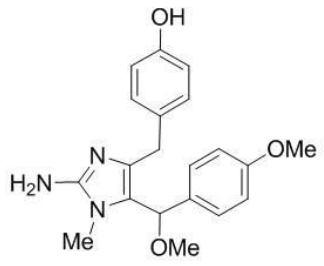
^{1}H and ^{13}C NMR Spectra of

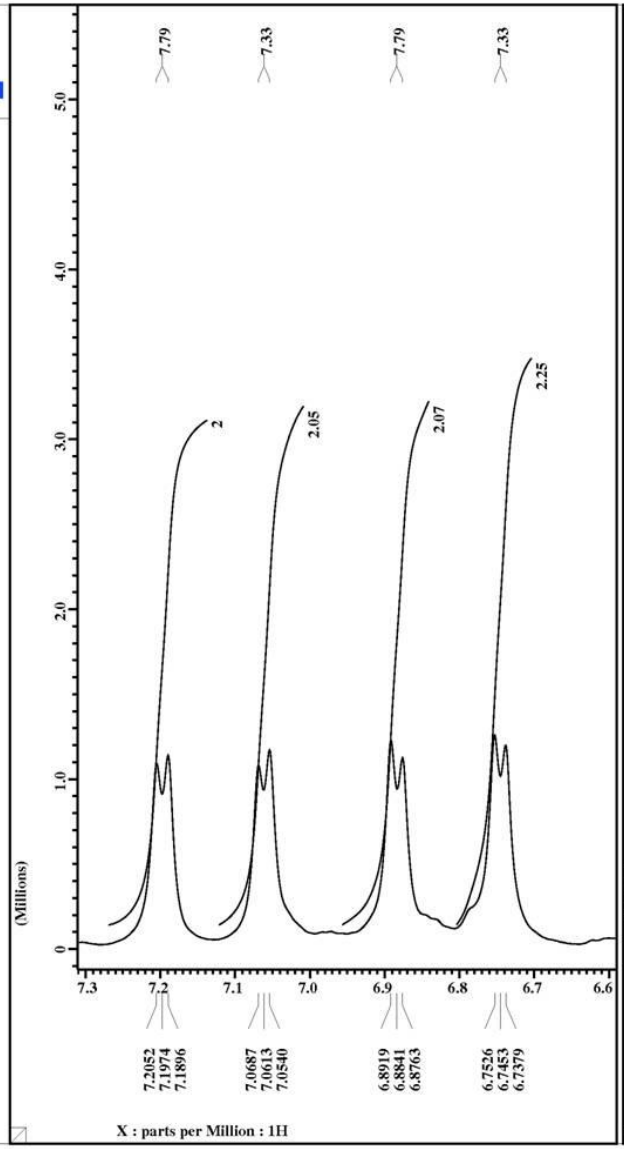
2-Amino-4-(4-hydroxybenzyl)-5-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**185**)

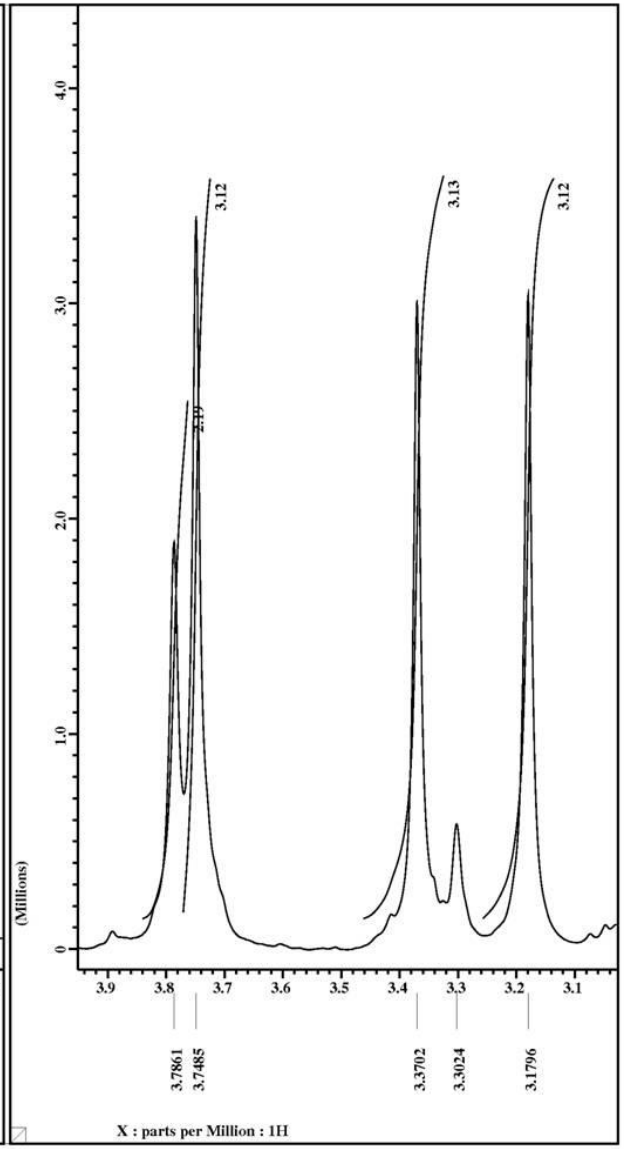


```
Filename          = IV_p_128_amine-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#298855
Solvent           = METHANOL-D3
Creation_time     = 18-JUL-2008 15:14:19
Revision_time     = 19-MAR-2010 00:41:21
Current_time      = 19-MAR-2010 00:41:45

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 13
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```

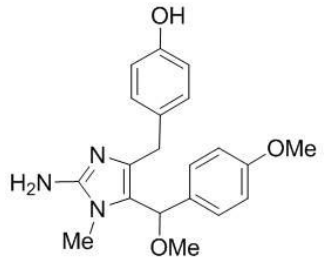
```
Filename          = IV_p_128_amine-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#298855
Solvent           = METHANOL-D3
Creation_time     = 18-JUL-2008 15:14:19
Revision_time     = 19-MAR-2010 00:42:13
Current_time      = 19-MAR-2010 00:42:22

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 13
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```

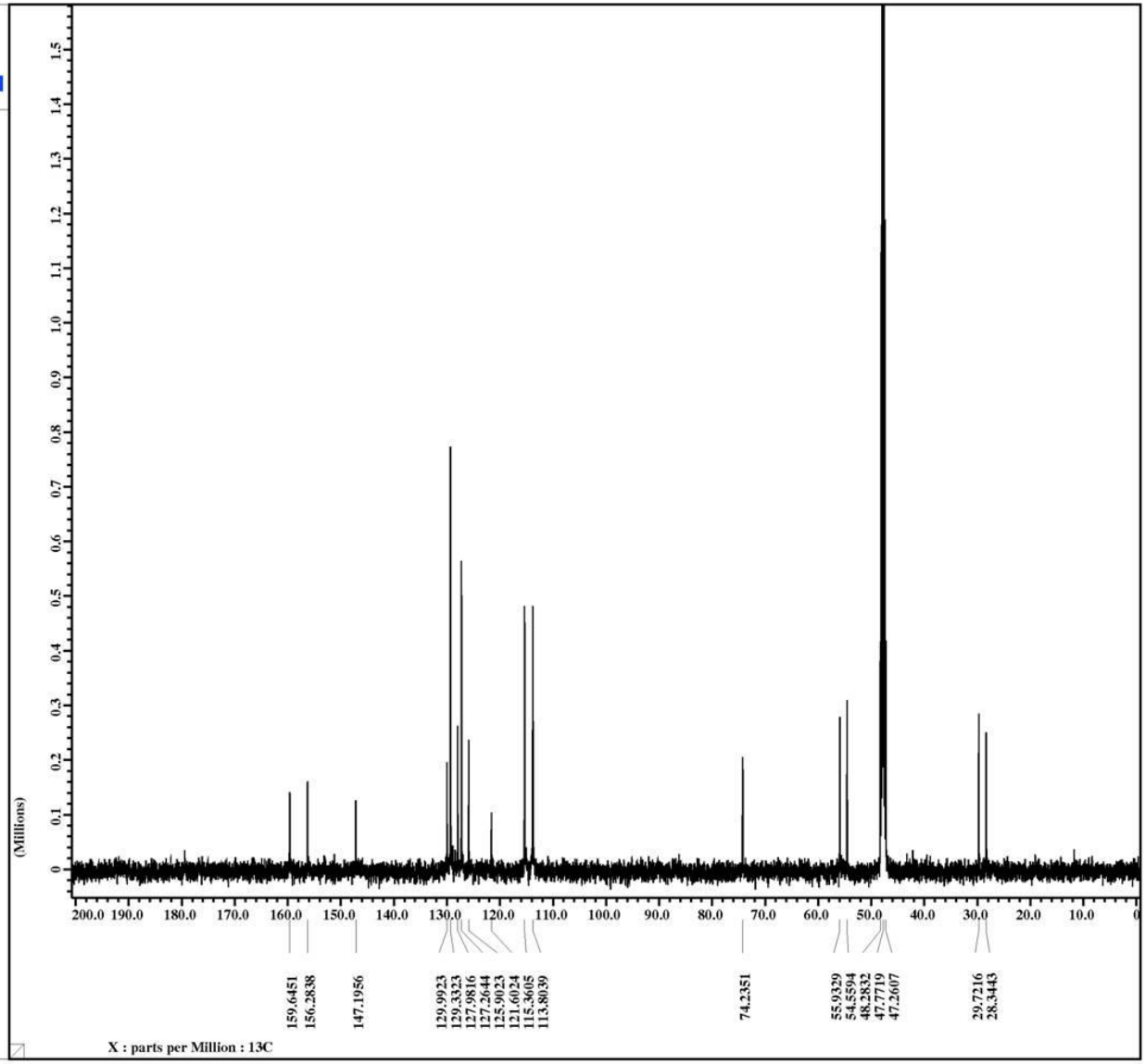






```
Filename          = IV_p_128_amine-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#299681
Solvent           = METHANOL-D3
Creation_time     = 18-JUL-2008 16:16:20
Revision_time     = 19-MAR-2010 00:44:07
Current_time      = 19-MAR-2010 00:44:26

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 707
Total_scans       = 707

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.9[dC]
Unblank_time      = 2[us]
```

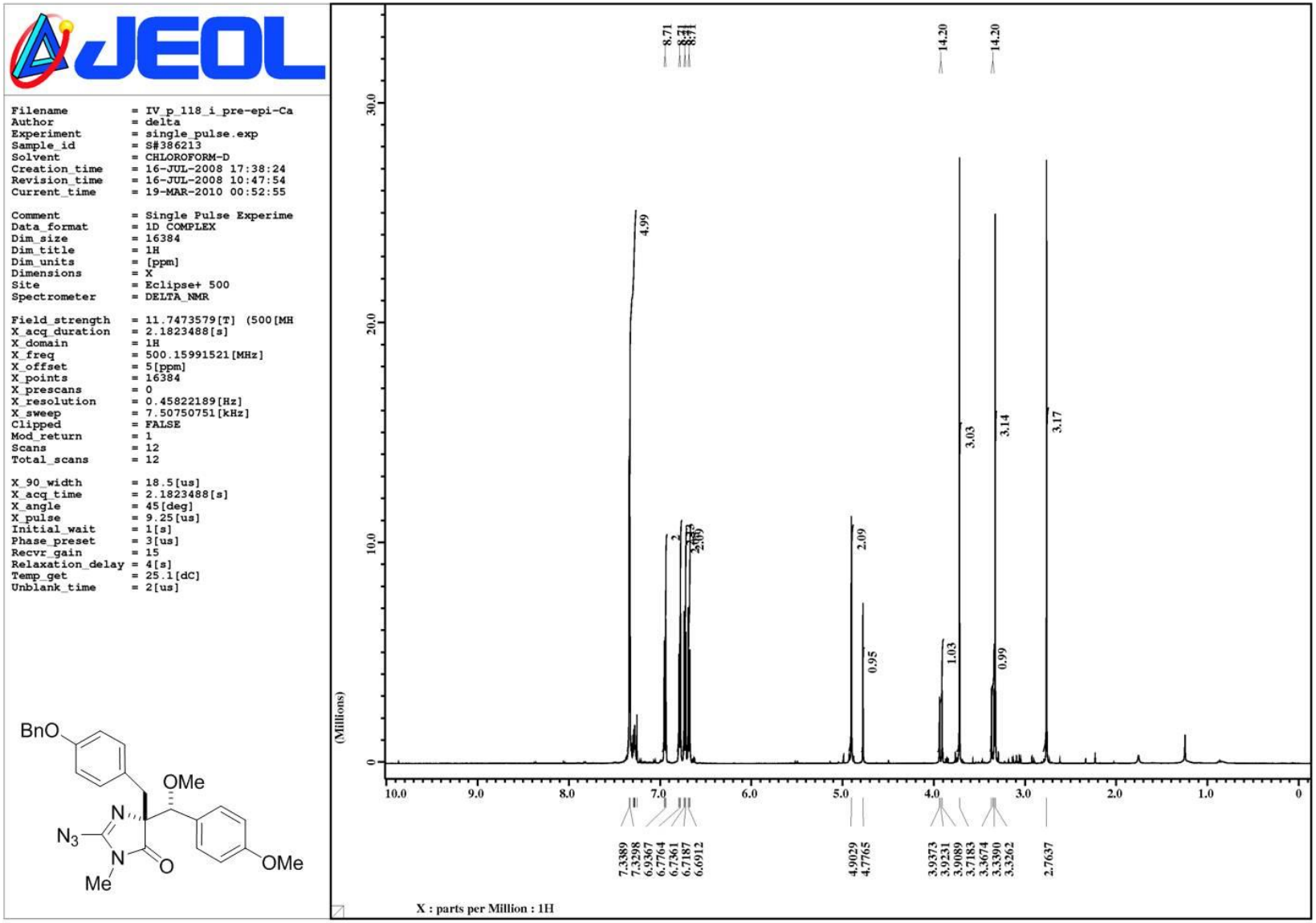




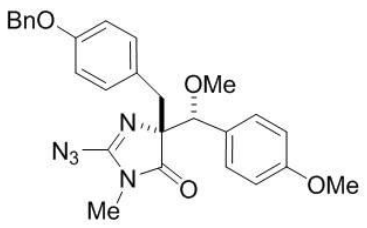
APPENDIX 65

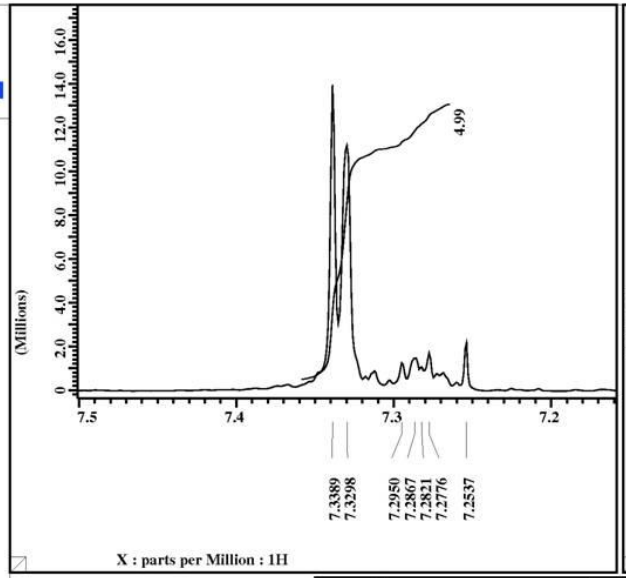
^{1}H and ^{13}C NMR Spectra of

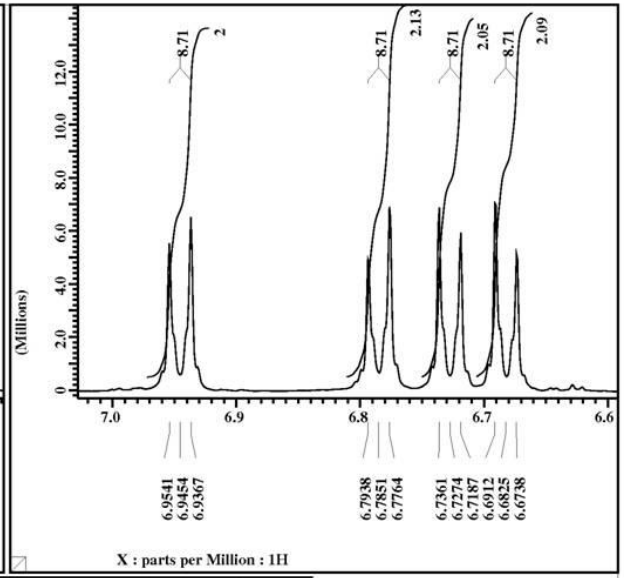
(*4R**, *8R**)-2-Azido-4-(4-benzyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1,5-dihydroimidazol-5-one (***epi*-186**)

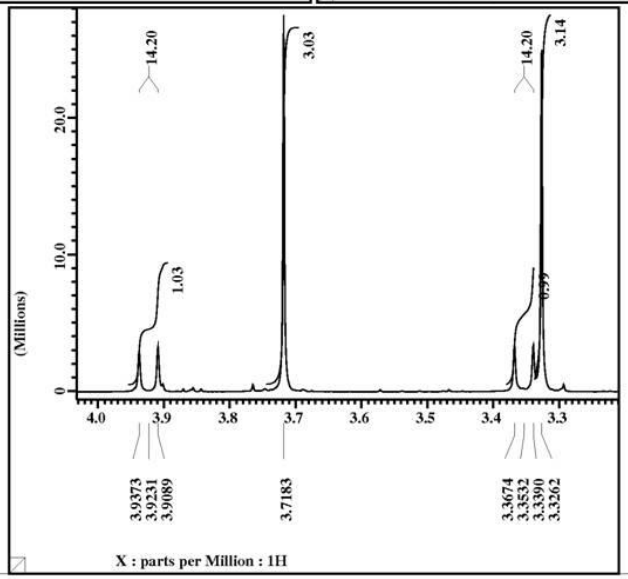

Filename = IV_p_118_i_pre-epi-Ca
Author = delta
Experiment = single_pulse.exp
Sample_id = S#386213
Solvent = CHLOROFORM-D
Creation_time = 16-JUL-2008 17:38:24
Revision_time = 16-JUL-2008 10:47:54
Current_time = 19-MAR-2010 00:52:55
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 15
Relaxation_delay = 4[s]
Temp_get = 25.1[dC]
Unblank_time = 2[us]
BnO
OMe
N
N3
N
O
Me
OMe
(Millions)
30.0
20.0
10.0
0
8.71
14.20
14.20
4.99
3.03
3.14
3.17
2.09
0.95
1.03
0.99
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
7.3389
7.3398
6.9367
6.7764
6.7361
6.7187
6.6912
4.9029
4.7765
3.9373
3.9231
3.9089
3.7183
3.3674
3.3390
3.3362
2.7637
X : parts per Million : 1H

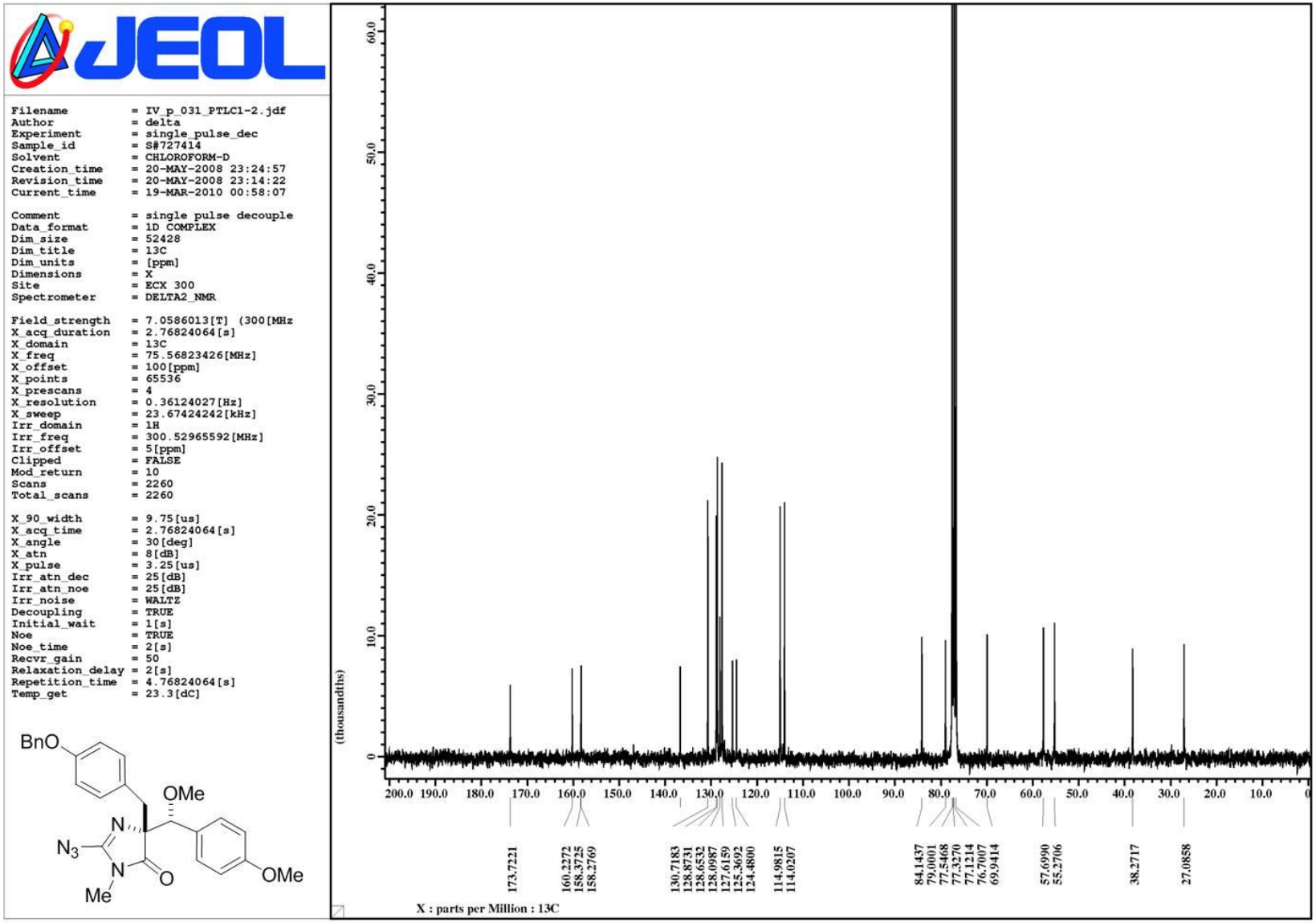
Filename	= IV_p_118_i_pre-epi-Ca
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#386213
Solvent	= CHLOROFORM-D
Creation_time	= 16-JUL-2008 17:38:24
Revision_time	= 16-JUL-2008 10:47:54
Current_time	= 19-MAR-2010 00:54:15
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 15
Relaxation_delay	= 4[s]
Temp_get	= 25.1[dC]
Unblank_time	= 2[us]





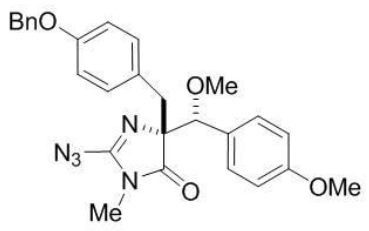


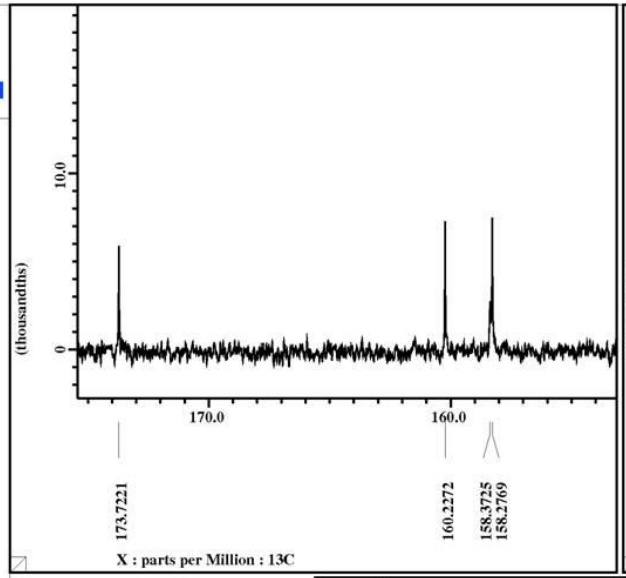


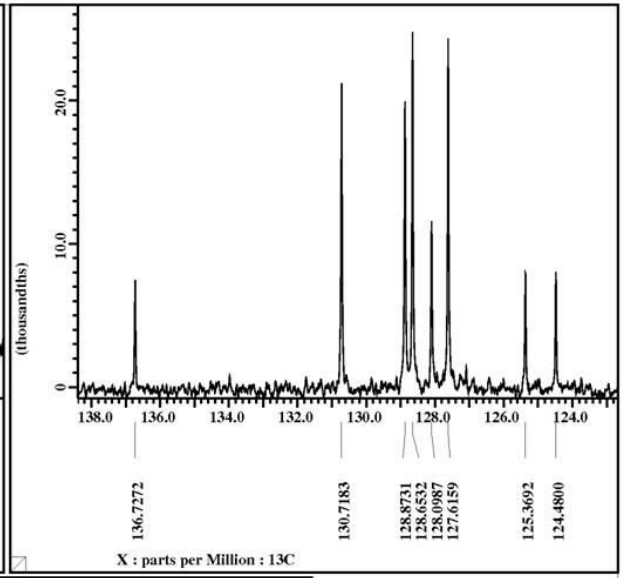


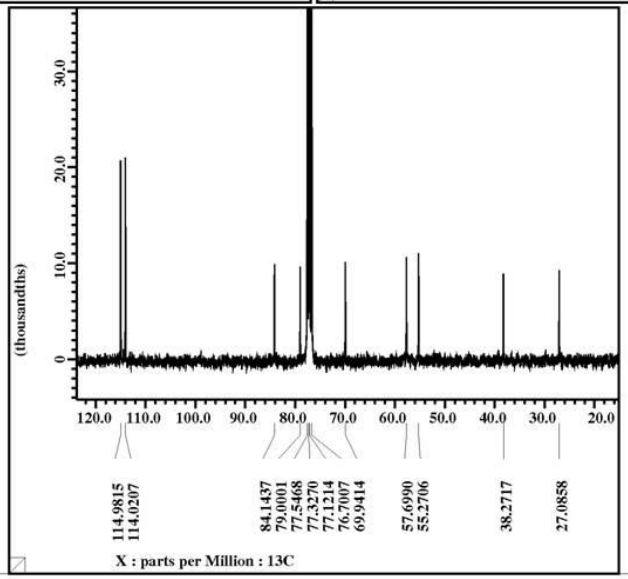
```
Filename          = IV_p_031_PTLC1-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#727414
Solvent           = CHLOROFORM-D
Creation_time     = 20-MAY-2008 23:24:57
Revision_time     = 20-MAY-2008 23:14:22
Current_time      = 19-MAR-2010 00:58:07

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 2260
Total_scans       = 2260

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```

```
Filename          = IV_p_031_PTLC1-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#727414
Solvent           = CHLOROFORM-D
Creation_time     = 20-MAY-2008 23:24:57
Revision_time     = 20-MAY-2008 23:14:22
Current_time      = 19-MAR-2010 00:57:37

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 2260
Total_scans       = 2260

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```







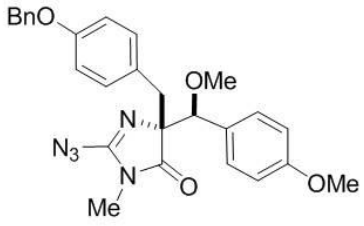


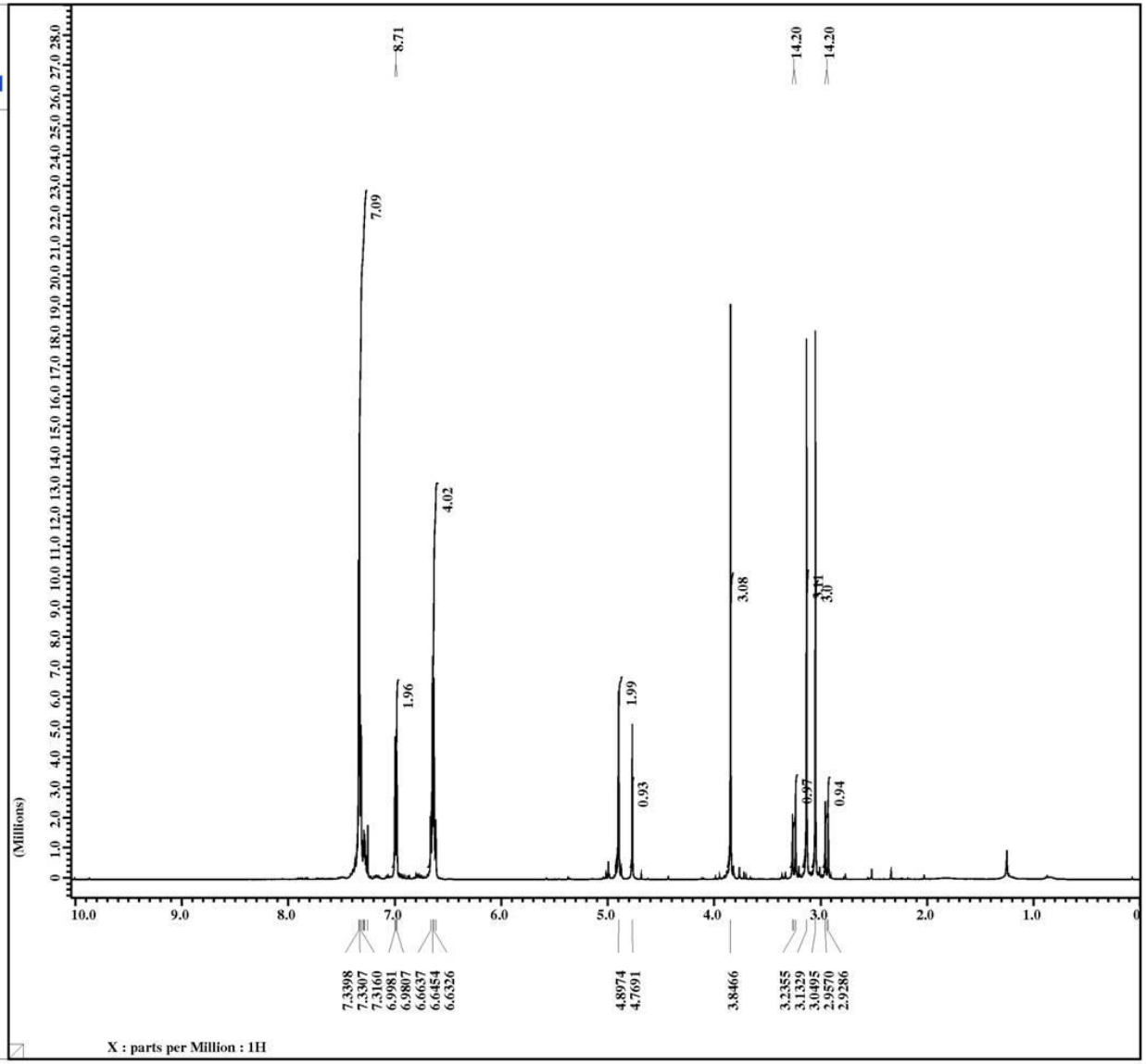
APPENDIX 66

^{1}H and ^{13}C NMR Spectra of

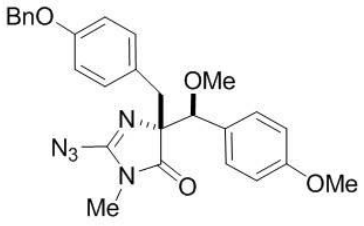
(*4R*, 8S**)-2-Azido-4-(4-benzyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1,5-dihydroimidazol-5-one (**186**)

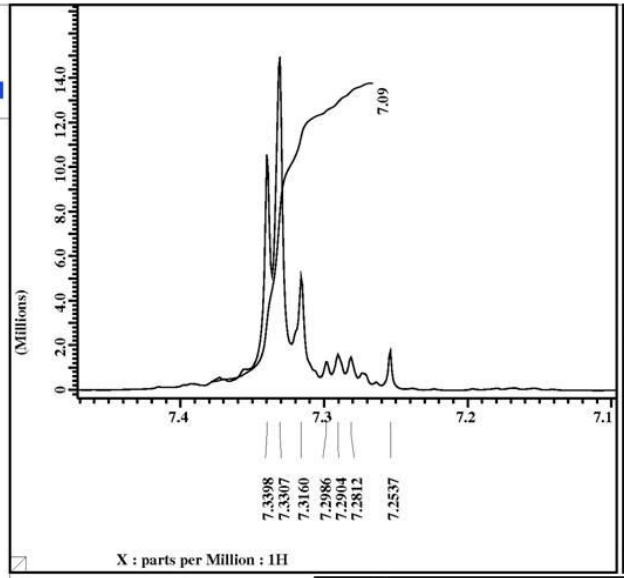
```
Filename          = IV_p_118_ii_pre-Calca
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#382103
Solvent           = CHLOROFORM-D
Creation_time     = 16-JUL-2008 17:31:35
Revision_time     = 19-MAR-2010 01:08:19
Current_time      = 19-MAR-2010 01:08:48

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 24.9[dC]
Unblank_time      = 2[us]
```

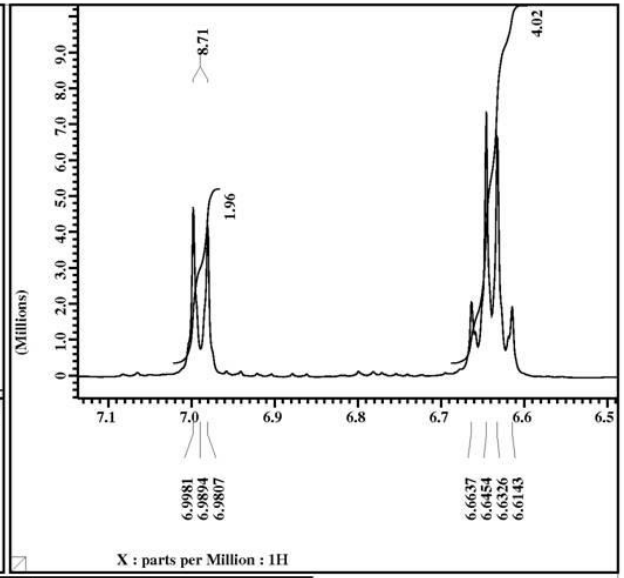


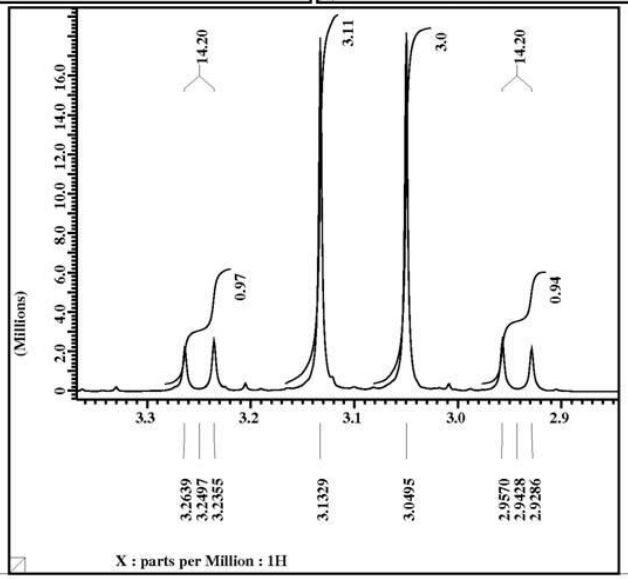


```
Filename          = IV_p_118_ii_pre-Calca
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#382103
Solvent           = CHLOROFORM-D
Creation_time     = 16-JUL-2008 17:31:35
Revision_time     = 19-MAR-2010 01:08:19
Current_time      = 19-MAR-2010 01:09:30

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 24.9[dC]
Unblank_time      = 2[us]
```

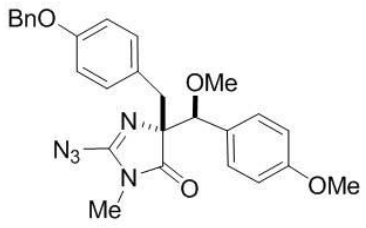


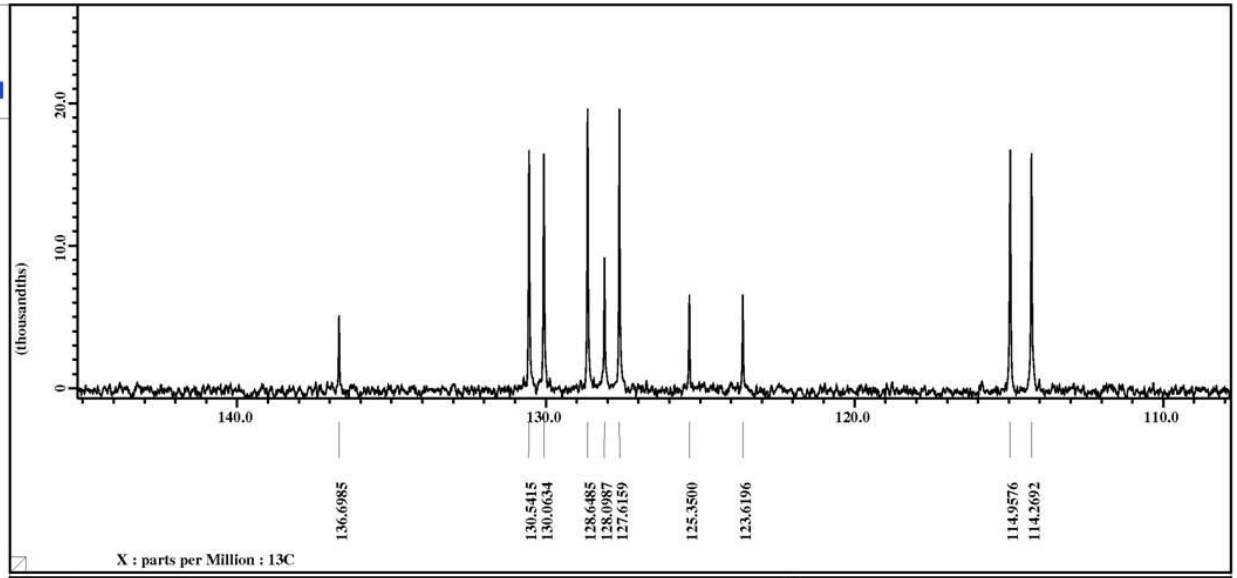


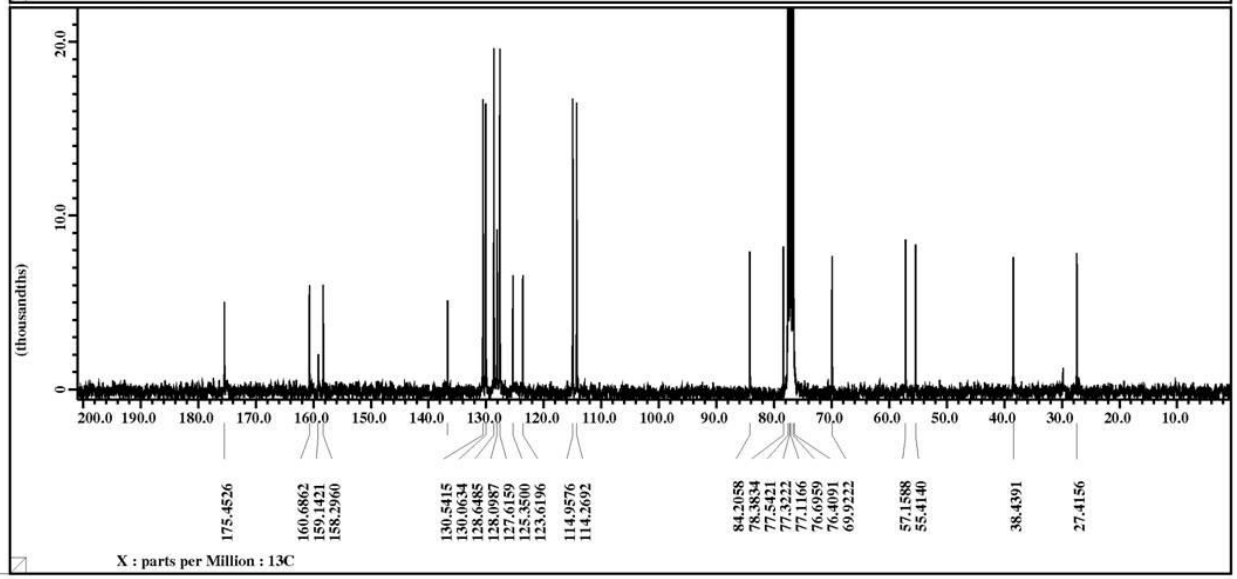




```
Filename          = IV_p_031_PTLC2-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 21-MAY-2008 04:47:56
Revision_time     = 21-MAY-2008 10:01:10
Current_time      = 19-MAR-2010 01:10:28

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 4000
Total_scans       = 4000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.1[dC]
```







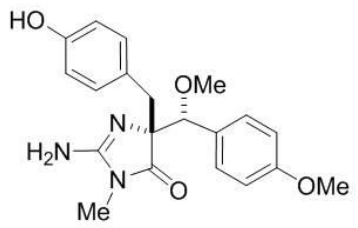
APPENDIX 67

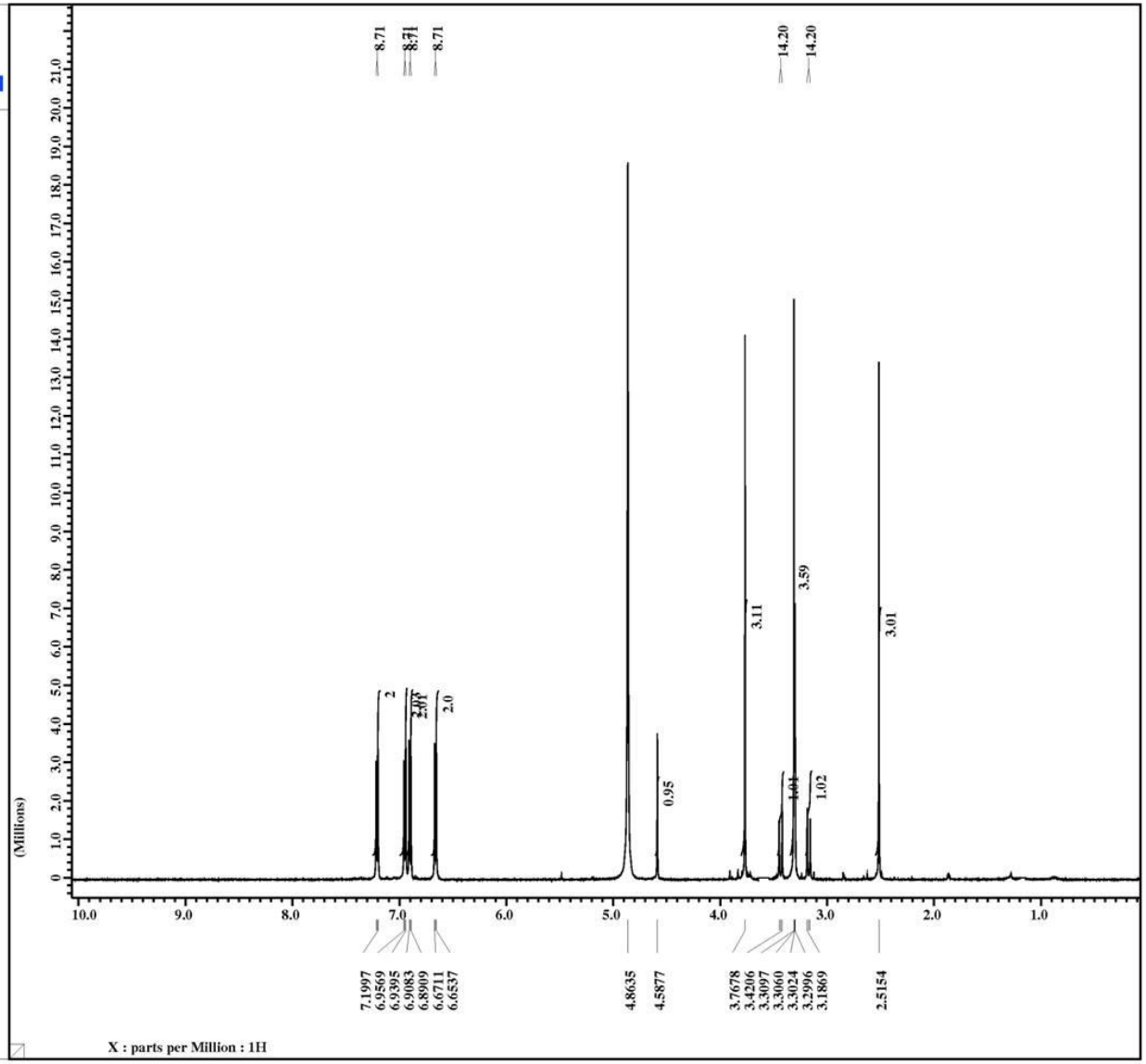
^{1}H and ^{13}C NMR Spectra of

(*4R**, *8R**)-2-Amino-4-(4-benzyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1,5-dihydroimidazol-5-one [***epi*-(13)**]:

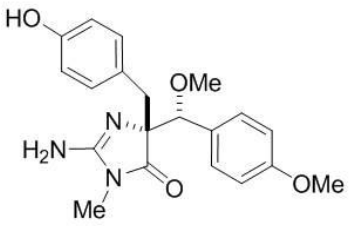
epi-Calcaridine A

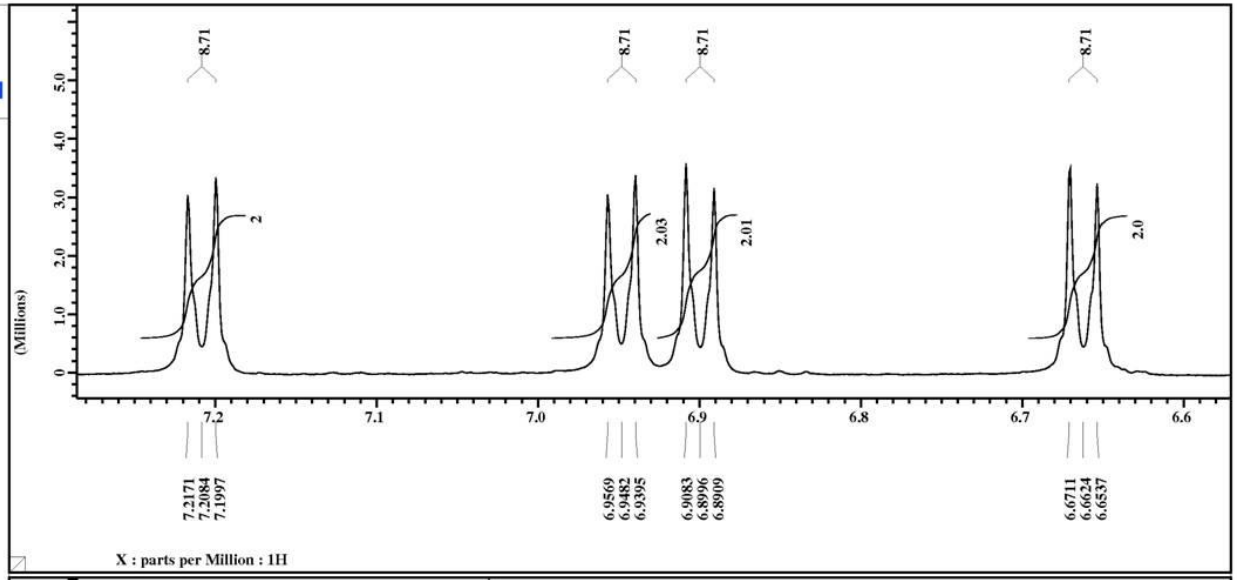
```
Filename          = IV_p_129_epi-Calcarid
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#607146
Solvent           = METHANOL-D3
Creation_time     = 21-JUL-2008 23:50:17
Revision_time     = 19-MAR-2010 01:24:52
Current_time      = 19-MAR-2010 01:25:26

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 19
Relaxation_delay  = 4[s]
Temp_get          = 24.8[dC]
Unblank_time      = 2[us]
```

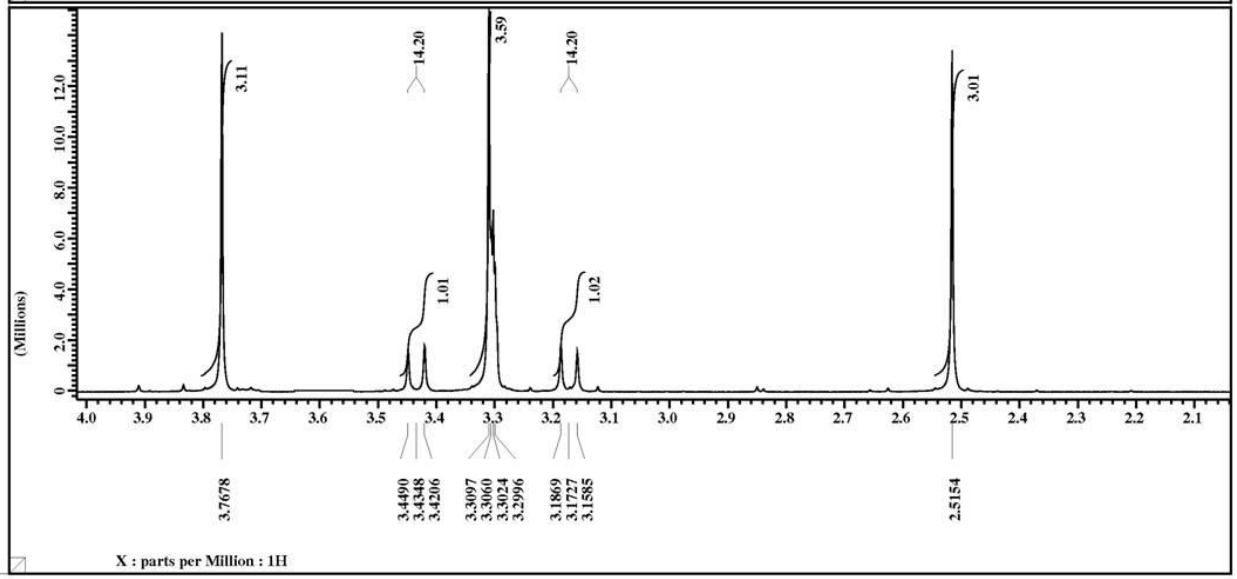




```
Filename          = IV_p_129_epi-Calcarid
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#607146
Solvent           = METHANOL-D3
Creation_time     = 21-JUL-2008 23:50:17
Revision_time     = 19-MAR-2010 01:24:52
Current_time      = 19-MAR-2010 01:26:00

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 19
Relaxation_delay  = 4[s]
Temp_get          = 24.8[dC]
Unblank_time      = 2[us]
```







JEOL
Filename = IV_p_129_epi-Calcarid
Author = delta
Experiment = single_pulse_dec
Sample_id = S#761824
Solvent = METHANOL-D3
Creation_time = 22-JUL-2008 14:00:39
Revision_time = 19-MAR-2010 01:26:28
Current_time = 19-MAR-2010 01:28:24
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.74735791[T] (500[MH
X_acq_duration = 2.08404481[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 7000
Total_scans = 7000
X_90_width = 14.2[us]
X_acq_time = 2.08404481[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 27.1[dC]
Unblank_time = 2[us]
(Thousands)
X : parts per Million : 13C
172.2356
160.4997
157.8366
156.8332
130.8851
129.0347
126.2037
124.0518
114.8836
113.4757
84.0672
73.5521
48.0238
47.8521
47.6842
47.5125
47.3408
47.1729
38.2680
24.1131
HO
OMe
OMe
N
O
N
Me
H_2N

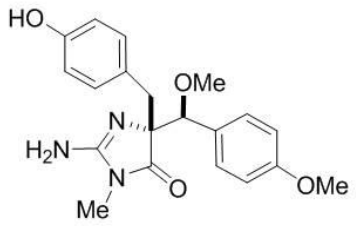
APPENDIX 68

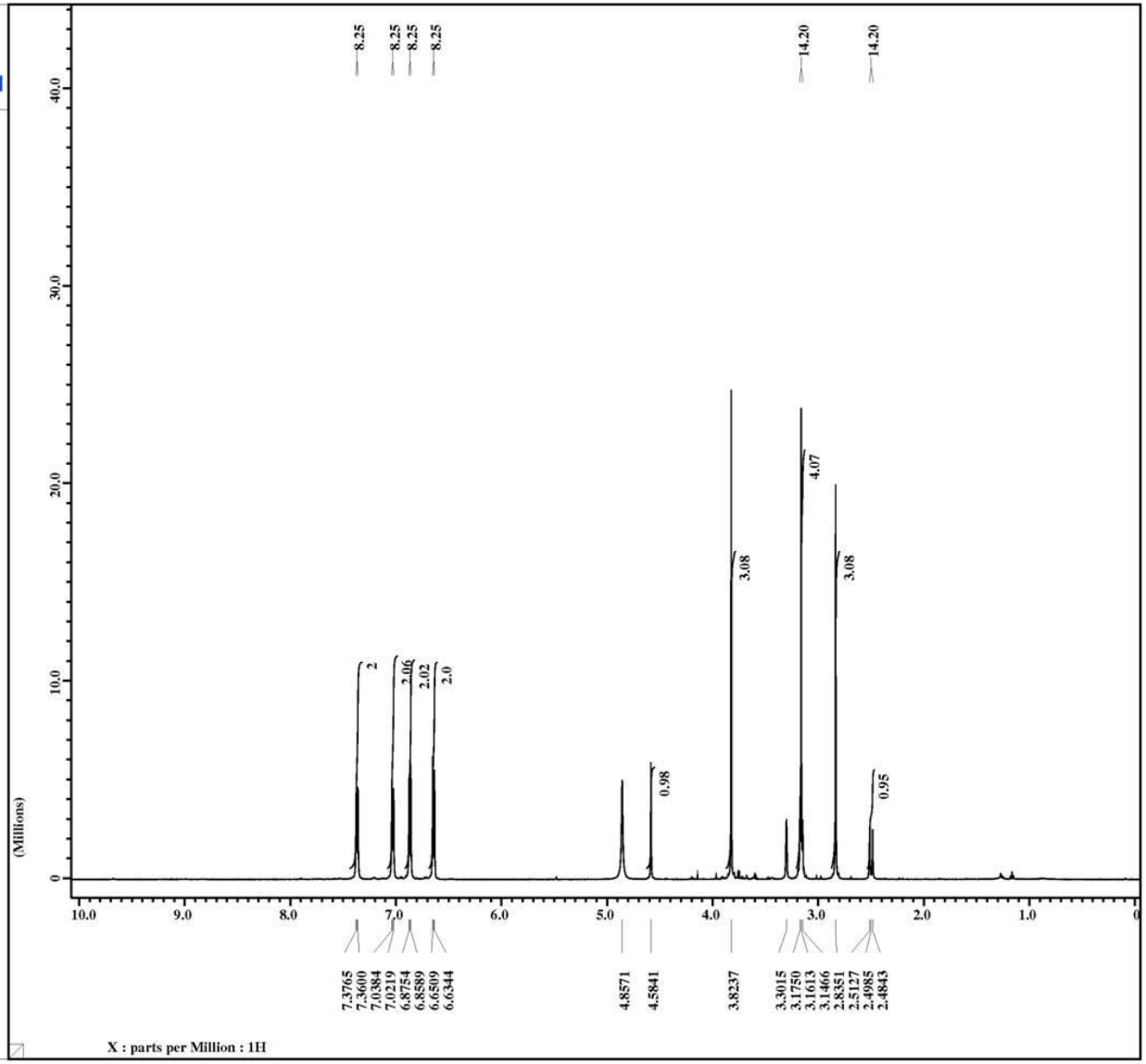
^{1}H and ^{13}C NMR Spectra of

(*4R*, 8S**)-2-Amino-4-(4-benzyloxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1,5-dihydroimidazol-5-one (**13**):

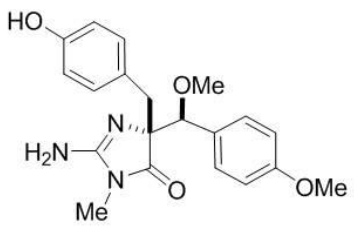
Calcaridine A

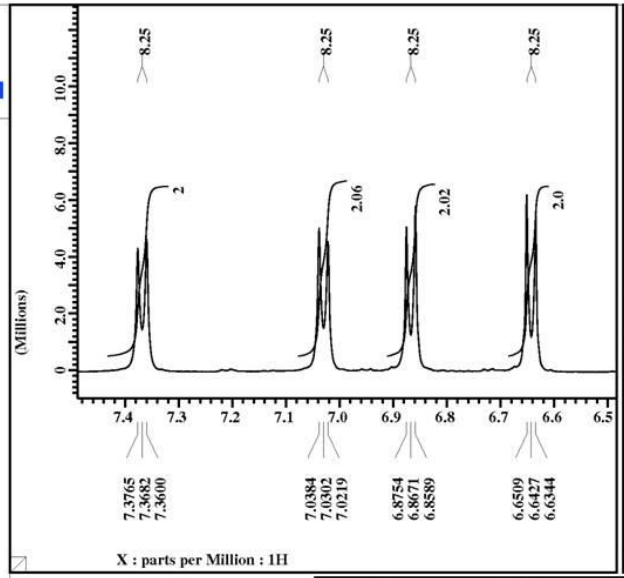
```
Filename          = IV_p_126_Calcaridine
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#807030
Solvent           = METHANOL-D3
Creation_time     = 18-JUL-2008 05:20:52
Revision_time     = 17-JUL-2008 22:29:05
Current_time      = 19-MAR-2010 01:36:54

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 17
Relaxation_delay  = 4[s]
Temp_get          = 24.9[dC]
Unblank_time      = 2[us]
```

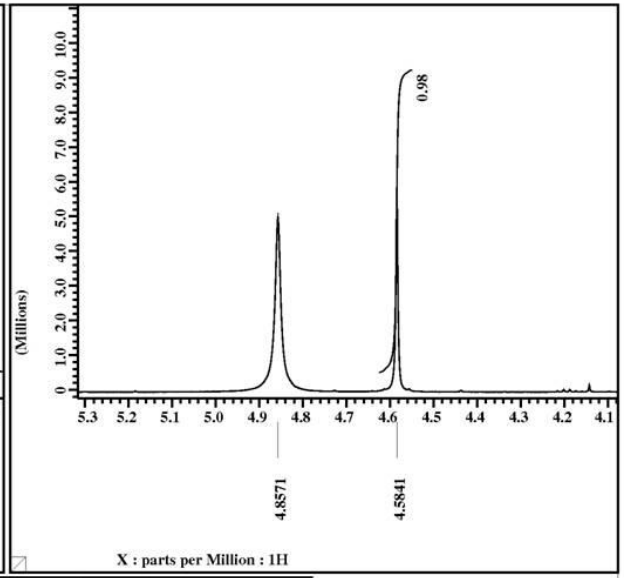


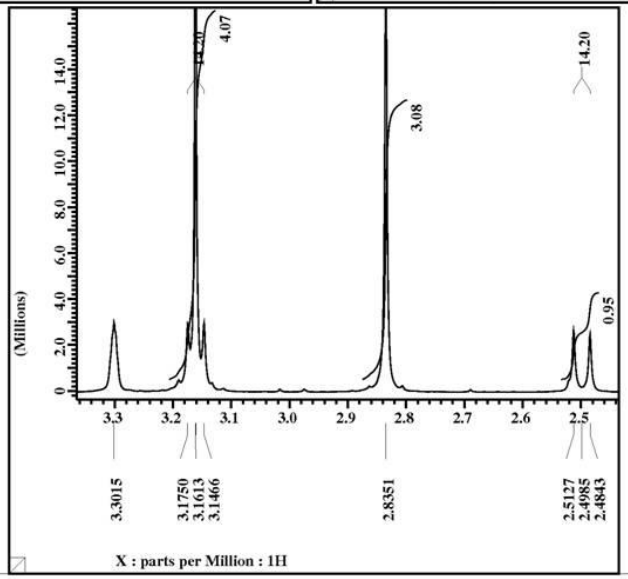


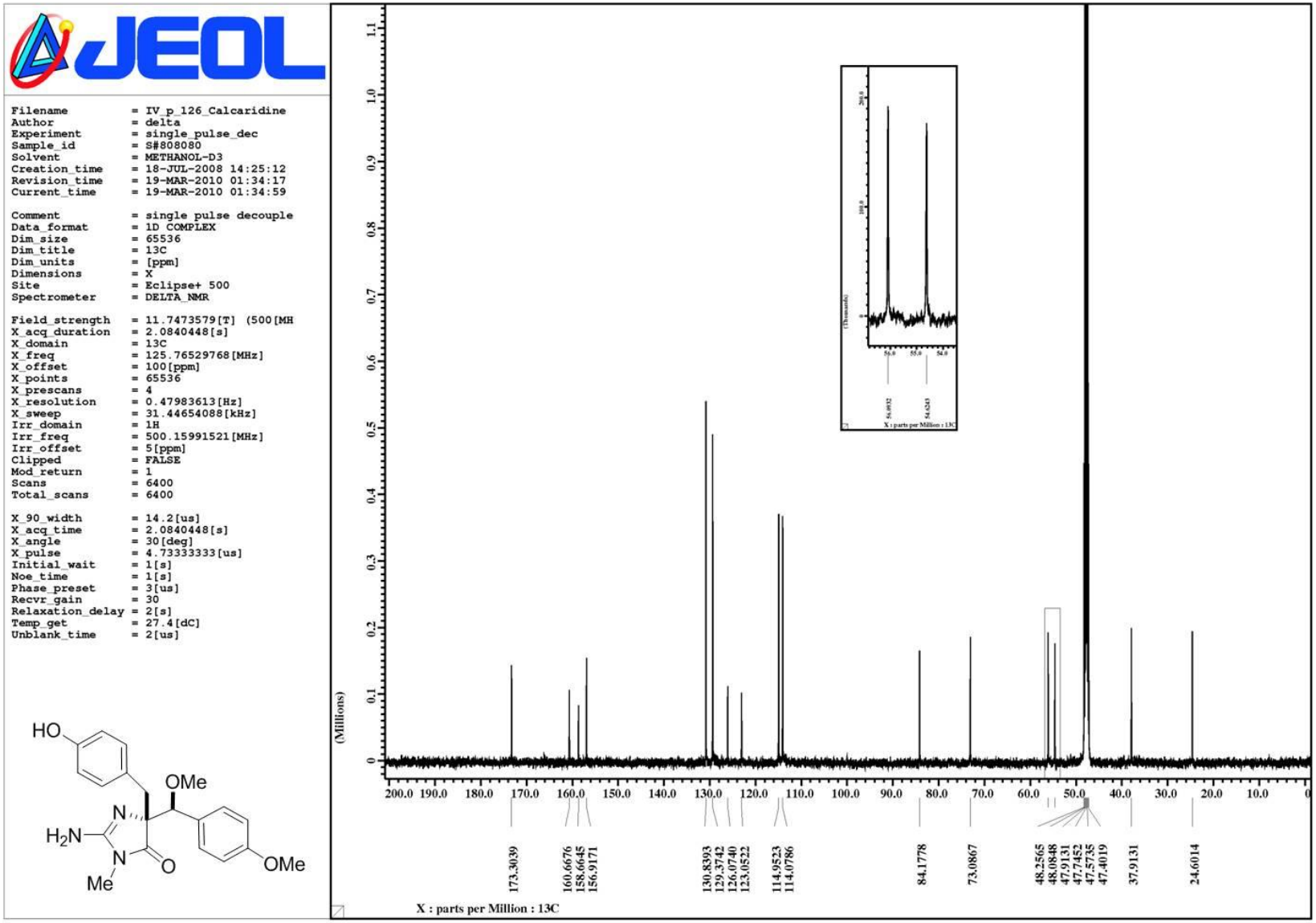
Filename	= IV_p_126_Calcaridine
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#807030
Solvent	= METHANOL-D3
Creation_time	= 18-JUL-2008 05:20:52
Revision_time	= 17-JUL-2008 22:29:05
Current_time	= 19-MAR-2010 01:37:51
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 17
Relaxation_delay	= 4[s]
Temp_get	= 24.9[dC]
Unblank_time	= 2[us]











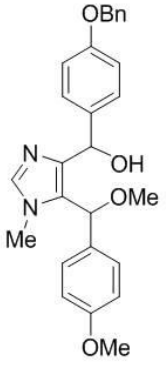
```
Filename          = IV_p_126_Calcaridine
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#808080
Solvent           = METHANOL-D3
Creation_time     = 18-JUL-2008 14:25:12
Revision_time     = 19-MAR-2010 01:34:17
Current_time      = 19-MAR-2010 01:34:59

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 6400
Total_scans       = 6400

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.4[dC]
Unblank_time      = 2[us]
```

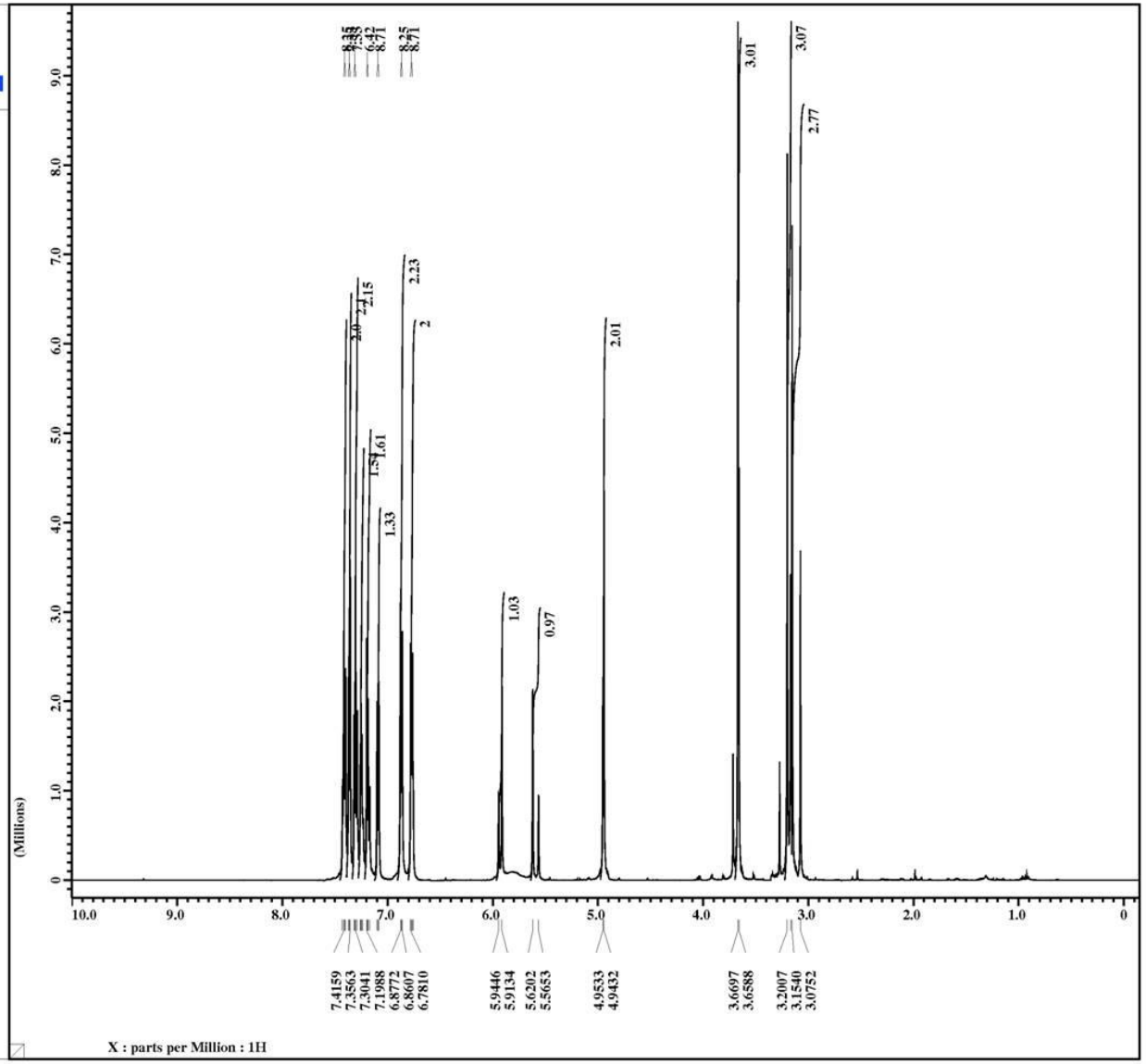
APPENDIX 69

^{1}H and ^{13}C NMR Spectra of

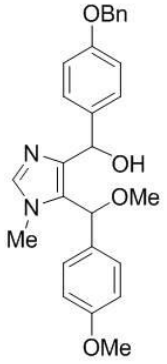
4-(4-Benzyloxyphenyl)hydroxymethyl-5-[methoxy-(4-methoxy)phenyl]methyl-1-methyl-1*H*-imidazole (**187**)

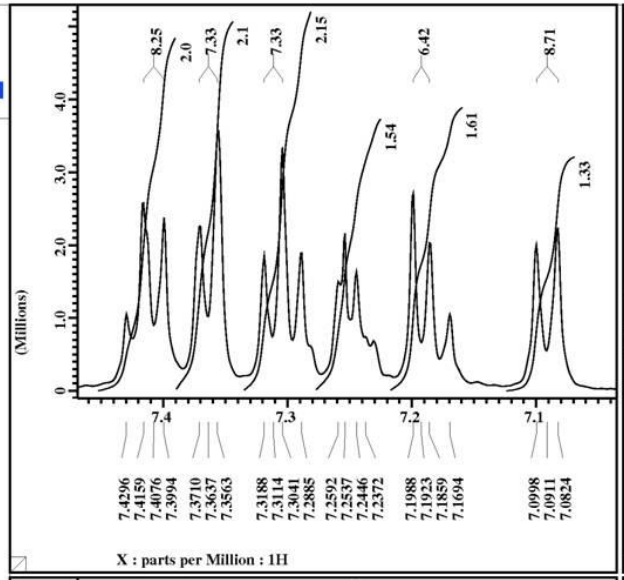
```
Filename          = IV_p_287_diBn-diol-de
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#460588
Solvent           = CHLOROFORM-D
Creation_time     = 24-OCT-2008 20:21:30
Revision_time     = 19-MAR-2010 01:48:26
Current_time      = 19-MAR-2010 01:48:42

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 8
Relaxation_delay  = 4[s]
Temp_get          = 26.1[dC]
Unblank_time      = 2[us]
```

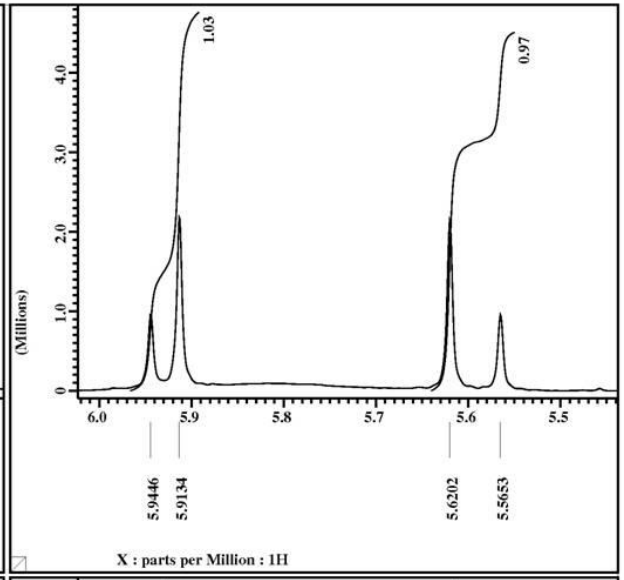


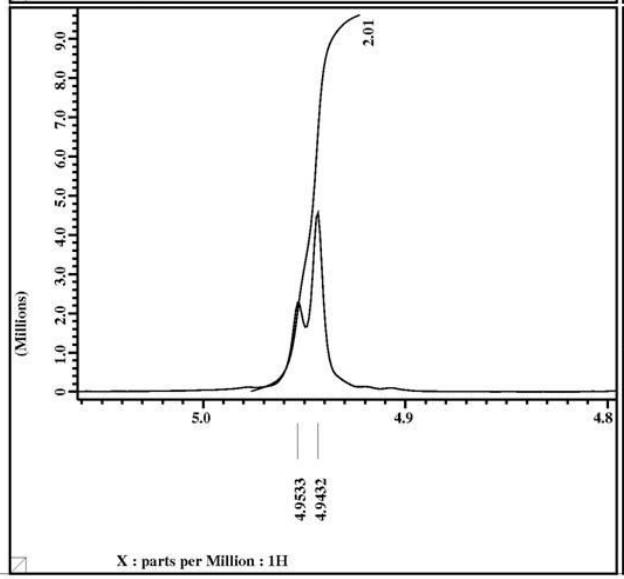


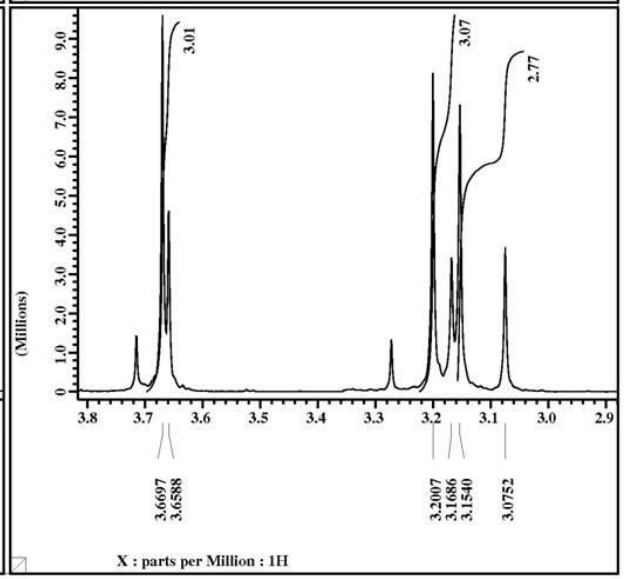
Filename	= IV_p_287_diBn-diol-de
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#460588
Solvent	= CHLOROFORM-D
Creation_time	= 24-OCT-2008 20:21:30
Revision_time	= 19-MAR-2010 01:48:26
Current_time	= 19-MAR-2010 01:49:28
Comment	= Single Pulse Experime
Data_format	= 1D REAL
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 8
Relaxation_delay	= 4[s]
Temp_get	= 26.1[dC]
Unblank_time	= 2[us]

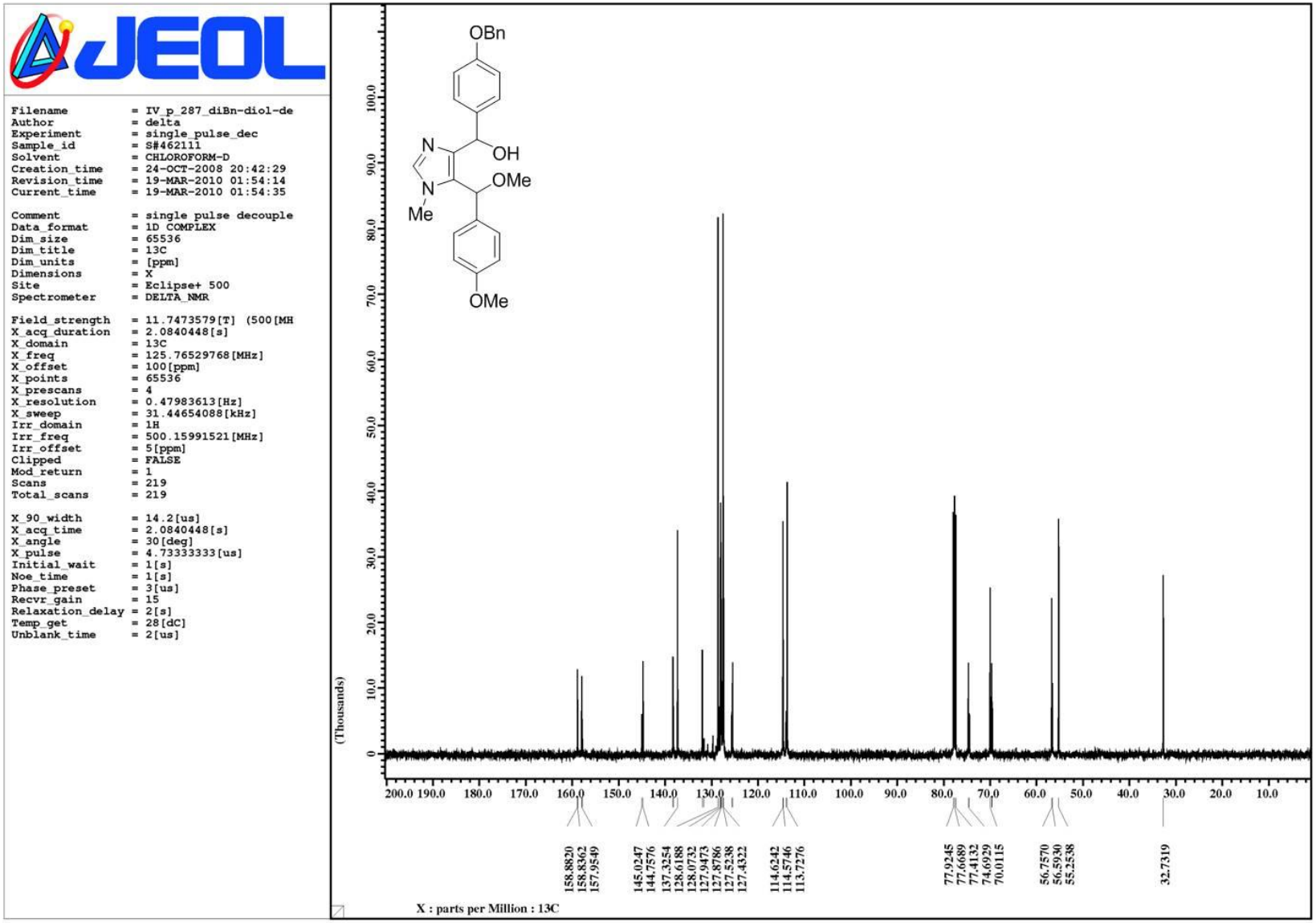




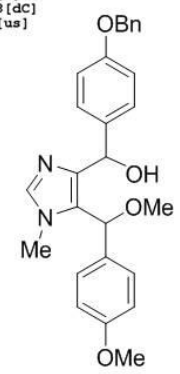


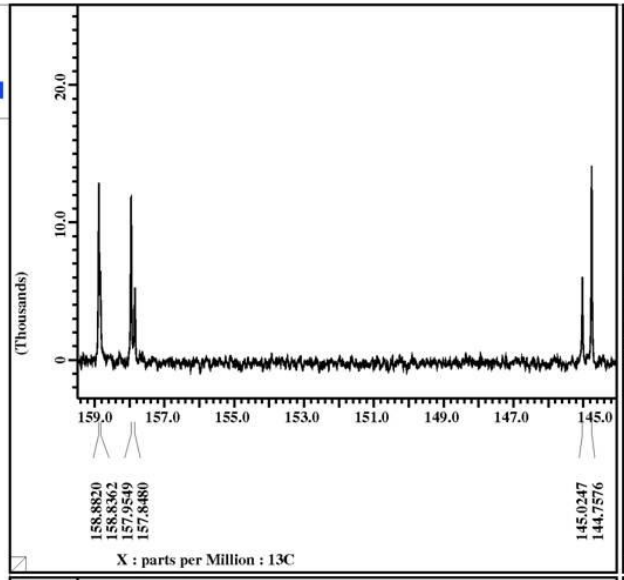


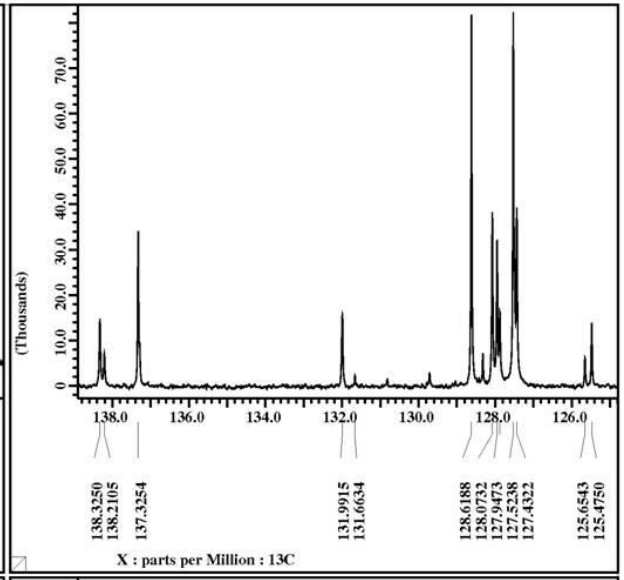


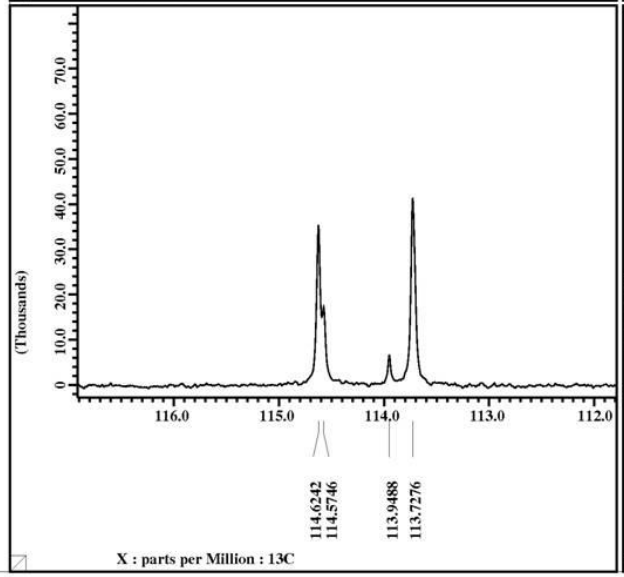

Filename = IV_p_287_diBn-diol-de
Author = delta
Experiment = single_pulse_dec
Sample_id = S#462111
Solvent = CHLOROFORM-D
Creation_time = 24-OCT-2008 20:42:29
Revision_time = 19-MAR-2010 01:54:14
Current_time = 19-MAR-2010 01:54:35
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 219
Total_scans = 219
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 15
Relaxation_delay = 2[s]
Temp_get = 28[dC]
Unblank_time = 2[us]
OBn
OH
OMe
N
N
Me
OMe
(Thousands)
158.8820
158.8362
157.9549
145.0247
144.7576
137.3254
128.6188
128.0732
127.9473
127.8786
127.5238
127.4322
114.6242
114.5746
113.7276
77.9245
77.6689
77.4132
74.6929
70.0115
56.7570
56.5930
55.2538
32.7319
X : parts per Million : 13C

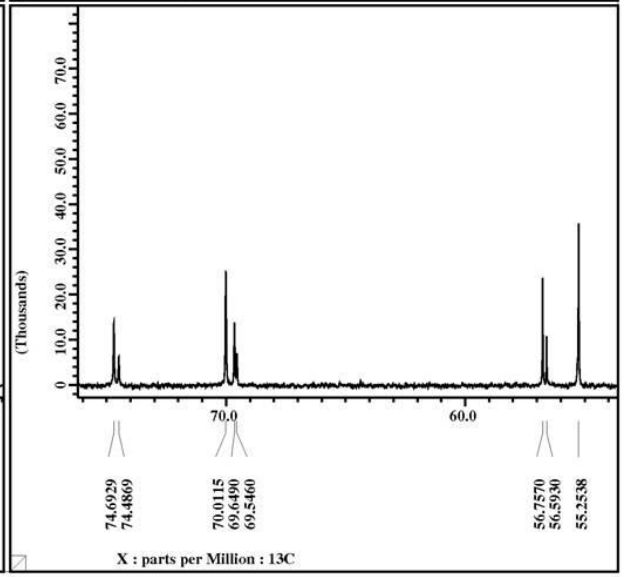
```
Filename          = IV_p_287_diBn-diol-de
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#462111
Solvent           = CHLOROFORM-D
Creation_time     = 24-OCT-2008 20:42:29
Revision_time     = 19-MAR-2010 01:55:24
Current_time      = 19-MAR-2010 01:55:52

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 219
Total_scans       = 219

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 2[s]
Temp_get          = 28[dC]
Unblank_time      = 2[us]
```











APPENDIX 70

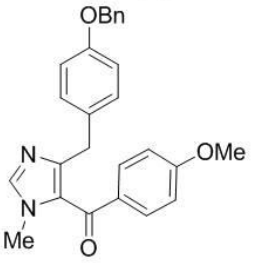
^{1}H and ^{13}C NMR Spectra of

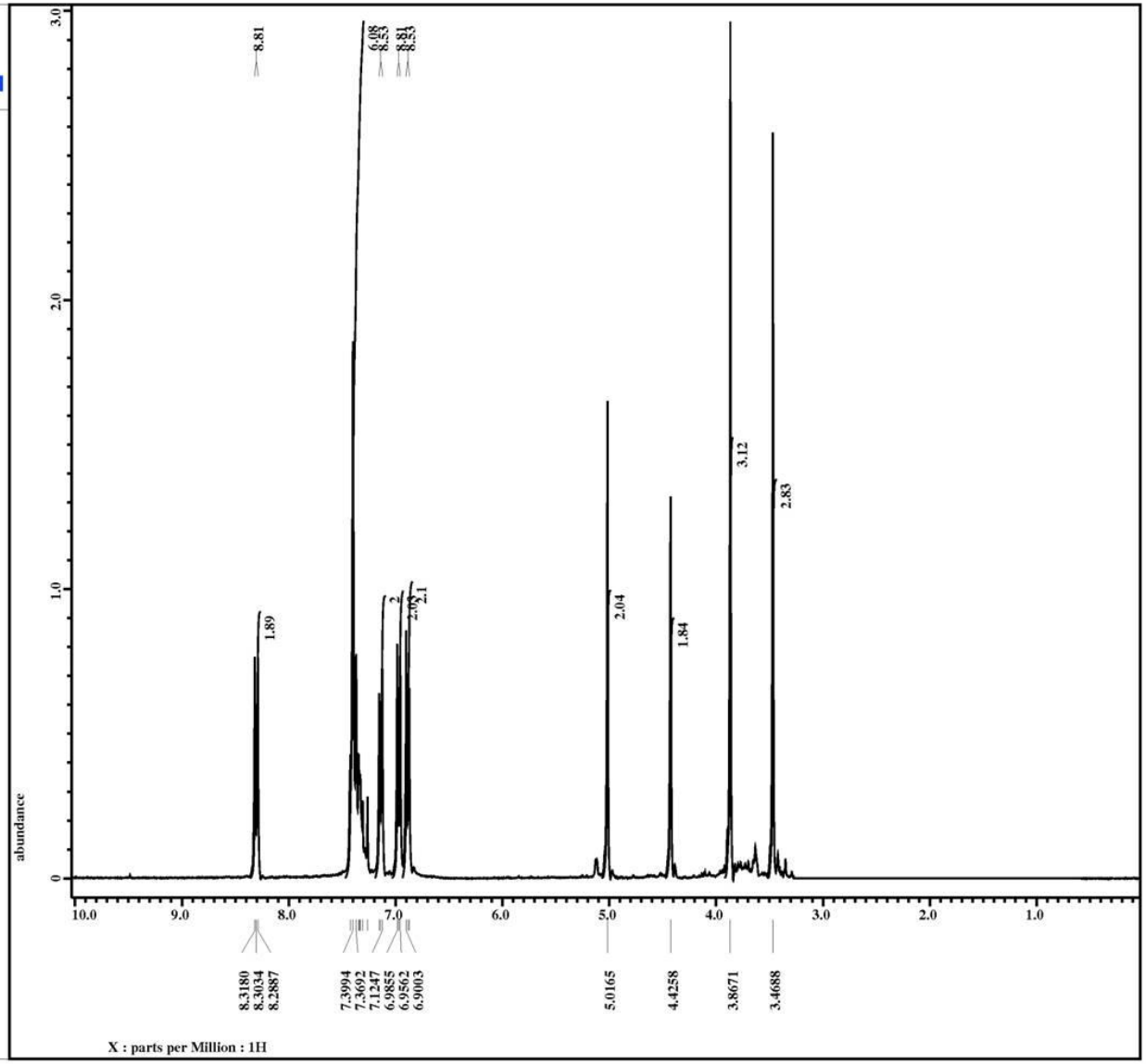
[4-(4-Benzyloxybenzyl)-1-methyl-1*H*-imidazol-5-yl]-(4-methoxyphenyl)-methanone

(**188**)

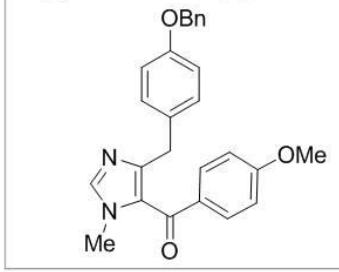
JEOL

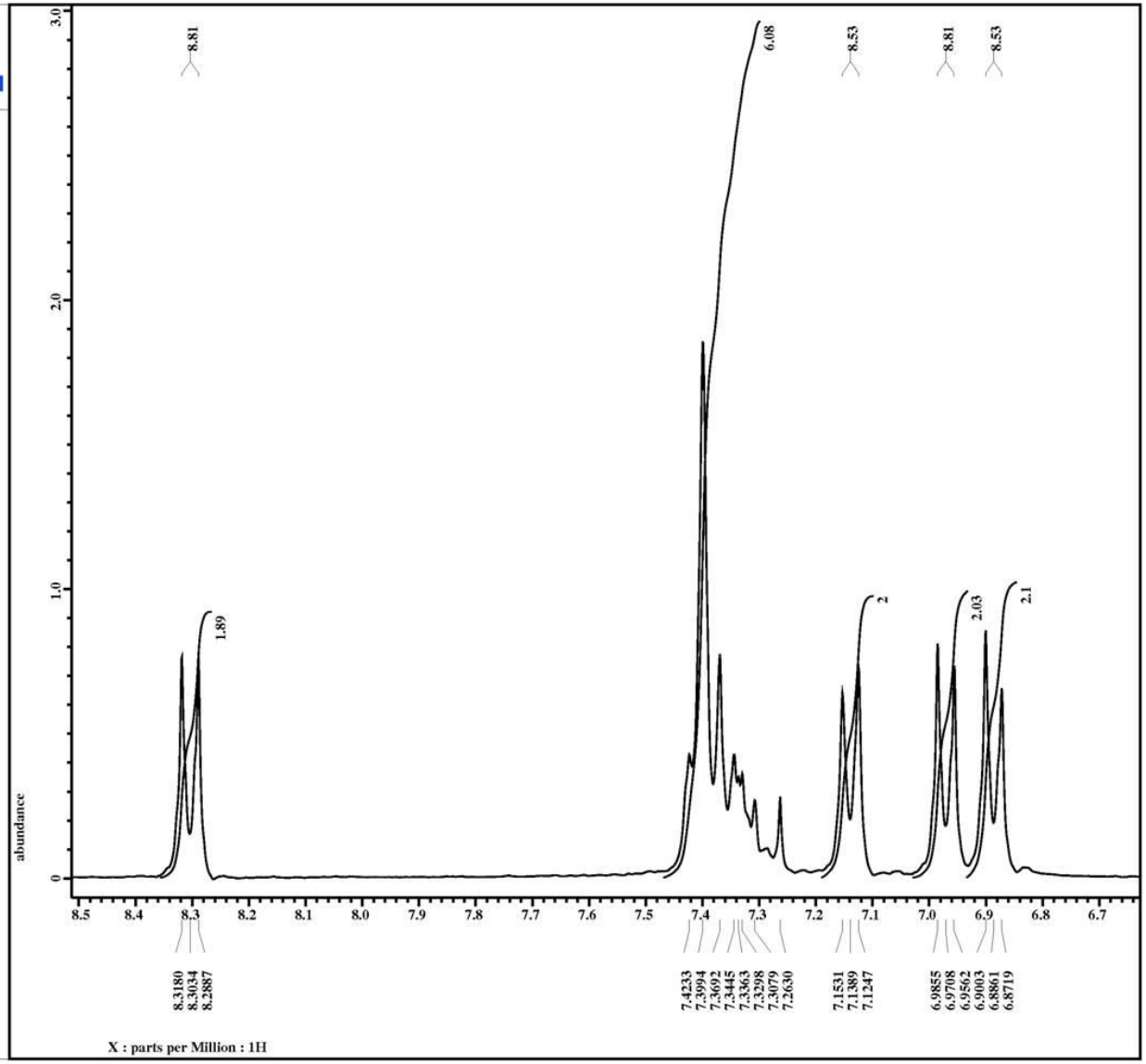
Filename	= V_p_017_i-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#803291
Solvent	= CHLOROFORM-D
Creation_time	= 19-NOV-2008 22:33:44
Revision_time	= 13-MAR-2010 20:42:59
Current_time	= 13-MAR-2010 20:43:24
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 38
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 22.9[dC]

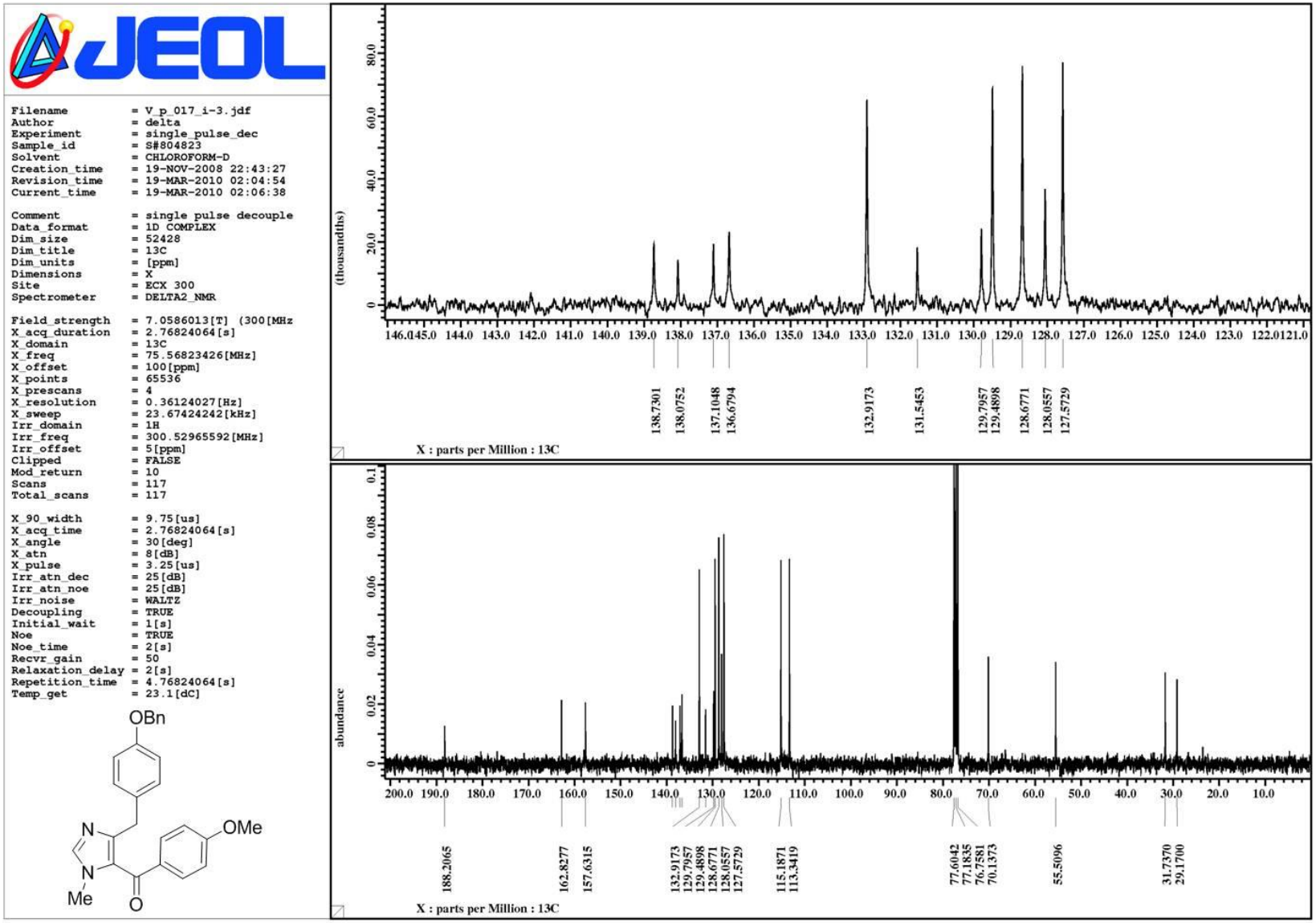




```
Filename          = V_p_017_i-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#803291
Solvent           = CHLOROFORM-D
Creation_time     = 19-NOV-2008 22:33:44
Revision_time     = 13-MAR-2010 20:42:59
Current_time      = 13-MAR-2010 20:43:44

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 38
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 22.9[dC]
```





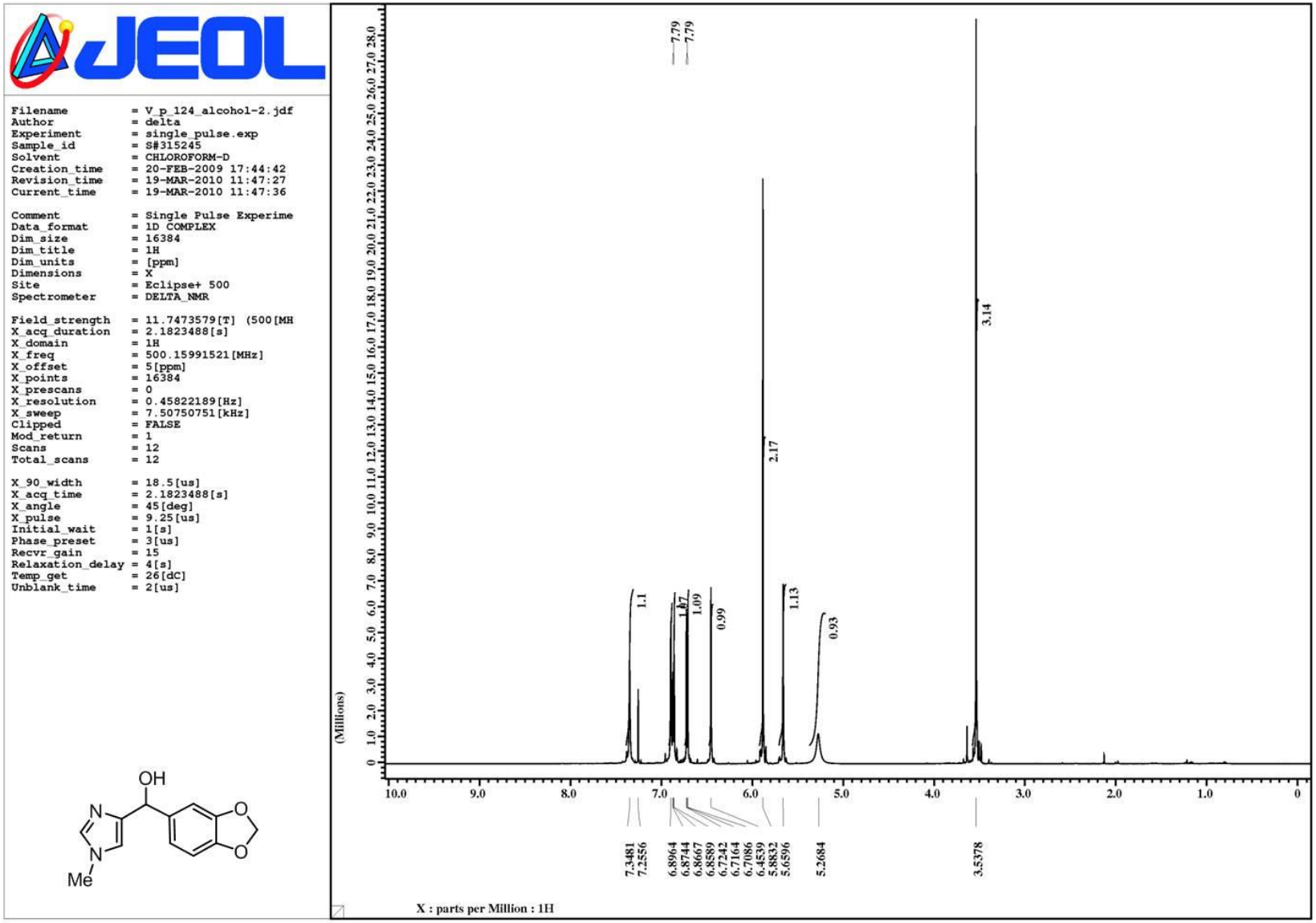


```
Filename          = V_p_017_i-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#804823
Solvent           = CHLOROFORM-D
Creation_time     = 19-NOV-2008 22:43:27
Revision_time     = 19-MAR-2010 02:04:54
Current_time      = 19-MAR-2010 02:06:38

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 117
Total_scans       = 117

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.1[dC]
```

APPENDIX 71

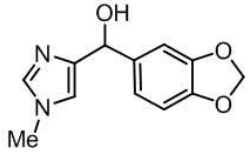
^{1}H and ^{13}C NMR Spectra of

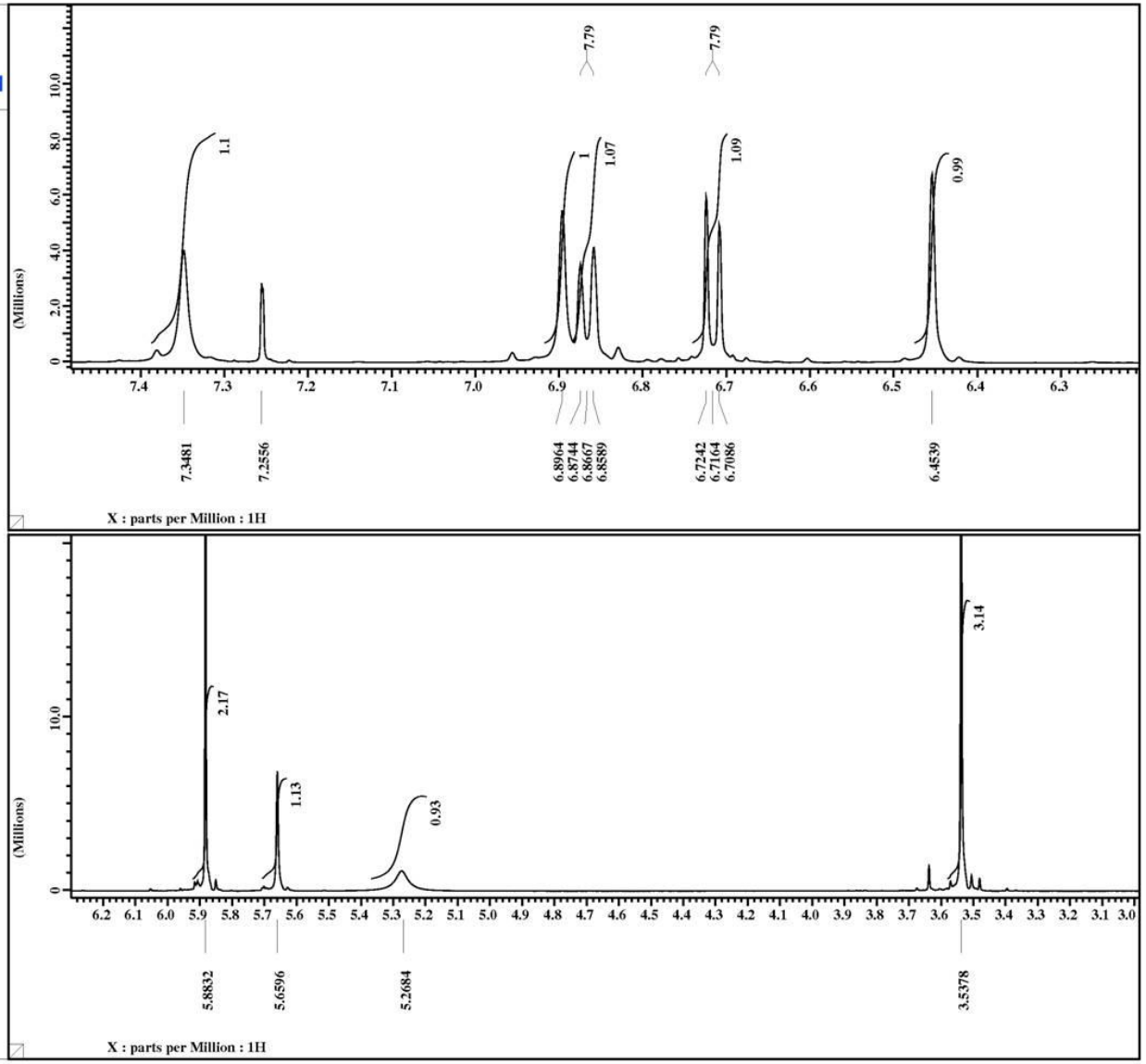
4-(Benzo[1,3]dioxol-5-yl)hydroxymethyl-1-methyl-1*H*-imidazole (**192**)

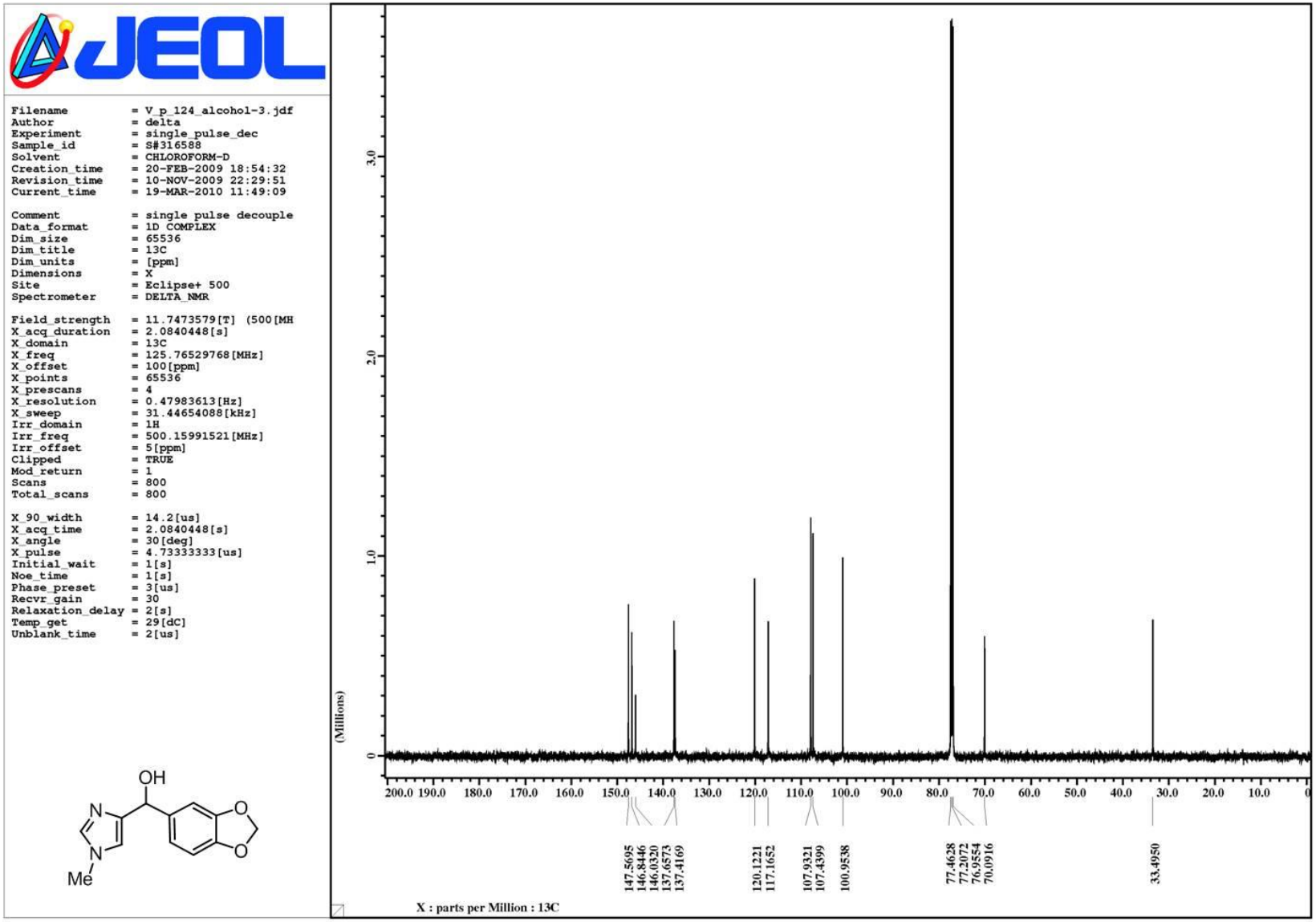


```
Filename          = V_p_124_alcohol-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#315245
Solvent           = CHLOROFORM-D
Creation_time     = 20-FEB-2009 17:44:42
Revision_time     = 19-MAR-2010 11:47:27
Current_time      = 19-MAR-2010 11:47:36

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 26[dC]
Unblank_time      = 2[us]
```

```
Filename          = V_p_124_alcohol-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#315245
Solvent           = CHLOROFORM-D
Creation_time     = 20-FEB-2009 17:44:42
Revision_time     = 19-MAR-2010 11:47:27
Current_time      = 19-MAR-2010 11:48:17

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 26[dC]
Unblank_time      = 2[us]
```







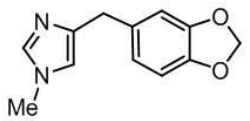
```
Filename          = V_p_124_alcohol-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#316588
Solvent           = CHLOROFORM-D
Creation_time     = 20-FEB-2009 18:54:32
Revision_time     = 10-NOV-2009 22:29:51
Current_time      = 19-MAR-2010 11:49:09

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 800
Total_scans       = 800

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29[dC]
Unblank_time      = 2[us]
```

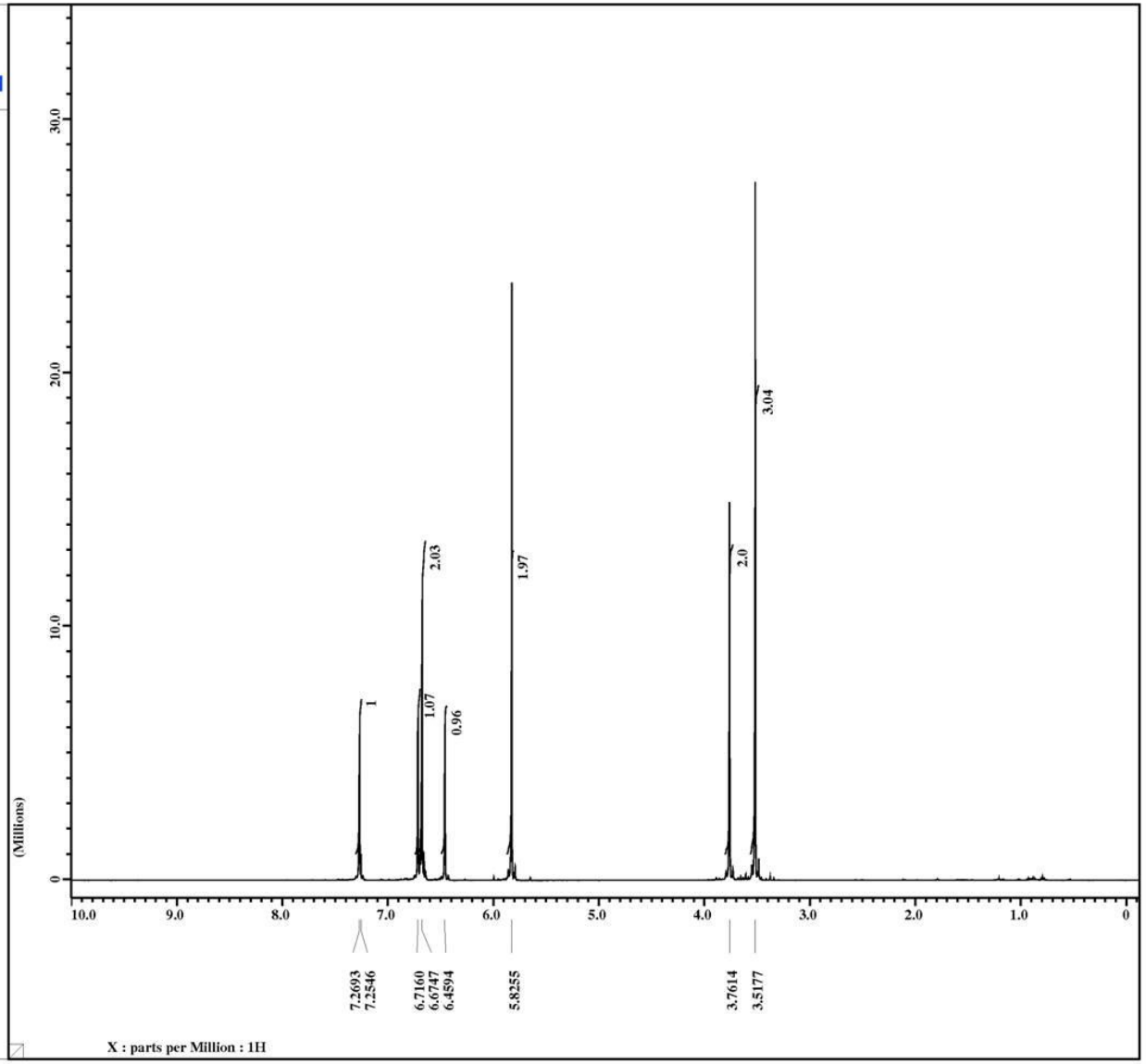
APPENDIX 72

^{1}H and ^{13}C NMR Spectra of

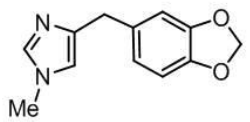
4-(Benzo[1,3]dioxol-5-yl)methyl-1-methyl-1*H*-imidazole (**193**)

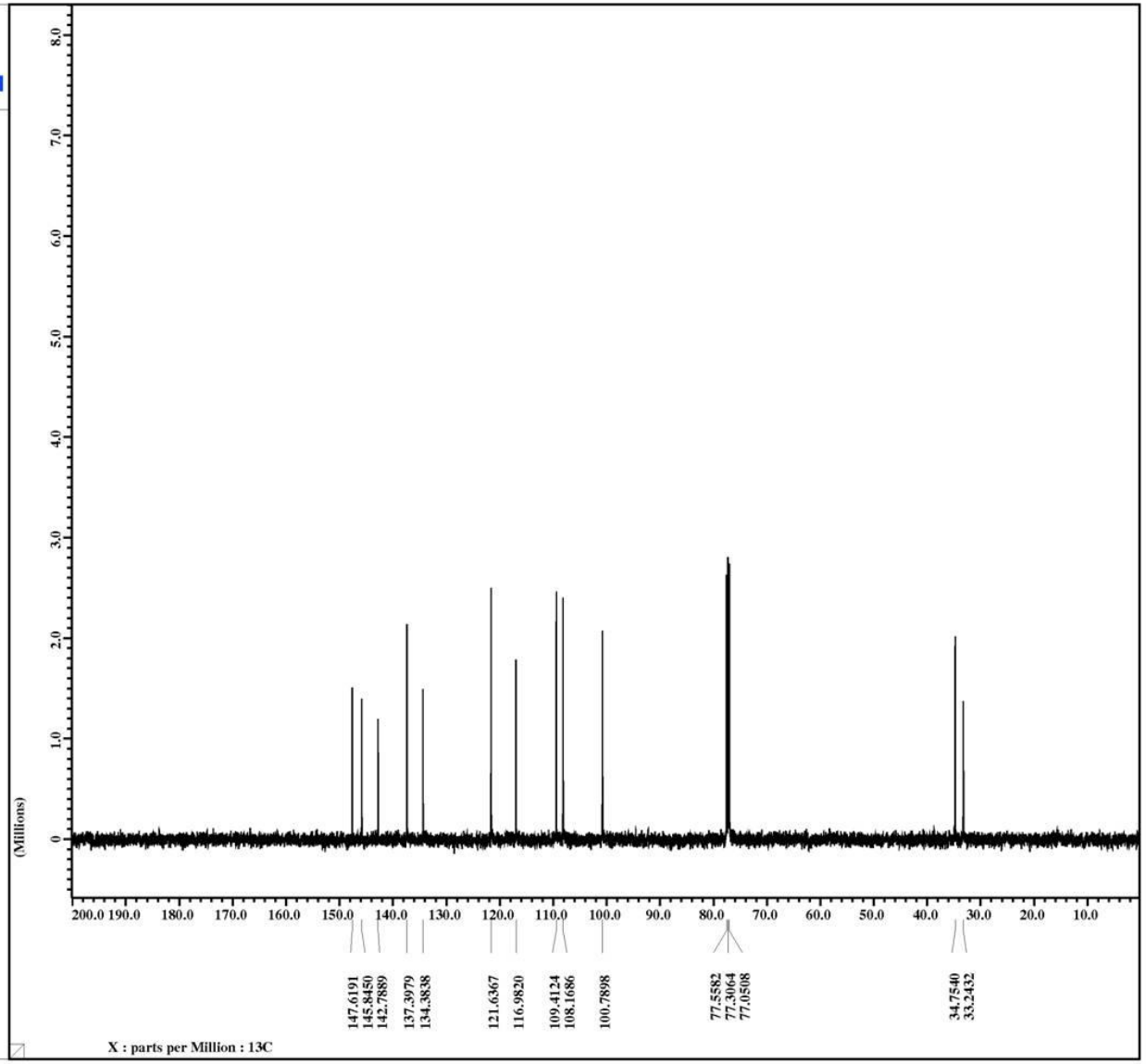
```
Filename          = V_p_126_product-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#408249
Solvent           = CHLOROFORM-D
Creation_time     =  2-MAR-2009 20:39:30
Revision_time     = 19-MAR-2010 11:59:57
Current_time      = 19-MAR-2010 12:00:13

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 12
Relaxation_delay  = 4[s]
Temp_get          = 26[dC]
Unblank_time      = 2[us]
```





```
Filename          = V_p_126_product-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#410318
Solvent           = CHLOROFORM-D
Creation_time     =  2-MAR-2009 20:47:28
Revision_time     =  2-MAR-2009 11:31:07
Current_time      = 19-MAR-2010 12:01:58

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 57
Total_scans       = 57

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28[dC]
Unblank_time      = 2[us]
```



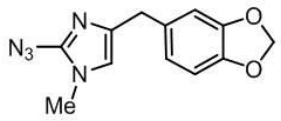


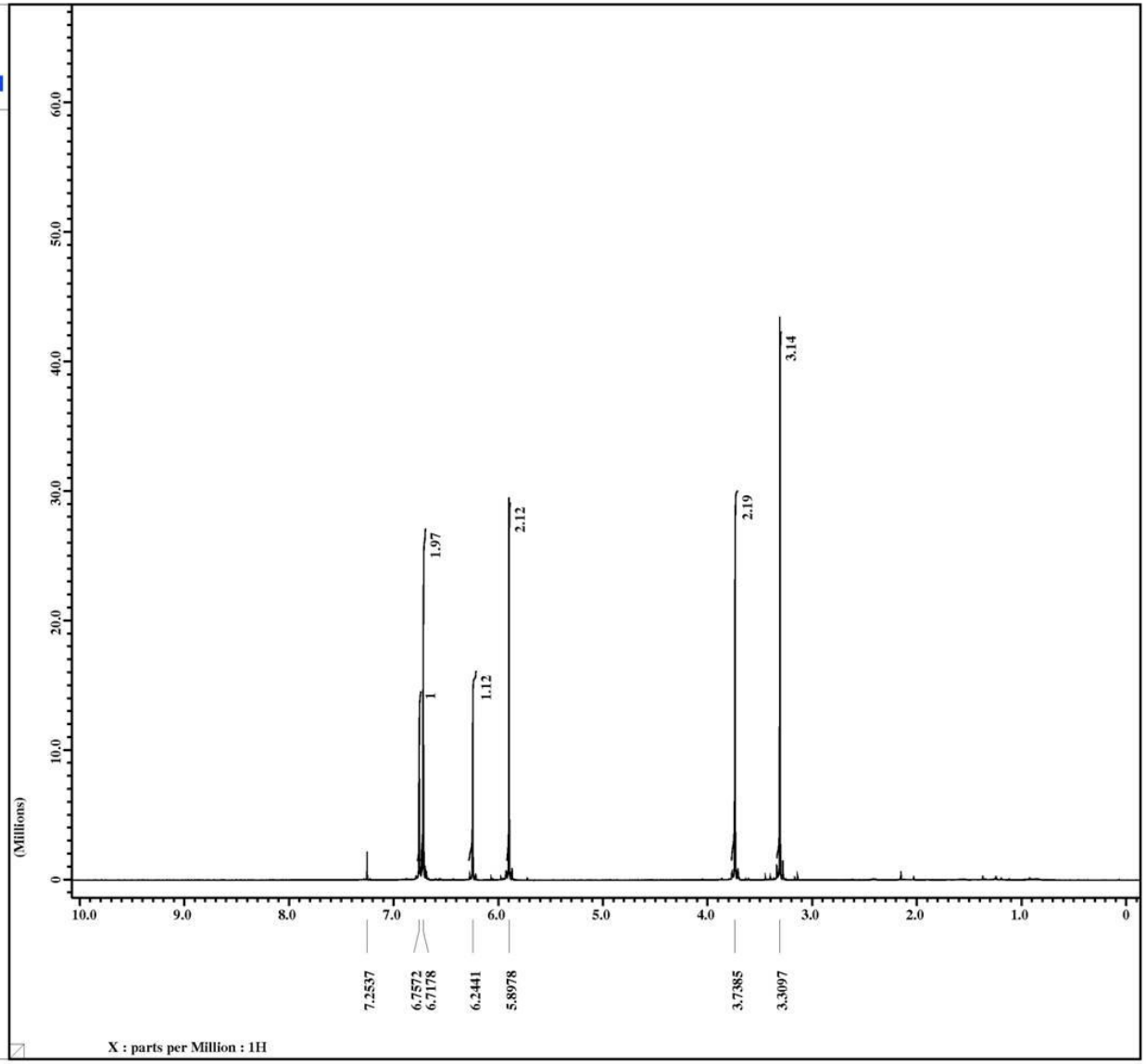
APPENDIX 73

^{1}H and ^{13}C NMR Spectra of

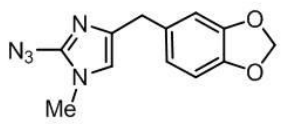
2-Azido-4-(benzo[1,3]dioxol-5-yl)methyl-1-methyl-1*H*-imidazole (**194**)

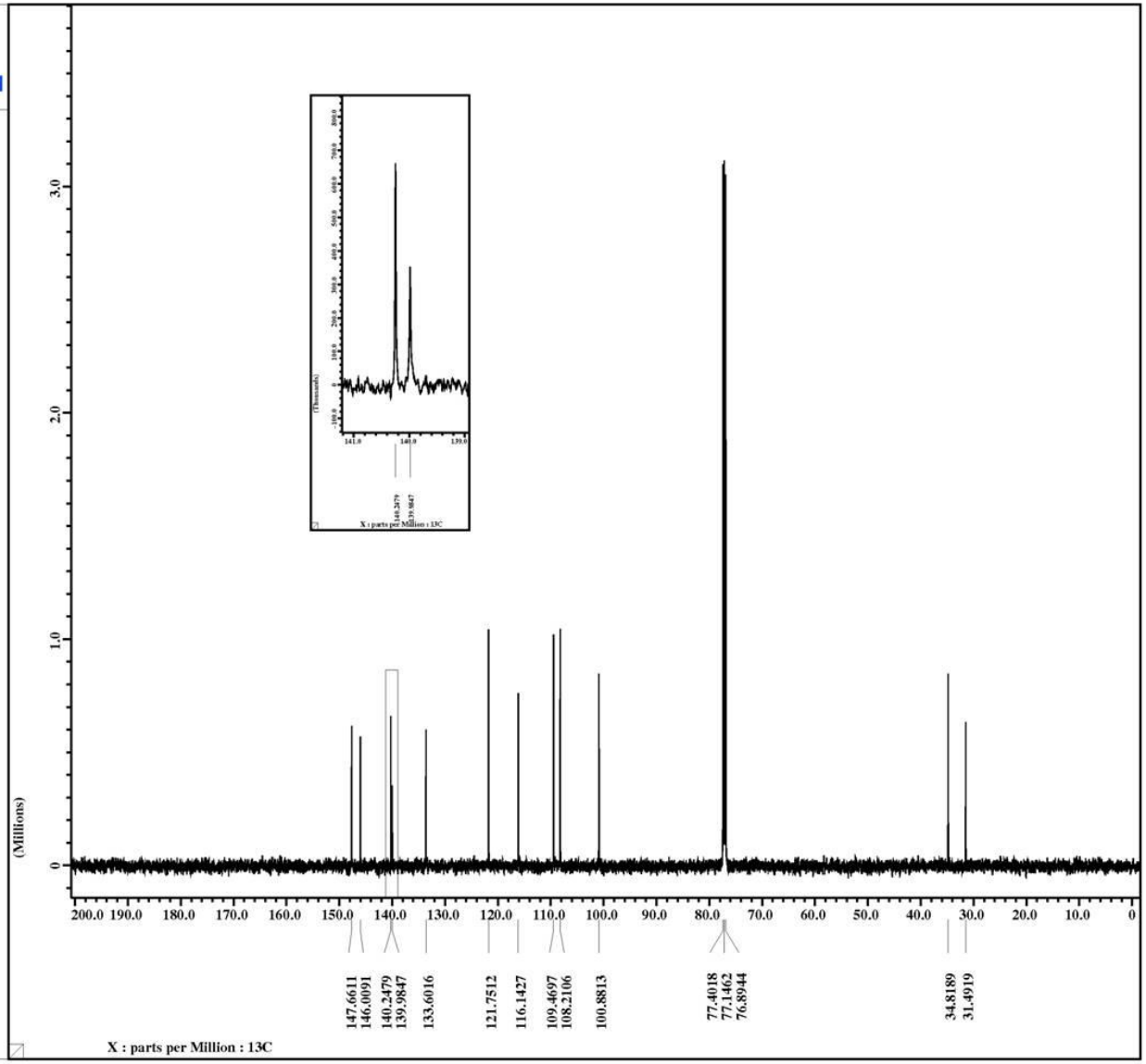
Filename	= V_p_128_azide-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#610216
Solvent	= CHLOROFORM-D
Creation_time	= 4-MAR-2009 02:18:30
Revision_time	= 19-MAR-2010 12:06:59
Current_time	= 19-MAR-2010 12:07:05
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 17
Relaxation_delay	= 4[s]
Temp_get	= 25.9[dC]
Unblank_time	= 2[us]





```
Filename          = V_p_128_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     =  3-MAR-2009 22:41:36
Revision_time     =  3-MAR-2009 16:49:00
Current_time      = 19-MAR-2010 12:05:39

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 339
Total_scans       = 339

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.7[dC]
Unblank_time      = 2[us]
```



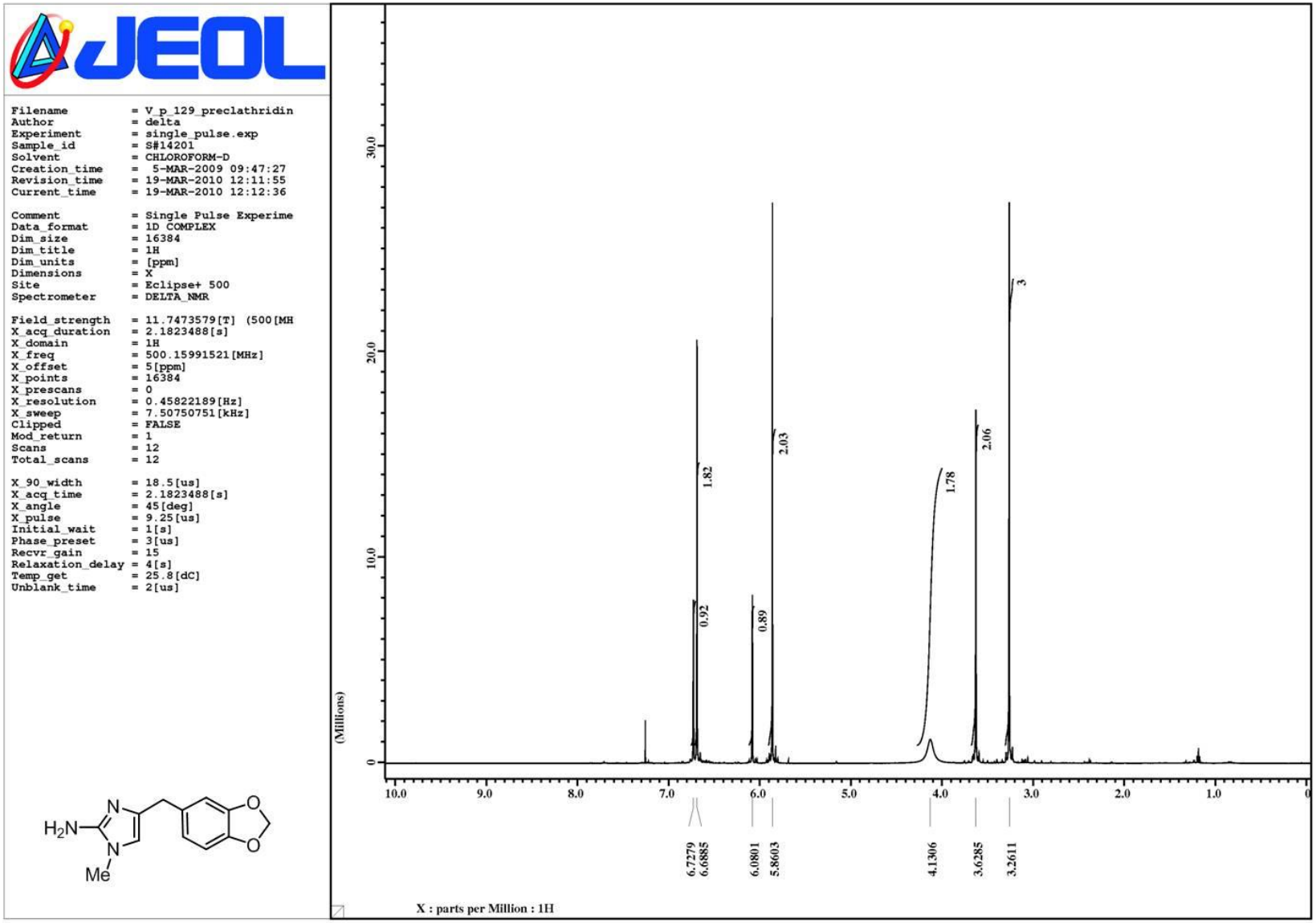


APPENDIX 74

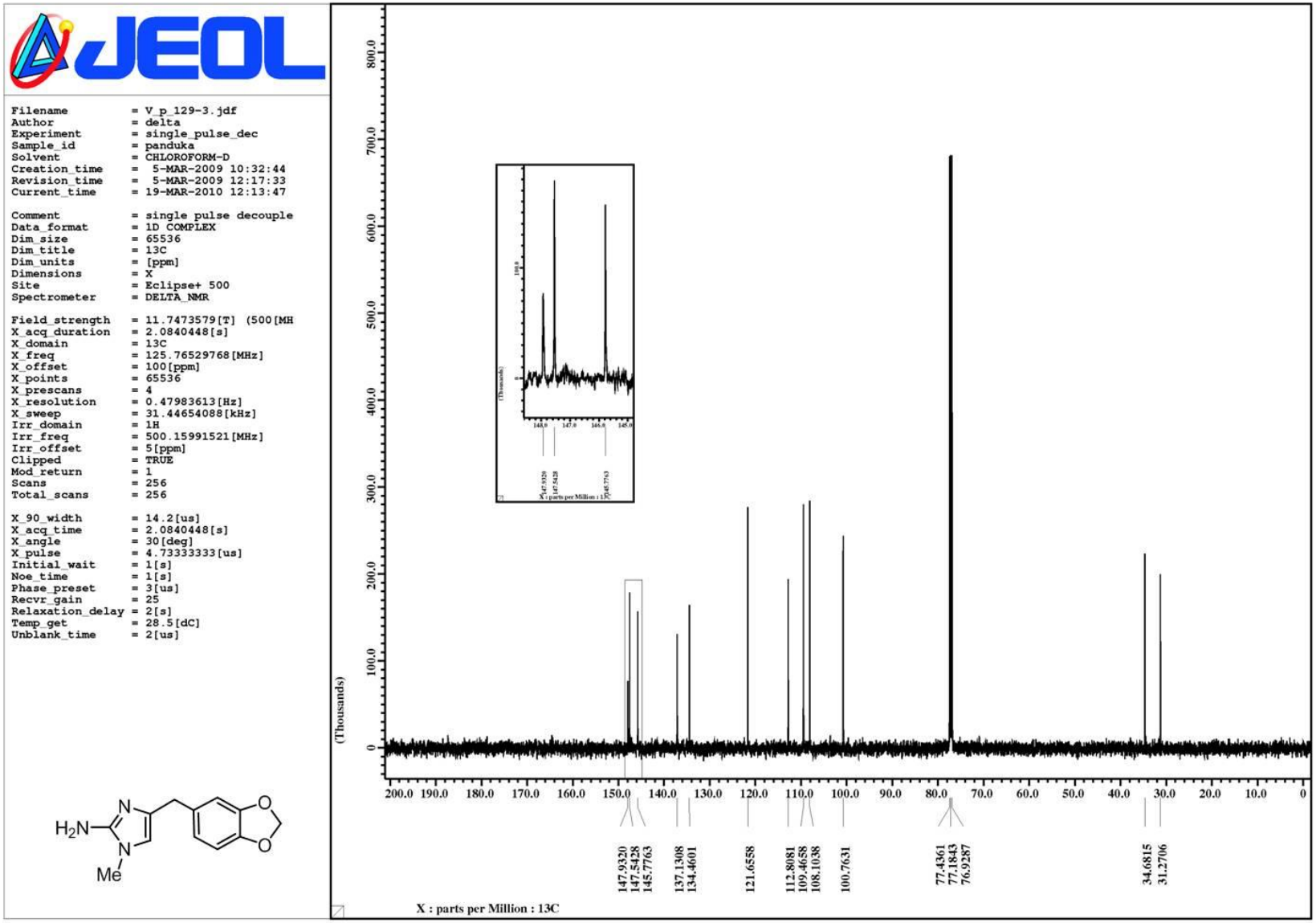
^{1}H and ^{13}C NMR Spectra of

2-amino-4-(benzo[1,3]dioxol-5-yl)methyl-1-methyl-1*H*-imidazole (**7b**):

Preclathridine A



```
Filename          = V_p_129_preclathridin
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#14201
Solvent           = CHLOROFORM-D
Creation_time     =  5-MAR-2009 09:47:27
Revision_time     = 19-MAR-2010 12:11:55
Current_time      = 19-MAR-2010 12:12:36

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```



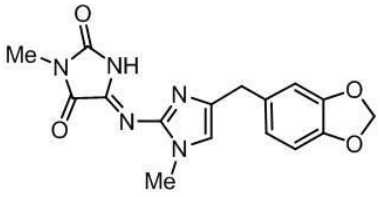
```
Filename          = V_p_129-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     =  5-MAR-2009 10:32:44
Revision_time     =  5-MAR-2009 12:17:33
Current_time      = 19-MAR-2010 12:13:47

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T]  (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 256
Total_scans       = 256

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 25
Relaxation_delay  = 2[s]
Temp_get          = 28.5[dC]
Unblank_time      = 2[us]
```

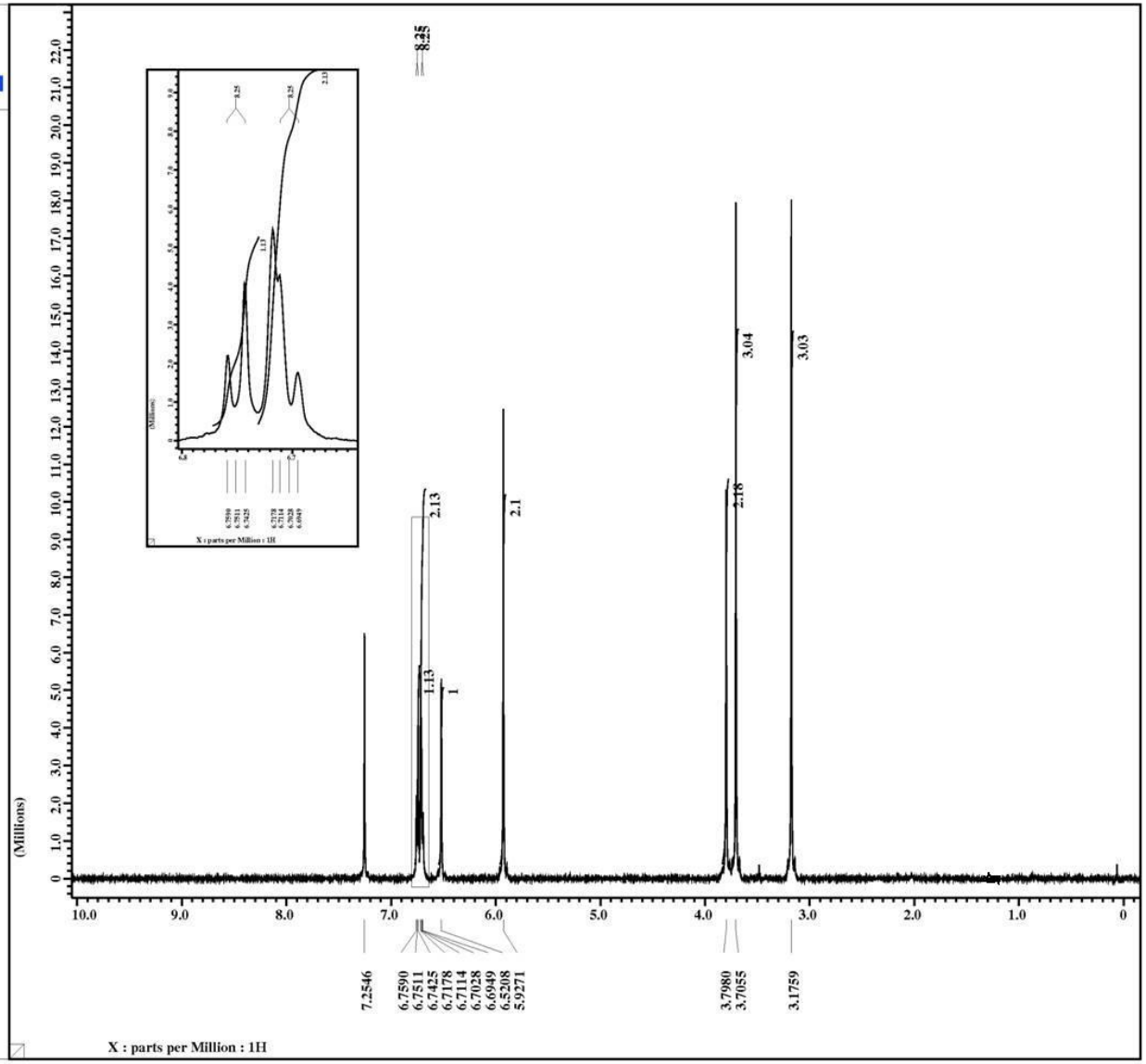
APPENDIX 75

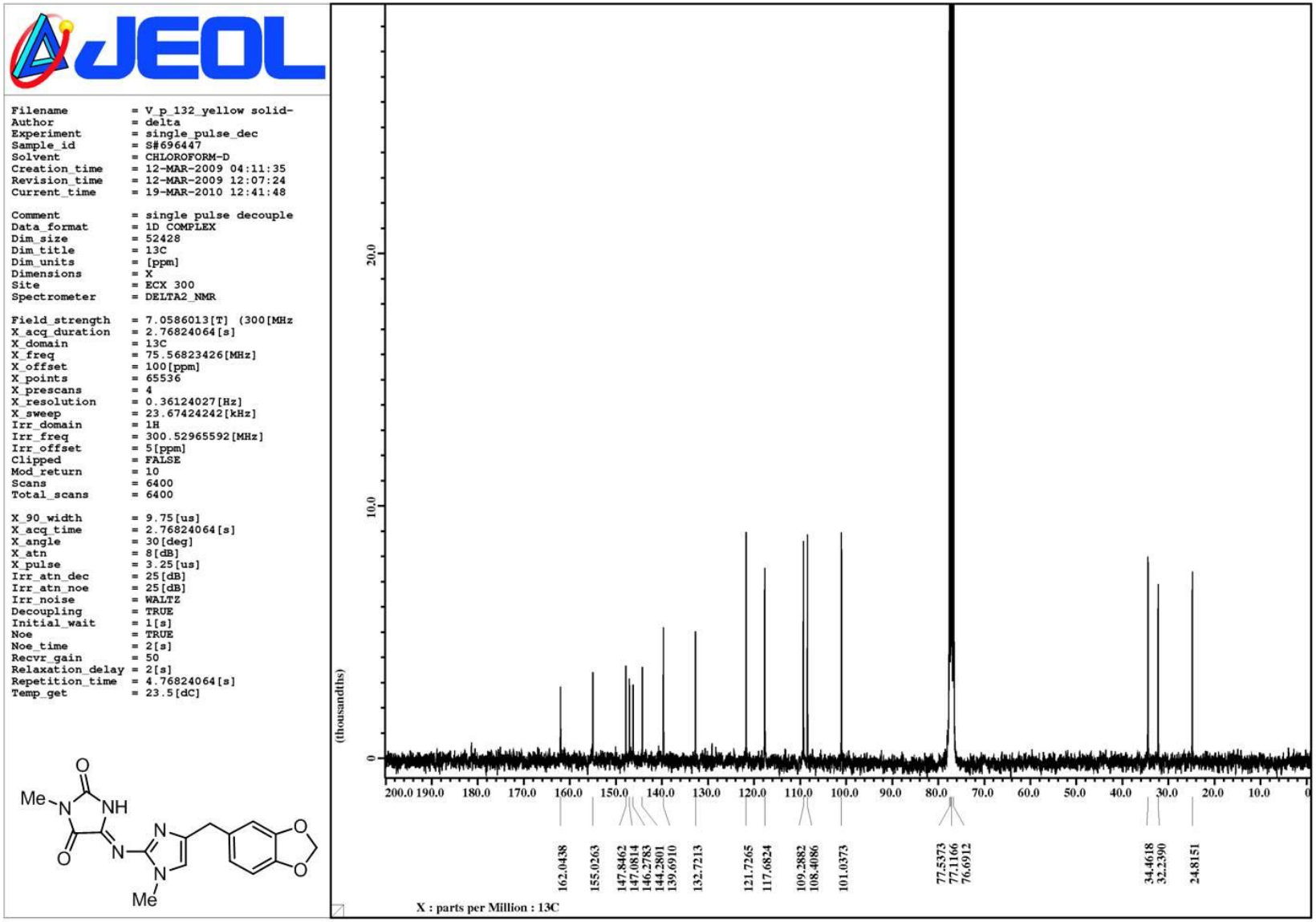
^{1}H and ^{13}C NMR Spectra of

2-(3-methylimidazolidine-2,4-dione)imino-4-(4-Benzo[1,3]dioxol-5-ylmethyl-1-methyl-1*H*-imidazole (**8b**): Clathridine A

Filename	= V_p_130_clathridine A
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#700442
Solvent	= CHLOROFORM-D
Creation_time	= 12-MAR-2009 05:04:27
Revision_time	= 19-MAR-2010 12:39:36
Current_time	= 19-MAR-2010 12:40:42
Comment	= Single Pulse Experime
Data_format	= 1D REAL
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 25
Relaxation_delay	= 4[s]
Temp_get	= 26.1[dC]
Unblank_time	= 2[us]





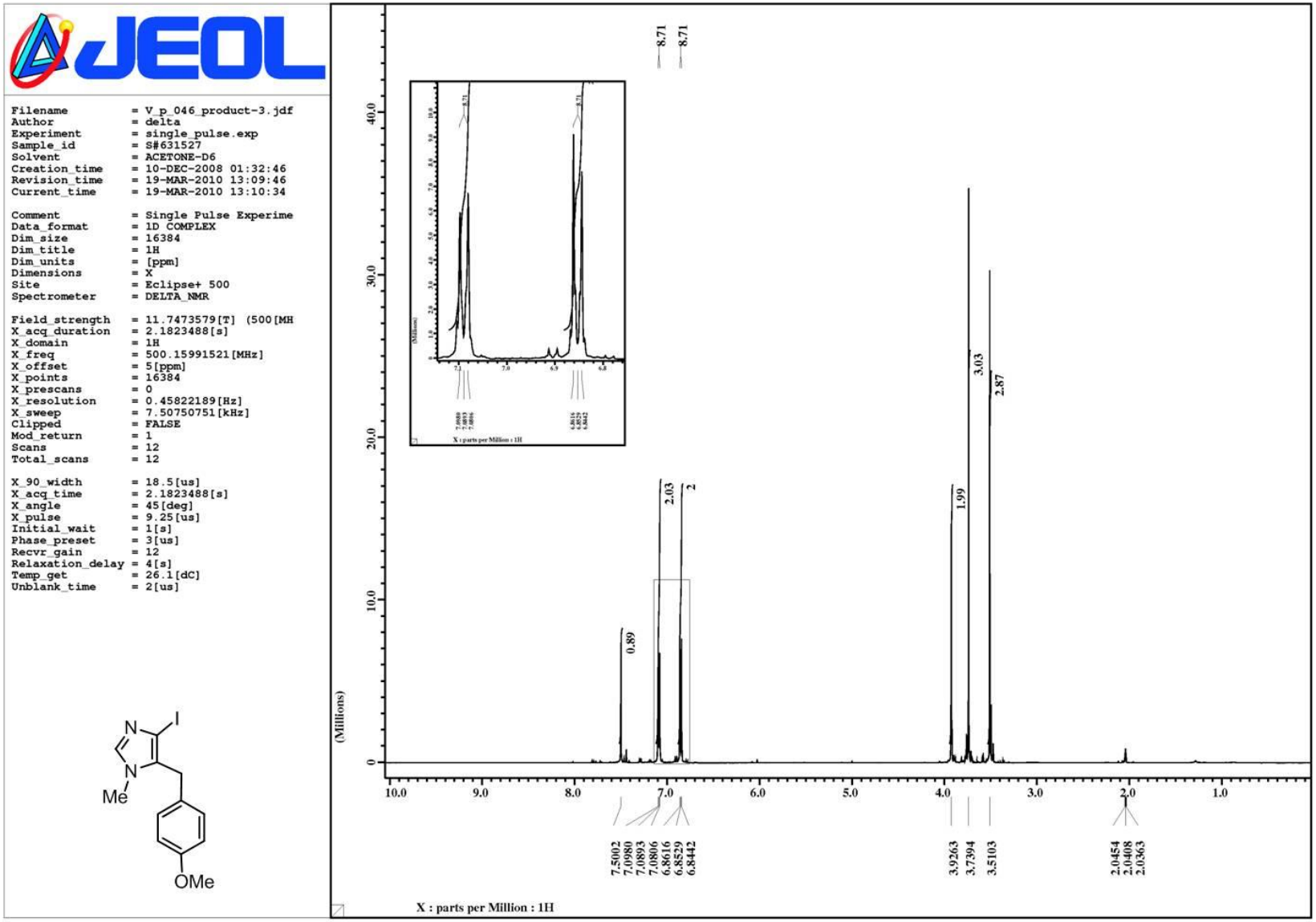


```
Filename          = V_p_132_yellow solid-
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#696447
Solvent           = CHLOROFORM-D
Creation_time     = 12-MAR-2009 04:11:35
Revision_time     = 12-MAR-2009 12:07:24
Current_time      = 19-MAR-2010 12:41:48

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```

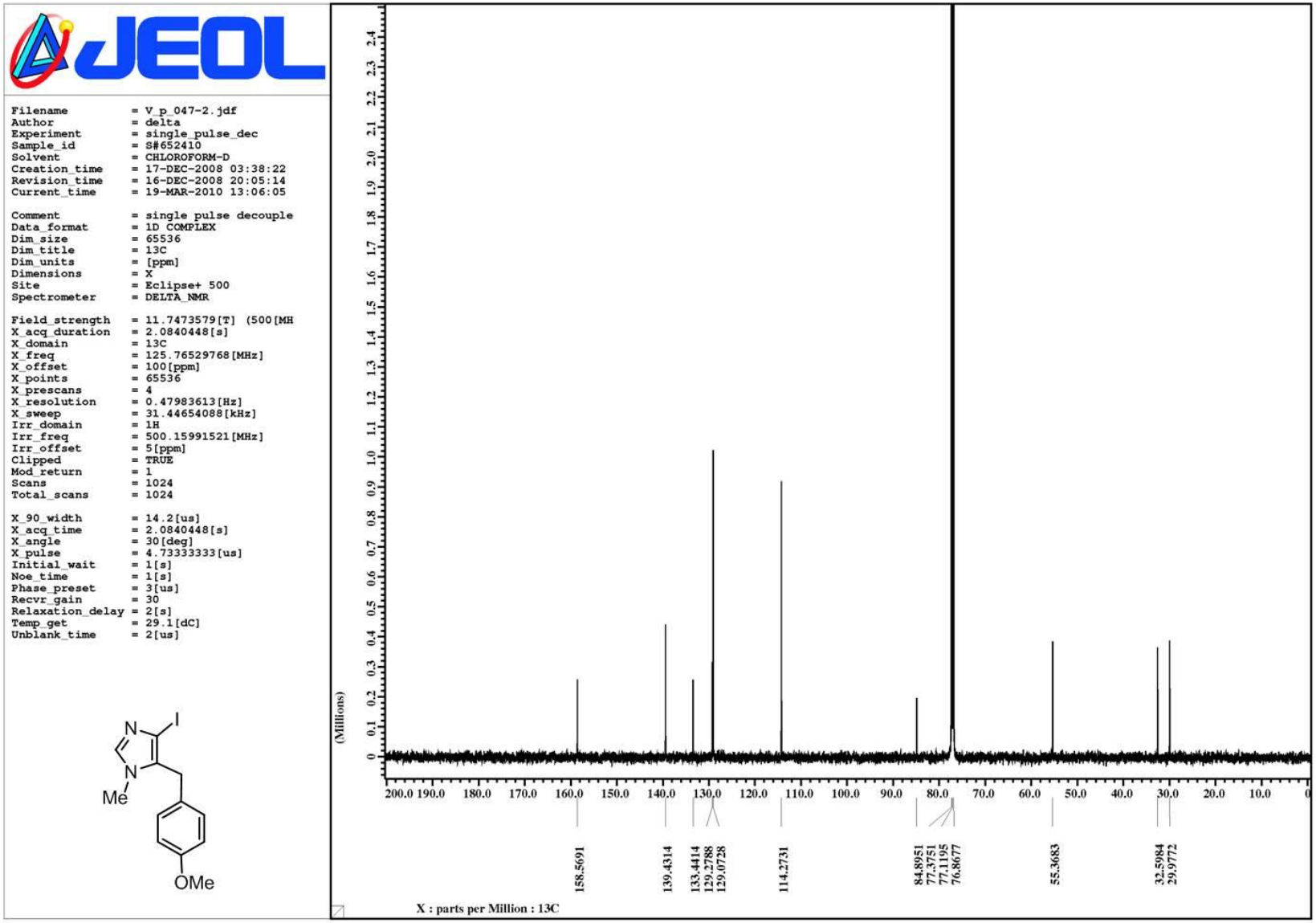
APPENDIX 76

^{1}H and ^{13}C NMR Spectra of

4-Iodo-5-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (**195**)



```
Filename          = V_p_046_product-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#631527
Solvent           = ACETONE-D6
Creation_time     = 10-DEC-2008 01:32:46
Revision_time     = 19-MAR-2010 13:09:46
Current_time      = 19-MAR-2010 13:10:34

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 12
Relaxation_delay  = 4[s]
Temp_get          = 26.1[dC]
Unblank_time      = 2[us]
```

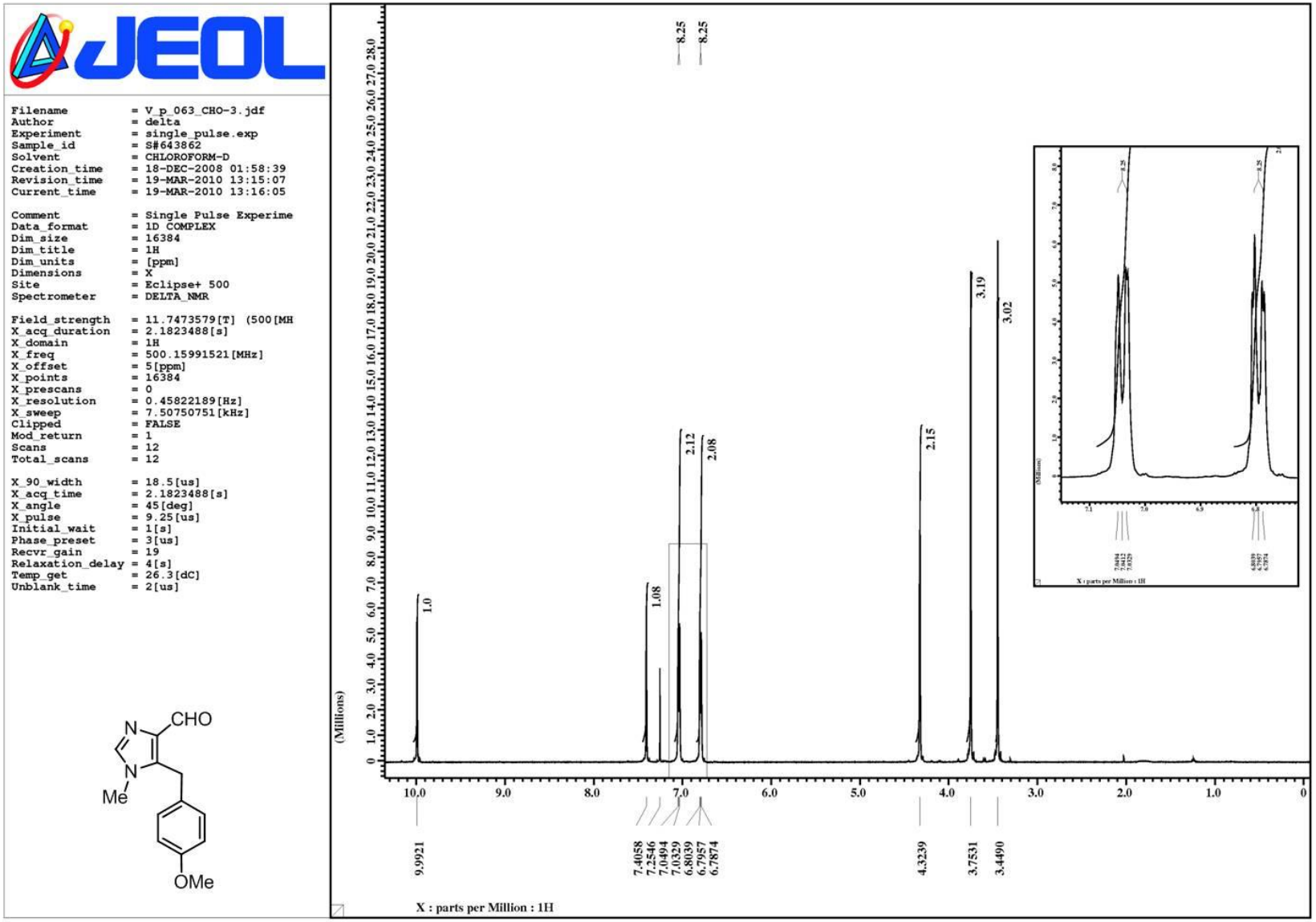


```
Filename          = V_p_047-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#652410
Solvent           = CHLOROFORM-D
Creation_time     = 17-DEC-2008 03:38:22
Revision_time     = 16-DEC-2008 20:05:14
Current_time      = 19-MAR-2010 13:06:05

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.1[dC]
Unblank_time      = 2[us]
```

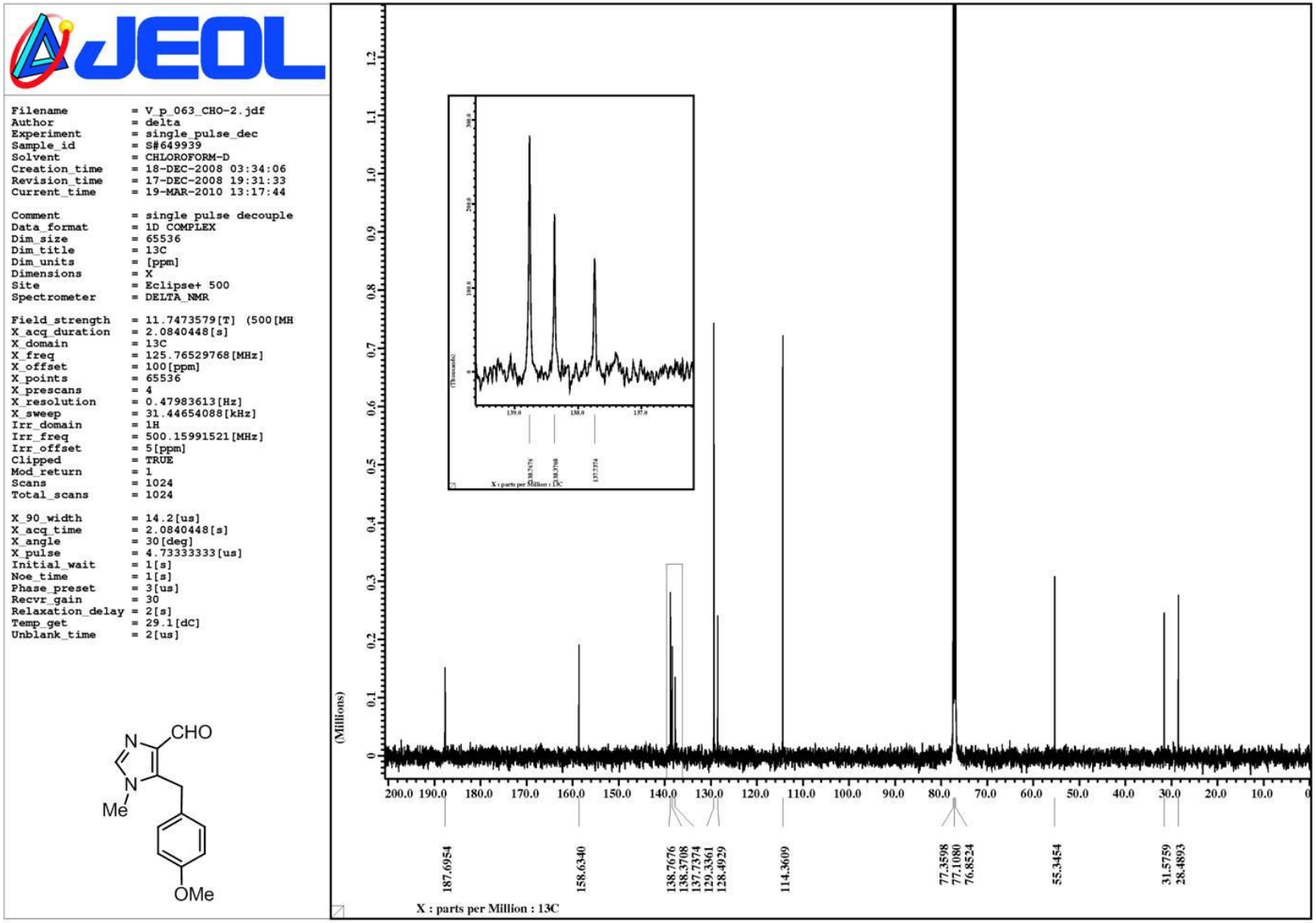
APPENDIX 77

^{1}H and ^{13}C NMR Spectra of

5-(4-Methoxybenzyl-1-methyl-1*H*-imidazole-4-carbaldehyde (**196**)



```
Filename          = V_p_063_CHO-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#643862
Solvent           = CHLOROFORM-D
Creation_time     = 18-DEC-2008 01:58:39
Revision_time     = 19-MAR-2010 13:15:07
Current_time      = 19-MAR-2010 13:16:05

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 19
Relaxation_delay  = 4[s]
Temp_get          = 26.3[dC]
Unblank_time      = 2[us]
```



```
Filename          = V_p_063_CHO-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#649939
Solvent           = CHLOROFORM-D
Creation_time     = 18-DEC-2008 03:34:06
Revision_time     = 17-DEC-2008 19:31:33
Current_time      = 19-MAR-2010 13:17:44

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.1[dC]
Unblank_time      = 2[us]
```


(Millions)

187.6954
158.6340
138.7676
138.3708
137.7374
129.3361
128.4929
114.3609
77.3598
77.1080
76.8524
55.3454
31.5759
28.4893

X : parts per Million : 13C

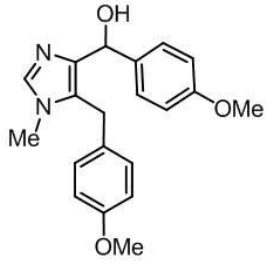
APPENDIX 78

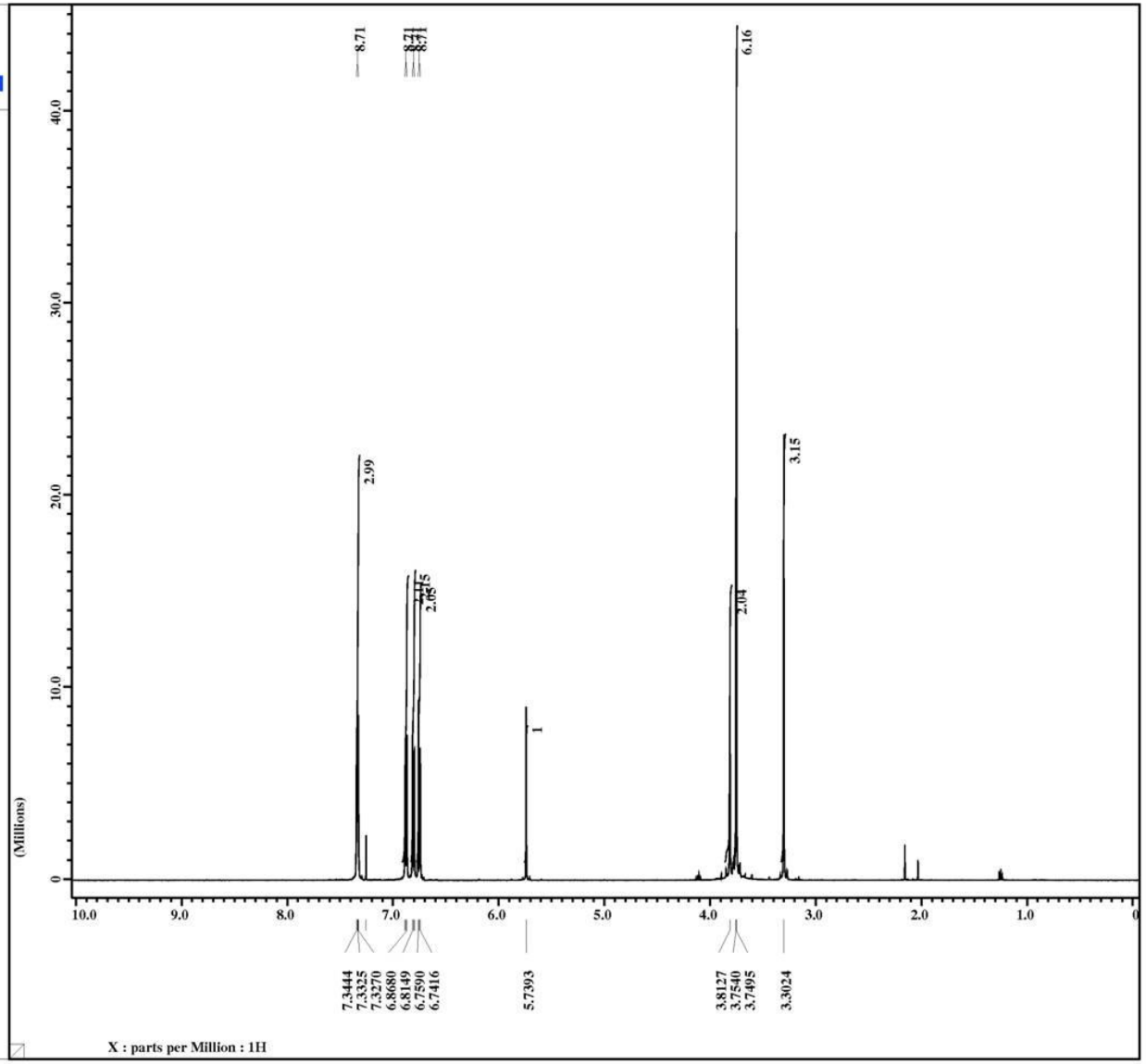
^{1}H and ^{13}C NMR Spectra of

4-[hydroxy-(4-methoxyphenyl)]methyl-5-(4-methoxybenzyl)-1-methyl-1*H*-imidazole

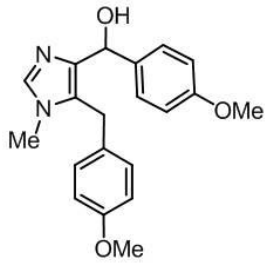
(**197**)

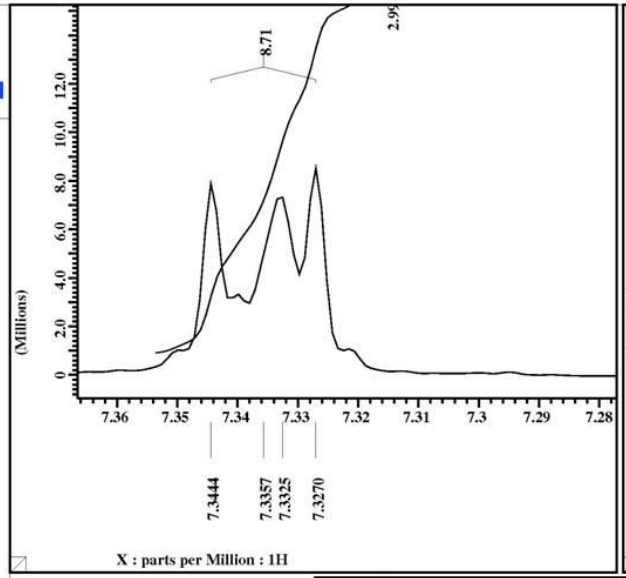
Filename	= V_p_067_alcohol-2.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#716858
Solvent	= CHLOROFORM-D
Creation_time	= 19-DEC-2008 04:00:35
Revision_time	= 18-DEC-2008 20:01:24
Current_time	= 19-MAR-2010 13:21:32
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 17
Relaxation_delay	= 4[s]
Temp_get	= 25.9[dC]
Unblank_time	= 2[us]

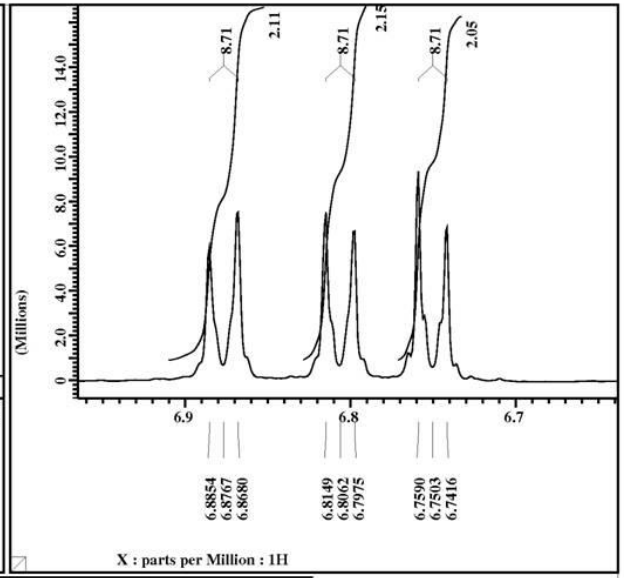


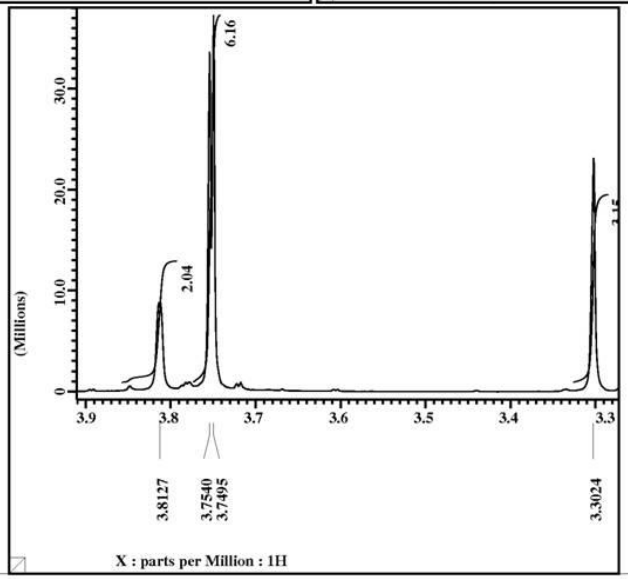


```
Filename          = V_p_067_alcohol-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#716858
Solvent           = CHLOROFORM-D
Creation_time     = 19-DEC-2008 04:00:35
Revision_time     = 18-DEC-2008 20:01:24
Current_time      = 19-MAR-2010 13:22:02

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 17
Relaxation_delay  = 4[s]
Temp_get          = 25.9[dC]
Unblank_time      = 2[us]
```

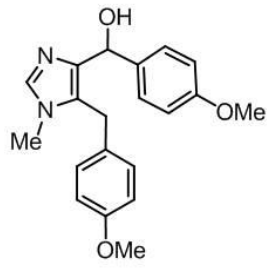


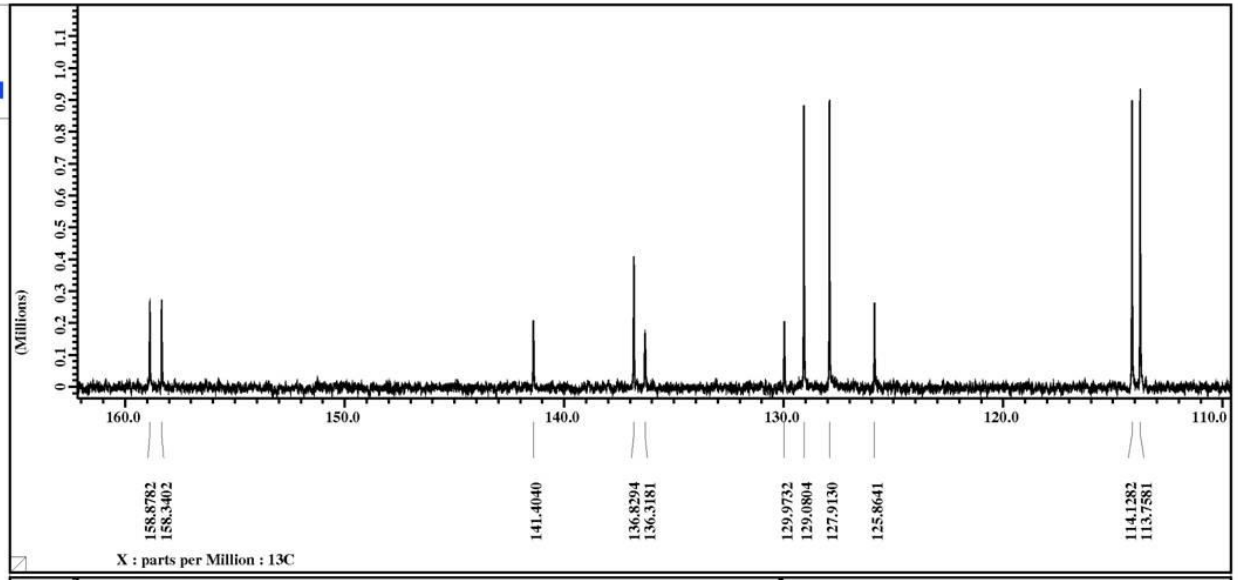


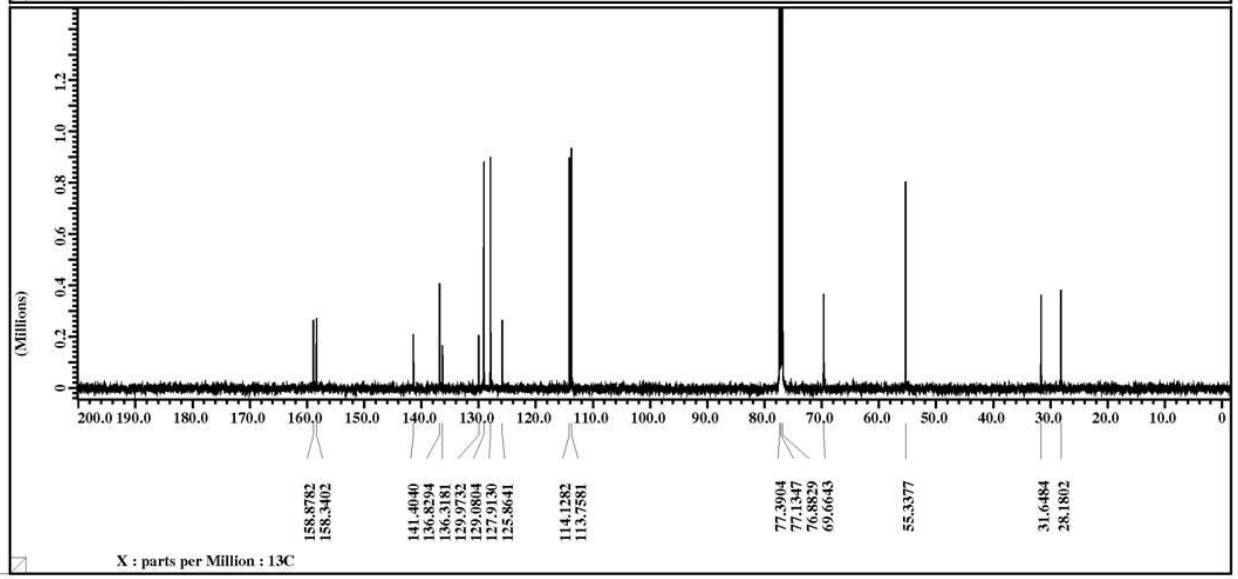




```
Filename          = V_p_067_alcohol-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#718437
Solvent           = CHLOROFORM-D
Creation_time     = 19-DEC-2008 05:29:36
Revision_time     = 19-DEC-2008 09:41:18
Current_time      = 19-MAR-2010 13:22:42

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.9[dC]
Unblank_time      = 2[us]
```



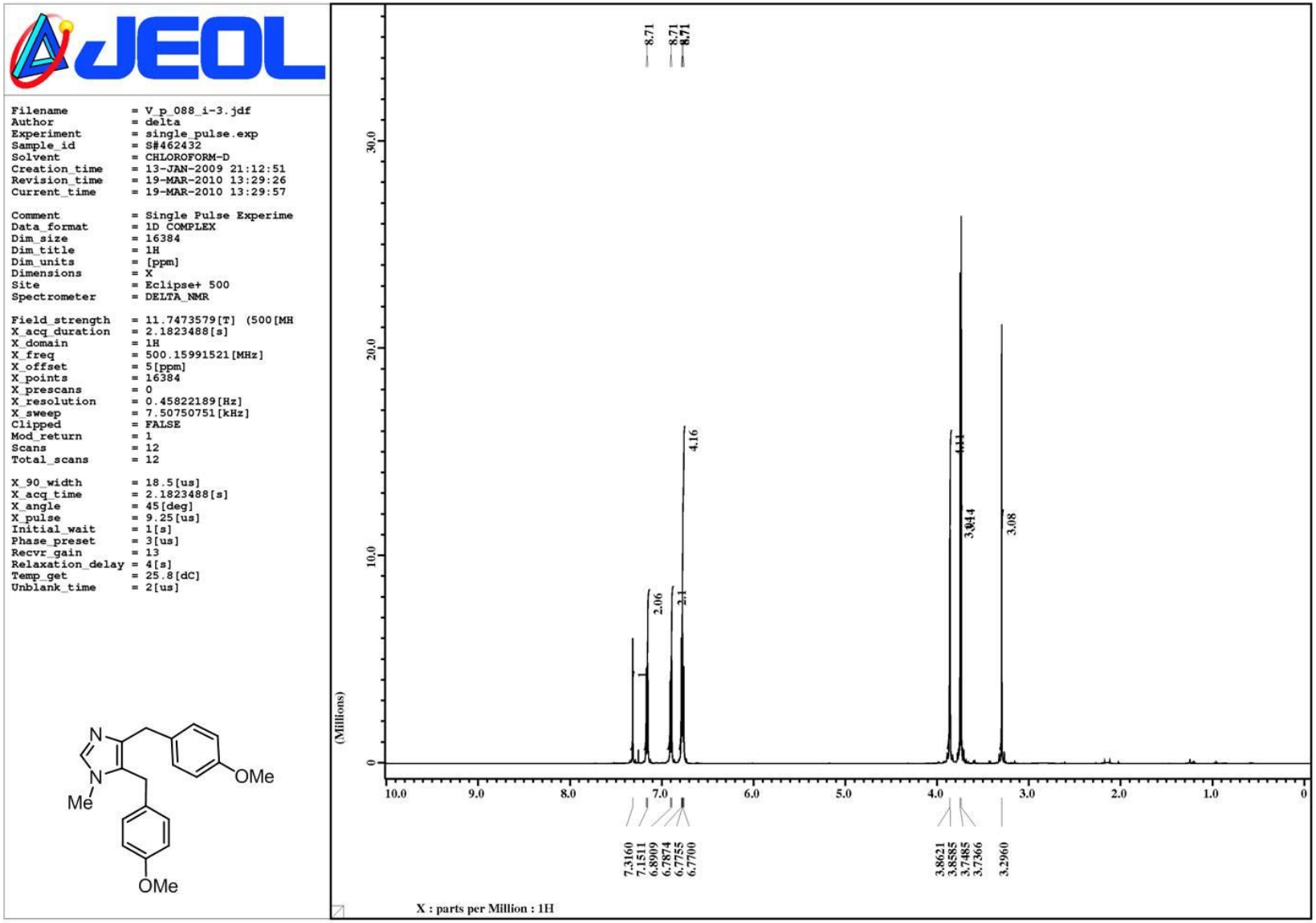




APPENDIX 79

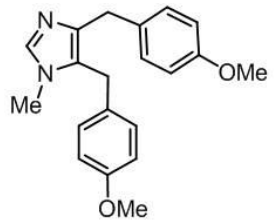
^{1}H and ^{13}C NMR Spectra of

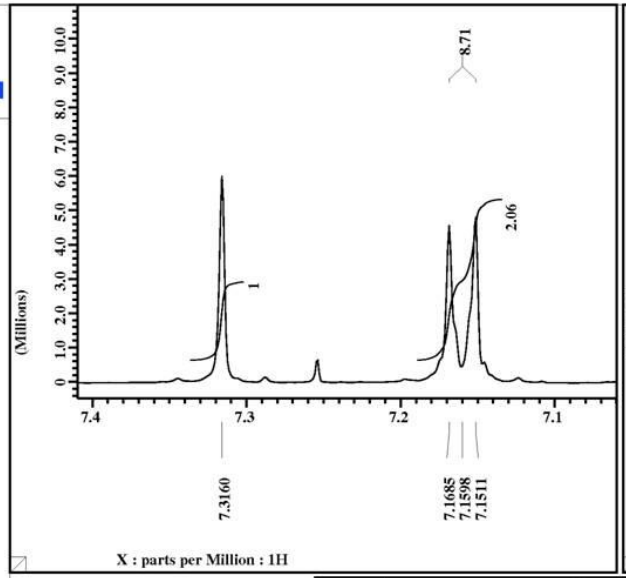
4,5-Bis(4-methoxybenzyl)-1-methyl-1*H*-imidazole (**198**)

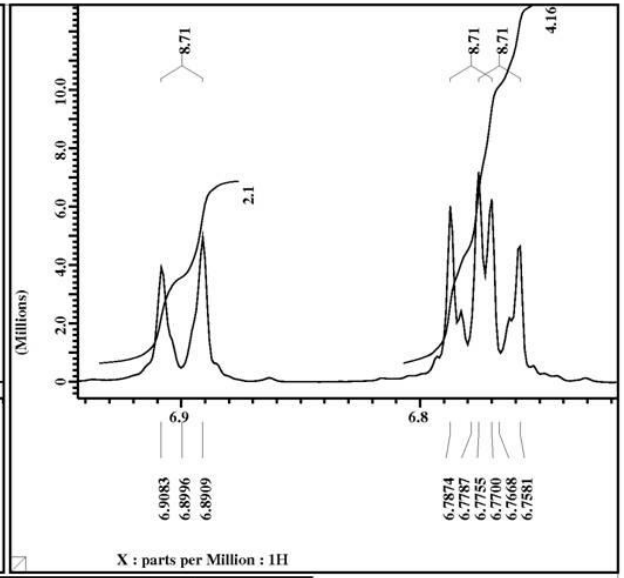


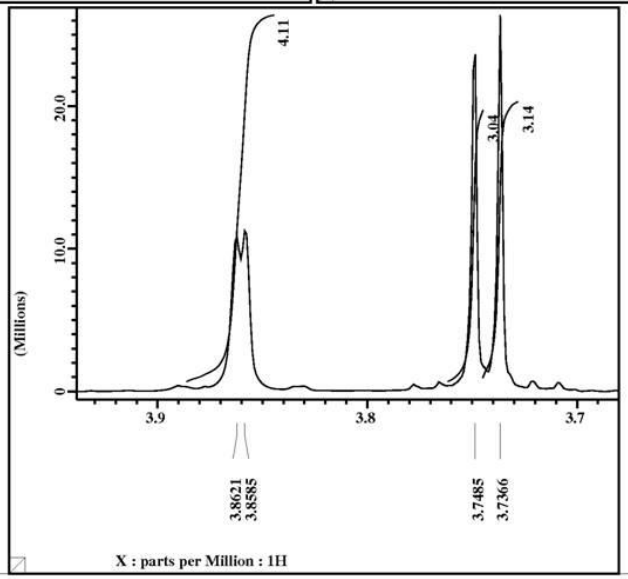
```
Filename          = V_p_088_i-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#462432
Solvent           = CHLOROFORM-D
Creation_time     = 13-JAN-2009 21:12:51
Revision_time     = 19-MAR-2010 13:29:26
Current_time      = 19-MAR-2010 13:29:57

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 13
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```

Filename	= V_p_088_i-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#462432
Solvent	= CHLOROFORM-D
Creation_time	= 13-JAN-2009 21:12:51
Revision_time	= 19-MAR-2010 13:29:26
Current_time	= 19-MAR-2010 13:30:32
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 13
Relaxation_delay	= 4[s]
Temp_get	= 25.8[dC]
Unblank_time	= 2[us]

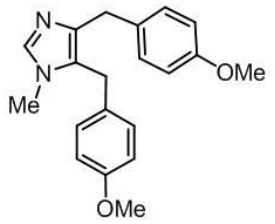


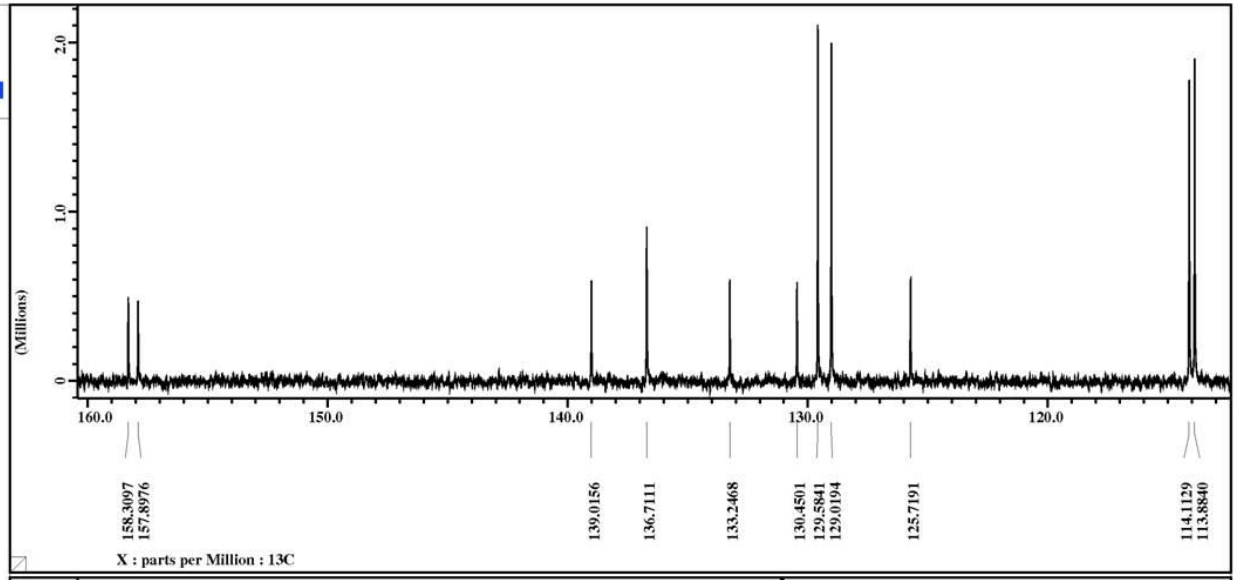


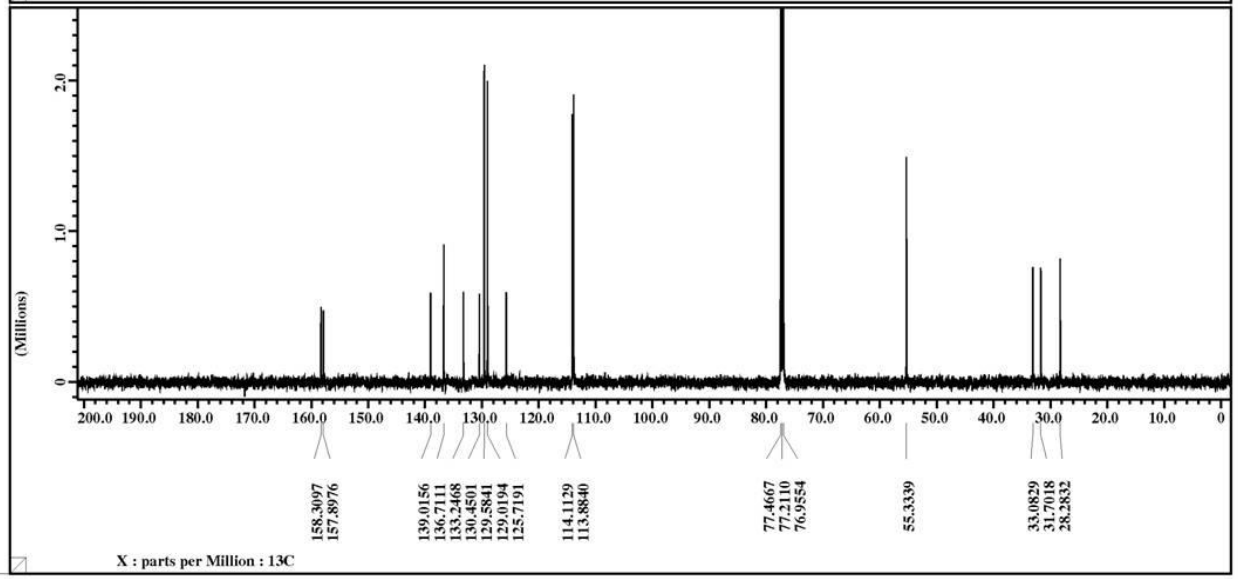




```
Filename          = V_p_088_i-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#463955
Solvent           = CHLOROFORM-D
Creation_time     = 13-JAN-2009 21:32:09
Revision_time     = 13-JAN-2009 13:13:10
Current_time      = 19-MAR-2010 13:31:15

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 203.0
Total_scans       = 203.0

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.6[dC]
Unblank_time      = 2[us]
```





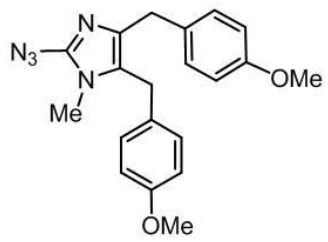


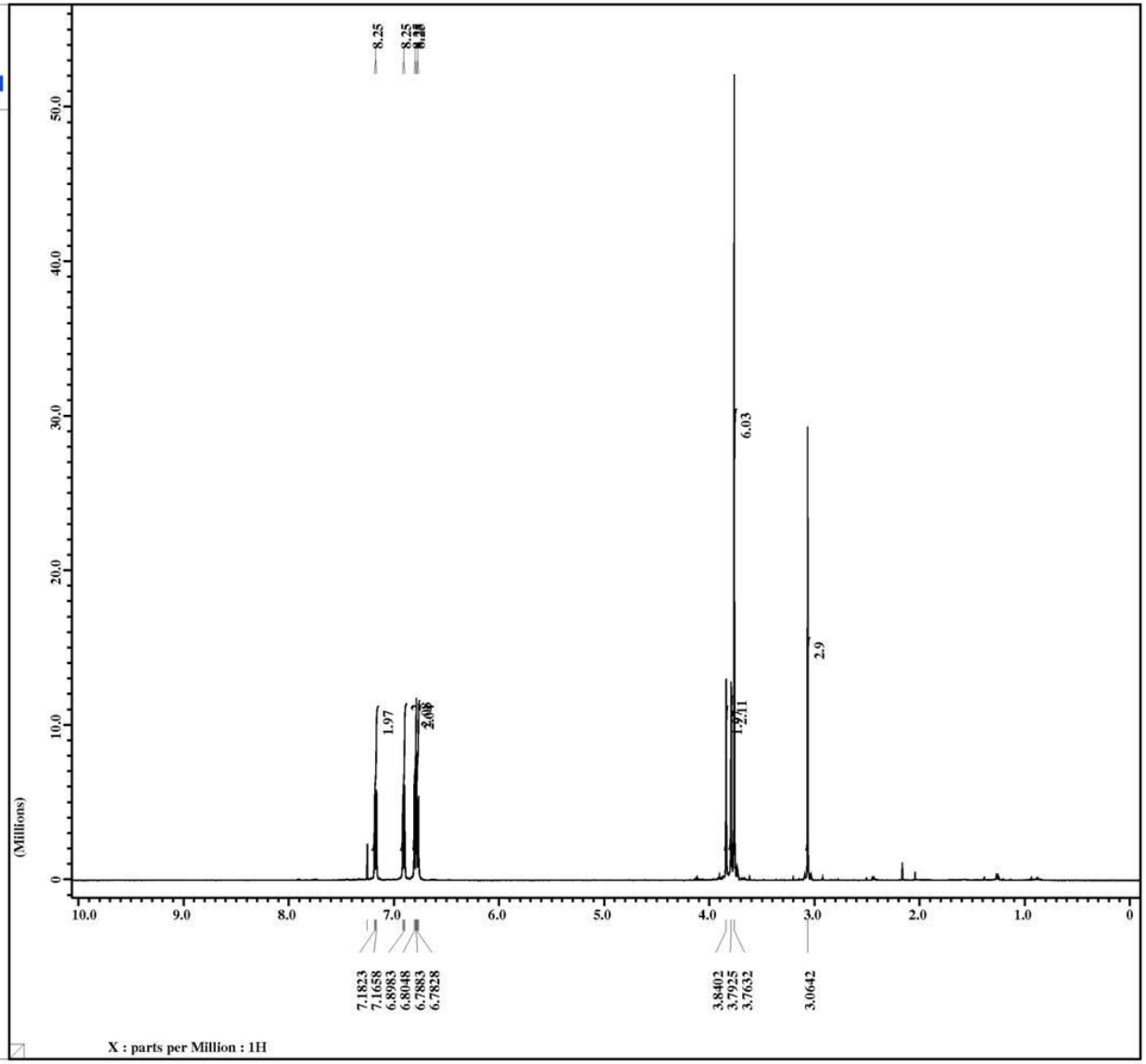
APPENDIX 80

^{1}H and ^{13}C NMR Spectra of

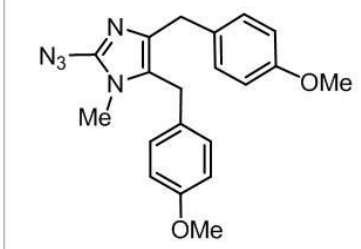
2-Azido-4,5-bis(4-methoxybenzyl)-1-methyl-1*H*-imidazole (**199**)

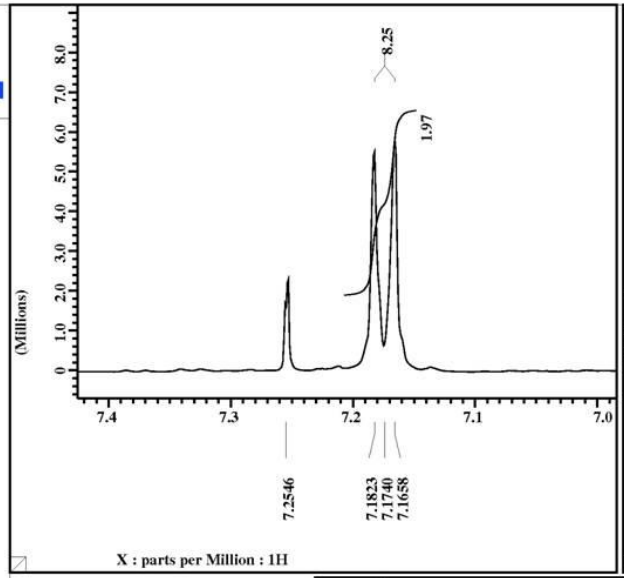
Filename	= V_p_091_azide-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#648334
Solvent	= CHLOROFORM-D
Creation_time	= 15-JAN-2009 02:23:26
Revision_time	= 19-MAR-2010 13:36:43
Current_time	= 19-MAR-2010 13:36:56
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 16
Relaxation_delay	= 4[s]
Temp_get	= 26.5[dC]
Unblank_time	= 2[us]

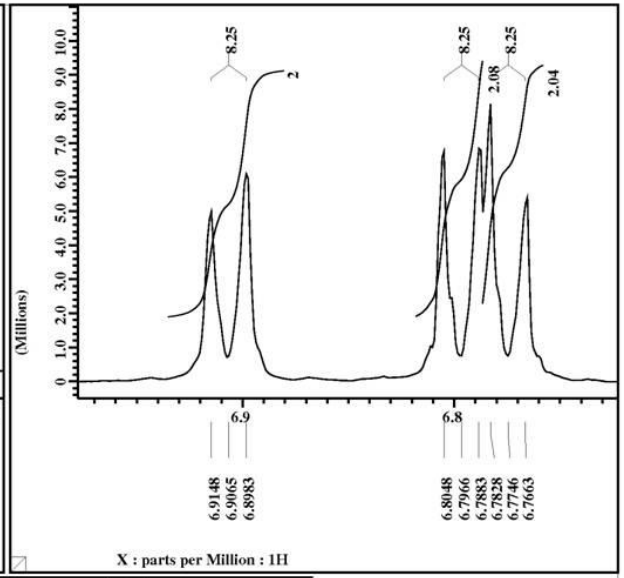


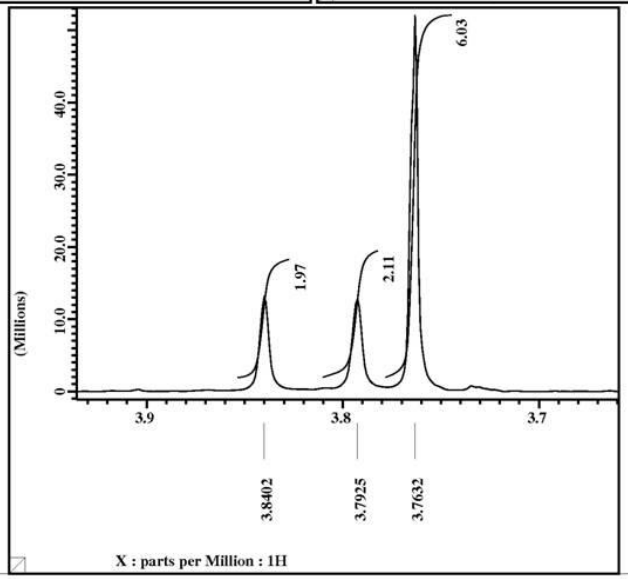


```
Filename          = V_p_091_azide-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#648334
Solvent           = CHLOROFORM-D
Creation_time     = 15-JAN-2009 02:23:26
Revision_time     = 19-MAR-2010 13:36:43
Current_time      = 19-MAR-2010 13:37:33

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 16
Relaxation_delay  = 4[s]
Temp_get          = 26.5[dC]
Unblank_time      = 2[us]
```

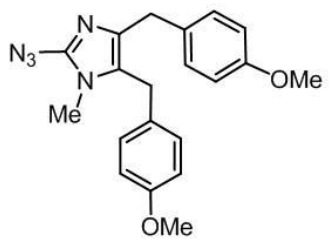


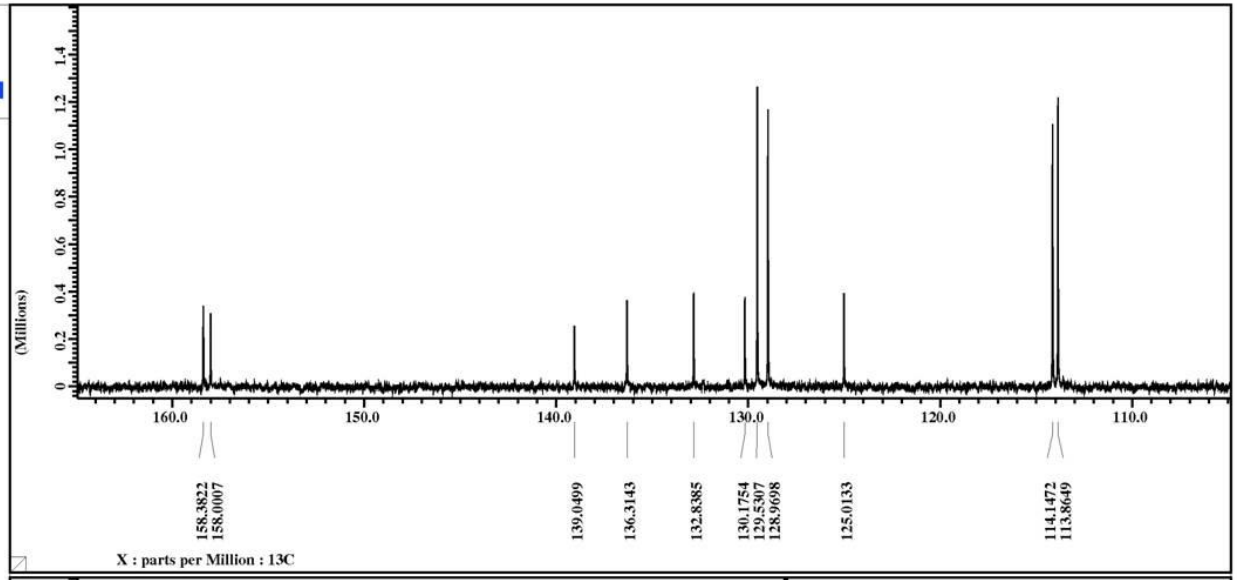


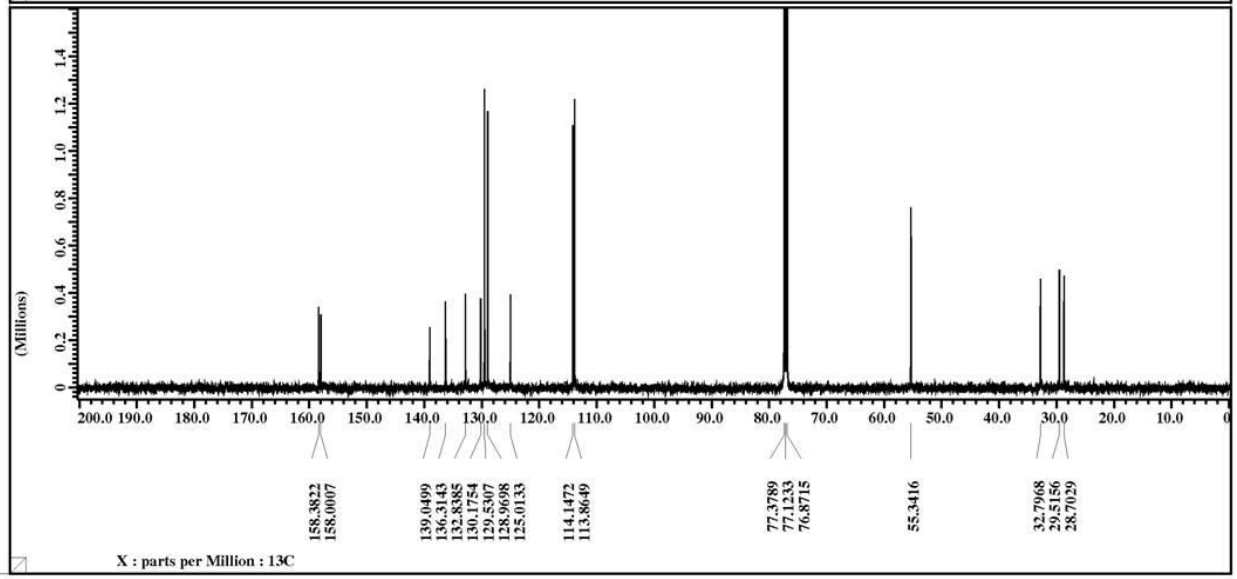




```
Filename          = V_p_091_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#649872
Solvent           = CHLOROFORM-D
Creation_time     = 15-JAN-2009 03:52:06
Revision_time     = 14-JAN-2009 19:31:58
Current_time      = 19-MAR-2010 13:38:22

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.3[dC]
Unblank_time      = 2[us]
```





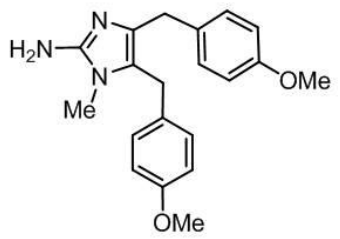


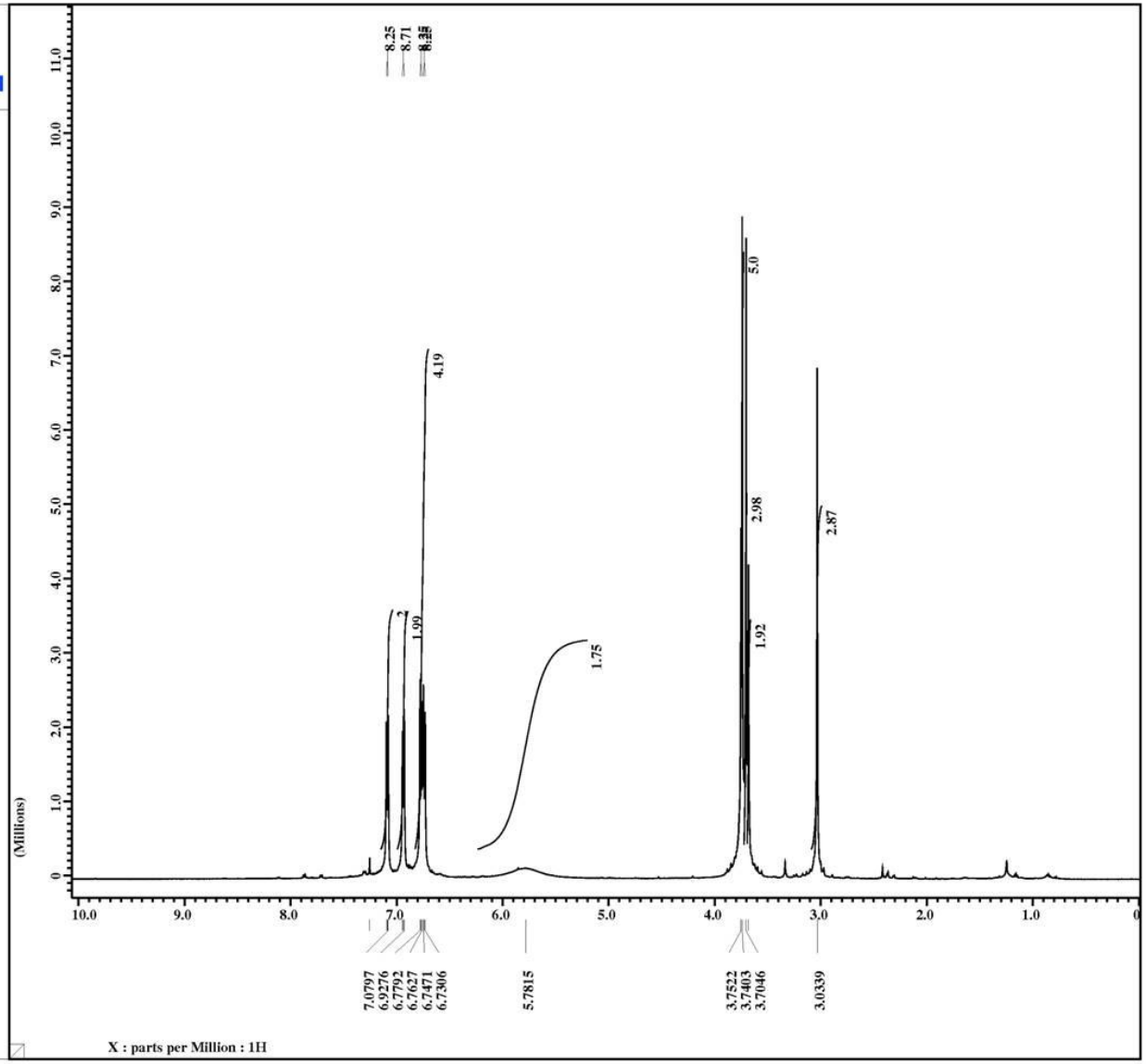
APPENDIX 81

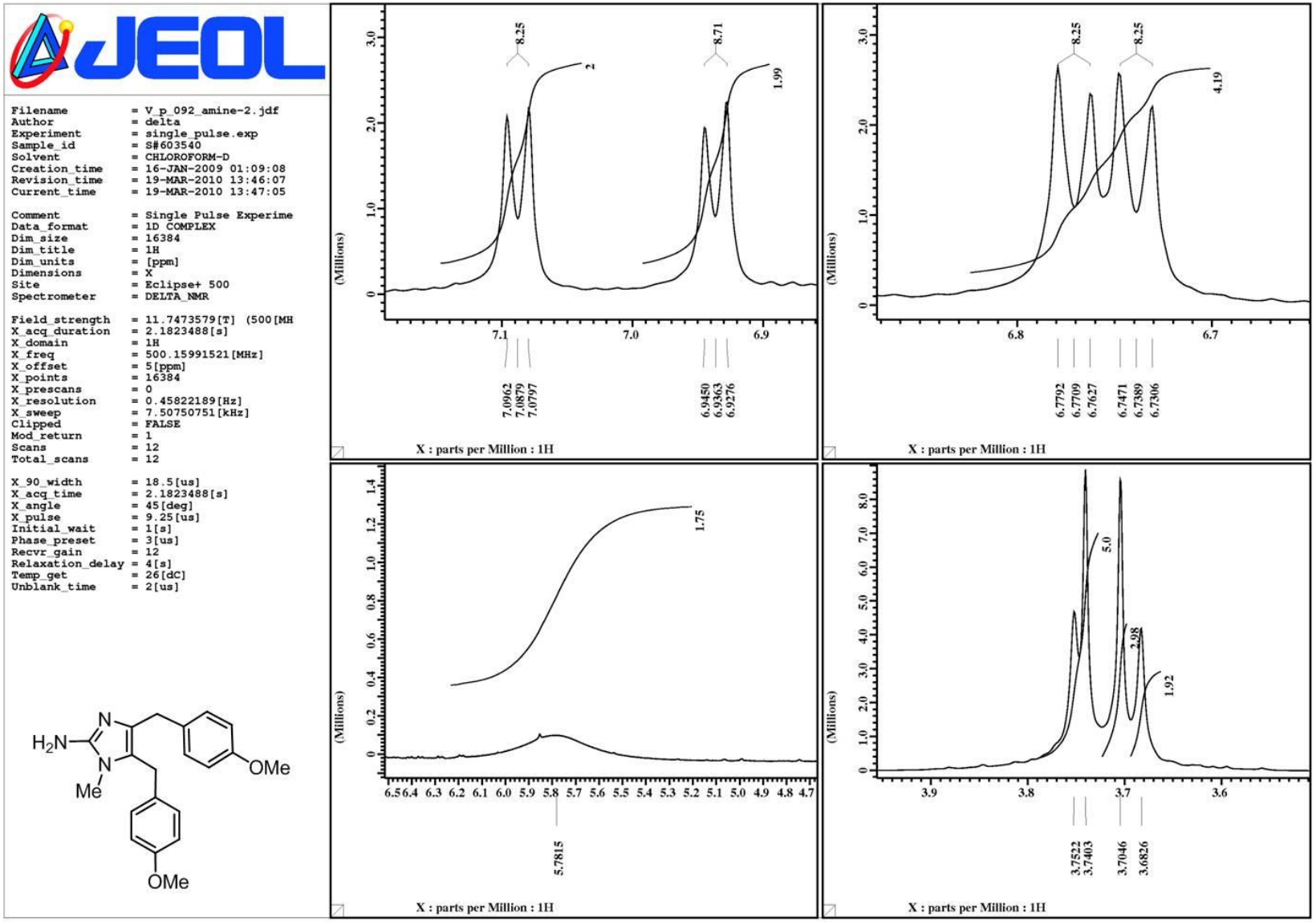
^{1}H and ^{13}C NMR Spectra of

2-Amino-4,5-bis(4-methoxybenzyl)-1-methyl-1*H*-imidazole (**200**)

Filename	= V_p_092_amine-2.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#603540
Solvent	= CHLOROFORM-D
Creation_time	= 16-JAN-2009 01:09:08
Revision_time	= 19-MAR-2010 13:46:07
Current_time	= 19-MAR-2010 13:46:20
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 12
Relaxation_delay	= 4[s]
Temp_get	= 26[dC]
Unblank_time	= 2[us]

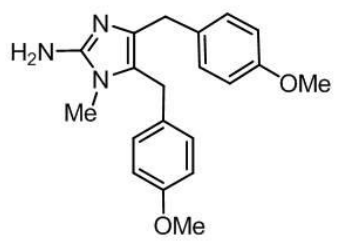


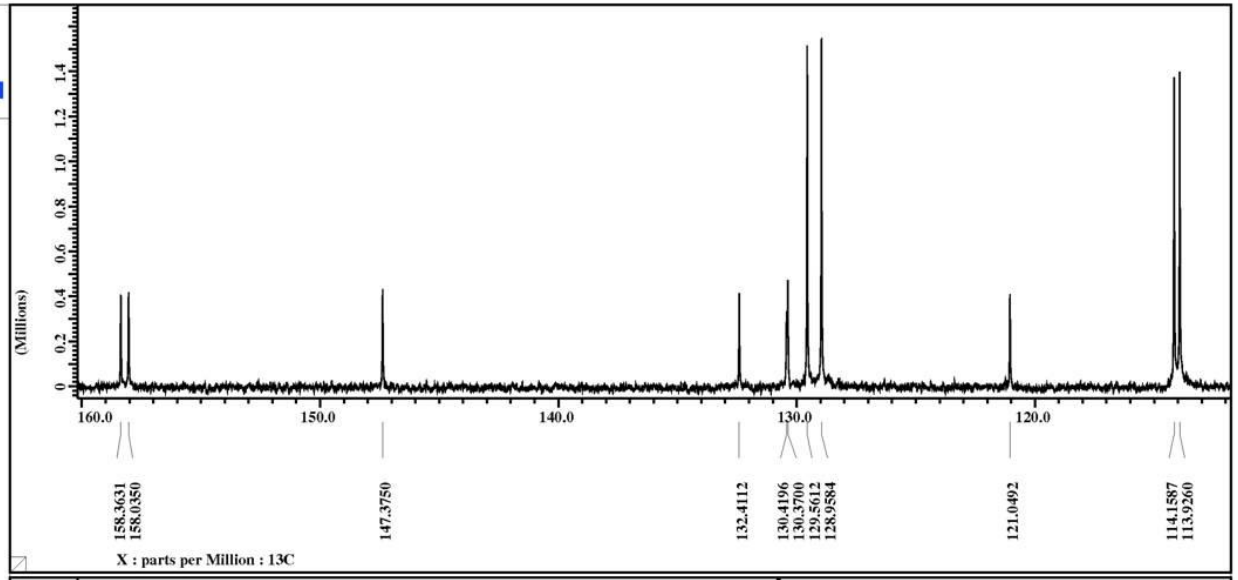


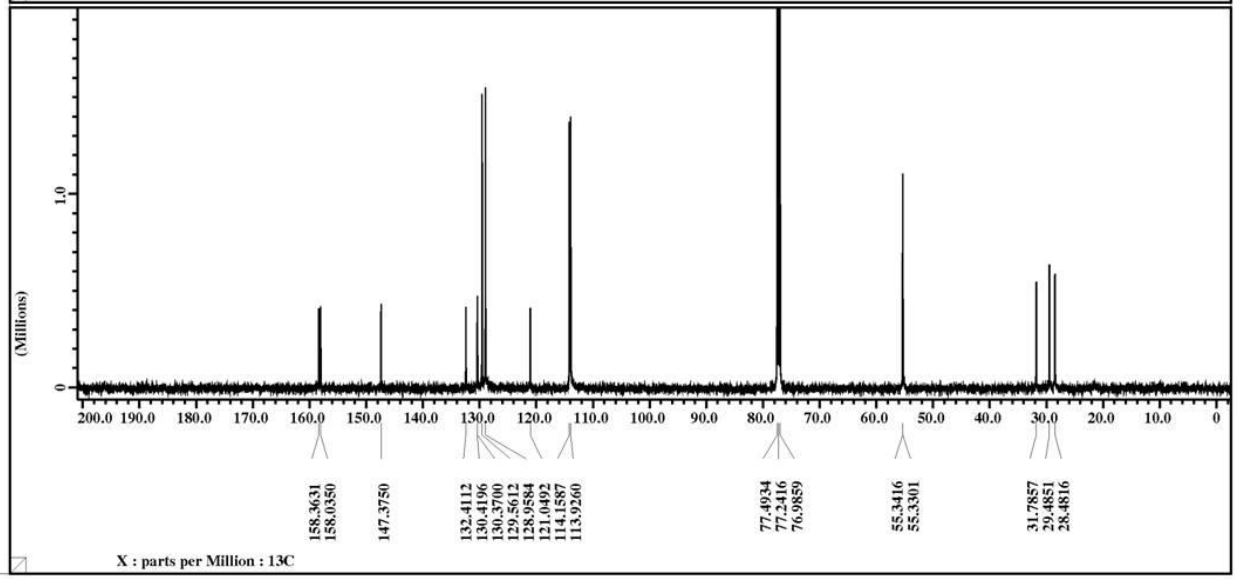


```
Filename          = V_p_092_amine-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#603540
Solvent           = CHLOROFORM-D
Creation_time     = 16-JAN-2009 01:09:08
Revision_time     = 19-MAR-2010 13:46:07
Current_time      = 19-MAR-2010 13:47:05

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 12
Relaxation_delay  = 4[s]
Temp_get          = 26[dC]
Unblank_time      = 2[us]
```

```
Filename          = V_p_092_amine-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#605054
Solvent           = CHLOROFORM-D
Creation_time     = 16-JAN-2009 02:22:24
Revision_time     = 15-JAN-2009 18:03:36
Current_time      = 19-MAR-2010 13:47:39

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 838
Total_scans       = 838

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.2[dC]
Unblank_time      = 2[us]
```





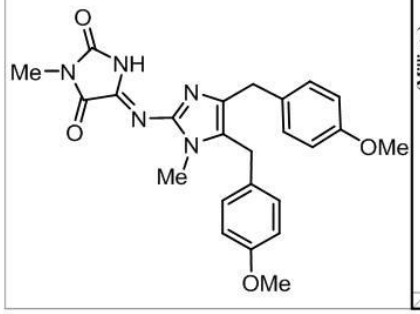


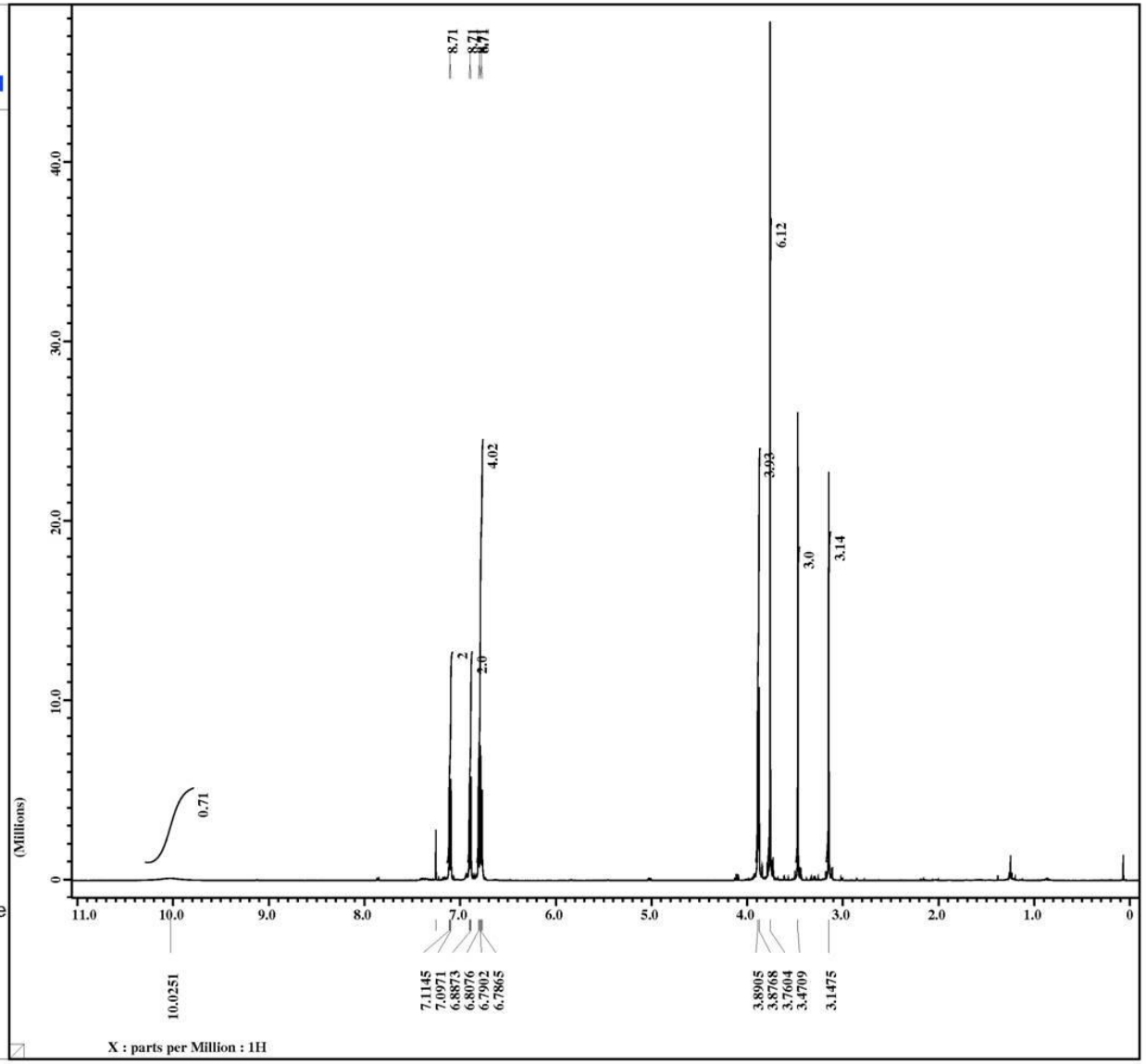
APPENDIX 82

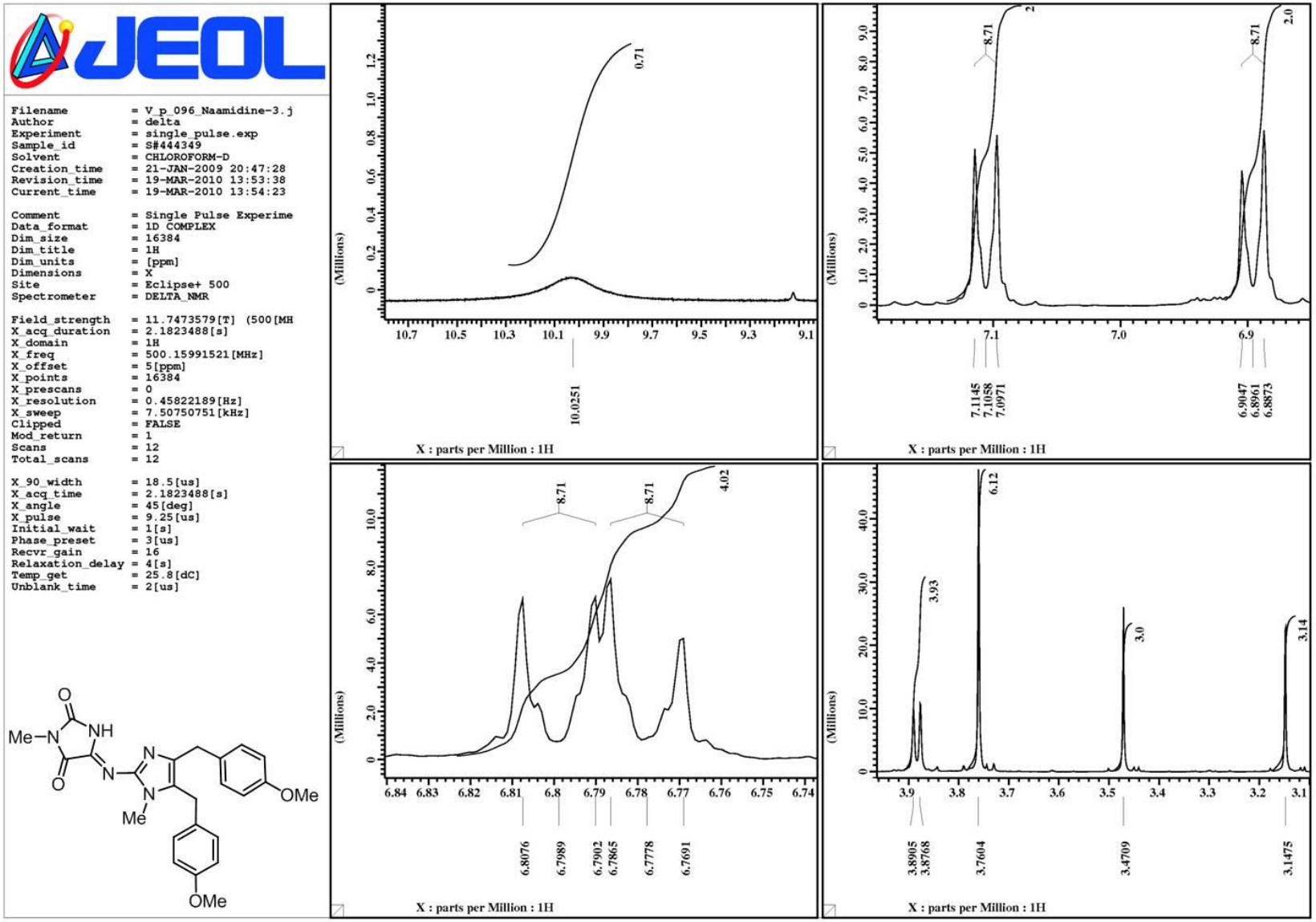
^{1}H and ^{13}C NMR Spectra of

4,5-Bis(4-methoxybenzyl)-1-methyl-2-(3-methylimidazolidine-2,4-dione)imino--1*H*-imidazole (**2f**): Naamidine G

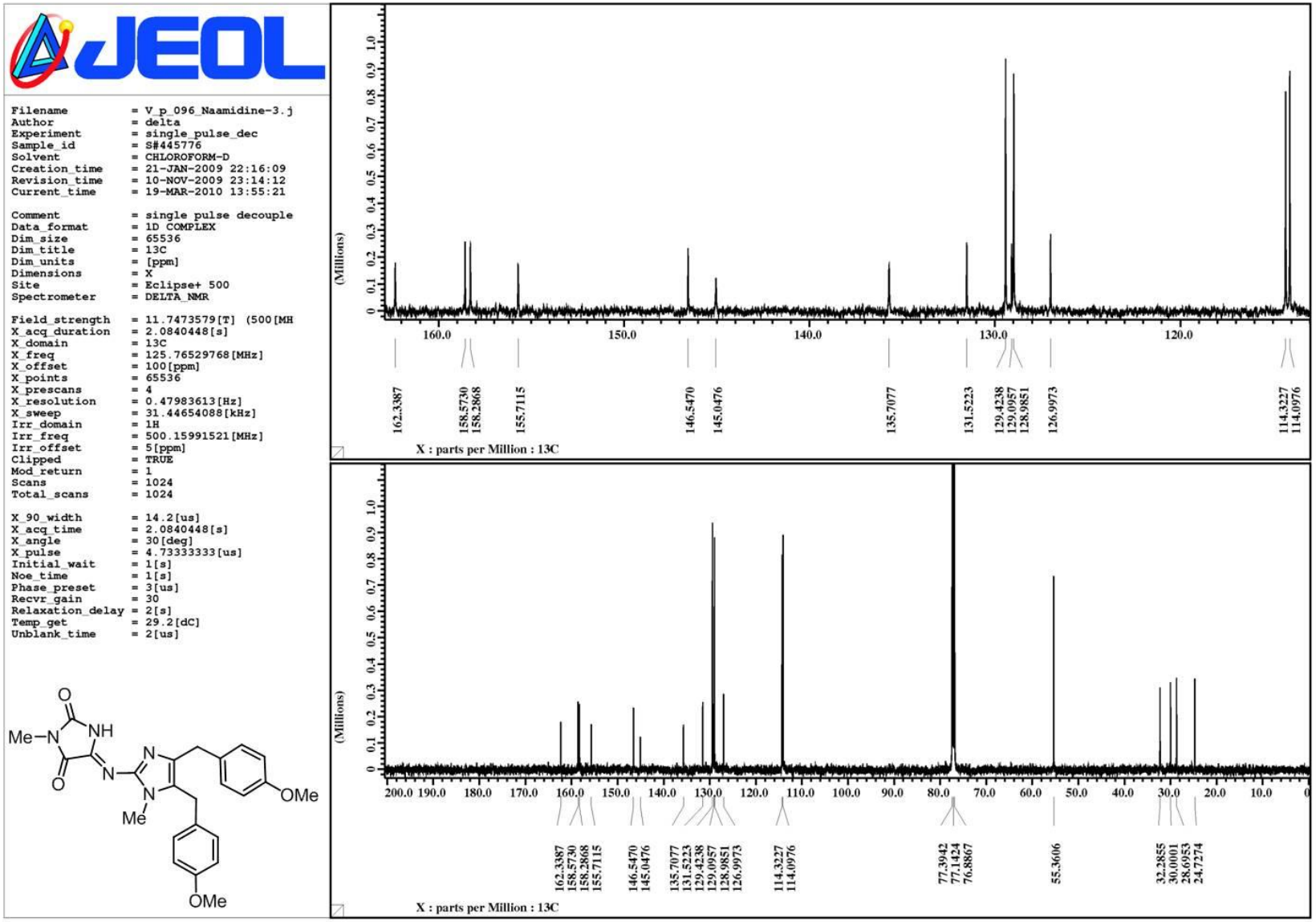
```
Filename          = V_p_096_Naamidine-3.j
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#444349
Solvent           = CHLOROFORM-D
Creation_time     = 21-JAN-2009 20:47:28
Revision_time     = 19-MAR-2010 13:53:02
Current_time      = 19-MAR-2010 13:53:10

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 16
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```







```
Filename          = V_p_096_Naamidine-3.j
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#444349
Solvent           = CHLOROFORM-D
Creation_time     = 21-JAN-2009 20:47:28
Revision_time     = 19-MAR-2010 13:53:38
Current_time      = 19-MAR-2010 13:54:23

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 16
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```



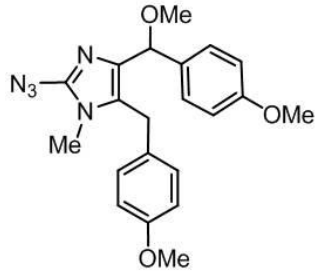
```
Filename          = V_p_096_Naamidine-3.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#445776
Solvent           = CHLOROFORM-D
Creation_time     = 21-JAN-2009 22:16:09
Revision_time     = 10-NOV-2009 23:14:12
Current_time      = 19-MAR-2010 13:55:21

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.2[dC]
Unblank_time      = 2[us]
```

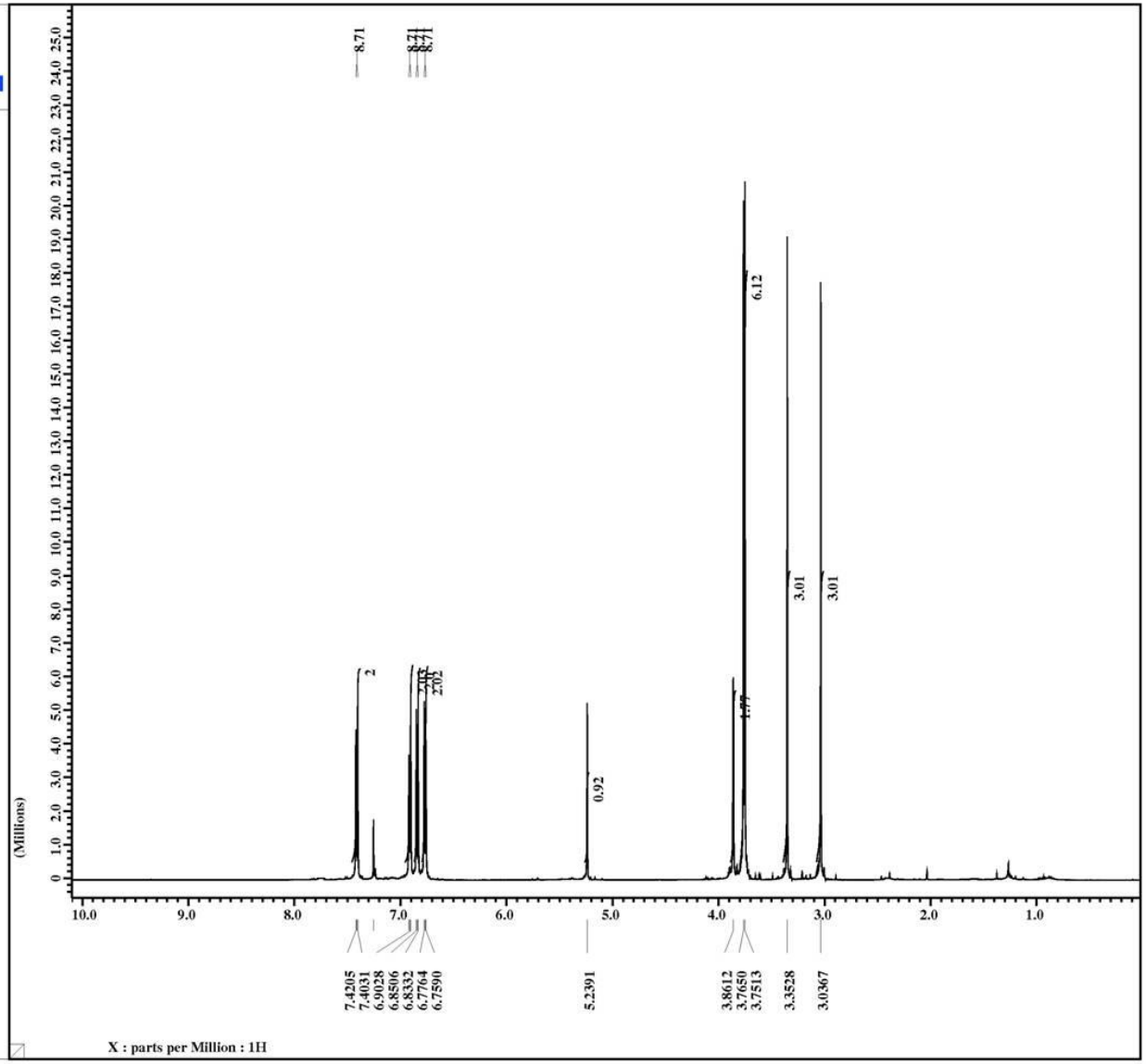
APPENDIX 83

^{1}H and ^{13}C NMR Spectra of

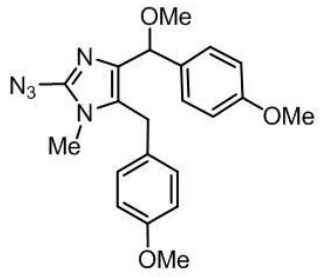
2-Azido-5-[4-methoxybenzyl]-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**201**)

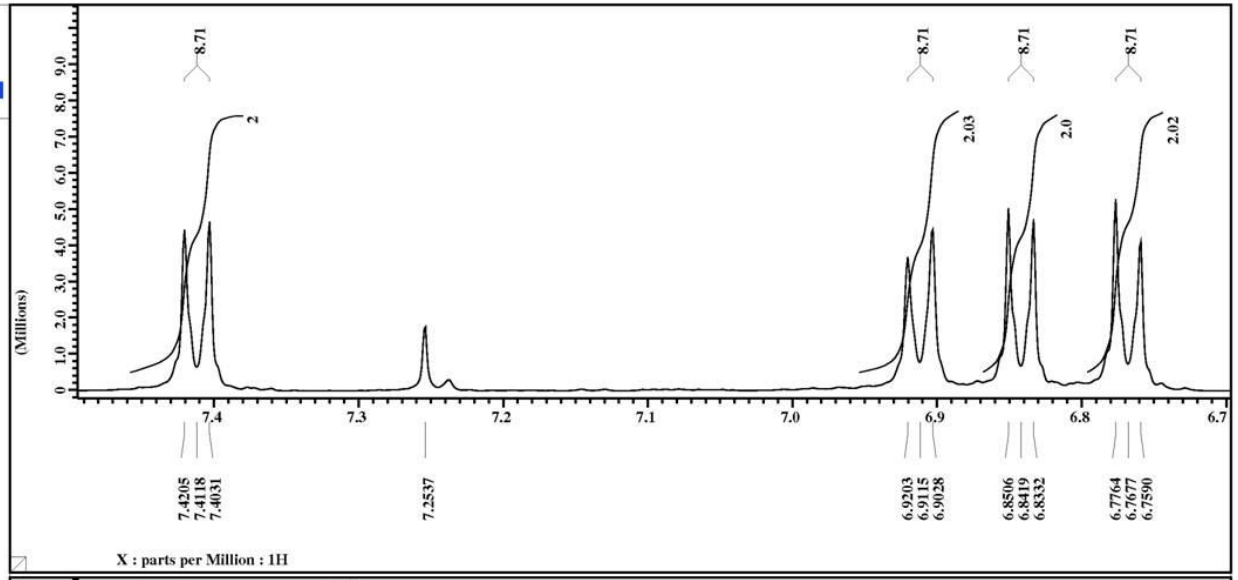
Filename	= V_p_036_Azide-4.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#624321
Solvent	= CHLOROFORM-D
Creation_time	= 27-NOV-2008 01:16:27
Revision_time	= 19-MAR-2010 14:30:42
Current_time	= 19-MAR-2010 14:30:51
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 12
Relaxation_delay	= 4[s]
Temp_get	= 25.5[dC]
Unblank_time	= 2[us]

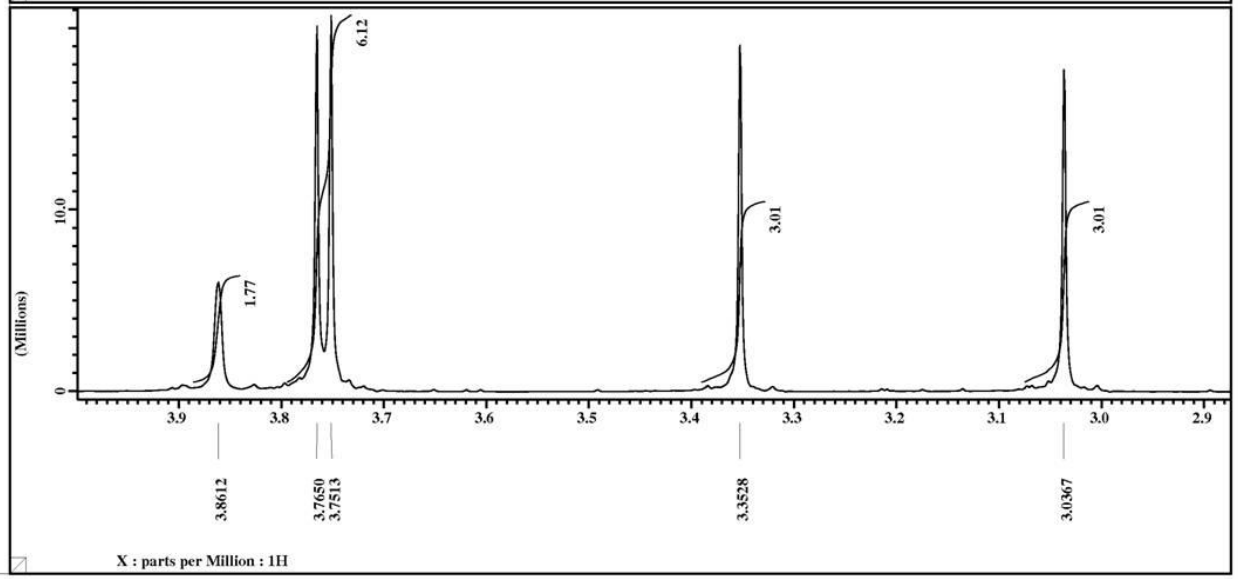




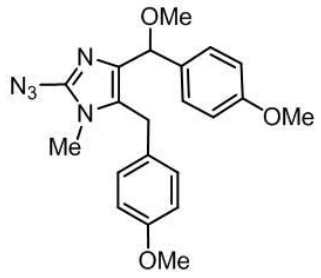
```
Filename          = V_p_036_Azide-4.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#624321
Solvent           = CHLOROFORM-D
Creation_time     = 27-NOV-2008 01:16:27
Revision_time     = 19-MAR-2010 14:30:42
Current_time      = 19-MAR-2010 14:31:47

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 12
Relaxation_delay  = 4[s]
Temp_get          = 25.5[dC]
Unblank_time      = 2[us]
```

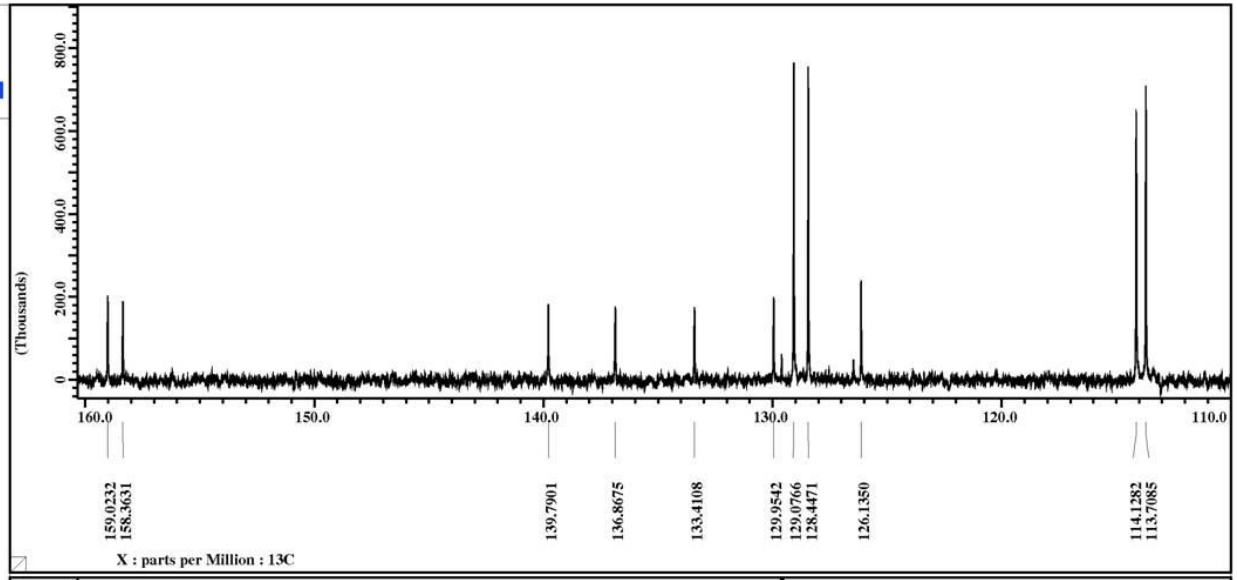


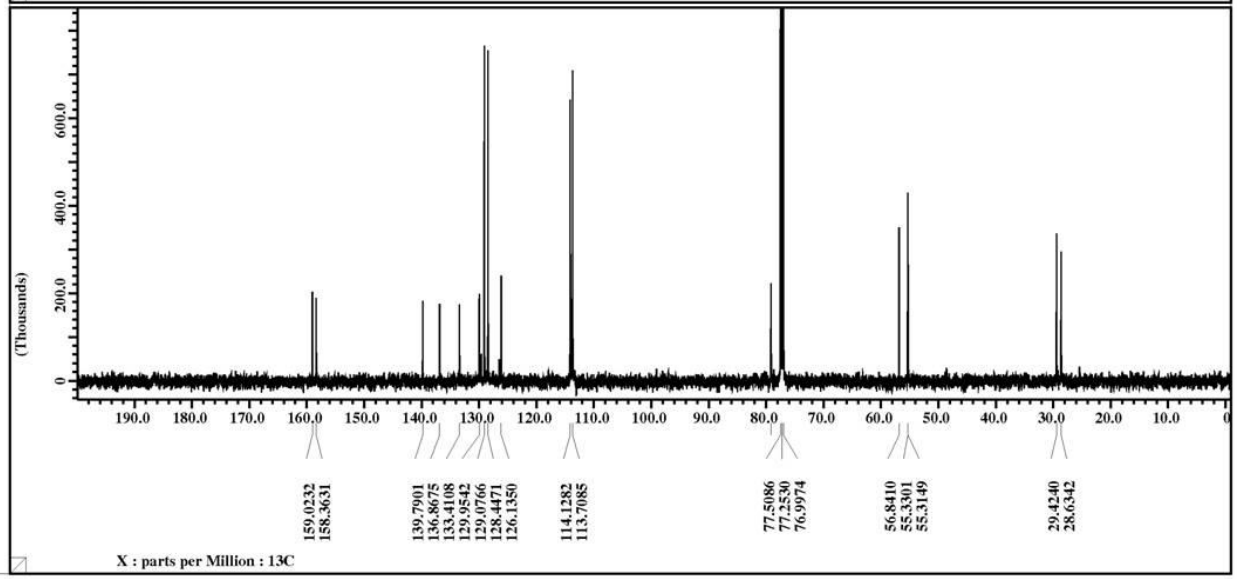




```
Filename          = V_p_036_Azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#626027
Solvent           = CHLOROFORM-D
Creation_time     = 27-NOV-2008 01:30:20
Revision_time     = 26-NOV-2008 17:37:22
Current_time      = 19-MAR-2010 14:32:54

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 136
Total_scans       = 136

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 26
Relaxation_delay  = 2[s]
Temp_get          = 28[dC]
Unblank_time      = 2[us]
```



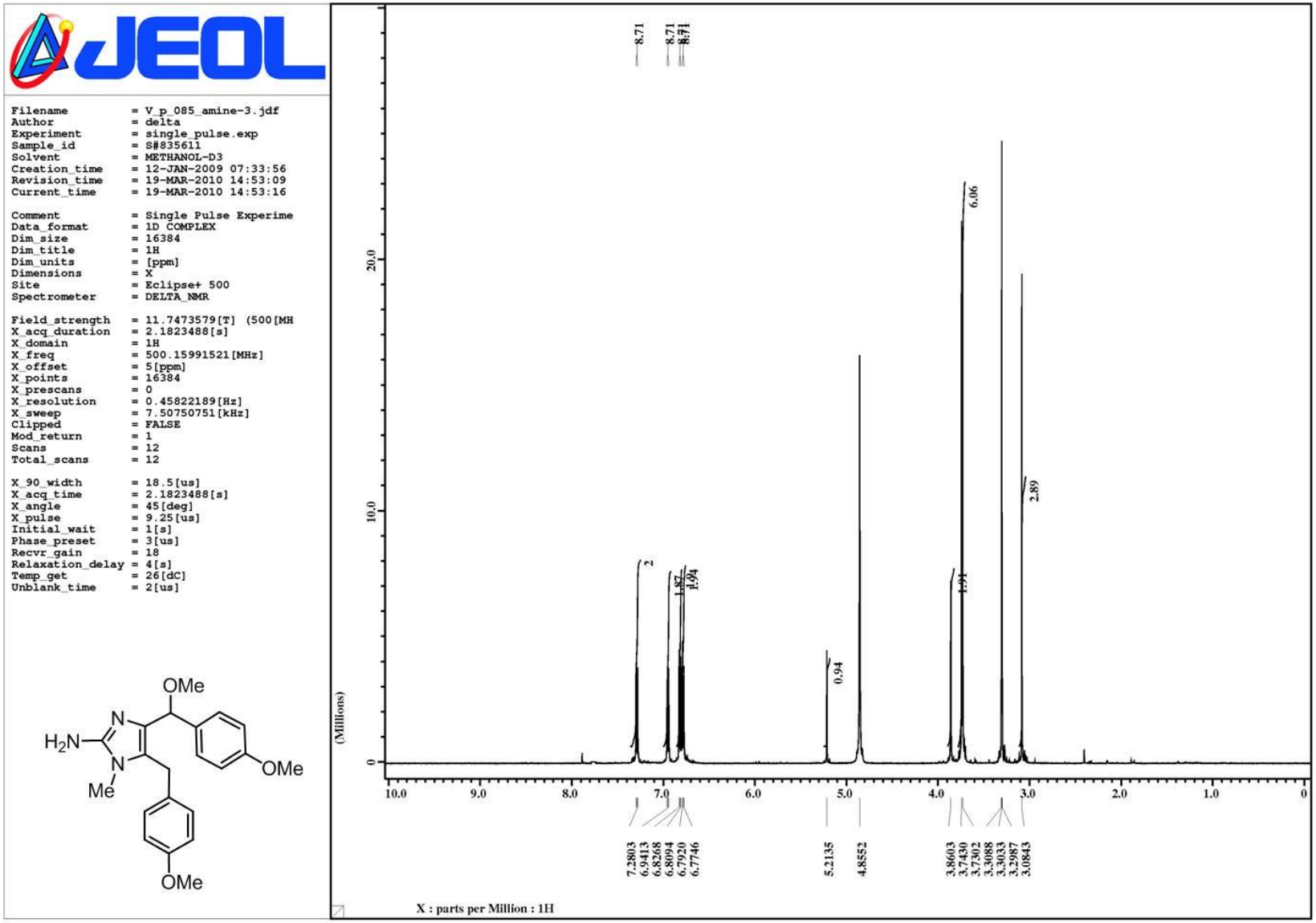




APPENDIX 84

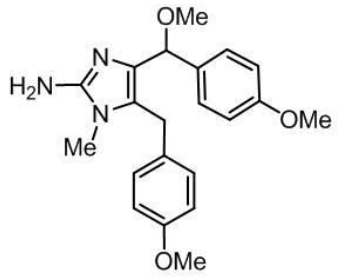
^{1}H and ^{13}C NMR Spectra of

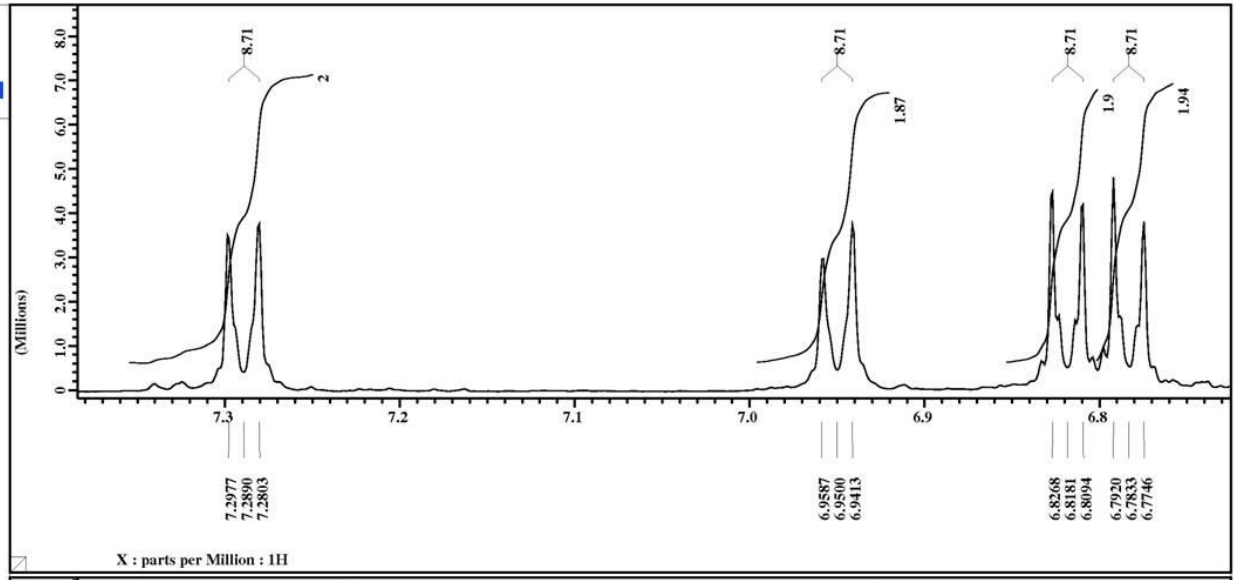
2-Amino-5-(4-methoxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole (**202**)

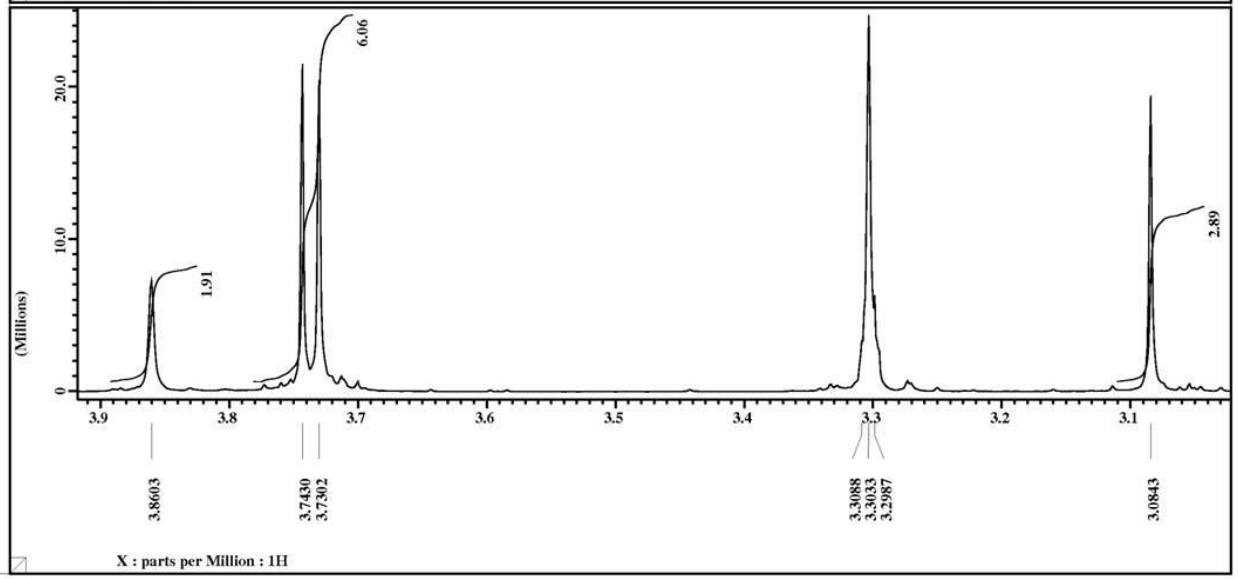


```
Filename          = V_p_085_amine-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#835611
Solvent           = METHANOL-D3
Creation_time     = 12-JAN-2009 07:33:56
Revision_time     = 19-MAR-2010 14:53:09
Current_time      = 19-MAR-2010 14:53:16

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 4[s]
Temp_get          = 26[dC]
Unblank_time      = 2[us]
```

```
Filename          = V_p_085_amine-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#835611
Solvent           = METHANOL-D3
Creation_time     = 12-JAN-2009 07:33:56
Revision_time     = 19-MAR-2010 14:53:09
Current_time      = 19-MAR-2010 14:53:45

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 4[s]
Temp_get          = 26[dC]
Unblank_time      = 2[us]
```







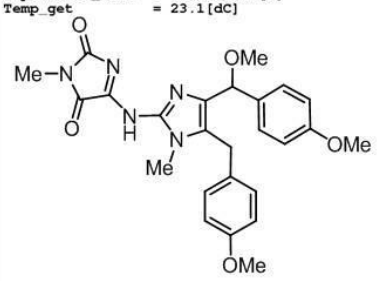
JEOL
Filename = V_p_037_amine-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#748994
Solvent = METHANOL-D3
Creation_time = 3-DEC-2008 10:26:10
Revision_time = 19-MAR-2010 14:49:13
Current_time = 19-MAR-2010 14:49:40
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 4000
Total_scans = 4000
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 29.1[dC]
Unblank_time = 2[us]
OMe
OMe
OMe
Me
H2N
(Thousands)
X : parts per Million : 13C
159.0041
158.3898
148.9507
133.8191
131.8389
130.7859
128.7447
127.9320
123.0217
113.6284
113.0980
78.5121
55.4064
54.3648
48.1993
47.6880
47.1768
28.0543
27.7186

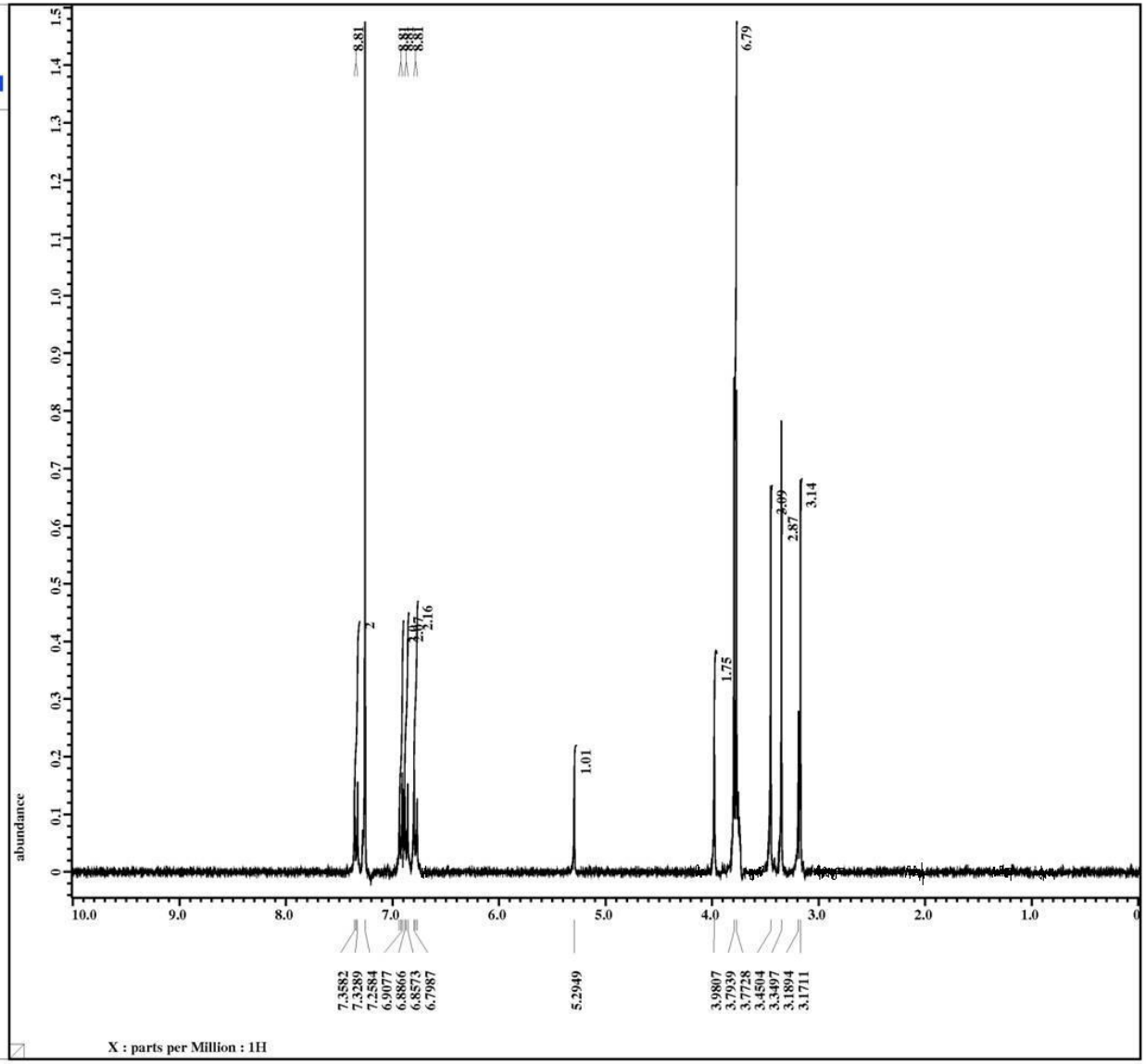
APPENDIX 85

^{1}H NMR Spectrum of

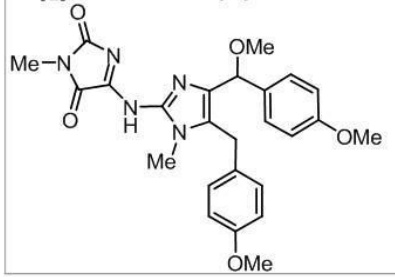
5-(4-Methoxybenzyl)-4-[methoxy-(4-methoxyphenyl)]methyl-1-methyl-2-(3-methylimidazolidine-2,4-dione)imino-1*H*-imidazole (**4d**): 4-Methoxynaamidine G

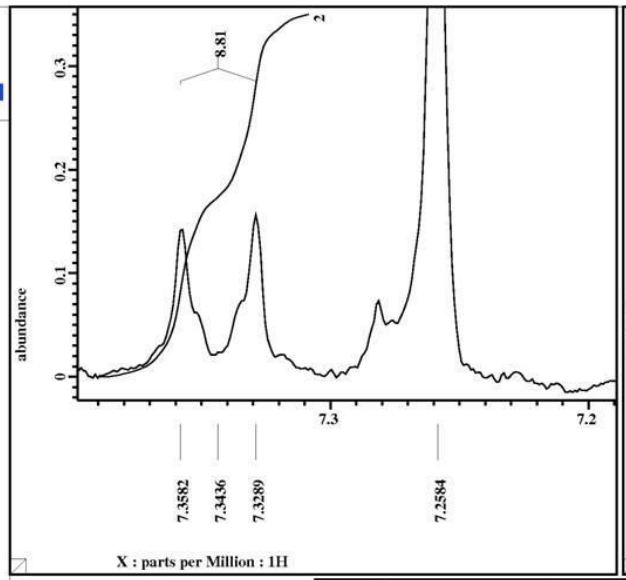
```
Filename          = V_p_098_i_14-methoxyn
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#584864
Solvent           = CHLOROFORM-D
Creation_time     = 27-JAN-2009 16:36:42
Revision_time     = 19-MAR-2010 15:03:19
Current_time      = 19-MAR-2010 15:03:38

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```

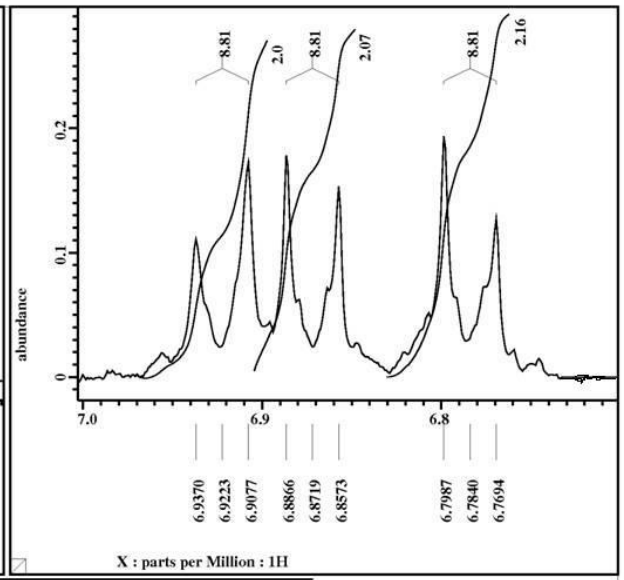


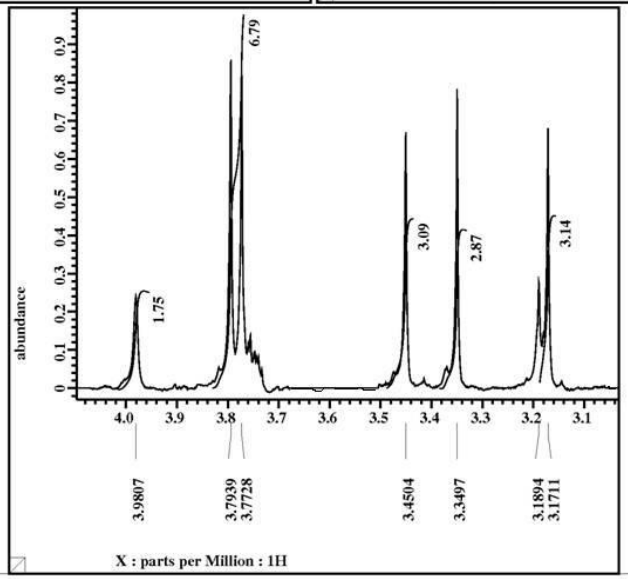


Filename	= V_p_098_i_14-methoxyn
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#584864
Solvent	= CHLOROFORM-D
Creation_time	= 27-JAN-2009 16:36:42
Revision_time	= 19-MAR-2010 15:04:22
Current_time	= 19-MAR-2010 15:04:57
Comment	= single_pulse
Data_format	= 1D REAL
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 50
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.1[dC]









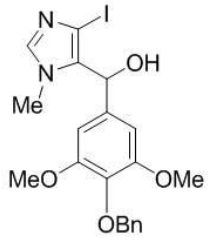
APPENDIX 86

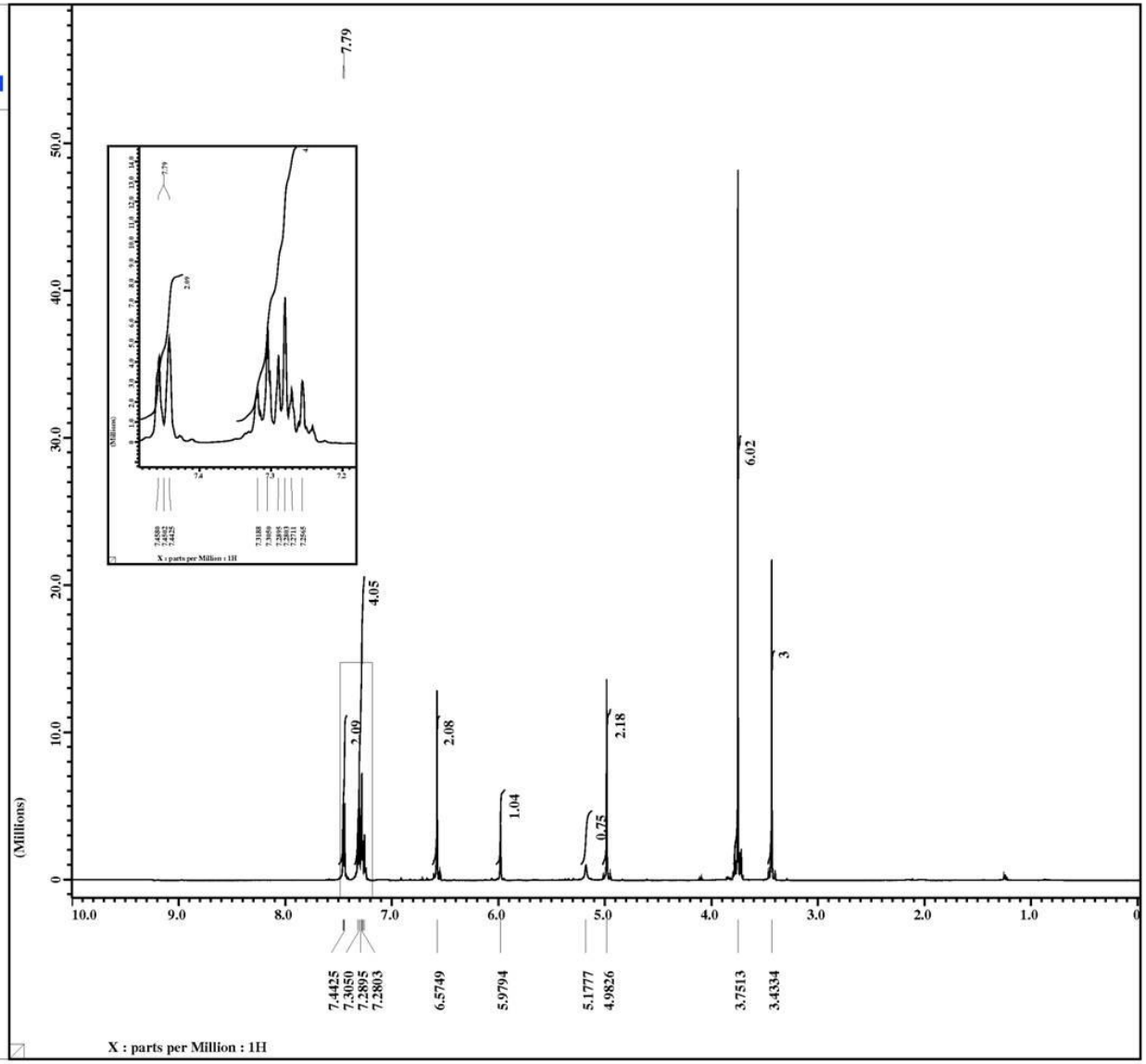
^{1}H and ^{13}C NMR Spectra of

5-(4-Benzyloxy-3,5-dimethoxyphenyl)hydroxymethyl-4-iodo-1-methyl-1*H*-imidazole

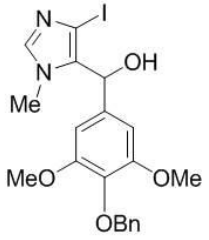
(**204**)

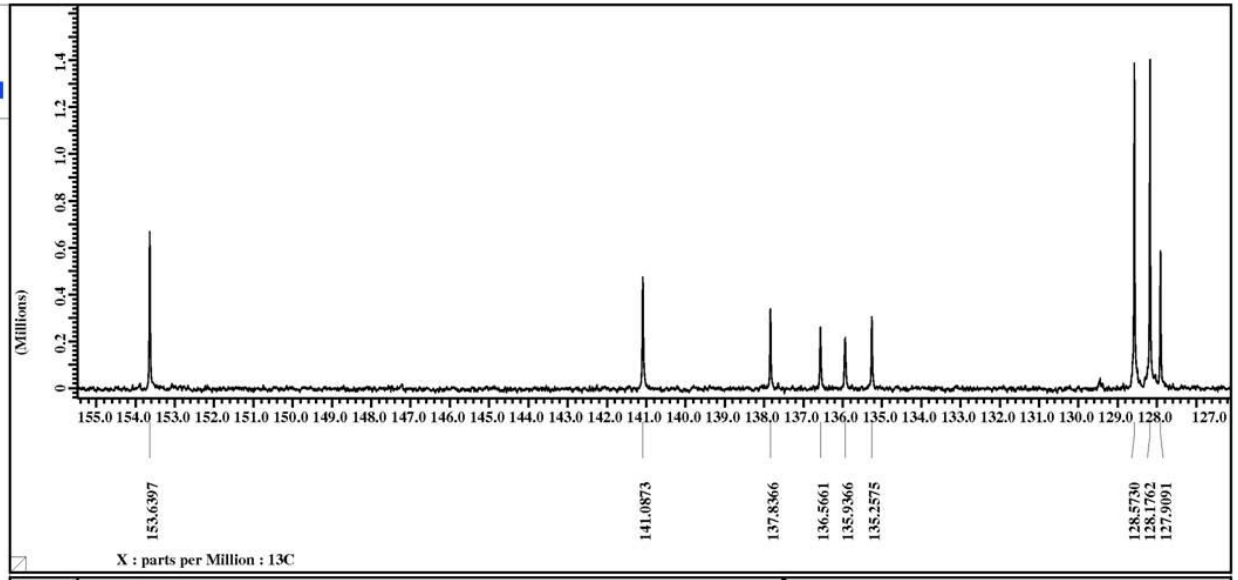
Filename	= V_p_127_alcohol-4.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#754188
Solvent	= CHLOROFORM-D
Creation_time	= 6-MAR-2009 06:22:58
Revision_time	= 19-MAR-2010 15:42:48
Current_time	= 19-MAR-2010 15:43:30
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 16
Relaxation_delay	= 4[s]
Temp_get	= 25.9[dC]
Unblank_time	= 2[us]

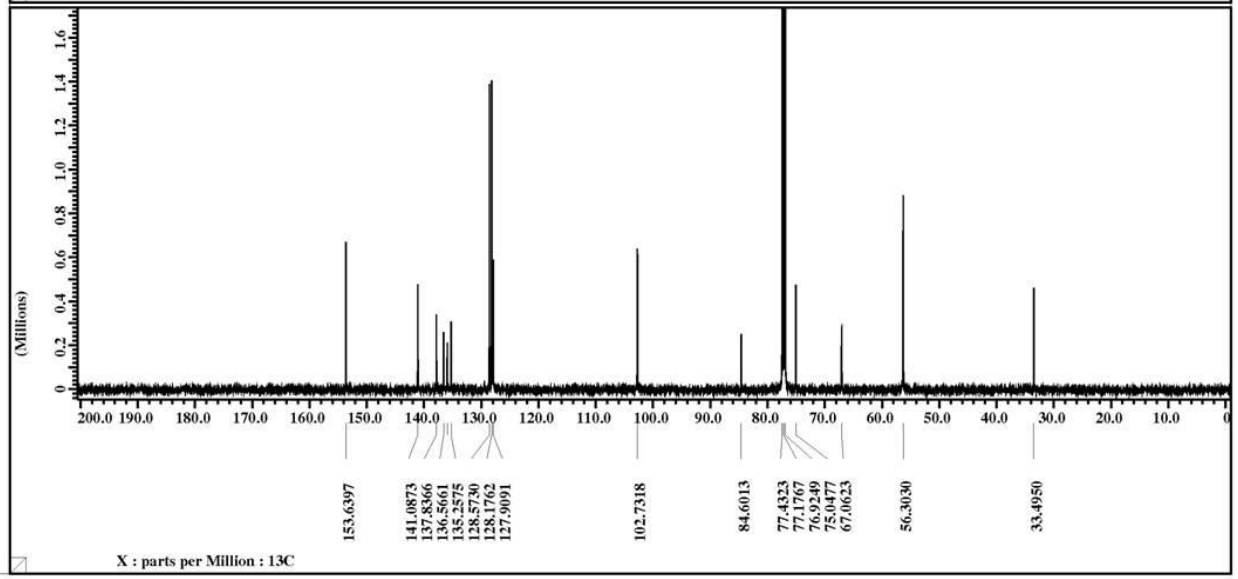




```
Filename          = V_p_127_alcohol-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#759557
Solvent           = CHLOROFORM-D
Creation_time     =  6-MAR-2009 09:31:40
Revision_time     =  8-MAR-2009 19:29:32
Current_time      = 19-MAR-2010 15:45:19

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 2135
Total_scans       = 2135

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.1[dC]
Unblank_time      = 2[us]
```



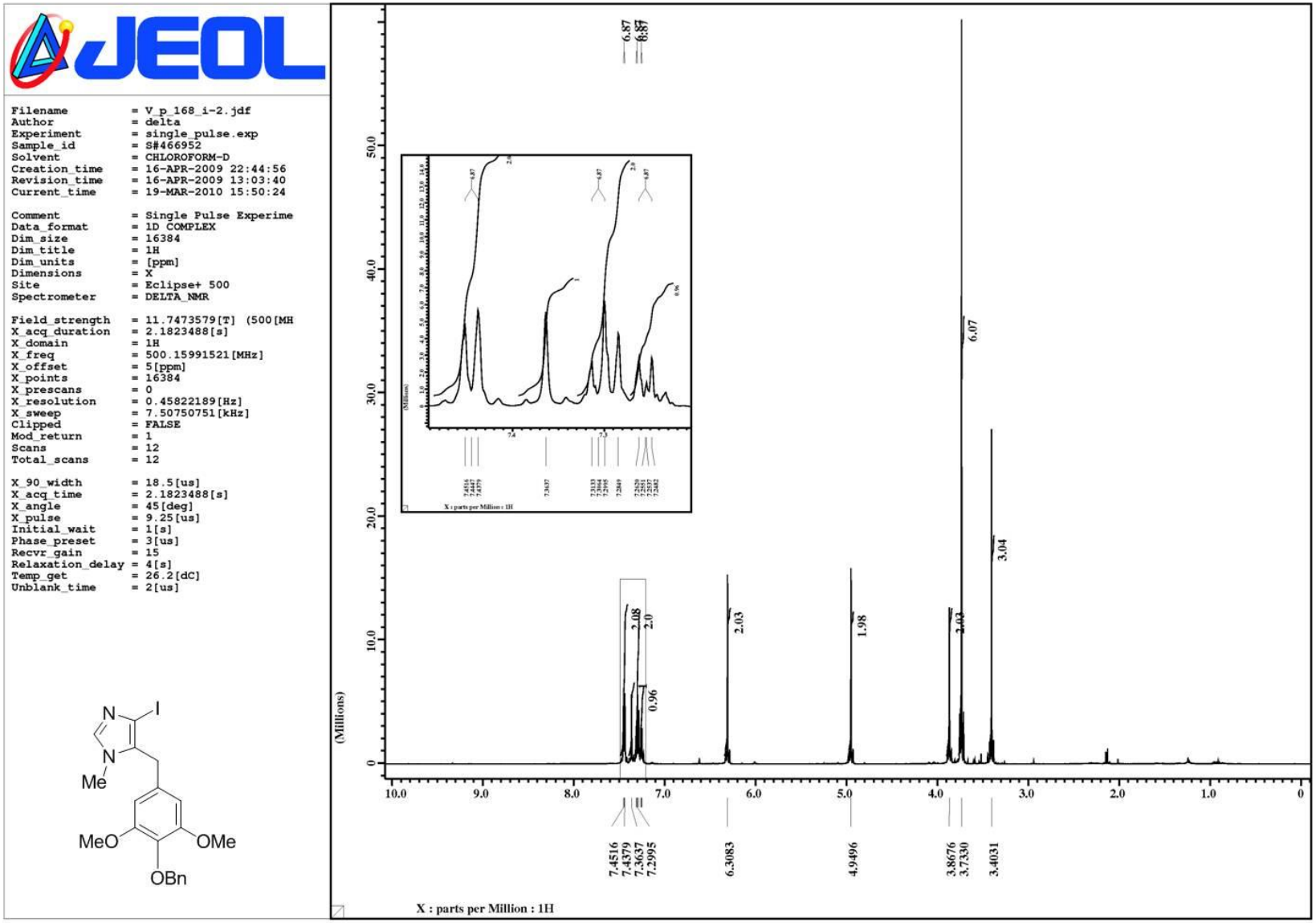




APPENDIX 87

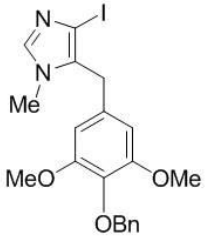
^{1}H and ^{13}C NMR Spectra of

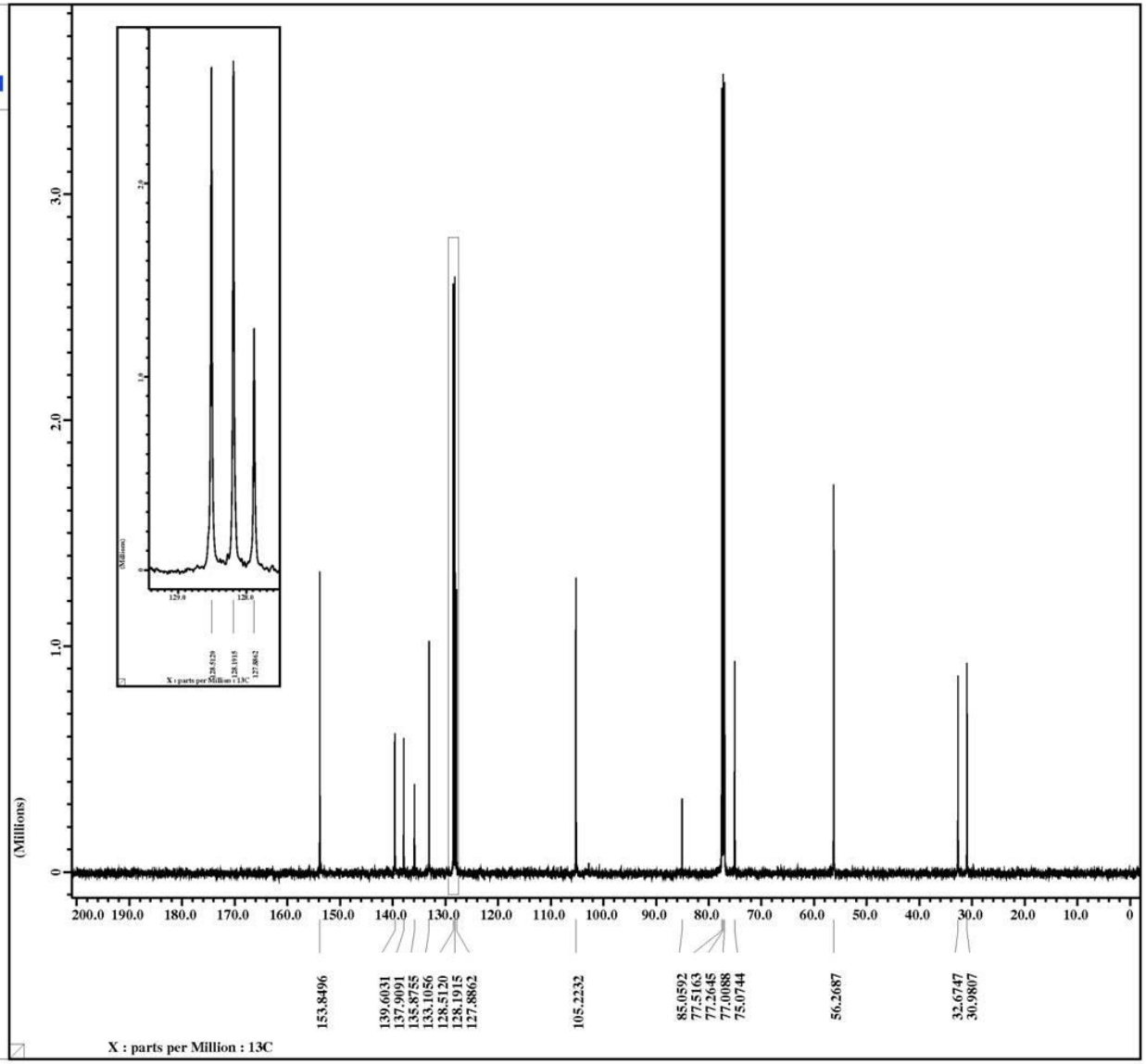
5-(4-Benzyloxy-3,5-dimethoxybenzyl)-4-iodo-1-methyl-1*H*-imidazole (**205a**)



```
Filename          = V_p_168_i-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#466952
Solvent           = CHLOROFORM-D
Creation_time     = 16-APR-2009 22:44:56
Revision_time     = 16-APR-2009 13:03:40
Current_time      = 19-MAR-2010 15:50:24

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 26.2[dC]
Unblank_time      = 2[us]
```

```
Filename          = V_p_168_i-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#468144
Solvent           = CHLOROFORM-D
Creation_time     = 17-APR-2009 00:15:23
Revision_time     = 19-MAR-2010 15:52:04
Current_time      = 19-MAR-2010 15:52:45

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T]  (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.3[dC]
Unblank_time      = 2[us]
```



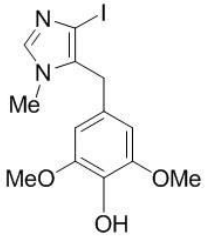


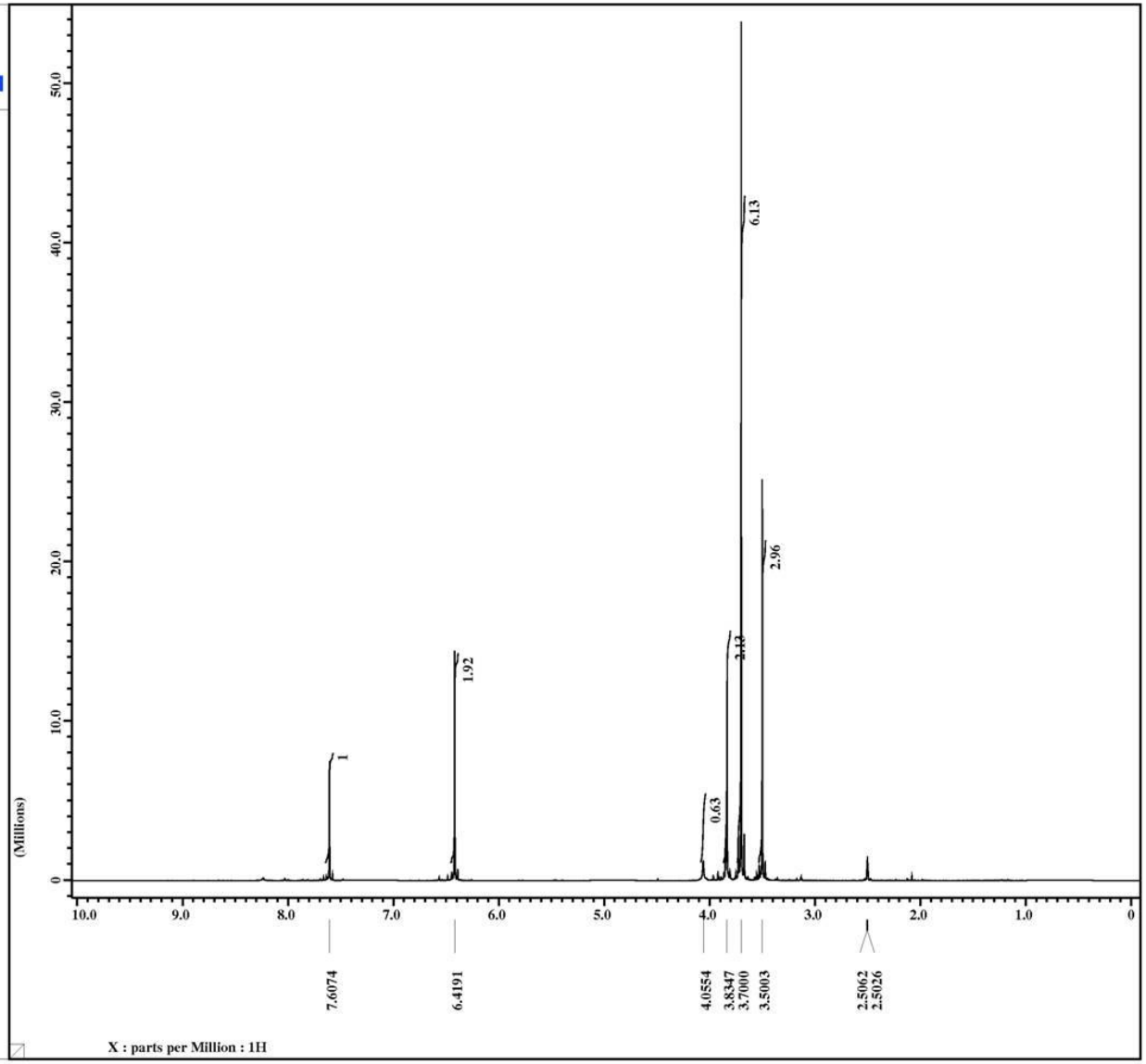
APPENDIX 88

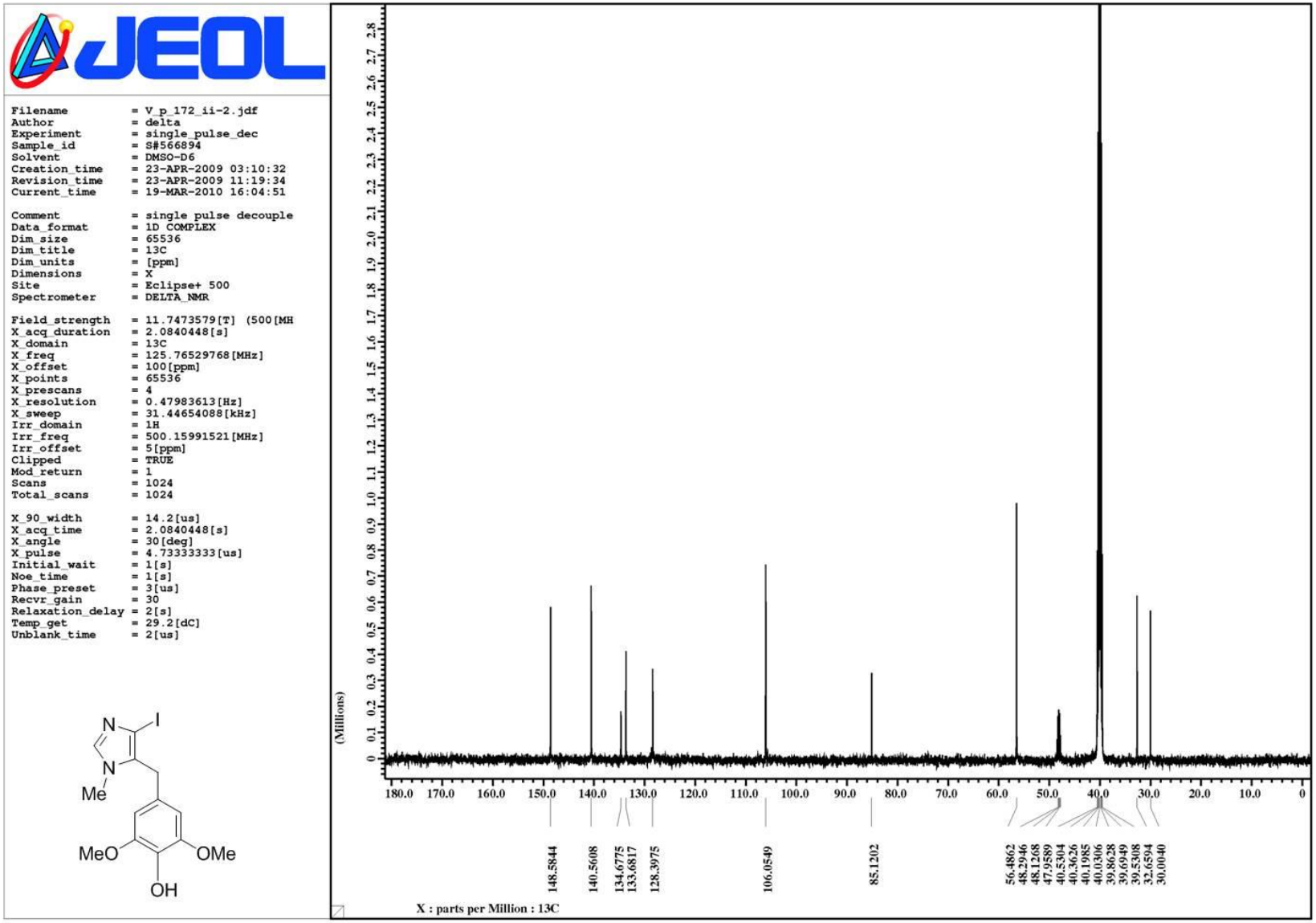
^{1}H and ^{13}C NMR Spectra of

5-[(3,5-Dimethoxy-4-hydroxy)benzyl]-4-iodo-1-methyl-1*H*-imidazole (**205b**)

```
Filename          = V_p_172_ii-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#565697
Solvent           = DMSO-D6
Creation_time     = 23-APR-2009 01:41:45
Revision_time     = 22-APR-2009 15:47:35
Current_time      = 19-MAR-2010 16:02:38

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 26.3[dC]
Unblank_time      = 2[us]
```





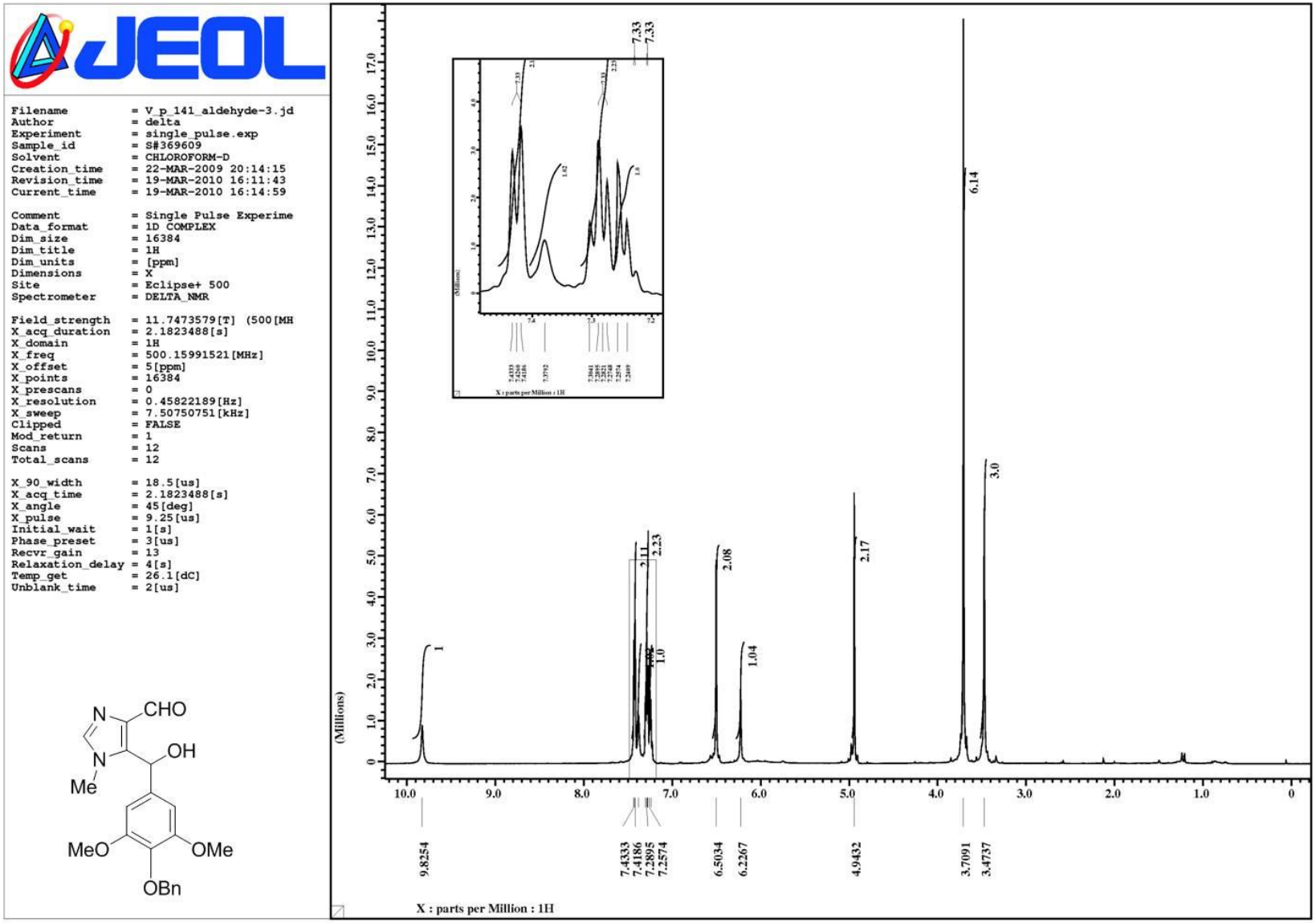


```
Filename          = V_p_172_ii-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#566894
Solvent           = DMSO-D6
Creation_time     = 23-APR-2009 03:10:32
Revision_time     = 23-APR-2009 11:19:34
Current_time      = 19-MAR-2010 16:04:51

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.2[dC]
Unblank_time      = 2[us]
```

APPENDIX 89

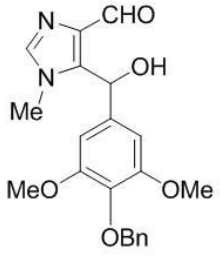
^{1}H and ^{13}C NMR Spectra of

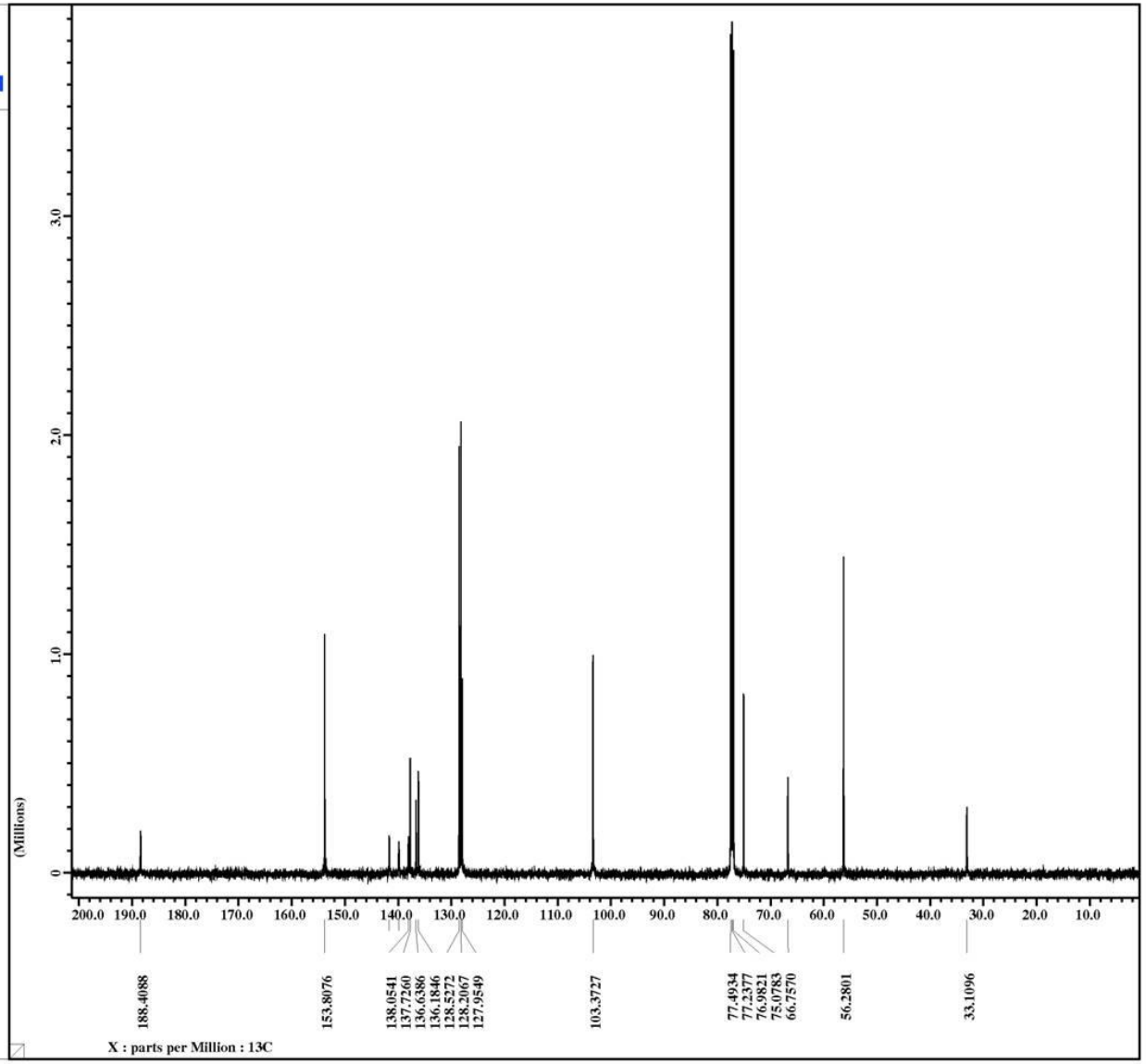
5-(4-Benzyloxy-3,5-dimethoxyphenyl)hydroxymethyl-1-methyl-1*H*-imidazole-4-carbaldehyde (**207**)



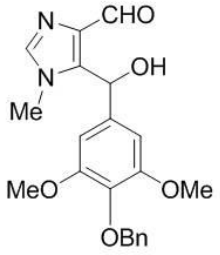
```
Filename          = V_p_141_aldehyde-3.jd
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#369609
Solvent           = CHLOROFORM-D
Creation_time     = 22-MAR-2009 20:14:15
Revision_time     = 19-MAR-2010 16:11:43
Current_time      = 19-MAR-2010 16:14:59

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 13
Relaxation_delay  = 4[s]
Temp_get          = 26.1[dC]
Unblank_time      = 2[us]
```

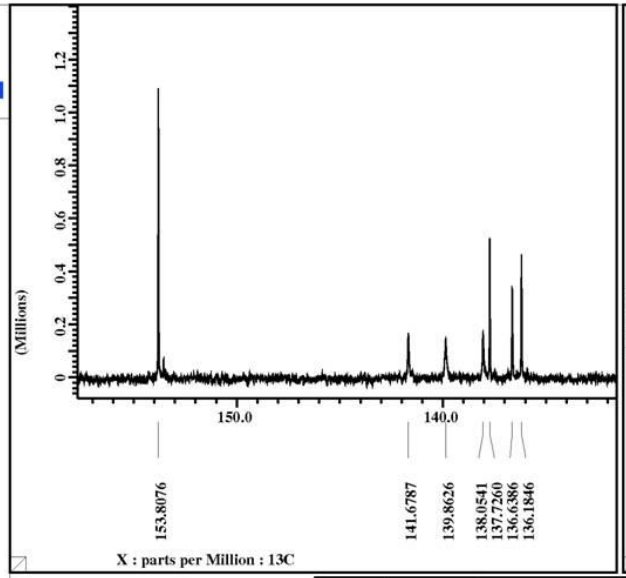
```
Filename          = V_p_141_aldehyde-2.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#371154
Solvent           = CHLOROFORM-D
Creation_time     = 22-MAR-2009 21:43:12
Revision_time     = 22-MAR-2009 14:20:59
Current_time      = 19-MAR-2010 16:17:07

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.2[dC]
Unblank_time      = 2[us]
```

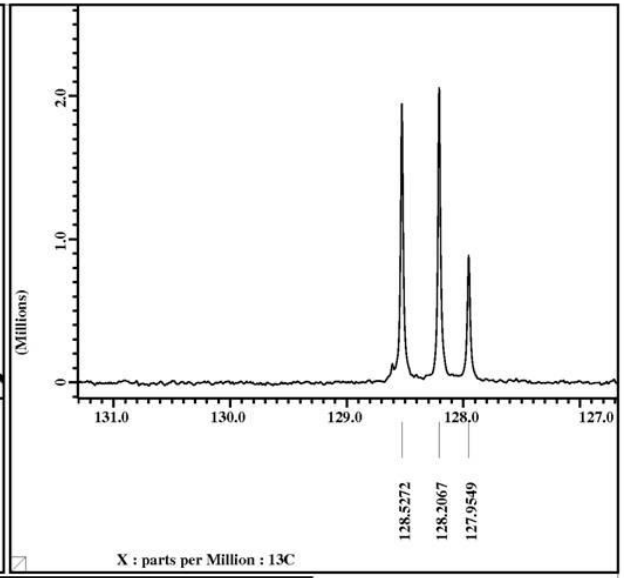


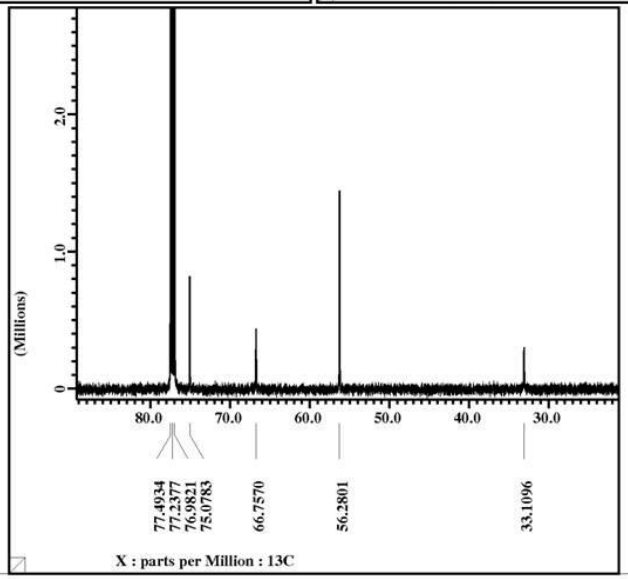


```
Filename          = V_p_141_aldehyde-2.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#371154
Solvent           = CHLOROFORM-D
Creation_time     = 22-MAR-2009 21:43:12
Revision_time     = 22-MAR-2009 14:20:59
Current_time      = 19-MAR-2010 16:18:05

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 1024
Total_scans       = 1024

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.2[dC]
Unblank_time      = 2[us]
```







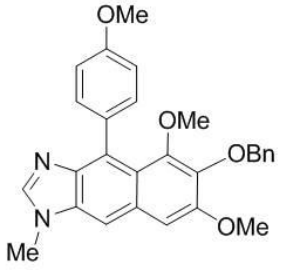


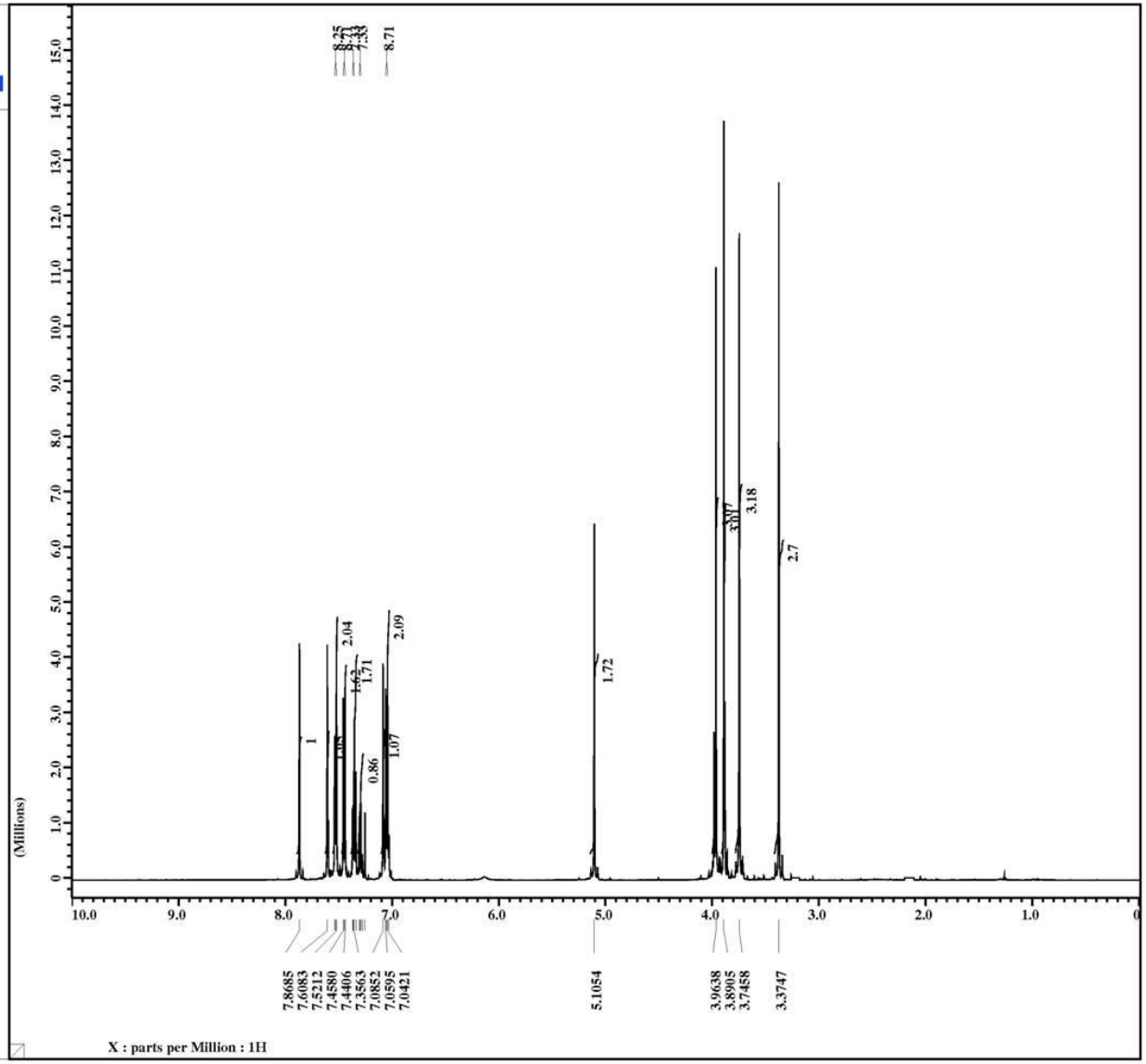
APPENDIX 90

^{1}H and ^{13}C NMR Spectra of

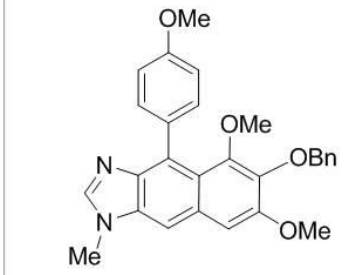
6-Benzyloxy-5,7-dimethoxy-4-(4-methoxyphenyl)-1-methyl-1*H*-naphtho[2,3-*d*]imidazole (**209**)

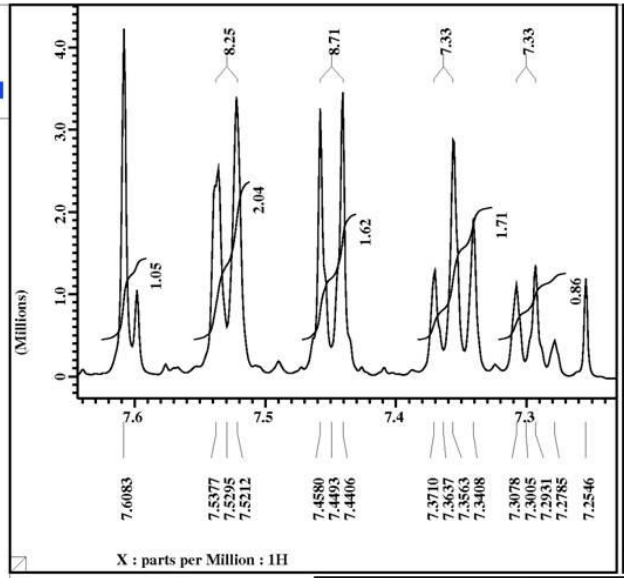
Filename	= V_p_147-4.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#502180
Solvent	= CHLOROFORM-D
Creation_time	= 29-MAR-2009 00:07:16
Revision_time	= 19-MAR-2010 16:40:07
Current_time	= 19-MAR-2010 16:40:22
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 11
Relaxation_delay	= 4[s]
Temp_get	= 26.2[dC]
Unblank_time	= 2[us]

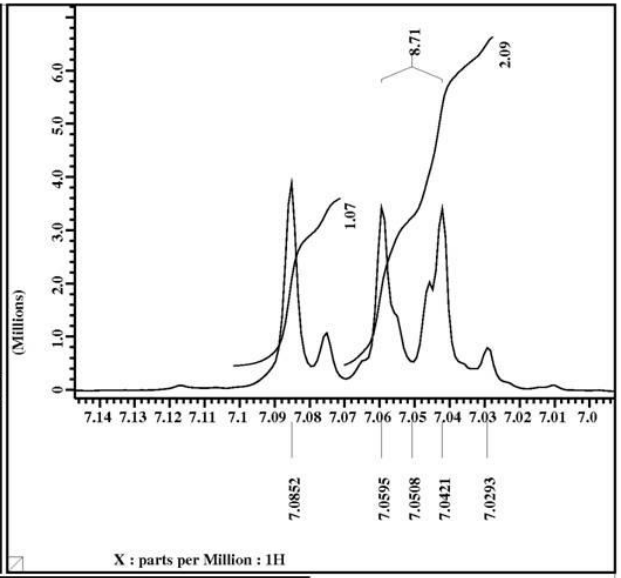


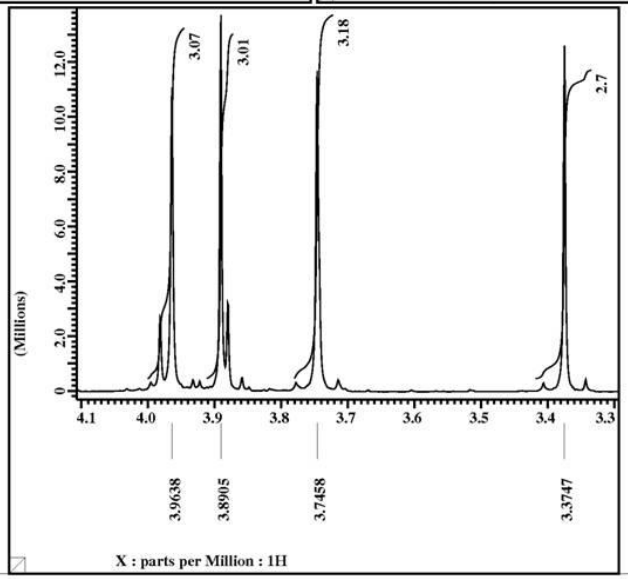


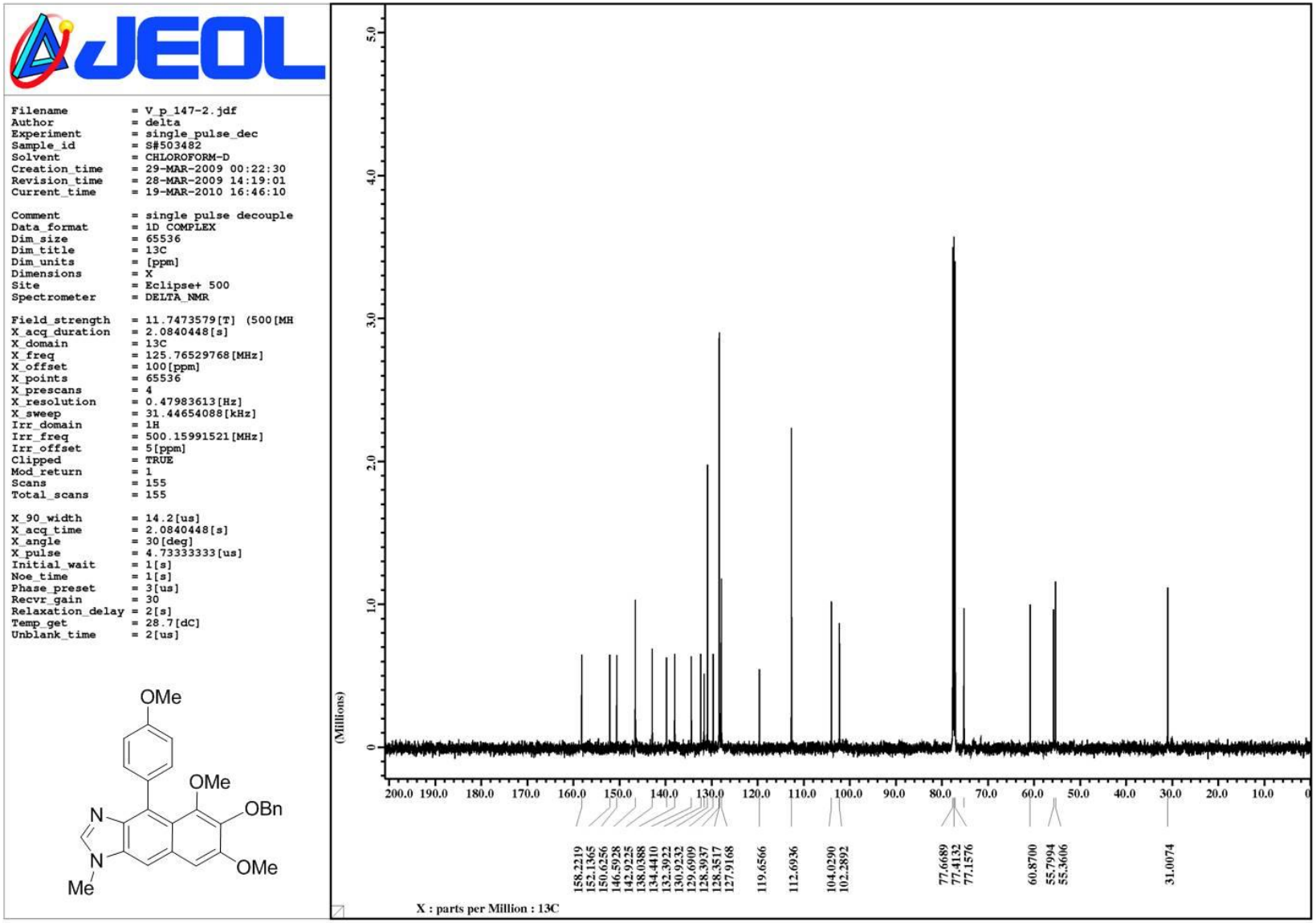
```
Filename          = V_p_147-4.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#502180
Solvent           = CHLOROFORM-D
Creation_time     = 29-MAR-2009 00:07:16
Revision_time     = 19-MAR-2010 16:40:07
Current_time      = 19-MAR-2010 16:41:13

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 11
Relaxation_delay  = 4[s]
Temp_get          = 26.2[dC]
Unblank_time      = 2[us]
```









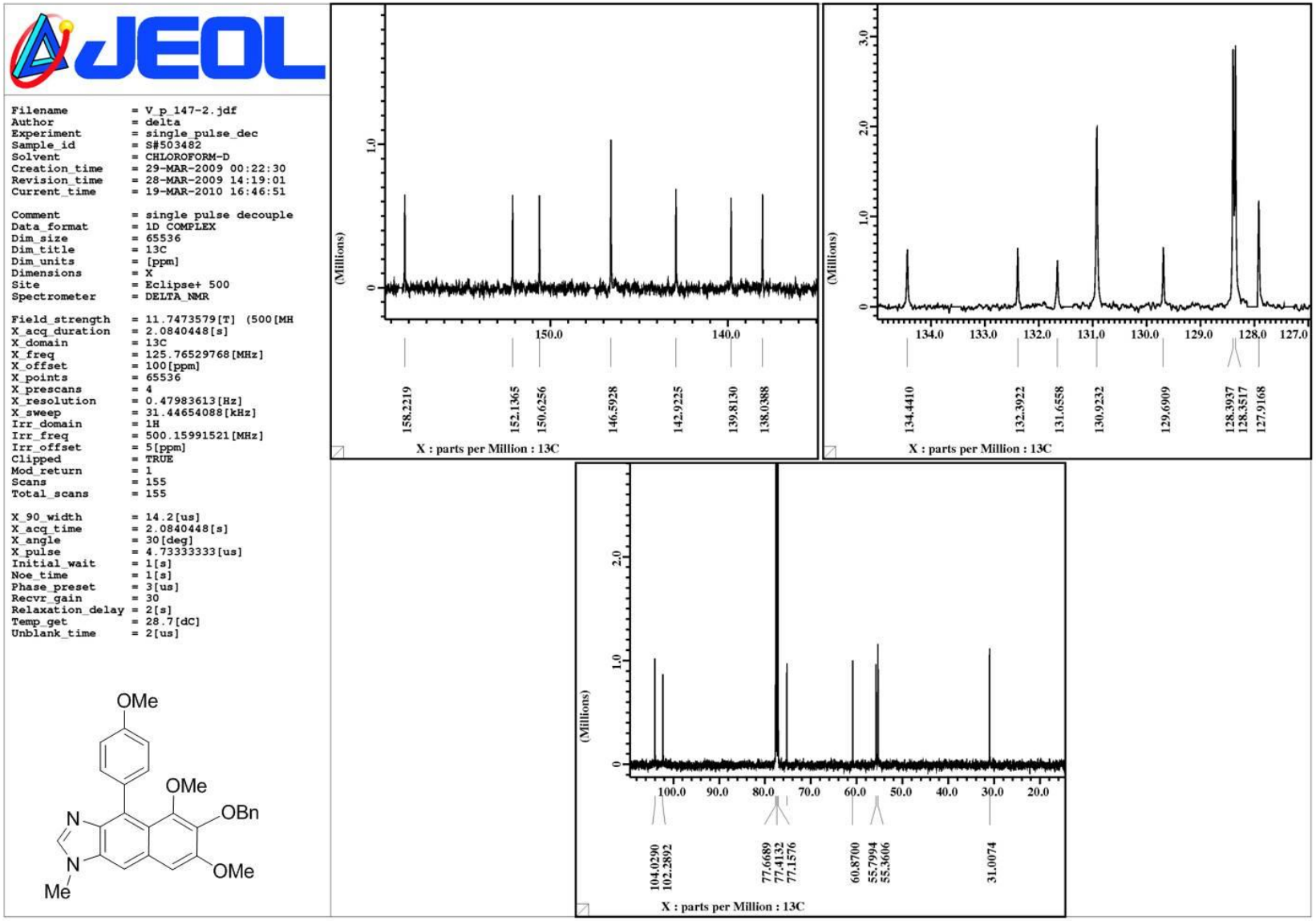


Filename = V_p_147-2.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#503482
Solvent = CHLOROFORM-D
Creation_time = 29-MAR-2009 00:22:30
Revision_time = 28-MAR-2009 14:19:01
Current_time = 19-MAR-2010 16:46:10

Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR

Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 155
Total_scans = 155

X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 28.7[dC]
Unblank_time = 2[us]



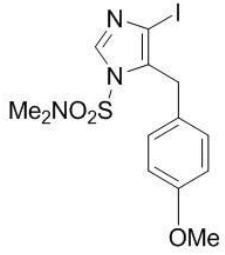
```
Filename          = V_p_147-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#503482
Solvent           = CHLOROFORM-D
Creation_time     = 29-MAR-2009 00:22:30
Revision_time     = 28-MAR-2009 14:19:01
Current_time      = 19-MAR-2010 16:46:51

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 155
Total_scans       = 155

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.7[dC]
Unblank_time      = 2[us]
```

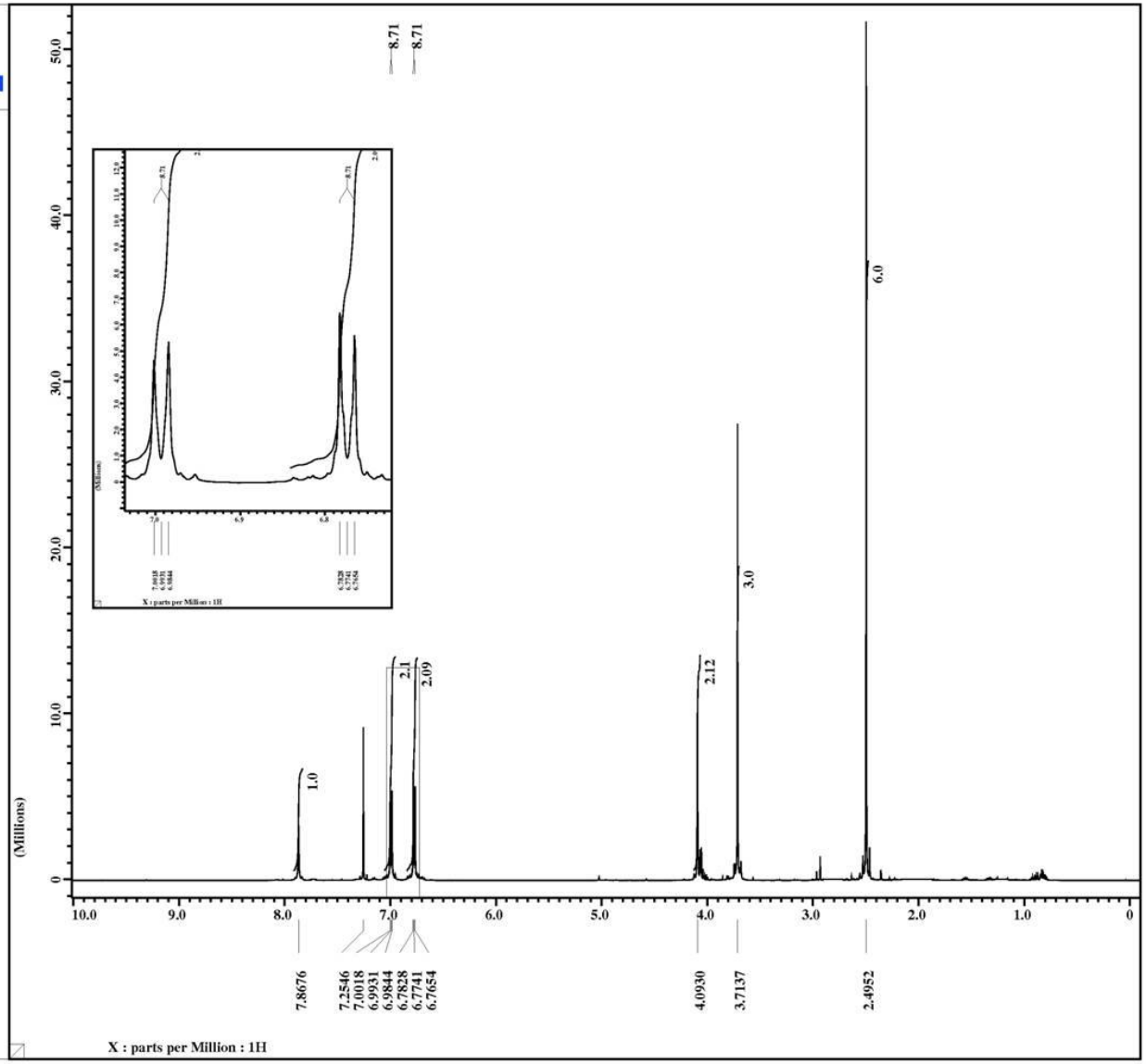
APPENDIX 91

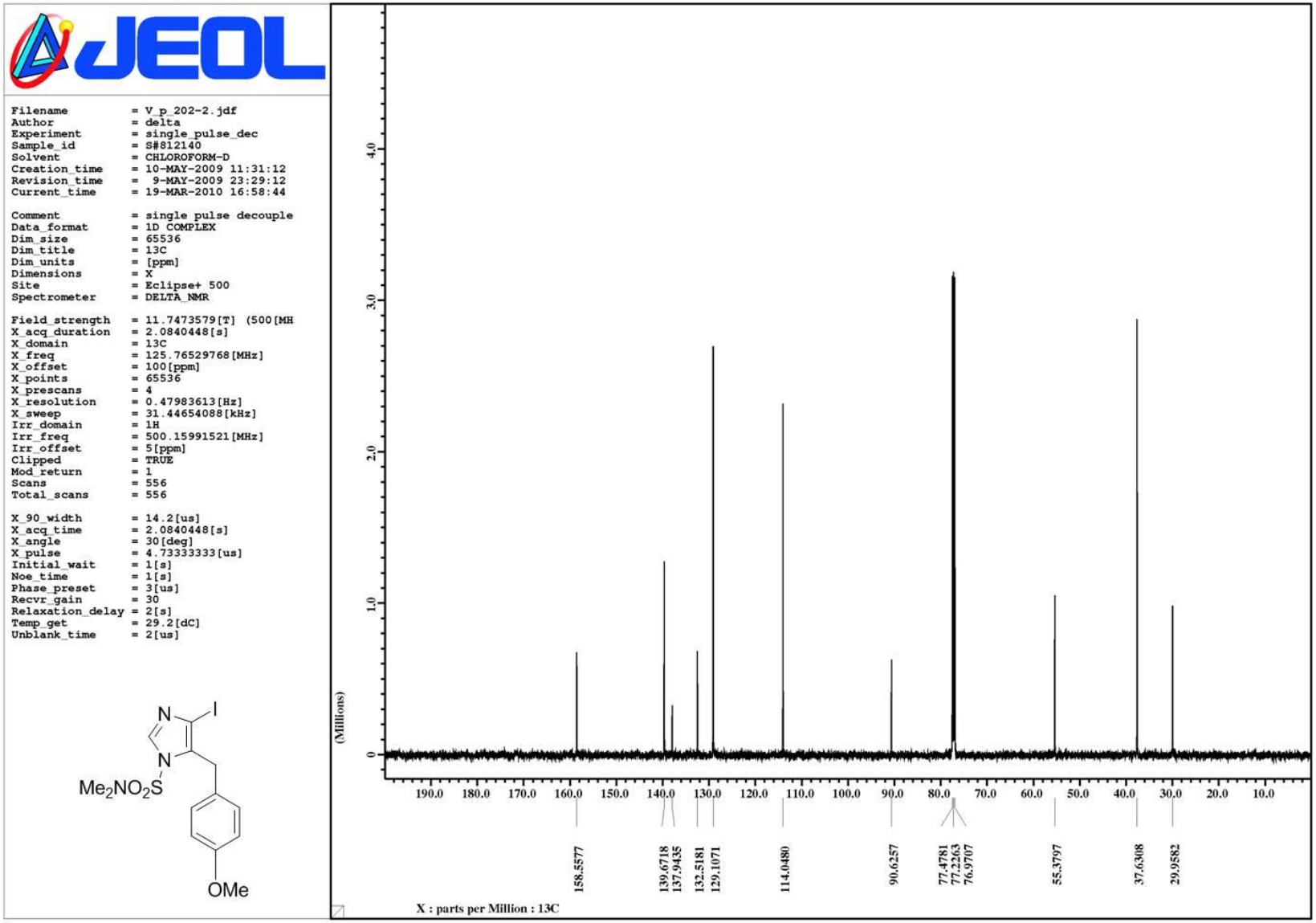
^{1}H and ^{13}C NMR Spectra of

1-(*N*,*N*-dimethylsulfonyl)-4-iodo-5-(4-methoxybenzyl)-1*H*-imidazole (**211**)

```
Filename          = V_p_194-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#770612
Solvent           = CHLOROFORM-D
Creation_time     =  6-MAY-2009 08:48:55
Revision_time     = 19-MAR-2010 16:57:01
Current_time      = 19-MAR-2010 16:57:59

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 12
Relaxation_delay  = 4[s]
Temp_get          = 26.6[dC]
Unblank_time      = 2[us]
```





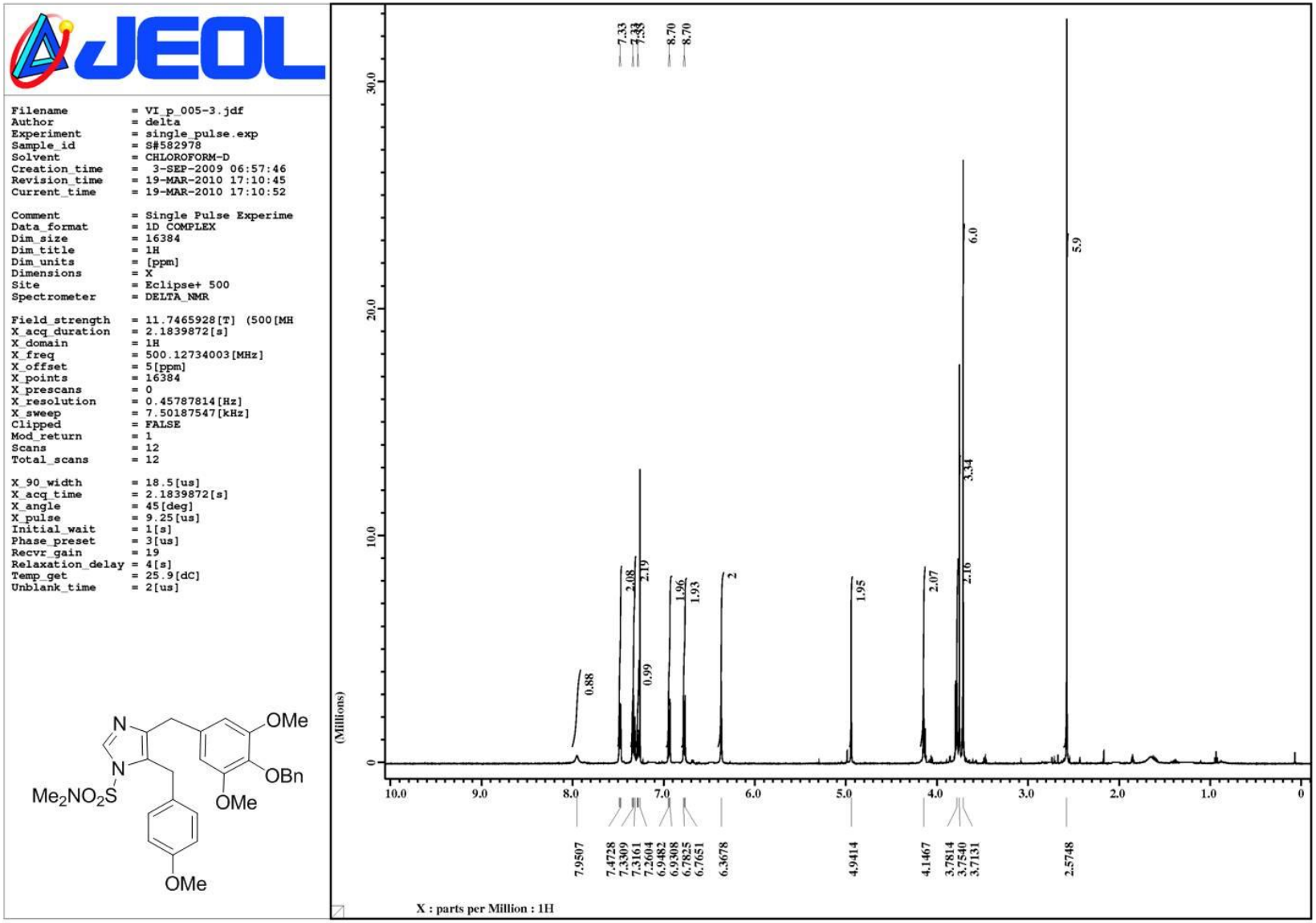


```
Filename          = V_p_202-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#812140
Solvent           = CHLOROFORM-D
Creation_time     = 10-MAY-2009 11:31:12
Revision_time     =  9-MAY-2009 23:29:12
Current_time      = 19-MAR-2010 16:58:44

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 556
Total_scans       = 556

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 29.2[dC]
Unblank_time      = 2[us]
```

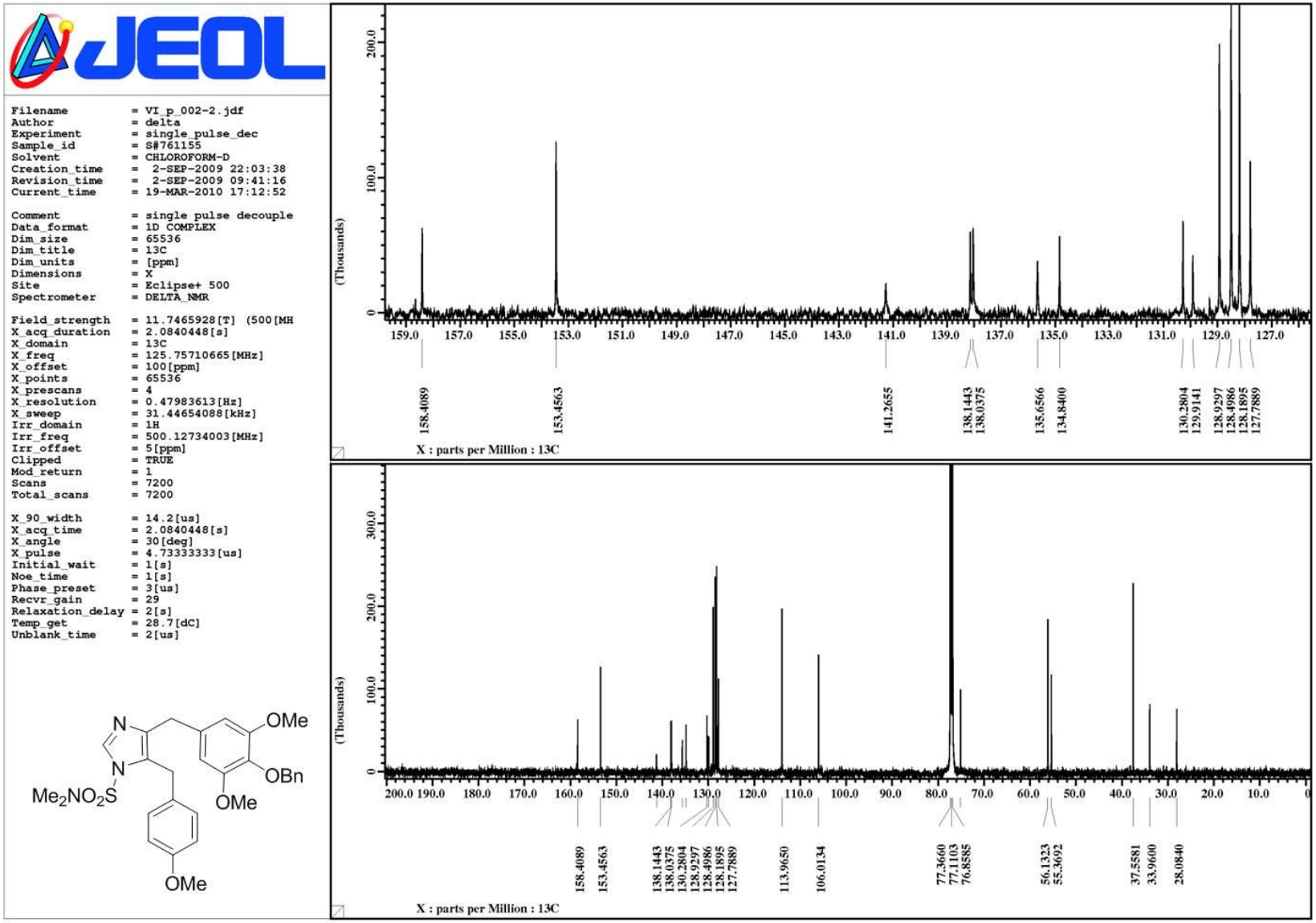
APPENDIX 92

^{1}H and ^{13}C NMR Spectra of

4-(4-Benzyloxy-3,5-dimethoxybenzyl)-1-(*N*,*N*-dimethylsulfonyl)-5-(4-methoxybenzyl)-1*H*-imidazole (**212**)


Filename = VI_p_005-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#582978
Solvent = CHLOROFORM-D
Creation_time = 3-SEP-2009 06:57:46
Revision_time = 19-MAR-2010 17:10:45
Current_time = 19-MAR-2010 17:10:52
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7465928[T] (500[MH
X_acq_duration = 2.1839872[s]
X_domain = 1H
X_freq = 500.12734003[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45787814[Hz]
X_sweep = 7.50187547[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1839872[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 19
Relaxation_delay = 4[s]
Temp_get = 25.9[dC]
Unblank_time = 2[us]
N
N
Me2NO2S
OMe
OBn
OMe
OMe
(Millions)
30.0
20.0
10.0
0
7.33
7.33
7.33
8.70
8.70
6.0
5.9
3.34
2.19
2.08
1.96
1.93
2
1.95
2.07
2.16
0.88
0.99
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
7.9507
7.4728
7.3309
7.3161
7.2604
6.9482
6.9308
6.7825
6.7651
6.3678
4.9414
4.1467
3.7814
3.7540
3.7131
2.5748
X : parts per Million : 1H

JEOL
Filename = VI_p_005-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#582978
Solvent = CHLOROFORM-D
Creation_time = 3-SEP-2009 06:57:46
Revision_time = 19-MAR-2010 17:10:45
Current_time = 19-MAR-2010 17:11:50
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7465928[T] (500[MH
X_acq_duration = 2.1839872[s]
X_domain = 1H
X_freq = 500.12734003[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45787814[Hz]
X_sweep = 7.50187547[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1839872[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 19
Relaxation_delay = 4[s]
Temp_get = 25.9[dC]
Unblank_time = 2[us]
OMe
OBn
OMe
OMe
N
N
Me2NO2S
Millions
X : parts per Million : 1H
0.88
7.9507
8.70
1.96
8.70
1.93
6.9482
6.9395
6.9308
6.7825
6.7738
6.7651
7.33
7.33
2.08
0.99
7.4875
7.4802
7.4728
7.3456
7.3382
7.3309
7.3161
7.2906
7.2833
7.2760
7.2604
2.07
4.1467
6.0
2.16
3.34
3.7814
3.7540
3.7131



Filename = VI_p_002-2.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#761155
Solvent = CHLOROFORM-D
Creation_time = 2-SEP-2009 22:03:38
Revision_time = 2-SEP-2009 09:41:16
Current_time = 19-MAR-2010 17:12:52

Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR

Field_strength = 11.7465928[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.75710665[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.12734003[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 7200
Total_scans = 7200

X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 29
Relaxation_delay = 2[s]
Temp_get = 28.7[dC]
Unblank_time = 2[us]

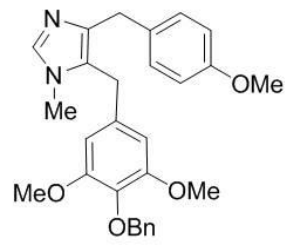
APPENDIX 93

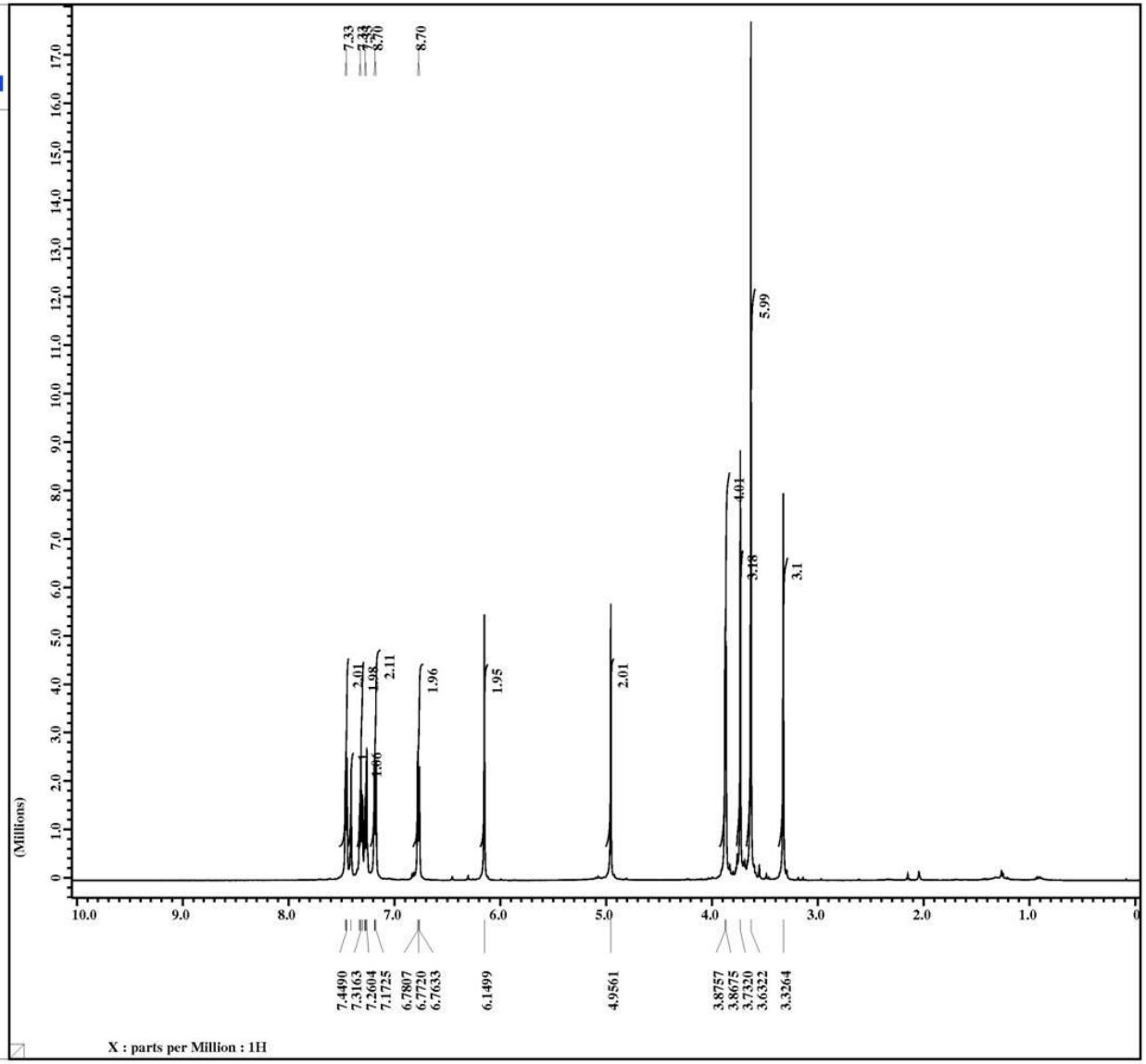
^{1}H and ^{13}C NMR Spectra of

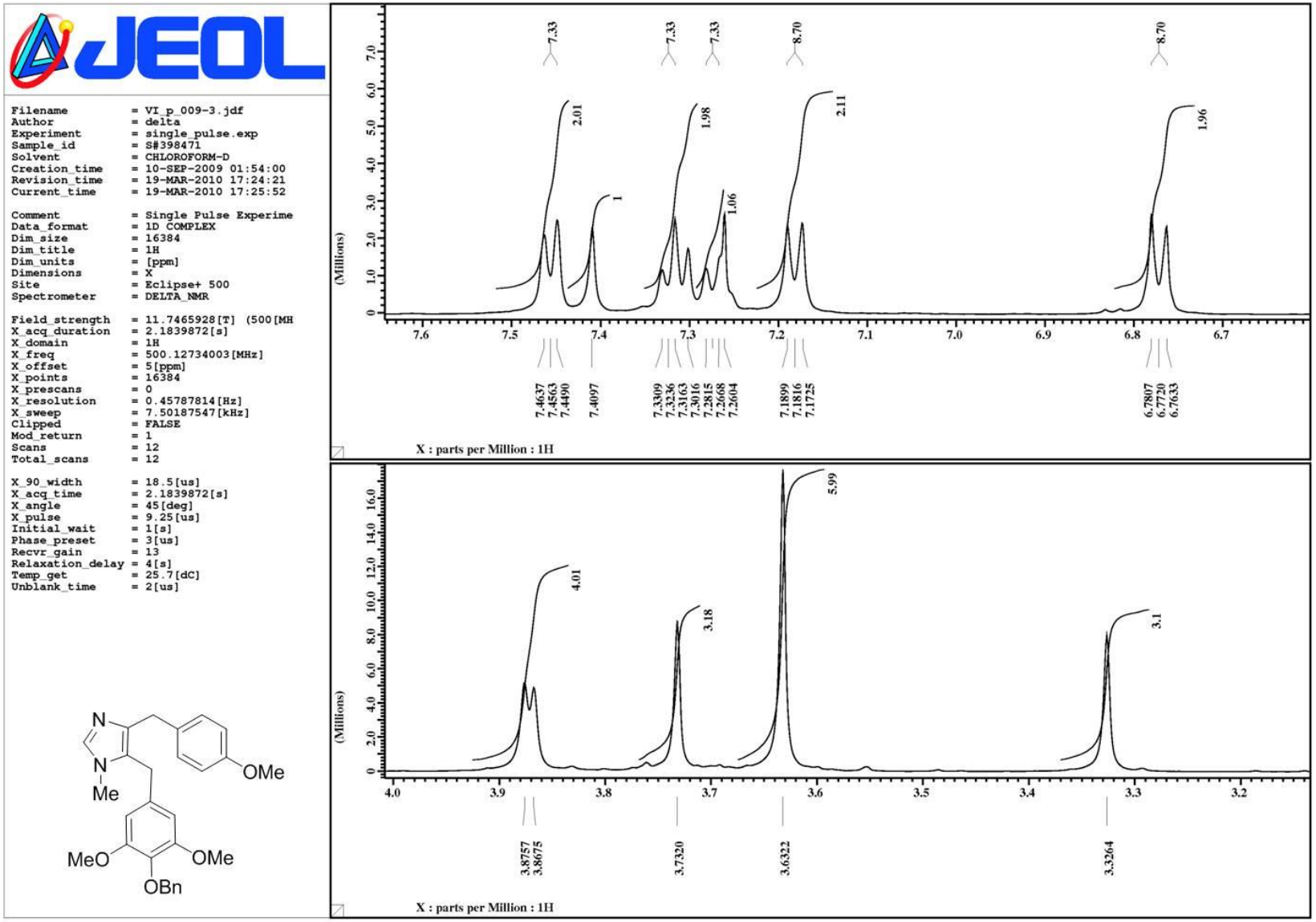
5-(4-Benzyloxy-3,5-dimethoxybenzyl)-4-(4-methoxybenzyl)-1-methyl-1*H*-imidazole

(**214**)

Filename	= VI_p_009-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#398471
Solvent	= CHLOROFORM-D
Creation_time	= 10-SEP-2009 01:54:00
Revision_time	= 19-MAR-2010 17:24:21
Current_time	= 19-MAR-2010 17:25:13
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7465928[T] (500[MH
X_acq_duration	= 2.1839872[s]
X_domain	= 1H
X_freq	= 500.12734003[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45787814[Hz]
X_sweep	= 7.50187547[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1839872[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 13
Relaxation_delay	= 4[s]
Temp_get	= 25.7[dC]
Unblank_time	= 2[us]

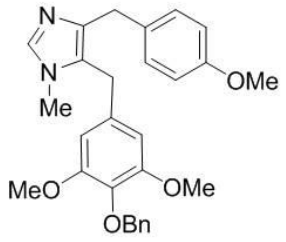


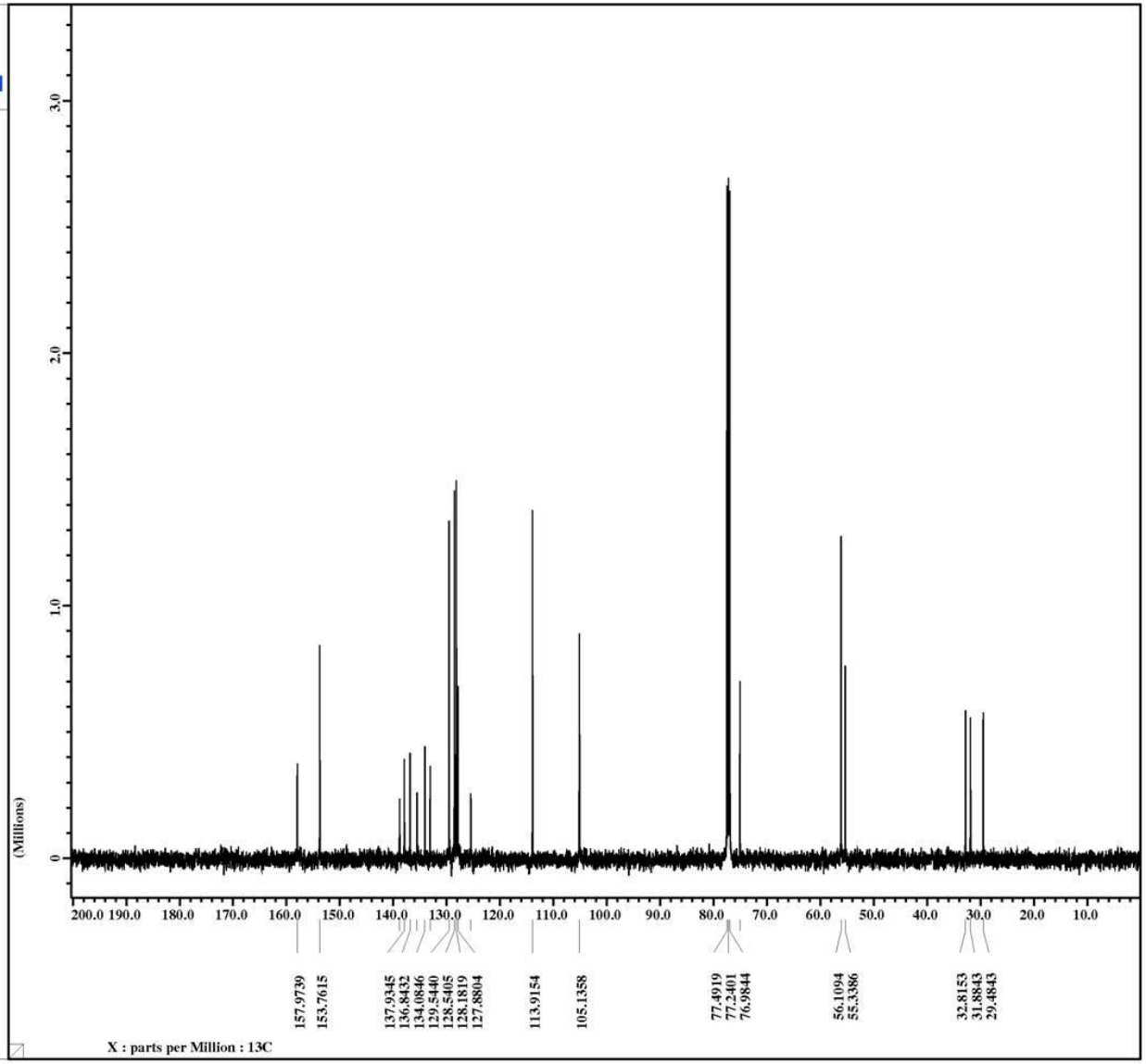




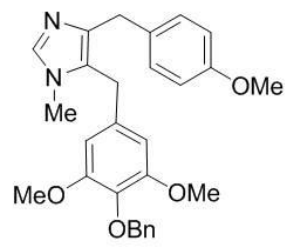
```
Filename          = VI_p_009-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#398471
Solvent           = CHLOROFORM-D
Creation_time     = 10-SEP-2009 01:54:00
Revision_time     = 19-MAR-2010 17:24:21
Current_time      = 19-MAR-2010 17:25:52

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.1839872[s]
X_domain          = 1H
X_freq            = 500.12734003[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45787814[Hz]
X_sweep           = 7.50187547[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1839872[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 13
Relaxation_delay  = 4[s]
Temp_get          = 25.7[dC]
Unblank_time      = 2[us]
```

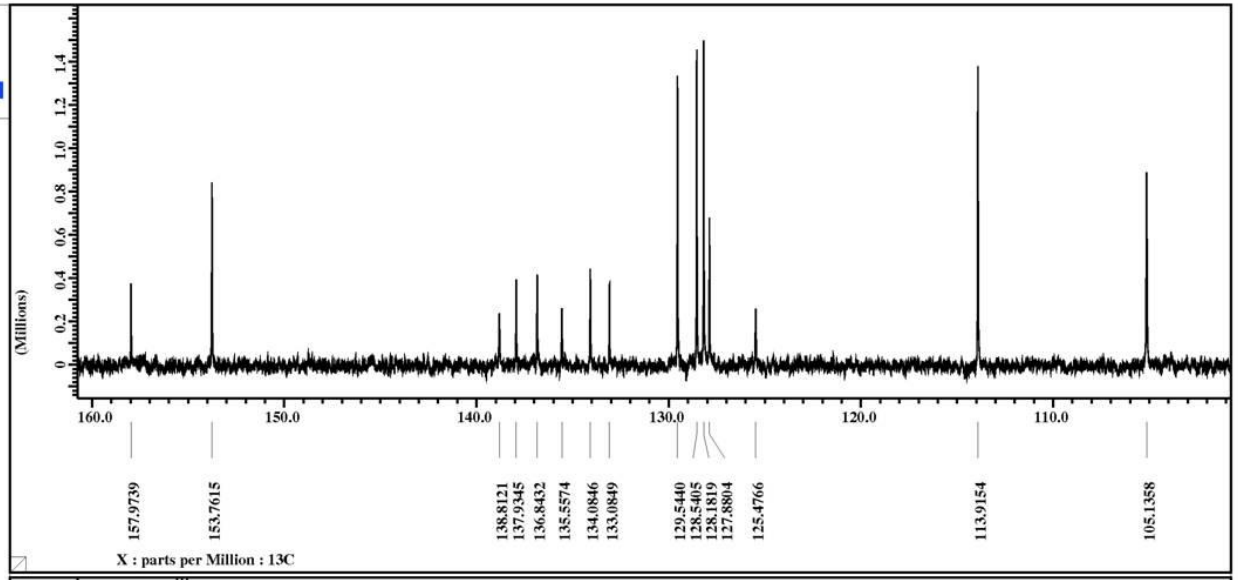
```
Filename          = VI_p_009-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#399474
Solvent           = CHLOROFORM-D
Creation_time     = 10-SEP-2009 02:15:16
Revision_time     =  9-SEP-2009 11:29:32
Current_time      = 19-MAR-2010 17:27:50

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.75710665[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.12734003[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 227
Total_scans       = 227

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.8[dC]
Unblank_time      = 2[us]
```

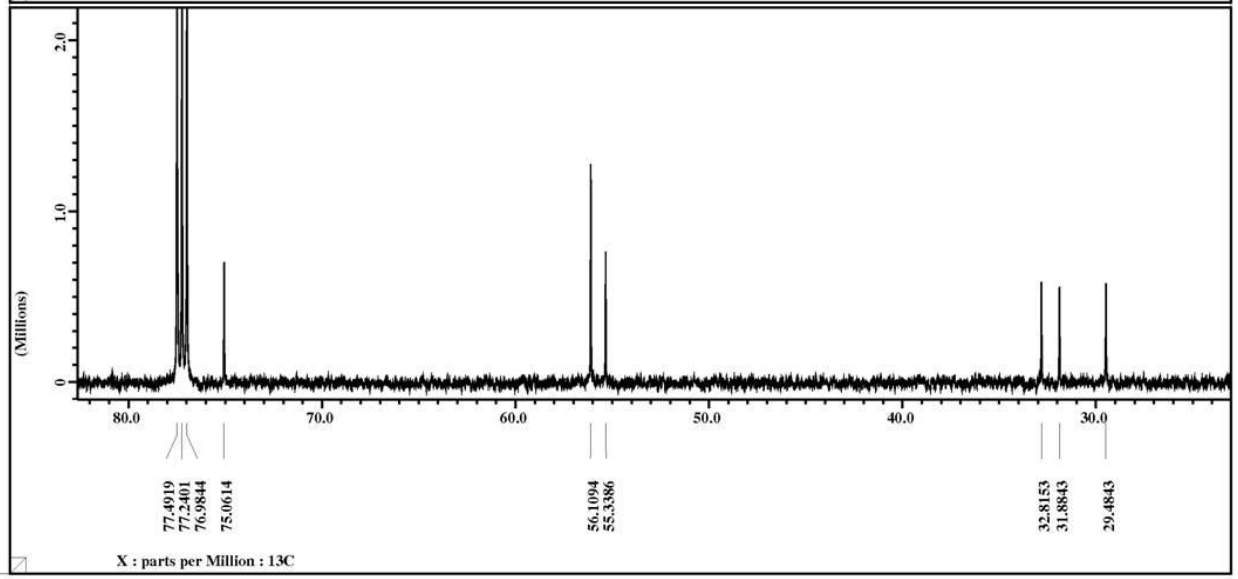




```
Filename          = VI_p_009-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#399474
Solvent           = CHLOROFORM-D
Creation_time     = 10-SEP-2009 02:15:16
Revision_time     =  9-SEP-2009 11:29:32
Current_time      = 19-MAR-2010 17:28:11

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.75710665[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.12734003[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 227
Total_scans       = 227

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.8[dC]
Unblank_time      = 2[us]
```





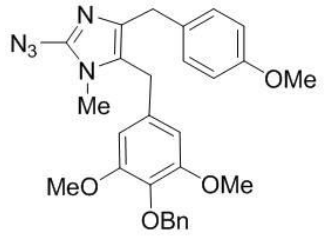


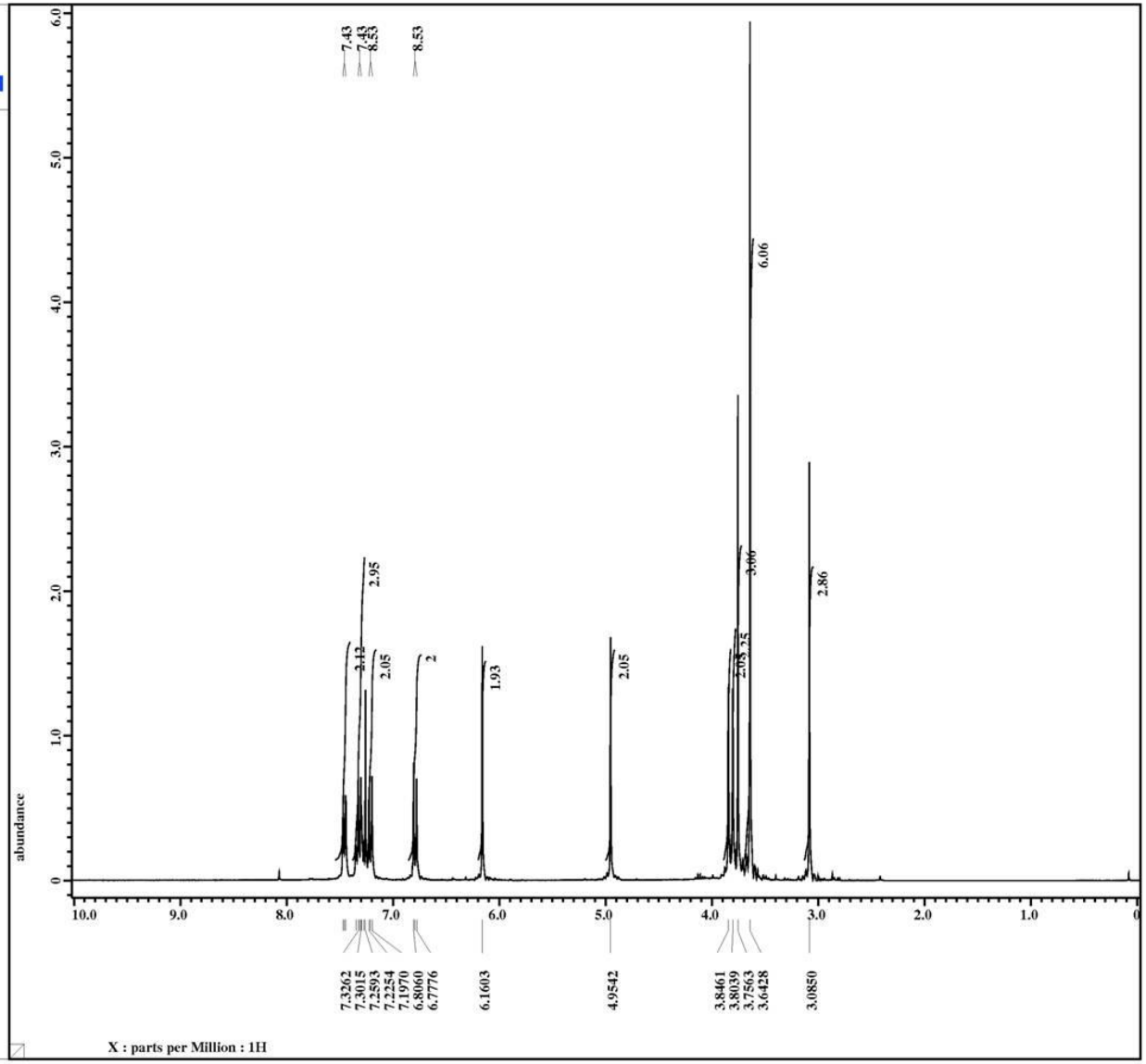
APPENDIX 94

^{1}H and ^{13}C NMR Spectra of

2-Azido-5-(4-benzyloxy-3,5-dimethoxybenzyl)-4-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (**216**)

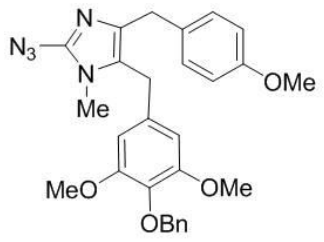
Filename	= VI_p_018_azide-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#644155
Solvent	= CHLOROFORM-D
Creation_time	= 12-SEP-2009 17:53:16
Revision_time	= 19-MAR-2010 17:35:47
Current_time	= 19-MAR-2010 17:37:02
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 38
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.2[dC]

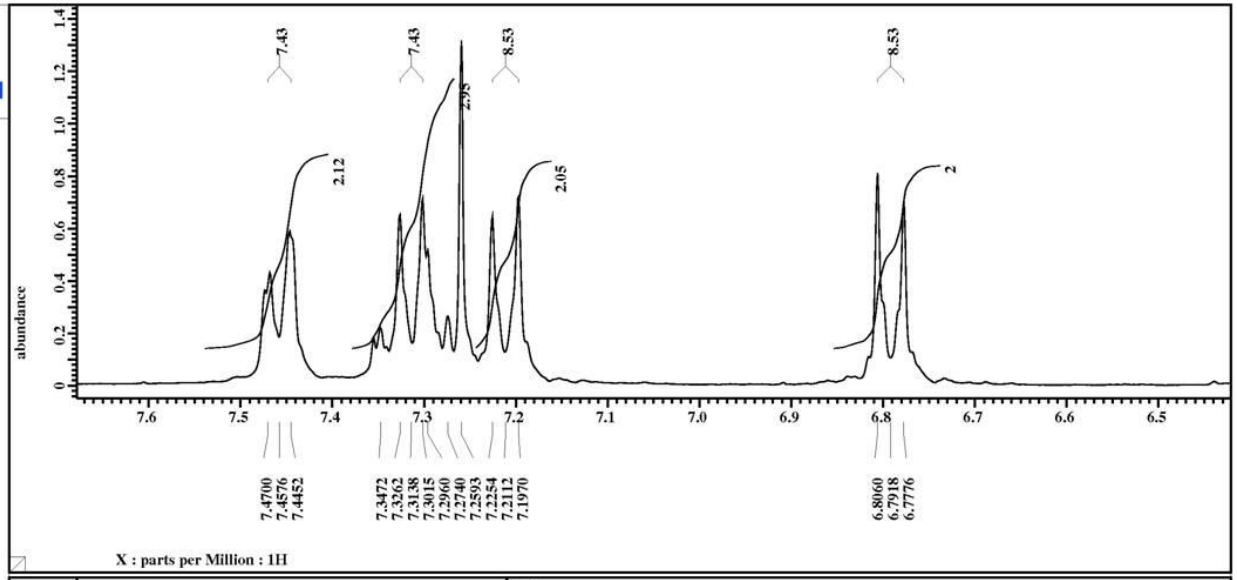


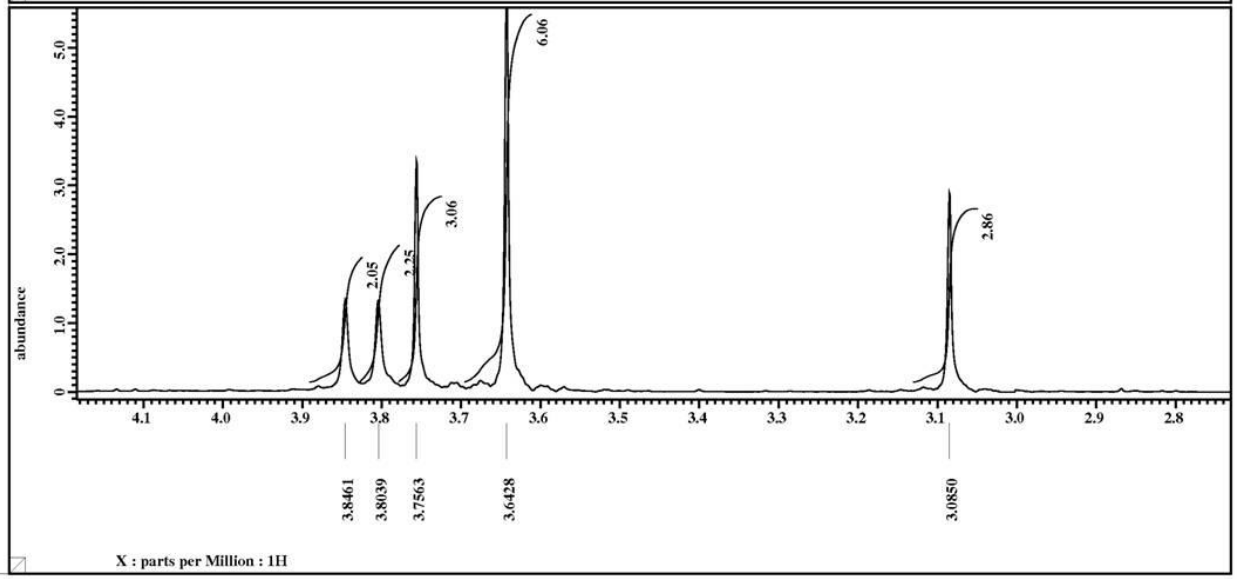


JEOL

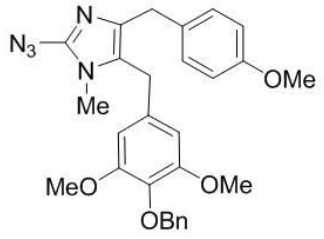
```
Filename          = VI_p_018_azide-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#644155
Solvent           = CHLOROFORM-D
Creation_time     = 12-SEP-2009 17:53:16
Revision_time     = 19-MAR-2010 17:35:47
Current_time      = 19-MAR-2010 17:37:28

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 38
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.2[dC]
```

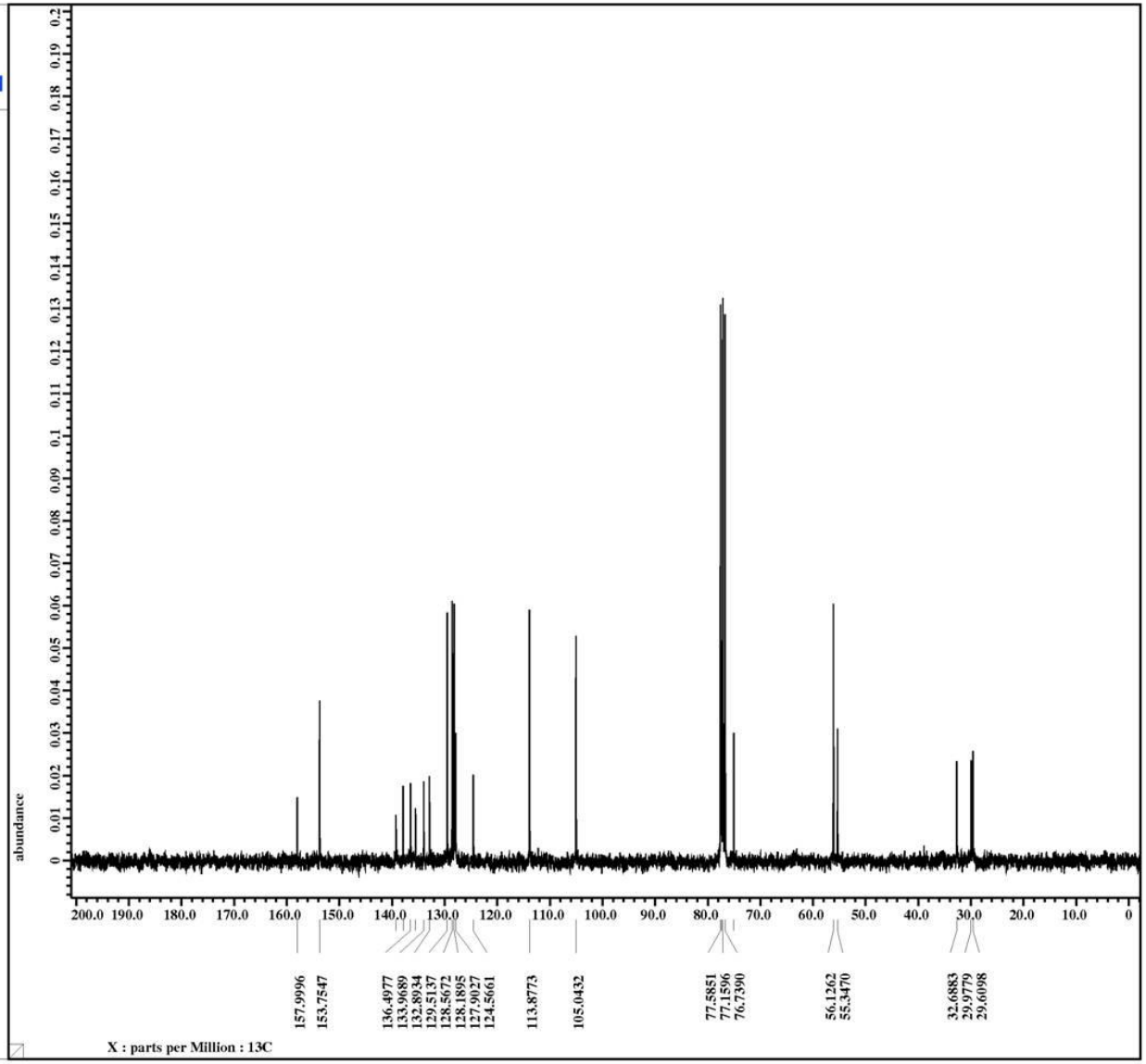




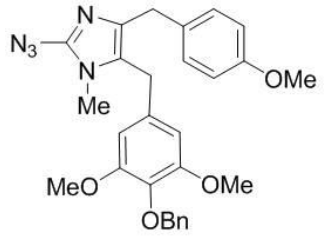


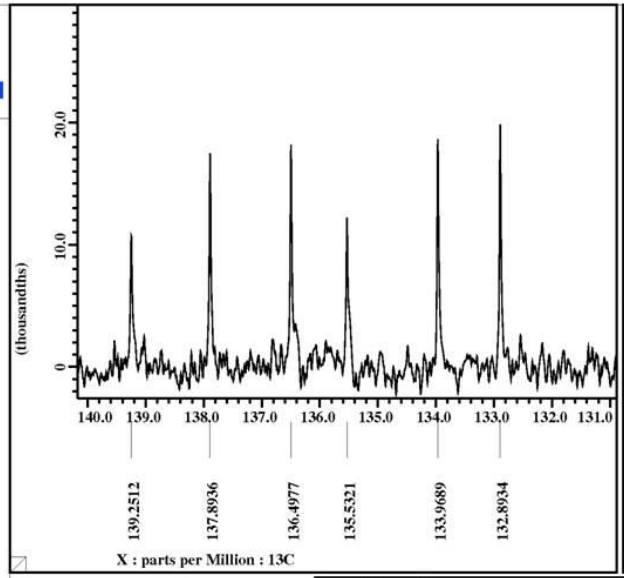
```
Filename          = VI_p_018_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#645616
Solvent           = CHLOROFORM-D
Creation_time     = 12-SEP-2009 18:12:46
Revision_time     = 12-SEP-2009 18:15:44
Current_time      = 19-MAR-2010 17:39:38

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 240
Total_scans       = 240

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```

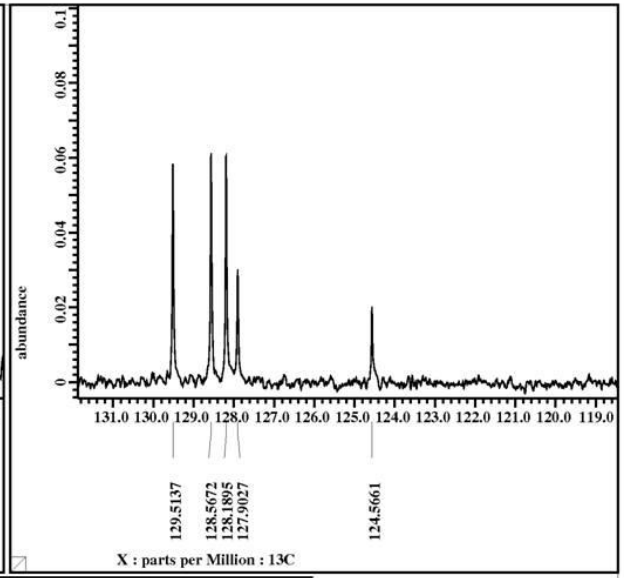


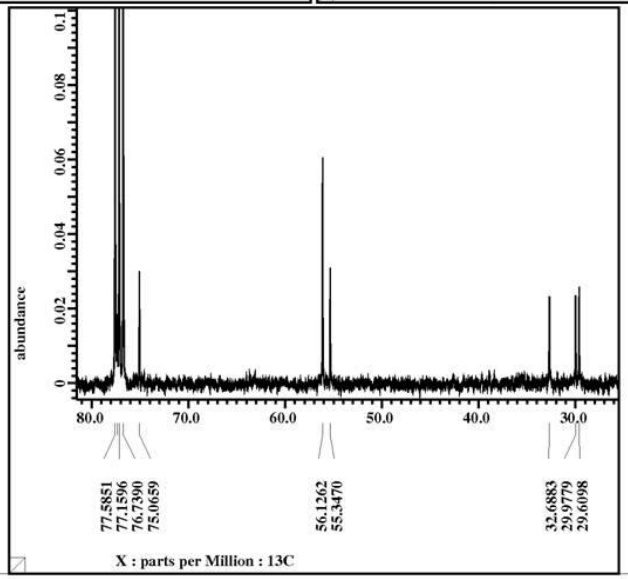


```
Filename          = VI_p_018_azide-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#645616
Solvent           = CHLOROFORM-D
Creation_time     = 12-SEP-2009 18:12:46
Revision_time     = 12-SEP-2009 18:15:44
Current_time      = 19-MAR-2010 17:39:23

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 240
Total_scans       = 240

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.3[dC]
```







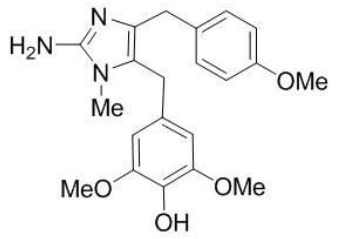


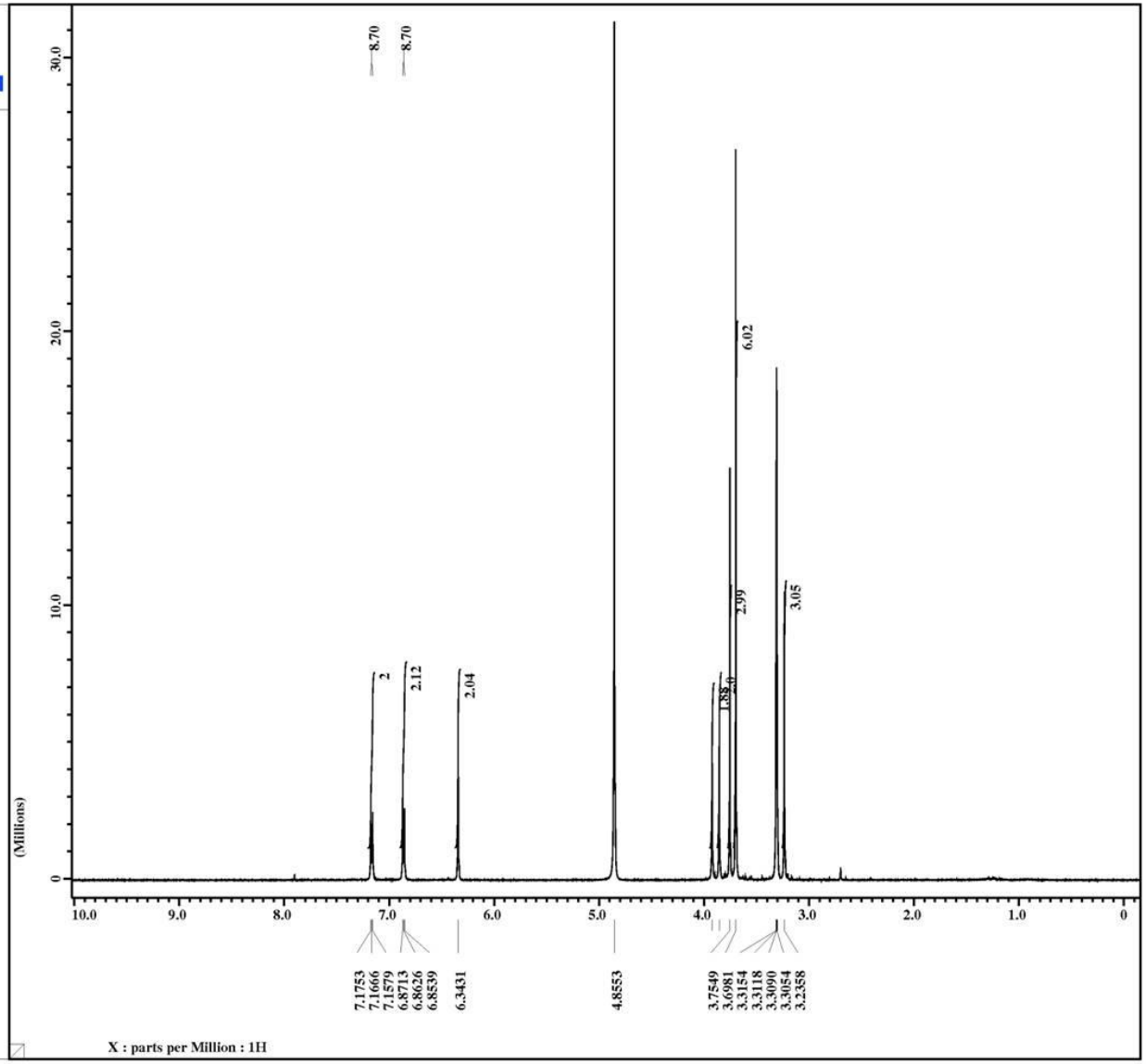
APPENDIX 95

^{1}H and ^{13}C NMR Spectra of

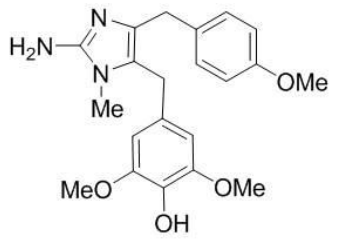
2-Amino-5-(3,5-dimethoxy-4-hydroxybenzyl)-4-(4-methoxybenzyl)-1-methyl-1*H*-imidazole (**1h**): Naamine G

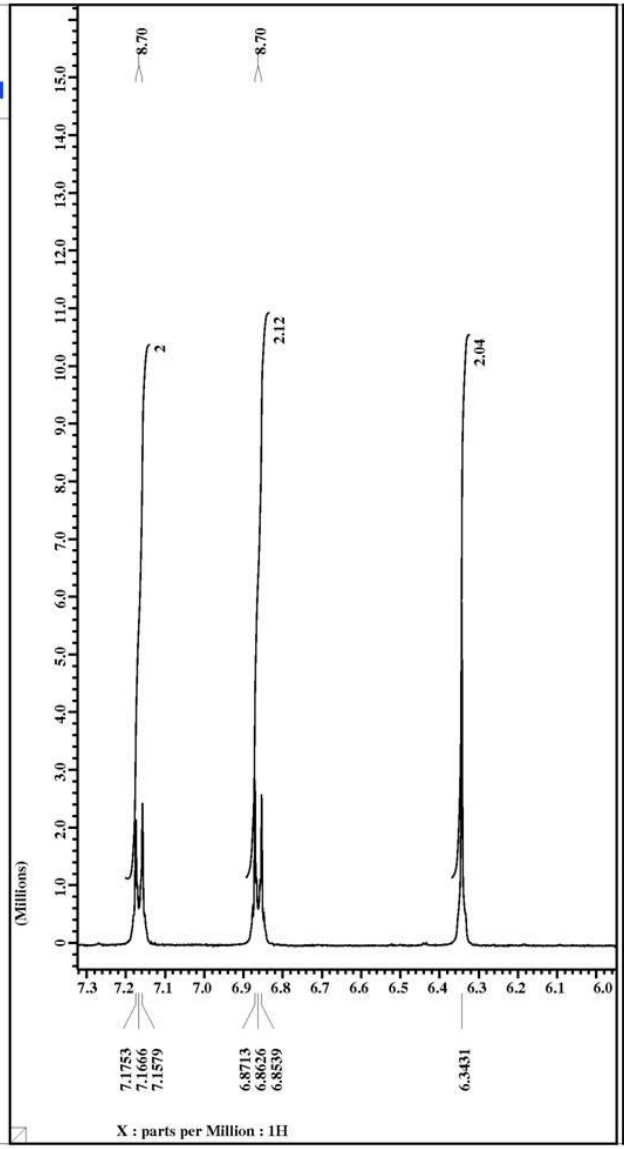
```
Filename          = VI_p_020_Naamine G-4.
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#780024
Solvent           = METHANOL-D3
Creation_time     = 17-SEP-2009 12:33:36
Revision_time     = 19-MAR-2010 17:50:29
Current_time      = 19-MAR-2010 17:50:49

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.1839872[s]
X_domain          = 1H
X_freq            = 500.12734003[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45787814[Hz]
X_sweep           = 7.50187547[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1839872[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```

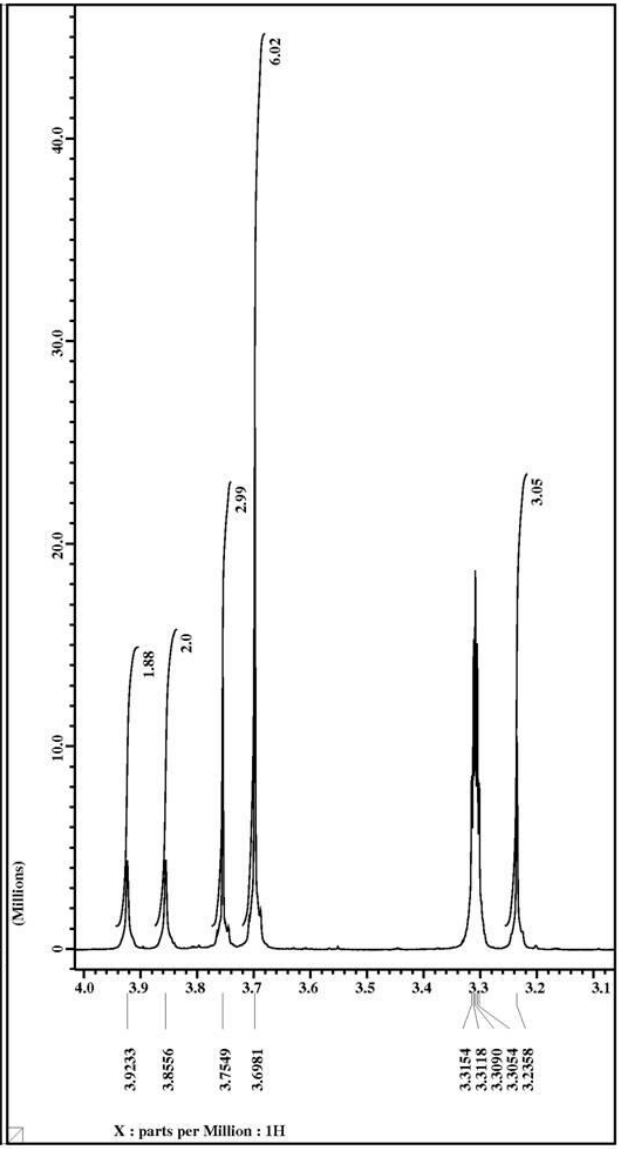




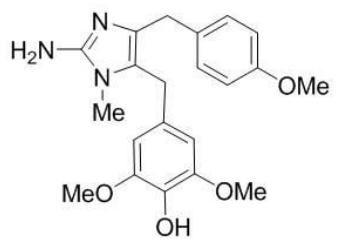
```
Filename          = VI_p_020_Naamine G-4.
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#780024
Solvent           = METHANOL-D3
Creation_time     = 17-SEP-2009 12:33:36
Revision_time     = 19-MAR-2010 17:50:29
Current_time      = 19-MAR-2010 17:51:51

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.1839872[s]
X_domain          = 1H
X_freq            = 500.12734003[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45787814[Hz]
X_sweep           = 7.50187547[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1839872[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 25.8[dC]
Unblank_time      = 2[us]
```

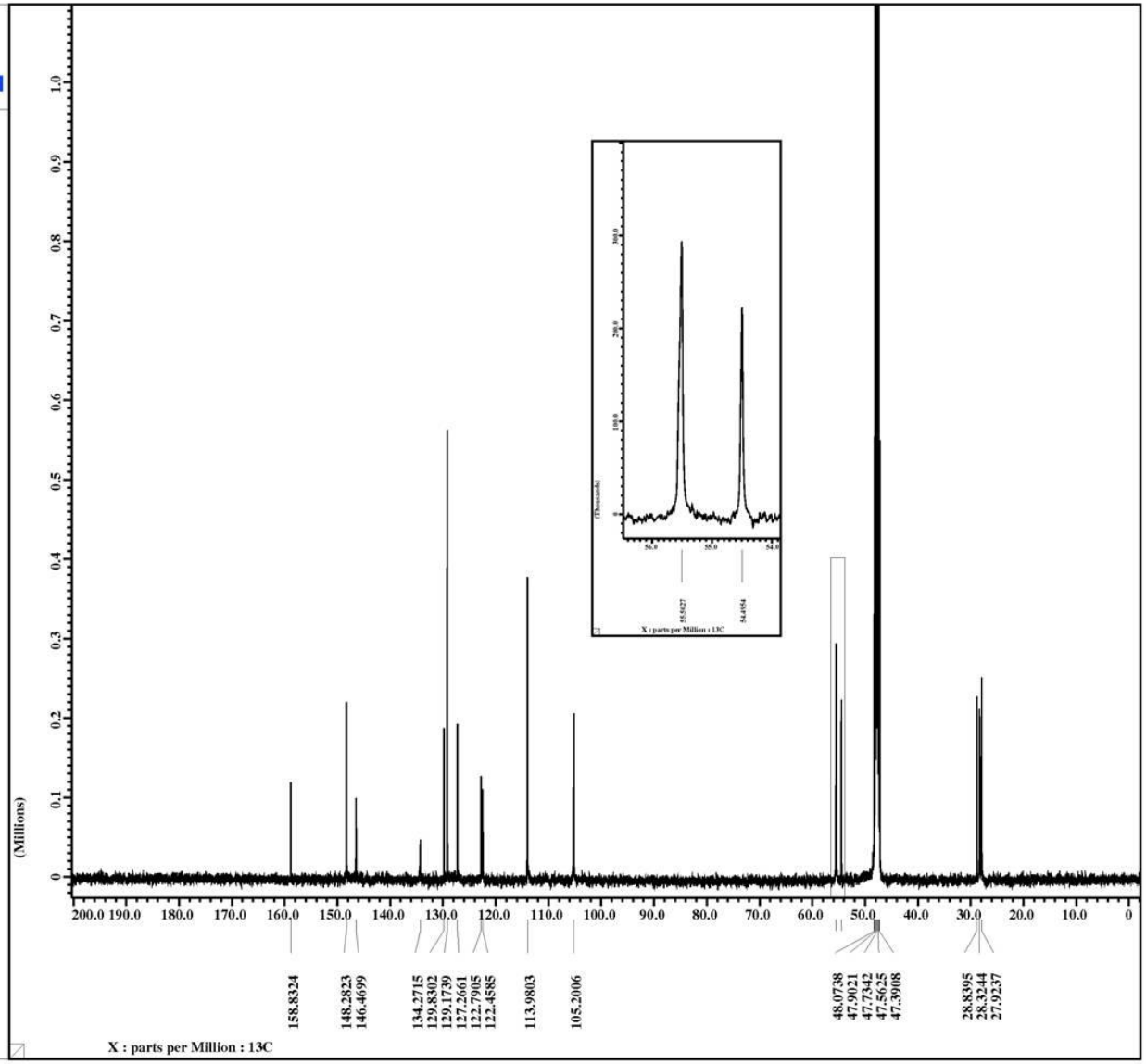






```
Filename          = VI_p_020_naamine G-3.
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#804773
Solvent           = METHANOL-D3
Creation_time     = 18-SEP-2009 22:18:01
Revision_time     = 18-SEP-2009 09:40:16
Current_time      = 19-MAR-2010 17:49:47

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.75710665[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.12734003[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 6400
Total_scans       = 6400

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.1[dC]
Unblank_time      = 2[us]
```

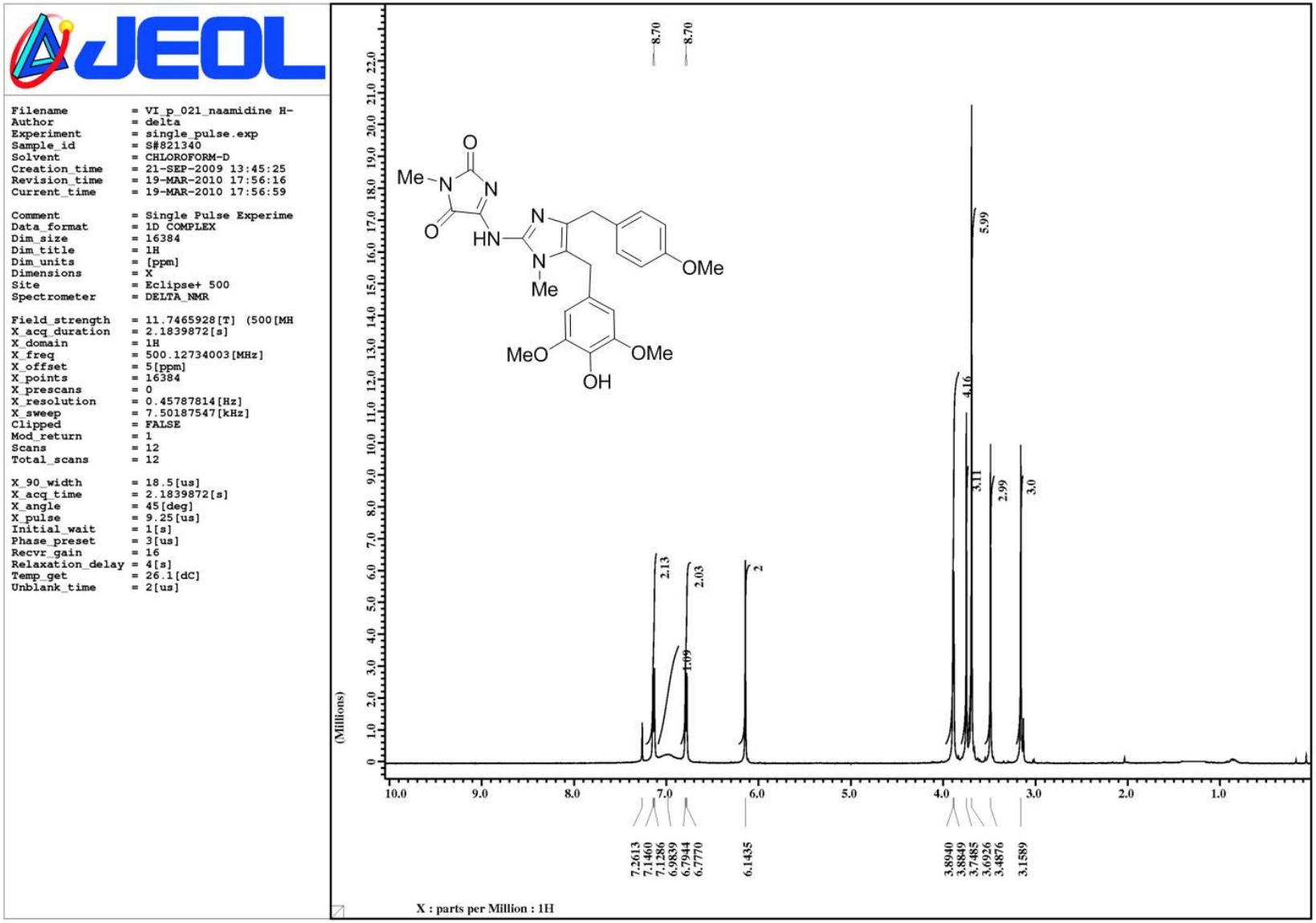




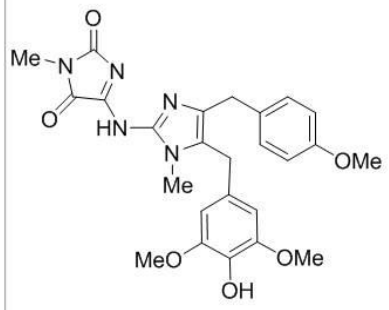
APPENDIX 96

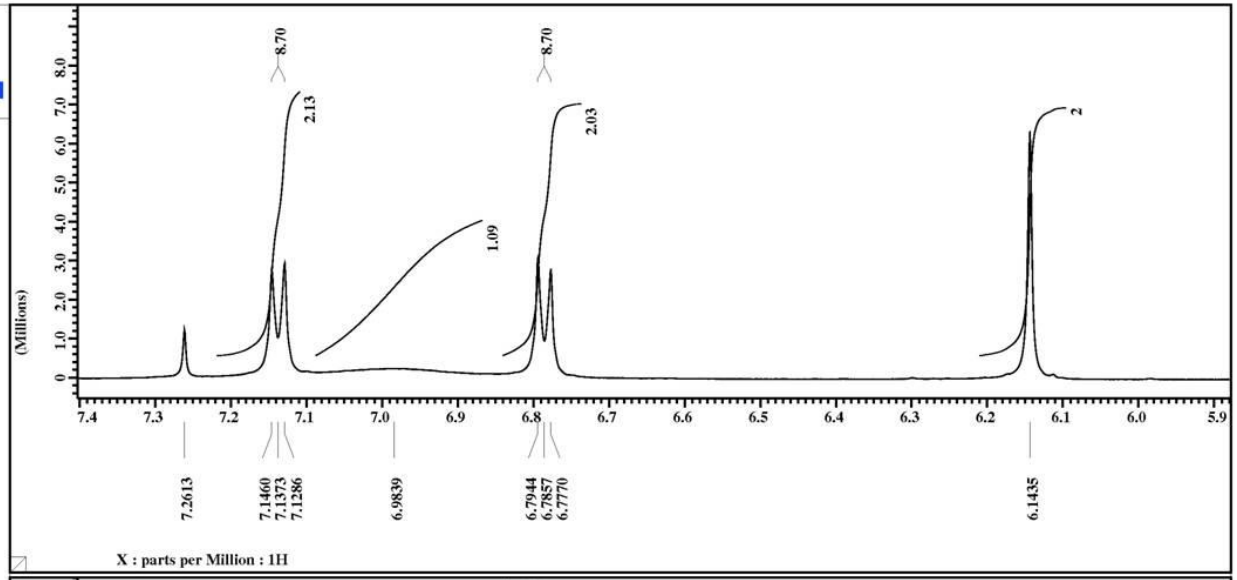
^{1}H and ^{13}C NMR Spectra of

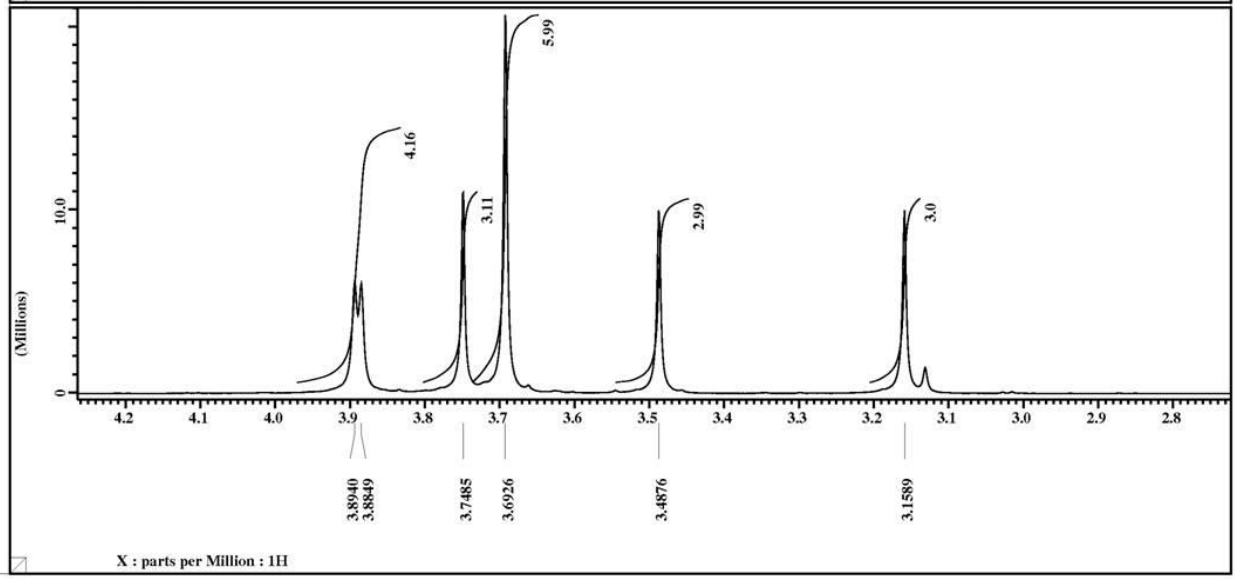
5-(3,5-Dimethoxy-4-hydroxybenzyl)-4-(4-methoxybenzyl)-1-methyl-2-(3-methylimidazolidine-2,4-dione)imino-1*H*-imidazole (**2g**): Naamidine H

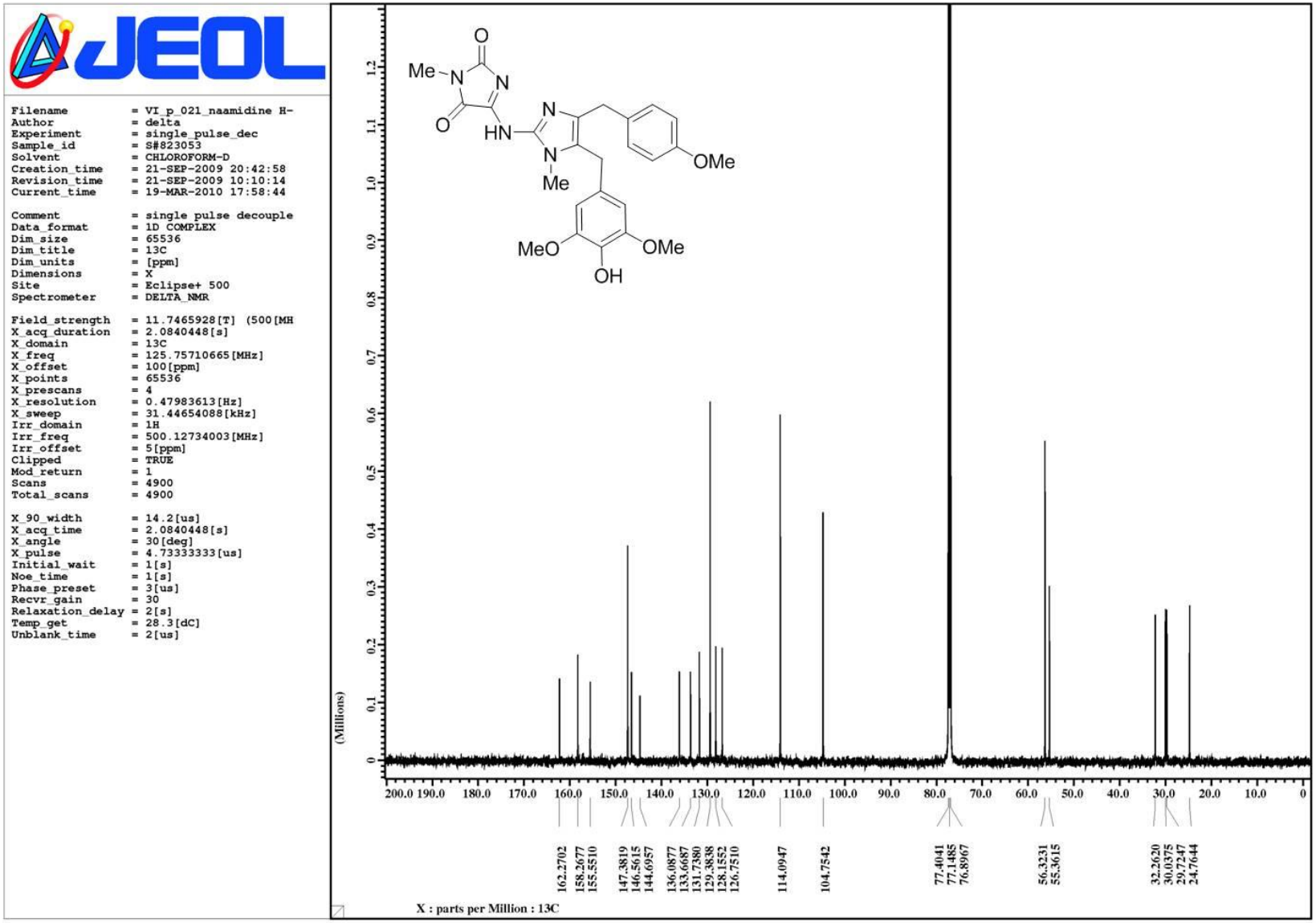

Filename = VI_p_021_naamidine H-
Author = delta
Experiment = single_pulse.exp
Sample_id = S#821340
Solvent = CHLOROFORM-D
Creation_time = 21-SEP-2009 13:45:25
Revision_time = 19-MAR-2010 17:56:16
Current_time = 19-MAR-2010 17:56:59
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7465928[T] (500[MH
X_acq_duration = 2.1839872[s]
X_domain = 1H
X_freq = 500.12734003[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45787814[Hz]
X_sweep = 7.50187547[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1839872[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 16
Relaxation_delay = 4[s]
Temp_get = 26.1[dC]
Unblank_time = 2[us]
Me
N
O
HN
OMe
MeO
OH
8.70
8.70
5.99
4.16
3.11
2.99
3.0
2.13
2.03
2
1.09
(Millions)
7.2613
7.1460
7.1286
6.9839
6.7944
6.7770
6.1435
3.8940
3.8849
3.7485
3.6926
3.4876
3.1589
X : parts per Million : 1H

```
Filename          = VI_p_021_naamidine H-
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#821340
Solvent           = CHLOROFORM-D
Creation_time     = 21-SEP-2009 13:45:25
Revision_time     = 19-MAR-2010 17:56:16
Current_time      = 19-MAR-2010 17:57:19

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7465928[T] (500[MH
X_acq_duration    = 2.1839872[s]
X_domain          = 1H
X_freq            = 500.12734003[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45787814[Hz]
X_sweep           = 7.50187547[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1839872[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 16
Relaxation_delay  = 4[s]
Temp_get          = 26.1[dC]
Unblank_time      = 2[us]
```





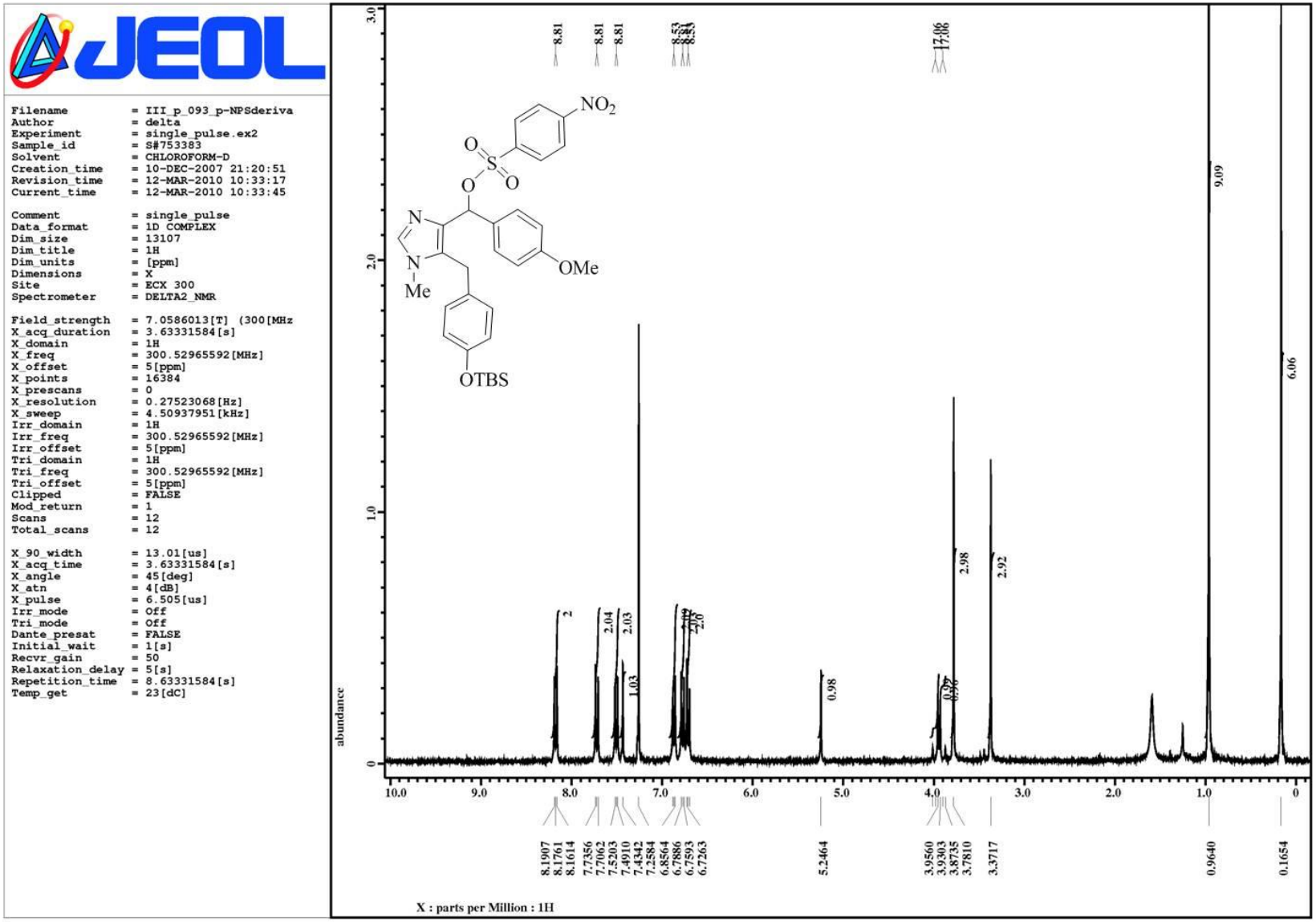



Filename = VI_p_021_naamidine H-
Author = delta
Experiment = single_pulse_dec
Sample_id = S#823053
Solvent = CHLOROFORM-D
Creation_time = 21-SEP-2009 20:42:58
Revision_time = 21-SEP-2009 10:10:14
Current_time = 19-MAR-2010 17:58:44
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7465928[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.75710665[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.12734003[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 4900
Total_scans = 4900
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 28.3[dC]
Unblank_time = 2[us]
Me
N
O
HN
OMe
MeO
OH
(Millions)
200.0 190.0 180.0 170.0 160.0 150.0 140.0 130.0 120.0 110.0 100.0 90.0 80.0 70.0 60.0 50.0 40.0 30.0 20.0 10.0 0
162.2702
158.2677
155.5510
147.3819
146.5615
144.6957
136.0877
133.6687
131.7380
129.3838
128.1552
126.7510
114.0947
104.7542
77.4041
77.1485
76.8967
56.3231
55.3615
32.2620
30.0375
29.7247
24.7644
X : parts per Million : 13C

APPENDIX 97

^{1}H and ^{13}C NMR Spectra of

4-Nitrobenzenesulfonyl {5-[4-*tert*-butyldimethylsilanyloxybenzyl]-1-methyl-1*H*-imidazol-4-yl}-(4-methoxyphenyl)methanoate (**79c**)


Filename = III_p_093_p-NPSderiva
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#753383
Solvent = CHLOROFORM-D
Creation_time = 10-DEC-2007 21:20:51
Revision_time = 12-MAR-2010 10:33:17
Current_time = 12-MAR-2010 10:33:45
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 50
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23[dC]
NO2
O
S
O
O
N
N
Me
OMe
OTBS
abundance
8.81
8.81
8.81
9.09
6.06
2.98
2.92
2.04
2.03
1.03
0.98
2
8.1907
8.1761
8.1614
7.7356
7.7062
7.5203
7.4910
7.4342
7.2584
6.8564
6.7886
6.7593
6.7263
5.2464
3.9560
3.9303
3.8735
3.7810
3.3717
0.9640
0.1654
X : parts per Million : 1H

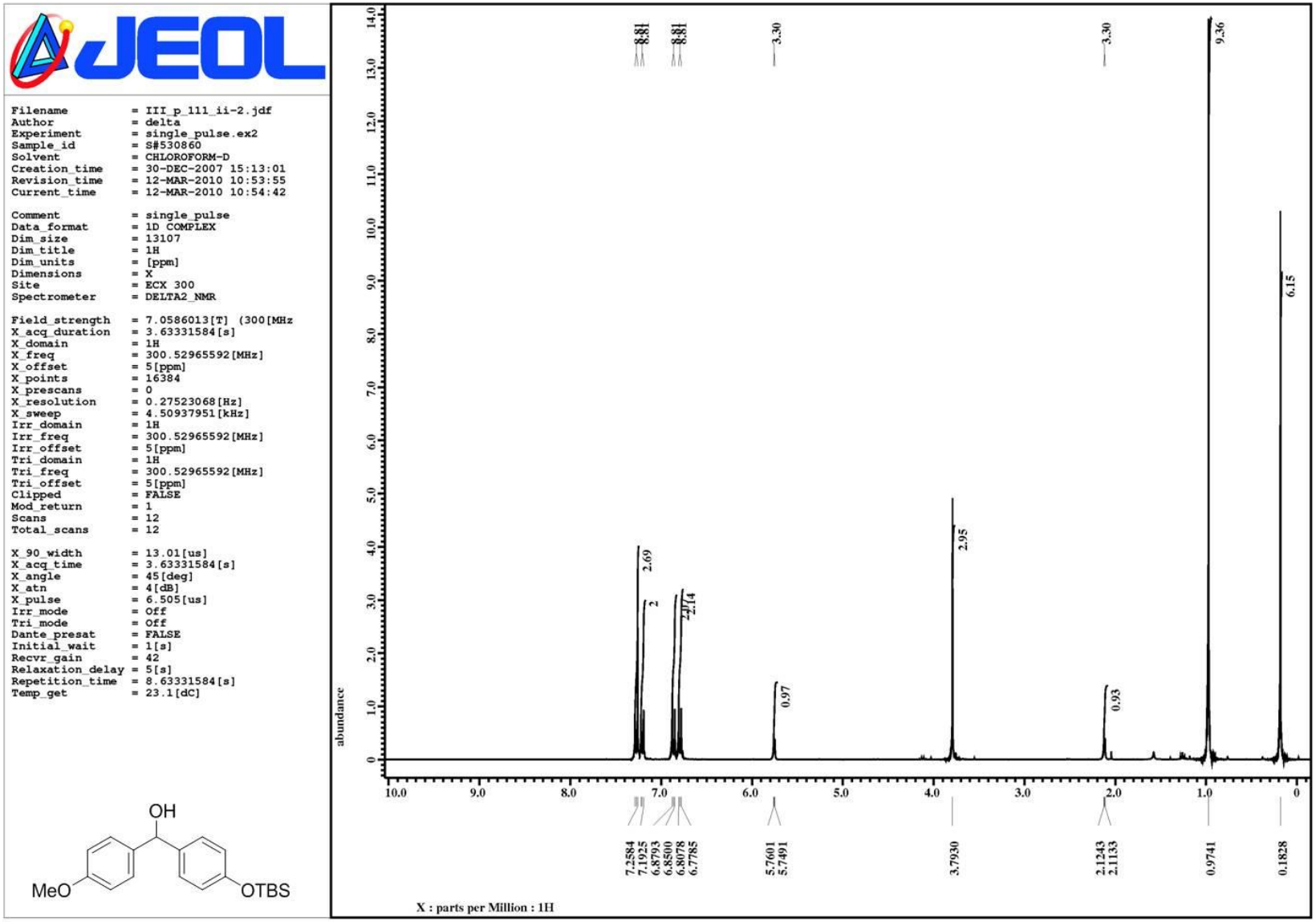
JEOL
Filename = III_p_093_p-NPSderiva
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#753383
Solvent = CHLOROFORM-D
Creation_time = 10-DEC-2007 21:20:51
Revision_time = 12-MAR-2010 10:33:17
Current_time = 12-MAR-2010 10:35:37
Comment = single pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 50
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23[dC]
abundance
X : parts per Million : 1H
8.81
2.04
2.03
1.03
8.1907
8.1761
8.1614
7.7356
7.7209
7.7062
7.5203
7.5057
7.4910
7.4342
17.06
0.99
0.96
4.0127
3.9844
3.9560
3.9303
3.9019
3.8735
8.53
2.03
2.0
6.8848
6.8706
6.8564
6.7886
6.7739
6.7593
6.7263
6.7121
6.6979
2.98
2.92
3.7810
3.3717
OMe
OTBS
Me
NO2

JEOL
Filename = III_p_093_I-4.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#756239
Solvent = CHLOROFORM-D
Creation_time = 11-DEC-2007 04:32:41
Revision_time = 20-MAR-2010 23:08:59
Current_time = 20-MAR-2010 23:10:22
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 5365
Total_scans = 5365
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23[dC]
NO2
OMe
OTBS
Me
(thousandths)
X : parts per Million : 13C
160.1699
154.7108
150.5567
143.8499
137.7836
132.2672
131.4688
130.9908
130.6371
129.4372
129.2603
123.7869
123.2276
120.5554
113.8342
69.3151
55.3709
32.0621
28.6107
25.7330
18.2852
-4.3544

APPENDIX 98

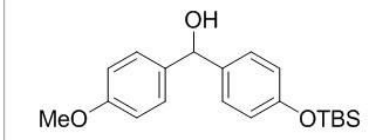
^{1}H and ^{13}C NMR Spectra of

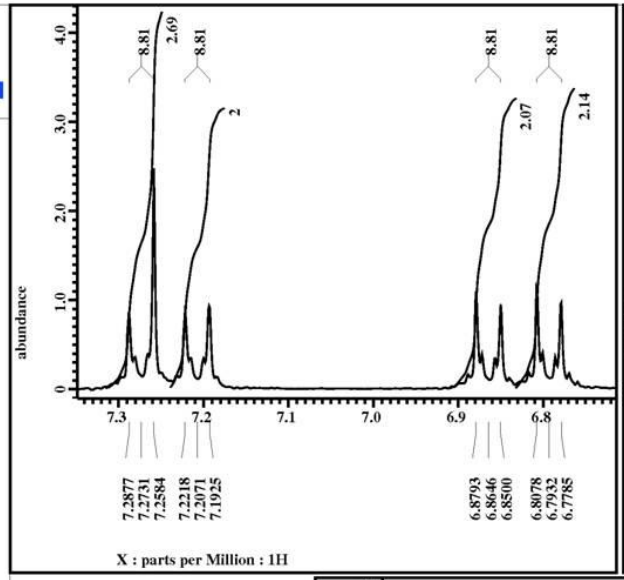
4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxybenzyl alcohol (**227**)

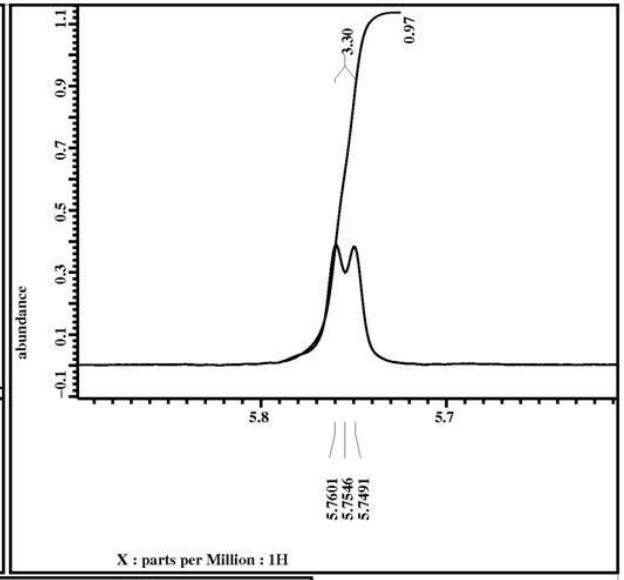


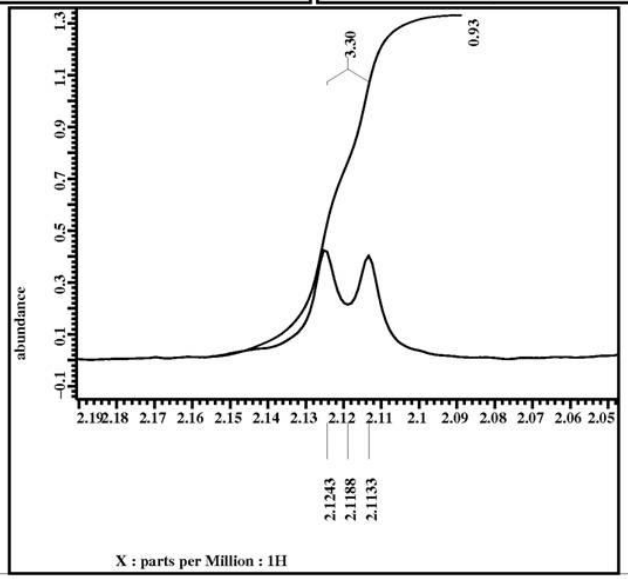
```
Filename          = III_p_111_ii-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#530860
Solvent           = CHLOROFORM-D
Creation_time     = 30-DEC-2007 15:13:01
Revision_time     = 12-MAR-2010 10:53:55
Current_time      = 12-MAR-2010 10:54:42

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 42
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```

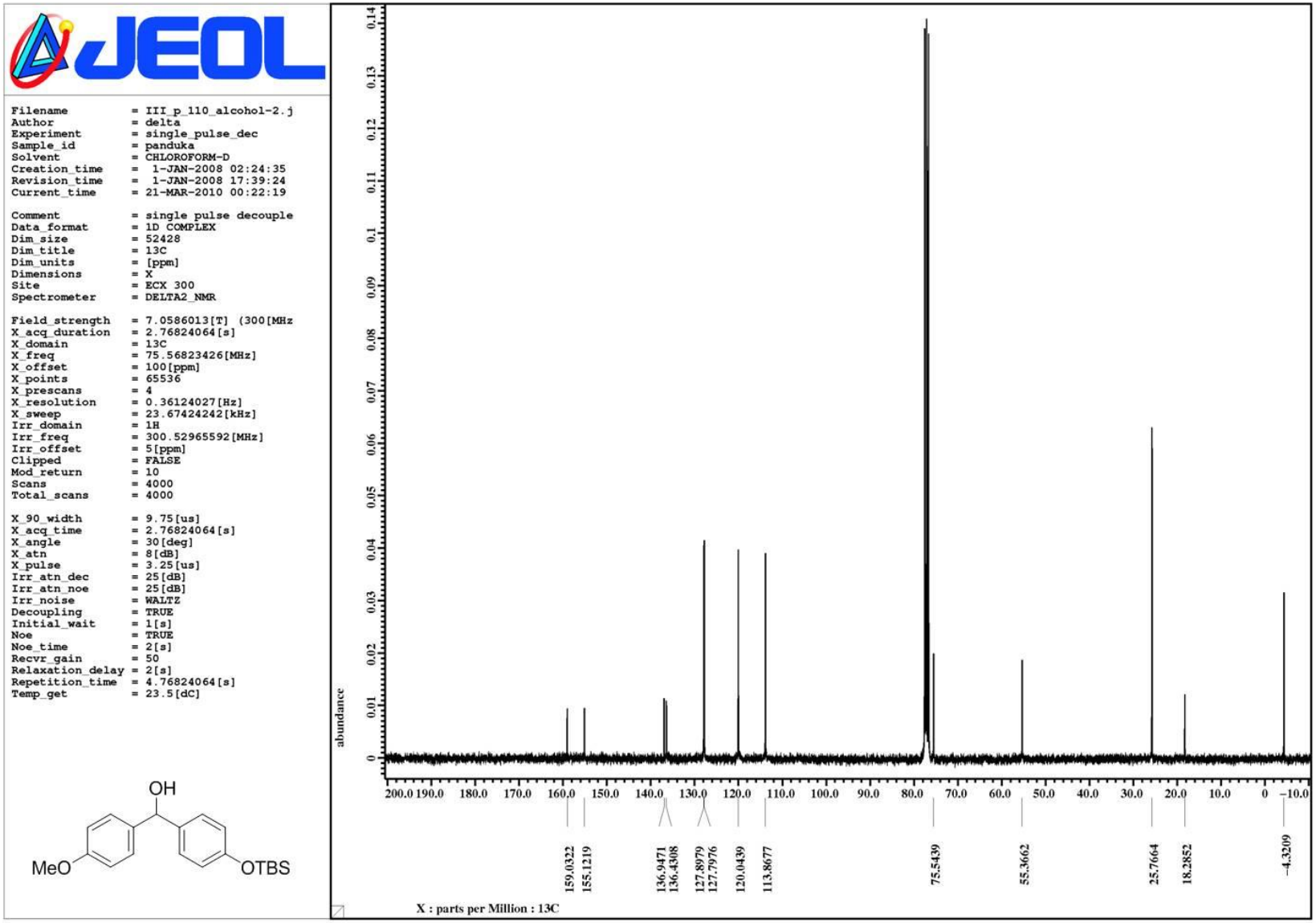
```
Filename          = III_p_111_ii-2.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#530860
Solvent           = CHLOROFORM-D
Creation_time     = 30-DEC-2007 15:13:01
Revision_time     = 12-MAR-2010 10:53:55
Current_time      = 12-MAR-2010 10:55:35

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 42
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```











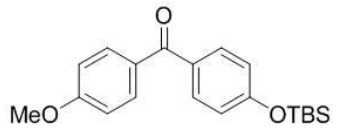
```
Filename          = III_p_110_alcohol-2.j
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     =  1-JAN-2008 02:24:35
Revision_time     =  1-JAN-2008 17:39:24
Current_time      = 21-MAR-2010 00:22:19

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 4000
Total_scans       = 4000

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.5[dC]
```

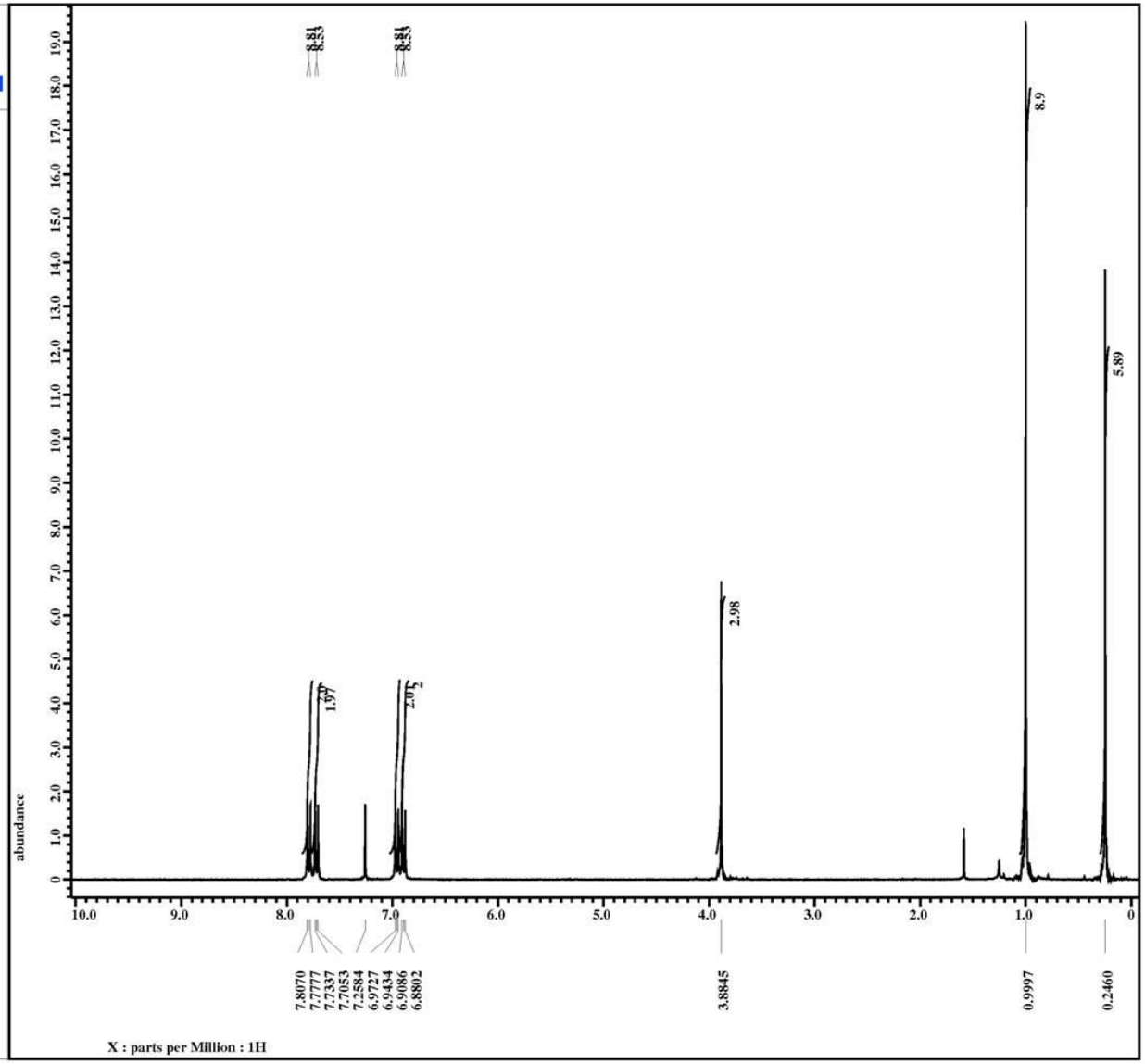
APPENDIX 99

^{1}H and ^{13}C NMR Spectra of

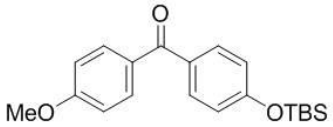
4-*tert*-Butyldimethylsilanyloxyphenyl-(4-methoxyphenyl)methanone (**228**)

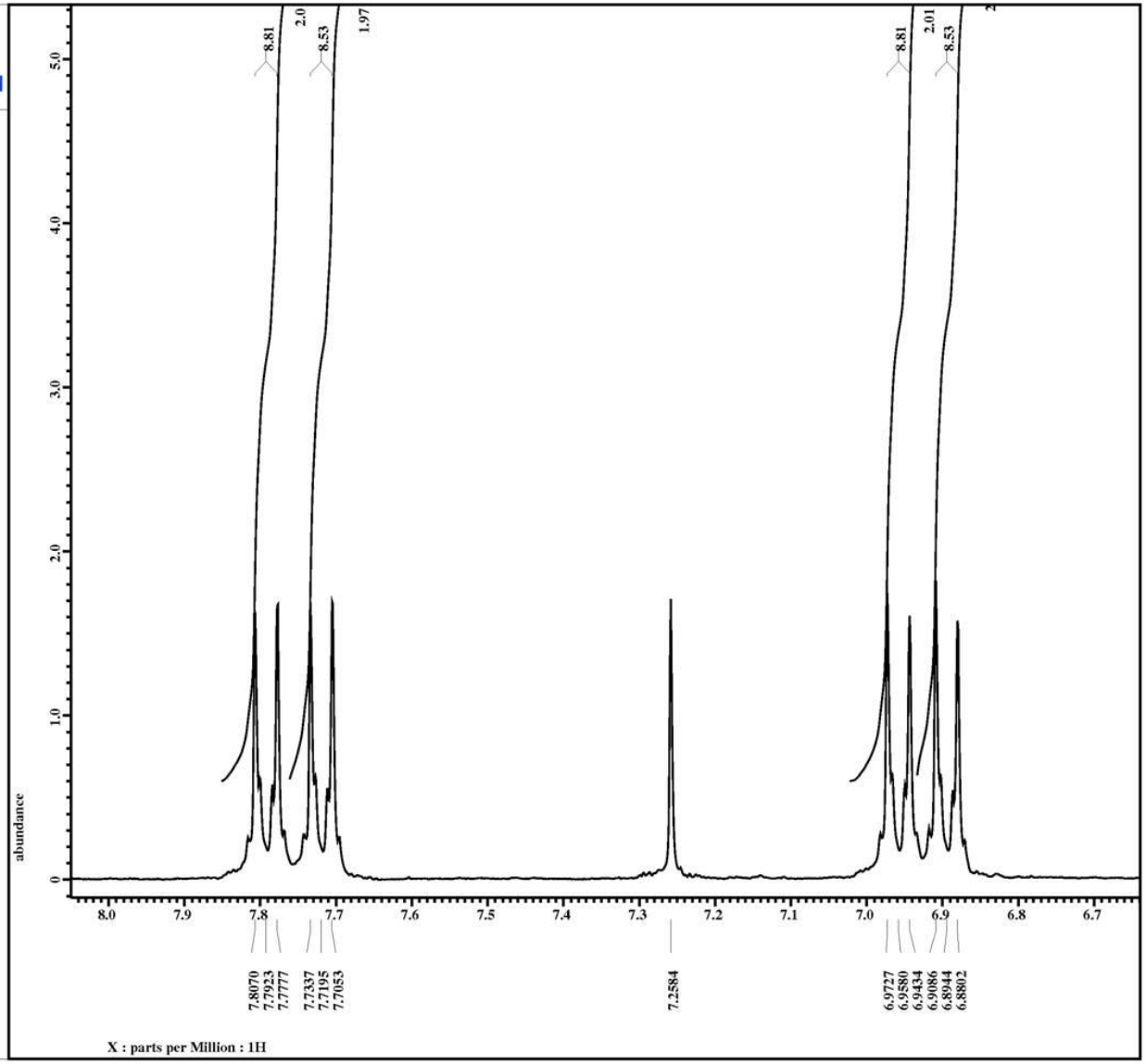
```
Filename          = III_p_112_ketone-2.jd
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#405704
Solvent           = CHLOROFORM-D
Creation_time     = 31-DEC-2007 11:43:57
Revision_time     = 12-MAR-2010 11:32:47
Current_time      = 12-MAR-2010 11:33:20

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 50
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.1[dC]
```





Filename	= III_p_112_ketone-2.jd
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#405704
Solvent	= CHLOROFORM-D
Creation_time	= 31-DEC-2007 11:43:57
Revision_time	= 12-MAR-2010 11:32:47
Current_time	= 12-MAR-2010 11:33:03
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 50
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.1[dC]



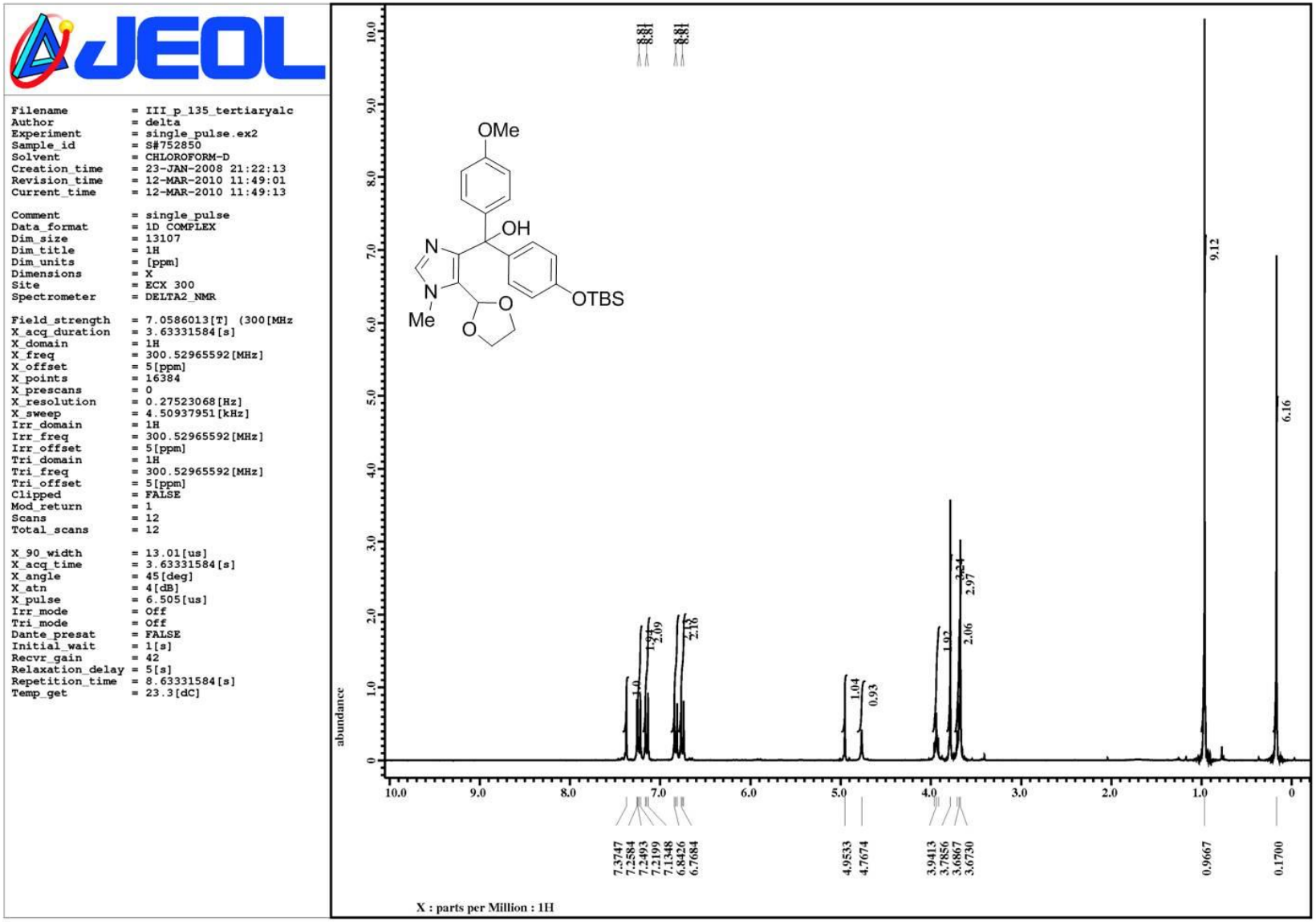


JEOL
Filename = III_p_112_ketone-2.jd
Author = delta
Experiment = single_pulse_dec
Sample_id = S#408910
Solvent = CHLOROFORM-D
Creation_time = 31-DEC-2007 13:07:11
Revision_time = 31-DEC-2007 12:41:44
Current_time = 21-MAR-2010 01:20:38
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 1000
Total_scans = 1000
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.2[dC]
O
OTBS
MeO
(thousandths)
194.6933
162.9329
159.5867
132.3580
132.2098
131.4210
130.8044
119.7284
113.5331
77.5325
77.1118
76.6864
55.5622
25.7091
18.3521
-4.2492
X : parts per Million : 13C

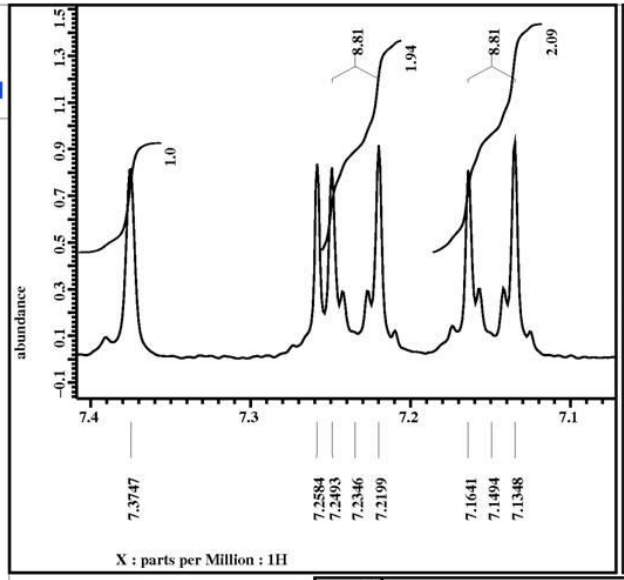
APPENDIX 100

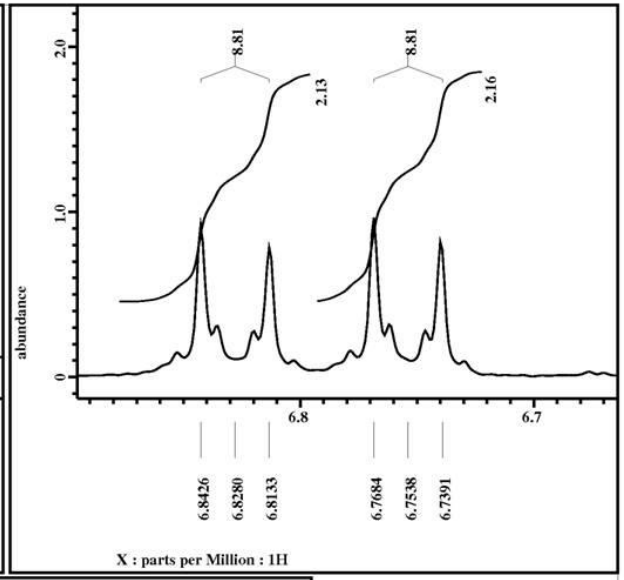
^{1}H and ^{13}C NMR Spectra of

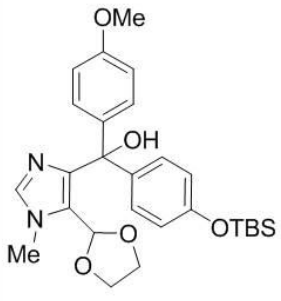
4-tert-Butyldimethysilanyloxyphenyl-(5-[1,3]dioxolan-2-yl-1-methyl-1H-imidazol-4-yl)-(4-methoxy-phenyl)-methanol (**229**)

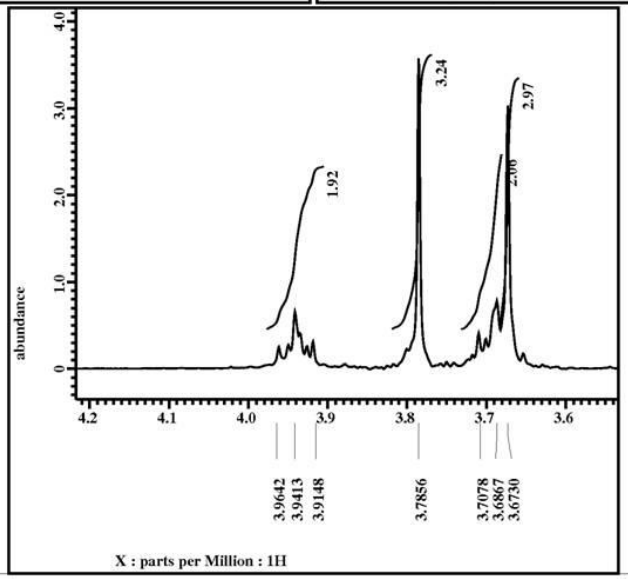

Filename = III_p_135_tertiaryalc
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#752850
Solvent = CHLOROFORM-D
Creation_time = 23-JAN-2008 21:22:13
Revision_time = 12-MAR-2010 11:49:01
Current_time = 12-MAR-2010 11:49:13
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 42
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.3[dC]
OMe
OH
N
N
Me
O
O
OTBS
abundance
8.81
8.81
8.81
8.81
9.12
6.16
1.0
1.94
2.09
2.13
2.16
1.04
0.93
1.92
2.06
2.97
3.24
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
7.3747
7.2584
7.2493
7.2199
7.1348
6.8426
6.7684
4.9533
4.7674
3.9413
3.7856
3.6867
3.6730
0.9667
0.1700
X : parts per Million : 1H

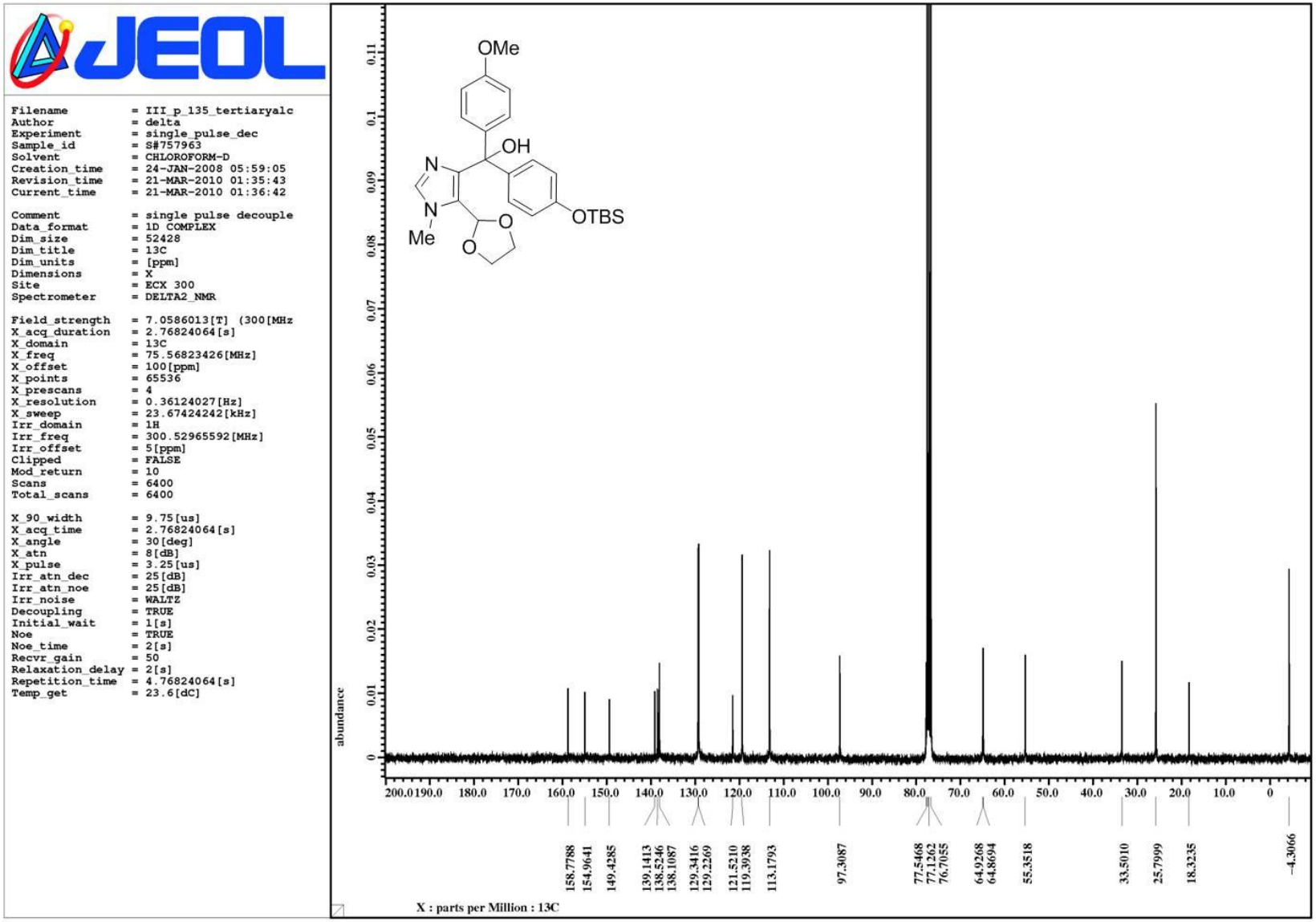
Filename	= III_p_135_tertiaryalc
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#752850
Solvent	= CHLOROFORM-D
Creation_time	= 23-JAN-2008 21:22:13
Revision_time	= 12-MAR-2010 11:49:40
Current_time	= 12-MAR-2010 11:50:27
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 42
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23.3[dC]



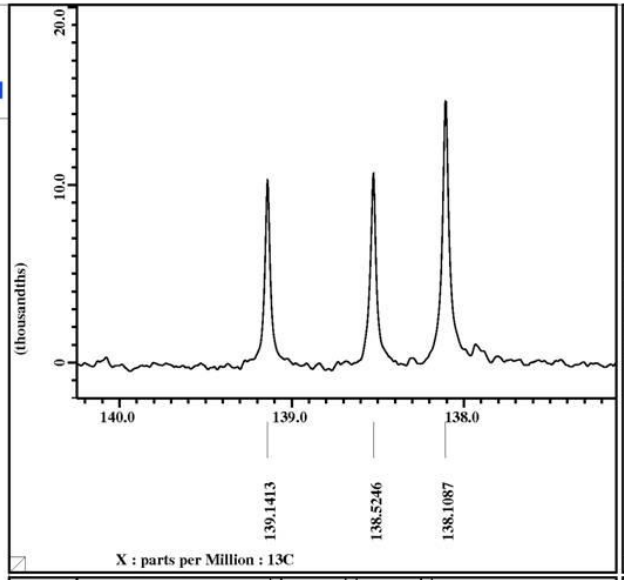


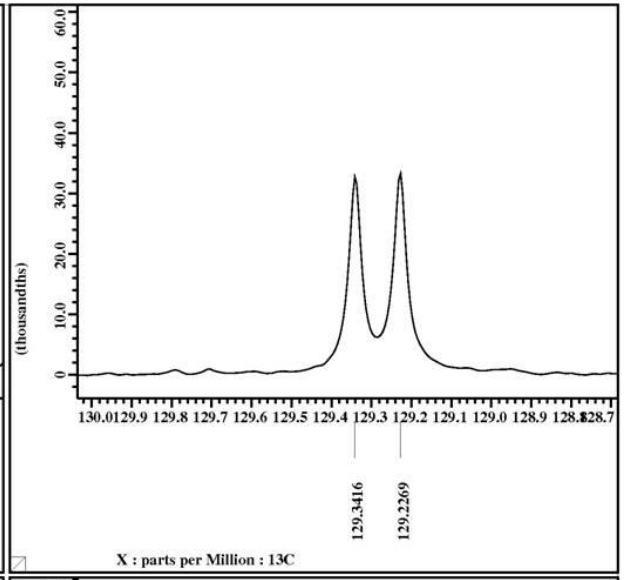


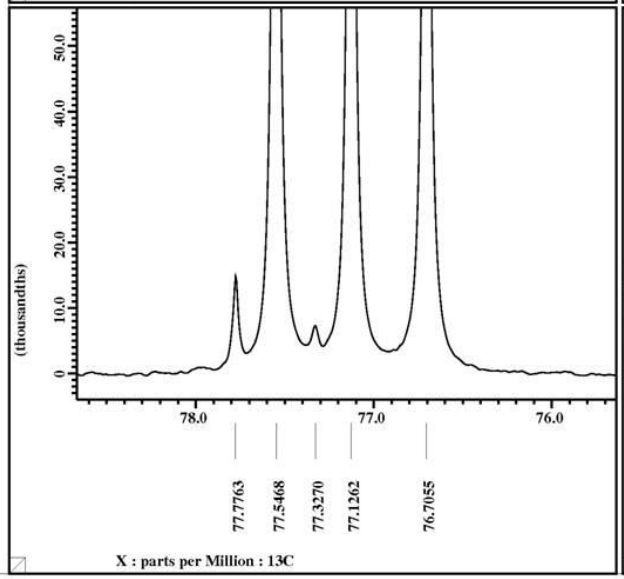


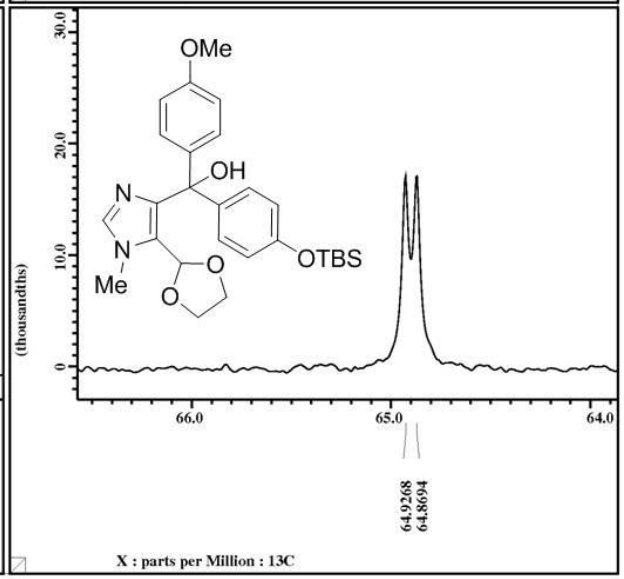

Filename = III_p_135_tertiaryalc
Author = delta
Experiment = single_pulse_dec
Sample_id = S#757963
Solvent = CHLOROFORM-D
Creation_time = 24-JAN-2008 05:59:05
Revision_time = 21-MAR-2010 01:35:43
Current_time = 21-MAR-2010 01:36:42
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.6[dC]
OMe
OH
N
N
Me
O
O
OTBS
abundance
0
0.01
0.02
0.03
0.04
0.05
0.06
0.07
0.08
0.09
0.1
0.11
200.0 190.0 180.0 170.0 160.0 150.0 140.0 130.0 120.0 110.0 100.0 90.0 80.0 70.0 60.0 50.0 40.0 30.0 20.0 10.0 0
158.7788
154.9641
149.4285
139.1413
138.5246
138.1087
129.3416
129.2269
121.5210
119.3938
113.1793
97.3087
77.5468
77.1262
76.7055
64.9268
64.8694
55.3518
33.5010
25.7999
18.3235
-4.3066
X : parts per Million : 13C

```
Filename          = III_p_135_tertiaryalc
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#757963
Solvent           = CHLOROFORM-D
Creation_time     = 24-JAN-2008 05:59:05
Revision_time     = 21-MAR-2010 01:35:43
Current_time      = 21-MAR-2010 01:38:08

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 23.6[dC]
```





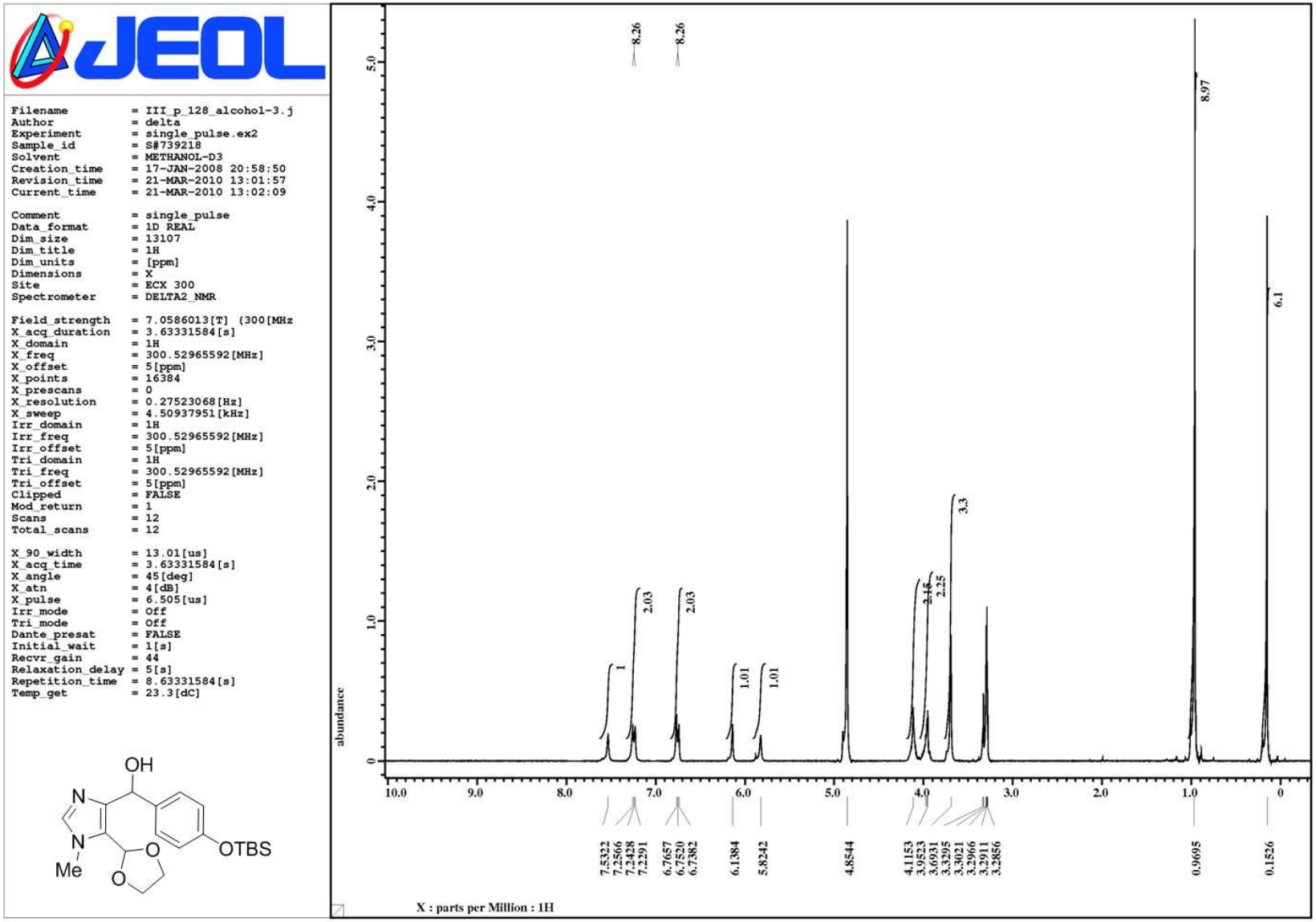




APPENDIX 101

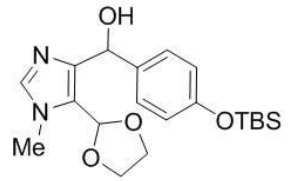
^{1}H Spectrum of

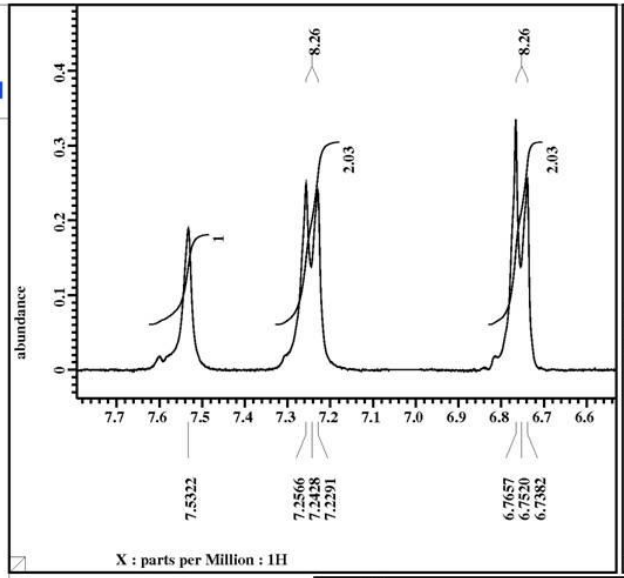
4-*tert*-Butyl-dimethylsilanyloxyphenyl-(5-[1,3]dioxolan-2-yl-1-methyl-1H-imidazol-4-yl)methanol (**230**)

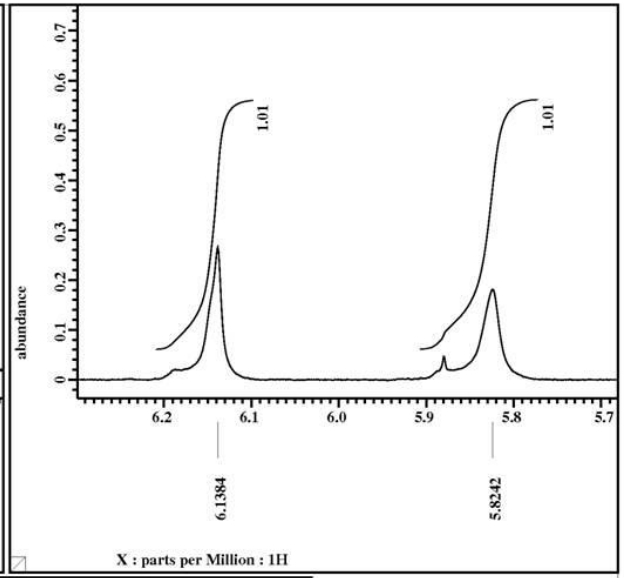

Filename = III_p_128_alcohol-3.j
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#739218
Solvent = METHANOL-D3
Creation_time = 17-JAN-2008 20:58:50
Revision_time = 21-MAR-2010 13:01:57
Current_time = 21-MAR-2010 13:02:09
Comment = single_pulse
Data_format = 1D REAL
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 44
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.3[dC]
OH
N
N
Me
O
O
OTBS
abundance
8.26
8.26
8.97
6.1
3.3
2.25
2.18
2.03
2.03
1
1.01
1.01
7.5322
7.2566
7.2428
7.2291
6.7657
6.7520
6.7382
6.1384
5.8342
4.8544
4.1153
3.9523
3.6931
3.3295
3.3021
3.2966
3.2911
3.2856
0.9695
0.1526
X : parts per Million : 1H

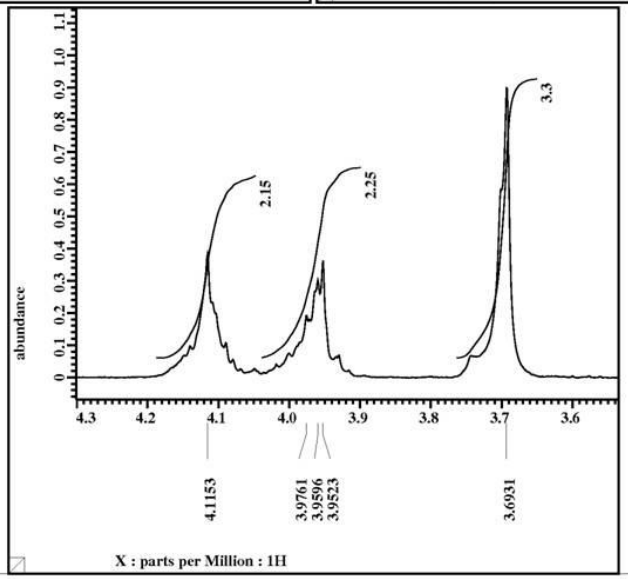
JEOL

```
Filename          = III_p_128_alcohol-3.j
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#739218
Solvent           = METHANOL-D3
Creation_time     = 17-JAN-2008 20:58:50
Revision_time     = 21-MAR-2010 13:02:46
Current_time      = 21-MAR-2010 13:03:00

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 44
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.3[dC]
```







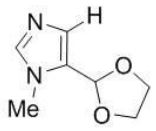


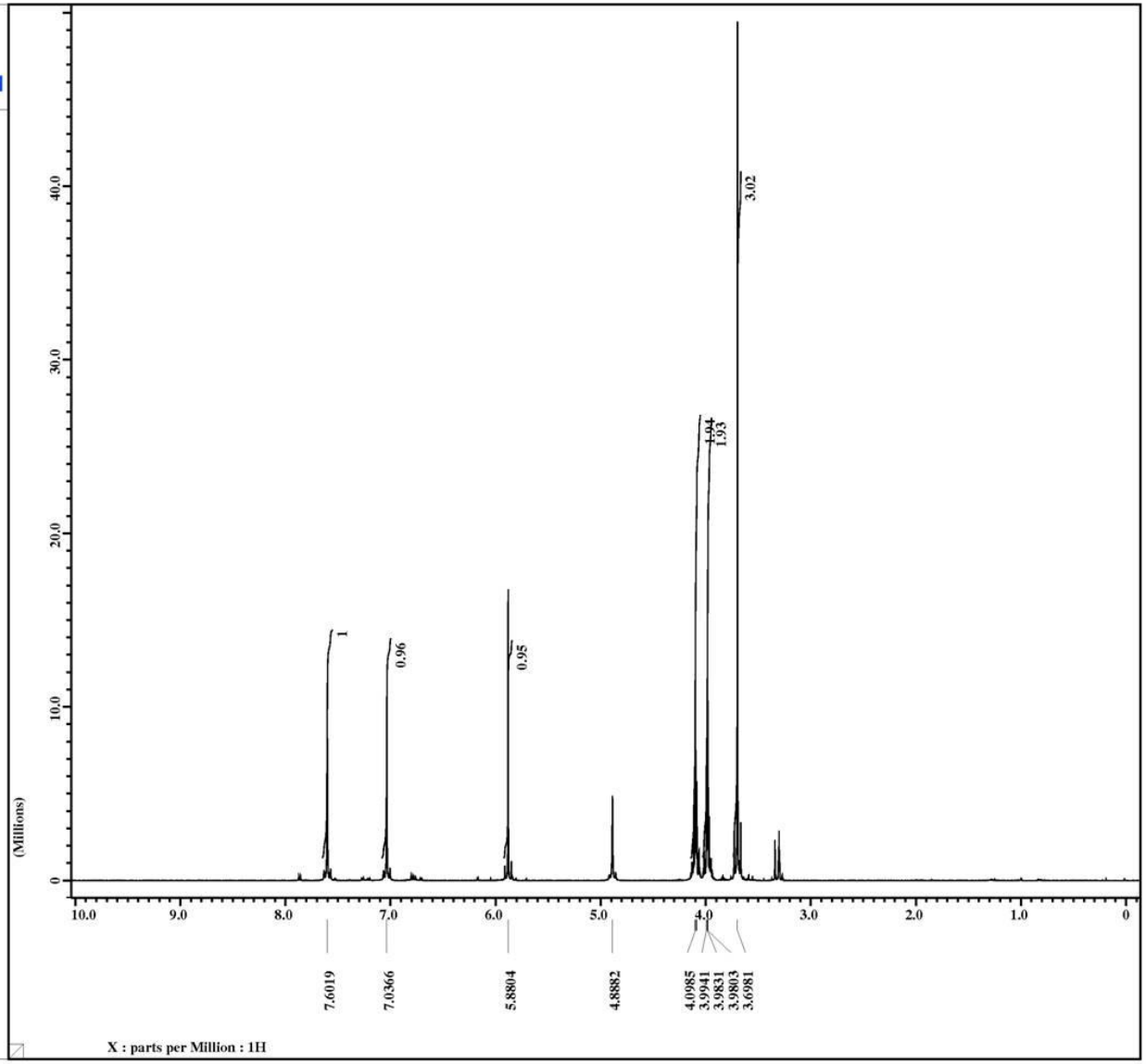
APPENDIX 102

^{1}H Spectrum of

5-[1,3]dioxolan-2-yl-1-methyl-1*H*-imidazole (**231**)

```
Filename          = III_p_139_alcohol-3.j
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#382210
Solvent           = METHANOL-D3
Creation_time     =  5-FEB-2008 16:15:11
Revision_time     = 21-MAR-2010 13:13:39
Current_time      = 21-MAR-2010 13:14:13

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 15
Relaxation_delay  = 4[s]
Temp_get          = 25.1[dC]
Unblank_time      = 2[us]
```

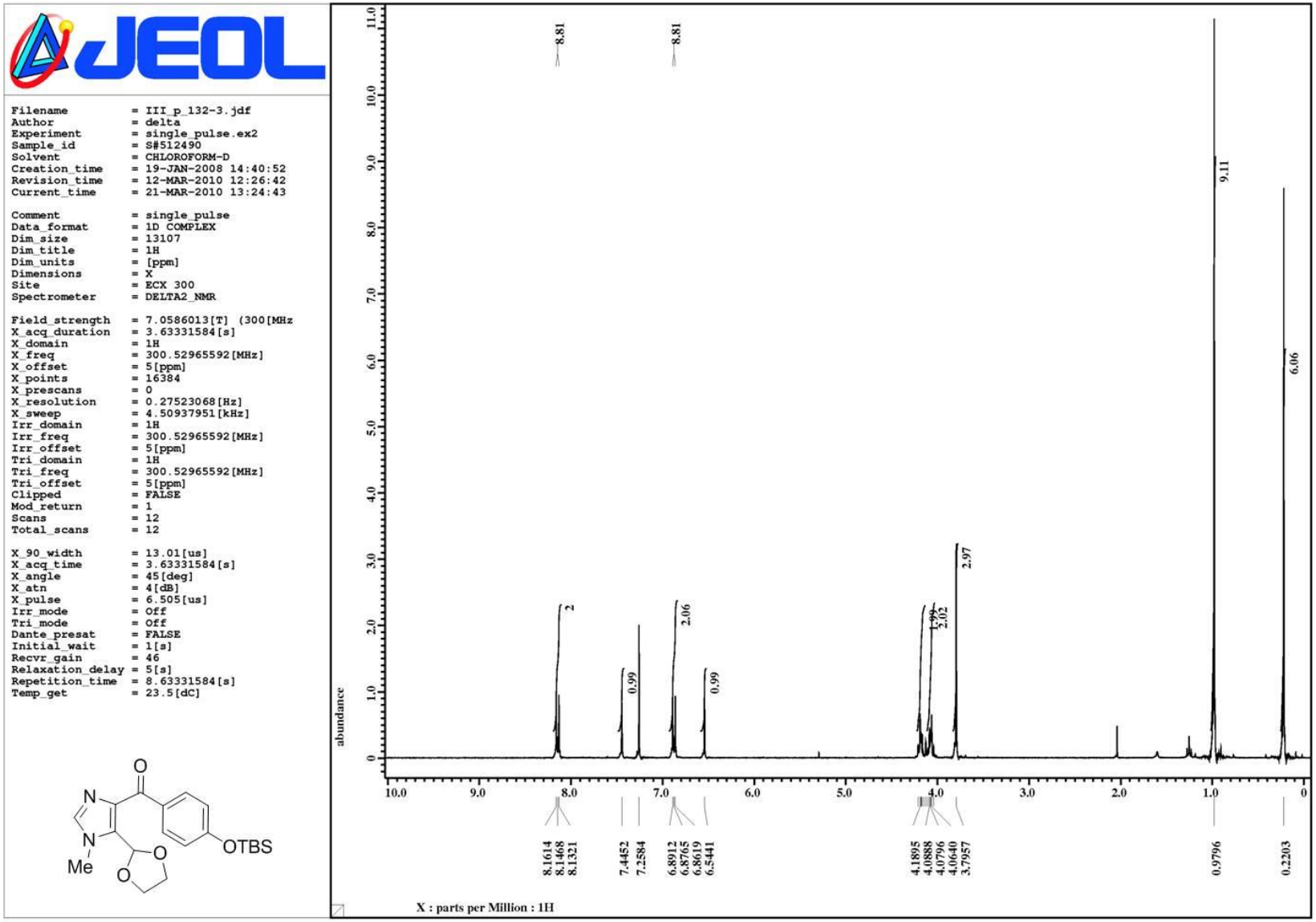




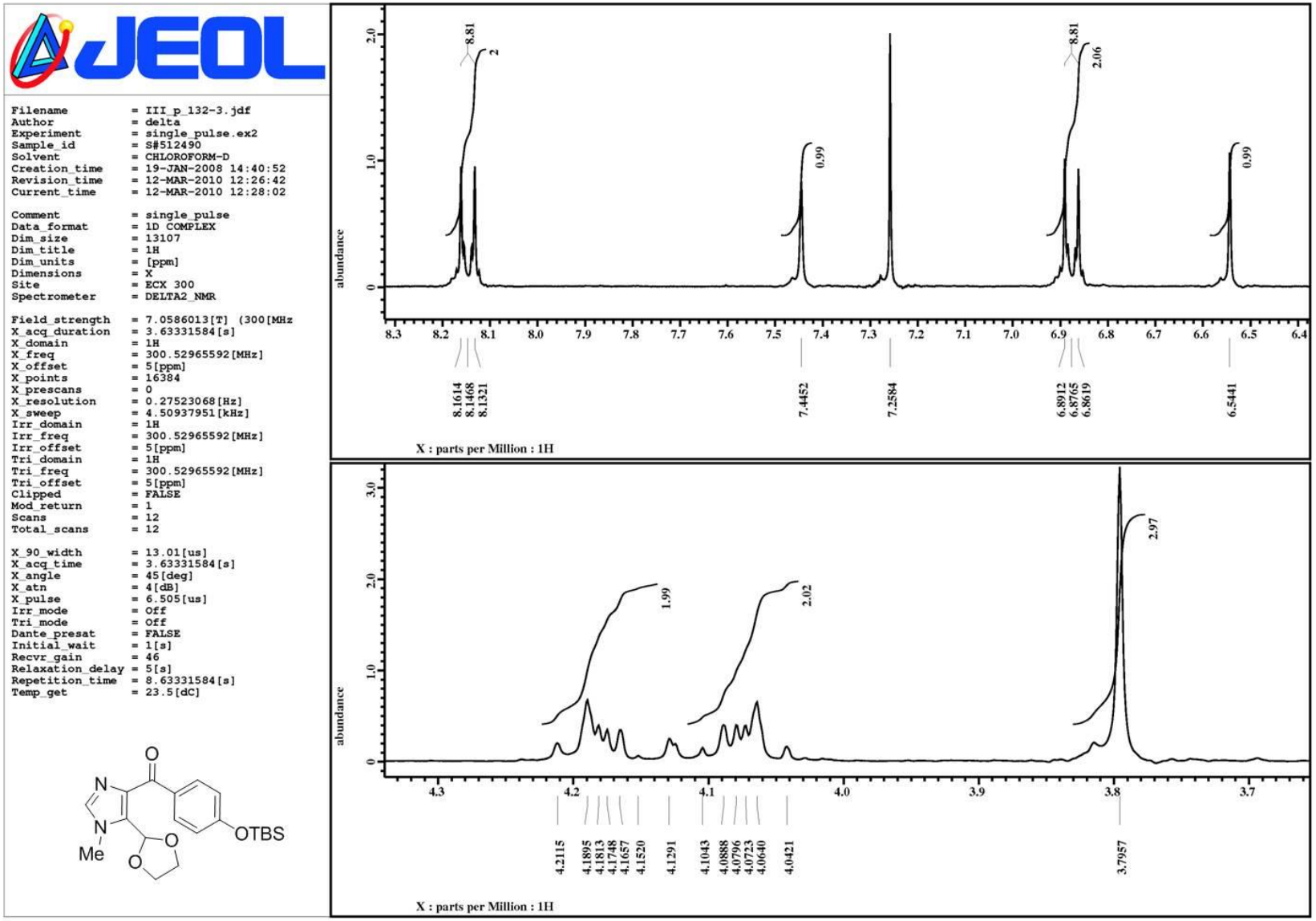
APPENDIX 103

^{1}H and ^{13}C NMR Spectra of

4-*tert*-Butyldimethylsilanyloxyphenyl-(5-[1,3]dioxolan-2-yl-1-methyl-1H-imidazol-4-yl)methanone (**232**)



```
Filename          = III_p_132-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#512490
Solvent           = CHLOROFORM-D
Creation_time     = 19-JAN-2008 14:40:52
Revision_time     = 12-MAR-2010 12:26:42
Current_time      = 21-MAR-2010 13:24:43

Comment           = single_pulse
Data_format       = 1D COMPLEX
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 46
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23.5[dC]
```

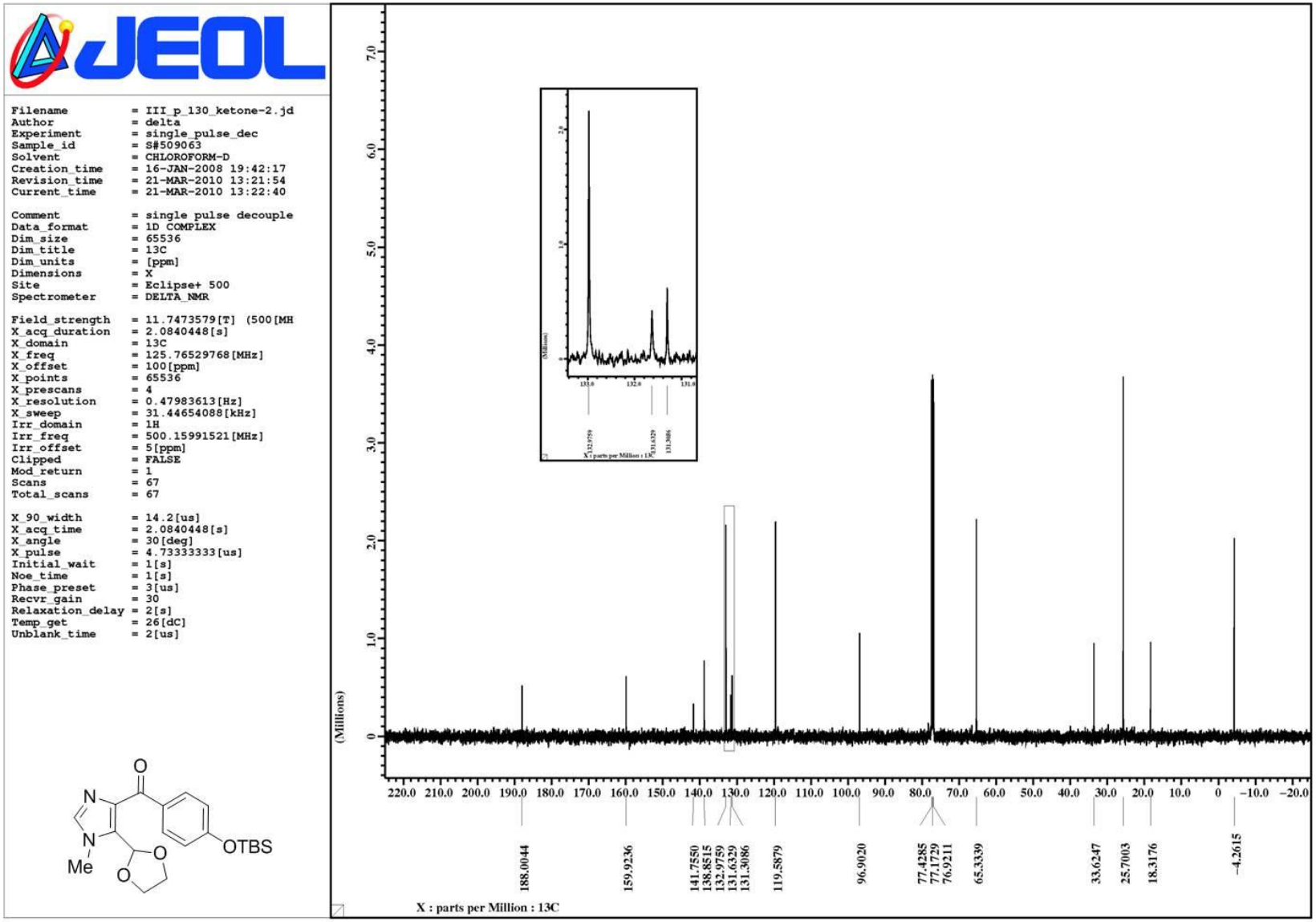


Filename = III_p_132-3.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#512490
Solvent = CHLOROFORM-D
Creation_time = 19-JAN-2008 14:40:52
Revision_time = 12-MAR-2010 12:26:42
Current_time = 12-MAR-2010 12:28:02

Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR

Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12

X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 46
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.5[dC]

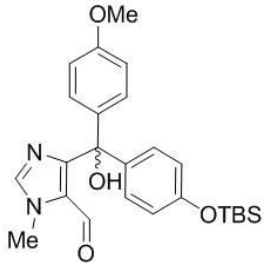

Filename = III_p_130_ketone-2.jd
Author = delta
Experiment = single_pulse_dec
Sample_id = S#509063
Solvent = CHLOROFORM-D
Creation_time = 16-JAN-2008 19:42:17
Revision_time = 21-MAR-2010 13:21:54
Current_time = 21-MAR-2010 13:22:40
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 67
Total_scans = 67
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 26[dC]
Unblank_time = 2[us]
O
N
N
Me
O
O
OTBS
(Millions)
188.0044
159.9236
141.7550
138.8515
132.9759
131.6329
131.3086
119.5879
96.9020
77.4285
77.1729
76.9211
65.3339
33.6247
25.7003
18.3176
-4.2615
X : parts per Million : 13C

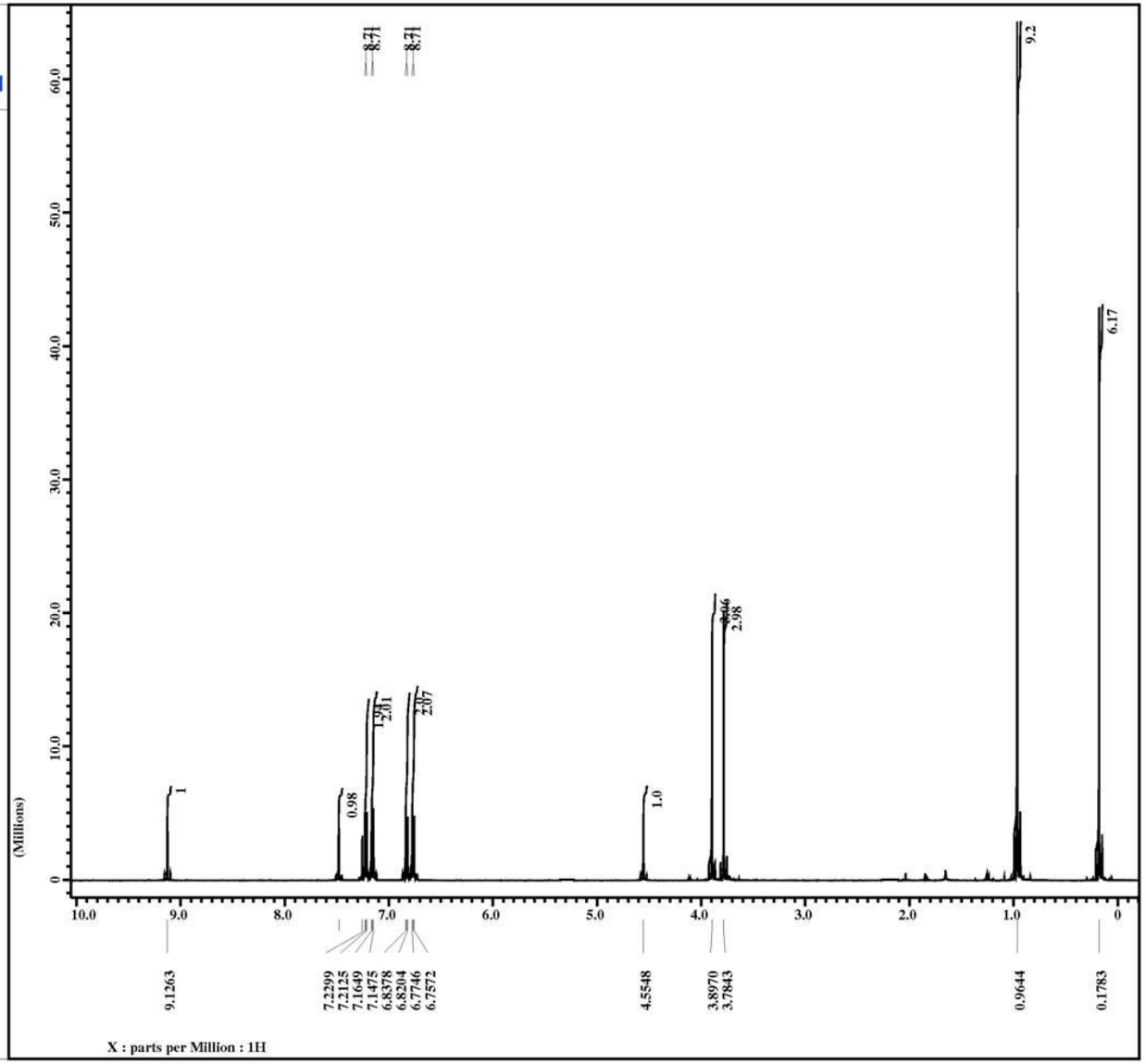
APPENDIX 104

^{1}H and ^{13}C NMR Spectra of

4-[(4-*tert*-Butyldimethylsilanyloxyphenyl)-hadroxy-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole-5-carbaldehyde (**233a**)

Filename	= III_p_168_i-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#540299
Solvent	= CHLOROFORM-D
Creation_time	= 12-FEB-2008 20:43:07
Revision_time	= 12-MAR-2010 16:15:00
Current_time	= 12-MAR-2010 16:15:32
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 17
Relaxation_delay	= 4[s]
Temp_get	= 25.2[dC]
Unblank_time	= 2[us]





JEOL
Filename = III_p_168_i-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#540299
Solvent = CHLOROFORM-D
Creation_time = 12-FEB-2008 20:43:07
Revision_time = 12-MAR-2010 16:17:25
Current_time = 12-MAR-2010 16:18:02
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 17
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
OMe
OH
OTBS
N
N
Me
O
(Millions)
8.71
1.94
8.71
2.01
7.2546
7.2299
7.2212
7.2125
7.1649
7.1562
7.1475
X : parts per Million : 1H
(Millions)
8.71
2.0
8.71
2.07
6.8378
6.8291
6.8204
6.7746
6.7659
6.7572
X : parts per Million : 1H

JEOL
Filename = III_p_168_i-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = panduka
Solvent = CHLOROFORM-D
Creation_time = 13-FEB-2008 10:27:10
Revision_time = 21-MAR-2010 13:44:20
Current_time = 21-MAR-2010 13:44:59
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 4800
Total_scans = 4800
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 27[dC]
Unblank_time = 2[us]
OMe
OH
OTBS
N
N
Me
O
(Millions)
182.4836
159.5726
159.1452
155.3566
141.2170
138.4089
137.8900
129.1453
129.1262
127.4627
119.6108
113.4643
77.3675
77.1156
76.8600
55.3301
35.1318
25.7384
18.2680
-4.3226
X : parts per Million : 13C

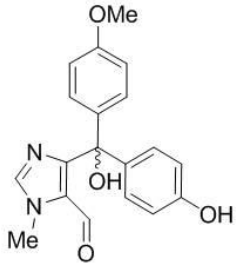
JEOL
Filename = III_p_168_i-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = panduka
Solvent = CHLOROFORM-D
Creation_time = 13-FEB-2008 10:27:10
Revision_time = 21-MAR-2010 13:44:20
Current_time = 21-MAR-2010 13:45:39
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.74735791[T] (500[MH
X_acq_duration = 2.08404481[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 4800
Total_scans = 4800
X_90_width = 14.2[us]
X_acq_time = 2.08404481[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 27[dC]
Unblank_time = 2[us]
(Thousands)
(Millions)
X : parts per Million : 13C
159.5726
159.1452
155.3566
141.2170
138.4089
137.8900
129.1453
129.1262
78.7601
77.3675
77.1156
76.8600
55.3301
OMe
OH
OTBS
N
N
Me
O

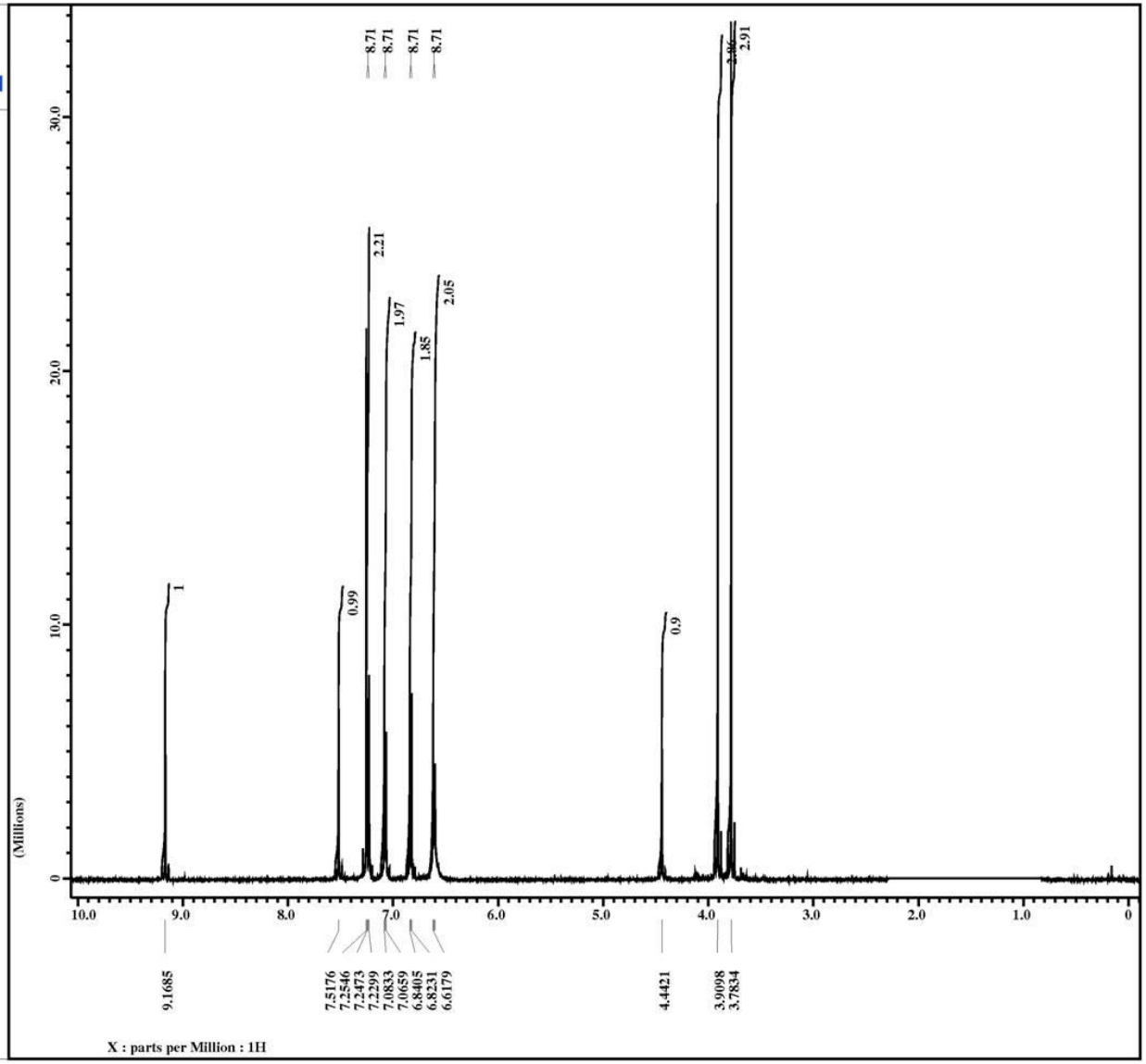
APPENDIX 105

^{1}H and ^{13}C NMR Spectra of

4-[Hydroxy-(4-hydroxyphenyl)-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazole-5-carbaldehyde (**233b**)

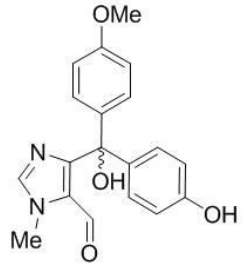
Filename	= III_p_169_i-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#691201
Solvent	= CHLOROFORM-D
Creation_time	= 14-FEB-2008 00:50:22
Revision_time	= 12-MAR-2010 16:32:13
Current_time	= 12-MAR-2010 16:32:26
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 23
Relaxation_delay	= 4[s]
Temp_get	= 25.1[dC]
Unblank_time	= 2[us]

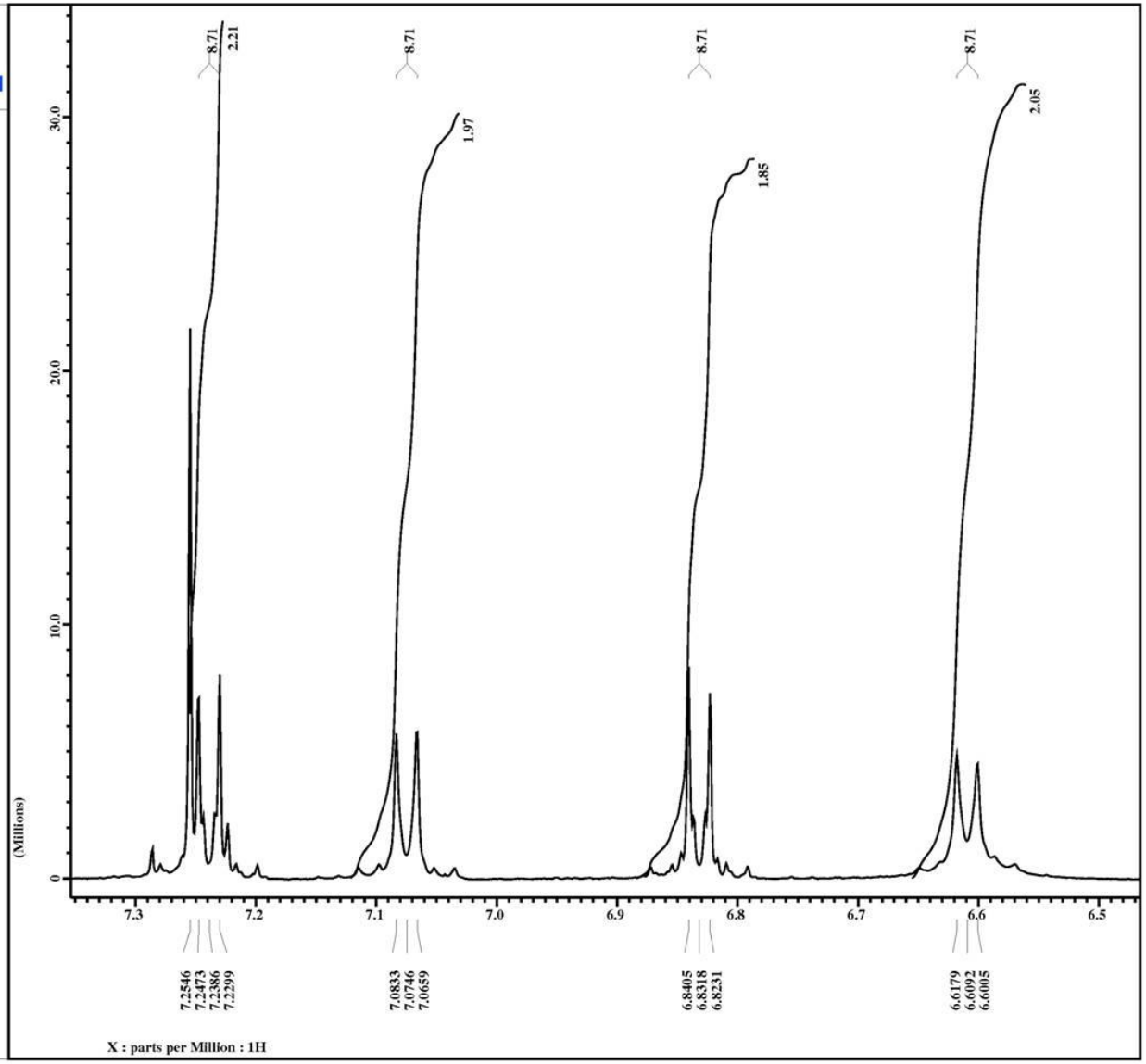


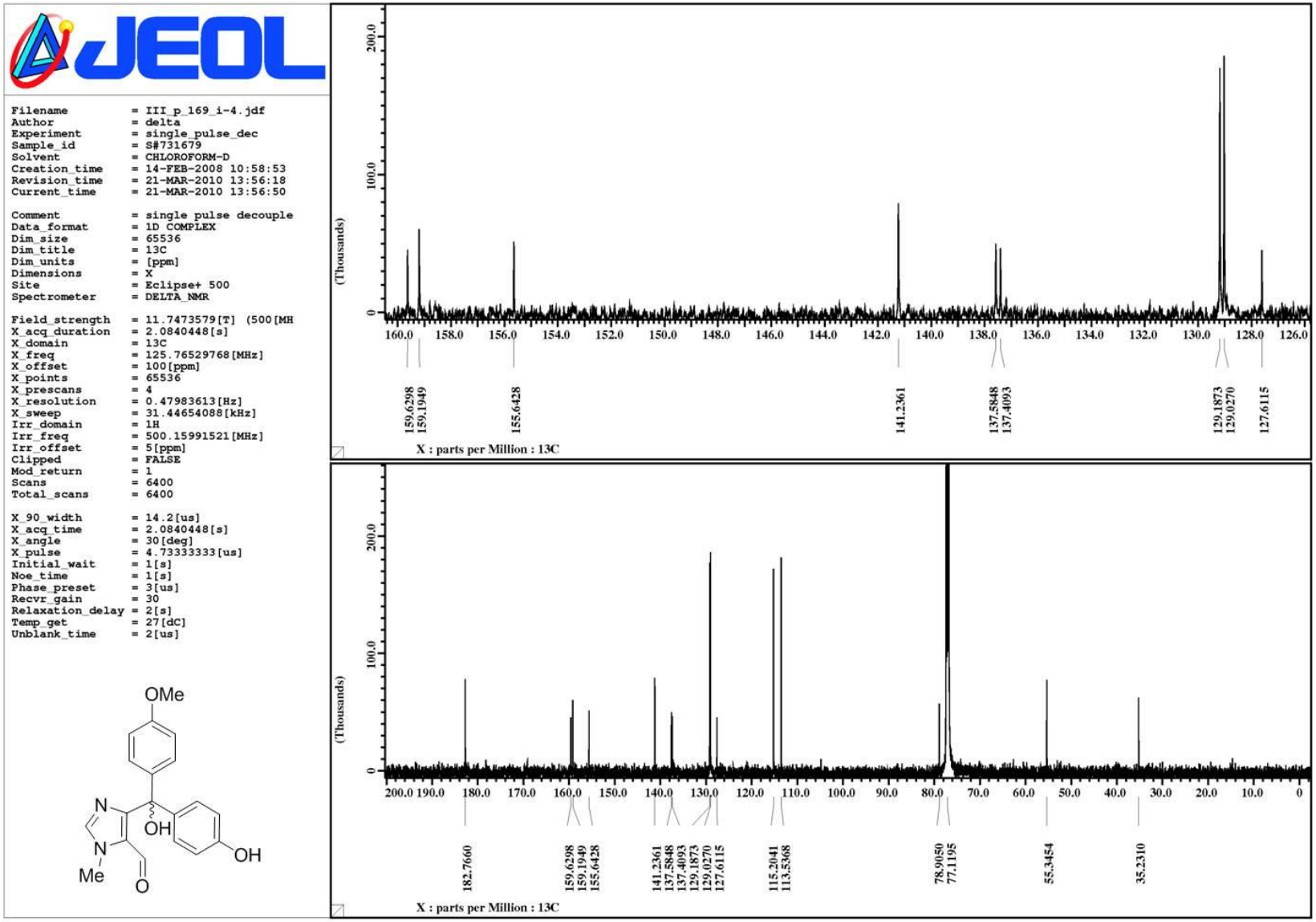


JEOL

Filename	= III_p_169_i-3.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#691201
Solvent	= CHLOROFORM-D
Creation_time	= 14-FEB-2008 00:50:22
Revision_time	= 12-MAR-2010 16:32:13
Current_time	= 12-MAR-2010 16:32:43
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 23
Relaxation_delay	= 4[s]
Temp_get	= 25.1[dC]
Unblank_time	= 2[us]







```
Filename          = III_p_169_i-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#731679
Solvent           = CHLOROFORM-D
Creation_time     = 14-FEB-2008 10:58:53
Revision_time     = 21-MAR-2010 13:56:18
Current_time      = 21-MAR-2010 13:56:50

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 6400
Total_scans       = 6400

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27[dC]
Unblank_time      = 2[us]
```

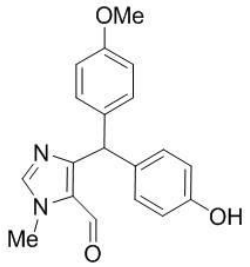
APPENDIX 106

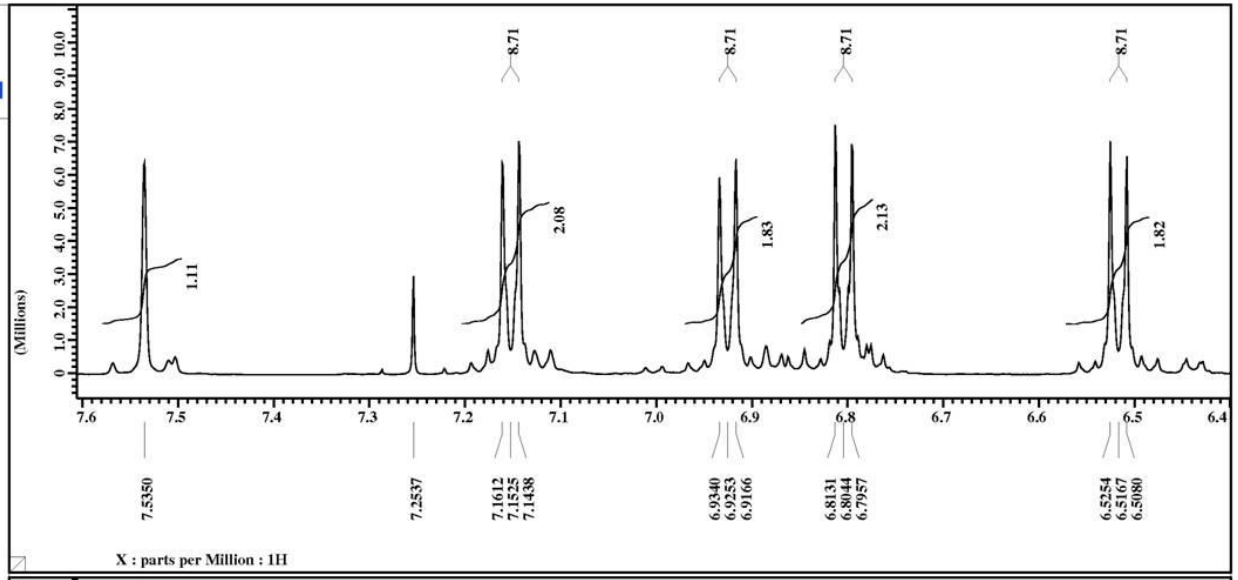
^{1}H and ^{13}C NMR Spectra of

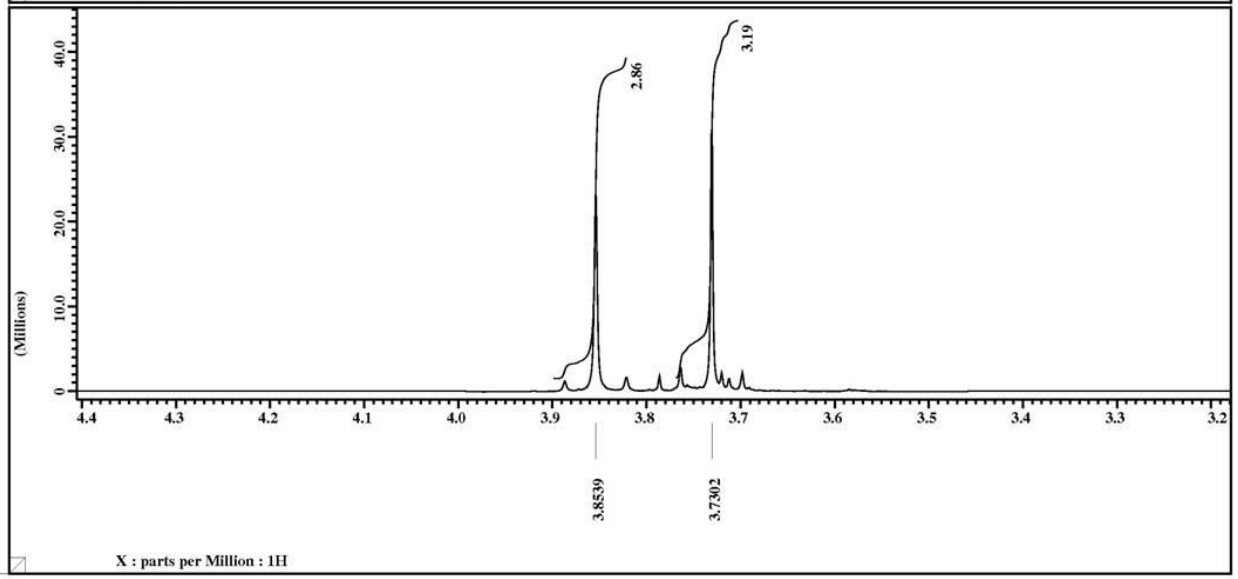
4-[4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl]methyl-1-methyl-1*H*-imidazole-5-carbaldehyde (**234**)

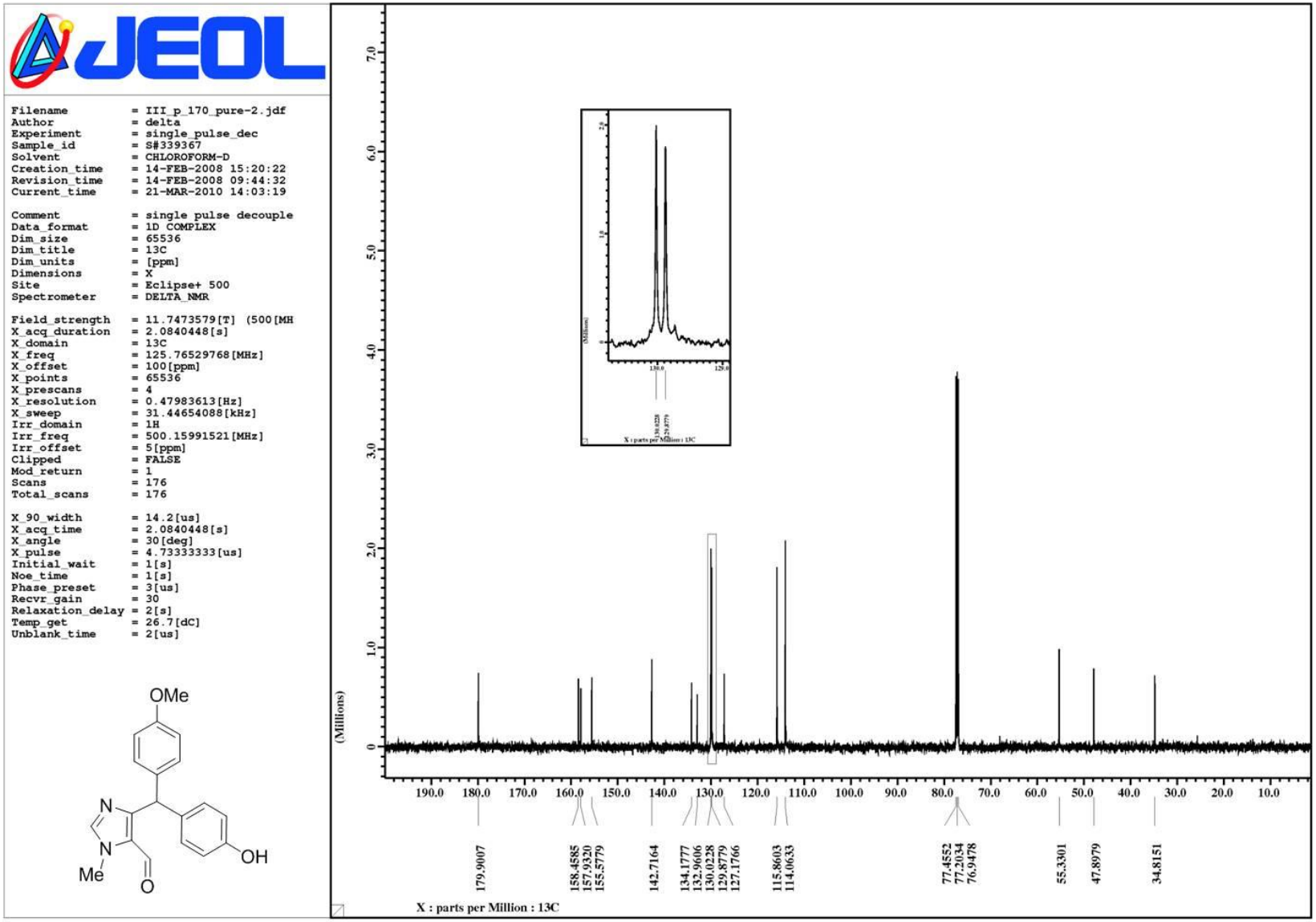
JEOL
Filename = III_p_170_pure-2.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#339306
Solvent = CHLOROFORM-D
Creation_time = 14-FEB-2008 15:03:23
Revision_time = 12-MAR-2010 16:51:43
Current_time = 21-MAR-2010 14:04:31
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 14
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
OMe
OH
N
N
Me
O
(Millions)
X : parts per Million : 1H
9.6760
7.5350
7.2537
7.1612
7.1438
6.9340
6.9166
6.8131
6.7957
6.5254
6.5080
5.7439
3.8539
3.7302
8.71
8.71
8.71
8.71
1.0
1.11
2.08
1.83
2.13
1.82
0.94
2.86
3.19

```
Filename          = III_p_170_pure-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#339306
Solvent           = CHLOROFORM-D
Creation_time     = 14-FEB-2008 15:03:23
Revision_time     = 12-MAR-2010 16:51:43
Current_time      = 21-MAR-2010 14:04:57

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 14
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```









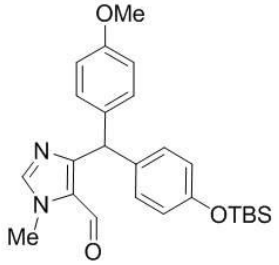
```
Filename          = III_p_170_pure-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#339367
Solvent           = CHLOROFORM-D
Creation_time     = 14-FEB-2008 15:20:22
Revision_time     = 14-FEB-2008 09:44:32
Current_time      = 21-MAR-2010 14:03:19

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 176
Total_scans       = 176

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.7[dC]
Unblank_time      = 2[us]
```

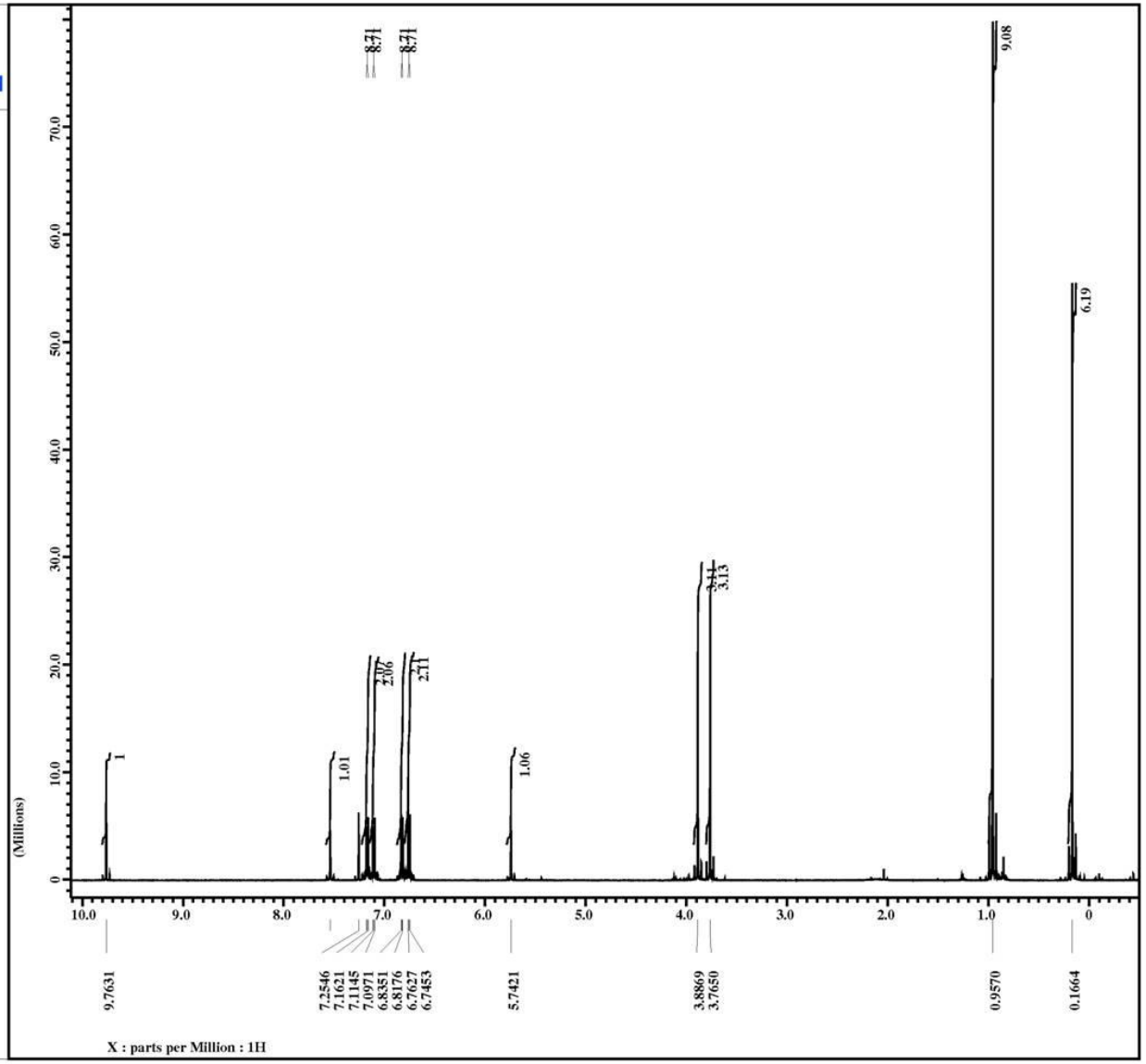
APPENDIX 107

^{1}H and ^{13}C NMR Spectra of

5-[4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl]methyl-1-methyl-1*H*-imidazole-4-carbaldehyde (**226**)

```
Filename          = IV_p_002_product-3.jd
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#701810
Solvent           = CHLOROFORM-D
Creation_time     =  2-MAY-2008 01:40:46
Revision_time     = 12-MAR-2010 18:06:34
Current_time      = 12-MAR-2010 18:08:38

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 18
Relaxation_delay  = 4[s]
Temp_get          = 25.2[dC]
Unblank_time      = 2[us]
```

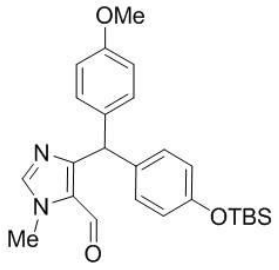


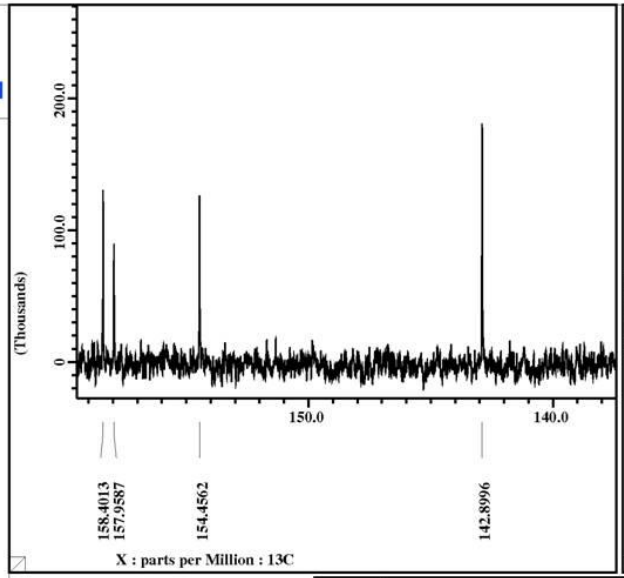


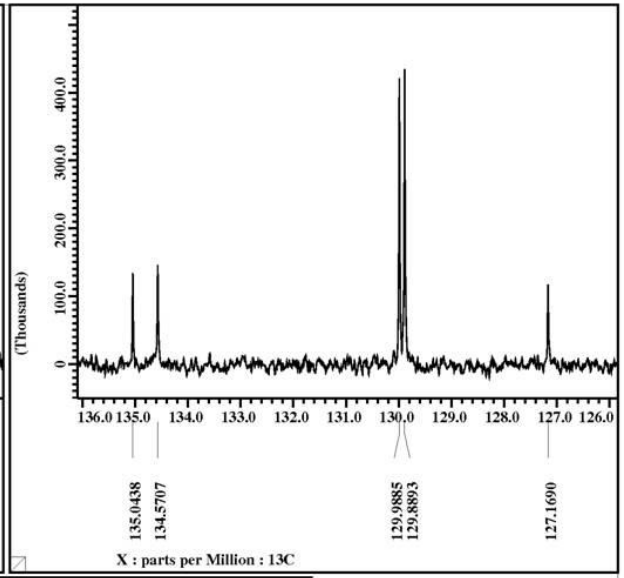
JEOL
Filename = IV_p_002_product-3.jd
Author = delta
Experiment = single_pulse.exp
Sample_id = S#701810
Solvent = CHLOROFORM-D
Creation_time = 2-MAY-2008 01:40:46
Revision_time = 12-MAR-2010 18:06:34
Current_time = 12-MAR-2010 18:09:04
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 18
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
OMe
OTBS
N
N
Me
O
(Millions)
X : parts per Million : 1H
2.11
2.1
2.07
2.06
8.71
8.71
8.71
8.71
7.1793
7.1708
7.1621
7.1145
7.1058
7.0971
6.8351
6.8263
6.8176
6.7627
6.7536
6.7453

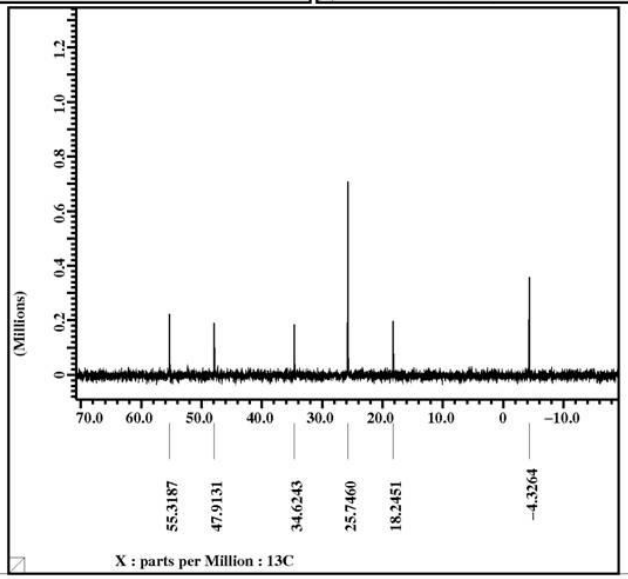
JEOL
Filename = IV_p_002_product-2.jd
Author = delta
Experiment = single_pulse_dec
Sample_id = S#703714
Solvent = CHLOROFORM-D
Creation_time = 2-MAY-2008 03:58:51
Revision_time = 2-MAY-2008 11:41:13
Current_time = 21-MAR-2010 01:05:39
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 1600
Total_scans = 1600
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 26.7[dC]
Unblank_time = 2[us]
OMe
OTBS
N
N
Me
O
(Millions)
X : parts per Million : 13C
179.4886
158.4013
157.9587
154.4562
142.8996
135.0438
134.5707
129.9885
129.8893
127.1690
120.0267
113.9641
77.3675
77.1156
76.8600
55.3187
47.9131
34.6243
25.7460
18.2451
-4.3264

```
Filename          = IV_p_002_product-2.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#703714
Solvent           = CHLOROFORM-D
Creation_time     =  2-MAY-2008 03:58:51
Revision_time     =  2-MAY-2008 11:41:13
Current_time      = 21-MAR-2010 01:05:21

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 1600
Total_scans       = 1600

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.7[dC]
Unblank_time      = 2[us]
```







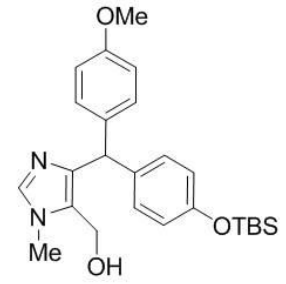


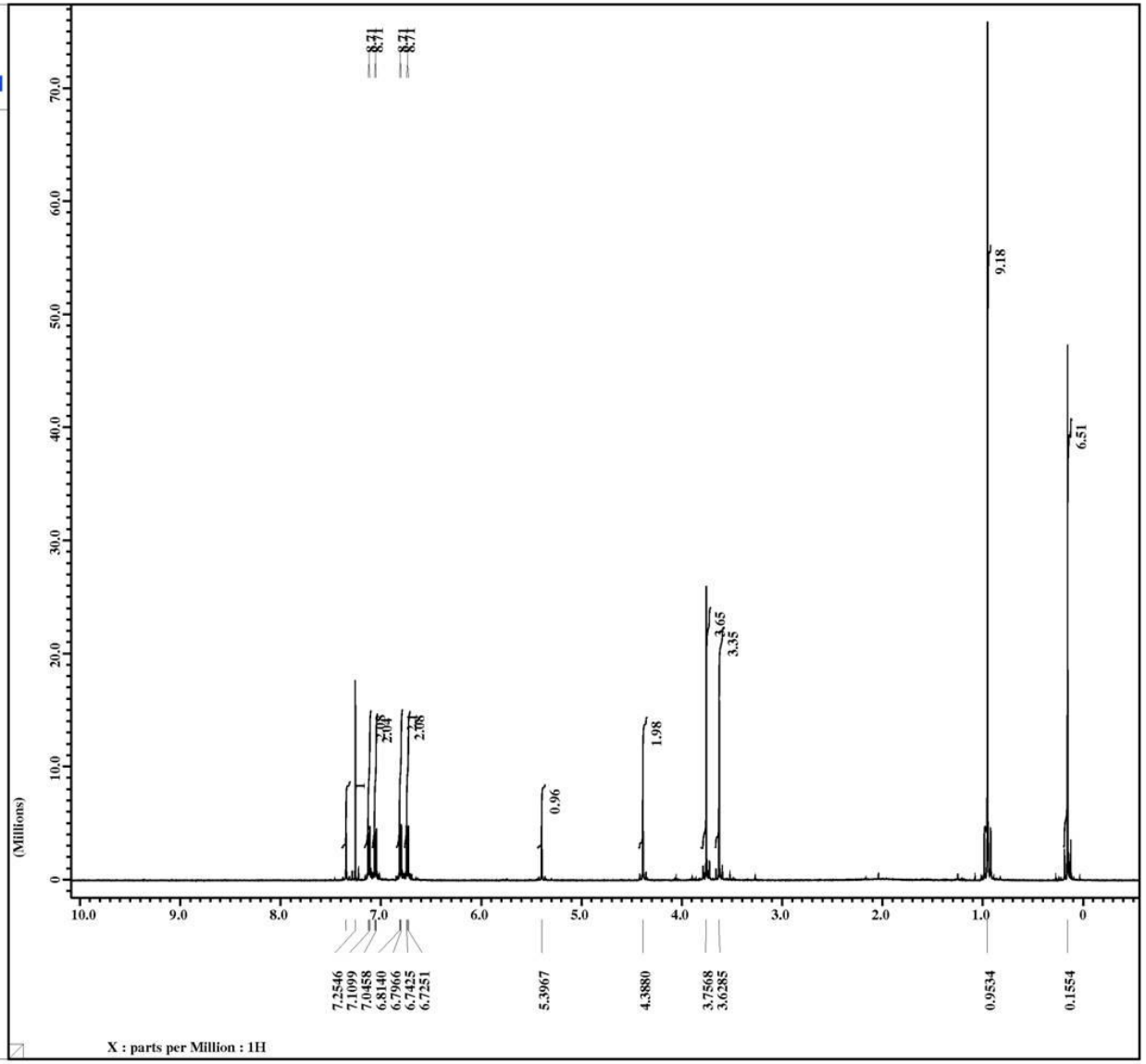
APPENDIX 108

^{1}H and ^{13}C NMR Spectra of

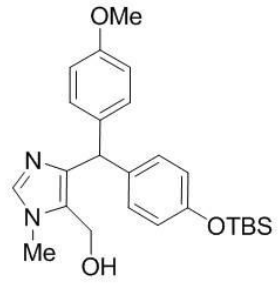
4-(4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl)methyl-5-hydroxymethyl-1-methyl-1*H*-imidazole (**225**)

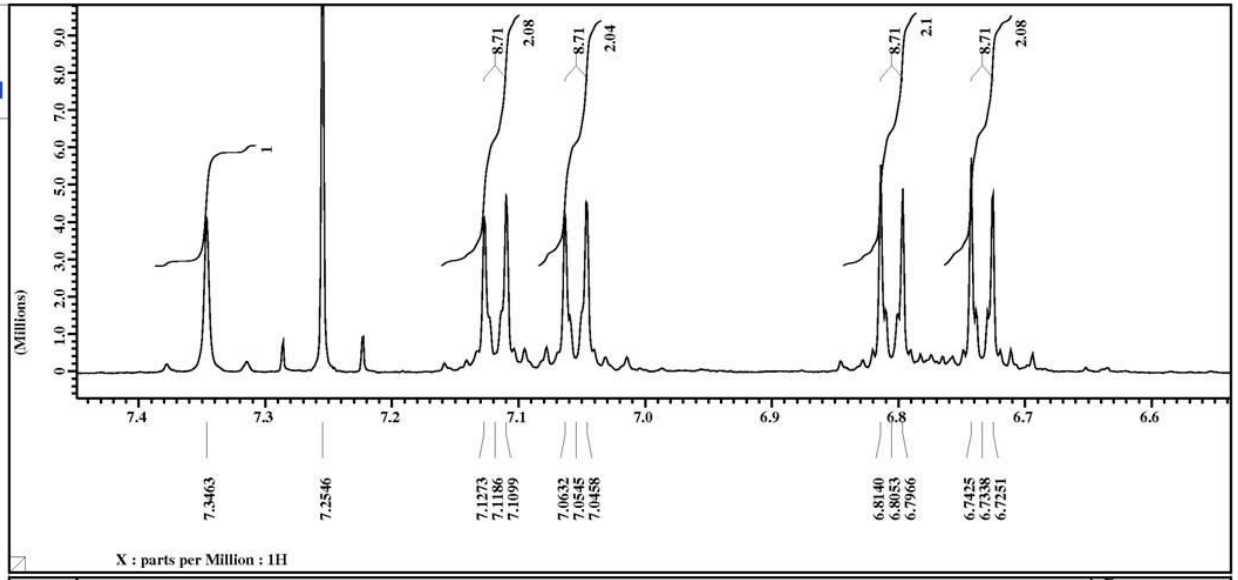
```
Filename          = III_p_137_i-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#700884
Solvent           = CHLOROFORM-D
Creation_time     = 26-JAN-2008 01:00:18
Revision_time     = 12-MAR-2010 18:24:45
Current_time      = 21-MAR-2010 00:54:36

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 22
Relaxation_delay  = 4[s]
Temp_get          = 24.9[dC]
Unblank_time      = 2[us]
```

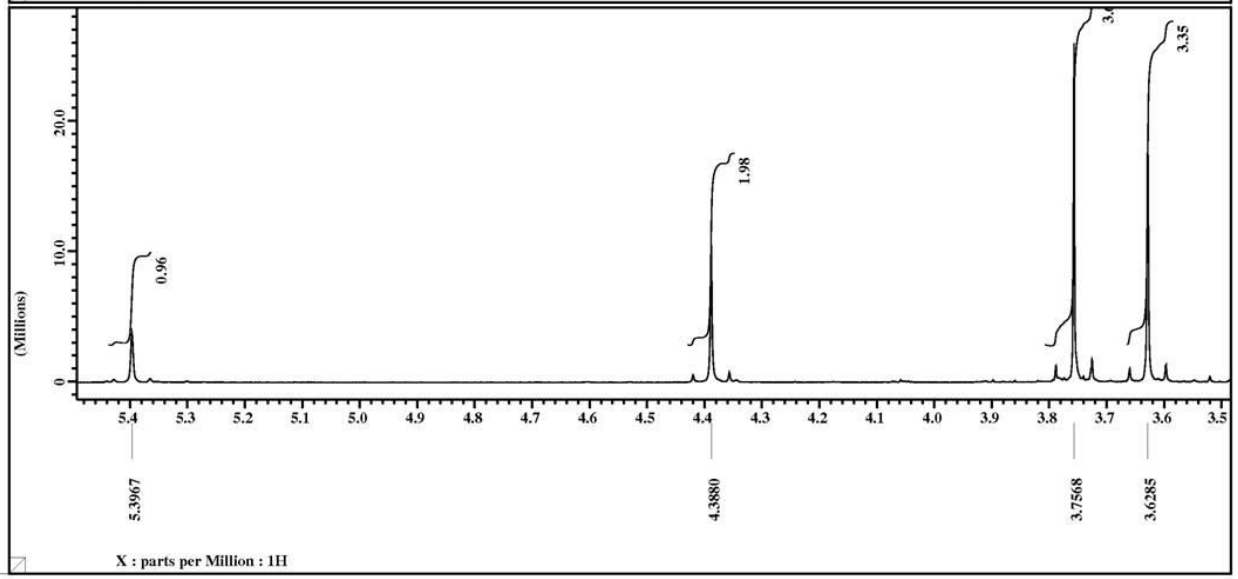




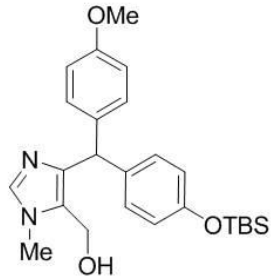
```
Filename          = III_p_137_i-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#700884
Solvent           = CHLOROFORM-D
Creation_time     = 26-JAN-2008 01:00:18
Revision_time     = 12-MAR-2010 18:24:45
Current_time      = 21-MAR-2010 00:55:06

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 22
Relaxation_delay  = 4[s]
Temp_get          = 24.9[dC]
Unblank_time      = 2[us]
```

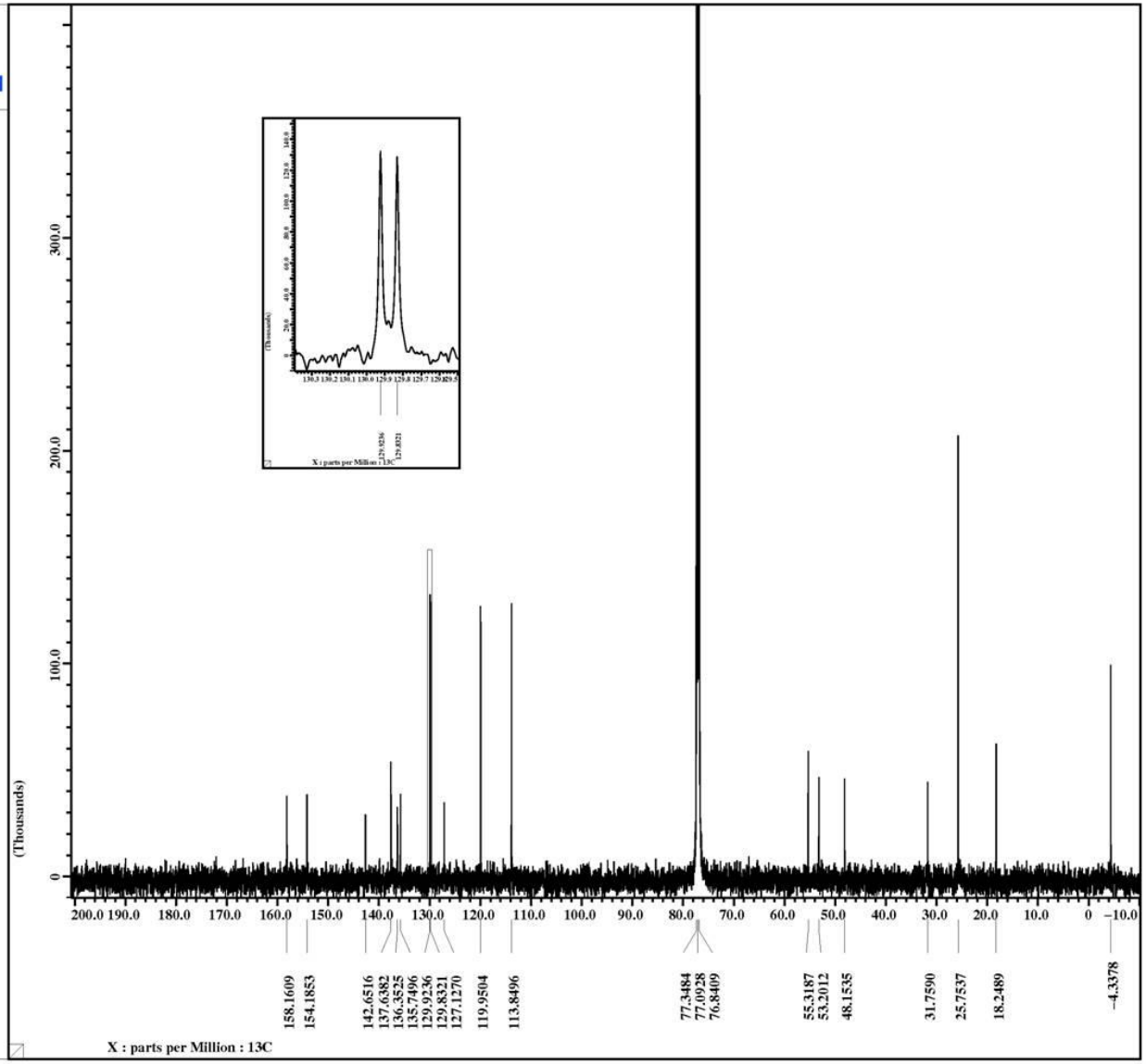






```
Filename          = III_p_137_i-2.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#702457
Solvent           = CHLOROFORM-D
Creation_time     = 26-JAN-2008 08:06:16
Revision_time     = 21-MAR-2010 00:50:49
Current_time      = 21-MAR-2010 00:51:53

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 5000
Total_scans       = 5000

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 26.9[dC]
Unblank_time      = 2[us]
```





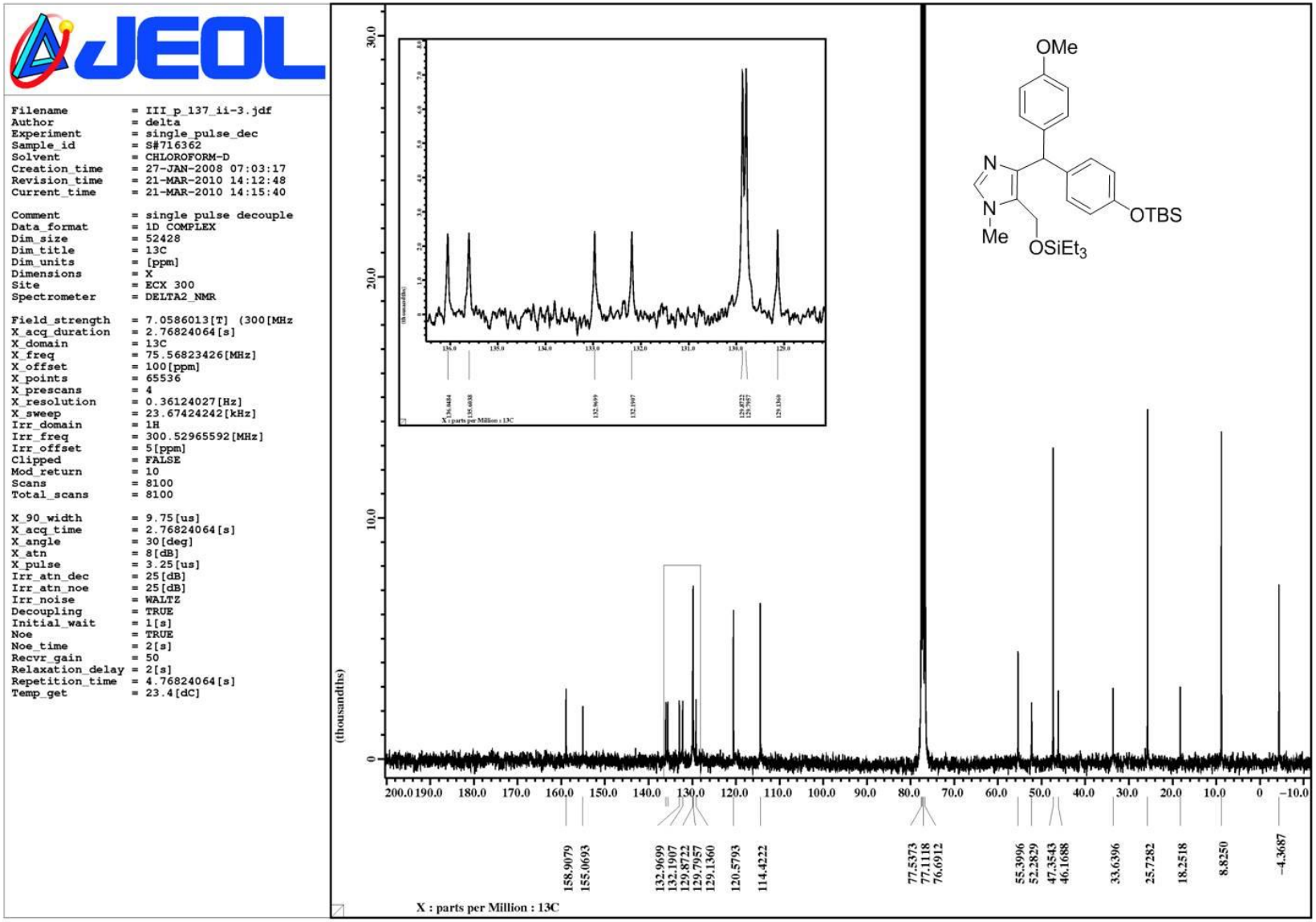
APPENDIX 109

^{1}H and ^{13}C NMR Spectra of

4-(4-*tert*-Butyldimethylsilanyloxyphenyl-4-methoxyphenyl)methyl-5-triethylsilanyloxymethyl-1-methyl-1*H*-imidazole (**235**)

JEOL
Filename = III_p_137_ii-2.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#395508
Solvent = CHLOROFORM-D
Creation_time = 25-JAN-2008 11:26:49
Revision_time = 12-MAR-2010 18:41:50
Current_time = 12-MAR-2010 18:42:14
Comment = single pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.509377951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 46
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.3[dC]
OMe
OTBS
OSiEt3
N
N
Me
7.16
8.99
9.07
5.95
6.09
6.85
2.1
0.96
2.51
abundance
X : parts per Million : 1H
7.9352
7.2584
7.0267
6.9663
6.8408
6.8115
6.7813
5.4945
4.3040
3.7737
3.7563
3.2288
3.2050
3.1803
2.1665
1.3743
1.3505
1.3257
0.9557
0.1636

JEOL
Filename = III_p_137_ii-2.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#395508
Solvent = CHLOROFORM-D
Creation_time = 25-JAN-2008 11:26:49
Revision_time = 12-MAR-2010 18:47:42
Current_time = 12-MAR-2010 18:49:05
Comment = single pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 46
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.3[dC]
OMe
OTBS
OSiEt3
Me
N
N
abundance
X : parts per Million : 1H
7.9352
7.2584
7.0560
7.0414
7.0267
6.9947
6.9805
6.9663
6.8408
6.8261
6.8115
6.7813
6.7671
6.7529
8.81
2.09
8.53
2.22
8.81
2.28
8.53
2.51
1
6.85
6.09
7.16
3.7737
3.7563
3.2288
3.2169
3.2050
3.1803
3.1555
8.99
1.3743
1.3624
1.3505
1.3257


Filename = III_p_137_ii-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#716362
Solvent = CHLOROFORM-D
Creation_time = 27-JAN-2008 07:03:17
Revision_time = 21-MAR-2010 14:12:48
Current_time = 21-MAR-2010 14:15:40
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 8100
Total_scans = 8100
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.4[dC]
OMe
N
N
Me
OSiEt3
OTBS
(thousandths)
158.9079
155.0693
132.9699
132.1907
129.8722
129.7957
129.1360
120.5793
114.4222
77.5373
77.1118
76.6912
55.3996
52.2829
47.3543
46.1688
33.6396
25.7282
18.2518
8.8250
−4.3687
X : parts per Million : 13C

APPENDIX 110

^{1}H and ^{13}C NMR Spectra of

5-(4-*tert*-Butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-4-iodo-1*H*-imidazole (**236**)

Filename = VI_p_029-2.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#17248
Solvent = CHLOROFORM-D
Creation_time = 7-NOV-2009 00:34:42
Revision_time = 7-NOV-2009 00:33:19
Current_time = 7-NOV-2009 00:33:28
Content = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.05860131[T] (300[MH
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
tri_freq = 300.52965592[MHz]
tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 40
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23[dC]
Delay_of_start = 1.999999974[s]
Actual_start_time = 7-NOV-2009 00:32:58
End_time = 7-NOV-2009 00:34:43
Local_time = 7-NOV-2009 00:34:41
VI_p_029-2.jdf
abundance
X : parts per Million : 1H
I
DMAS
OTBS

VI_p_029-3.jdf
Filename = VI_p_029-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#18547
Solvent = CHLOROFORM-D
Creation_time = 7-NOV-2009 09:04:57
Revision_time = 9-NOV-2009 23:37:40
Current_time = 9-NOV-2009 23:38:04
Content = single pulse decoupl
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.05860131[T] (300[MH
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 22.7[dC]
Delay_of_start = 1.999999974[s]
Actual_start_time = 7-NOV-2009 00:36:00
End_time = 7-NOV-2009 09:05:41
Local_time = 7-NOV-2009 09:04:56
abundance
X : parts per Million : 13C
-4.330
18.314
25.762
30.045
37.564
76.701
77.126
77.547
90.640
120.288
129.141
129.858
132.654
139.724
154.634
DMAS
OTBS

APPENDIX 111

^{1}H and ^{13}C NMR Spectra of

5-(4-*tert*-Butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-4-formyl-1*H*-imidazole (**237**)

VI_p_030-aldehyde-2.jdf
Filename = VI_p_030-aldehyde-2.
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#852428
Solvent = CHLOROFORM-D
Creation_time = 9-NOV-2009 23:46:58
Revision_time = 9-NOV-2009 23:47:06
Current_time = 9-NOV-2009 23:47:15
Content = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MH
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 38
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.5[dC]
Delay_of_start = 1.99999974[s]
Actual_start_time = 9-NOV-2009 23:45:14
End_time = 9-NOV-2009 23:46:59
Local_time = 9-NOV-2009 23:46:58
abundance
X : parts per Million : 1H
10.037
7.948
7.259
6.985
6.970
6.956
6.739
6.725
6.711
4.527
2.561
0.945
0.139
8.53
8.53
1
1.03
2.02
2.16
1.98
6.16
9.14
6.13
CHO
OTBS
DMAS
N
N

VI_p_030_aldehyde-3.jdf
Filename = VI_p_030_aldehyde-3.
Author = delta
Experiment = single_pulse_dec
Sample_id = S#854251
Solvent = CHLOROFORM-D
Creation_time = 10-NOV-2009 08:16:29
Revision_time = 10-NOV-2009 13:40:35
Current_time = 10-NOV-2009 13:41:17
Content = single pulse decoupl
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MH
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.5[dC]
Delay_of_start = 1.999999974[s]
Actual_start_time = 9-NOV-2009 23:47:32
End_time = 10-NOV-2009 08:17:13
Local_time = 10-NOV-2009 08:16:29
CHO
OTBS
N
N
DMAS
abundance
187.480
154.716
138.969
138.682
137.047
129.480
129.260
120.374
77.556
77.131
76.710
37.703
28.386
25.747
18.295
-4.354
X : parts per Million : 13C

APPENDIX 112

^{1}H and ^{13}C NMR Spectra of

5-(4-*tert*-Butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-4-hydroxymethyl-1*H*-imidazole (**238**)

Filename = VI_p_031_alcohol-2.jd
Author = delta
Experiment = single_pulse.exp
Sample_id = S#803765
Solvent = CHLOROFORM-D
Creation_time = 13-NOV-2009 13:31:55
Revision_time = 12-NOV-2009 22:24:55
Current_time = 12-NOV-2009 22:25:01
Content = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7465928[T] (500[MH
X_acq_duration = 2.1839872[s]
X_domain = 1H
X_freq = 500.12734003[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45787814[Hz]
X_sweep = 7.50187547[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1839872[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 19
Relaxation_delay = 4[s]
Temp_get = 25.8[dC]
Unblank_time = 2[us]
VI_p_031_alcohol-2.jdf
(Millions)
30.0
20.0
10.0
0
X : parts per Million : 1H
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
7.9343
7.2613
6.9903
6.9739
6.7587
6.7551
6.7413
2.242
2.166
2.15
1.919
6.015
9.191
5.934
4.5358
4.1595
2.5537
0.9589
0.1541
I
OH
OTBS
N
N
DMAS

VI_p_031_alcohol-2.jdf
Filename = VI_p_031_alcohol-2.j
Author = delta
Experiment = single_pulse_dec
Sample_id = S#829584
Solvent = CHLOROFORM-D
Creation_time = 19-NOV-2009 07:36:20
Revision_time = 19-NOV-2009 22:33:04
Current_time = 20-NOV-2009 21:49:33
Content = single pulse decoupl
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.05860131[T] (300[MH
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.568234261[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.8[dC]
Delay_of_start = 1.99999974[s]
Actual_start_time = 18-NOV-2009 23:07:23
End_time = 19-NOV-2009 07:37:04
Local_time = 19-NOV-2009 07:36:20
abundance
X : parts per Million : 13C
154.543
141.225
138.180
130.685
129.069
126.779
120.345
77.547
77.126
76.701
57.140
37.569
28.137
25.757
18.309
-4.350
OH
OTBS
N
N
DMAS

APPENDIX 113

^{1}H and ^{13}C NMR Spectra of

4-Bromomethyl-5-(4-*tert*-butyldimethylsilanyloxybenzyl)-1-*N*,*N*-dimethylsulfonyl-1*H*-imidazole (**239a**)

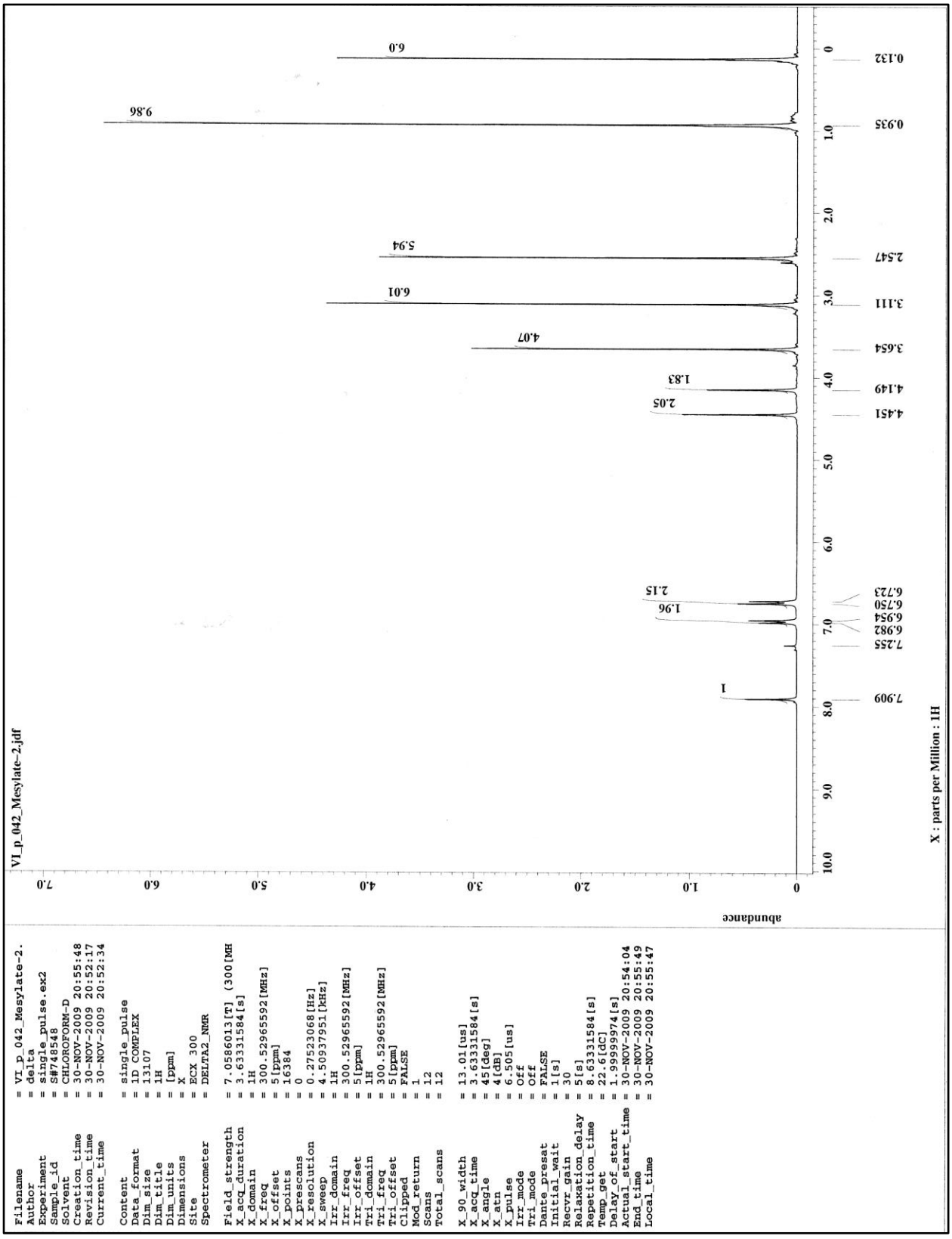
VI_p_036_bromide-2.jdf
Filename = VI_p_036_bromide-2.j
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#780597
Solvent = CHLOROFORM-D
Creation_time = 24-NOV-2009 21:48:18
Revision_time = 24-NOV-2009 21:46:56
Current_time = 24-NOV-2009 21:47:01
Content = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MH
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 50
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.4[dC]
Delay_of_start = 1.99999974[s]
Actual_start_time = 24-NOV-2009 21:46:33
End_time = 24-NOV-2009 21:48:18
Local_time = 24-NOV-2009 21:48:18
abundance
X : parts per Million : 1H
6.11
8.84
6
1.98
1.94
2.2
2.04
0.89
7.932
7.259
7.016
6.986
6.779
6.750
4.365
4.169
2.571
0.962
0.160
Br
OTBS
N
N
DMAS

VI_p_03f_bromide-2.jdf
Filename = VI_p_036_bromide-2.j
Author = delta
Experiment = single_pulse_dec
Sample_id = S#784300
Solvent = CHLOROFORM-D
Creation_time = 25-NOV-2009 06:21:31
Revision_time = 25-NOV-2009 12:32:34
Current_time = 25-NOV-2009 12:34:28
Content = single pulse decoupl
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MH
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.3[dC]
Delay_of_start = 1.99999974[s]
Actual_start_time = 24-NOV-2009 21:52:33
End_time = 25-NOV-2009 06:22:14
Local_time = 25-NOV-2009 06:21:30
abundance
154.720
138.309
129.872
129.189
128.118
120.455
77.533
77.112
76.686
37.602
28.472
25.757
25.245
18.319
-4.340
X : parts per Million : 13C
Br
N
N
DMAS
OTBS

APPENDIX 114

^{1}H and ^{13}C NMR Spectra of

Unknown compound (**239b**)

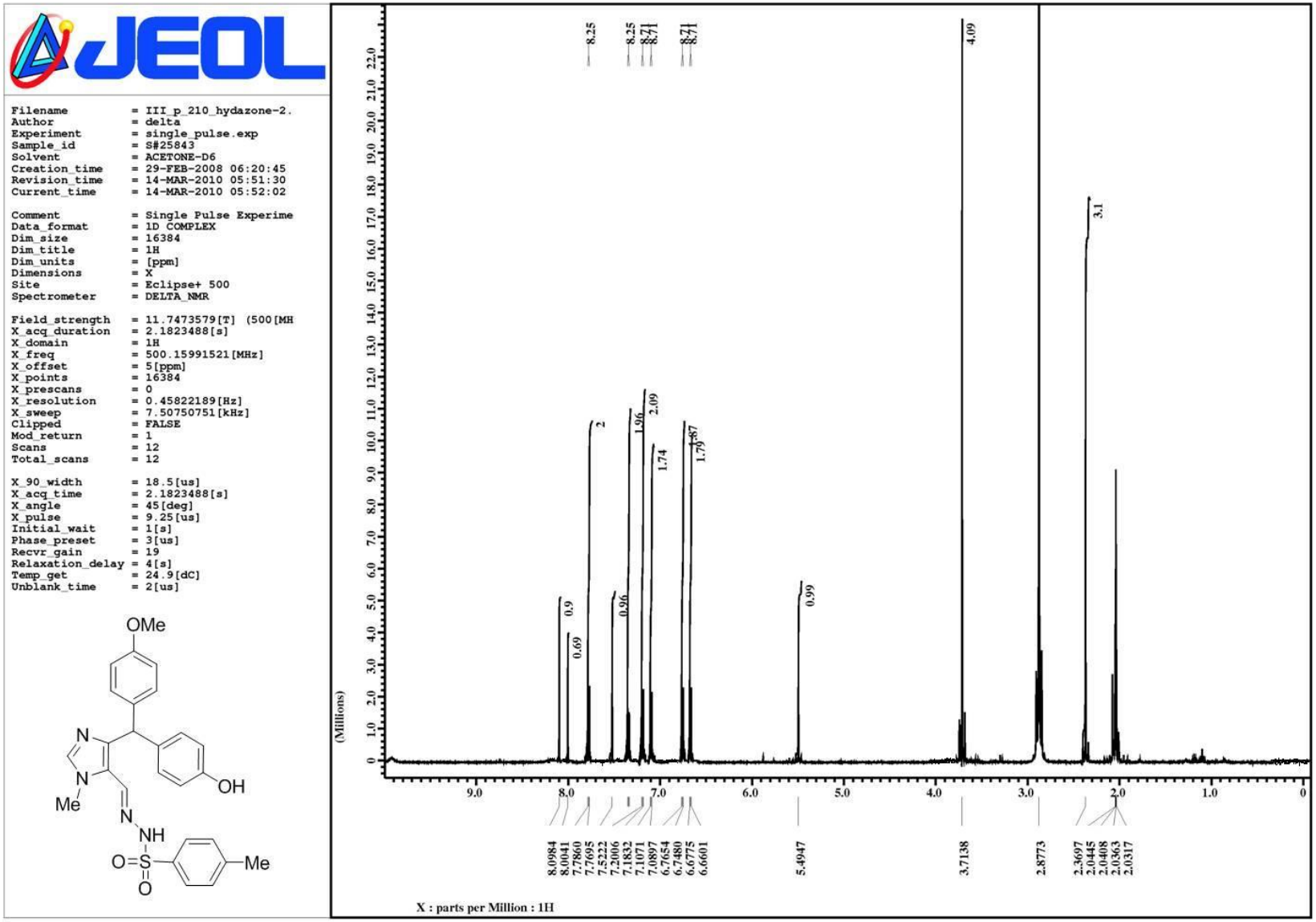

VI_p_042_Mesylate-2.jdf
Filename = VI_p_042_Mesylate-2.
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#748548
Solvent = CHLOROFORM-D
Creation_time = 30-NOV-2009 20:55:48
Revision_time = 30-NOV-2009 20:52:17
Current_time = 30-NOV-2009 20:52:34
Content = single_pulse
Data_format = 1D COMPLEX
Dim_size = 131107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX_300
Spectrometer = DEFTA2_NMR
Field_strength = 7.05860137[T] (300[MH
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 30
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 22.6[dC]
Delay_of_start = 1.99999974[s]
Actual_start_time = 30-NOV-2009 20:54:04
End_time = 30-NOV-2009 20:55:49
Local_time = 30-NOV-2009 20:55:47
abundance
X : parts per Million : 1H
9.86
6.0
5.94
6.01
4.07
1.83
2.05
2.15
1.96
I
7.909
7.255
6.982
6.954
6.750
6.723
4.451
4.149
3.654
3.111
2.547
0.935
0.132

Filename = VI_p_042_Mesylate-2.
Author = delta
Experiment = single_pulse_dec
Sample_id = S#749867
Solvent = CHLOROFORM-D
Creation_time = 30-NOV-2009 21:04:10
Revision_time = 30-NOV-2009 21:00:02
Current_time = 30-NOV-2009 21:00:17
Content = single pulse decoupl
Data_format = 1D COMPLEX
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.05860137[T] (300[MH
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 100
Total_scans = 100
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.1[dC]
Delay_of_start = 1.99999974[s]
Actual_start_time = 30-NOV-2009 20:55:54
End_time = 30-NOV-2009 21:59:54
Local_time = 30-NOV-2009 21:04:09
VI_p_042_Mesylate-2.jdf
abundance
0.4
0.3
0.2
0.1
0
200.0
190.0
180.0
170.0
160.0
150.0
140.0
130.0
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0
-10.0
154.668
138.309
138.080
129.992
129.179
128.414
120.412
77.652
77.227
76.801
52.684
38.496
37.593
31.684
28.319
25.747
18.300
-4.359
X : parts per Million : 13C

APPENDIX 115

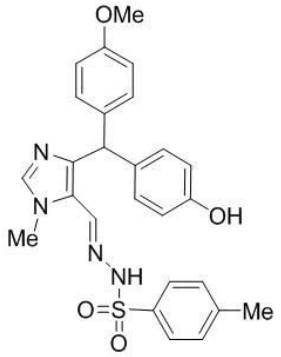
^{1}H and ^{13}C NMR Spectra of

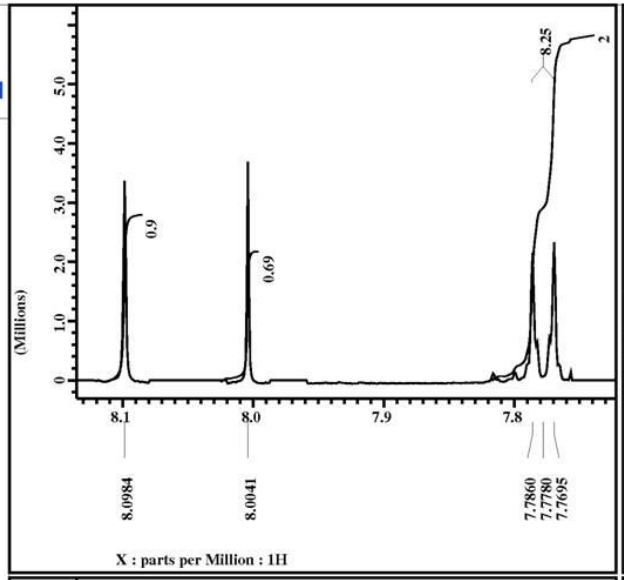
5-[4-Hydroxyphenyl-(4-methoxyphenyl)]methyl-1-methyl-1*H*-imidazol-4-yl-methylene)-4-methyphenylsulfonylhydrazone (**241**)

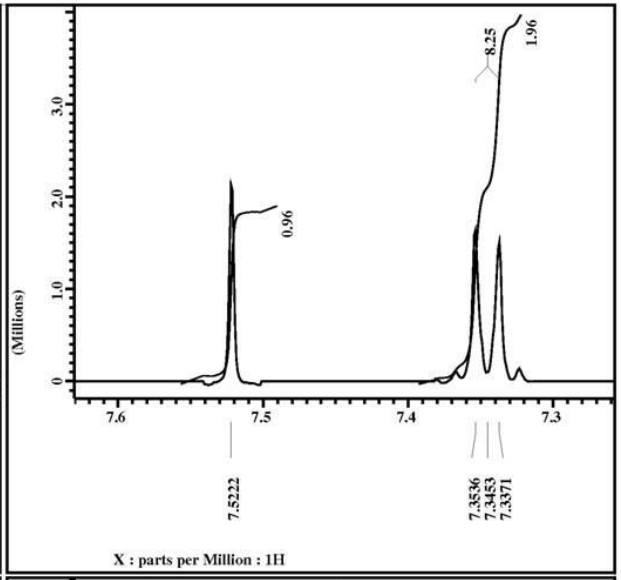

Filename = III_p_210_hydazone-2.
Author = delta
Experiment = single_pulse.exp
Sample_id = S#25843
Solvent = ACETONE-D6
Creation_time = 29-FEB-2008 06:20:45
Revision_time = 14-MAR-2010 05:51:30
Current_time = 14-MAR-2010 05:52:02
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 19
Relaxation_delay = 4[s]
Temp_get = 24.9[dC]
Unblank_time = 2[us]
OMe
N
N
Me
OH
N
NH
O=S=O
Me
(Millions)
8.25
8.25
8.71
8.71
8.71
8.71
4.09
3.1
2
2.09
1.96
1.74
1.87
1.79
0.9
0.69
0.96
0.99
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
8.0984
8.0041
7.7860
7.7695
7.5222
7.2006
7.1832
7.1071
7.0897
6.7654
6.7480
6.6775
6.6601
5.4947
3.7138
2.8773
2.3697
2.0445
2.0408
2.0363
2.0317
X : parts per Million : 1H

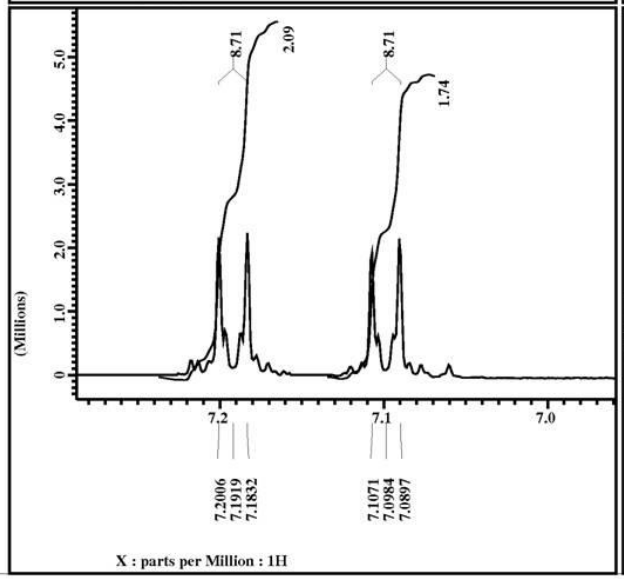
JEOL

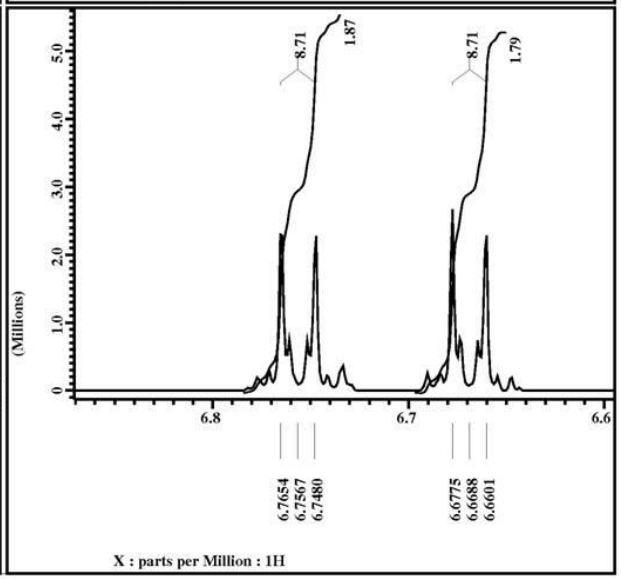
Filename	= III_p_210_hydazone-2.
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#25843
Solvent	= ACETONE-D6
Creation_time	= 29-FEB-2008 06:20:45
Revision_time	= 14-MAR-2010 05:51:30
Current_time	= 14-MAR-2010 05:55:17
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 19
Relaxation_delay	= 4[s]
Temp_get	= 24.9[dC]
Unblank_time	= 2[us]











JEOL
Filename = III_p_210_hydazone-2.
Author = delta
Experiment = single_pulse_dec
Sample_id = S#26388
Solvent = ACETONE-D6
Creation_time = 29-FEB-2008 14:51:04
Revision_time = 29-FEB-2008 10:32:08
Current_time = 21-MAR-2010 14:30:10
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 6000
Total_scans = 6000
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 29
Relaxation_delay = 2[s]
Temp_get = 26.5[dC]
Unblank_time = 2[us]
OMe
OH
N
NH
N
Me
O=S=O
Me
(Thousands)
X : parts per Million : 13C
148.5806
143.8420
140.7554
138.5615
136.5509
136.3410
134.9484
129.9694
129.5803
127.8939
(Thousands)
X : parts per Million : 13C
158.1113
155.7115
148.5806
143.8420
140.7554
138.5615
136.5509
136.3410
134.9484
129.9694
129.5803
127.8939
122.1861
114.7386
113.2621
78.3900
54.6014
46.8143
33.6514
20.6259

APPENDIX 116

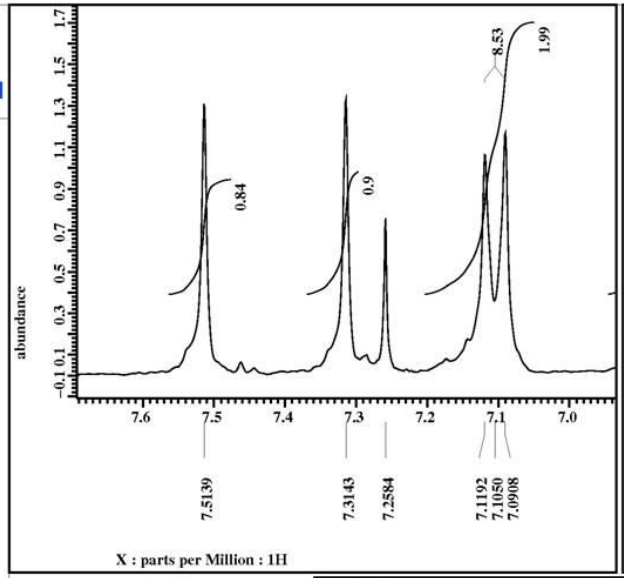
^{1}H and ^{13}C NMR Spectra of

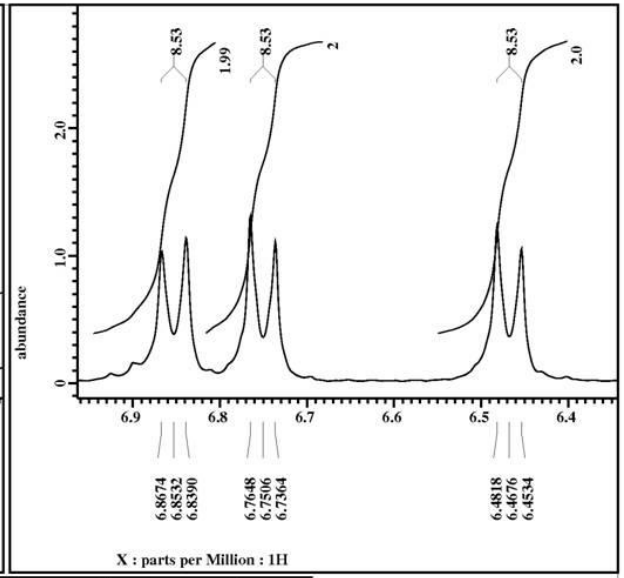
5-(4-Hydroxyphenyl-4-methoxyphenyl)methyl-1-methyl-1*H*-imidazol-4-yl-methylenehydrazone (**242**)

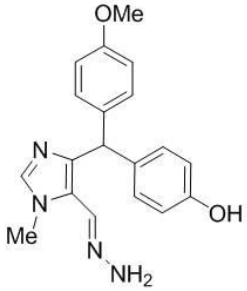
JEOL
Filename = III_p_265_hydrazone-4
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#730060
Solvent = CHLOROFORM-D
Creation_time = 3-APR-2008 19:30:34
Revision_time = 14-MAR-2010 06:24:38
Current_time = 14-MAR-2010 06:24:55
Comment = single pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 30
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 21.6[dC]
OMe
OH
NH2
N
N
Me
8.53
8.53
5.94
2.0
1.99
1.99
0.9
0.84
0.87
5.4469
3.6776
3.6464
7.5139
7.3143
7.1192
7.0908
6.8674
6.8390
6.7648
6.7364
6.4818
6.4534
abundance
X : parts per Million : 1H

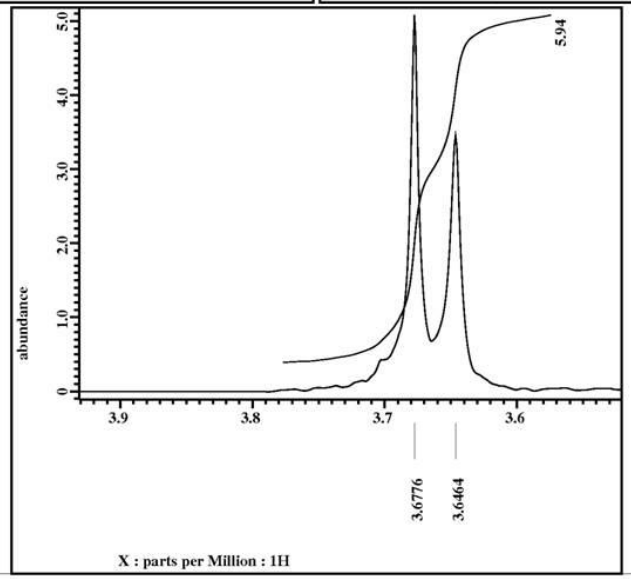
JEOL

Filename	= III_p_265_hydrazone-4
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#730060
Solvent	= CHLOROFORM-D
Creation_time	= 3-APR-2008 19:30:34
Revision_time	= 14-MAR-2010 06:24:38
Current_time	= 14-MAR-2010 06:25:42
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 30
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 21.6[dC]

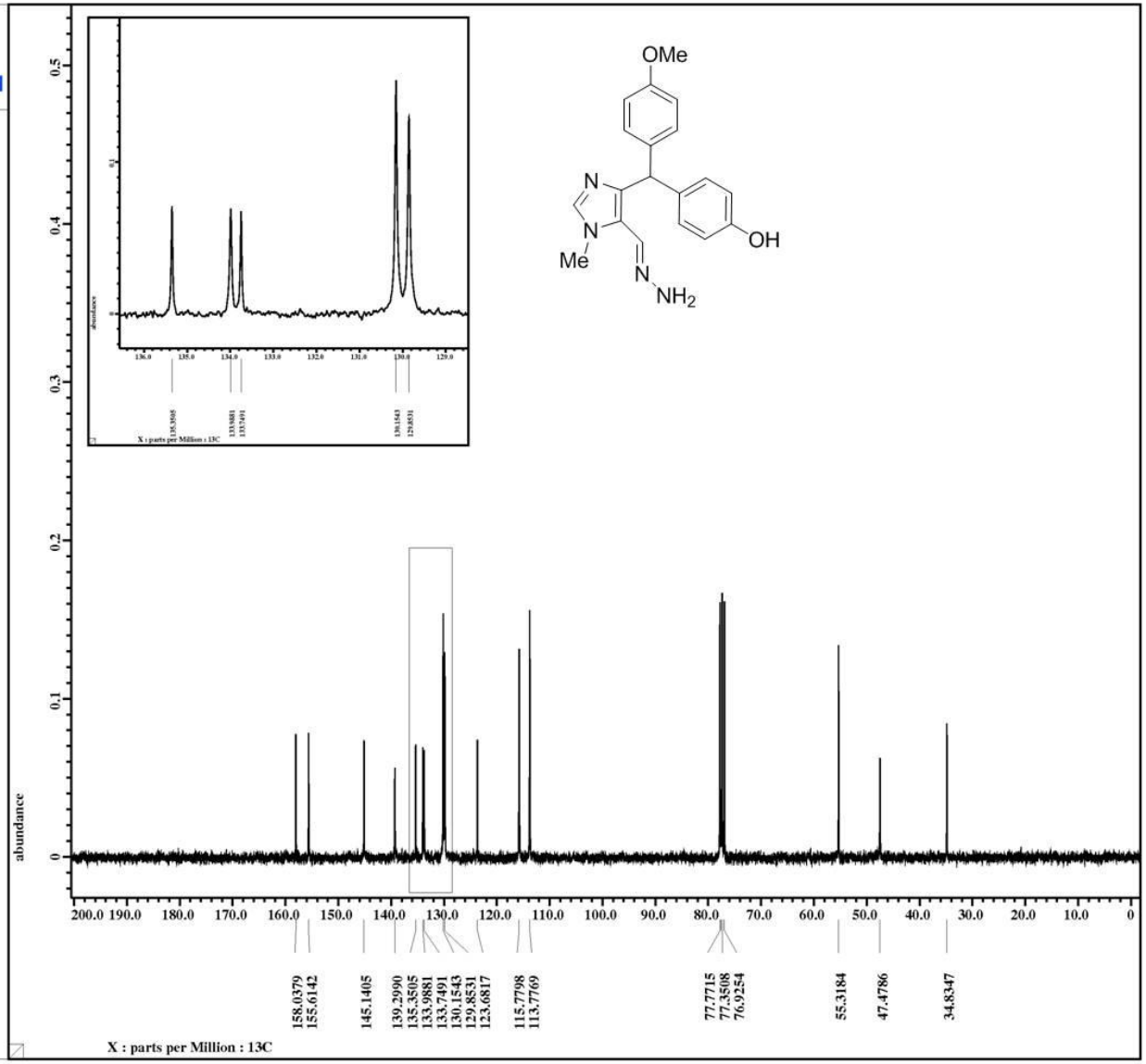








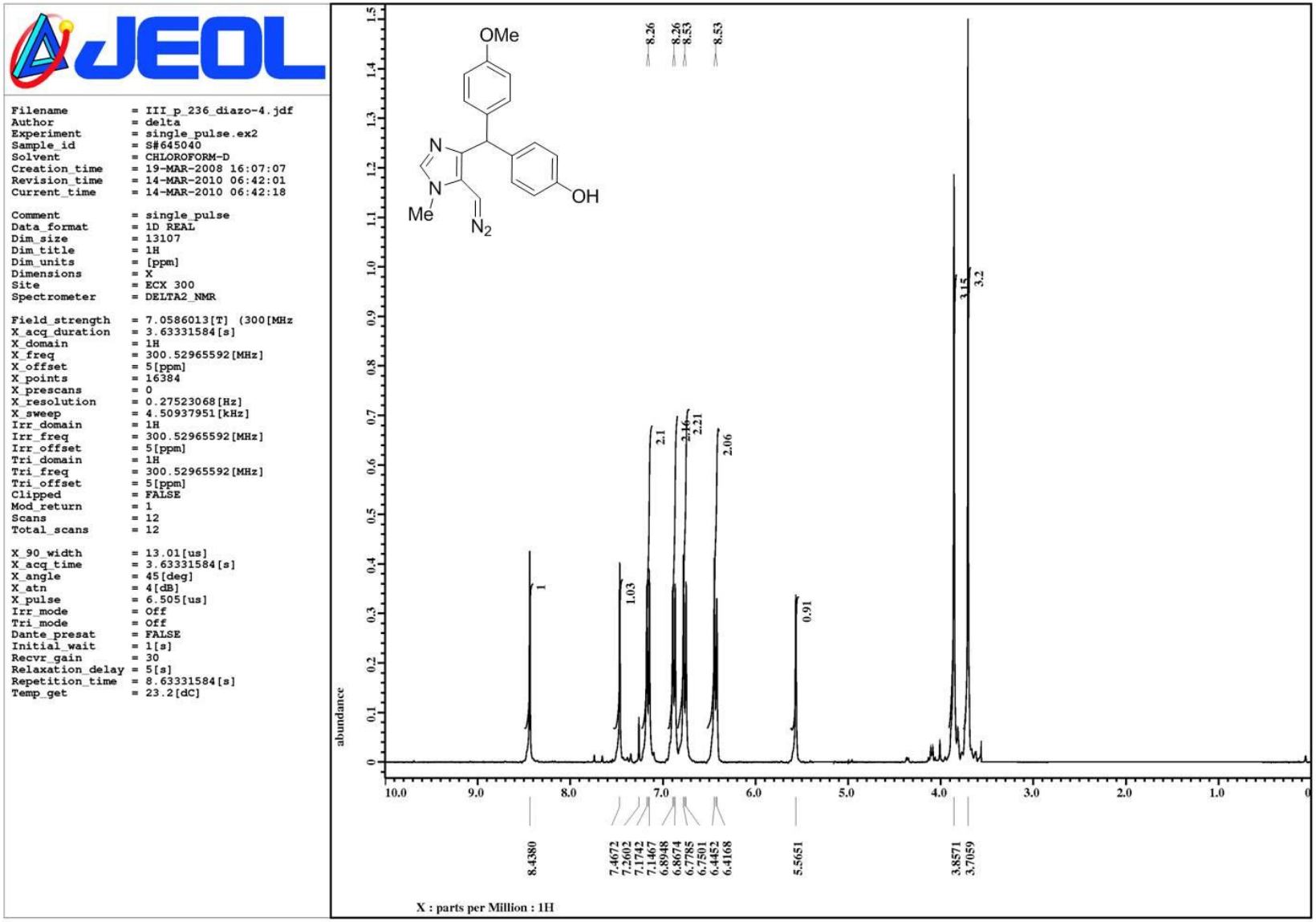
Filename	= III_p_265_hydrazone-2
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= panduka
Solvent	= CHLOROFORM-D
Creation_time	= 3-APR-2008 19:43:16
Revision_time	= 3-APR-2008 20:31:55
Current_time	= 21-MAR-2010 14:37:19
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 52428
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 2.76824064[s]
X_domain	= 13C
X_freq	= 75.56823426[MHz]
X_offset	= 100[ppm]
X_points	= 65536
X_prescans	= 4
X_resolution	= 0.36124027[Hz]
X_sweep	= 23.67424242[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 149
Total_scans	= 149
X_90_width	= 9.75[us]
X_acq_time	= 2.76824064[s]
X_angle	= 30[deg]
X_atn	= 8[dB]
X_pulse	= 3.25[us]
Irr_atn_dec	= 25[dB]
Irr_atn_noe	= 25[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 4.76824064[s]
Temp_get	= 22[dC]



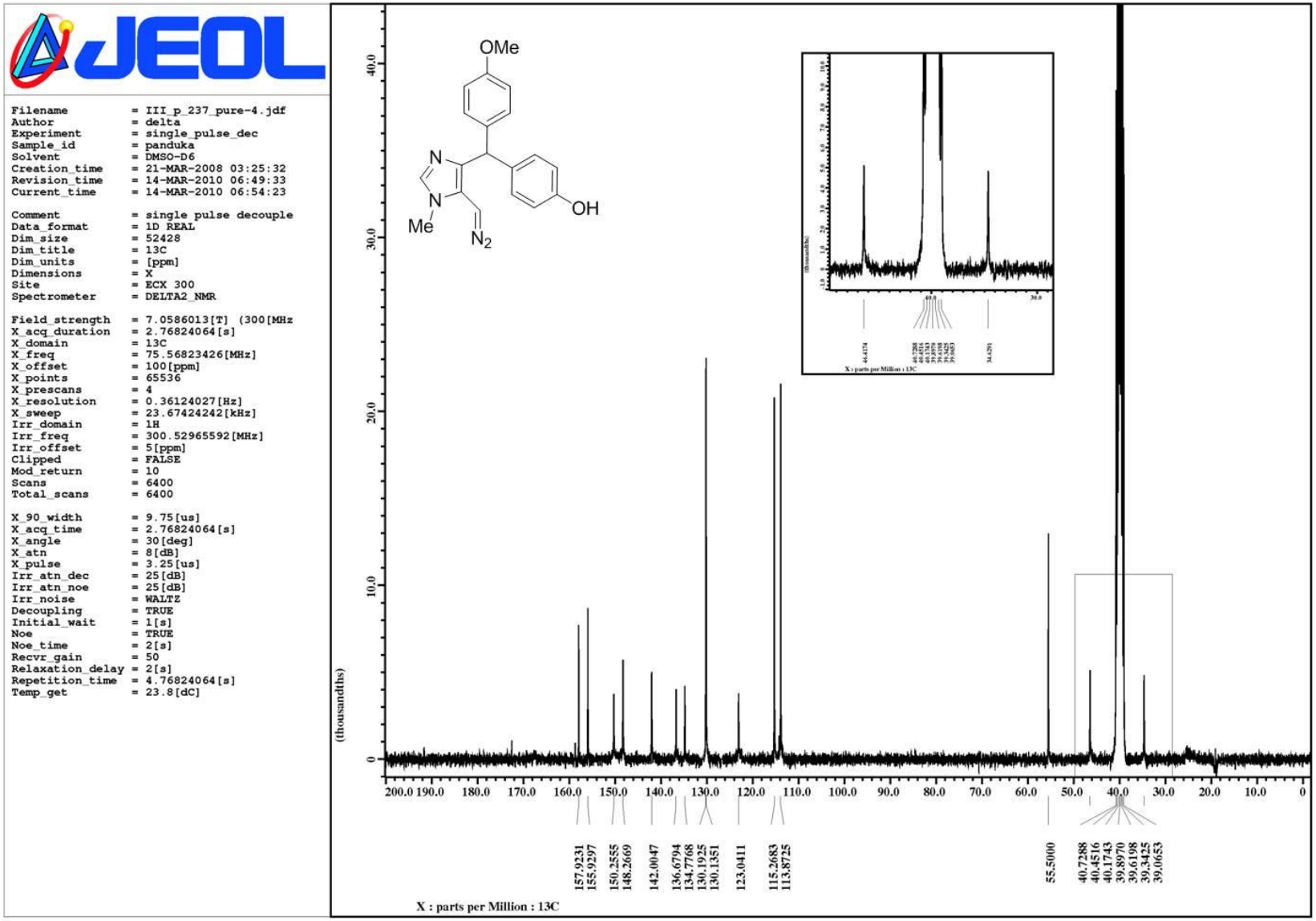
APPENDIX 117

^{1}H and ^{13}C NMR Spectra of

4-(4-Hydroxyphenyl-4-methoxyphenyl)methyl-5-diazomethyl-1-methyl-1*H*-imidazole (**243**)


JEOL
Filename = III_p_236_diazo-4.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#645040
Solvent = CHLOROFORM-D
Creation_time = 19-MAR-2008 16:07:07
Revision_time = 14-MAR-2010 06:42:01
Current_time = 14-MAR-2010 06:42:18
Comment = single_pulse
Data_format = 1D REAL
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 30
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.2[dC]
OMe
OH
N
N
Me
N2
8.26
8.26
8.53
8.53
3.15
3.2
2.1
2.16
2.21
2.06
1
1.03
0.91
abundance
8.4380
7.4672
7.2602
7.1742
7.1467
6.8948
6.8674
6.7785
6.7501
6.4452
6.4168
5.5651
3.8571
3.7059
X : parts per Million : 1H

JEOL
Filename = III_p_236_diazo-4.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#645040
Solvent = CHLOROFORM-D
Creation_time = 19-MAR-2008 16:07:07
Revision_time = 14-MAR-2010 06:42:01
Current_time = 14-MAR-2010 06:42:34
Comment = single_pulse
Data_format = 1D REAL
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 30
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 23.2[dC]
OMe
OH
N2
Me
abundance
X : parts per Million : 1H
8.4380
7.4672
7.2602
7.1742
7.1604
7.1467
6.8948
6.8811
6.8674
6.7785
6.7643
6.7501
6.4452
6.4310
6.4168
1.03
2.1
2.16
2.21
2.06
8.26
8.26
8.53
8.53


JEOL
Filename = III_p_237_pure-4.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = panduka
Solvent = DMSO-D6
Creation_time = 21-MAR-2008 03:25:32
Revision_time = 14-MAR-2010 06:49:33
Current_time = 14-MAR-2010 06:54:23
Comment = single pulse decouple
Data_format = 1D REAL
Dim_size = 52428
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 2.76824064[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.36124027[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 9.75[us]
X_acq_time = 2.76824064[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 4.76824064[s]
Temp_get = 23.8[dC]
OMe
N
N
Me
N2
OH
(thousandths)
40.0
30.0
20.0
10.0
0
200.0 190.0 180.0 170.0 160.0 150.0 140.0 130.0 120.0 110.0 100.0 90.0 80.0 70.0 60.0 50.0 40.0 30.0 20.0 10.0 0
157.9231
155.9297
150.2555
148.2669
142.0047
136.6794
134.7768
130.1925
130.1351
123.0411
115.2683
113.8725
55.5000
40.7288
40.4516
40.1743
39.8970
39.6198
39.3425
39.0653
X : parts per Million : 13C

APPENDIX 118

^{1}H and ^{13}C NMR Spectra of

2-iodo-3-oxo-1-phenyl-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (**256**)

JEOL
Filename = IV_p_086_sm-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#572462
Solvent = CHLOROFORM-D
Creation_time = 18-JUN-2008 22:32:20
Revision_time = 22-MAR-2010 18:19:01
Current_time = 22-MAR-2010 18:19:13
Comment = Single Pulse Experime
Data_format = 1D REAL
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 20
Relaxation_delay = 4[s]
Temp_get = 25.4[dC]
Unblank_time = 2[us]
3.31
2.21
2.11
1.89
3.18
0.71
0.31
7.3948
7.3838
7.3692
7.2537
5.8914
5.8859
5.8300
5.8273
5.8099
5.2483
2.8241
2.7600
2.0454
1.9419
(Millions)
X : parts per Million : 1H
AcO
I
O

JEOL
Filename = IV_p_086_sm-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#572462
Solvent = CHLOROFORM-D
Creation_time = 18-JUN-2008 22:32:20
Revision_time = 22-MAR-2010 18:19:01
Current_time = 22-MAR-2010 18:19:49
Comment = Single Pulse Experime
Data_format = 1D REAL
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 20
Relaxation_delay = 4[s]
Temp_get = 25.4[dC]
Unblank_time = 2[us]
AcO
Millions
X : parts per Million : 1H
3.18
1.89
7.4241
7.4223
7.4177
7.4067
7.3948
7.3838
7.3756
7.3692
7.3618
7.3573
7.2537
7.2391
7.2363
7.2253
7.2217
0.71
0.31
5.5543
5.5524
5.2483
2.11
2.0
5.9537
5.9464
5.9336
5.9262
5.9116
5.9061
5.8914
5.8859
5.8722
5.8520
5.8300
5.8273
5.8099
3.31
2.21
2.8241
2.7600
2.0454
1.9419

JEOL
Filename = IV_p_086_sm(054_produ
Author = delta
Experiment = single_pulse_dec
Sample_id = S#791705
Solvent = CHLOROFORM-D
Creation_time = 20-JUN-2008 13:42:30
Revision_time = 22-MAR-2010 18:10:55
Current_time = 22-MAR-2010 18:11:19
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 6400
Total_scans = 6400
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 28.5[dC]
Unblank_time = 2[us]
(Thousands)
X : parts per Million : 13C
201.1520
201.0299
180.1448
178.9354
170.4997
170.4691
132.4913
129.6947
128.3678
127.9473
127.2720
126.3029
105.3758
104.8035
77.3598
77.1080
76.8524
63.6513
63.3728
51.7780
51.4576
47.5659
46.7227
21.1524
21.0150
AcO
I

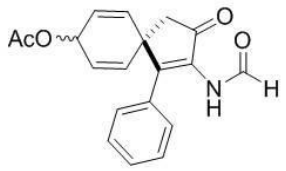
JEOL
Filename = IV_p_086_sm(054_produ
Author = delta
Experiment = single_pulse_dec
Sample_id = S#791705
Solvent = CHLOROFORM-D
Creation_time = 20-JUN-2008 13:42:30
Revision_time = 22-MAR-2010 18:10:55
Current_time = 22-MAR-2010 18:13:15
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.74735791[T] (500[MH
X_acq_duration = 2.08404481[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 6400
Total_scans = 6400
X_90_width = 14.2[us]
X_acq_time = 2.08404481[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 28.5[dC]
Unblank_time = 2[us]
AcO
I
(Thousands)
X : parts per Million : 13C
201.1520
201.0299
180.1448
178.9354
170.4997
170.4691
135.1964
134.9713
133.7809
132.4913
129.8893
129.6947
128.2678
128.0007
127.9473
127.2720
126.3029
125.3834
104.8035
105.3758
63.6513
63.3728
51.7780
51.4576
47.5659
46.7227
21.1524
21.0150

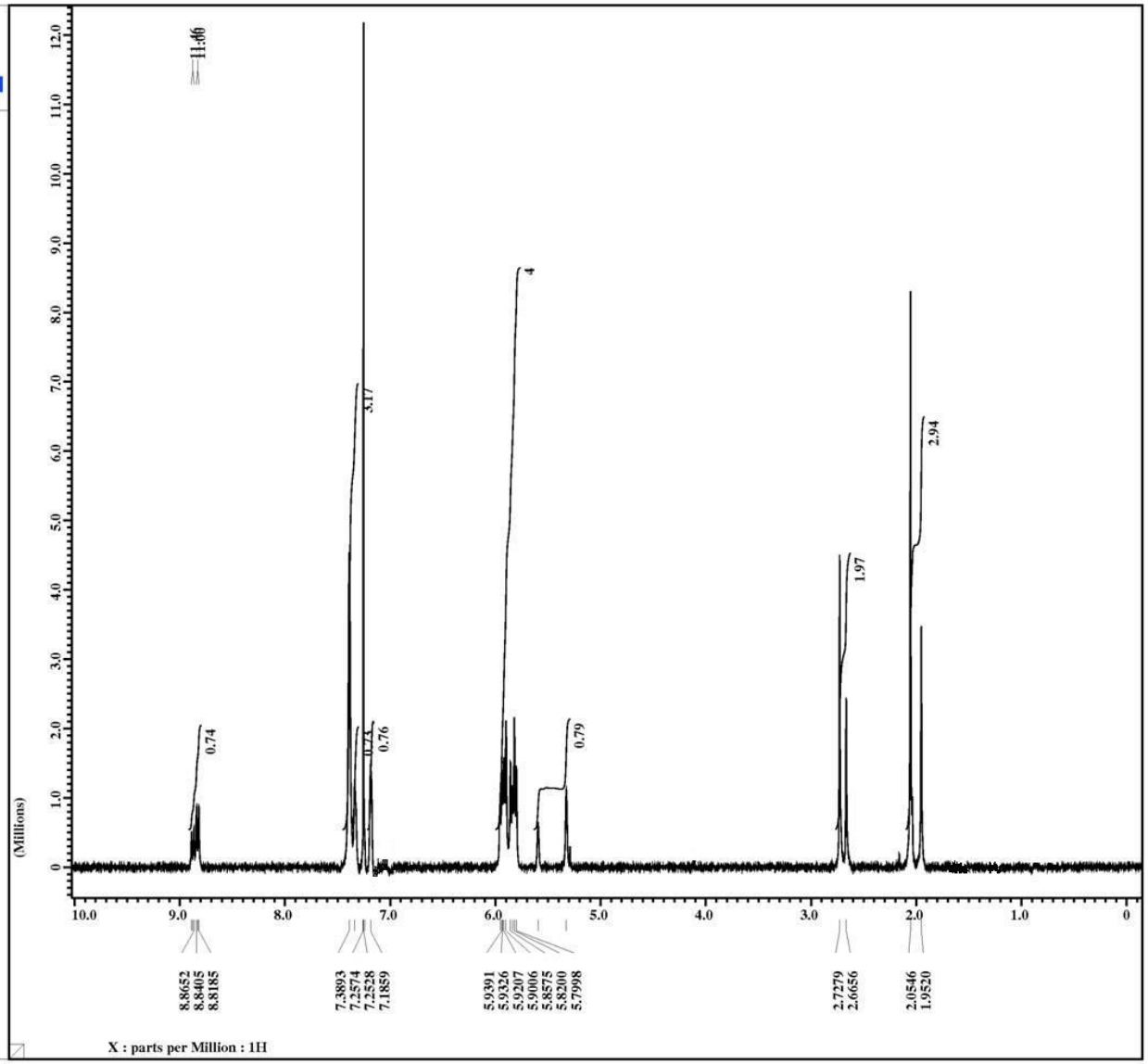
APPENDIX 119

^{1}H and ^{13}C NMR Spectra of

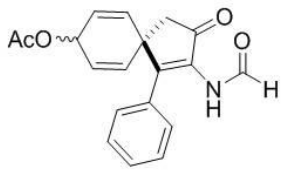
2-formylamino-3-oxo-1-phenyl-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (**259**)

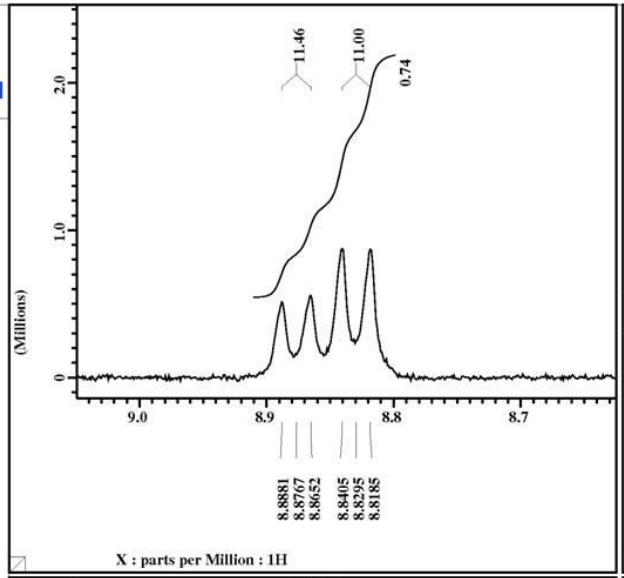
```
Filename          = IV_p_089_ii-4.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#424761
Solvent           = CHLOROFORM-D
Creation_time     = 25-JUN-2008 18:29:59
Revision_time     = 21-MAR-2010 17:26:49
Current_time      = 21-MAR-2010 17:26:58

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 25
Relaxation_delay  = 4[s]
Temp_get          = 24.8[dC]
Unblank_time      = 2[us]
```

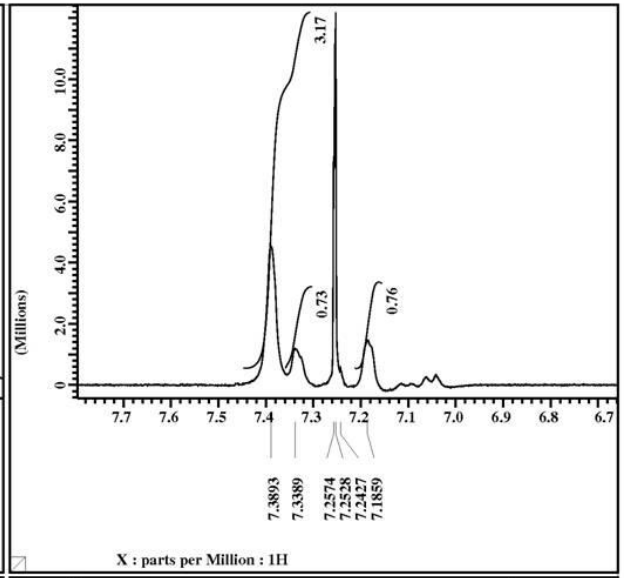


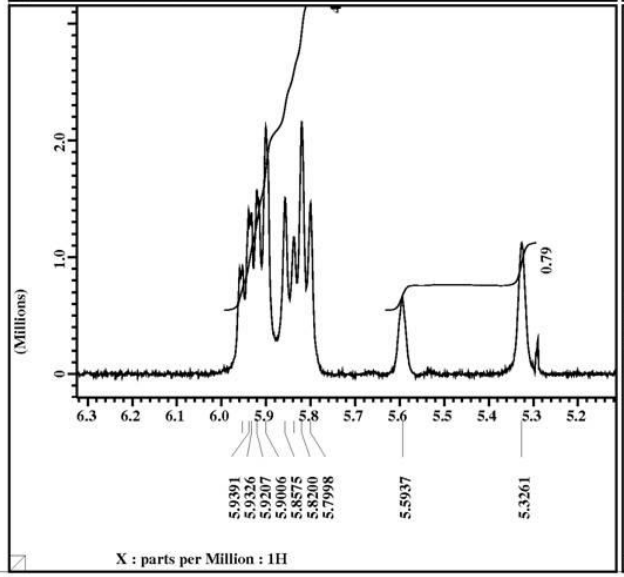


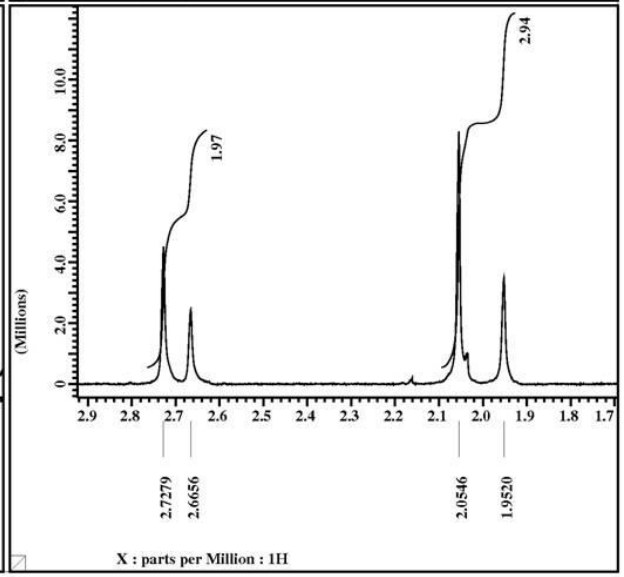
```
Filename          = IV_p_089_ii-4.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#424761
Solvent           = CHLOROFORM-D
Creation_time     = 25-JUN-2008 18:29:59
Revision_time     = 21-MAR-2010 17:27:01
Current_time      = 21-MAR-2010 17:27:38

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 25
Relaxation_delay  = 4[s]
Temp_get          = 24.8[dC]
Unblank_time      = 2[us]
```



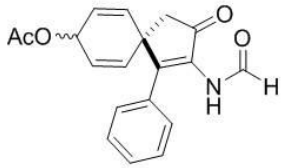


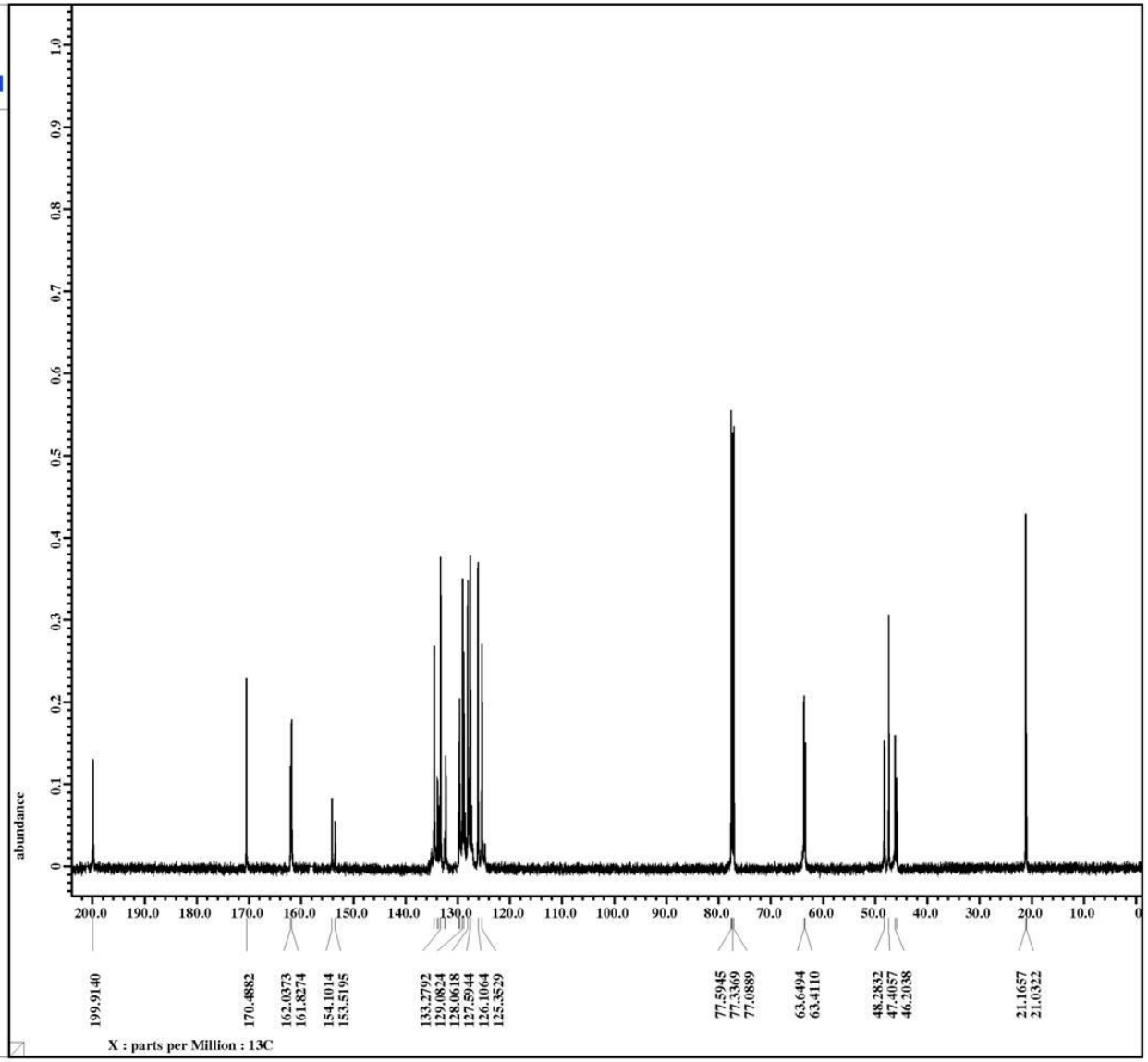




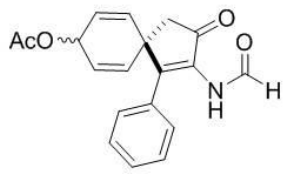


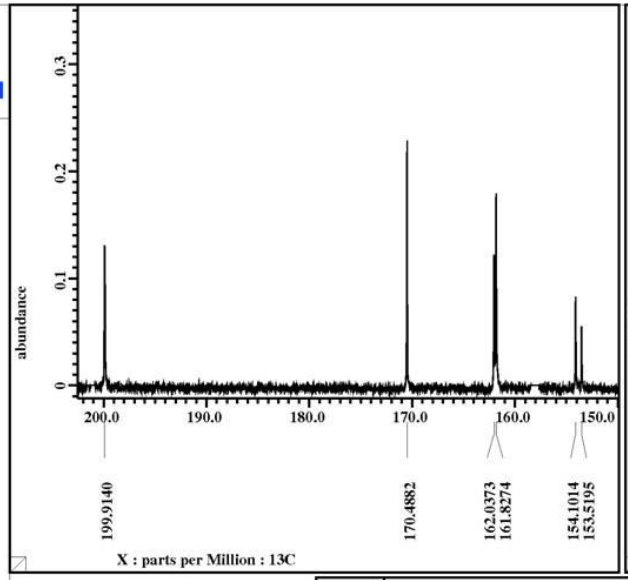
Filename	= VI_p_069_product-2.jd
Author	= delta
Experiment	= single_pulse_dec
Sample_id	= S#767231
Solvent	= CHLOROFORM-D
Creation_time	= 15-APR-2010 12:03:54
Revision_time	= 15-APR-2010 12:17:31
Current_time	= 15-APR-2010 12:18:25
Comment	= single pulse decouple
Data_format	= 1D COMPLEX
Dim_size	= 26214
Dim_title	= 13C
Dim_units	= [ppm]
Dimensions	= X
Site	= ECA 500
Spectrometer	= JNM-ECA500
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 0.83361792[s]
X_domain	= 13C
X_freq	= 125.76529768[MHz]
X_offset	= 100[ppm]
X_points	= 32768
X_prescans	= 4
X_resolution	= 1.19959034[Hz]
X_sweep	= 39.3081761[kHz]
Irr_domain	= 1H
Irr_freq	= 500.15991521[MHz]
Irr_offset	= 5.0[ppm]
Clipped	= FALSE
Mod_return	= 10
Scans	= 900
Total_scans	= 900
X_90_width	= 13.3[us]
X_acq_time	= 0.83361792[s]
X_angle	= 30[deg]
X_atn	= 9[dB]
X_pulse	= 4.43333333[us]
Irr_atn_dec	= 16.5[dB]
Irr_atn_noe	= 16.5[dB]
Irr_noise	= WALTZ
Decoupling	= TRUE
Initial_wait	= 1[s]
Noe	= TRUE
Noe_time	= 2[s]
Recvr_gain	= 50
Relaxation_delay	= 2[s]
Repetition_time	= 2.83361792[s]
Temp_get	= 26.7[dC]

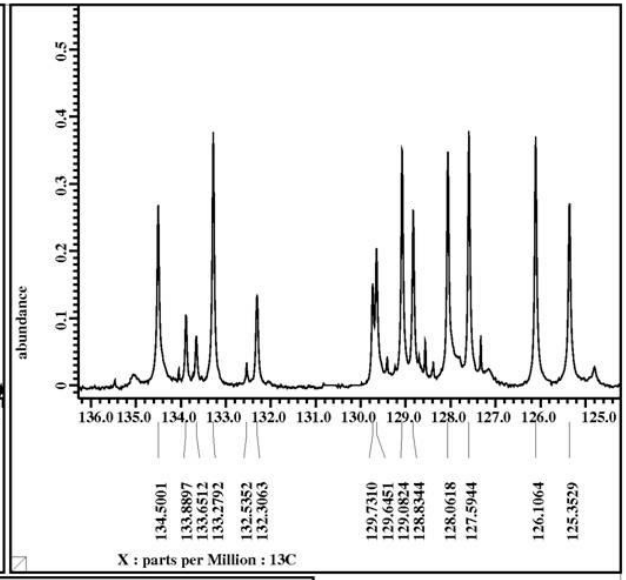


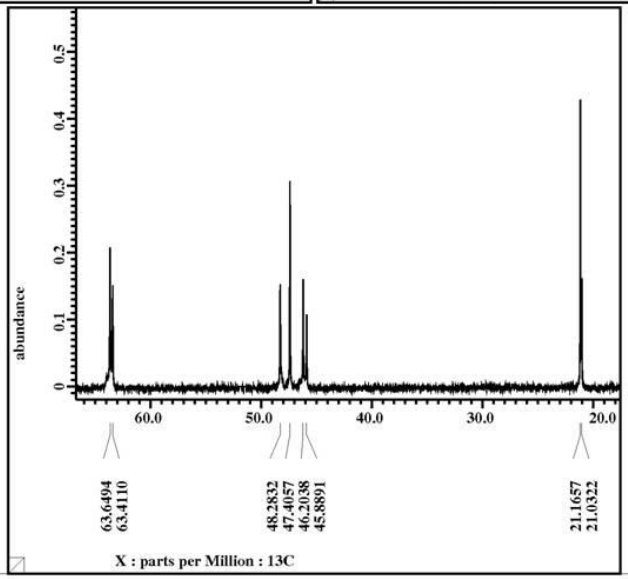


```
Filename          = VI_p_069_product-2.jd
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#767231
Solvent           = CHLOROFORM-D
Creation_time     = 15-APR-2010 12:03:54
Revision_time     = 15-APR-2010 12:17:31
Current_time      = 15-APR-2010 12:19:51

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 26214
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECA 500
Spectrometer      = JNM-ECA500

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 0.83361792[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 32768
X_prescans        = 4
X_resolution      = 1.19959034[Hz]
X_sweep           = 39.3081761[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5.0[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 900
Total_scans       = 900

X_90_width        = 13.3[us]
X_acq_time        = 0.83361792[s]
X_angle           = 30[deg]
X_atn             = 9[dB]
X_pulse           = 4.43333333[us]
Irr_atn_dec       = 16.5[dB]
Irr_atn_noe       = 16.5[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 2.83361792[s]
Temp_get          = 26.7[dC]
```









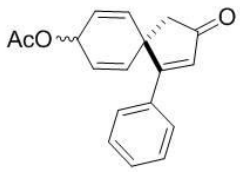
APPENDIX 120

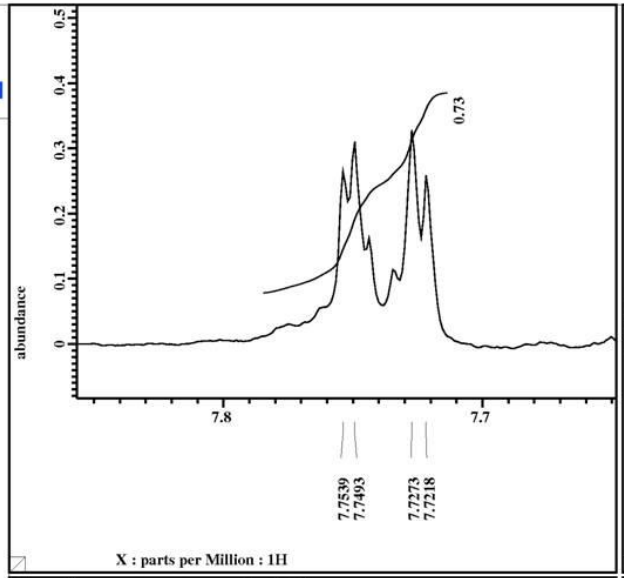
^{1}H and ^{13}C NMR Spectra of

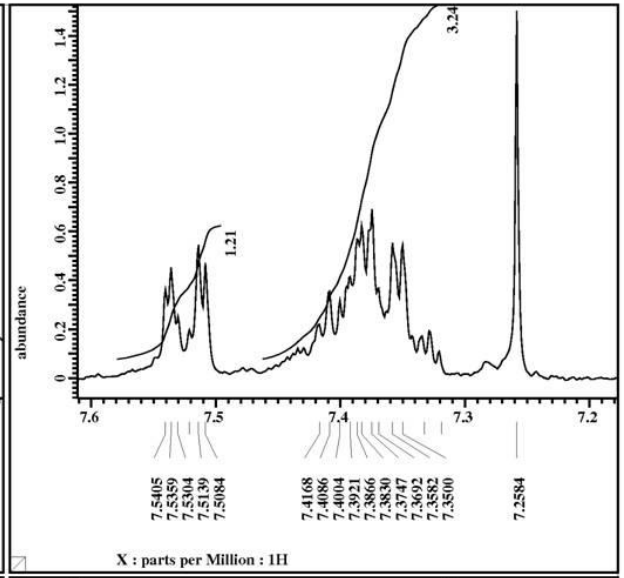
3-Oxo-1-phenyl-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (**260**)

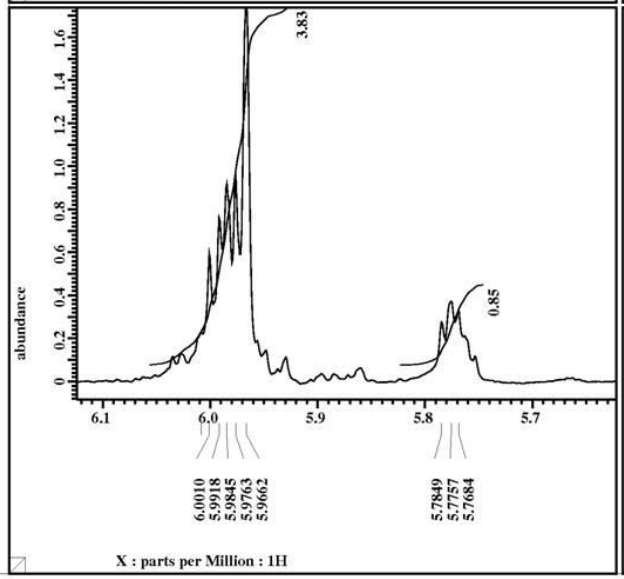
JEOL
Filename = IV_p_089_i-3.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#708028
Solvent = CHLOROFORM-D
Creation_time = 24-JUN-2008 19:59:08
Revision_time = 21-MAR-2010 17:43:39
Current_time = 21-MAR-2010 17:43:47
Comment = single_pulse
Data_format = 1D REAL
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 3.63331584[s]
X_domain = 1H
X_freq = 300.52965592[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.27523068[Hz]
X_sweep = 4.50937951[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Tri_domain = 1H
Tri_freq = 300.52965592[MHz]
Tri_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 10
Total_scans = 10
X_90_width = 13.01[us]
X_acq_time = 3.63331584[s]
X_angle = 45[deg]
X_atn = 4[dB]
X_pulse = 6.505[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 46
Relaxation_delay = 5[s]
Repetition_time = 8.63331584[s]
Temp_get = 21.9[dC]
AcO
abundance
X : parts per Million : 1H
2.87
1.88
3.83
0.85
1.0
3.24
1.21
0.73
7.5139
7.3866
7.3830
7.3747
7.3582
7.2584
6.5798
6.5056
5.9918
5.9845
5.9763
5.9662
2.6720
2.5996
2.1353
2.1133

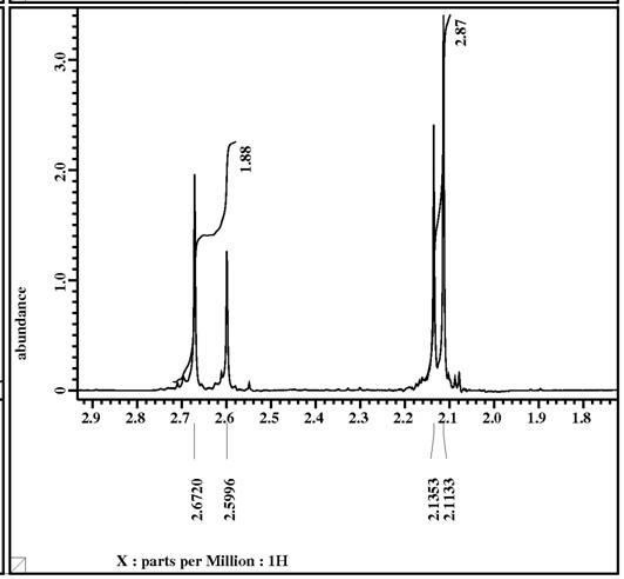
Filename	= IV_p_089_i-3.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#708028
Solvent	= CHLOROFORM-D
Creation_time	= 24-JUN-2008 19:59:08
Revision_time	= 21-MAR-2010 17:45:43
Current_time	= 21-MAR-2010 17:48:41
Comment	= single_pulse
Data_format	= 1D REAL
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 10
Total_scans	= 10
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 46
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 21.9[dC]

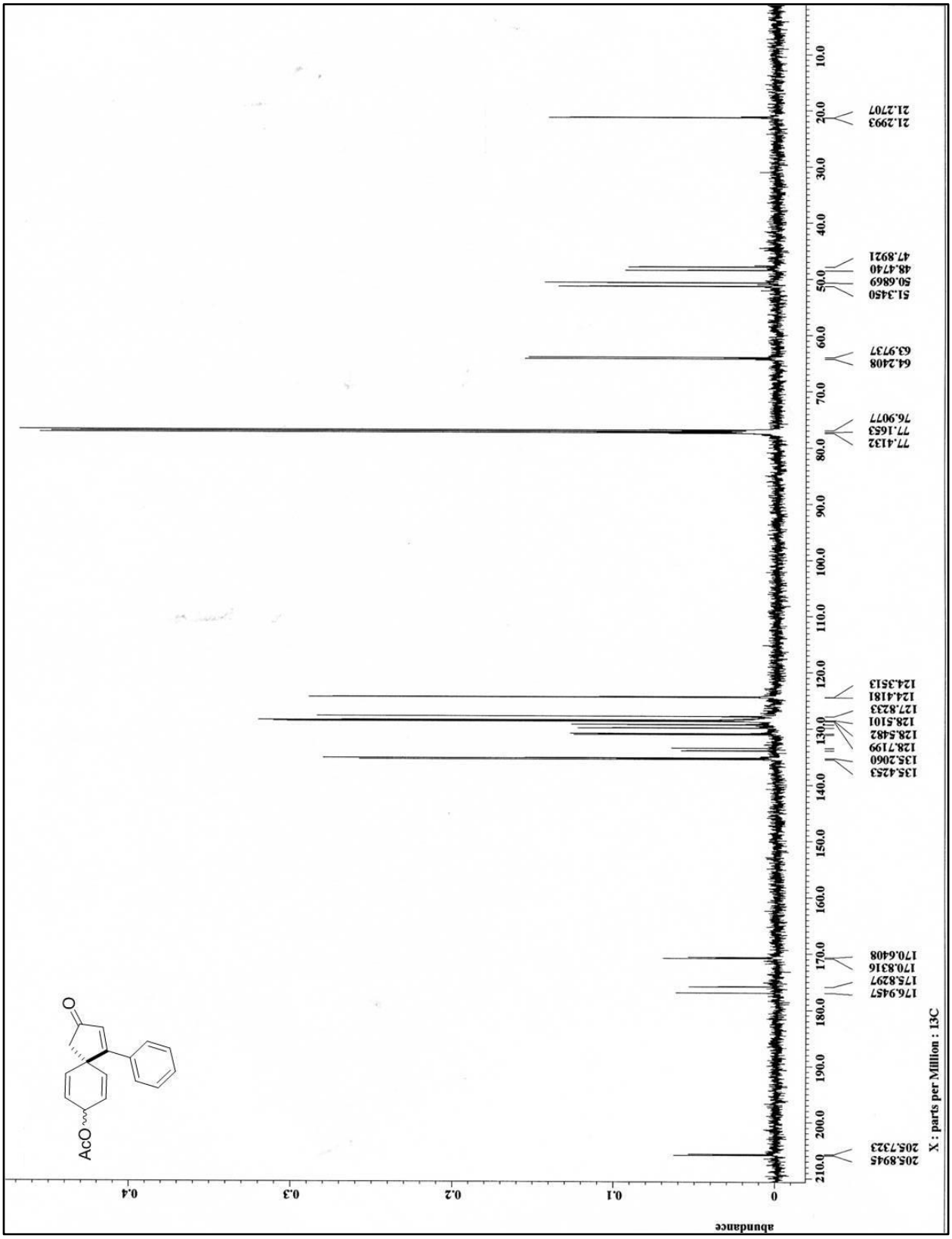











AcO
O
205.8945
205.7323
176.9457
175.8297
170.8316
170.6408
135.4253
135.2060
128.7199
128.5482
128.5101
127.8233
124.4181
124.3513
77.4132
77.1653
76.9077
64.2408
63.9737
51.3450
50.6869
48.4740
47.8921
21.2993
21.2707
X : parts per Million : 13C
abundance

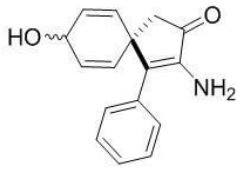
Filename = IV_p_235_i-2.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = 81349355
Solvent = CHLOROFORM-D
Creation_time = 12-MAR-2010 00:56:41
Revision_time = 11-MAR-2010 10:01:52
Current_time = 11-MAR-2010 10:02:27
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_Size = 26214
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = RCA 500
Spectrometer = JNM-ECA500
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 0.83361792[s]
X_domain = 13C
X_freq = 125.765297768[MHz]
X_offset = 100[ppm]
X_points = 32768
X_prescans = 4
X_resolution = 1.19959034[Hz]
X_sweep = 39.3081761[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5.0[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 350
Total_scans = 350
X_90_width = 12.82[us]
X_acq_time = 0.83361792[s]
X_angle = 30[deg]
X_atn = 9[dB]
X_pulse = 4.273333333[us]
Irr_atn_dec = 18[dB]
Irr_atn_noe = 18[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 2.83361792[s]
Temp_get = 22.5[dC]
abundance
X : parts per Million : 13C
IV_p_235_i-2.jdf
205.8945
205.7323
176.9457
175.8297
170.8316
170.6408
124.4181
124.3513
64.2408
63.9737
21.2993
21.2707
AcO

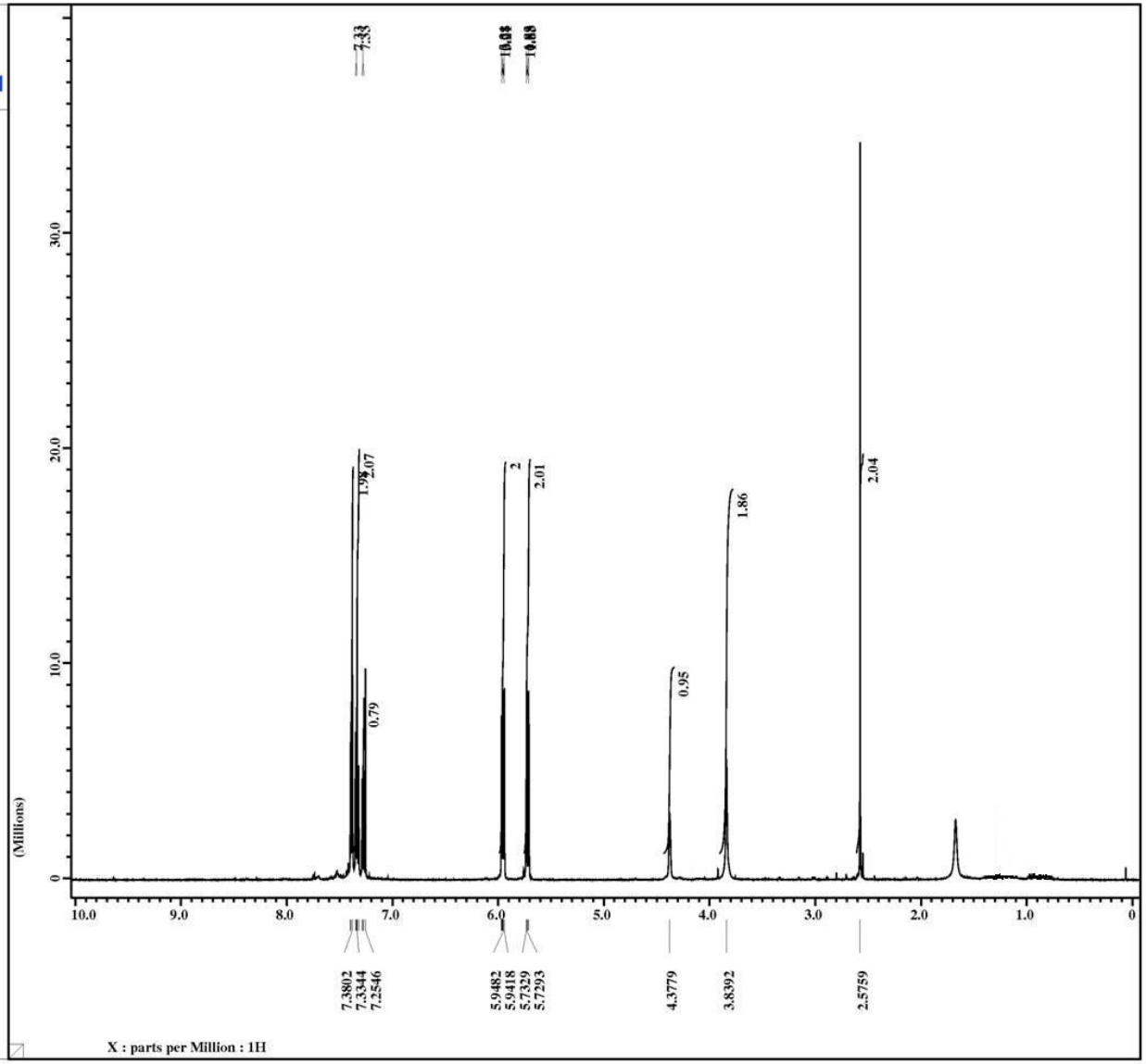
APPENDIX 121

^{1}H and ^{13}C NMR Spectra of

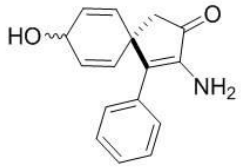
3-Amino-8-hydroxy-4-phenyl-spiro[4.5]deca-3,6,9-trien-2-one (**261**)

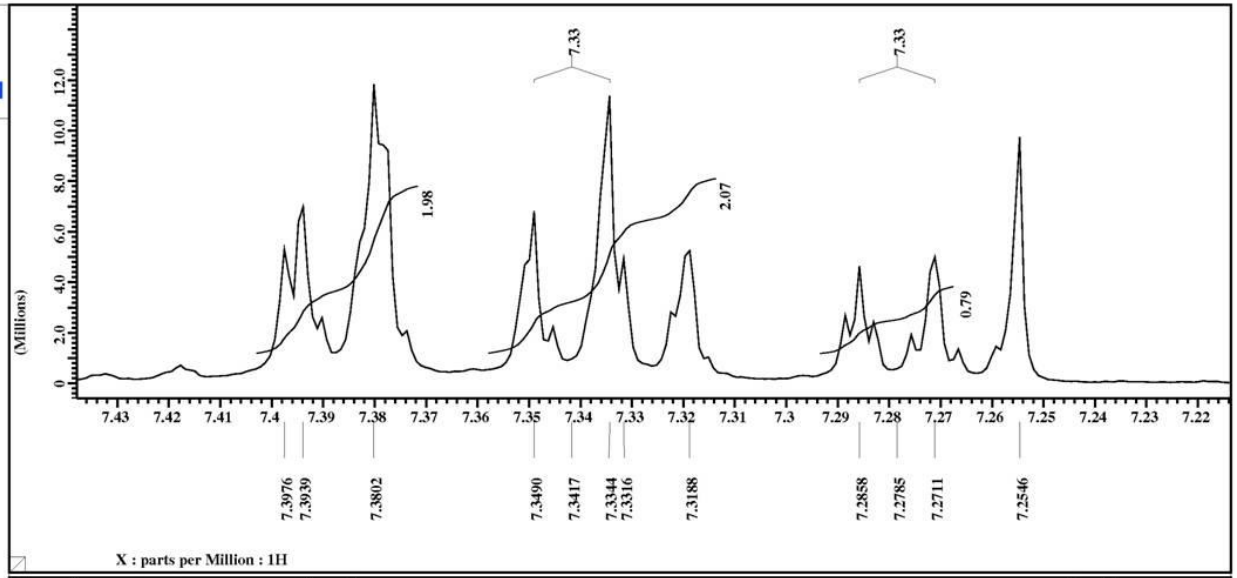
Filename	= IV_p_114_ii-2.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#778738
Solvent	= CHLOROFORM-D
Creation_time	= 13-JUL-2008 04:30:21
Revision_time	= 21-MAR-2010 18:03:49
Current_time	= 21-MAR-2010 18:04:01
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 20
Relaxation_delay	= 4[s]
Temp_get	= 24.7[dC]
Unblank_time	= 2[us]

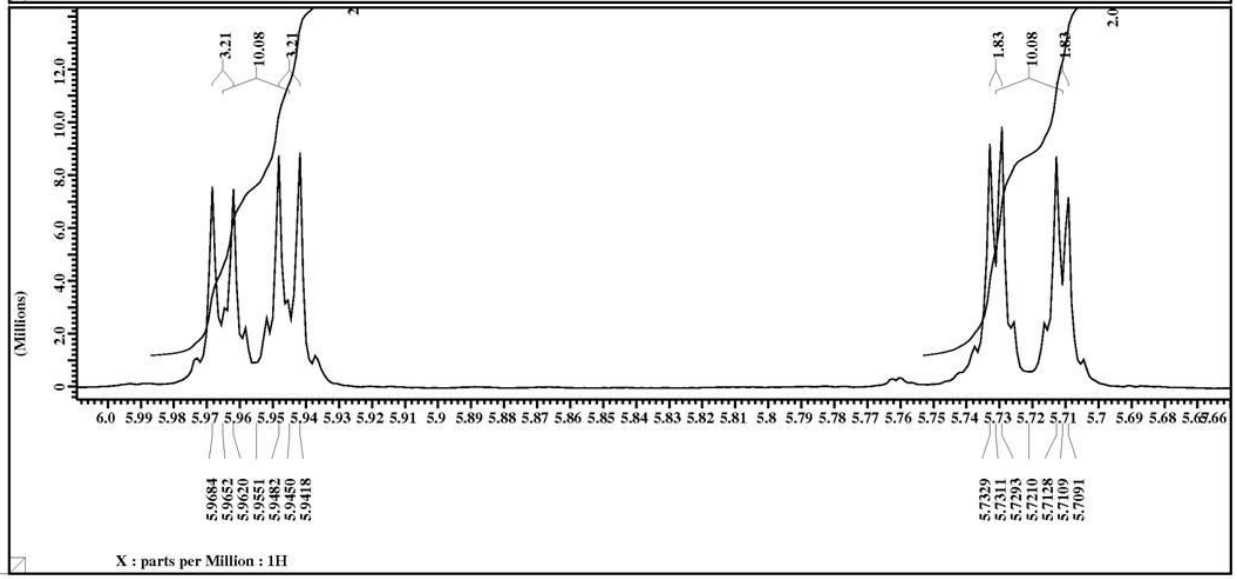


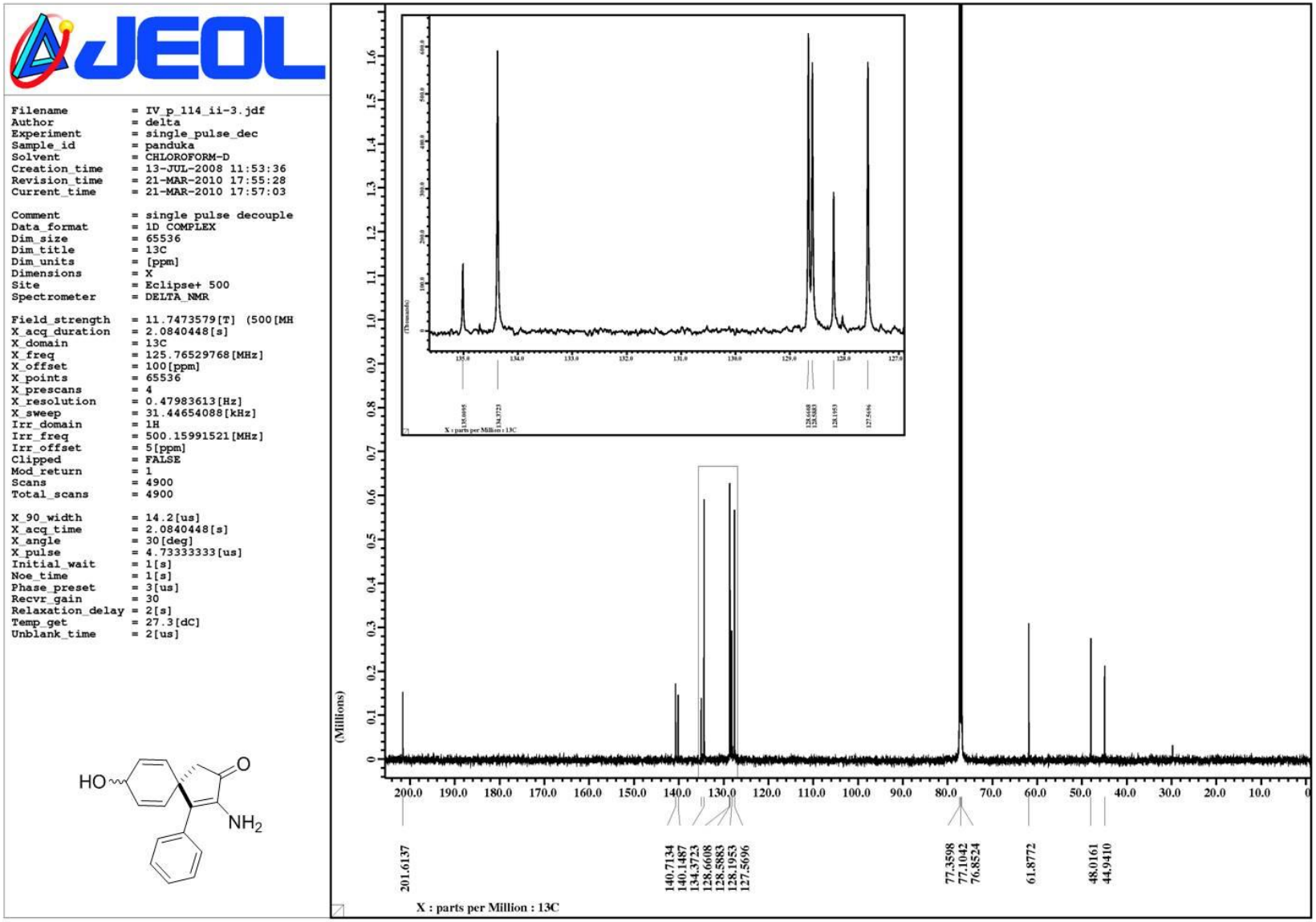


```
Filename          = IV_p_114_ii-2.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#778738
Solvent           = CHLOROFORM-D
Creation_time     = 13-JUL-2008 04:30:21
Revision_time     = 21-MAR-2010 18:03:49
Current_time      = 21-MAR-2010 18:04:47

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 24.7[dC]
Unblank_time      = 2[us]
```









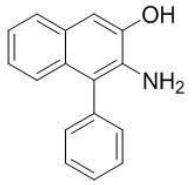
```
Filename          = IV_p_114_ii-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 13-JUL-2008 11:53:36
Revision_time     = 21-MAR-2010 17:55:28
Current_time      = 21-MAR-2010 17:57:03

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 4900
Total_scans       = 4900

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.3[dC]
Unblank_time      = 2[us]
```

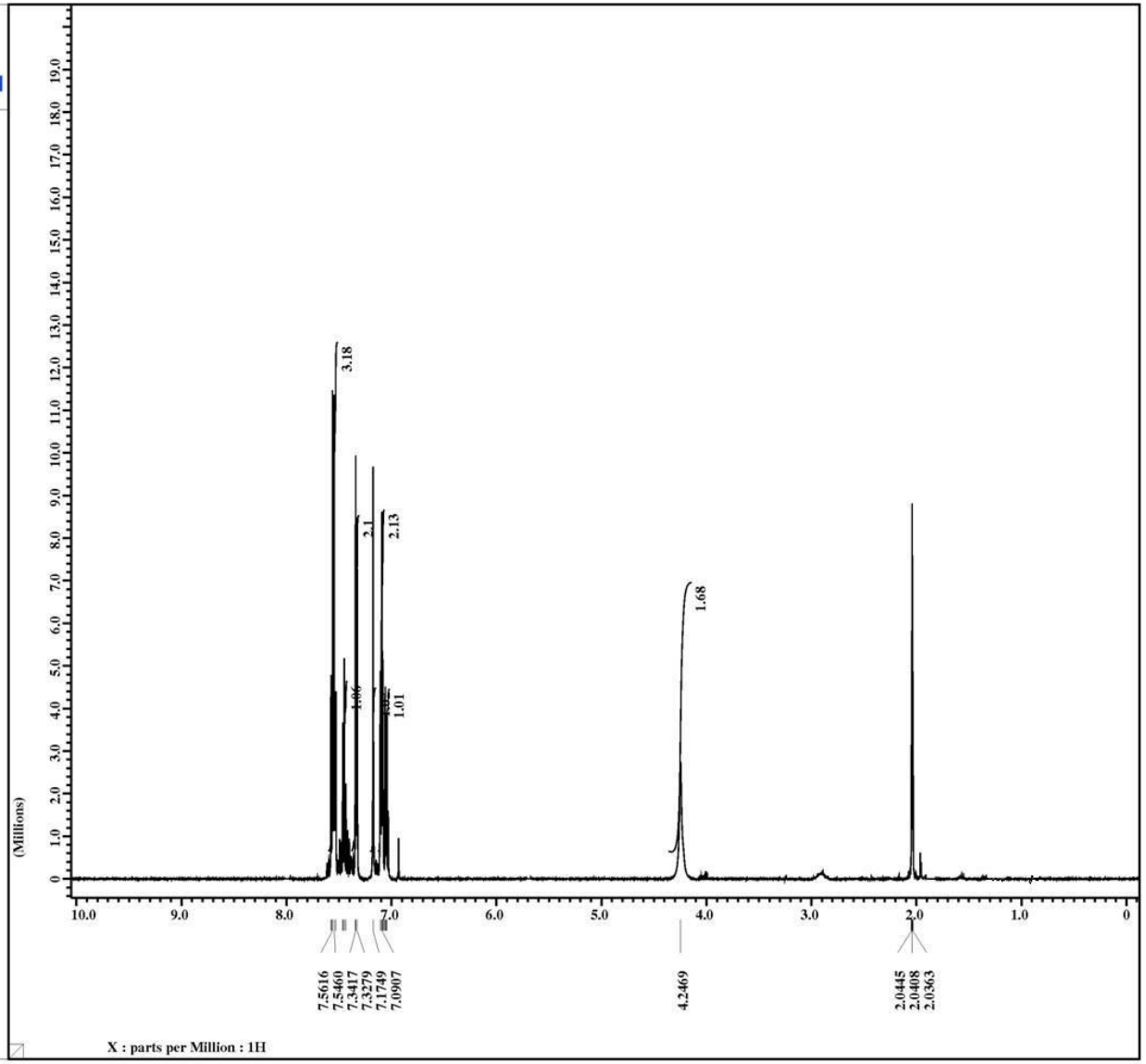
APPENDIX 122

^{1}H and ^{13}C NMR Spectra of

3-Amino-4-phenyl-2-naphthol (**262**)

```
Filename          = IV_p_120_crude-3.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#595038
Solvent           = ACETONE-D6
Creation_time     = 16-JUL-2008 23:26:23
Revision_time     = 21-MAR-2010 18:33:39
Current_time      = 21-MAR-2010 18:33:47

Comment           = Single Pulse Experime
Data_format       = 1D REAL
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 20
Relaxation_delay  = 4[s]
Temp_get          = 24.8[dC]
Unblank_time      = 2[us]
```





JEOL
Filename = IV_p_120_crude-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = s#595038
Solvent = ACETONE-D6
Creation_time = 16-JUL-2008 23:26:23
Revision_time = 21-MAR-2010 18:33:39
Current_time = 21-MAR-2010 18:34:00
Comment = Single Pulse Experime
Data_format = 1D REAL
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 20
Relaxation_delay = 4[s]
Temp_get = 24.8[dC]
Unblank_time = 2[us]
OH
NH2
(Millions)
3.18
1.06
2.1
1.02
2.13
1.01
7.5762
7.5726
7.5616
7.5460
7.5322
7.5295
7.4635
7.4608
7.4480
7.4333
7.3444
7.3417
7.3279
7.1749
7.1071
7.0943
7.0907
7.0879
7.0806
7.0613
7.0577
7.0439
X : parts per Million : 1H

JEOL
Filename = brown solid-3.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#519022
Solvent = METHANOL-D3
Creation_time = 19-APR-2010 14:15:39
Revision_time = 19-APR-2010 15:09:04
Current_time = 19-APR-2010 15:11:00
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 26214
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 1.38412032[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 32768
X_prescans = 4
X_resolution = 0.72248054[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 140
Total_scans = 140
X_90_width = 9.75[us]
X_acq_time = 1.38412032[s]
X_angle = 30[deg]
X_atn = 8[dB]
X_pulse = 3.25[us]
Irr_atn_dec = 25[dB]
Irr_atn_noe = 25[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 3.38412032[s]
Temp_get = 24.8[dC]
OH
NH2
abundance
133.7012
130.5940
128.9018
128.8540
128.7775
127.2096
125.6608
123.4714
122.7256
122.0851
120.8135
X : parts per Million : 13C
abundance
145.8528
137.5159
130.5940
128.9018
128.8540
128.7775
127.2096
125.6608
123.4714
122.7256
122.0851
120.8135
107.6389
47.9901
47.7129
47.4261
X : parts per Million : 13C

APPENDIX 123

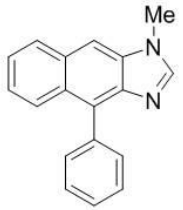
^{1}H and ^{13}C NMR Spectra of

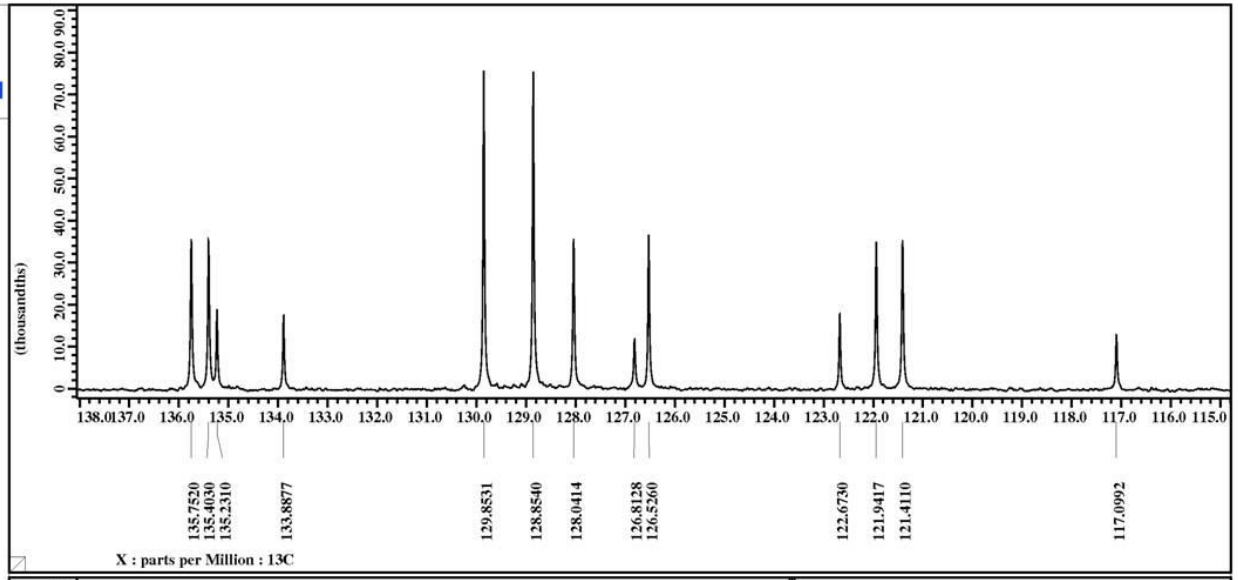
1-Methyl-4-phenyl-1*H*-naphtho[2,3-*d*]imidazole (**263**)

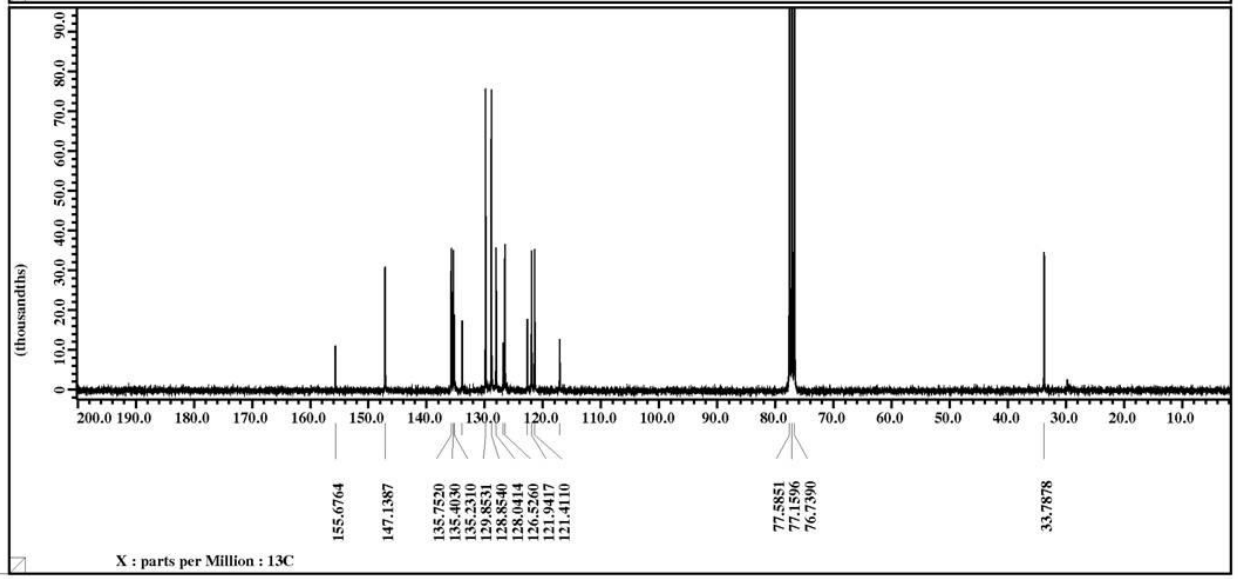
JEOL
Filename = IV_p_131_ii-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#633669
Solvent = CHLOROFORM-D
Creation_time = 24-JUL-2008 00:35:16
Revision_time = 21-MAR-2010 18:53:26
Current_time = 21-MAR-2010 18:53:39
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 22
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
Me
N
N
(Millions)
X : parts per Million : 1H
10.08
9.16
7.79
7.79
7.29
10.08
7.79
2.09
2.14
1.17
1.17
1.18
1.14
1
1.02
1.04
8.7150
8.7049
8.6948
8.2789
8.2606
8.0435
7.9784
7.9628
7.5542
7.5387
7.5231
7.4663
7.3417
7.2546
7.0110
3.23
4.2121

JEOL
Filename = IV_p_131_ii-3.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#633669
Solvent = CHLOROFORM-D
Creation_time = 24-JUL-2008 00:35:16
Revision_time = 21-MAR-2010 18:54:16
Current_time = 21-MAR-2010 18:55:21
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 22
Relaxation_delay = 4[s]
Temp_get = 25.2[dC]
Unblank_time = 2[us]
Me
N
N
(Millions)
X : parts per Million : 1H

```
Filename          = IV_p_131-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#761966
Solvent           = CHLOROFORM-D
Creation_time     = 31-JUL-2008 05:59:54
Revision_time     = 21-MAR-2010 18:56:36
Current_time      = 21-MAR-2010 18:57:26

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 6400
Total_scans       = 6400

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 22.1[dC]
```







APPENDIX 124

^{1}H and ^{13}C NMR Spectra of

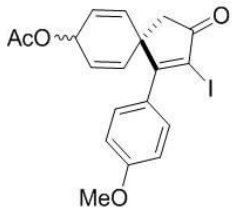
2-iodo-1-(4-methoxy-phenyl)-3-oxo-spiro[4.5]deca-1,6,9-trien-8-yl-acetate

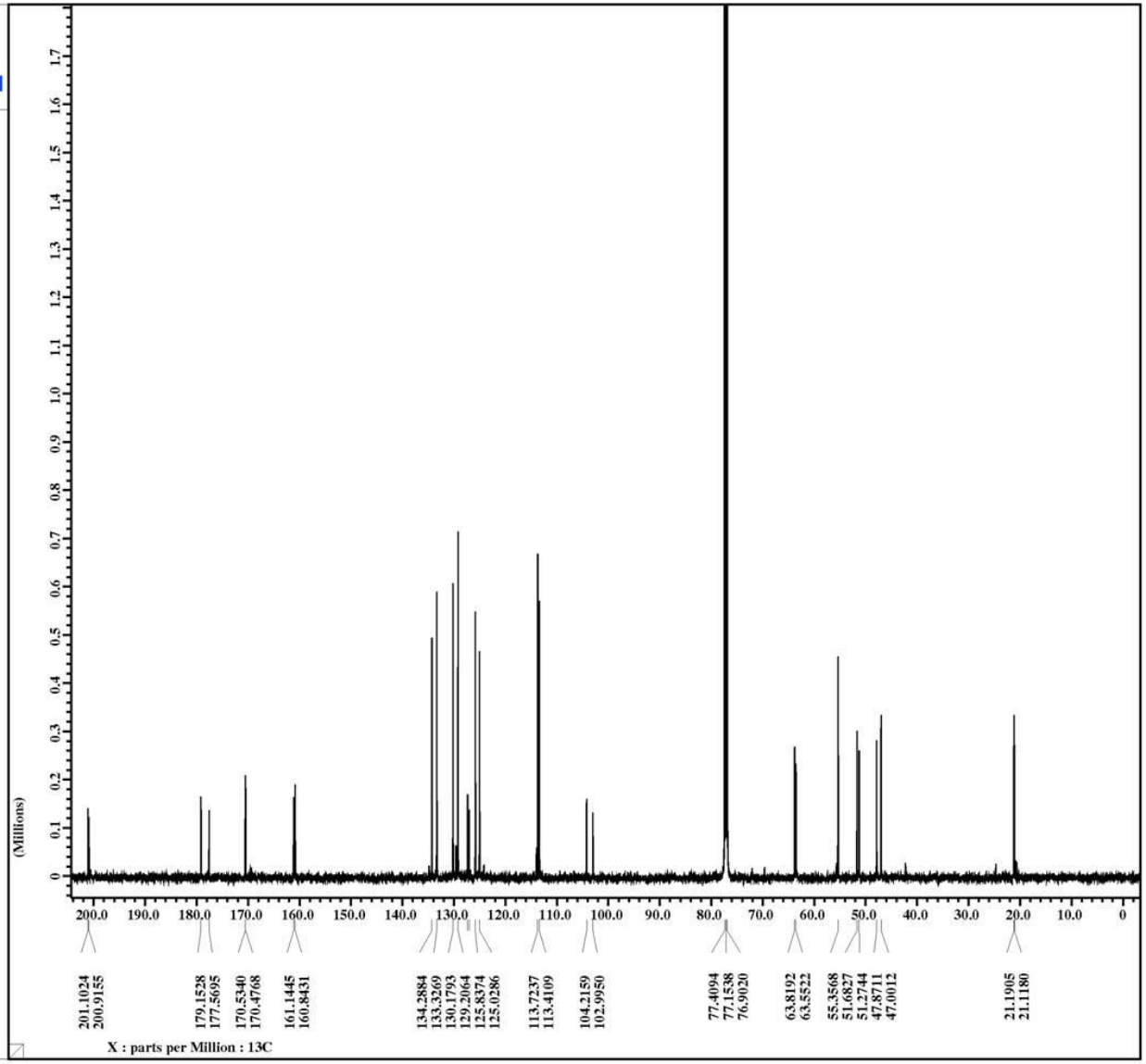
(**251**)

JEOL
Filename = IV_p_231_spiro acetat
Author = delta
Experiment = single_pulse.exp
Sample_id = S#766535
Solvent = CHLOROFORM-D
Creation_time = 19-SEP-2008 04:50:17
Revision_time = 22-MAR-2010 18:00:56
Current_time = 22-MAR-2010 18:01:11
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 17
Relaxation_delay = 4[s]
Temp_get = 25.5[dC]
Unblank_time = 2[us]
(Millions)
X : parts per Million : 1H
AcO
MeO

JEOL
Filename = IV_p_231_spiro acetat
Author = delta
Experiment = single_pulse.exp
Sample_id = S#766535
Solvent = CHLOROFORM-D
Creation_time = 19-SEP-2008 04:50:17
Revision_time = 22-MAR-2010 18:01:54
Current_time = 22-MAR-2010 18:02:13
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 17
Relaxation_delay = 4[s]
Temp_get = 25.5[dC]
Unblank_time = 2[us]
AcO
MeO
(Millions)
X : parts per Million : 1H

```
Filename          = IV_p_231_spiro acetat
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#754436
Solvent           = CHLOROFORM-D
Creation_time     = 20-SEP-2008 11:25:33
Revision_time     = 21-SEP-2008 13:17:03
Current_time      = 22-MAR-2010 18:02:42

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = TRUE
Mod_return        = 1
Scans             = 4900
Total_scans       = 4900

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.8[dC]
Unblank_time      = 2[us]
```





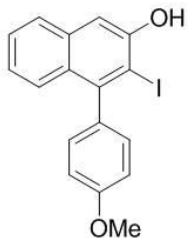
JEOL
Filename = IV_p_231_spiro acetat
Author = delta
Experiment = single_pulse_dec
Sample_id = S#754436
Solvent = CHLOROFORM-D
Creation_time = 20-SEP-2008 11:25:33
Revision_time = 21-SEP-2008 13:17:03
Current_time = 22-MAR-2010 18:04:30
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 65536
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.0840448[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 65536
X_prescans = 4
X_resolution = 0.47983613[Hz]
X_sweep = 31.44654088[kHz]
Irr_domain = 1H
Irr_freq = 500.159991521[MHz]
Irr_offset = 5[ppm]
Clipped = TRUE
Mod_return = 1
Scans = 4900
Total_scans = 4900
X_90_width = 14.2[us]
X_acq_time = 2.0840448[s]
X_angle = 30[deg]
X_pulse = 4.73333333[us]
Initial_wait = 1[s]
Noe_time = 1[s]
Phase_preset = 3[us]
Recvr_gain = 30
Relaxation_delay = 2[s]
Temp_get = 28.8[dC]
Unblank_time = 2[us]
AcO
I
MeO
X : parts per Million : 13C
(Thousands)
(Millions)
179.1528
177.5695
201.1024
200.9155
170.5340
170.4768
161.1445
160.8431
134.2884
133.3269
130.1793
129.2064
127.3483
127.0278
125.8374
125.0286
113.7237
113.4109
104.2159
102.9950
63.8192
63.5522
55.3568
51.6827
51.2744
21.1905
21.1180

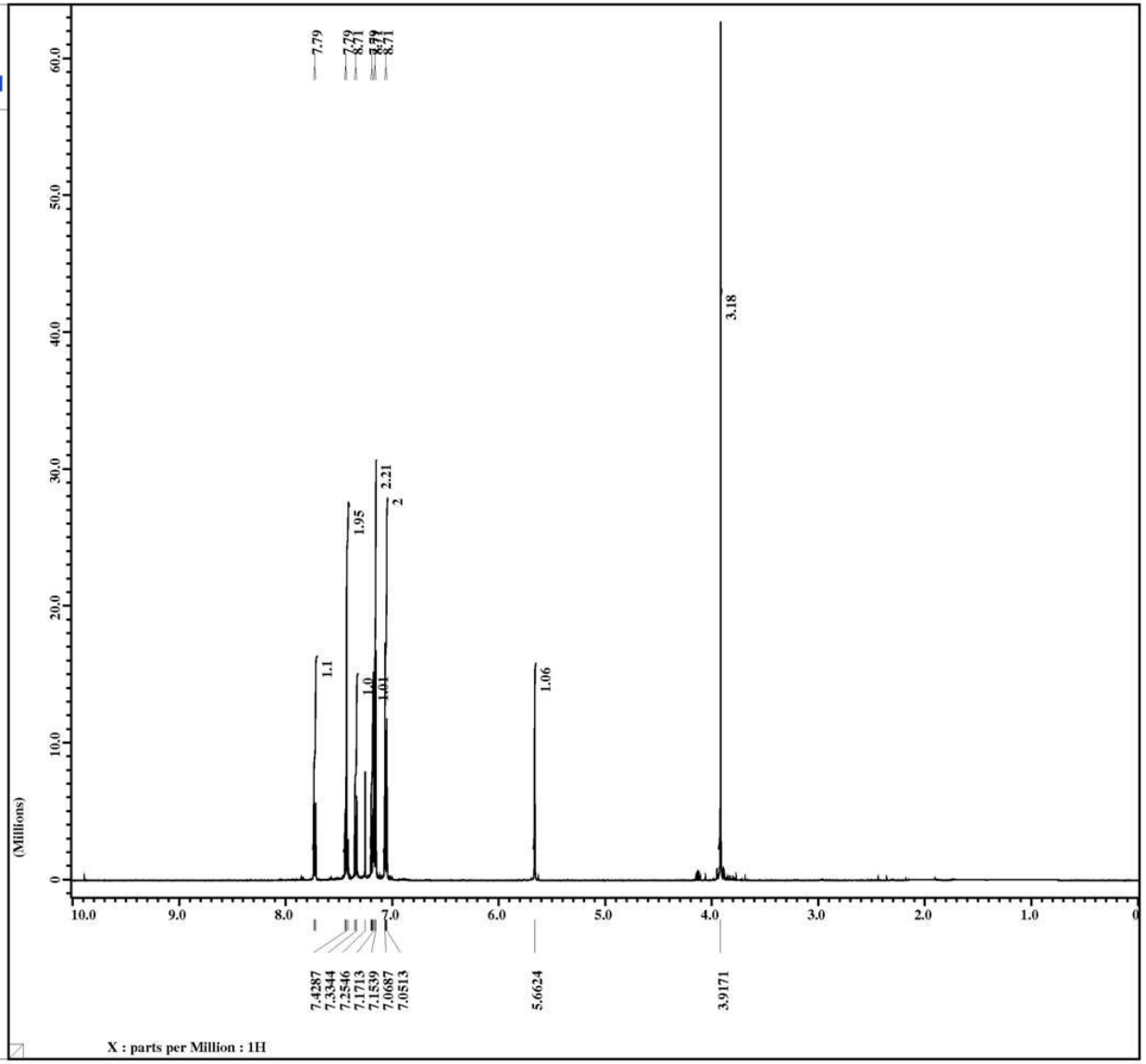
APPENDIX 125

^{1}H, ^{13}C and DEPT NMR Spectra of

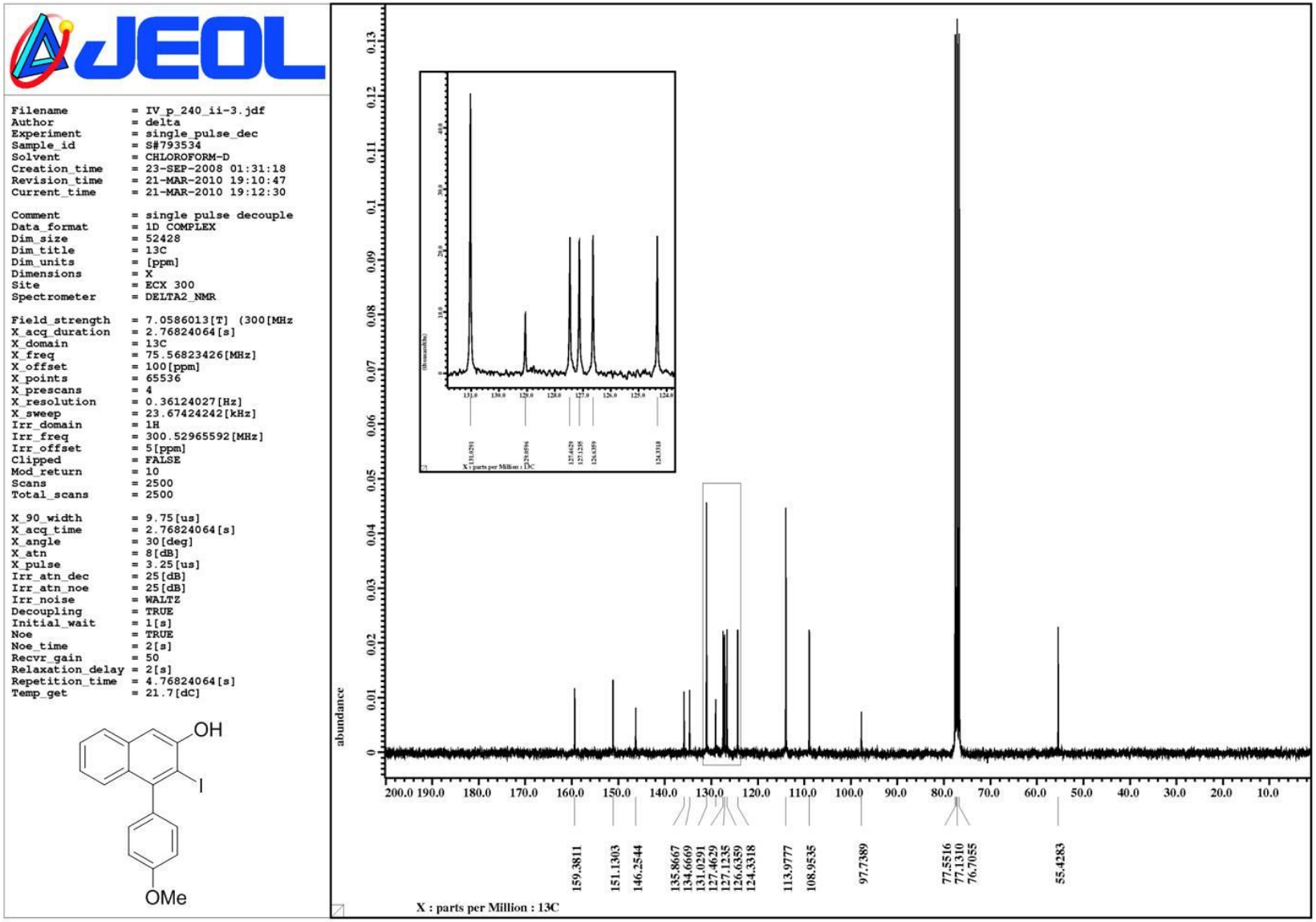
3-Iodo-4-(4-methoxy-phenyl)-naphthalen-2-ol (**271**)

```
Filename          = IV_p_240_ii-4.jdf
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#772537
Solvent           = CHLOROFORM-D
Creation_time     = 23-SEP-2008 05:02:38
Revision_time     = 21-MAR-2010 19:15:05
Current_time      = 21-MAR-2010 19:15:22

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 22
Relaxation_delay  = 4[s]
Temp_get          = 24.8[dC]
Unblank_time      = 2[us]
```



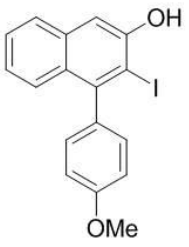


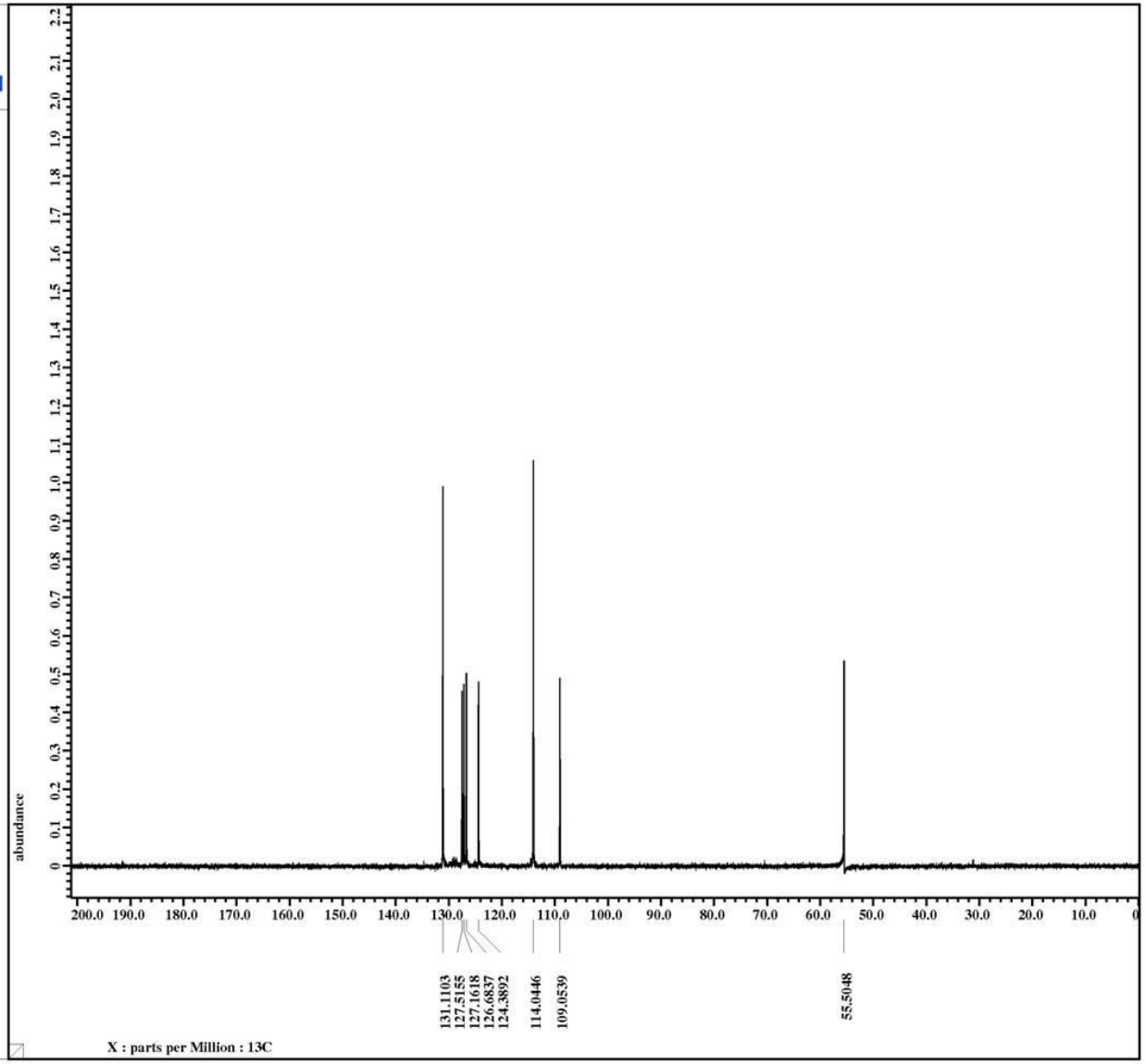
JEOL
Filename = IV_p_240_ii-4.jdf
Author = delta
Experiment = single_pulse.exp
Sample_id = S#772537
Solvent = CHLOROFORM-D
Creation_time = 23-SEP-2008 05:02:38
Revision_time = 21-MAR-2010 19:15:05
Current_time = 21-MAR-2010 19:15:39
Comment = Single Pulse Experime
Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 500
Spectrometer = DELTA_NMR
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 2.1823488[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.45822189[Hz]
X_sweep = 7.50750751[kHz]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 18.5[us]
X_acq_time = 2.1823488[s]
X_angle = 45[deg]
X_pulse = 9.25[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 22
Relaxation_delay = 4[s]
Temp_get = 24.8[dC]
Unblank_time = 2[us]
OH
I
OMe
(Millions)
X : parts per Million : 1H
7.79
8.71
7.79
8.71
7.79
8.71
1.1
1.95
1.0
1.01
2.21
2
7.7356
7.7278
7.7201
7.4443
7.4365
7.4287
7.4150
7.3518
7.3431
7.3344
7.2546
7.1997
7.1969
7.1859
7.1832
7.1804
7.1713
7.1539
7.0687
7.0641
7.0549
7.0513



```
Filename          = IV_p_240_ii-3.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#793534
Solvent           = CHLOROFORM-D
Creation_time     = 23-SEP-2008 01:31:18
Revision_time     = 21-MAR-2010 19:10:47
Current_time      = 21-MAR-2010 19:12:30

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 52428
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 2.76824064[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.36124027[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 10
Scans             = 2500
Total_scans       = 2500

X_90_width        = 9.75[us]
X_acq_time        = 2.76824064[s]
X_angle           = 30[deg]
X_atn             = 8[dB]
X_pulse           = 3.25[us]
Irr_atn_dec       = 25[dB]
Irr_atn_noe       = 25[dB]
Irr_noise         = WALTZ
Decoupling        = TRUE
Initial_wait      = 1[s]
Noe               = TRUE
Noe_time          = 2[s]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Repetition_time   = 4.76824064[s]
Temp_get          = 21.7[dC]
```

```
Filename          = IV_p_240_ii-3.jdf
Author            = delta
Experiment        = dept.ex2
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 28-SEP-2008 22:33:49
Revision_time     = 21-MAR-2010 19:17:14
Current_time      = 21-MAR-2010 19:17:24

Comment           = DEPT with decoupling
Data_format       = 1D COMPLEX
Dim_size          = 26214
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 1.38412032[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 32768
X_prescans        = 4
X_resolution      = 0.72248054[Hz]
X_sweep           = 23.67424242[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 500
Total_scans       = 500

X_acq_time        = 1.38412032[s]
X_atn             = 8[dB]
X_pulse           = 9.75[us]
Irr_atn           = 4[dB]
Irr_atn_dec       = 25[dB]
Irr_noise         = WALTZ
Irr_pulse         = 13.01[us]
Decoupling        = TRUE
Initial_wait      = 1[s]
J_constant        = 140[Hz]
Recvr_gain        = 50
Relaxation_delay  = 2[s]
Selection_angle   = 135[deg]
Selection_pulse   = 19.515[us]
Temp_get          = 21.7[dC]
```

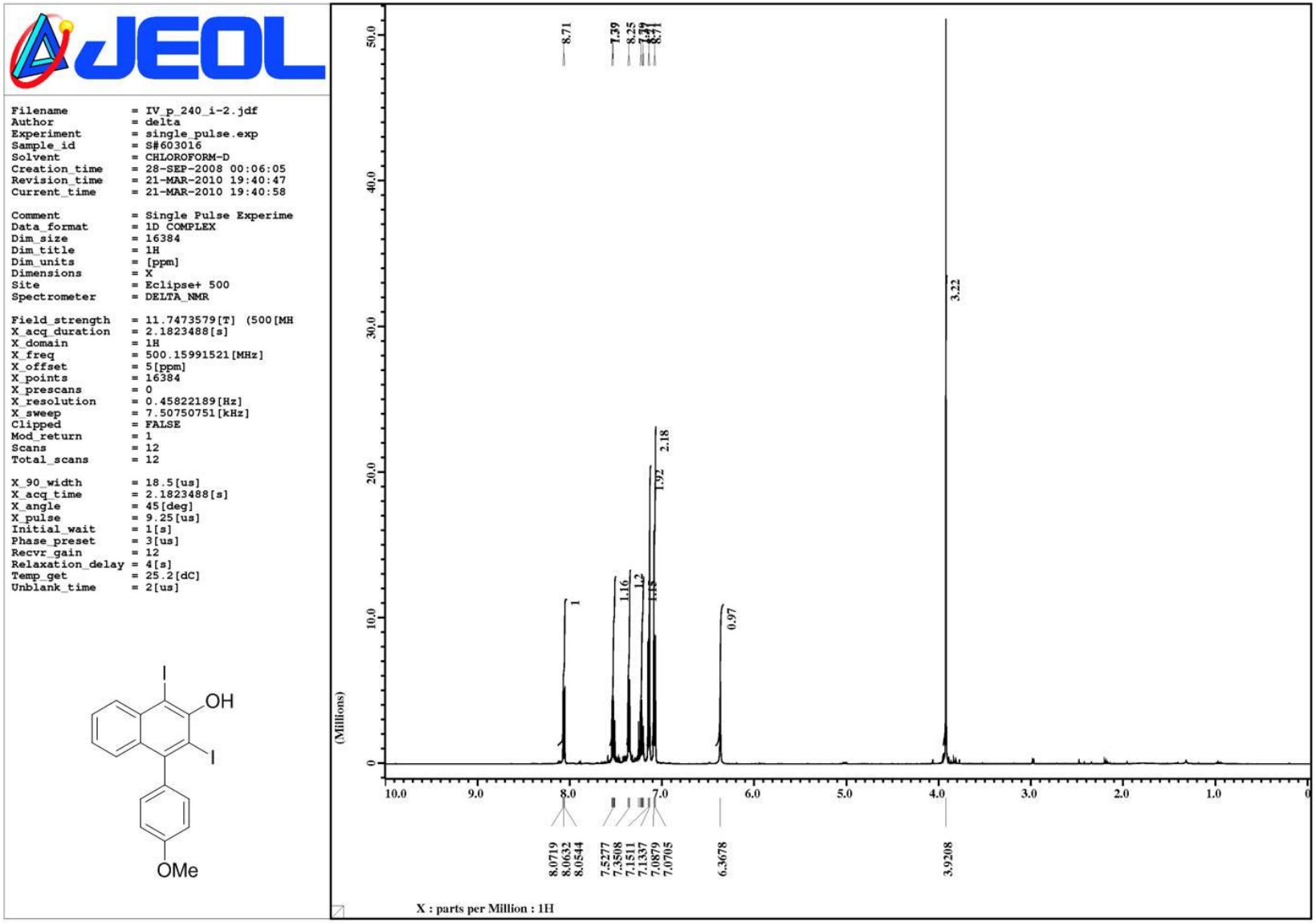




APPENDIX 126

^{1}H, ^{13}C and DEPT NMR Spectra of

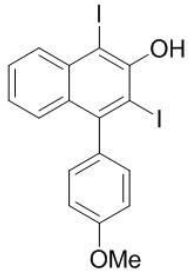
1,3-Diiodo-4-(4-methoxy-phenyl)-naphthalen-2-ol (**272**)

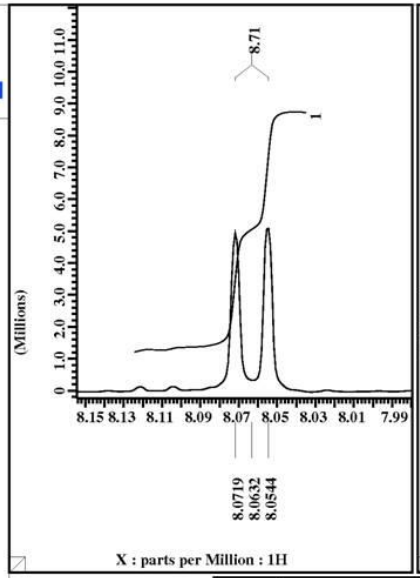


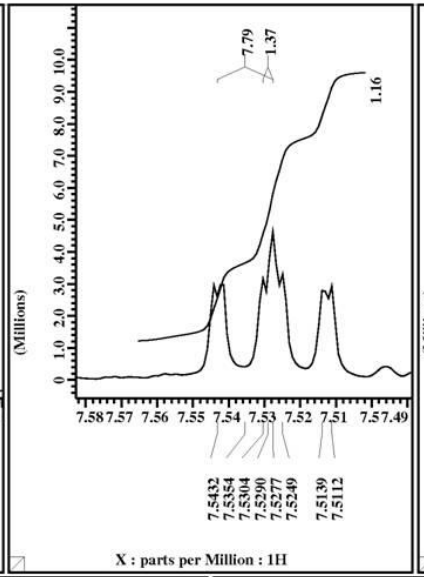
Filename	= IV_p_240_i-2.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#603016
Solvent	= CHLOROFORM-D
Creation_time	= 28-SEP-2008 00:06:05
Revision_time	= 21-MAR-2010 19:40:47
Current_time	= 21-MAR-2010 19:40:58
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 12
Relaxation_delay	= 4[s]
Temp_get	= 25.2[dC]
Unblank_time	= 2[us]

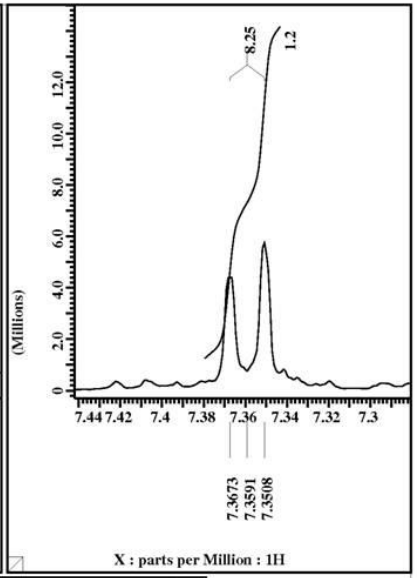
JEOL

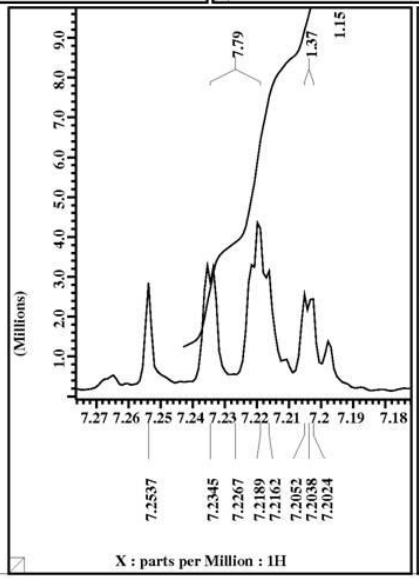
Filename	= IV_p_240_i-2.jdf
Author	= delta
Experiment	= single_pulse.exp
Sample_id	= S#603016
Solvent	= CHLOROFORM-D
Creation_time	= 28-SEP-2008 00:06:05
Revision_time	= 21-MAR-2010 19:40:47
Current_time	= 21-MAR-2010 19:42:21
Comment	= Single Pulse Experime
Data_format	= 1D COMPLEX
Dim_size	= 16384
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 500
Spectrometer	= DELTA_NMR
Field_strength	= 11.7473579[T] (500[MH
X_acq_duration	= 2.1823488[s]
X_domain	= 1H
X_freq	= 500.15991521[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.45822189[Hz]
X_sweep	= 7.50750751[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 12
Total_scans	= 12
X_90_width	= 18.5[us]
X_acq_time	= 2.1823488[s]
X_angle	= 45[deg]
X_pulse	= 9.25[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 12
Relaxation_delay	= 4[s]
Temp_get	= 25.2[dC]
Unblank_time	= 2[us]

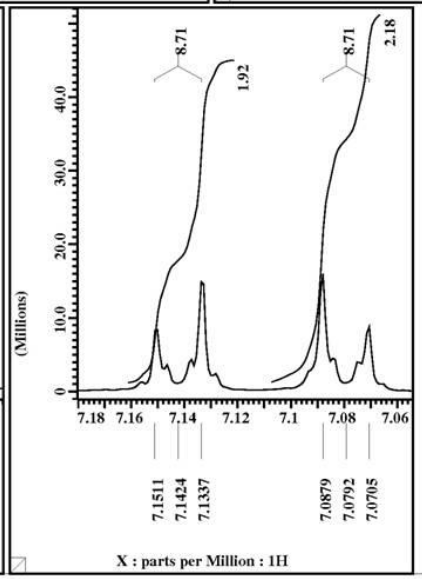




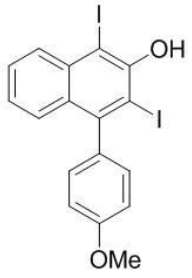


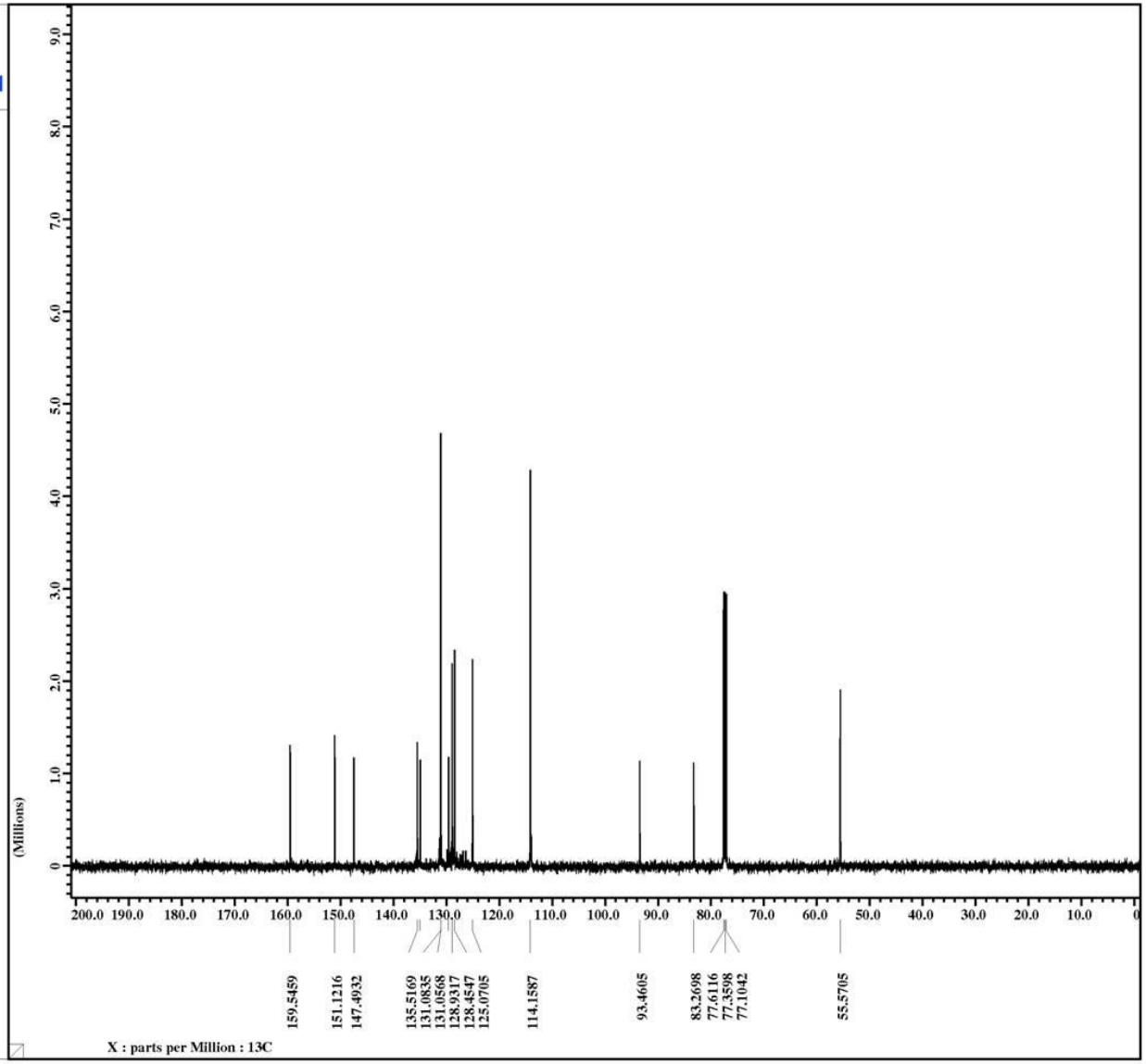




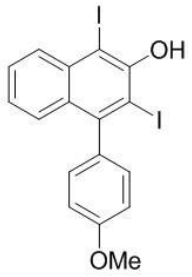


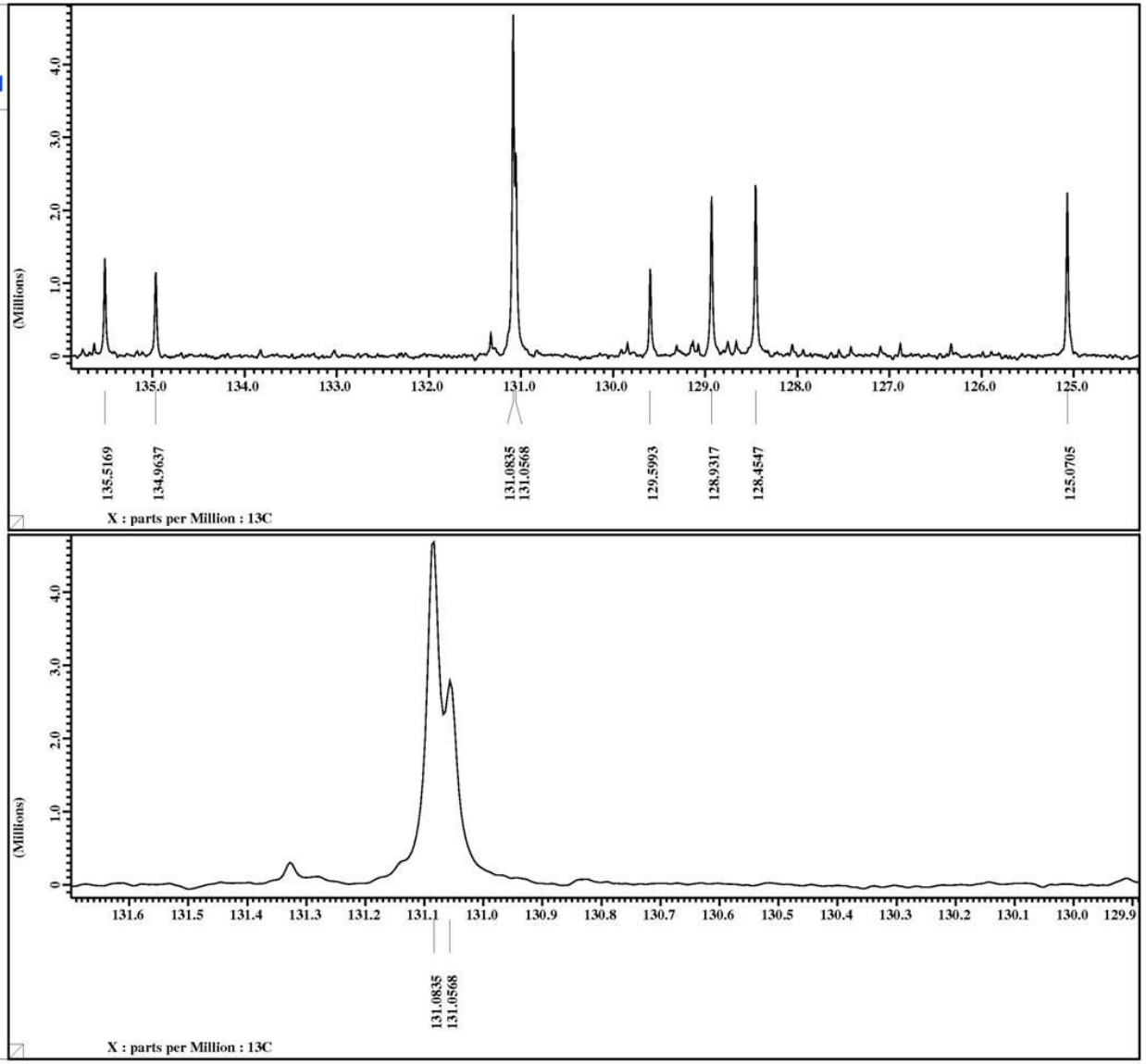
```
Filename          = IV_p_240_i-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 28-SEP-2008 00:22:34
Revision_time     = 21-MAR-2010 19:43:17
Current_time      = 21-MAR-2010 19:46:47

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 169
Total_scans       = 169

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.6[dC]
Unblank_time      = 2[us]
```

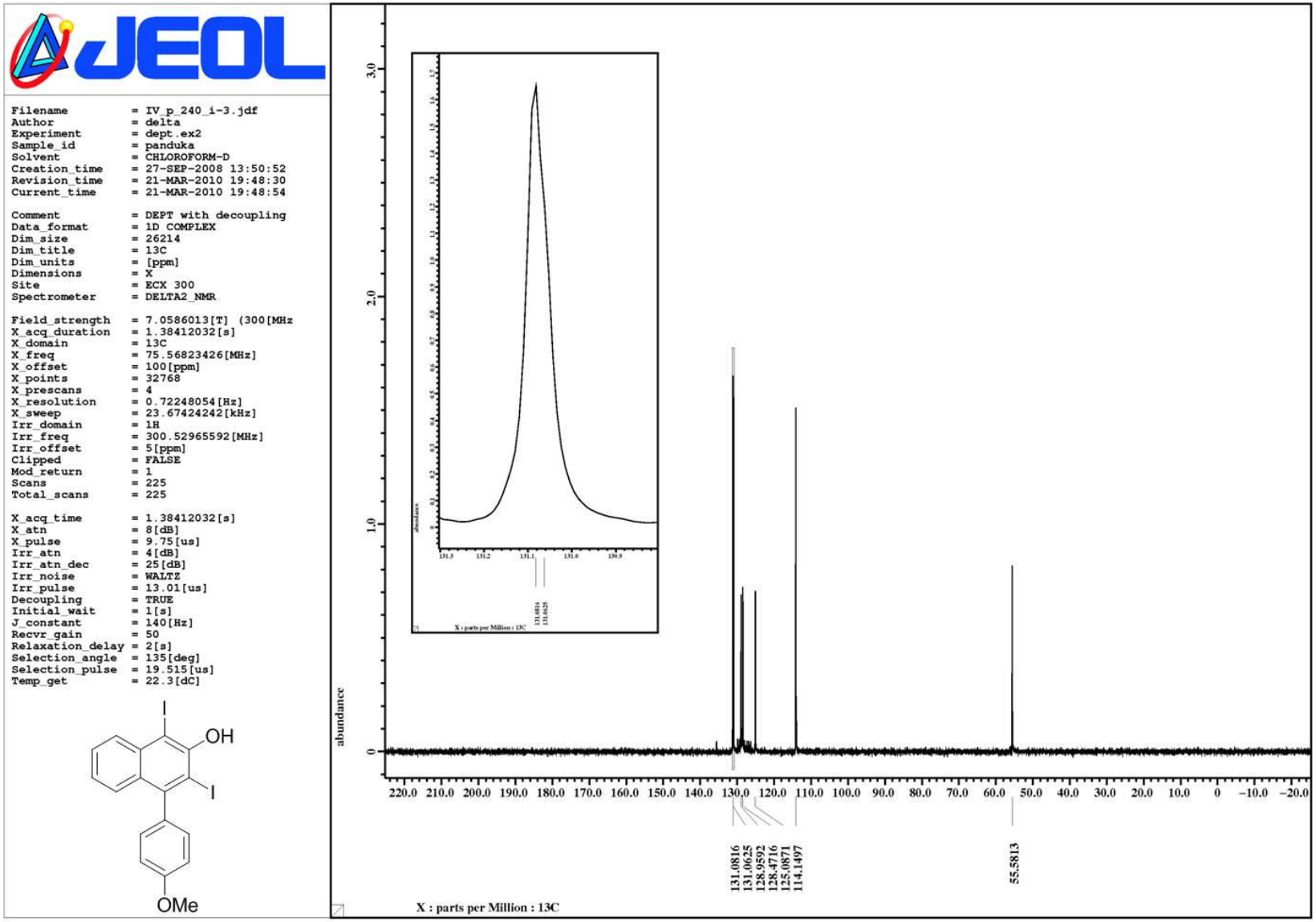




```
Filename          = IV_p_240_i-4.jdf
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = panduka
Solvent           = CHLOROFORM-D
Creation_time     = 28-SEP-2008 00:22:34
Revision_time     = 21-MAR-2010 19:43:17
Current_time      = 21-MAR-2010 19:47:16

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 169
Total_scans       = 169

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 27.6[dC]
Unblank_time      = 2[us]
```






Filename = IV_p_240_i-3.jdf
Author = delta
Experiment = dept.ex2
Sample_id = panduka
Solvent = CHLOROFORM-D
Creation_time = 27-SEP-2008 13:50:52
Revision_time = 21-MAR-2010 19:48:30
Current_time = 21-MAR-2010 19:48:54
Comment = DEPT with decoupling
Data_format = 1D COMPLEX
Dim_size = 26214
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECX 300
Spectrometer = DELTA2_NMR
Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 1.38412032[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 32768
X_prescans = 4
X_resolution = 0.72248054[Hz]
X_sweep = 23.67424242[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 225
Total_scans = 225
X_acq_time = 1.38412032[s]
X_atn = 8[dB]
X_pulse = 9.75[us]
Irr_atn = 4[dB]
Irr_atn_dec = 25[dB]
Irr_noise = WALTZ
Irr_pulse = 13.01[us]
Decoupling = TRUE
Initial_wait = 1[s]
J_constant = 140[Hz]
Recvr_gain = 50
Relaxation_delay = 2[s]
Selection_angle = 135[deg]
Selection_pulse = 19.515[us]
Temp_get = 22.3[dC]
I
OH
I
OMe
abundance
3.0
2.0
1.0
0
220.0 210.0 200.0 190.0 180.0 170.0 160.0 150.0 140.0 130.0 120.0 110.0 100.0 90.0 80.0 70.0 60.0 50.0 40.0 30.0 20.0 10.0 0 −10.0 −20.0
131.0816
131.0625
128.9592
128.4716
125.0871
114.1497
55.5813
X : parts per Million : 13C

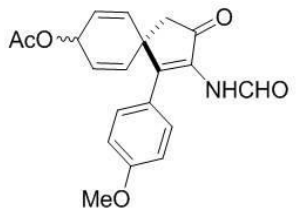
APPENDIX 127

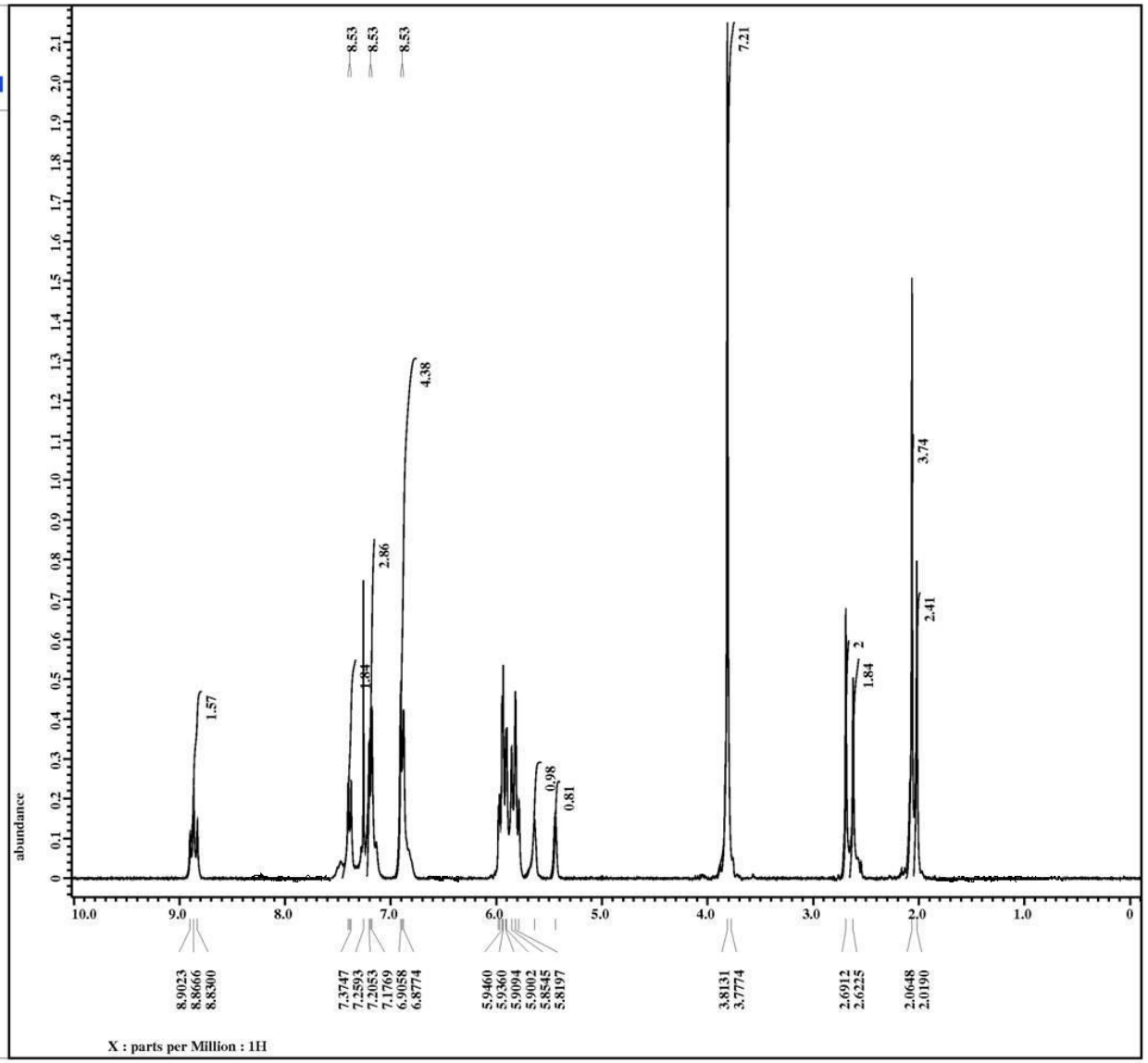
^{1}H and ^{13}C NMR Spectra of

2-formylamino-1-(4-methoxy-phenyl)-3-oxo-spiro[4.5]deca-1,6,9-trien-8-yl-acetate

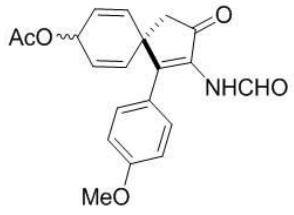
(**250**)

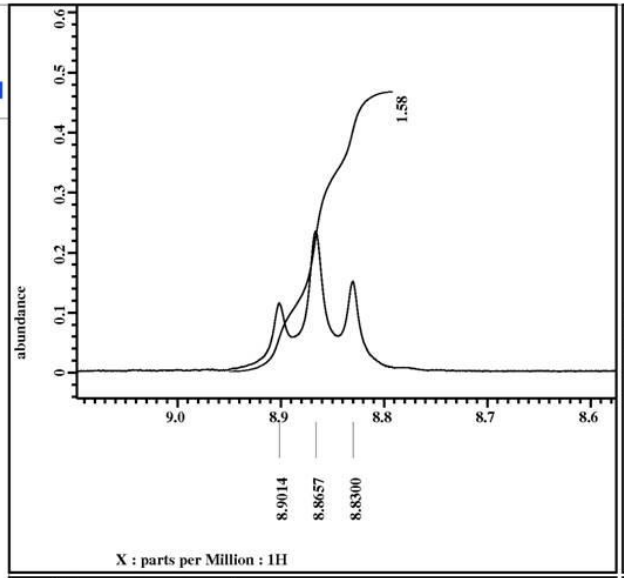
```
Filename          = IV_p_286_ii-3.jdf
Author            = delta
Experiment        = single_pulse.ex2
Sample_id         = S#568452
Solvent           = CHLOROFORM-D
Creation_time     = 27-OCT-2008 16:02:01
Revision_time     = 14-MAR-2010 19:59:49
Current_time      = 14-MAR-2010 20:00:05

Comment           = single_pulse
Data_format       = 1D REAL
Dim_size          = 13107
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = ECX 300
Spectrometer      = DELTA2_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 3.63331584[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.27523068[Hz]
X_sweep           = 4.50937951[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Tri_domain        = 1H
Tri_freq          = 300.52965592[MHz]
Tri_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 9
Total_scans       = 9

X_90_width        = 13.01[us]
X_acq_time        = 3.63331584[s]
X_angle           = 45[deg]
X_atn             = 4[dB]
X_pulse           = 6.505[us]
Irr_mode          = Off
Tri_mode          = Off
Dante_presat      = FALSE
Initial_wait      = 1[s]
Recvr_gain        = 40
Relaxation_delay  = 5[s]
Repetition_time   = 8.63331584[s]
Temp_get          = 23[dC]
```

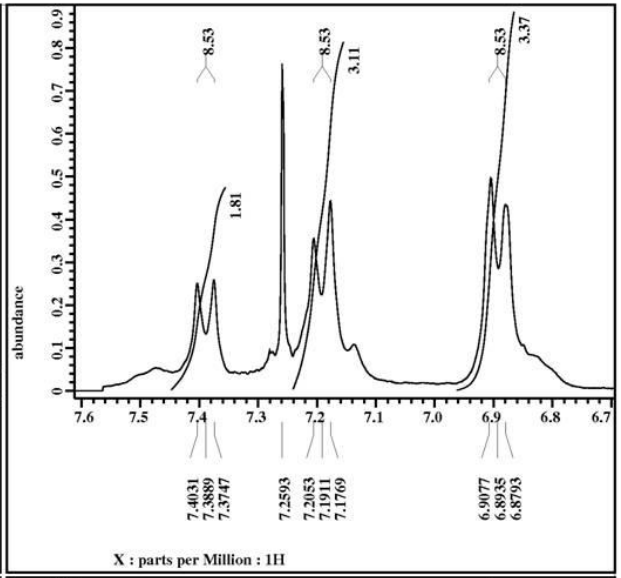


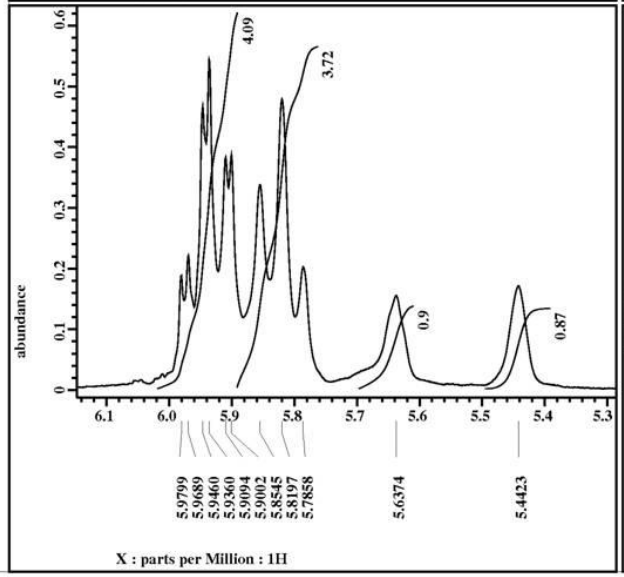


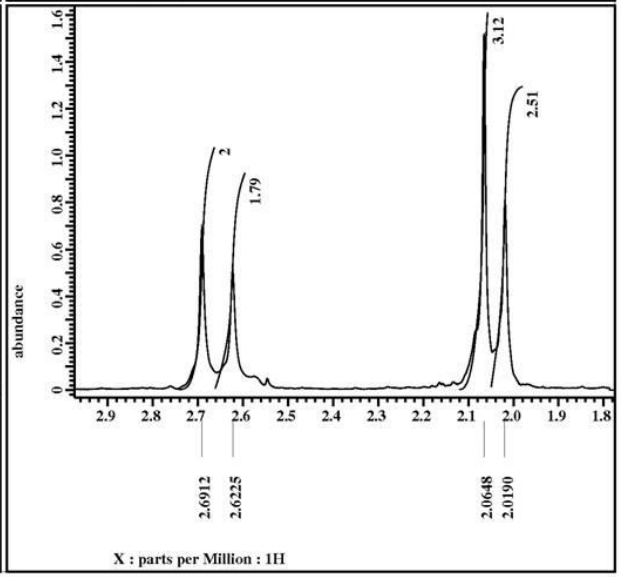
Filename	= IV_p_286_ii-6.jdf
Author	= delta
Experiment	= single_pulse.ex2
Sample_id	= S#568452
Solvent	= CHLOROFORM-D
Creation_time	= 27-OCT-2008 16:02:01
Revision_time	= 14-MAR-2010 20:27:33
Current_time	= 14-MAR-2010 20:28:15
Comment	= single_pulse
Data_format	= 1D COMPLEX
Dim_size	= 13107
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= ECX 300
Spectrometer	= DELTA2_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 3.63331584[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 16384
X_prescans	= 0
X_resolution	= 0.27523068[Hz]
X_sweep	= 4.50937951[kHz]
Irr_domain	= 1H
Irr_freq	= 300.52965592[MHz]
Irr_offset	= 5[ppm]
Tri_domain	= 1H
Tri_freq	= 300.52965592[MHz]
Tri_offset	= 5[ppm]
Clipped	= FALSE
Mod_return	= 1
Scans	= 9
Total_scans	= 9
X_90_width	= 13.01[us]
X_acq_time	= 3.63331584[s]
X_angle	= 45[deg]
X_atn	= 4[dB]
X_pulse	= 6.505[us]
Irr_mode	= Off
Tri_mode	= Off
Dante_presat	= FALSE
Initial_wait	= 1[s]
Recvr_gain	= 40
Relaxation_delay	= 5[s]
Repetition_time	= 8.63331584[s]
Temp_get	= 23[dC]

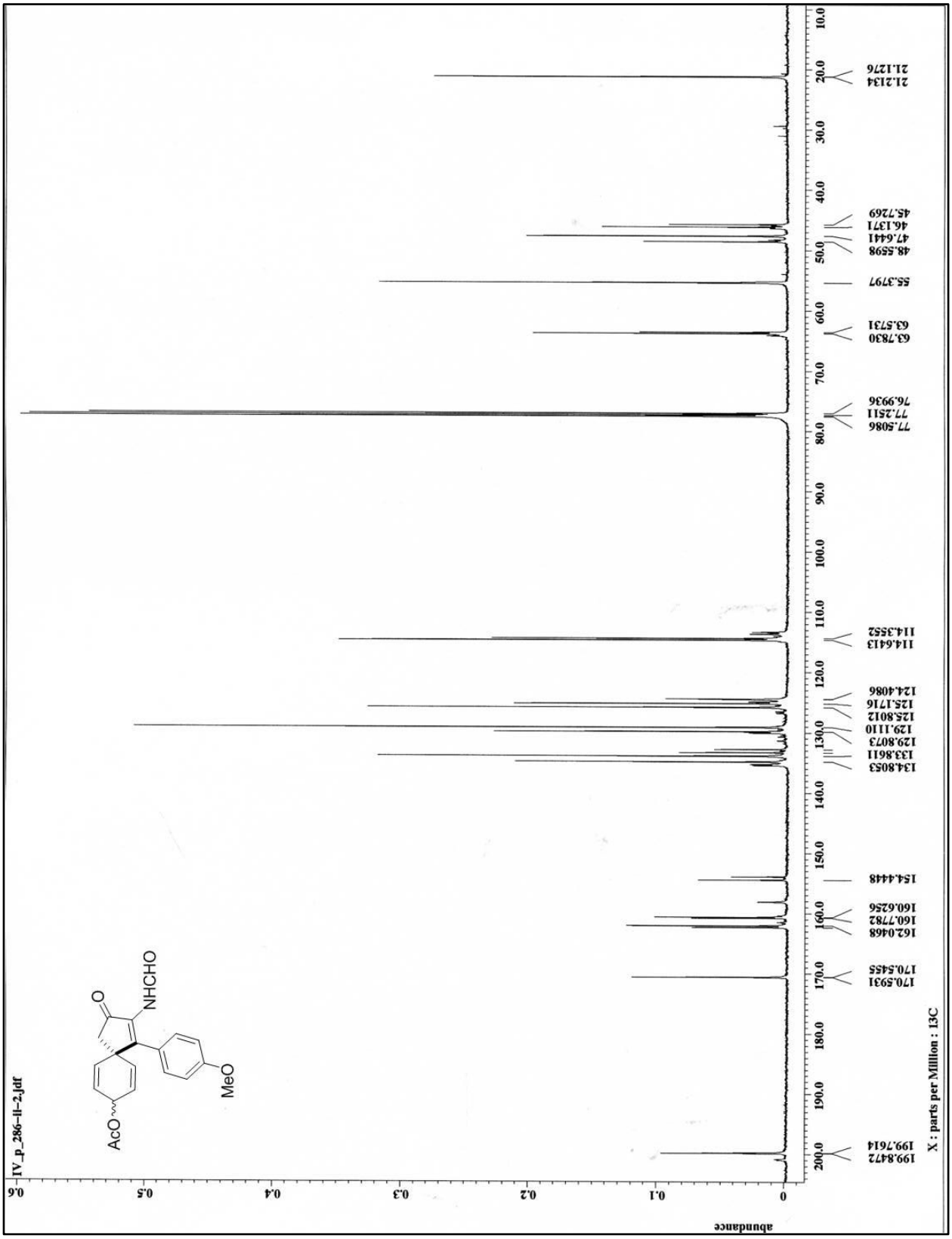











IV_p_286-II-2.jdf
X : parts per Million : 13C
abundance
199.8472
199.7614
170.5931
170.5455
162.0468
160.7782
160.6256
154.4448
134.8053
133.8611
129.8073
129.1110
125.8012
125.1716
124.4086
114.6413
114.3552
77.5086
77.2511
76.9936
63.7830
63.5731
55.3797
48.5598
47.6441
46.1371
45.7269
21.2134
21.1276
NHCHO
MeO
AcO

IV_p_286-II_copy-2.jdf
abundance
X : parts per Million : 13C
199.8472
199.7614
170.5931
170.5455
162.3234
162.0468
160.7782
160.6256
154.4448
153.9011
134.8053
133.8611
133.2983
132.7546
129.8073
129.1110
125.8012
125.1716
124.4849
124.4086
114.6413
114.3552
63.7830
63.5731
55.3797
48.5598
47.6441
46.1371
45.7269
21.2134
21.1276
NHCHO
O
AcO
MeO

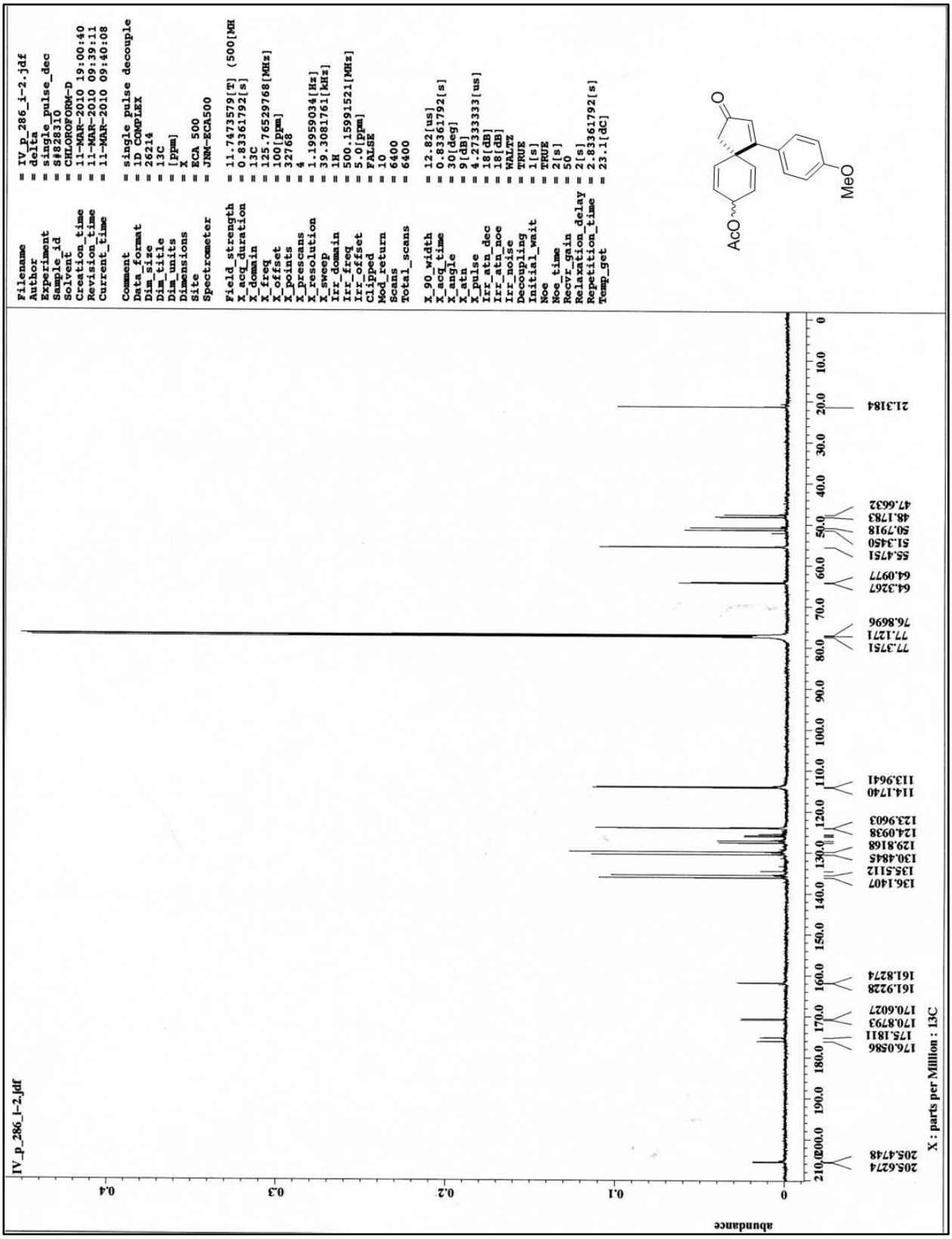
APPENDIX 128

^{1}H and ^{13}C NMR Spectra of

1-(4-Methoxy-phenyl)-3-oxo-spiro[4.5]deca-1,6,9-trien-8-yl-acetate (**276**)

Filename = IV_p_286_i-2.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = s9826647
Solvent = CHLOROFORM-D
Creation_time = 11-MAR-2010 13:57:23
Revision_time = 10-MAR-2010 23:01:50
Current_time = 11-MAR-2010 09:46:24
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECA 500
Spectrometer = JNM-ECA500
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 1.74587904[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5.0[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.57277737[Hz]
X_sweep = 9.38438438[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5.0[ppm]
Tri_domain = 1H
Tri_freq = 500.15991521[MHz]
Tri_offset = 5.0[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.8[us]
X_acq_time = 1.74587904[s]
X_angle = 45[deg]
X_atn = 2[dB]
X_pulse = 6.9[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 32
Relaxation_delay = 5[s]
Repetition_time = 6.74587904[s]
Temp_get = 22.5[dC]
IV_p_286_i-2.jdf
abundance
X : parts per Million : 1H
7.7359
7.7175
7.5297
7.5125
7.2537
6.8667
6.8495
6.5013
6.4578
5.9860
5.9803
5.9723
5.9665
3.8296
3.8205
2.6375
2.5630
2.1462
2.1141
2.75
1.93
2
4.05
6.44
7.38
1.88
1.57
0.85
0.83
0.81
0.80
AcO
MeO

Filename = IV_p_286_i-2.jdf
Author = delta
Experiment = single_pulse.ex2
Sample_id = S#826647
Solvent = CHLOROFORM-D
Creation_time = 11-MAR-2010 13:57:23
Revision_time = 10-MAR-2010 23:01:50
Current_time = 11-MAR-2010 09:47:04
Comment = single_pulse
Data_format = 1D COMPLEX
Dim_size = 13107
Dim_title = 1H
Dim_units = [ppm]
Dimensions = X
Site = ECA 500
Spectrometer = JNM-ECA500
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 1.74587904[s]
X_domain = 1H
X_freq = 500.15991521[MHz]
X_offset = 5.0[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 0.57277737[Hz]
X_sweep = 9.38438438[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5.0[ppm]
Tri_domain = 1H
Tri_freq = 500.15991521[MHz]
Tri_offset = 5.0[ppm]
Clipped = FALSE
Mod_return = 1
Scans = 12
Total_scans = 12
X_90_width = 13.8[us]
X_acq_time = 1.74587904[s]
X_angle = 45[deg]
X_atn = 2[dB]
X_pulse = 6.9[us]
Irr_mode = Off
Tri_mode = Off
Dante_presat = FALSE
Initial_wait = 1[s]
Recvr_gain = 32
Relaxation_delay = 5[s]
Repetition_time = 6.74587904[s]
Temp_get = 22.5[dC]
IV_p_286_i-2.jdf
abundance
X : parts per Million : 1H
7.7359
7.7175
7.5297
7.5125
7.2537
6.8667
6.8495
6.5013
6.4578
5.9860
5.9803
5.9723
5.9665
5.8257
5.8211
5.7638
2
1.93
4.05
0.87
0.85
7.38
1.06
0.8
AcO
MeO


Filename = IV_p_286_i-2.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#828310
Solvent = CHLOROFORM-D
Creation_time = 11-MAR-2010 19:00:40
Revision_time = 11-MAR-2010 09:39:11
Current_time = 11-MAR-2010 09:40:08
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 26214
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECA 500
Spectrometer = JNM-ECA500
Field_strength = 11.74735791[T] (500[MH
X_acq_duration = 0.83361792[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 32768
X_prescans = 4
X_resolution = 1.19959034[Hz]
X_sweep = 39.30817614[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5.0[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 12.82[us]
X_acq_time = 0.83361792[s]
X_angle = 30[deg]
X_atn = 9[dB]
X_pulse = 4.27333333[us]
Irr_atn_dec = 18[dB]
Irr_atn_noe = 18[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 2.83361792[s]
Temp_get = 23.1[dC]
AcO
MeO
IV_p_286_i-2.jdf
abundance
X : parts per Million : 13C
205.6274
205.4748
176.0586
175.1811
170.8793
170.6027
161.9228
161.8274
136.1407
135.5112
130.4845
129.8168
124.0938
123.9603
114.1740
113.9641
77.3751
77.1271
76.8696
64.3267
64.0977
55.4751
51.3450
50.7918
48.1783
47.6632
21.3184

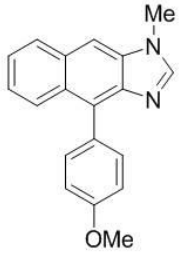
Filename = IV_p_286_i-2.jdf
Author = delta
Experiment = single_pulse_dec
Sample_id = S#828310
Solvent = CHLOROFORM-D
Creation_time = 11-MAR-2010 19:00:40
Revision_time = 11-MAR-2010 09:43:52
Current_time = 11-MAR-2010 09:44:03
Comment = single pulse decouple
Data_format = 1D COMPLEX
Dim_size = 26214
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = ECA 500
Spectrometer = JNM-ECA500
Field_strength = 11.7473579[T] (500[MH
X_acq_duration = 0.83361792[s]
X_domain = 13C
X_freq = 125.76529768[MHz]
X_offset = 100[ppm]
X_points = 32768
X_prescans = 4
X_resolution = 1.19959034[Hz]
X_sweep = 39.3081761[kHz]
Irr_domain = 1H
Irr_freq = 500.15991521[MHz]
Irr_offset = 5.0[ppm]
Clipped = FALSE
Mod_return = 10
Scans = 6400
Total_scans = 6400
X_90_width = 12.82[us]
X_acq_time = 0.83361792[s]
X_angle = 30[deg]
X_atn = 9[dB]
X_pulse = 4.273333333[us]
Irr_atn_dec = 18[dB]
Irr_atn_noe = 18[dB]
Irr_noise = WALTZ
Decoupling = TRUE
Initial_wait = 1[s]
Noe = TRUE
Noe_time = 2[s]
Recvr_gain = 50
Relaxation_delay = 2[s]
Repetition_time = 2.83361792[s]
Temp_get = 23.1[dC]
IV_p_286_I-2.jdf
(thousandths)
X : parts per Million : 13C
205.6274
205.4748
176.0586
175.1811
170.8793
170.6027
161.9228
161.8274
IV_p_286_I-2.jdf
abundance
X : parts per Million : 13C
64.3267
64.0977
55.4751
51.3450
50.7918
48.1783
47.6632
IV_p_286_I-2.jdf
abundance
X : parts per Million : 13C
136.1407
135.5112
134.5383
130.4845
129.8168
127.6707
127.1270
126.1636
125.8679
125.5246
124.0938
123.9603
114.1740
113.9641
AcO
MeO

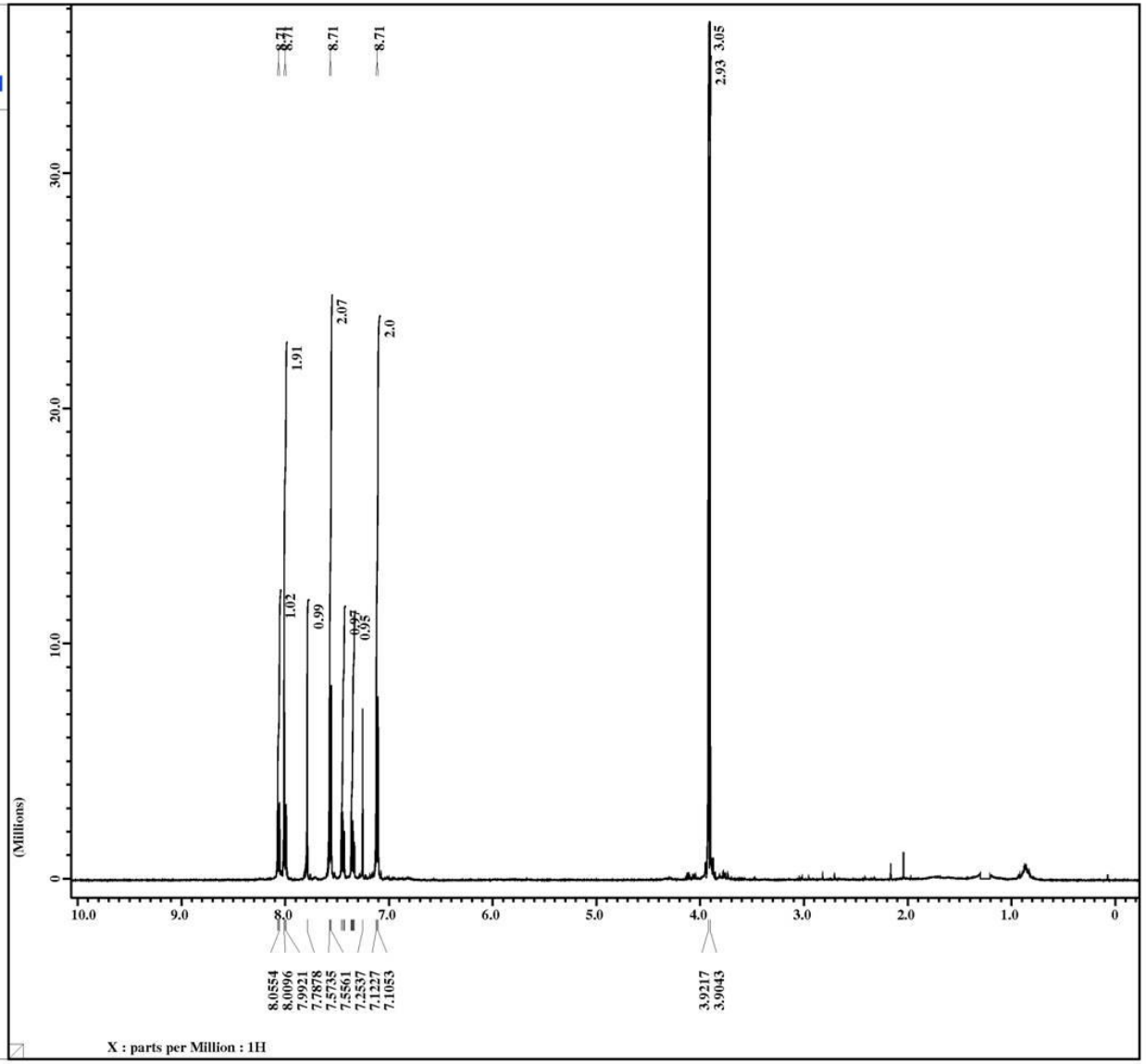
APPENDIX 129

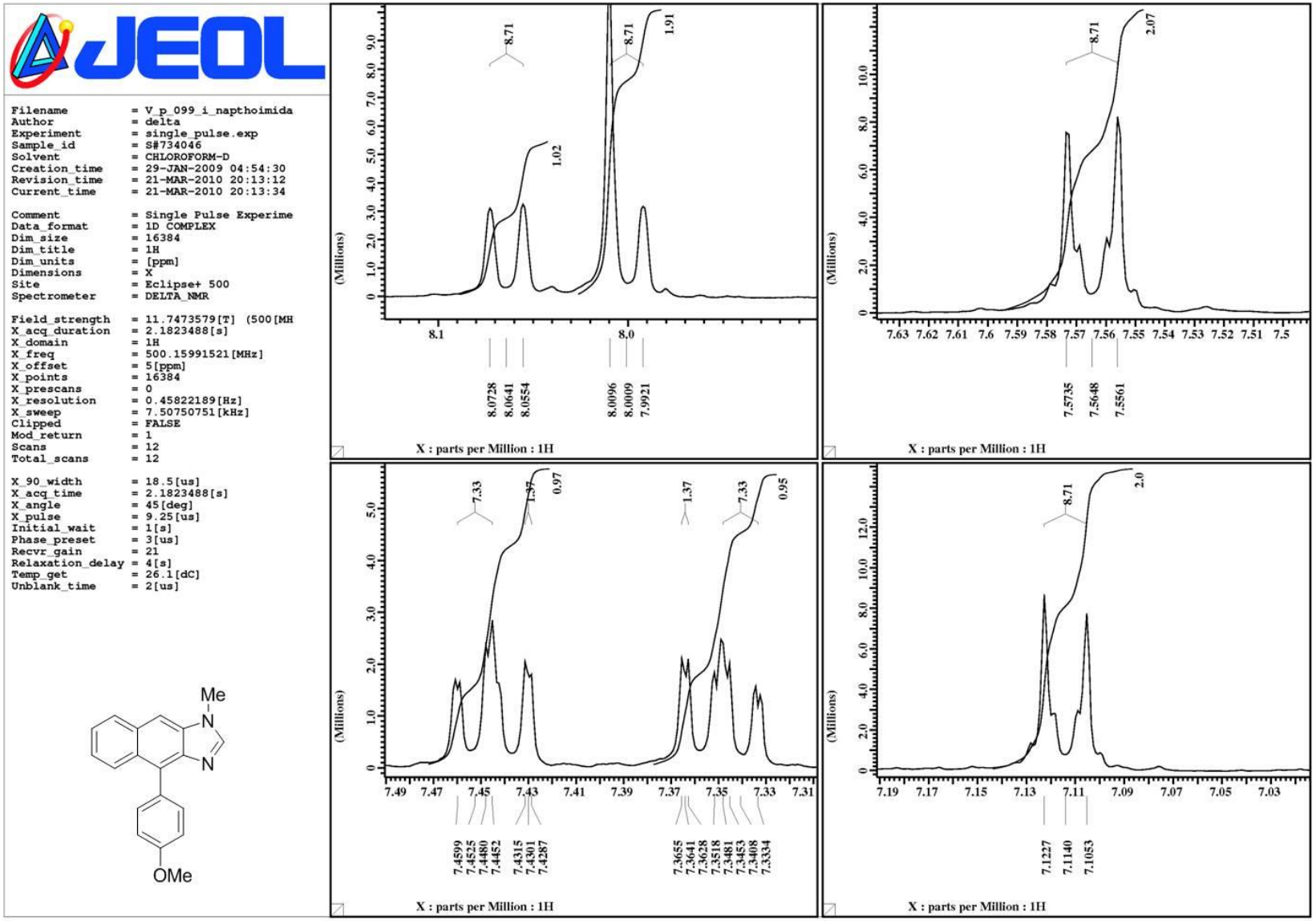
^{1}H and ^{13}C NMR Spectra of

1-Methyl-4-(4-methoxyphenyl)-1*H*-naphtho[2,3-*d*]imidazole (**278**)

```
Filename          = V_p_099_i_napthoimida
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#734046
Solvent           = CHLOROFORM-D
Creation_time     = 29-JAN-2009 04:54:30
Revision_time     = 21-MAR-2010 20:05:18
Current_time      = 21-MAR-2010 20:05:37

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 21
Relaxation_delay  = 4[s]
Temp_get          = 26.1[dC]
Unblank_time      = 2[us]
```

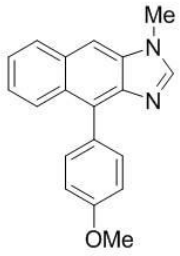


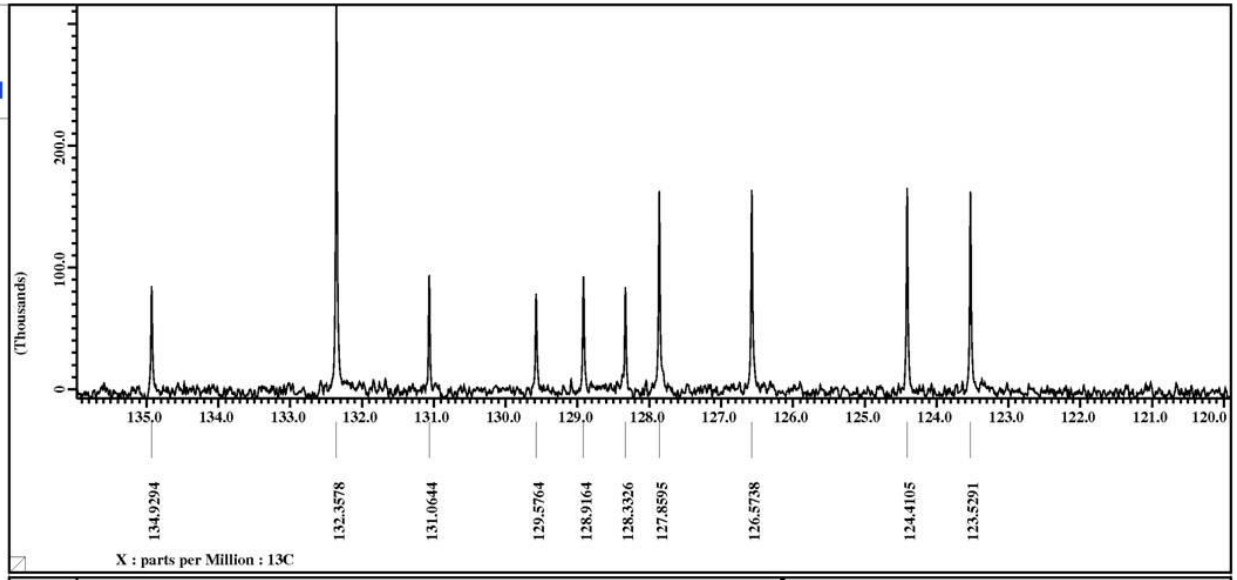


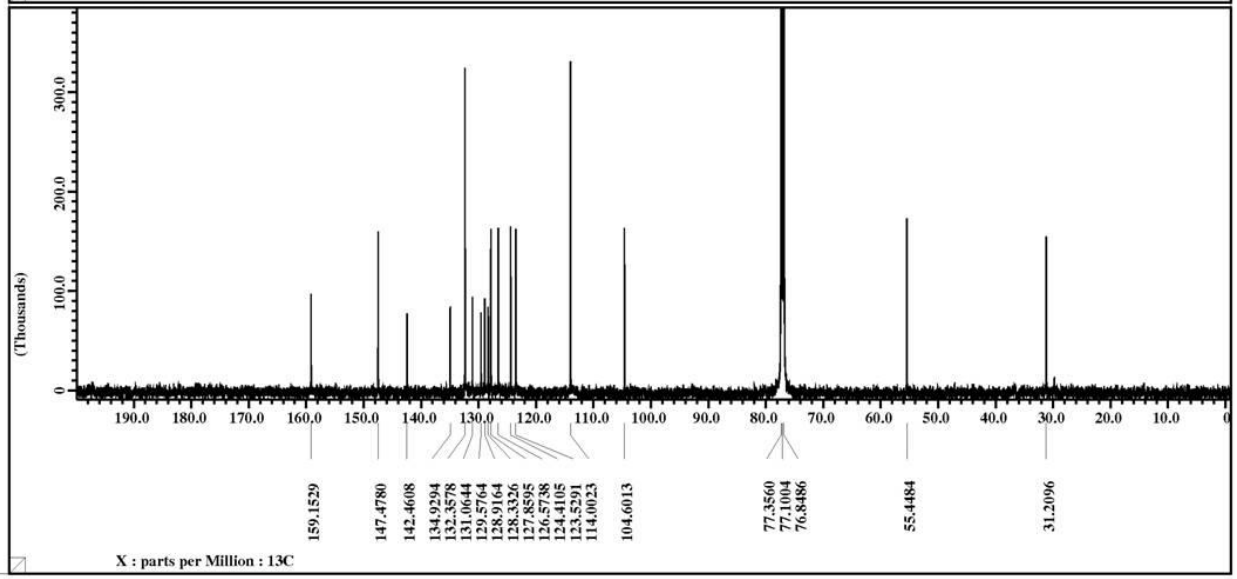


```
Filename          = V_p_099_i_napthoimida
Author            = delta
Experiment        = single_pulse.exp
Sample_id         = S#734046
Solvent           = CHLOROFORM-D
Creation_time     = 29-JAN-2009 04:54:30
Revision_time     = 21-MAR-2010 20:13:12
Current_time      = 21-MAR-2010 20:13:34

Comment           = Single Pulse Experime
Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.1823488[s]
X_domain          = 1H
X_freq            = 500.15991521[MHz]
X_offset          = 5[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 0.45822189[Hz]
X_sweep           = 7.50750751[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 12
Total_scans       = 12

X_90_width        = 18.5[us]
X_acq_time        = 2.1823488[s]
X_angle           = 45[deg]
X_pulse           = 9.25[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 21
Relaxation_delay  = 4[s]
Temp_get          = 26.1[dC]
Unblank_time      = 2[us]
```

```
Filename          = V_p_099_i_napthoimida
Author            = delta
Experiment        = single_pulse_dec
Sample_id         = S#735513
Solvent           = CHLOROFORM-D
Creation_time     = 29-JAN-2009 15:08:26
Revision_time     = 29-JAN-2009 10:11:56
Current_time      = 21-MAR-2010 20:14:57

Comment           = single pulse decouple
Data_format       = 1D COMPLEX
Dim_size          = 65536
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 500
Spectrometer      = DELTA_NMR

Field_strength    = 11.7473579[T] (500[MH
X_acq_duration    = 2.0840448[s]
X_domain          = 13C
X_freq            = 125.76529768[MHz]
X_offset          = 100[ppm]
X_points          = 65536
X_prescans        = 4
X_resolution      = 0.47983613[Hz]
X_sweep           = 31.44654088[kHz]
Irr_domain        = 1H
Irr_freq          = 500.15991521[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 1
Scans             = 7200
Total_scans       = 7200

X_90_width        = 14.2[us]
X_acq_time        = 2.0840448[s]
X_angle           = 30[deg]
X_pulse           = 4.73333333[us]
Initial_wait      = 1[s]
Noe_time          = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 30
Relaxation_delay  = 2[s]
Temp_get          = 28.6[dC]
Unblank_time      = 2[us]
```







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BIOGRAPHICAL INFORMATION

Panduka Koswatta, born in Matale, Central Province, Sri Lanka, obtained his B.S in special degree in chemistry with a first class honor in 2003, from The University of Peradeniya, Peradeniya, Sri Lanka. After working nearly two years in The University of Peradeniya, and six months in The Open University of Sri Lanka as an assistant lecturer, he moved to The United States in 2005 and began his doctoral study at the University of Texas at Arlington with Professor Carl J. Lovely. He studied oxidative chemistry of tetrahydroimidazole derivatives with Davis' aryl *N*-sulfonyloxaziridine and applied this chemistry during "the total synthesis of 2-aminoimidazole alkaloids from *Leucetta* and *Clathrina* sponges", which is the title of this work. He obtained his doctorate in 2010.