UNIVERSITY OF TEXAS, ARLINGTON

Variability and Temporal Trends of Semivolatile Organic Compounds in Biological and Environmental Media

DISSERTATION

Submitted in partial satisfaction of the requirements for the degree of

DOCTOR OF PHILOSOPHY

in concentration on Environmental Science

by

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To my wife, daughter, mother, older brother, and deceased father

TABLE OF CONTENTS

LIST OF FIGURES

Page

LIST OF TABLES

Page

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2. H Park, **K Kim***, J Yoon, KK Kim, SM Park, JS Ha. 2015. Fabrication of patterned flexible graphene devices via facile direct transfer of as-grown bi-layer graphene. *Applied Surface Science* 328 (2015): 235-240

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ABSTRACTS OF THE DISSERTATION

Variability and Temporal Trends of Semivolatile Organic Compounds
in Biological and Environmental Media

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Exposure to semivolatile organic compounds (SVOCs) in indoor environments and its potential impact on human health have been receiving increased public attention, because people in developed countries spend over 80% of their time indoors, SVOC levels are several orders of magnitude higher indoors than outdoors, and many SVOCs have various toxicities and endocrine disrupting potential. Concentrations of SVOCs in biological (e.g., human serum) and environmental (e.g., household dust, indoor air) media can help us better understand human exposure to SVOCs. For example, SVOC concentrations in biological media collected over several years may shed light on temporal trends of exposure due to the changes in consumer use or regulations. In addition, SVOC concentrations in indoor dust repeatedly collected in the same home may allow us to examine temporal variability of exposure via non-dietary dust ingestion. Lastly, SVOC concentrations in upholstered home furniture with frequent skin contact may improve our understanding of exposure via direct skin contact with furniture surfaces, particularly for infants and young children.

My dissertation studies include three sub-studies: (1) examining temporal trends and determinants of concentrations of SVOCs in blood serum samples, (2) examining temporal variability of dust concentrations and factors affecting dust concentrations for SVOCs, and (3) evaluating couch polyurethane foam (PUF) for a potential passive sampler of SVOCs.

In the first study, I utilized measured concentrations of per- and polyfluoroalkyl substances (PFAS), one class of SVOCs, in serum collected from California mothers with a young child. Then, I used multiple regression to estimate geometric means of PFAS concentrations for each sampling year (2009-2016), with adjustment for sampling year and other population characteristics that may affect PFAS concentrations in maternal serum. I observed that perfluorooctanoate (PFOA), perfluorooctane sulfonate (PFOS), and perfluorohexane sulfonate (PFHxS) decreased over the study period, consistent with results of some studies reported for other U.S. populations and other studies outside the USA. My study showed that body burden of some common long-chain PFAS decreased over the study period among California mothers with a young child.

In the second study, I utilized measured concentrations of a wide range of SVOCs in dust collected three times from the same home during a period of 22 months. To test for within-home temporal variability of SVOC concentrations in household dust, I computed intraclass correlation coefficients (ICCs), a ratio of between-home variance to total variance (within-home variance + between-home variance). Among 26 compounds that were detected in more than 50% of the samples at all three visits, 20 compounds had ICCs above 0.50 and 6 compounds had ICCs below 0.50. For 19 out of 26 compounds, correlation coefficients between spring and fall (r = 0.48-0.98) were higher than those between summer and winter (r = 0.09-0.92), implying seasonal effects on dust concentrations. My study showed that within-home temporal variability of dust

concentrations was small (ICC > 0.50) for most SVOCs, but dust concentrations may vary over time for some SVOCs with seasonal variations in source rates, such as product use.

In the third study, I utilized measured concentrations of non-flame retardant SVOCs in couch PUF at three different depths. Then, I examined concentration changes with depths and developed predictive equations for the PUF-air partition coefficient ($K_{PUF\text{-}air}$). Among 29 detected compounds, 11 compounds were detected in more than 50% of the samples at all depths. Among the 11 compounds, concentrations of phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate decreased with depth. Among the studied SVOCs, calculated log $K_{PUF\text{-}air}$ values varied from 2.46 (dimethyl phthalate) to 7.80 (homosalate), and K_{oa} (r^2 = 0.62) was a stronger predictor of $K_{PUF\text{-}air}$ than VP (r^2 = 0.47). My study showed that couch PUF can absorb many SVOCs but may not be an effective passive sampling medium for those that were less frequently detected in couch PUF and had low correlation coefficients between concentrations in dust and PUF.

CHAPTER 1: INTRODUCTION

1.1. Background

1.1.1. Semivolatile organic compounds in indoor environments

Definition and common chemical classes

Semivolatile organic compounds (SVOCs) are a group of organic compounds with a range of vapor pressure between 10^{-9} and 10 Pa (Weschler and Nazaroff 2008), lower than those of volatile organic compounds (VOCs). Analytically justified definitions of SVOCs are relatively incomplete compared to those of VOCs (Salthammer et al. 2018). SVOCs are introduced into indoor residential settings in the form of consumer products, building materials, furnishings, pesticides, and combustion by-products (Lucattini et al. 2018; Weschler and Nazaroff 2008). SVOCs are classified to various use categories or chemical classes depending on their uses, including phthalates, polybrominated diphenyl ethers (PBDEs), organophosphorus flame retardants (OPFRs), cosmetics, fragrances, ultraviolet (UV) filters, polychlorinated biphenyls (PCBs), per- and polyfluoroalkyl substances (PFASs), biocides, and polycyclic aromatic hydrocarbons (PAHs).

Sources

SVOCs have a variety of indoor sources, as they are used for multiple purposes (Figure 1.1). For example, phthalates are mainly used as plasticizers, released from products such as personal care products, solvents, adhesives, building materials, cables, toys, and food packaging (Zota et al. 2014). PBDEs were typically added as flame retardants to electronic device casing (e.g., computers, TV sets), upholstery, carpet, and polyurethane foam used as cushioning in furniture and bedding (Fromme et al. 2016). PFAS are widely used in food packaging, paints, cookware, waterproof clothing, stain-resistant fabrics, and firefighting foam (Buck et al. 2011; Lindstrom et al. 2011). PAHs have multiple indoor and outdoor sources, including cooking,

smoking, use of candles, infiltration of outdoor air, and residential heating (Cao et al. 2019; Chuang et al. 1999). Biocides such as fungicides and pesticides are transported throughout the indoor environments during or after product use (Bennett and Furtaw 2004). Cosmetic ingredients, fragrance ingredients, and UV filters are used as mixtures with various types of personal care products (e.g., lotions, sunscreen creams, fragrances).

Figure 1.1. Sources of semivolatile organic compounds (SVOCs) in indoor environments.

Distribution in indoor environments

Once SVOCs are introduced into the indoor environment from their sources, they are redistributed over time among several indoor compartments, including the gas phase, airborne particles, settled dust, human skin, and other indoor surfaces (Figure 1.2) (Little et al. 2012; Weschler and Nazaroff 2010, 2012, 2017). For example, plasticizers, flame retardants, UV

filters, and other additives are commonly found in household dust and indoor surfaces, indicating that they migrate from the products originally containing them (Melymuk et al. 2016; Sukiene et al. 2016; Venier et al. 2016; Wei et al. 2017; Wei et al. 2016a, b). The fraction of particle-phase SVOCs increases with increasing concentrations of airborne particles, leading to a decrease in the fraction of SVOCs in the gas phase (Weschler and Carslaw 2018). Among those indoor compartments, settled dust is commonly used as surrogates for human exposure to some SVOCs (Butte and Heinzow 2002). Thus, several studies investigated concentration profiles of SVOCs in indoor dust to understand residential chemical exposure (Gaspar et al. 2014; Mitro et al. 2016; Shin et al. 2020).

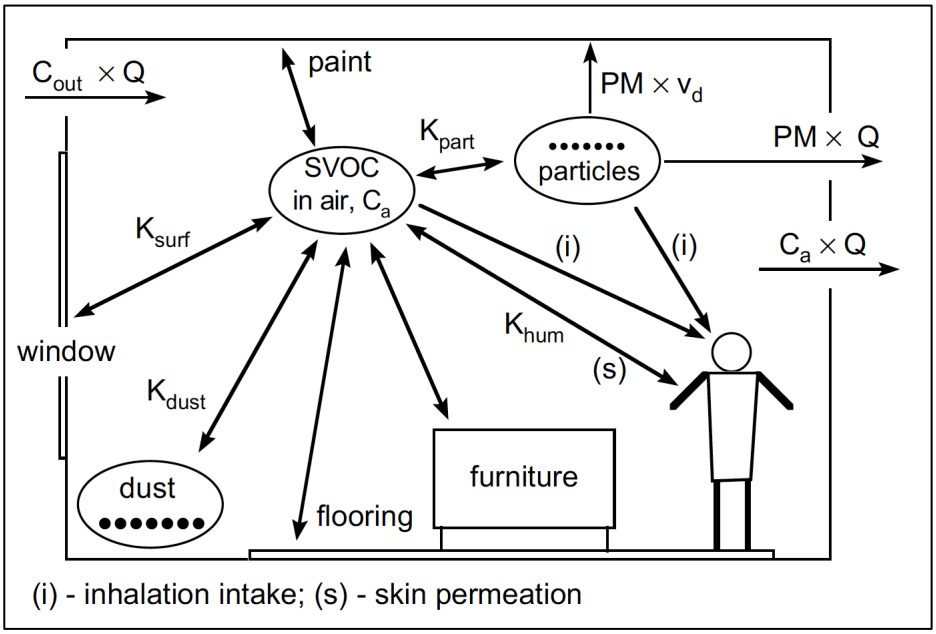

Figure 1.2. Indoor partitioning of SVOCs (adapted from Weschler and Nazaroff 2008).

Human exposure

Because SVOCs are redistributed indoors over time, human exposure to SVOCs occurs via non-dietary exposure routes including inhalation, dermal absorption, and dust ingestion (Little et al. 2012; Weschler and Nazaroff 2008). Exposure via inhalation depends on the concentration of SVOCs in both the gas-phase and airborne particles. Dermal absorption takes

place via molecular transfer from air to human skin or via direct application of skin care products on the skin (e.g., lotions, sunblock creams, fragrances). For compounds with relatively low volatility (e.g., di(2-ethylhexyl) phthalate, 2,2',4,4'-tetrabromodiphenyl ether), more than 90% of the exposure occurred via dust ingestion for young children (Gaspar et al. 2014; Little et al. 2012). Moreover, the proportion of intake from dust ingestion is larger for young children than adults (Little et al. 2012; Lorber and Egeghy 2011).

Detection in human fluids

Chemicals detected in human fluids such as urine or blood are evidence of exposure to SVOCs. The U.S. National Center for Health Statistics conducts a biannual survey research program of the National Health and Nutrition Examination Survey (NHANES) for reporting nationwide human exposure to environmental chemicals including some SVOCs, based on measurement of concentrations in blood, serum, and urine samples (Centers for Disease Control and Prevention 2019).

Toxicities and Health effects

Many SVOCs have various toxicities such as neurotoxicity (Colt et al. 2005; Colt 2006; Colt et al. 2009; Kamel and Hoppin 2004; Munoz-Quezada et al. 2013; Viel et al. 2015), carcinogenicity (Knafla et al. 2006; Shi et al. 2018), and endocrine disrupting potential (Birnbaum and Staskal 2004; Howdeshell 2002; Iwasaki et al. 2002; Jacobson and Jacobson 1996; Sharpe 2005). In addition, SVOCs have various adverse health effects. For example, some phthalates have been related to children's allergic symptoms (Bornehag et al. 2004; Oie et al. 1997), neurodevelopmental disorders (Cho et al. 2010; Kim et al. 2009), male reproductive disorder (Swan et al. 2005), and altered quality of semen (Hauser et al. 2006; Hauser et al. 2007).

Increasing levels of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in cord blood were associated with a decrease in birth weight (Apelberg et al. 2007) and higher levels of PFOS and PFOA were associated with attention deficit/hyperactivity disorder (ADHD) in children (Hoffman et al. 2010). Higher concentration of some PBDE congeners in children's serum were associated with ADHD symptoms and poorer social competence (Gascon et al. 2011) and those in breast milk was inversely associated with scores on a cognitive scale (Chao et al. 2011). Chemical structure of some SVOCs are known to be similar with those of human hormones, leading to the disruption of endocrine activities. SVOCs, recognized as endocrine disrupting compounds, include some PBDEs (Birnbaum and Staskal 2004), PCBs (Iwasaki et al. 2002; Jacobson and Jacobson 1996), phthalates (Sharpe 2005), and pesticides (Howdeshell 2002).

Regulation

Due to the increased public's concern over the toxicities and adverse health effects of SVOCs, there have been several regulatory actions in the United States, against the use, production, and emissions of such SVOCs. For example, 3M, one of the largest worldwide manufacturers of PFOS, ceased the production of PFOS and its precursors in 2002 (EPA 2017b). Emissions of PFOA from factories and product content were also reduced via a 2005/2010 PFOA Stewardship Program established by the U.S. Environmental Protection Agency (US EPA) (EPA 2017b). Moreover, the use of several phthalates, including benzyl butyl phthalate, di-n-butyl phthalate (DBP), and di(2-ethylhexyl) phthalate (DEHP), was prohibited in children's items by the Consumer Product Safety Improvement Act in the U.S. in 2008 (Zota et al. 2014). Although the production of PCBs, used as heat transfer fluids, was peaked in early 1970s, their use was restricted in the U.S. in 1978 (Weschler 2009). For PBDEs, some U.S. states banned

penta- and octa-BDEs and four out of those states also banned the sale of certain products containing deca-BDE in the U.S. in 2004 (EPA 2017a). Due to those enacted regulations, decreased temporal trends in body burdens of the regulated compounds were observed in the U.S. population (Kato et al. 2011; Zota et al. 2014).

1.1.2. Case Studies

Temporal trends and variability of SVOC concentrations in various media

SVOC concentrations in various biological and environmental media can vary over time due to several reasons. Among them, SVOC concentrations in biological media (e.g., serum, urine) changed over several years due to nationwide regulatory actions. As the U.S. federal government enacted legislation in 2008 against the use of several phthalates in child care articles, Zota et al. (Zota et al. 2014) observed that concentrations of some phthalate biomarkers (i.e., monoethyl phthalate, mono-n-butyl phthalate, monobenzyl phthalate, sum of four DEHP metabolites (ΣDEHP)) decreased between 2001-2002 and 2009-2010. In addition, serum concentrations of PFOS, PFOA, perfluorohexane sulfonate (PFHxS), and 2-(N-methyl-perfluorooctanesulfonamido) acetate (Me-FOSAA) have been observed to decline in the U.S. since another nationwide regulatory actions against the use, emissions, and production of these PFAS (Gribble et al. 2015; Hurley et al. 2018). In addition, SVOC concentrations in environmental media (e.g., air, dust) can vary over time due to indoor human activities. For example, substantial increases in SVOC air concentrations within a few hours were associated with episodic human activities (e.g., cooking and cleaning) in residential areas (Kristensen et al. 2019) and ventilation frequency affected dust concentrations of some phthalates over a year (Wei et al. 2019).

Polyurethane foam (PUF) as a sampling medium for SVOCs

Reliable sampling methods for assessing human exposure to SVOCs indoors are needed in environmental health studies (Dodson et al. 2019). Active or passive air samplers are typically used to measure SVOC concentrations in indoor air (Newton et al. 2016; Shoeib and Harner 2002c). Active air samplers are known to be accurate, but it is obtrusive, noisy, and relatively expensive because it requires pump and extra parts to draw air across a sorbent (Bohlin et al. 2008; Tuduri et al. 2012). In contrast, passive air samplers are cheap, simple to set up and operate, and relatively unobtrusive (Bohlin et al. 2008). PUF has high sorption capacity for organic compounds (Tromp et al. 2019; Zhao et al. 2004). Thus, it has been widely used as a sampling medium for SVOCs in both active (Gouin et al. 2005; Hayward et al. 2010; Moeckel et al. 2009; Newton et al. 2016) and passive (Jaward et al. 2004; Shoeib and Harner 2002c; Strandberg et al. 2018) sampling systems.

1.2. Rationale for the dissertation

Temporal trends of PFAS concentrations in human serum

Studies of PFAS exposure and its temporal changes have been mostly limited to pregnant women or adults (Table 1.1) (Bjerregaard-Olesen et al. 2016; Gribble et al. 2015; Hurley et al. 2018; Okada et al. 2013; Shu et al. 2018). Little is known for young children about temporal trends of PFAS exposure. For young children (e.g., 1 to 5 years old), their blood samples are not easy to obtain due to parents' limited consent on sampling their child's blood and child's intentional refusal of blood sampling, making it difficult to observe the temporal trends of PFAS concentrations in young children's serum. As young children likely share a large portion of PFAS exposure sources (e.g., drinking water, food, house dust) with their mothers

(Koppen et al. 2019; Song et al. 2013), studying PFAS exposure for mothers with a young child may help understand early childhood exposure to PFAS.

Table 1.1. Previous studies on temporal trends of PFAS concentrations in blood samples.

Studies	**Okada et al. 2013**	**Gribble et al. 2015**	**Hurley et al. 2017**	**Bjerregaard-Olesen et al. 2018**	**Shu et al. 2018**
Location	Japan	USA	USA	Denmark	Sweden
Target population	Pregnant women	Non-pregnant women	Non-pregnant women	Pregnant women	Pregnant women
Study period	2003-2011	2003-2013	2011-2015	2008-2013	2007-2010
Exposure media	Plasma	Serum	Serum	Serum	Serum
Main findings	PFOA (↓) PFOS (↓) PFNA (↑) PFDA (↑)	PFOA (↓) PFOS (↓) PFHxS (↓) PFNA (↓) PFDA (↓) PFUA (↓)	PFOA (↓) PFOS (↓) PFHxS (↓) MeFOSAA (↓) PFNA (mixed) PFDA (mixed)	PFOA (↓) PFOS (↓) PFHxS (↓) PFNA (↓) PFDA (↓) PFUA (↓)	PFOA (↓) PFOS (↓) PFHxS (↓) PFNA (↓) PFDA (↓) PFUA (mixed)

Abbreviation: perfluorooctanoate (**PFOA**), perfluorooctane sulfonate (**PFOS**), perfluorohexane sulfonate (**PFHxS**), perfluorononanoate (**PFNA**), perfluorodecanoate (**PFDA**), perfluoroundecanoate (**PFUA**), 2-(N-methyl-perfluorooctanesulfonamido) acetate (**Me-FOSAA**)

Temporal variability of SVOC concentrations in household dust

SVOCs are fairly persistent in indoor environments (Shin et al. 2013; Weschler and Nazaroff 2008) and their temporal variability in indoor dust concentrations is known to be smaller than that of indoor air concentrations (Egeghy et al. 2005). Egeghy et al. found that measurements of some SVOCs in dust were generally more consistent over time than those in indoor air, based on their repetitive measurements (2-3 times) over 12 months (Egeghy et al. 2005). However, different degrees of temporal variability of SVOC dust concentrations were observed between chemical classes or within the same chemical class (Table 1.2) (Bennett et al.

2015; Cao et al. 2014; Egeghy et al. 2005; Whitehead et al. 2012). In addition, studied compounds of the previous studies were limited mostly to flame retardants, PAHs, and PCBs (already banned in the U.S.). Seasonal factors (e.g., different emission strength, ventilation frequency) may also affect temporal variability in dust concentrations of some classes of SVOCs (Cao et al. 2019). Therefore, further studies are needed to investigate temporal variability of dust concentrations for a wide range of SVOCs and other determinants affecting the temporal variability of dust concentrations.

Table 1.2. Previous studies on temporal variability of SVOC indoor dust concentrations.

Studies	**Whitehead et al. 2012**	**Cao et al. 2014**	**Egeghy et al. 2005**	**Bennett et al. 2015**
Chemicals included	9 PAHs, 9 PCBs, nicotine	27 PBDEs, 10 OPFRs	1 PAH, 2 pesticides	6 PBDEs
Sample matrix	Household dust	Office dust	Household dust	Household dust
Number of homes or offices	21	3	49	24
Number of measurements	1-7 (median of 3)	16-23	2-3	2
Time period	~15 months	5-9 months	12 months	~12 months
Main findings	ICC range: 0.13-0.72	PBDEs: Relatively stable with seasons OPFRs: More sensitive with seasons	ICC range: 0.34-0.76	Spearman's correlation coefficient range: 0.34-0.70

Abbreviation: polycyclic aromatic hydrocarbons (**PAHs**), polychlorinated biphenyls (**PCBs**), polybrominated diphenyl ethers (**PBDEs**), organophosphate flame retardants (**OPFRs**), intraclass correlation coefficient (**ICC**)

Evaluating couch polyurethane foam (PUF) for a potential passive sampler of SVOCs

PUF in active or passive air samplers are used for collecting environmental pollutants in indoor air, but deployment of those samplers requires significant cost and may create burdens for residents in the homes where air samplers are deployed. Moreover, passive air samplers should be deployed for several hundreds of days to ensure chemical equilibrium between air and PUF (Shoeib and Harner 2002c). Because PUF is widely used in home furniture (e.g., pillow, beds, couch pads, mat) as a padding material, there is evidence that PUF in the home furniture can absorb other SVOCs commonly detected indoors. For example, PUF in an infant crib mattress is not known as a source of plasticizers, but among the ten used infant crib mattresses that were in contact with mattress covers with detectable plasticizers, at least one plasticizer was detected in nine mattress PUF samples (Boor et al. 2015). However, a holding capacity of upholstered home furniture PUF is mostly unknown because SVOC partitioning between PUF and air were determined in chamber studies under the controlled laboratory conditions or outdoor field studies using purposefully-designed PUF samplers (Abdollahi et al. 2017; Bidleman et al. 2016; Francisco et al. 2017; Kamprad and Goss 2007; Parnis et al. 2016; Saini et al. 2019; Tromp et al. 2019). Thus, SVOC partitioning with direct concentration measurements in upholstered home furniture PUF may improve our understanding of home furniture PUF's actual holding capacity for indoor SVOCs.

1.3. Research objectives and hypothesis

The main objectives of this dissertation were (1) to examine temporal changes of SVOCs concentration in biological and environmental media and (2) to evaluate couch PUF for a potential passive sampler of SVOCs. Specific research objectives and their corresponding hypotheses are outlined below:

1) Assess temporal trends of PFAS maternal serum concentrations. Identify determinants of PFAS maternal serum concentrations among maternal characteristics.

a) H_0: PFAS serum concentrations did not change over time.

b) H_1: PFAS serum concentrations changed over time.

2) Investigate within-home temporal variability of dust concentrations for a wide range of SVOCs. Examine whether sampling seasons or sampling visits affected dust concentrations.

a) H_0: SVOC dust concentrations did not change with seasons or visits.

b) H_1: SVOC dust concentrations changed with seasons or visits.

3) Evaluate couch PUF for a potential passive sampler of indoor SVOCs. Calculate $K_{PUF\text{-}air}$ using direct measurements of SVOC concentrations in PUF and dust.

a) H_0: SVOC concentrations in couch PUF did not change with depths.

b) H_1: SVOC concentrations in couch PUF changed with depths.

1.4. Overview of dissertation

The first phase of my research is to investigate temporal trends and variability of SVOC concentrations in biological (i.e., maternal serum) and environmental media (i.e., household dust). The second phase is to evaluate couch PUF for a potential passive sampler of SVOCs.

In Chapter 2, I examined temporal trends of PFAS maternal serum concentrations over 8 years (2008-2016). I also identified determinants affecting PFAS maternal serum concentrations among several population characteristics. I used a multiple regression model with sampling year as a main predictor, adjusted for covariates (e.g., breastfeeding duration, maternal age, parity).

Lastly, I compared PFAS maternal serum levels in our study population with those in the general female population in the U.S. National Health and Nutrition Examination Survey (NHANES).

In Chapter 3, I examined temporal variability of dust concentrations for a wide range of SVOCs over 22 months by calculating intraclass correlation coefficients. I also examined the effect of sampling seasons on SVOC dust levels. In addition, I examined if there were significant differences in SVOC dust concentrations between subgroups of two household characteristics (i.e., household income and presence of children).

In Chapter 4, I evaluated couch PUF for a potential passive sampler of SVOCs. I examined changes in SVOC concentrations in couch PUF with depths and calculated PUF-air partition coefficients ($K_{PUF\text{-}air}$) using a partitioning relationship and measured concentrations in PUF and dust. In addition, I compared my calculated $K_{PUF\text{-}air}$ values with those measured or estimated in previous studies. I developed predictive equations for $K_{PUF\text{-}air}$ with readily available chemical properties (i.e., octanol-air partition coefficient (K_{oa}), vapor pressure). Lastly, I calculated the holding capacity of couch PUF for studied SVOCs.

In Chapter 5, I summarized the main results, conclusions, implications, and recommendations from Chapter 2 through Chapter 4.

CHAPTER 2: TEMPORAL TRENDS AND DETERMINANTS OF SERUM CONCENTRATIONS OF PER- AND POLYFLUOROALKYL SUBSTANCES AMONG NORTHERN CALIFORNIA MOTHERS WITH A YOUNG CHILD, 2009-2016

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2.1. Introduction

2.1.1. Background

Per- and polyfluoroalkyl substances (PFAS) are a group of fluorinated compounds, widely used for industrial and commercial applications including food packaging, paints, cookware, waterproof clothing, stain-resistant fabrics and firefighting foams (Buck et al. 2011; Lindstrom et al. 2011). PFAS migrate from food-contact paper or food packaging into foods (Begley et al. 2005; Begley et al. 2008) and are likely to be transferred from consumer products to indoor air and household dust (Eriksson and Karrman 2015; Winkens et al. 2017; Yao et al. 2018). Thus, PFAS have been also detected in most of the serum samples of the general population worldwide (Eriksson et al. 2017; Haug et al. 2009; Hurley et al. 2018; Jain 2018; Kato et al. 2011; Nost et al. 2014). Based on laboratory animal toxicity testing, some of the common long-chain PFAS including perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) are known to have neurotoxicity (Austin et al. 2003; Harada et al. 2005), reproductive toxicity (Butenhoff et al. 2004; Vandenheuvel et al. 1992), hepatotoxicity (Kudo et al. 1999; Liu et al. 1996; Seacat et al. 2002; Vandenheuvel et al. 1992), and developmental toxicity (Lau et al. 2003; Lau et al. 2004; Lau et al. 2006; Thibodeaux et al. 2003).

Increasing public health concern over toxicity from PFAS exposure in the early 2000s led to regulatory and voluntary efforts in restricting the production, use, and emissions of the common long-chain PFAS. One of the largest manufacturers of PFOS worldwide, 3M, voluntarily ceased the production of PFOS and its precursors in 2002 (EPA 2017b). The European Union (EU) restricted use of PFOS in finished and semi-finished products in 2006 (EU 2007). The U.S. Environmental Protection Agency (EPA) established a 2005/2010 PFOA Stewardship Program for reducing emissions from factories and product content of PFOA and its

precursors (EPA 2017b). Consequently, the eight largest companies in the PFOA industry achieved complete elimination of PFOA from their production in 2015. In addition to PFOS and PFOA, national and/or regional regulations were enacted in several countries against the use of other long-chain PFAS in consumer products (Wang et al. 2017), including perfluorohexane sulfonate (PFHxS), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUA), and 2-(N-methyl-perfluorooctanesulfonamido) acetate (Me-FOSAA). As a result of the efforts for reducing exposures to some long-chain PFAS and their precursors since the early 2000s, serum concentrations of PFOS, PFOA, PFHxS, and Me-FOSAA (a derivative of PFOS) declined whereas those of PFNA, PFDA, and PFUA showed mixed time trends in the United States and some European countries (Bjerregaard-Olesen et al. 2016; Gribble et al. 2015; Hurley et al. 2018; Okada et al. 2013; Shu et al. 2018).

2.1.2. Statement of research problem

Children's early life exposure to PFAS is of public health concern. However, studies of PFAS exposure and its temporal changes have been limited to date mostly to adults and pregnant women. For young children (e.g., 1 to 5 years old), their blood samples are not easy to obtain because of parents' limited consent on sampling their child's blood and child's intentional refusal of blood sampling. As young children likely share a large portion of PFAS exposure sources (e.g., drinking water, food, house dust) with their mothers (Koppen et al. 2019; Song et al. 2013), studying PFAS exposure for mothers with a young child may help understand early childhood exposure to PFAS.

2.1.3. Objectives

In this study, we used 450 serum samples collected from 450 mothers when the child was 2 to 5 years old in a case-control study to (1) assess temporal trends of PFAS maternal serum concentrations and (2) identify determinants of PFAS maternal serum concentrations.

2.2. Methods

2.2.1. Study population and sample collection

This study included mothers participating in CHARGE (***CH***ildhood ***A***utism ***R***isk from ***G***enetics and ***E***nvironment), a population-based case-control study designed to identify causes and contributing factors for autism (Hertz-Picciotto et al. 2006). Children with autism were primarily recruited through the California Department of Developmental Services, as well as from other studies and various clinics, by self- or provider referrals. The general population group (controls) were identified from state birth files and were frequency-matched to the age, sex, and catchment area distribution of the autism cases. Details of study design, recruitment, eligibility for cases and controls, sample size, exposure data, and developmental diagnosis are available elsewhere (Hertz-Picciotto et al. 2006).

CHARGE started collecting serum aliquots in 2009. For the current study, we selected 450 mothers with sufficient volume of available blood serum for quantification of PFAS at the time of enrollment. Among the mothers who enrolled since 2009, approximately 36% of them were not included in the current study because they provided insufficient volume of available serum or had a child who had no diagnosis or incomplete diagnosis, was pending for diagnosis, or received a final diagnosis other than autism spectrum disorder or typical development by February 2017. We included samples collected between 2009 and 2016 in the current study. Two mothers who provided samples in January 2017 were also included in a batch of 2016. The mean

age of the child at the time of blood collection from mothers was just under four years (average ± Std. Dev.: 46.5 ± 9.5 months); the youngest and oldest children were 25 months and 61 months old, respectively. Serum was separated from the blood and placed in a -80°C freezer within 24 hours until analysis.

2.2.2. Quantification of PFAS in serum

We shipped 0.5 mL serum aliquots to the Centers for Disease Control and Prevention (CDC) for analysis. At CDC, we quantified serum concentrations of nine PFAS using solid-phase extraction linked with reversed-phase high-performance liquid chromatography-isotope dilution tandem mass spectrometry (Kato et al. 2011). We applied strict quality control/quality assurance protocols to the analytical measurements. In addition to study samples, we analyzed 38 duplicates for quality assurance. Depending on the analyte and concentration, median relative standard deviations for 19 pairs of blind duplicates ranged from 0 to 6%, except for PFHxS (17%) and PFDA (20%).

The nine PFAS quantified in this study were: PFOA, PFOS, PFHxS, PFNA, PFDA, PFUA, perfluorododecanoate (PFDoDA), Me-FOSAA, and 2-(N-ethyl-perfluorooctane sulfonamido) acetate (Et-FOSAA). For all PFAS, the limit of detection (LOD) was 0.1 ng/mL (nanograms per milliliter = ppb).

2.2.3. Statistical analysis

We conducted all statistical analyses using STATA/IC 15.1 (StataCorp LLC, College Station, TX, USA). For concentrations below the LOD, we assigned a value of LOD divided by the square root of 2 (Hornung 1990). We used ln-transformed PFAS serum concentrations in the

regression to account for skewed distributions. To test for significant changes in PFAS serum concentrations over time, we used multiple regression with adjustment for maternal race/ethnicity (white, Hispanic, others), maternal age (in years), breastfeeding duration (in months), maternal education (less than college, bachelor, graduate or professional), pre-pregnancy body mass index (BMI) (in kg/m^2), homeownership (yes, no), and parity (1-7). After checking crude temporal trends from plots of the sampling time (years) versus yearly average PFAS serum concentrations, we centered our sampling years (2009-2016) at 2013 to robustly account for nonlinear time trends of PFAS serum concentrations. For PFOA, PFOS, and PFHxS, we included only a linear term (year) in the regression models. For PFNA and PFDA, we included both linear and quadratic terms (year and year^2) in the regression models. The regression models used in this study were as follows:

For PFOA, PFOS, and PFHxS,

$$\ln[C] = (\beta_1 \cdot year) + (\beta_2 \cdot race) + (\beta_3 \cdot age) + (\beta_4 \cdot BF) + (\beta_5 \cdot Edu) + (\beta_6 \cdot BMI) + (\beta_7 \cdot Home) + (\beta_8 \cdot Parity) + \beta_0 \quad [1]$$

For PFNA and PFDA,

$$\ln[C] = (\beta_1 \cdot year) + (\beta_2 \cdot year^2) + (\beta_3 \cdot race) + (\beta_4 \cdot age) + (\beta_5 \cdot BF) + (\beta_6 \cdot Edu) + (\beta_7 \cdot BMI) + (\beta_8 \cdot Home) + (\beta_9 \cdot Parity) + \beta_0 \quad [2]$$

where C is the PFAS serum concentration in ng/mL, β_i are the regression coefficients (i = 0 to 8 for Equation 1, i = 0 to 9 for Equation 2), *year* is sampling year, *race* is maternal race/ethnicity, *age* is maternal age, *BF* is breastfeeding duration, *Edu* is education, *BMI* is pre-pregnancy body mass index, *Home* is homeownership, and *Parity* is number of completed pregnancies beyond 20 weeks of gestation. We obtained the regression coefficients from the above regression models

and computed yearly-specific fractions for categorical covariates (*race, Home,* and *Edu*) and averages for continuous covariates (*age, BF*, *BMI*, and *Parity*) from our samples.

To estimate the least square mean (LSM) which is the mean of ln[*C*] for each sampling year, we used the estimated regression coefficients and the computed yearly-specific fractions and averages of the selected covariates. We also computed the least square geometric mean (LSGM) of PFAS serum concentrations for each sampling year, as exp(LSM) with 95% confidence intervals (CIs) as exp(LSM ± 1.97·SE_{LSM}), where SE_{LSM} is the standard error of the LSM. To construct CIs, we used a critical value 1.97 from the t-distribution, based on the degree of freedom given the numbers of both the study participants and the selected covariates. To examine the relative concentration changes over our study period, we computed average annual percent changes of PFAS serum concentrations using an equation [exp(aRGR) – 1] × 100% with 95% CIs as [exp(aRGR ± 1.97·SE_{β}) – 1], where aRGR is the average relative growth rate and SE_{β} is the standard error of time-related regression coefficients. For PFOA, PFOS, and PFHxS with β_1 only in the models, β_1 is equal to aRGR. For PFNA and PFDA with both β_1 and β_2 in the models, we estimated aRGR using a formula: aRGR = β_1 - β_2. We obtained this formula by partially differentiating Equation 2 with respect to a variable '*year*' to get the relative growth rate (RGR = $\beta_1 + 2\beta_2 \cdot$year). Then, we averaged RGR over the centered-year interval [-4, 3], finally resulting in the formula aRGR = β_1 - β_2 (Hurley et al. 2018). For PFUA, PFDoDA, Me-FOSAA, and Et-FOSAA, which were detected less than 50% in our serum samples, we did not perform the regression analyses.

We also used multiple regression to examine the association between maternal PFAS serum concentrations and the covariates adjusted in the above regression. To examine whether maternal PFAS serum concentrations varied by different subgroups of categorical covariates

(i.e., maternal race/ethnicity, education, and homeownership), we conducted stratified analyses. To investigate the effects of continuous covariates (i.e., maternal age, breastfeeding duration, pre-pregnancy BMI, and parity) on maternal PFAS serum concentrations, we estimated percent changes of PFAS maternal serum concentrations per one unit increase using the regression coefficients (β) of each covariate obtained from Equation 1 or 2. Then, by assigning the regression coefficients to the equation $[\exp(\beta) - 1] \times 100\%$, we computed the percent changes for each PFAS per one unit increase of each covariate.

To compare PFAS serum concentrations between CHARGE mothers and the general female population in the U.S. National Health and Nutrition Examination Survey (NHANES) (Centers for Disease Control and Prevention 2019), we computed geometric means (GMs) and 95% CIs of PFAS serum concentrations in CHARGE and NHANES by restricting our comparison to the same age range (i.e., 16-47 years of age in the two populations). Because NHANES documents biannual biomonitoring data, we grouped the 8-yearly data (2009-2016) every two years, resulting in the four cycles (i.e., 2009-2010, 2011-2012, 2013-2014, 2015-2016), which are matched with NHANES survey cycles. Five PFAS detected in more than 50% of the samples (i.e., PFOA, PFOS, PFHxS, PFNA, PFDA) were included in the comparison, while the remaining four PFAS (i.e., PFUA, PFDoDA, Me-FOSAA, Et-FOSAA) were not included because of their relatively low detection frequencies (<50%) in the CHARGE subjects (Table 2.2).

2.3. Results

2.3.1. Characteristics of CHARGE mothers

The average age of the mothers at the time of blood serum collection was 30.3 years old, ranging from 16 to 47 years old. The average breastfeeding duration between the delivery of the participating child (or index child) and blood collection was 8.0 months. Among 450 mothers, 128 mothers delivered one younger sibling of the index child and 11 mothers delivered two younger siblings of the index child. The mothers included in the study were 59.3% white, 20.2% Hispanic, and 20.4% others (28% black, 48% Asian, and 24% multiracial). Summary statistics of other maternal characteristics are available in Table 2.1.

Table 2.1. Characteristics of 450 CHARGE mothers included in the current study.

	n	**%**
Race/ethnicity		
white	267	59.3
Hispanic	91	20.2
others [a]	92	20.4
Education		
Less than college	239	53.1
Bachelor	149	33.1
Graduate or professional	62	13.8
Homeownership		
Yes	274	60.9
No	159	35.3
Missing	17	3.8
	Mean	**SD**
Age (year)	30.3	5.8
Breastfeeding duration (month)	8.0	8.4
Pre-pregnancy BMI (kg/m^2)	26.3	6.3
Parity	1.8	1.0

[a] Includes 28% black, 48% Asian, and 24% multiracial

Abbreviation: body mass index (**BMI**), standard deviation (**SD**)

2.3.2. PFAS maternal serum concentrations

Four PFAS were detected in more than 90% of the samples: PFOA (100%), PFOS (100%), PFHxS (98%), and PFNA (95%) (Table 2.2). PFDA, PFUA, and Me-FOSAA were detected in 68%, 35%, and 46% of the samples, respectively. The remaining two PFAS were detected in less than 5% of the samples: PFDoDA (3%) and Et-FOSAA (1%). The highest GM was observed in PFOS (3.29 ng/mL), followed by PFOA, PFNA, PFHxS, and PFDA (1.10, 0.49, 0.46, and 0.16 ng/mL, respectively). For PFOA, PFOS, PFHxS, and PFNA, the GMs of CHARGE mothers were lower than those of the general female population reported in NHANES during most of survey cycles (Figure 2.1).

Table 2.2. Distributions of PFAS maternal serum concentrations (ng/mL) collected from 450 CHARGE mothers.

	% detect [a]	Minimum	Median	GM	Maximum	Standard Deviation
PFOA	100	0.22	1.07	1.10	8.07	0.98
PFOS	100	0.37	3.20	3.29	23.80	3.05
PFHxS	98	<LOD	0.50	0.46	4.70	0.49
PFNA	95	<LOD	0.50	0.49	3.40	0.42
PFDA	68	<LOD	0.20	0.16	2.10	0.20
PFUA	35	<LOD	<LOD	*	1.70	*
PFDoDA	3	<LOD	<LOD	*	0.60	*
Me-FOSAA	46	<LOD	<LOD	*	2.40	*
Et-FOSAA	1	<LOD	<LOD	*	0.30	*

[a] Detection frequency was calculated based on 450 mothers recruited by CHARGE during 2009-2016. Limit of detection was 0.1 ng/mL for all compounds.
* Not calculated: The proportion of results above the limit of detection was too low to provide a valid result.

Abbreviation: geometric mean (**GM**), perfluorooctanoate (**PFOA**), perfluorooctane sulfonate (**PFOS**), perfluorohexane sulfonate (**PFHxS**), perfluorononanoate (**PFNA**), perfluorodecanoate (**PFDA**), perfluoroundecanoate (**PFUA**), perfluorododecanoate (**PFDoDA**), 2-(N-methyl-perfluorooctanesulfonamido) acetate (**Me-FOSAA**), 2-(N-ethyl-perfluorooctanesulfonamido) acetate (**Et-FOSAA**)

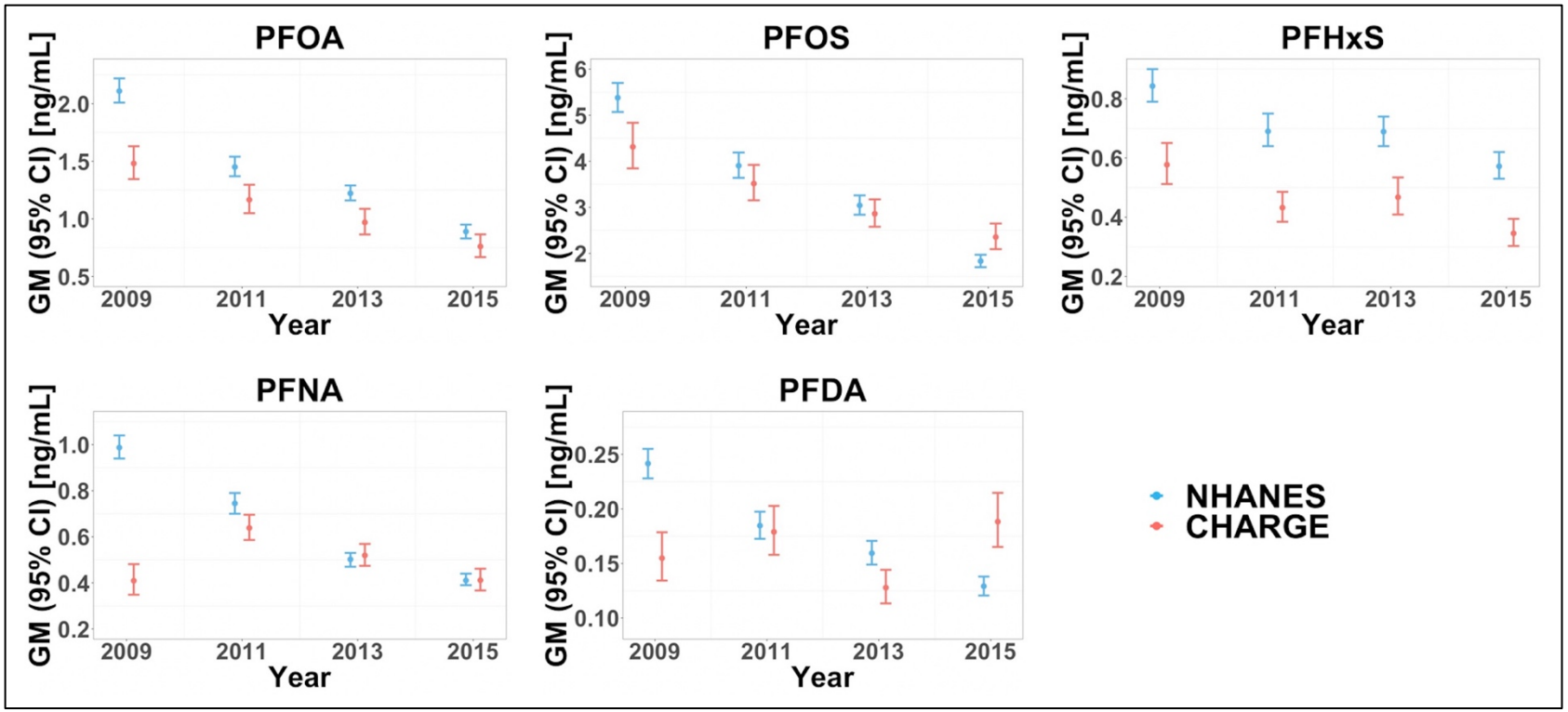

Figure 2.1. Geometric means (GMs) and 95% confidence interval (CI) of PFAS serum concentrations from CHARGE (450 mothers living in Northern California) and NHANES (U.S. female population of 16 to 47 years of age) during 4 sampling cycles (2009-2010, 2011-2012, 2013-2014, and 2015-2016).

2.3.3. Temporal trends of PFAS maternal serum concentrations

After adjusting for the selected covariates, the LSGMs of PFOA, PFOS, and PFHxS linearly decreased during the study period [percent change (95% confidence interval): -10.7% (-12.7%, -8.7%); -10.8% (-12.9%, -8.5%); -8.0% (-10.5%, -5.5%), respectively] (Figure 2.2). The LSGMs of PFNA increased in early study years and decreased in later study years. On the other hand, the LSGM of PFDA decreased in early study years and increased in later study years (Table S2.1).

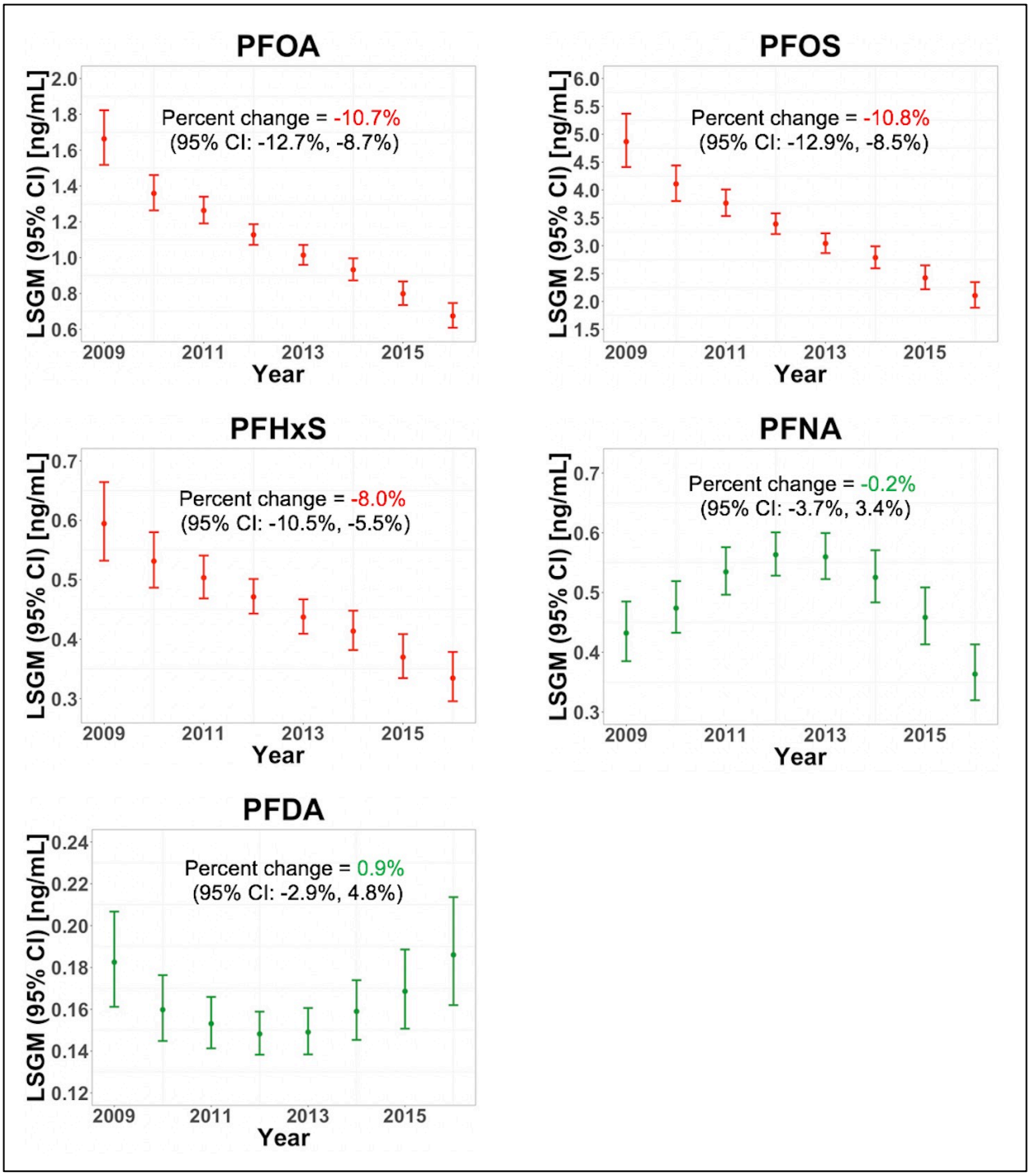

Figure 2.2. Least square geometric means (LSGMs) of PFAS maternal serum concentrations during our study period and annual percent change (95% confidence interval). The LSGMs in this figure were estimated from our regression models after adjusting for selected covariates.

2.3.4. PFAS maternal serum concentrations by maternal characteristics

When analyses were stratified by race/ethnicity, we found that concentration patterns were different among different race/ethnicity groups. For example, although three PFAS showed linearly decreased trends among all mothers (i.e., PFOA, PFOS, PFHxS), LSGMs did not change or relatively slowly decreased over our study period among mothers who are neither white nor Hispanic, compared to white or Hispanic mothers (Figure S2.2, Table 2.3). When stratified by maternal education, we found that LSGMs of PFOS, PFHxS, and PFDA were the highest and declined relatively rapidly among mothers who had graduate or professional degrees (Figure S2.3), compared to mothers with less education (Table 2.3). For PFDA, LSGMs varied little in the 'less than college' subgroup. When stratified by homeownership, we observed that mothers who owned a home tended to have higher LSGMs of PFOA, PFOS, and PFDA than those who did not own a home (Results were not shown because of similar patterns with Figure S2.2). For PFDA, LSGMs did not significantly vary among mothers who did not own a home during our study period (Table 2.3).

For PFOA and PFDA, LSGMs increased with mother's age [percent change (95% CIs) per one year of age: 1.2% (0.1%, 2.3%); 1.7% (0.3%, 3.2%), respectively] (Table 2.4). LSGMs of PFOA, PFOS, PFHxS, PFNA, and PFDA decreased with breastfeeding duration [percent change (95% CIs) per one month of breastfeeding: -2.8% (-3.5%, -2.2%); -2.0% (-2.6%, -1.3%); -1.8% (-2.6%, -1.1%); -1.5% (-2.3%, -0.7%); -1.3% (-2.1%, -0.5%), respectively]. We also found that LSGMs of PFNA and PFDA decreased with pre-pregnancy BMI [percent change (95% CIs) per one unit increase in the pre-pregnancy BMI (in kg/m^2): -1.5% (-2.5%, -0.4%); -1.9% (-3.0%, -0.8%), respectively]. In addition, LSGMs of PFOS and PFHxS decreased with increasing parity [percent change (95% CIs) per each additional pregnancy: -8.6% (-14.1%, -2.8%); -14.5% (-20.2%, -8.3%), respectively].

Table 2.3. Average annual percent change in serum concentrations and 95% confidence interval among subgroups of each categorical covariate.

	Subgroups	Average annual percent change with 95% confidence interval [a,b]				
		PFOA	PFOS	PFHxS	PFNA	PFDA
Race/ ethnicity	white	-11.4* (-14.1, -8.7)	-10.5* (-13.4, -7.5)	-8.6* (-11.9, -5.2)	1.2 (-3.5, 6.1)	2.0 (-2.8, 7.1)
	Hispanic	-11.9 (-16.6, -6.9)	-14.2* (-18.8, -9.3)	-9.8* (-14.9, -4.3)	-6.5 (-14.5, 2.1)	-1.7 (-10.5, 8.0)
	Others [c]	-7.7* (-13.0, -2.2)	-9.3* (-14.6, -3.7)	-3.3 (-10.0, 4.0)	2.4 (-6.1, 11.5)	0.6 (-8.5, 10.5)
Education	Less than college	-12.4* (-15.4, -9.4)	-11.4* (-14.5, -8.2)	-7.7* (-11.3, -4.0)	-3.2 (-8.4, 2.2)	0.7 (-4.8, 6.5)
	Bachelor	-9.2* (-12.9, -5.4)	-9.9* (-13.6, -5.9)	-6.3* (-10.6, -1.7)	2.4 (-3.2, 8.2)	2.0 (-4.6, 9.0)
	Graduate or professional	-10.8* (-15.9, -5.3)	-12.9* (-18.8, -6.7)	-12.8* (-19.7, -5.2)	1.9 (-8.6, 13.6)	-2.7 (-11.5, 7.0)
Home-ownership	No	-9.2* (-12.9, -5.3)	-10.1* (-13.7, -6.4)	-4.3 (-9.0, 0.7)	-3.7 (-9.7, 2.5)	1.6 (-5.1, 8.7)
	Yes	-11.6* (-14.1, -9.1)	-11.3* (-14.0, -8.4)	-9.8* (-12.7, -6.8)	1.3 (-3.2, 6.0)	0.1 (-4.5, 4.9)

[a] For PFOA, PFOS, and PFHxS, we provided significance (*) of p-value of the null hypothesis that the regression coefficient (β) of 'year' equals to zero.

[b] For PFNA and PFDA, we provided significance (*) of p-value of the null hypothesis that the regression coefficient (β) of 'year^2' equals to zero.

[c] Includes 28% black, 48% Asian, and 28% multiracial.

* p-value < 0.05

Table 2.4. Percent changes of PFAS maternal serum concentrations per one-unit increase of each covariate.

Covariate	Compound	β [a]	SE_β [b]	*p*-value [c]	% change per month	95% confidence interval
Maternal age (in year)	PFOA	0.012	0.006	0.04	1.2	(0.1%, 2.3%)
	PFOS	0.010	0.006	0.09	1.0	(-0.2%, 2.2%)
	PFHxS	0.006	0.007	0.41	0.6	(-0.8%, 1.9%)
	PFNA	0.011	0.007	0.11	1.1	(-0.2%, 2.5%)
	PFDA	0.017	0.007	0.02	1.7	(0.3%, 3.2%)
	PFUA	0.014	0.006	0.02	1.4	(0.2%, 2.5%)
	Me-FOSAA	0.011	0.008	0.16	1.1	(-0.4%, 2.6%)
Breastfeeding duration (in month)	PFOA	-0.029	0.003	< 0.01	-2.8	(-3.5%, -2.2%)
	PFOS	-0.020	0.003	< 0.01	-2.0	(-2.6%, -1.3%)
	PFHxS	-0.018	0.004	< 0.01	-1.8	(-2.6%, -1.1%)
	PFNA	-0.015	0.004	< 0.01	-1.5	(-2.3%, -0.7%)
	PFDA	-0.013	0.004	< 0.01	-1.3	(-2.1%, -0.5%)
	PFUA	-0.004	0.003	0.19	-0.4	(-1.1%, 0.2%)
	Me-FOSAA	-0.001	0.004	0.78	-0.1	(-1.0%, 0.8%)
Pre-pregnancy BMI (in kg/m^2)	PFOA	-0.005	0.004	0.26	-0.5	(-1.3%, 0.4%)
	PFOS	-0.006	0.005	0.17	-0.6	(-1.5%, 0.3%)
	PFHxS	-0.001	0.005	0.88	-0.1	(-1.1%, 0.9%)
	PFNA	-0.015	0.005	< 0.01	-1.5	(-2.5%, -0.4%)
	PFDA	-0.019	0.006	< 0.01	-1.9	(-3.0%, -0.8%)
	PFUA	-0.017	0.004	< 0.01	-1.7	(-2.5%, -0.8%)
	Me-FOSAA	-0.007	0.006	0.26	-0.7	(-1.8%, 0.5%)
Parity	PFOA	-0.026	0.030	0.39	-2.5	(-8.1%, 3.4%)
	PFOS	-0.090	0.032	< 0.01	-9.0	(-14.1%, -2.8%)
	PFHxS	-0.156	0.036	< 0.01	-15.6	(-20.2%, -8.3%)
	PFNA	-0.028	0.037	0.45	-2.8	(-9.6%, 4.6%)
	PFDA	-0.007	0.039	0.86	-0.7	(-8.0%, 7.2%)
	PFUA	-0.025	0.031	0.41	-2.5	(-8.2%, 3.6%)
	Me-FOSAA	-0.030	0.041	0.47	-2.9	(-10.5%, 5.3%)

[a] β represents the regression coefficient of each covariate in the regression models.

[b] SE_β represents the standard error of the regression coefficient (β).

[c] p-value of the null hypothesis that the regression coefficient (β) of each covariate equals to zero.

Abbreviation: perfluorooctanoate (**PFOA**), perfluorooctane sulfonate (**PFOS**), perfluorohexane sulfonate (**PFHxS**), perfluorononanoate (**PFNA**), perfluorodecanoate (**PFDA**), perfluoroundecanoate (**PFUA**), and 2-(N-methyl-perfluorooctanesulfonamido) acetate (**Me-FOSAA**)

2.4. Discussion

In this study, we used samples from mothers with a young child to examine temporal changes of PFAS maternal serum concentrations and to identify determinants of PFAS maternal serum concentrations. Our results showed that PFOA, PFOS, and PFHxS serum concentrations of mothers with a young child decreased during our study period. On the other hand, PFNA and PFDA maternal serum concentrations exhibited mixed time trends over the same period. Longer breastfeeding duration was associated with decreased maternal serum concentrations of PFOA, PFOS, PFHxS, PFNA, and PFDA, which were detected most frequently in our study subjects. Older mothers tended to have higher maternal serum concentrations of PFOA and PFDA. We also observed that pre-pregnancy BMI, parity, maternal education, race/ethnicity, and homeownership affected maternal serum concentrations of some PFAS.

The decreasing trends of PFOA, PFOS, and PFHxS observed in our study are consistent with those of other studies reported for other U.S. populations (Table 2.5) (Gribble et al. 2015; Hurley et al. 2018; Olsen et al. 2012). The results of these studies and our current study suggest that serum concentrations of PFOA, PFOS, and PFHxS are decreasing in the U.S. general population. Furthermore, the decreasing trends of PFOA, PFOS, and PFHxS in this study are also consistent with those of other studies outside the USA (Bjerregaard-Olesen et al. 2016; Eriksson et al. 2017; Okada et al. 2013; Shu et al. 2018) (Table 2.5). Serum concentrations of PFNA and PFDA showed mixed trends in the present study. Three studies reported downward trends of PFNA and PFDA serum concentrations within the U.S. (Gribble et al. 2015; Hurley et al. 2018; Jain 2018), whereas one study reported increasing trends (Olsen et al. 2012). The inconsistent results of PFNA and PFDA among studies may partially relate to the differences in study populations and study periods.

Table 2.5. Summary of temporal trends in PFAS serum concentrations from previous studies.

	Studies	Location	Population	Period	PFOA	PFOS	PFHxS	PFNA	PFDA	PFUA	Me-FOSAA
USA	Olsen et al. 2012 [a]	6 States	General adults	2000-2010	↓	↓	↓	↑	↑	↑	-
	Gribble et al. 2015	South Carolina	African American Gullah	2003-2013	↓	↓	↓	↓	↓	↓	-
	Hurley et al. 2018	California	California teachers	2011-2015	↓	↓	↓	↓	↓	-	↓
	Jain et al. 2018 [b]	All States	General adults	2003-2014	↓	↓	↓	↓	↓	-	-
	This study	California	Mothers with young children	2009-2016	↓	↓	↓	↑ (2009~2012), and then ↓	No significant trend	↓ (2009~2013), and then ↑	↓
Outside USA	Glynn et al. 2012	Sweden	Primiparous women	1996-2010	↓	↓	↑	↑	↑	↑	-
	Okada et al. 2013	Japan	Pregnant women	2003-2011	↓	↓	-	↑	↑	No significant trend	-
	Bjerregaard-Olesen et al. 2018	Denmark	Pregnant women	2008-2013	↓	↓	↓	↓	↓	↓	-
	Eriksson et al. 2017	Australia	General people	2002-2013	↓	↓	↓	↑ (2002~2006), and then ↓	↑ (2002~2008), and then ↓	↑ (2002~2006), and then ↓	-
	Shu et al. 2018	Sweden	Pregnant women	2007-2010	↓	↓	↓	No significant trend	No significant trend	No significant trend	-

[a] Olsen et al. (2012) quantified PFAS concentrations in plasma samples collected from California, Maryland, Massachusetts, Minnesota, North Carolina, and Oregon.
[b] Jain et al. (2018) used PFAS serum concentrations reported in National Health and Nutrition Examination Survey (NHANES).

Different annual percent changes may reflect physiological differences or different exposure sources of individual PFAS. For example, for PFOA and PFOS that were relatively strictly regulated during similar time periods and have relatively similar biological elimination half-lives, we observed that the rates of decrease were almost identical for PFOA (-10.7% per year) and PFOS (-10.8% per year). However, PFHxS serum concentrations were decreasing rather slowly (-8.0% per year) compared to PFOA and PFOS in the same population, although PFHxS was also phased out in the early 2000s with PFOS (Calafat et al. 2007). The slower rate of decrease in PFHxS serum concentrations observed in this study is also consistent with results of previous studies (Bjerregaard-Olesen et al. 2016; Gribble et al. 2015; Hurley et al. 2018). One possible reason for the slower decrease of PFHxS serum concentrations is its longer biological elimination half-life (5.3 - 15.5 years) (Y Li et al. 2018; Olsen et al. 2007; Worley et al. 2017) than those of PFOA (2.4 - 3.9 years) (Bartell et al. 2010; Gomis et al. 2016; Y Li et al. 2018; Olsen et al. 2007; Russell et al. 2015; Worley et al. 2017) and PFOS (3.3 - 5.4 years) (Y Li et al. 2018; Olsen et al. 2007; Worley et al. 2017). Moreover, because PFHxS was used in some Scotchgard formulation to treat carpets and furniture (Beesoon et al. 2012) and these consumer items tend to have long product lives, it is likely that PFHxS exposure may continue for years after it was phased out for use.

Our study identified several determinants of maternal PFAS serum concentrations (Tables 2.3 and 2.4, Figures S2.1 and S2.2). For example, longer breastfeeding duration was associated with decreased PFAS maternal serum concentrations for the five most frequently detected compounds (i.e., PFOA, PFOS, PFHxS, PFNA, PFDA) in the current study (Table 2.4). This is additional evidence that lactation is a major excretion route of PFAS for nursing mothers, as presented in previous studies (Mogensen et al. 2015; Papadopoulou et al. 2016). We observed

differences in the PFAS maternal serum concentrations and rates of decrease among different maternal race/ethnicity. These differences might be, in part, due to differences in dietary habits or metabolism by different race/ethnicity (Jain 2014; Johnson 1997). Higher maternal serum concentrations of PFOA and PFDA were associated with increased maternal age. This is consistent with a previous study reporting higher serum concentrations in older people (Bjermo et al. 2013). Older mothers are likely to have greater chances of PFAS exposure than younger mothers before the common long-chain PFAS were phased out. In addition, mothers with high parity had relatively lower serum concentrations of PFOS and PFHxS. Multiple placental transfers of PFAS and subsequent lactational transfers may contribute to the low serum concentrations of PFOS and PFHxS among the CHARGE mothers with high parity (Bjermo et al. 2013; Brantsaeter et al. 2013; Park et al. 2019). Mothers who received higher education tended to have higher serum concentrations of PFOS, PFHxS, and PFDA in early study years, which were consistent with results of a previous study (Bjermo et al. 2013). The fact that women in the United States with higher education were known to consume more fast foods that likely contain PFAS in packaging materials may explain this finding (Hidaka et al. 2018). Mothers who had higher pre-pregnancy BMI also had higher maternal serum concentrations of PFNA and PFDA, which were consistent with results of previous studies (Fei et al. 2007; Olsen et al. 1998).

There are limitations and strengths of this study. We cannot ascertain that our study results (e.g., decreasing trends in maternal PFAS serum concentrations, determinants of maternal PFAS serum concentrations) are applicable to the U.S. general female population, because our study population was limited to a select group of Northern California mothers with a young child. For PFOA, PFOS, PFHxS, and PFNA, serum concentrations of the CHARGE mothers were overall lower than those of the NHANES participants. The differences in PFAS serum

concentrations between our study and NHANES (Figure 2.1) may relate to breastfeeding practices and child birth of our study population. For PFNA and PFDA showing mixed trends in our study, their concentrations were decreasing in NHANES. Furthermore, temporal trends of PFAS serum concentrations within same participants over time were not examined in the current study because our samples were from a case-control study with a single sample for each individual woman. In spite of these limitations, results in our study can help us understand exposure to common long-chain PFAS and their time trends among mothers with a young child. From the investigation of the relationship between breastfeeding duration and maternal serum concentrations, which has not been thoroughly evaluated by previous studies on PFAS temporal trends, we found that breastfeeding duration was inversely associated (p-values <0.01) with maternal serum concentrations of the five most dominant compounds (i.e., PFOA, PFOS, PFHxS, PFNA, PFDA) in the current study.

While there is a public health need to estimate PFAS body burden levels in young children, PFAS concentrations in cord blood are problematic, as evidenced by the weak to moderate correlations between PFAS concentrations in cord blood and those in 3-year-old child's serum: 0.45 for PFOA, 0.21 for PFOS, and 0.46 for PFHxS (Kingsley et al. 2018). Among infants who exclusively breastfed without consuming other foods, their PFAS serum concentrations increased 27.8% for PFOA, 29.2% for PFOS, 20.8% for PFNA, and 18.1% for PFDA per month of breastfeeding (Mogensen et al. 2015), suggesting that breastfeeding duration would be an important predictor of young children's PFAS exposure. PFAS serum concentrations of mothers with a young child were positively associated with those of their young child (Wu et al. 2015). Our results further support the importance of accounting for

breastfeeding histories in a pharmacokinetic model with PFAS serum concentrations collected from mothers with a young child to help estimate or reconstruct early childhood exposure.

CHAPTER 3: TEMPORAL VARIABILITY OF INDOOR DUST CONCENTRATIONS OF SEMIVOLATILE ORGANIC COMPOUNDS

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3.1. Introduction

3.1.1. Background

Exposure to semivolatile organic compounds (SVOCs) in indoor environments and its potential impact on human health have been receiving increased public attention, because people in developed countries spend over 80% of their time indoors (Klepeis et al. 2001) and SVOC levels are several orders of magnitude higher indoors than outdoors (Bennett et al. 2002b; Nazaroff 2008; Shin et al. 2012). SVOCs are introduced into indoor residential settings in the form of consumer products, building materials, furnishings, pesticides, and combustion by-products (Bennett and Furtaw 2004; Weschler and Nazaroff 2008). When indoor SVOCs are released from their original sources, they are redistributed over time among the gas phase, airborne particles, settled dust and other indoor surfaces (Bennett and Furtaw 2004; Weschler and Nazaroff 2010). Consequently, residents can be exposed to indoor SVOCs via inhalation, dermal uptake, and dust ingestion (Little et al. 2012). Of interest, for young children who crawl and play on the floor and have frequent hand-to-mouth activity, dust ingestion has been determined to be a major non-dietary exposure route for several classes of SVOCs including phthalates (Mitro et al. 2016; Pelletier et al. 2017), polybrominated diphenyl ethers (PBDEs) (Pelletier et al. 2017; Wilford et al. 2005), organophosphate flame retardants (OPFRs) (Zhou et al. 2017), per- and polyfluoroalkyl substances (PFAS) (Egeghy and Lorber 2011; Lorber and Egeghy 2011; Mitro et al. 2016), polycyclic aromatic hydrocarbons (PAHs) (Gevao et al. 2007), and polychlorinated biphenyls (PCBs) (L Li et al. 2018).

The measured indoor dust concentrations have been found to have positive correlations with concentrations in biological samples (e.g., serum, urine) for PBDEs (Bennett et al. 2015; Johnson et al. 2013), OPFRs (Dodson et al. 2014; Fromme et al. 2014; Meeker et al. 2013), PCBs (Rudel et al. 2008), bisphenol A (Liu et al. 2019), and pyrethroid pesticides (Trunnelle et

al. 2014). Moreover, epidemiologic studies using indoor dust concentrations as surrogates for human exposure found positive associations between dust concentrations of tributyl phosphate and incidence of asthma and allergies (Araki et al. 2014) as well as di(2-ethylhexyl) phthalate (DEHP) and incidence of allergies (Bamai et al. 2016). It was also found that increasing levels of biocides (i.e., organochlorines, residential insecticides) in dust were known to be associated with increasing risk of non-Hodgkin lymphoma (Colt et al. 2005; Colt 2006; Colt et al. 2009). This suggests that for young children who are exposed to SVOCs primarily via dust ingestion, indoor dust concentrations collected from a single visit can be used as surrogates for exposure to some SVOCs, particularly if diet and personal care product use are not significant exposure pathways.

3.1.2. Statement of research problem

SVOCs are fairly persistent indoors (Shin et al. 2013; Weschler and Nazaroff 2008) and therefore the temporal variability of SVOC dust concentrations is typically smaller than that of concentrations in indoor air (Egeghy et al. 2005). In addition, compared to volatile organic compounds that are typically released into air from dynamic indoor sources, many SVOCs are slowly released from building materials, furnishings and other indoor solid items and have a strong tendency to partition to settled dust. Thus, SVOC dust levels are relatively stable over time. However, several studies observed different degrees of temporal variability of SVOC dust concentrations across chemical classes or within the same chemical class (Bennett et al. 2015; Cao et al. 2014; Egeghy et al. 2005; Whitehead et al. 2012). Episodic occupant activities (e.g., cooking, vacuuming, ventilation) and environmental factors (e.g., temperature, relative humidity, airborne particulate matter concentration) are also known to affect SVOC air and/or dust concentrations during a short period of time (Kristensen et al. 2019; Wei et al. 2019). Seasonal

factors (e.g., different product use pattern, ventilation frequency) may also play a role in temporal variability in measured dust concentrations (Cao et al. 2019). Thus, further studies are needed to extensively examine temporal variability of dust concentrations for a wide range of SVOCs detected in household dust and other determinants (e.g., chemical properties, seasonal difference in use patterns or emission sources) of the temporal variability of dust concentrations.

3.1.3. Objective

The objective of this study was to investigate temporal variability of dust concentrations for a wide range of SVOCs. Specifically, we quantified 47 compounds in dust samples repeatedly collected from 29 homes during a period of 22 months. The measured SVOCs represent a broad range of compounds across chemical properties and product use categories, and thus are ideal for evaluating the sources of any differences in temporal concentration patterns. We also examined whether sampling season affected measured dust concentrations using samples collected at two seasons with similar climate (spring and fall) and two seasons with opposite climate (summer and winter).

3.2 Methods

3.2.1 Dust sample collection

We recruited households in Northern California as part of an effort to examine the overall decrease of flame retardant (FR) concentrations in indoor dust after replacing old couches (assumed to be the primary sources of flame retardants in participating homes). Under the assumption that dust concentrations of non-FR SVOCs were not affected by the replacement of old couches, we examined the temporal variability of non-FR SVOCs. Households were selected

as part of one of two studies; one study where participants bought their couch by themselves and the other where the study team bought a couch. In total, 34 household replaced their couches, and of these, 29 completed three visits where a dust sample was collected over 22 months, from July 2015 to May 2018, for a total of 87 dust samples. The second visit was approximately 15.3 months apart from the first visit ($\sigma = \pm$ 2.6 months), allowing us to examine variability of dust concentrations during a long period of time. The third visit was approximately 6.3 months apart from the second visit ($\sigma = \pm$ 0.8 months), allowing us to examine variability of dust concentrations during a short period of time. All recruitment and data collection protocols were approved by the Institutional Review Board for the University of California at Davis (UC Davis). Participants provided informed consent before collection of any data.

For each home visit, samples were collected from the main living room under the assumption that the main living room is where dust concentrations are of interest for residential exposure assessment. One standardized protocol is to vacuum all surfaces except for under furniture and between cushions (Allen et al. 2008; Rudel et al. 2003), but we modified the protocol to also exclude vacuuming upholstered furniture. Dust was collected with a Eureka Mighty-Mite vacuum cleaner equipped with the standard crevice tool attachment (Model 3670), modified to capture dust in a 19×90 mm cellulose extraction thimble (Whatman Inc) (Allen et al. 2008; Rudel et al. 2003). The thimbles containing the dust samples were wrapped in pre-cleaned aluminum foil, placed in 50 ml polypropylene vials, shipped in a cooler to the central repository, and stored at -20 °C at UC Davis until analysis. The dust was shaken from each thimble into a 100-mesh stainless steel sieve and then sieved to obtain the fraction of dust smaller than 150 μm.

3.2.2. Target compounds

We used a target compound list developed through a project looking at widely-detected compounds in household dust (Moschet et al. 2018; Shin et al. 2020), including new compounds, originally detected through suspect screening or nontarget identification with high resolution mass spectrometry, for which we subsequently obtained standards. Using this expanded target compound list, we analyzed all compounds that were previously detected in dust via gas chromatography (GC). To find the most common and primary use category of studied compounds, we relied on use categorization in the U.S. Environmental Protection Agency (EPA)'s Consumer Product Chemical Profiles database (Goldsmith et al. 2014) and the U.S. National Library of Medicine's Household Product Database. Details of use categorization are available elsewhere (Shin et al. 2020). The selected compounds include ultraviolet [UV] filters, fragrances, and other ingredients of personal care products (PCPs); insecticide ingredients; and a variety of other compounds widely detected in homes (phenols, phthalates, other plasticizers, PAHs, and skin oils, those lipids found in the skin such as squalene). We excluded 11 compounds used as flame retardants from this analysis because replacing couches would affect dust concentrations of flame retardants. The selected compounds represent a range of chemical properties, ranging from relatively volatile compounds (e.g., dimethyl phthalate [DMP]) to those with a high tendency to partition to dust (e.g., DEHP). Chemical properties including the octanol-air partition coefficient (log K_{oa}) and use categories of our study compounds are listed in Table S3.1 of Supporting Information.

3.2.3. Sample analysis

Details of the analytical methods and settings are found elsewhere (Moschet et al. 2018; Shin et al. 2020). Briefly, we quantified concentrations of 47 compounds using GC quadrupole

time-of-flight (Q/TOF) mass spectrometry (MS) at UC Davis. Analysis was carried out on an Agilent 7890B gas chromatograph with a HP-5MS (30 m × 0.25 mm, 0.25 μm) column coupled to an Agilent Q/TOF 7200B instrument running in electron ionization (EI) mode. A 78 min run time with a linear temperature gradient from 35 to 325 °C was chosen to separate all selected compounds and all major peaks in the analysis of a dust extract. Absolute recovery and precision for each analyte are available in Table S3.1.

3.2.4. Statistical analysis

We performed all statistical analyses using STATA/IC 15.1 (StataCorp LLC, College Station, TX, USA) and R version 3.6.1. For all analyzed compounds, we provided summary statistics of dust concentrations for each of the three visits. For all other statistical analyses that require sufficient detection of the samples, we included compounds detected in more than 50% of the samples at all three visits. For concentrations below the limit of detection (LOD), we assigned a value of the LOD divided by the square root of 2 (Hornung 1990). We compared the distributions of dust concentrations for all three visits and performed a paired t-test of log-transformed concentrations to compare means between two visits (i.e., 1st visit versus 2nd visit, 2nd visit versus 3rd visit, 1st visit versus 3rd visit) and adjusted p-values for multiple comparison of means with a false discovery rate method (Benjamini et al. 2001). Sources of indoor SVOCs may vary with several household characteristics. Thus, we examined if there were significant differences in SVOC dust concentrations between groups of two household characteristics: household income (<$100K, $100K-$150K, >$150K) and presence of children (yes, no). We performed the Wilcoxon-Mann-Whitney test for a binary variable and the Kruskal-Wallis test for the other categorical variable.

To test for within-home temporal variability of SVOC dust concentrations during the three visits (~22 months), we computed the intraclass correlation coefficient (ICC) of individual compounds, a ratio of between-home variance to total variance (within-home variance + between-home variance). ICC ranges from 0 (no reproducibility of within-home measurements) to 1 (perfect reproducibility of within-home measurements) (Adibi et al. 2008; Rosner 2000). We used natural log (ln)-transformed concentrations to account for skewed distributions of dust concentrations and then calculated ICCs and 95% confidence intervals (CIs) using a mixed-effects model with sampling date for fixed effects (Shrout and Fleiss 1979). We examined the relationship between ICCs and log K_{oa} within the same use category or chemical class. Because K_{oa} is a strong predictor of SVOC partitioning between indoor air and dust (Weschler and Nazaroff 2010), it may affect within-home temporal variability of SVOC dust concentrations. For our study compounds, log K_{oa} values ranged from 5.68 for DMP to 11.74 for di-n-octyl phthalate (DnOP) and octocrylene.

Because we collected dust samples with different time intervals (i.e., ~15 months between the 1st and 2nd visits and ~6 months between the 2nd and 3rd visits), we also examined the effect of different sample collection time intervals on the magnitude of temporal variability of dust concentrations by computing Pearson correlation coefficients (r) of ln-transformed concentrations between two visits. To examine whether sampling season affected dust concentrations because of seasonal variations in ventilation or product use (e.g., UV filters, pesticides), we computed Pearson correlation coefficients of ln-transformed concentrations using samples collected (1) between spring (March to May) and fall (September to November) for similar climate (n = 13 homes) and (2) between summer (June to August) and winter (December to February) for opposite climate (n = 16 homes). To examine the differences of correlation

coefficients (1) between two different time intervals (1st and 2nd visits versus 2nd and 3rd visits) and (2) between two different sets of seasons (spring and fall versus summer and winter), we used the 'cocor' function in R which compares two correlations based on either independent or dependent groups (Diedenhofen and Musch 2015).

3.3. Results

3.3.1. Measured dust concentrations

Among 47 quantified compounds, 26 compounds were detected in more than 50% of the samples at all three visits and 15 compounds were detected in more than 90% of the samples at all three visits (Table 3.1). The highest median from all three visits was observed in acetyl tributyl citrate [ATBC] (7.9×10^6 ng/g), followed by dioctyl terephthalate [DOTP] (9.3×10^4 ng/g), DEHP (7.6×10^4 ng/g), squalene (4.9×10^4 ng/g), and cholesta-3,5-diene (1.8×10^4 ng/g). We observed a statistically significant difference of mean concentrations between visits for 6 compounds (p-value < 0.05, see Figure 3.1). For 13 SVOCs, dust concentrations were associated with household income or presence of children (Table 3.2). Homes with the lowest household income had the highest dust concentration for di-isobutyl phthalate (DiBP), DOTP, phenanthrene, pyrene, 2-benzylideneoctanal, galaxolide, tonalide and lilial, but had the lowest dust concentration for octocrylene (p-value < 0.05). Homes with children had higher dust concentrations than those without children for ATBC, DOTP, 2-benzylideneoctanal, galaxolide, tonalide, lilial, and triethyl citrate, but had lower dust concentrations for di-n-butyl phthalate (DBP), phenanthrene and pyrene (p-value < 0.05).

Among the 47 quantified compounds, 21 compounds were detected in less than 50% of the samples at least one visit (Table 3.1). Most of them were insecticide ingredients, PAHs, or

phenols (i.e., 7 out of 9 insecticide ingredients, 8 out of 10 PAHs, and all 5 phenols) with detection frequencies ranging between 1% and 57% among a total of 87 dust samples. The detection frequency of bis(2-ethylhexyl) adipate [DEHA] increased during three visits (38%, 55%, 59%, respectively) and the 75th percentile concentrations were relatively similar during three visits (10.6, 9.3, 9.1 µg/g of dust, respectively). P-cresol, phenol, and benzo(b)fluoranthene were detected in more than 50% of the samples in the first visit but were less detected in later visits (7% and 14% for p-cresol, 3% and 0% for phenol, 48% and 24% for benzo(b)fluoranthene, respectively).

Table 3.1. Median concentrations (ng/g of dust) of 47 detected compounds from 29 households.

Chemical class	Compounds	1st visit			2nd visit			3rd visit			All visits	
		LOD	DF (%)	Median	LOD	DF (%)	Median	LOD	DF (%)	Median	DF (%)	Median
Phthalates	Benzyl butyl phthalate	1000	100	5209	2500	97	7250	2500	97	6447	98	6280
	Di(2-ethylhexyl) phthalate	10000	100	76963	25000	100	69040	25000	100	77379	100	75637
	Di-isobutyl phthalate	1000	100	6996	1000	100	4595	1000	100	5943	100	5609
	Di-n-butyl phthalate	1000	100	5521	2500	72	3626	2500	76	4351	83	4257
	Di-n-octyl phthalate	1000	79	233	250	79	311	250	69	315	76	277
	Diethyl phthalate	100	100	846	250	100	824	250	100	786	100	796
	Dimethyl phthalate	50	100	98	100	69	123	100	52	105	74	110
Other plasticizers	Acetyl tributyl citrate	10000	100	7.9×10^6	100000	100	7.8×10^6	100000	100	8.1×10^6	100	7.9×10^6
	Bis(2-ethylhexyl) adipate	10000	38	-	5000	55	5209	5000	59	6060	51	7071
	Dioctyl terephthalate	2500	100	108870	10000	100	92947	10000	100	91331	100	92947
Insecticides	Bifenthrin	50	55	27	50	55	37	50	52	19	54	27
	Chlorpyrifos	50	21	-	50	17	-	50	10	-	16	-
	Cyhalothrin	250	0	-	500	3	-	500	7	-	3	-
	Cypermethrin	1000	21	-	2500	14	-	2500	17	-	17	-
	Deltamethrin	500	10	-	1000	7	-	1000	10	-	8	-
	Esfenvalerate	500	0	-	500	7	-	500	10	-	6	-
	Etofenprox	1000	3	-	1000	0	-	1000	3	-	2	-
	Permethrin	100	97	1276	500	90	1952	500	90	1764	92	1377
	Phenothrin	100	3	-	250	0	-	250	3	-	2	-
Phenols	2-Chlorophenol	500	0	-	250	3	-	250	3	-	2	-
	2-Nitrophenol	1000	3	-	2500	0	-	2500	0	-	1	-
	4-Chloro-3-methylphenol	1000	3	-	500	0	-	500	0	-	1	-
	para-Cresol	2500	55	1005	2500	7	-	2500	14	-	25	-
	Phenol	1000	97	962	5000	3	-	5000	0	-	33	-
PAHs	Anthracene	100	3	-	500	7	-	500	10	-	7	-
	Benzo(a)anthracene	100	14	-	500	3	-	500	0	-	6	-
	Benzo(a)pyrene	250	7	-	500	3	-	500	0	-	3	-
	Benzo(b)fluoranthene	100	100	48	250	48	-	250	24	-	57	71
	Benzo(g,h,i)perylene	1000	0	-	500	3	-	500	3	-	2	-
	Crysene	100	7	-	250	28	-	250	21	-	18	-

	Dibenzo(a,h)anthracene + Indeno(1,2,3-cd) pyrene	500	3	-	250	21	-	250	10	-	11	-
	Fluorene	100	10	-	100	0	-	100	0	-	3	-
	Phenanthrene	100	97	253	100	90	195	100	79	134	89	176
	Pyrene	50	100	167	50	97	193	50	100	181	99	173
Cosmetic ingredients	Benzyl benzoate	100	100	1131	500	86	992	500	86	958	91	987
	Glycerol tricaprylate	100	100	6273	500	90	12230	500	97	12211	95	10372
	Isopropyl myristate	100	100	3903	250	97	4790	250	100	4724	99	4669
	Lilial	50	100	129	25	86	125	25	86	105	91	125
	Triethyl citrate	100	100	463	500	86	913	500	86	1378	91	691
Fragrance ingredients	2-Benzylideneoctanal	500	93	404	250	79	554	250	79	526	84	506
	Galaxolide	50	100	246	100	83	468	100	90	365	91	308
	Tonalide	100	79	82	50	76	76	50	66	74	74	76
UV filters	Benzophenone	100	100	803	500	100	909	500	100	559	100	705
	Homosalate	250	100	4837	250	100	4650	250	100	4504	100	4650
	Octocrylene	100	100	15343	500	100	9415	500	100	8477	100	10095
Skin oils	Cholesta-3,5-diene	500	100	18121	1000	100	13226	1000	100	20513	100	18121
	Squalene	1000	100	49834	2500	100	36462	2500	100	50762	100	48797

Abbreviation: limit of detection (**LOD**), detection frequency (**DF**), polycyclic aromatic hydrocarbons (**PAH**), ultraviolet (**UV**)

Table 3.2. Median dust concentrations (ng/g of dust) of 26 compounds by household characteristics.

Compounds	Annual household income ($) [a]				Presence of child(ren)		
	< 100K (n = 7)	100K-150K (n = 10)	> 150K (n = 9)	p-value [b]	Yes (n = 21)	No (n = 8)	p-value [b]
Phthalates							
BBP	8,418	5,208	8,102	0.11	6,054	6,736	0.66
DBP	4,536	4,741	4,099	0.91	3,876	7,544	**0.02**
DEHP	87,634	68,435	77,379	0.12	73,787	77,171	0.58
DiBP	8,370	5,525	3,600	**<0.01**	6,428	4,753	0.15
DnOP	233	323	307	0.14	252	387	0.054
DEP	975	747	927	0.16	791	828	0.66
DMP	106	83	138	0.12	125	93	0.29
Other plasticizers							
ATBC	1.1×10^7	6.5×10^6	7.4×10^6	0.12	9.4×10^6	4.6×10^6	**<0.01**
DOTP	144,785	75,467	70,354	**<0.01**	115,875	56,015	**<0.01**
Insecticide ingredients							
Bifenthrin	18	50	18	0.23	45	18	0.13
Permethrin	2,829	1075	2,232	**0.04**	1764	1,131	0.56
Ultraviolet (UV) filters							
Benzophenone	580	856	805	0.10	780	710	0.53
Homosalate	3,179	5,302	5,405	0.06	4,988	4,293	0.24
Octocrylene	5,517	11,791	14,294	**0.01**	14,294	7244	0.08
Polycyclic aromatic hydrocarbons (PAHs)							
Phenanthrene	281	192	133	**0.03**	165	253	**0.04**
Pyrene	248	169	132	**0.01**	167	231	**0.01**
Fragrance ingredients							
2-Benzylideneoctanal	2,148	300	411	**<0.01**	894	178	**<0.01**
Galaxolide	965	155	277	**<0.01**	415	140	**<0.01**
Tonalide	182	66	75	**<0.01**	99	63	**0.01**
Cosmetic ingredients							
Benzyl benzoate	1,202	866	850	0.15	987	1,029	0.84
Glycerol tricaprylate	8,523	18,368	11,438	0.30	10,364	12,273	0.38
Isopropyl myristate	5,326	4,686	3,245	0.40	4,790	4,686	0.75
Lilial	269	62	88	**<0.01**	163	53	**<0.01**
Triethyl citrate	1,310	509	661	0.62	1048	391	**<0.01**
Skin oils							
Cholesta-3,5-diene	11,739	21,944	23,157	0.09	21,415	12,222	0.37
Squalene	40,278	49,719	49,834	0.84	58,097	38,258	0.19

[a] Among total 29 households, dust concentrations of 3 households were excluded in the calculation of median because residents of the three households did not know or refused to answer their incomes.

[b] P-value from the Kruskal-Wallis test or the Wilcoxon-Mann-Whitney test. P-values less than 0.05 were highlighted in bold.

Abbreviation: acetyl tributyl citrate (**ATBC**), benzyl butyl phthalate (**BBP**), diethyl phthalate (**DEP**), bis(2-ethylhexyl) phthalate (**DEHP**), di-isobutyl phthalate (**DiBP**), dimethyl phthalate (**DMP**), di-n-butyl phthalate (**DBP**), di-n-octyl phthalate (**DnOP**), dioctyl terephthalate (**DOTP**)

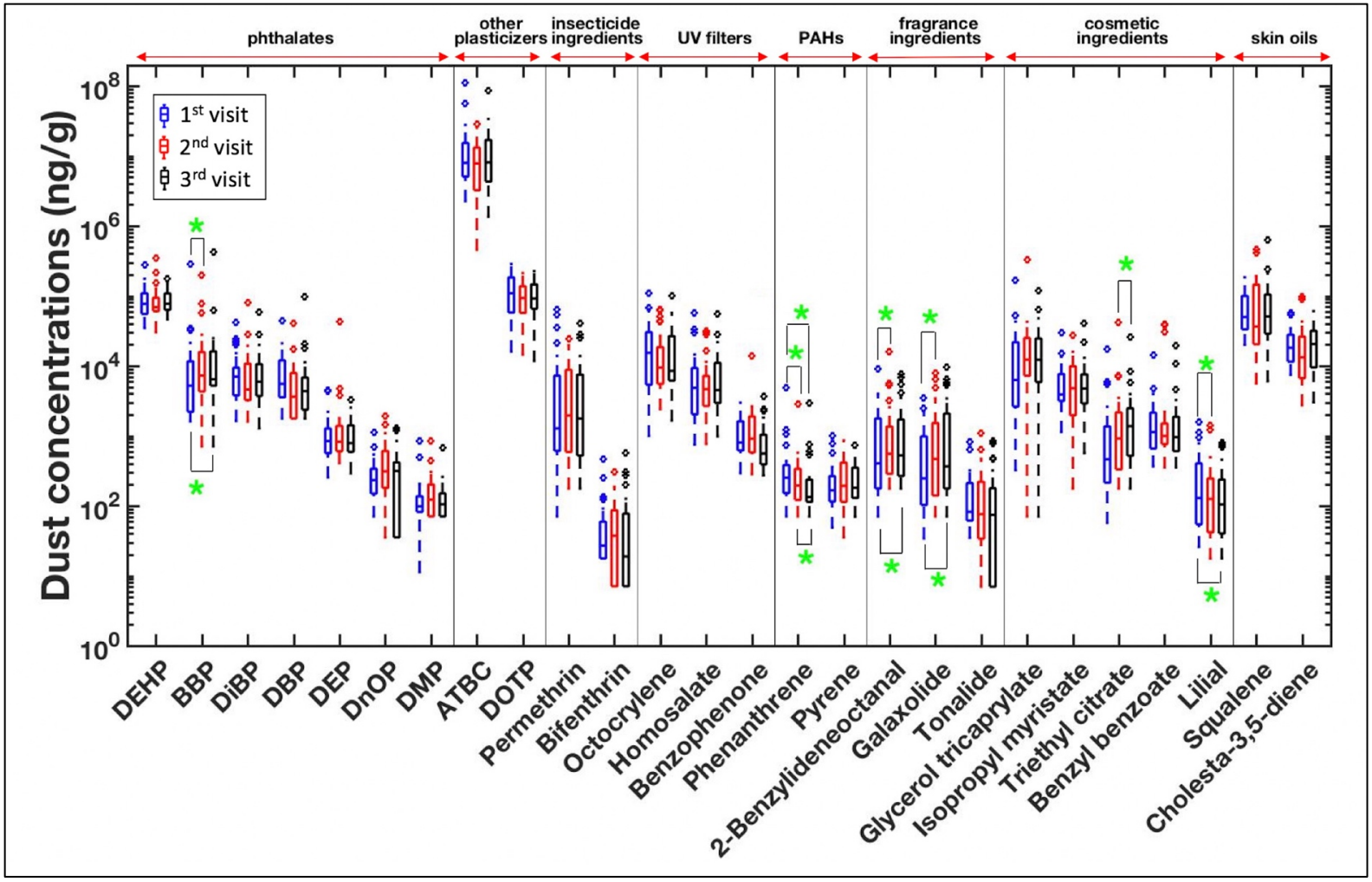

Figure 3.1. Distribution of dust concentrations (ng/g of dust) for 26 organic compounds detected in more than 50% of the samples at all three visits. P-value < 0.05 are marked (*).

Abbreviation: acetyl tributyl citrate (**ATBC**), benzyl butyl phthalate (**BBP**), diethyl phthalate (**DEP**), di(2-ethylhexyl) phthalate (**DEHP**), di-isobutyl phthalate (**DiBP**), dimethyl phthalate (**DMP**), di-n-butyl phthalate (**DBP**), di-n-octyl phthalate (**DnOP**), dioctyl terephthalate (**DOTP**)

3.3.2. Within-home variability during three visits

Among the 26 compounds, 5 compounds had ICCs above 0.75, 15 compounds had ICCs between 0.50 and 0.75, and 6 compounds had ICCs below 0.50 (Figure 3.2). Permethrin (used as an insecticide ingredient) had the highest ICC (= 0.90; 95% CI: 0.83, 0.95) and benzophenone (used as a UV filter) had the lowest ICC (= 0.30; 95% CI: 0.08, 0.54). Within the same use category or chemical class with only 2 or 3 compounds, ICCs were higher with higher values of log K_{oa} for non-phthalate plasticizers, insecticides and UV filters, whereas ICCs were lower with higher values of log K_{oa} for PAHs and skin oils. For phthalates, fragrance ingredients, and cosmetic ingredients, ICCs did not increase or decrease with increasing values of log K_{oa} within the same use category or chemical class.

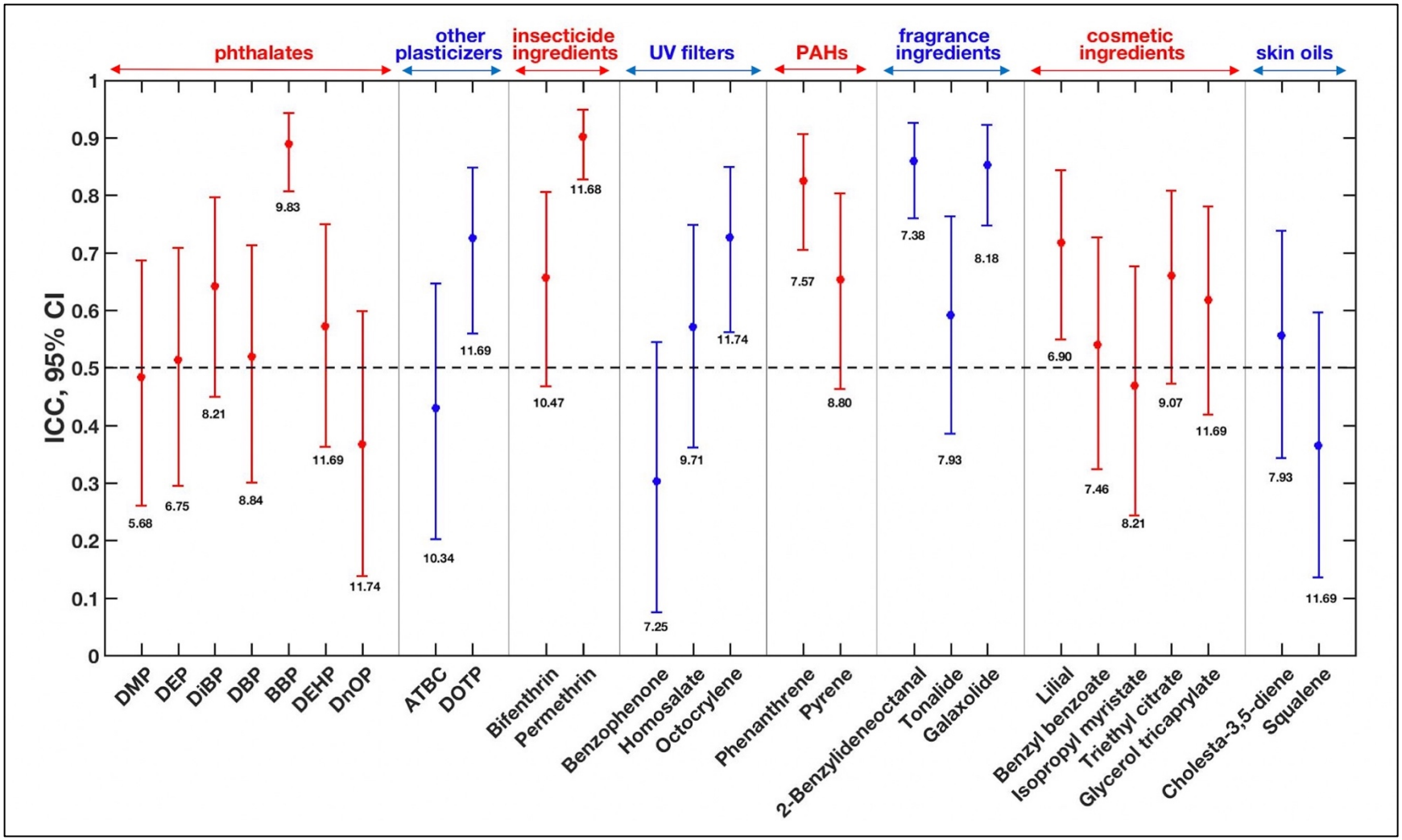

Figure 3.2. Intraclass correlation coefficients (ICCs) and 95% CIs of ln-transformed dust concentrations of 26 compounds across three sampling time points in the increasing order of the octanol-air partition coefficient (K_{oa}) within each class. The log K_{oa} value of each compound is placed below the lower bound of 95% CI.

3.3.3. Variability across four seasons

When examining the effect of sampling seasons (collected during similar climate or opposite climate) on the magnitude of temporal variability of dust concentrations, the correlation coefficients between spring and fall (r = 0.48-0.98) were higher than those between summer and winter (r = 0.09-0.92) for 19 compounds (Figure 3.3). The differences of correlation coefficients between spring-fall and summer-winter were statistically significant (p-value < 0.05) for DMP, DOTP, octocrylene, phenanthrene, and squalene. Among the 19 compounds, DMP, which is commonly used in insect repellents (Metcalf 2014), showed the largest difference between the two coefficients (0.09 versus 0.79) and squalene showed the second largest difference between the coefficients (0.31 versus 0.90). All insecticide ingredients, UV filters, PAHs, and skin oils had consistently lower correlation coefficients between summer and winter, compared to those between spring and fall. When comparing the distributions across four seasons, two compounds (i.e., ATBC, squalene) had variations in dust concentrations between seasons (p-value < 0.05, Figure 3.4). Mean ATBC concentrations were lower during summer than other seasons, and mean squalene concentrations were the highest during winter and the lowest during spring.

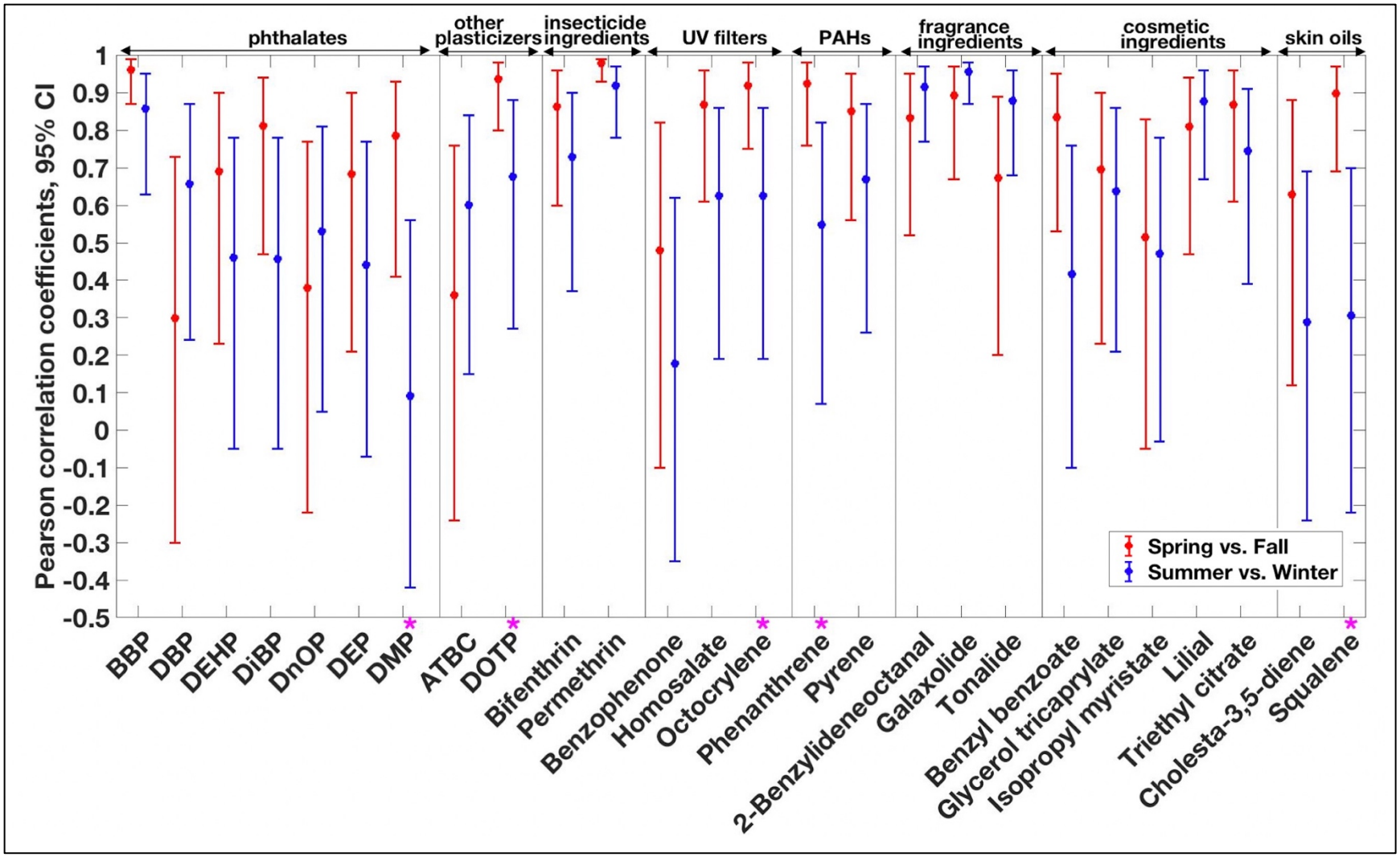

Figure 3.3. Pearson correlation coefficients of ln-transformed dust concentrations and 95% CIs between spring and fall (n = 13 homes) and between summer and winter (n = 16 homes). Compounds whose coefficients between spring-fall and summer-winter were different (p-value < 0.05) were marked (*).

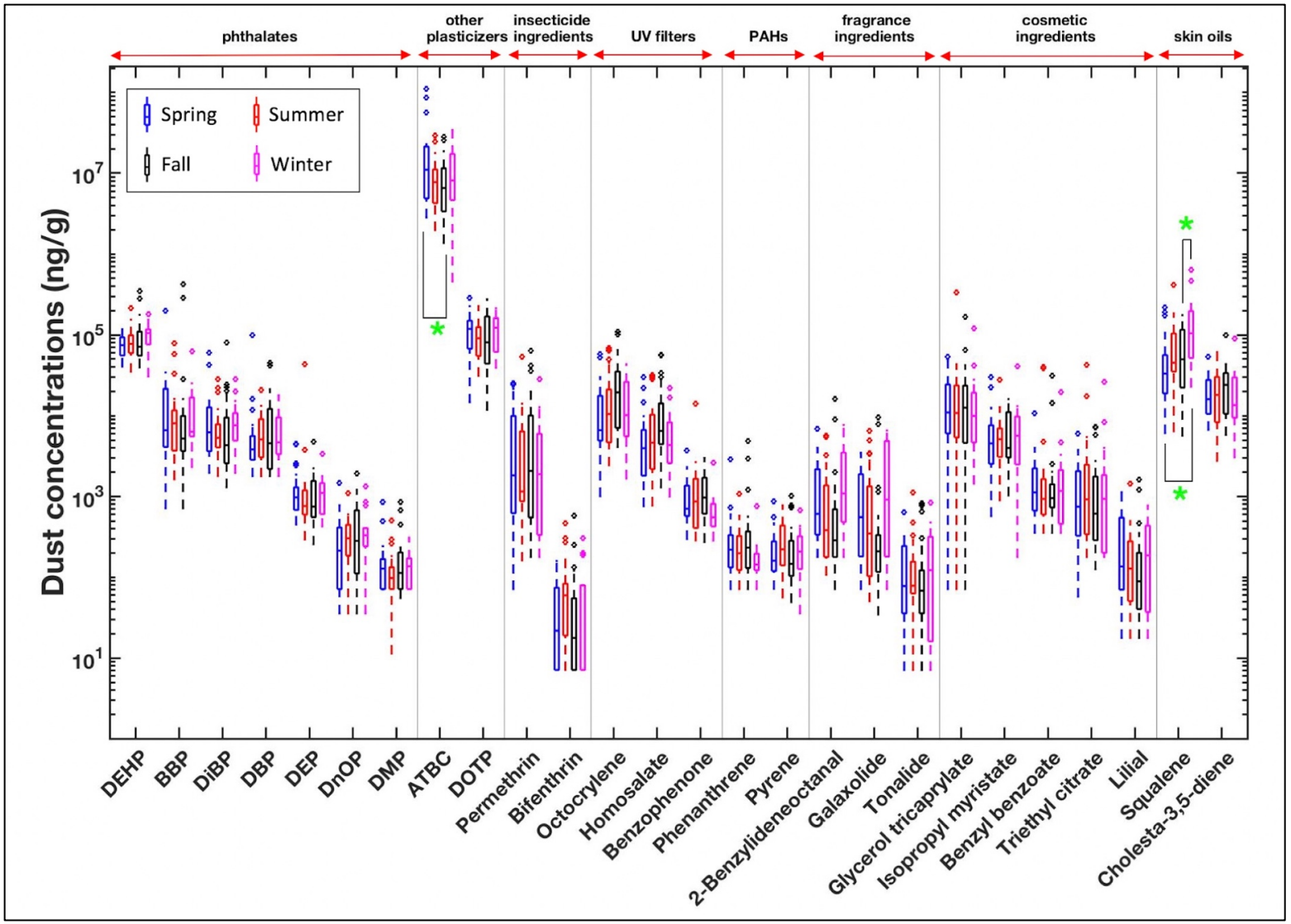

Figure 3.4. Distributions of dust concentrations (ng/g of dust) for 26 organic compounds detected in more than 50% of the samples for each of the four seasons. P-values < 0.05 are marked with asterisk (*).

3.3.4. Correlation between visits

Pearson correlation coefficients (r) of ln-transformed dust concentrations between two visits for the 26 compounds were positive: r = 0.26-0.90 between the 1st and 2nd visits and r = 0.34-0.95 between the 2nd and 3rd visits (Figure 3.5). Except for benzophenone and DnOP, the correlation coefficients between the first two visits with a long-time interval ($\mu \pm \sigma$ = 15.3 ± 2.6 months) was statistically significant (p-value < 0.05) for the other 24 compounds. Similarly, except for benzophenone, the correlation coefficients between the 2nd and 3rd visits with a short time interval ($\mu \pm \sigma$ = 6.3 ± 0.8 months) were statistically significant for the other 25 compounds. When the sample collection time interval between visits increased, the correlation coefficients tended to decrease for 19 out of 26 compounds. However, the differences of correlation coefficients between 1st-2nd visits and 2nd-3rd visits were not statistically significant (p-value > 0.05) for all 26 compounds.

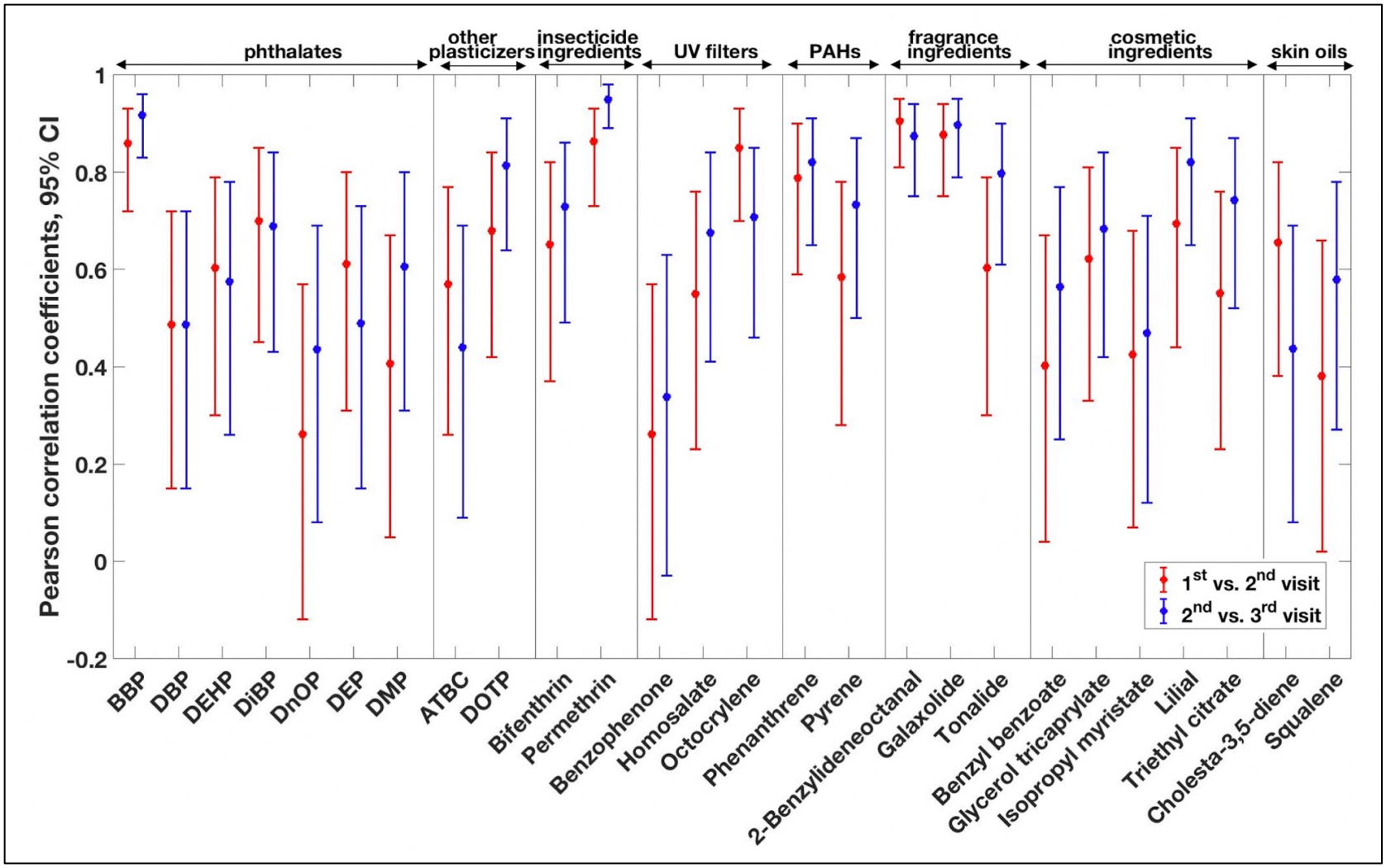

Figure 3.5. Pearson correlation coefficients of ln-transformed dust concentrations and 95% confidence intervals between visits with short (1st versus 2nd) and long (2nd versus 3rd) time intervals. An average time interval between the 1st and 2nd visits and between the 2nd and 3rd visits was 15.3 months (σ = 2.6 months) and 6.3 months (σ = 0.8 months), respectively. Differences in coefficients between 1st-2nd visits and 2nd-3rd visits were not statistically significant for all 26 compounds (p-value > 0.05).

3.4. Discussion

In this study, we examined temporal variability of dust concentrations for a wide range of SVOCs over a 22-month period and investigated the effect of sampling season, log K_{oa}, and sample collection time intervals on the magnitude of temporal variability of dust concentrations. Among the 26 compounds detected in more than 50% of the samples at all three visits, 20 compounds showed moderate ($0.50 \leq ICCs < 0.75$) to high ($ICCs \geq 0.75$) reproducibility of within-home measurements in dust concentrations, while the remaining 6 compounds showed relatively low reproducibility ($ICCs < 0.5$). Overall, dust concentrations of insecticide ingredients, PAHs, and fragrance ingredients were more stable over time, with those of skin oils (or constituents of skin surface lipids) being the least stable. For those compounds with high ICCs, dust concentrations collected from a single visit may serve as surrogates for exposure for approximately 2 years, particularly for infants and toddlers. We observed higher correlation coefficients of dust concentrations between spring and fall than those between summer and winter for 19 compounds, indicating seasonal effects on dust concentrations. When the sample collection time interval between visits increased, the correlation coefficients tended to decrease for 19 out of 26 compounds, but results were not statistically significant for any compounds.

Our study showed that dust concentrations may vary over time for some SVOCs with seasonal variations in source rates. Among the 6 compounds that had relatively low reproducibility of within-home measurements in dust concentrations ($ICCs < 0.5$), two compounds are associated with PCP use including benzophenone (UV filter) and isopropyl myristate (cosmetic ingredient) with a third related to skin sloughing, squalene (found in skin surface lipids). Among them, we observed that benzophenone had the lowest concentration during winter and squalene had the highest concentration during winter (see Figure 3.4). The high concentration of squalene in dust might be due to reduced levels of skin moisture during

winter, which would result in more skin flaking (Engebretsen et al. 2018). Squalene had the second biggest difference in its correlation coefficients (0.90 between spring and fall versus 0.31 between summer and winter) among the 26 compounds. Compared to the correlation coefficients between spring and fall, those between summer and winter tended to be smaller for all UV filters, skin oils, and PAHs in our samples. The reason for the difference between seasons for PAHs is less clear, as there are multiple sources, including cooking, smoking, use of candles, infiltration of outdoor air, and residential heating. Potential reasons for the differences include seasonal differences in ventilation rates or increased PAH emissions from residential heating such as wood burning during winter (Finardi et al. 2017). Our results further support that seasonal variations in source rates, either through increased product use (e.g., UV filters), increased skin flaking (e.g., squalene), or increased transport from outdoors through ventilation or residential heating (e.g., PAHs), may have increased temporal variability of these compound classes in dust concentrations.

In our study, we observed relatively low reproducibility for two phthalates (DnOP, DMP) and one non-phthalate plasticizer (ATBC), although DnOP and DMP did not have seasonal variations (see Figure 3.4 for the distributions across four seasons). For example, although we did not observe differences in means across four seasons for DMP, we observed a very low correlation coefficient between summer and winter (r = 0.09). Sensitivity to temperature changes is known to be another factor leading to temporal variations in the household dust concentrations of compounds with small K_{oa} (Bi et al. 2018; Cao et al. 2014; Weschler and Nazaroff 2010) because K_{oa} is inversely proportional to temperature (Shoeib and Harner 2002b) and compounds with small K_{oa} have relatively high affinity for air rather than dust. DMP and DEP have the smallest value and the second smallest value of log K_{oa} (= 5.68 and 6.75, respectively) among

phthalates and non-phthalate plasticizers. Thus, dust concentrations of these two compounds may vary over time, in part, due to intermittent human activities such as ventilation, combined with a greater fraction in air, resulting in poor reproducibility in dust concentrations. In addition, after equilibrium among indoor compartments (e.g., dust, gas phase, airborne particles) is broken due to vacuuming or ventilation, SVOCs with relatively high log K_{oa} values (DnOP = 11.7 and ATBC = 10.3, respectively) do not reach equilibrium within a short period of time (Weschler and Nazaroff 2008). Thus, intermittent human activities before sample collection may have caused increased temporal variability for DnOP and ATBC.

Comparing temporal variability of dust concentrations for a wide range of SVOCs allowed us to examine the degree of variability by the chemical property relevant to dust concentrations. K_{oa} is a strong predictor of SVOC partitioning between the gas phase and settled dust in indoor environments [8]. Although our comparison by chemical properties within the use category is limited due to the small number of compounds with >50% detection frequency, ranging from 2 to 7, we observed that log K_{oa} may play a role on the magnitude of temporal variability of dust concentrations for such SVOCs. However, we also observed that K_{oa} is not a governing factor determining the magnitude of temporal variability for other chemical classes or use categories. It is likely that dust concentrations of such SVOCs may vary over time due to other factors including intermittent human activities and seasonal differences in use patterns of consumer products. We found increasing reproducibility with increasing values of log K_{oa} among two non-phthalate plasticizers, two insecticides and three UV filters. Based on the U.S. EPA's CompTox Chemicals Dashboard (Phillips et al. 2017), the compounds in these three use categories tend to have a primary use, compared to other chemical classes or use categories that did not show particular trends and tend to have multiple uses (e.g., phthalates). In other words, if

emission sources or product use patterns were similar or relatively simple, reproducibility of dust concentrations is increasing with increasing values of log K_{oa}. On the other hand, for compounds with multiple uses or emission sources, we did not observe the effect of log K_{oa} on the reproducibility of dust concentrations.

We observed that dust concentrations of some SVOCs were associated with household characteristics (Table 3.2). For example, lower income in our population, which were correlated with apartments or townhouses, were associated with higher PAH concentrations in dust. Potential reasons for higher PAH concentrations in homes with low income include close proximity to heavy traffic or industrial sources, or tobacco smoke (Chuang et al. 1999). Higher concentrations of three fragrance ingredients, which are added to personal care and cleaning products, were observed in households with child(ren). Perfumes are used more frequently by the U.S. younger adults with children than elders (aged 60+) (Statista 2017). It is also likely that households with child(ren) are associated with increased use of cleaning products and personal care products.

Our measured dust concentrations reflected regulations of some phthalates in consumer products. For example, when comparing pooled SVOC concentrations in the U.S. dust collected between 2000 and 2015 by Mitro et al. (Mitro et al. 2016) and our dust concentrations collected in California between 2015 and 2018, we observed lower dust concentrations in our study for most phthalates (Table S3.2). Our lower concentrations may reflect the changes of phthalate use in consumer products due to the Consumer Product Safety Improvement Act that was enacted in the U.S. in 2008 to prohibit the use of several phthalates including benzyl butyl phthalate, DBP, and DEHP in children's items (Zota et al. 2014). Moreover, as DiBP is used as a substitute for DBP (Wittassek et al. 2007), we observed higher DiBP dust concentrations than Mitro et al. We

also observed increased detection frequencies of DEHA over three visits from 38% to 59% in our study, potentially due to increased use of consumer products that replace DEHP with DEHA.

CHAPTER 4: EVALUATING COUCH POLYURETHANE FOAM FOR A POTENTIAL PASSIVE SAMPLER OF SEMIVOLATILE ORGANIC COMPOUNDS

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Hyeong-Moo Shin

Luann Wong

Thomas M. Young

Deborah H. Bennett

4.1. Introduction

4.1.1. Background

Reliable sampling methods for assessing exposure to semivolatile organic compounds (SVOCs) in indoor environments are being demanded in environmental health studies (Dodson et al. 2019). Exposure to SVOCs released indoors is of concern because their levels are typically several orders of magnitude higher indoors than outdoors (Bennett et al. 2002a; Shin et al. 2012) and SVOCs have been shown to be toxic or to be associated with adverse health effects such as neurotoxicity (Colt et al. 2005; Colt 2006; Colt et al. 2009; Kamel and Hoppin 2004; Munoz-Quezada et al. 2013; Viel et al. 2015), carcinogenicity (Knafla et al. 2006; Shi et al. 2018), and endocrine disrupting potential (Birnbaum and Staskal 2004; Howdeshell 2002; Iwasaki et al. 2002; Jacobson and Jacobson 1996; Sharpe 2005). Polyurethane foam (PUF), a class of lightweight porous materials (Gama et al. 2018), has been widely used as a sampling medium for SVOCs in both active (Gouin et al. 2005; Hayward et al. 2010; Moeckel et al. 2009; Newton et al. 2016) and passive (Jaward et al. 2004; Shoeib and Harner 2002c; Strandberg et al. 2018) air sampling systems due to its high sorption capacity for organic compounds (Tromp et al. 2019; Zhao et al. 2004). PUF-based active or passive samplers are used to measure concentrations of SVOCs in indoor air (Newton et al. 2016; Shoeib and Harner 2002a). However, deploying purposeful active or passive samplers for SVOCs can add significant cost to studies in terms of extra trips to homes and may also create burdens for residents living in the homes. In addition, passive air samplers utilizing PUF material need to be deployed for a long period of time for SVOCs because of slow equilibrium of SVOCs in PUF disk (Shoeib and Harner 2002c).

PUF is also widely used in pillows, beds, and chair and sofa cushions in office and home environments. Thus, there is evidence that PUF in the home furniture can absorb other SVOCs commonly detected indoors. For example, PUF in an infant crib mattress is not known as a

source of plasticizers, but among the ten used infant crib mattresses that were in contact with mattress covers with detectable plasticizers, at least one plasticizer was detected in nine mattress PUF samples (Boor et al. 2015). This shows that mattress PUF can absorb plasticizers from its cover. Moreover, because PUF in upholstered home furniture is treated with flame retardants (FRs) to lower its flammability (Alaee et al. 2003; Blum and Ames 1977; Hale et al. 2002; Hammel et al. 2017; Stapleton et al. 2009), PUF in the home furniture is an emission source of flame retardants in indoor environments (Keimowitz et al. 2016; Stubbings et al. 2018) and PUF-based couch cushions are one of the flame-retarded home furniture with frequent skin contact. Thus, if couch PUF can be sampled, it may provide a simple way to assess chemical exposure with minimal cost (i.e., no extra trips to deploy samplers and collect them).

4.1.2. Statement of research problem

The partitioning relationship between PUF and air can be used to compute the chemical distribution between air and PUF for unmeasured SVOCs with known chemical properties. To date, SVOC partitioning between PUF and air were determined in chamber studies under the controlled laboratory conditions or outdoor field studies using purposefully-designed PUF samplers (Abdollahi et al. 2017; Bidleman et al. 2016; Francisco et al. 2017; Kamprad and Goss 2007; Parnis et al. 2016; Saini et al. 2019; Tromp et al. 2019). Chamber studies can control environmental conditions (e.g., temperature, humidity) and various indoor human activities that could disturb chemical equilibrium between PUF and air, such as ventilation, cooking, walking, sitting, and cleaning. However, because chamber studies typically use bare PUF disks, the PUF-air partition coefficient ($K_{PUF\text{-}air}$) from chamber studies may not represent common conditions of home furniture PUF such as upholstered couches. Because $K_{PUF\text{-}air}$ values vary with

environmental conditions (Francisco et al. 2017; Zhao et al. 2004), those derived from outdoor field studies may not represent relatively invariant indoor temperature and humidity. Thus, $K_{PUF\text{-}air}$ with direct concentration measurements in upholstered home furniture PUF may improve our understanding of home furniture PUF's actual holding capacity for indoor SVOCs.

4.1.3. Objective

The objectives of this study were to assess the potential utility of couch PUF as a passive sampling medium for indoor SVOCs and to compare the holding capacity of home furniture's PUF with that of other indoor media (i.e., air, dust, carpet). Specifically, we collected couch PUF samples at three different depths inside couch cushions, measured SVOC concentrations (C_{PUF}), and examined concentration changes with depth. Because we previously measured SVOC concentrations in dust (C_{dust}) collected in the same home when PUF was collected (Kim et al. revision under review) and the dust-air partition coefficient ($K_{dust\text{-}air} = C_{dust}/C_{air}$, where C_{air} is the air concentration) is a function of the octanol-air partition coefficient (K_{oa}), we calculated $K_{PUF\text{-}air}$ ($= C_{PUF}/C_{air} = C_{PUF} \times K_{dust\text{-}air}/C_{dust}$) using direct measurements of C_{PUF} and C_{dust}. Then, we explored predictive relationships of $K_{PUF\text{-}air}$ with K_{oa} or vapor pressure (VP). Lastly, we calculated the holding capacity of couch PUF for SVOCs.

4.2. Methods

4.2.1. Participant recruitment and couch PUF sample collection

As part of an effort to examine the overall decrease of FR concentrations in household dust after replacing old couches (assumed to be the primary sources of FRs in participating homes) with new ones, we recruited 14 homes in San Jose, California in July 2016. On the day

of couch replacement, the study team purchased and delivered a new couch to 11 homes, collected a sample from the top to the bottom of the whole cushion of the old couch, and labeled top and bottom on it. In the laboratory, we cut it into a 1-inch square through-cut to obtain individual samples at each depth (i.e., top, top-middle, and middle from couch cushion surfaces facing outward). Because two couches were removed in two homes, we collected samples from both, bringing the total number of sample sets (top, top-middle, and middle) to 13.

All recruitment and data collection protocols were approved by the Institutional Review Board for the University of California at Davis (UC Davis). Participants provided informed consent before collection of any data.

4.2.2. Target compounds

We focused on SVOCs that are assumed not to originate from couch PUF but rather be absorbed by the PUF, and thus excluded flame retardants. We used a compound list developed through a previous project searching for widely-detected compounds in household dust (Moschet et al. 2018; Shin et al. 2020) and then analyzed for the compounds detected in our previous studies as described below. The selected compounds included ultraviolet (UV) filters, fragrances, and other ingredients of personal care products (PCPs); insecticide ingredients; and a variety of other compounds widely detected in homes (phenols, phthalates, other plasticizers, PAHs, and squalene found in the skin). The selected compounds represent a range of chemical properties, ranging from relatively volatile compounds (e.g., phenol) to those with a high affinity to dust (e.g., di(2-ethylhexyl) phthalate [DEHP]). Chemical properties and use categories of the studied compounds were listed in Table S4.1.

4.2.3. Sample analysis

At UC Davis, we quantified concentrations of 64 compounds in three sections (top, top-middle, and middle) of each couch foam sample. The foam samples were cut using a pre-cleaned cutter and subsections (approximately 100 mg) were sonication-extracted using hexane:acetone (3:1 v:v) followed by acetone (100%). The supernatant was collected and combined into an evaporation tube and evaporated to 1 mL under nitrogen using a Turbovap (Biotage). The extract was then filtered through a polytetrafluoroethylene filter (0.2 μm). Half of the 1 mL extract was spiked with internal standard dibromooctafluorobisphenol (DBOFB) and 1 μL was injected for analysis. An Agilent 7890B gas chromatograph using a HP-5MS (30 m × 0.25 m, 0.25 μm) column coupled to an Agilent 7200B gas chromatography quadrupole time-of-flight mass spectrometry (GC-QTOF-MS) was used to acquire samples in electron ionization (EI) mode. Samples were acquired using a 78-minute method with a linear temperature gradient from 35 °C to 325 °C. A 13-point calibration curve was used to quantify target analytes using the Agilent MassHunter Quantitative Analysis (version B.09).

4.2.4. Statistical analysis

We performed all statistical analyses using R version 3.6.1. For all analyzed compounds, we provided summary statistics of measured concentrations in PUF samples for each depth. For concentrations below the limit of detection (LOD), we assigned a value of the LOD divided by the square root of 2 (Hornung 1990). For concentrations between the LOD and the limit of quantification (LOQ), we assigned a value of the LOQ divided by 2.

For compounds detected in more than 50% of the PUF samples at all three depths, we used natural logarithm (ln)-transformed concentrations in all statistical analyses because

distributions of concentrations were right-skewed for these compounds. We normalized the top-middle and middle concentrations to the top concentrations and then examined concentration changes with depth. We also performed a paired t-test to compare mean PUF concentrations between two depths (i.e., top versus top-middle, top-middle versus middle, top versus middle). For compounds detected in more than 50% of the top PUF samples and dust samples collected in the same home (Kim et al. revision under review), we computed the Pearson correlation coefficients (r) between PUF (C_{PUF}) and dust concentrations (C_{dust}) and computed the ratio of C_{PUF} to C_{dust}. For two couches that were removed in two homes, we compared distributions of the measured PUF concentrations (ng/g of PUF) at each depth (i.e., top, top-middle, middle) to examine the effect of upholstery materials (e.g., leather, microfiber, velvet) on PUF concentrations. Couch age may affect PUF concentrations, but age information was not available for those couches in the two homes.

4.2.5. Calculation of PUF-air partition coefficients ($K_{PUF\text{-}air}$)

To calculate $K_{PUF\text{-}air}$, we used the compounds detected in more than 50% of both the top PUF samples and dust samples. Because $K_{PUF\text{-}air}$ (= $C_{PUF}/C_{air} = C_{PUF} \times K_{dust\text{-}air}/C_{dust}$) is a function of $K_{dust\text{-}air}$ (m^3 of air/mg of dust), we calculated $K_{dust\text{-}air}$ using the following relationship (Little et al. 2012; Weschler and Nazaroff 2010).

$$K_{dust-air} = \frac{C_{dust}}{C_{air}} = f_{om\_dust} \times K_{oa} \,/\, \rho_{dust}$$

where f_{om_dust} is the fraction of organic matter associated with settled dust (unitless), K_{oa} is the octanol-air partition coefficient (unitless), and ρ_{dust} is the density of settled dust (mg/m^3). We used 0.2 and 2×10^9 mg/m^3 for f_{om_dust} and ρ_{dust}, respectively, reported elsewhere (Little et al. 2012; Weschler and Nazaroff 2010) and chemical-specific K_{oa} values available in the U.S.

Environmental Protection Agency's CompTox Chemistry Dashboard (Williams et al. 2017). The unit of PUF concentration was converted from ng/g of PUF to ng/m^3 of PUF using the bulk density of PUF (2.2×10^3 g/m^3) (Zhao et al. 2004) before calculating $K_{PUF\text{-}air}$ (m^3 of air/m^3 of PUF). In addition, we regressed our calculated $K_{PUF\text{-}air}$ on K_{oa} or VP (in Pa) to formulate predictive equations for $K_{PUF\text{-}air}$.

Predictive equations of $K_{PUF\text{-}air}$ with K_{oa} or vapor pressure (VP) were developed in two chamber studies (Francisco et al. 2017; Zhao et al. 2004) and another indoor field study with purposefully-designed PUF samplers (Shoeib and Harner 2002a). The $K_{PUF\text{-}air}$ values in these three studies are considered to be "true" partition coefficients, because they were calculated after "effective" (or 95% of) equilibrium was established between concentrations in air and PUF under the tight control on sampling conditions. On the other hand, those values calculated in the current study are considered to be "apparent" partition coefficients, because we could not confirm whether our system was at equilibrium at the time of sampling. Although the $K_{PUF\text{-}air}$ values in the current study may not be directly comparable to those in those three studies, we compared them each other to observe potential differences in PUF's holding capacity between upholstered couch PUF and bare PUF.

4.2.6. Calculation of holding capacity for SVOCs

The fugacity capacity is defined as the holding capacity of a material for a chemical substance based on the properties of both the material and the chemical (Bennett and Furtaw 2004). Because little is known for holding capacity of couch PUF for SVOCs, we used the following relationship to calculate the fugacity capacity of couch PUF.

$$Z_{PUF} = \frac{K_{PUF-air}}{R \times T}$$

where Z_{PUF} is the fugacity capacity of couch PUF (mol/m^3·Pa), R is the ideal gas constant (8.314 Pa·m^3/mol·K), and T is the ambient temperature (298 K). Then, we calculated the amount of SVOCs sorbed in couch PUF by multiplying the fugacity capacity by the representative volume of couch PUF (1 m^3). To compare the mass distribution of SVOCs in couch PUF with other indoor compartments (i.e., air, carpet, vinyl flooring, wall), we also multiplied the fugacity capacity and the representative volume of each compartment. For the other indoor compartments, equations and parameters used in calculating the fugacity capacity and the amount of sorbed SVOCs are available in Tables S4.3, S4.4, and S4.5.

4.3. Results

4.3.1. Measured PUF concentrations

Among 64 studied compounds, 29 compounds were detected in at least one of the 39 PUF samples (Table 4.1 and Table S4.2). Among the 29 detected compounds, 15, 13, and 11 compounds were detected in more than 50% of the samples in the top, top-middle, and middle sections, respectively, and 11 compounds were detected in more than 50% of the samples at all three depths. For 26 out of the 29 detected compounds, detection frequency (%) was the highest at the top section. The highest geometric mean concentration at the top section was observed in acetyl tributyl citrate (7.9×10^4 ng/g of PUF), followed by phenol (3.3×10^4 ng/g of PUF), homosalate (1.2×10^4 ng/g of PUF), 2-benzylideneoctanal (9.9×10^3 ng/g of PUF), and galaxolide (7.7×10^3 ng/g of PUF).

Among 64 studied compounds, 35 compounds were not detected in any samples and 18 compounds were detected in less than 50% of the samples at least from one depth (Table S4.2). Most of the non-detected compounds were insecticide ingredients (i.e., 16 out of 20), phenols

(i.e., 13 out of 18), and PAHs (i.e., 5 out of 10). Among those detected in less than 50% of the dust samples in the same home (Kim et al. revision under review), all were not detected or detected in less than 50% of the PUF samples as well, except for phenol and 4-chloro-3-methylphenol. Among those detected in more than 50% of the dust samples, 7 compounds (i.e., benzyl butyl phthalate, di-n-octyl phthalate, acetyl tributyl citrate, bifenthrin, permethrin, octocrylene, squalene) were not detected or detected in less than 50% of the PUF samples.

Table 4.1. Summary of measured concentrations for 29 SVOCs detected in PUF samples (n = 39) and dust samples (n = 11).

Use category or chemical class	Compound name	Couch PUF concentration (ng/g of PUF)					Dust concentration (ng/g of dust)			Correlation coefficient (r) [c]	C_{PUF}/C_{dust} [d]
		LOD	DF (%) [a]			GM [b] (n =13)	LOD	DF (%)	GM (n = 11)		
			top	top-middle	middle						
Phthalates	Benzyl butyl phthalate	500	31	8	8	443	1,000	100	7,715	-	-
	Di(2-ethylhexyl) phthalate	1,000	92	92	85	1,447	10,000	91	66,778	0.10	0.02
	Di-isobutyl phthalate	250	100	69	31	1,710	1,000	100	8,883	0.71*	0.19
	Di-n-butyl phthalate	500	100	54	23	850	1,000	100	8,213	0.32	0.10
	Di-n-octyl phthalate	100	8	8	8	-	1,000	73	143	-	-
	Diethyl phthalate	50	100	100	100	574	100	100	1,890	0.20	0.30
	Dimethyl phthalate	10	100	100	100	30	50	100	122	0.78*	0.25
Other plasticizers	Acetyl tributyl citrate	10,000	100	23	8	79,029	10,000	100	2.1×10^7	0.20	0.004
	Bis(2-ethylhexyl) adipate	1,000	8	8	8	-	10,000	55	13,088	-	-
Insecticide ingredients	Bioallethrin	100	46	8	0	299	100	0	-	-	-
	Esfenvalerate	100	23	15	15	-	500	0	354	-	-
	Permethrin	250	31	31	31	390	100	91	1,031	-	-
	Phenothrin	50	8	0	0	46	100	9	46	-	-
Phenols	2-Nitrophenol	100	0	8	8	-	1,000	0	-	-	-
	4-Chloro-3-methylphenol	100	100	100	85	7,186	1,000	0	-	-	-
	Cresol (o, m)	100	92	92	100	168	500	0	-	-	-
	para-Cresol	1,000	23	15	15	-	2,500	73	1,663	-	-
	Phenol	250	100	100	100	32,832	1,000	91	910	-0.12	36.1
Polycyclic aromatic hydrocarbons	Anthracene	10	23	8	8	-	100	0	71	-	-
	Benzo(a)anthracene	10	0	8	8	-	100	36	85	-	-
	Fluorene	10	46	46	46	52	100	9	-	-	-
	Phenanthrene	10	100	85	54	226	100	100	268	0.05	0.84
	Pyrene	5	100	46	38	123	50	100	168	-0.10	0.73
Fragrance ingredients	2-Benzylideneoctanal	100	100	100	85	9,850	500	100	2,062	0.46	4.78
	Galaxolide	5	100	100	100	7,712	50	100	1,363	0.66*	5.66
	Tonalide	10	100	92	77	1,092	100	91	324	0.51	3.37
Ultraviolet filters	Homosalate	25	100	100	92	12,041	250	100	2,416	0.91*	4.98
	Octocrylene	100	23	0	0	-	100	100	6,624	-	-

Skin oils	Squalene	1,000	46	15	0	3,975	1,000	100	71,984	-	-

[a] Detection frequency (%) at each depth (i.e., top, top-middle, middle) from 13 PUF samples each.

[b] Calculated with 13 PUF samples at the top section only.

[c] Pearson correlation coefficients (r) of ln-transformed concentrations between PUF (C_{PUF}) and dust (C_{dust}) for 13 compounds detected in more than 50% of the top PUF samples and dust samples. P-values < 0.05 were marked with asterisk (*).

[d] Ratios of couch PUF concentrations (C_{PUF}) to dust concentrations (C_{dust}).

Abbreviation: limit of detection (**LOD**), detection frequency (**DF**), geometric mean (**GM**), ortho-Cresol + meta-Cresol (**Cresol (o, m)**)

4.3.2. Changes in PUF concentrations inside couch cushions

Among the 11 compounds detected in more than 50% of the samples at all three depths, the geometric mean PUF concentrations of 6 compounds (i.e., diethyl phthalate (DEP), phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide and homosalate) decreased from top to middle, whereas those of ortho-cresol + meta-cresol [cresol (o, m)] increased from top to middle (Figure 4.1). The geometric mean concentrations of the other 4 compounds did not decrease or increase with depth. Similarly, the mean concentrations of 5 compounds (i.e., phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate) were different between two depths (p-value < 0.05) and decreased from top to middle (Figure 4.2). Cresol (o, m) had lower mean PUF concentrations in the top section than in the middle section, and its difference between two depths was borderline significant (p-value = 0.06). For PUF concentrations measured in two different couches in the same home, they were within one order of magnitude between couches in the top section for most compounds (Figure 4.3). Compared to the top section, the differences of the measured PUF concentrations between two couches were higher in the top-middle and middle sections for most compounds. In one of the two homes (Home B in Figure 4.3), concentrations were higher in one couch than the other for most compounds.

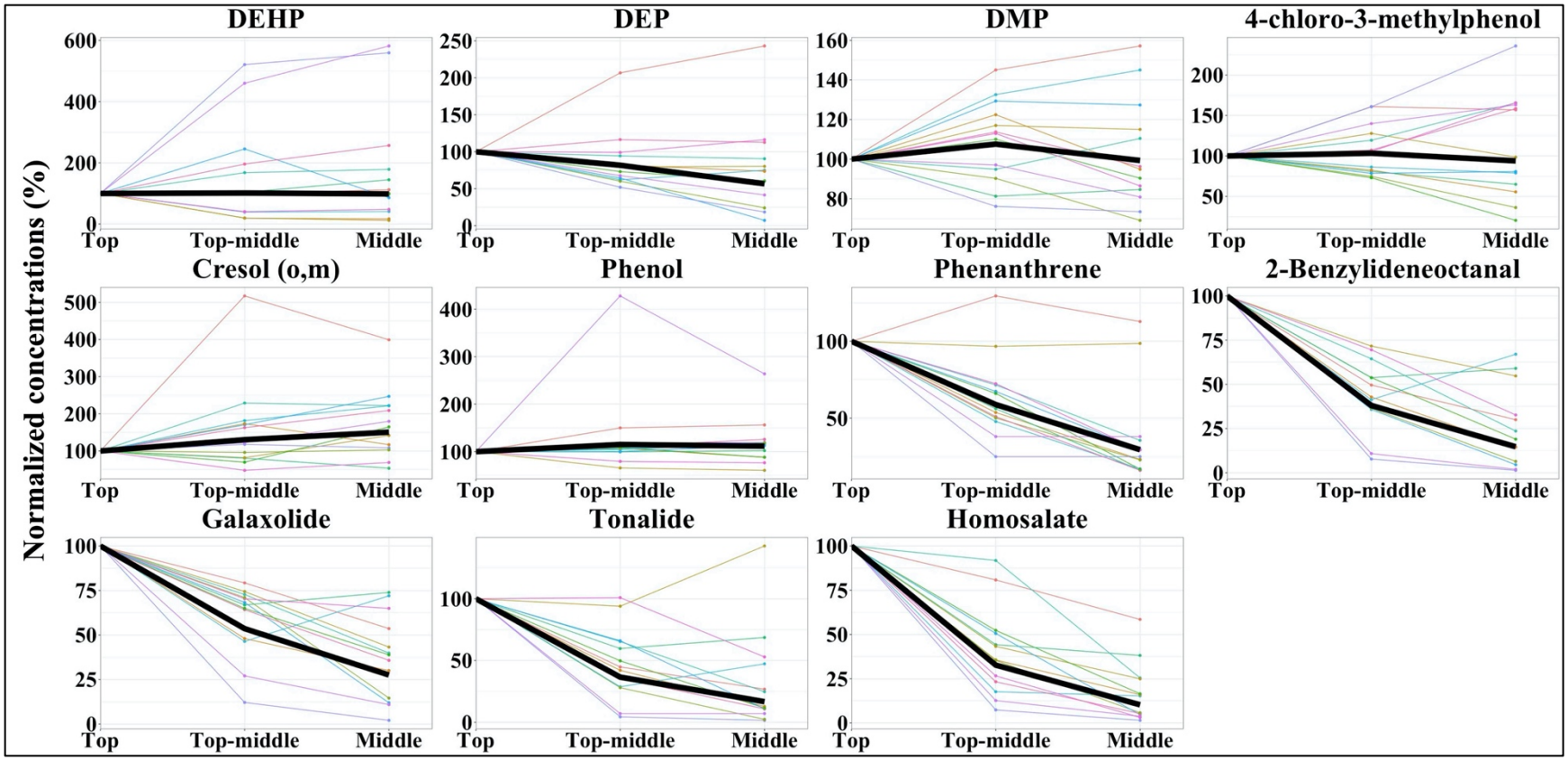

Figure 4.1. Measured PUF concentrations at three depths of couch cushions (i.e., top, top-middle, middle) after normalizing the top-middle and middle concentrations to the top concentrations. Black lines in bold represent normalized geometric mean concentrations.

Abbreviation: di(2-ethylhexyl) phthalate (**DEHP**), diethyl phthalate (**DEP**), dimethyl phthalate (**DMP**), ortho-cresol + meta-cresol (**Cresol (o, m)**)

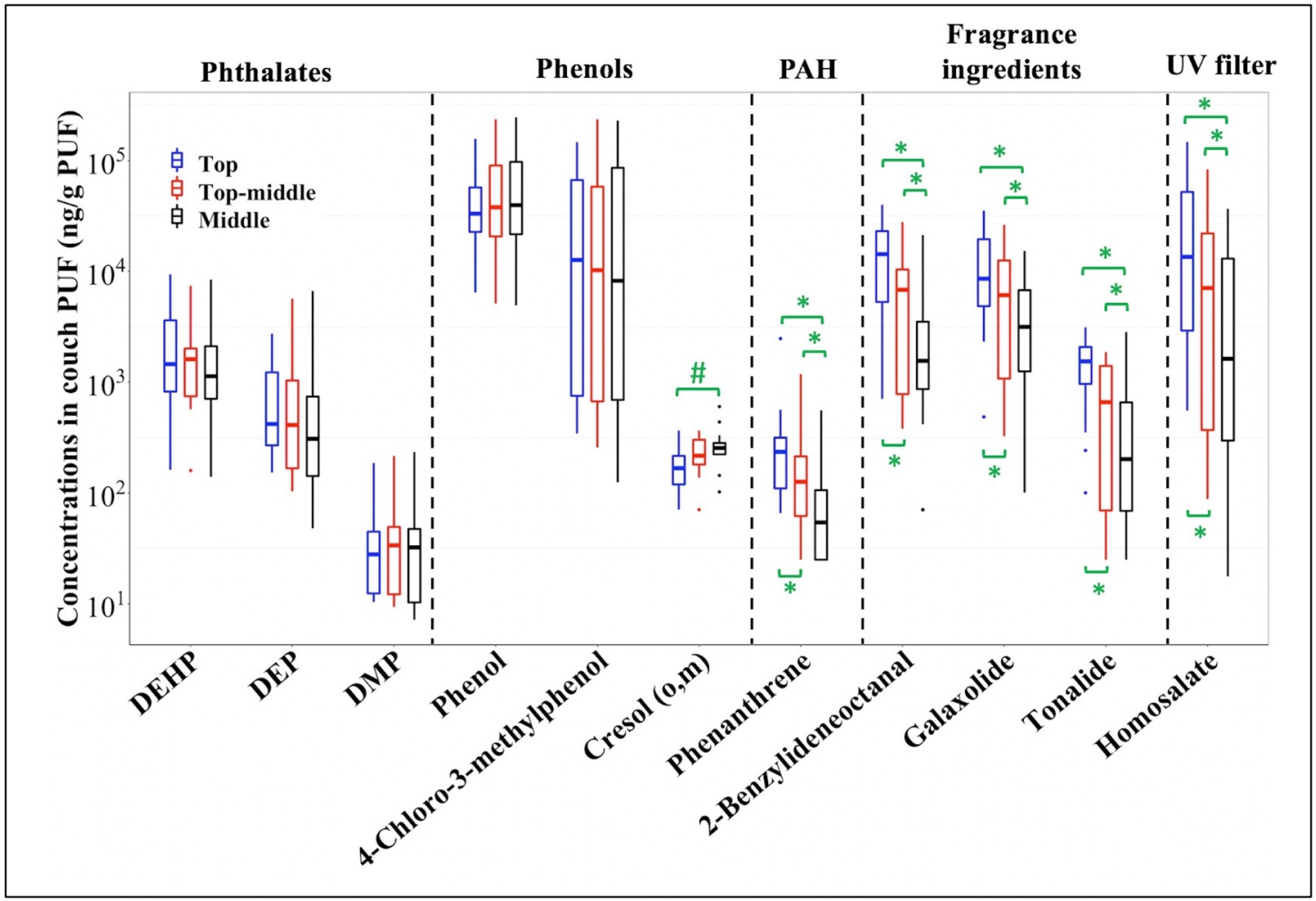

Figure 4.2. Distributions of the measured concentrations in couch PUF samples (n = 13 for each depth) for 11 compounds detected in more than 50% of the samples at all three depths (top, top-middle, middle). P-values < 0.05 were marked with asterisk (*). P-value of cresol (o, m) was 0.06 between top and middle, marked with asterisk (#).

Abbreviation: di(2-ethylhexyl) phthalate (**DEHP**), diethyl phthalate (**DEP**), dimethyl phthalate (**DMP**), polycyclic aromatic hydrocarbons (**PAHs**), ultraviolet (**UV**), ortho-cresol + meta-cresol (**Cresol (o, m)**)

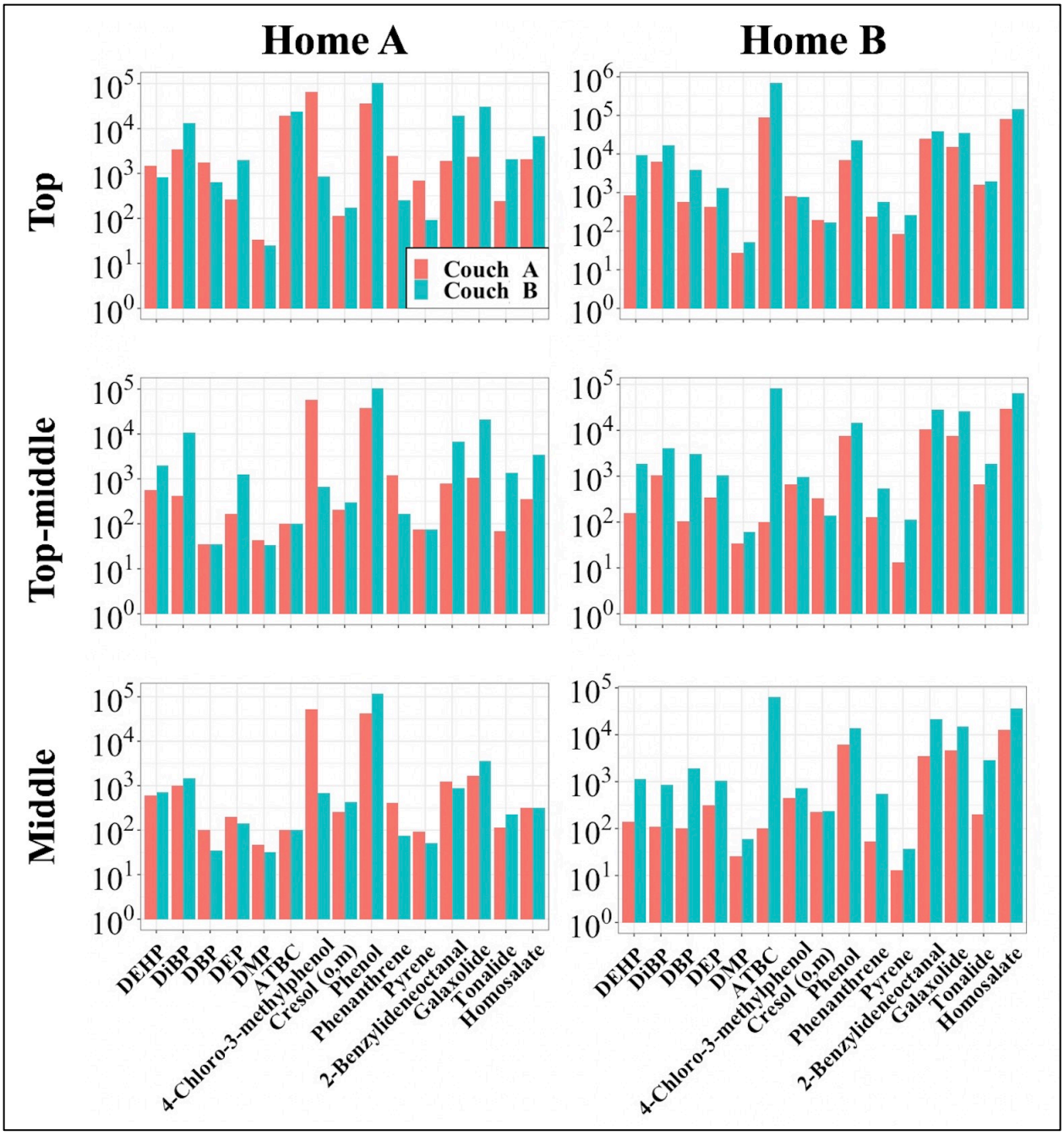

Figure 4.3. Distributions of the measured PUF concentration (ng/g of PUF) at each depth (i.e., top, top-middle, middle) between two different couches in the same home (the first column for Home A and the second column for Home B).

4.3.3. Association between PUF and dust concentrations

Among the 13 compounds detected in more than 50% of both the top PUF samples and dust samples, 4 compounds (i.e., di-isobutyl phthalate [DiBP], dimethyl phthalate [DMP], galaxolide, homosalate) showed statistically significant positive correlations between the two media (r = 0.66-0.91, p-value < 0.05, Table 4.1). Except for phenol and pyrene, correlation coefficients for the other compounds (i.e., DEHP, di-n-butyl phthalate [DBP], diethyl phthalate [DEP], acetyl tributyl phthalate, phenanthrene, 2-benzylideneoctanal, tonalide) were generally positive, but not statistically significant (r = 0.05-0.51, p-value > 0.05). For the same 13 compounds above, the ratios of C_{PUF} to C_{dust} varied by use category or chemical class. The ratios ranged from 0.004 to 0.30 for 5 phthalates and 1 other plasticizer and ranged from 3.4 to 5.7 for four skin applied compounds (3 fragrance ingredients and 1 UV filter). Phenol and 4-chloro-3-methylphenol had the highest ratio (= 36.1) and the second highest ratio (= 10.2), respectively, among all detected compounds.

4.3.4. Calculated $K_{PUF\text{-}air}$

Among the 13 compounds detected in more than 50% of both the top PUF samples and dust samples, calculated log $K_{PUF\text{-}air}$ values varied from 2.46 (DMP) and 7.80 (homosalate) (Table 4.2). The calculated log $K_{PUF\text{-}air}$ values increased with increasing log K_{oa} values (log $K_{PUF\text{-}air}$ = 0.64 log K_{oa} + 0.01, r^2 = 0.62) (Figure 4.4). The slopes of the regression equations and r^2 values increased when regressing by individual use categories (e.g., r^2 = 0.92 for 6 phthalates/ plasticizers and r^2 = 0.99 for 4 skin-applied compounds such as fragrance ingredients and UV filters) (Figure S4.1). Our regression slope for all 13 compounds was similar to those obtained in two previous studies: 0.64 for the current study, 0.64 for Shoeib and Harner (2002), and 0.66 for Francisco et al. (2017). Our calculated log $K_{PUF\text{-}air}$ values were on the same magnitude of those

of other studies for 4 skin-applied compounds (3 fragrance ingredients and 1 UV filter) but were approximately 1 to 3 orders of magnitude smaller than those of the other studies for other compounds (Table 4.2). On the other hand, the calculated log $K_{PUF\text{-}air}$ values decreased with increasing log VP values (log $K_{PUF\text{-}air}$ = -0.60 log VP + 3.98, r^2 = 0.47) (Figure 4.4). Our calculated log $K_{PUF\text{-}air}$ values were approximately 1 to 3 orders of magnitude lower than those of Zhao et al. (2004), except for phenol, 2-benzylideneoctanal, galaxolide, and homosalate.

Table 4.2. Two key chemical properties related to the partitioning between PUF and air, and calculated log $K_{PUF\text{-}air}$ values from this current study and those estimated in other studies using regression equations with the octanol-water partition coefficient (K_{oa}) or vapor pressure (VP) for 13 compounds detected in more than 50% of the top PUF samples and dust samples. All $K_{PUF\text{-}air}$ values are expressed in the unit of m^3 of air/m^3 of PUF.

Compound name	log K_{oa} (unitless)	-log VP (Pa)	log $K_{PUF\text{-}air}$			
			This study [a]	Shoeib and Harner [b]	Francisco et al. [c]	Zhao et al. [d]
Phthalates and other plasticizers						
Dimethyl phthalate	5.68	0.08	2.46	4.77	5.03	5.10
Diethyl phthalate	6.75	0.91	3.45	5.45	5.73	5.88
Di-isobutyl phthalate	8.21	2.37	4.72	6.38	6.69	7.24
Di-n-butyl phthalate	8.84	2.46	5.13	6.78	7.11	7.31
Acetyl tributyl citrate	10.34	3.67	5.10	7.73	8.09	8.44
Di(2-ethylhexyl) phthalate	11.69	4.42	7.35	8.59	8.98	9.14
Phenols and Polycyclic aromatic hydrocarbons						
Phenol	4.80	-1.30	3.63	4.21	4.45	3.82
Phenanthrene	7.57	1.29	4.77	5.97	6.27	7.06
Pyrene	8.80	2.24	6.00	6.75	7.08	9.62
Fragrance ingredients and Ultraviolet filters						
2-Benzylideneoctanal	7.38	1.02	5.43	5.85	6.15	5.98
Tonalide	7.93	2.24	5.94	6.20	6.51	7.11
Galaxolide	8.18	1.29	6.40	6.36	6.67	6.23
Homosalate	9.71	3.22	7.80	7.33	7.68	8.03

[a] Calculated using the direct measurements of C_{PUF} and C_{dust}.
[b] Estimated from a regression equation: $\log K_{PUF\text{-}air} = 0.64 \log K_{oa} + 1.15$ (Shoeib and Harner 2002a)
[c] Estimated from a regression equation: $\log K_{PUF\text{-}air} = 0.66 \log K_{oa} + 1.30$ (Francisco et al. 2017)
[d] Estimated from a regression equation: $\log K_{PUF\text{-}air} = -0.93 \log VP + 5.03$ (Zhao et al. 2004)

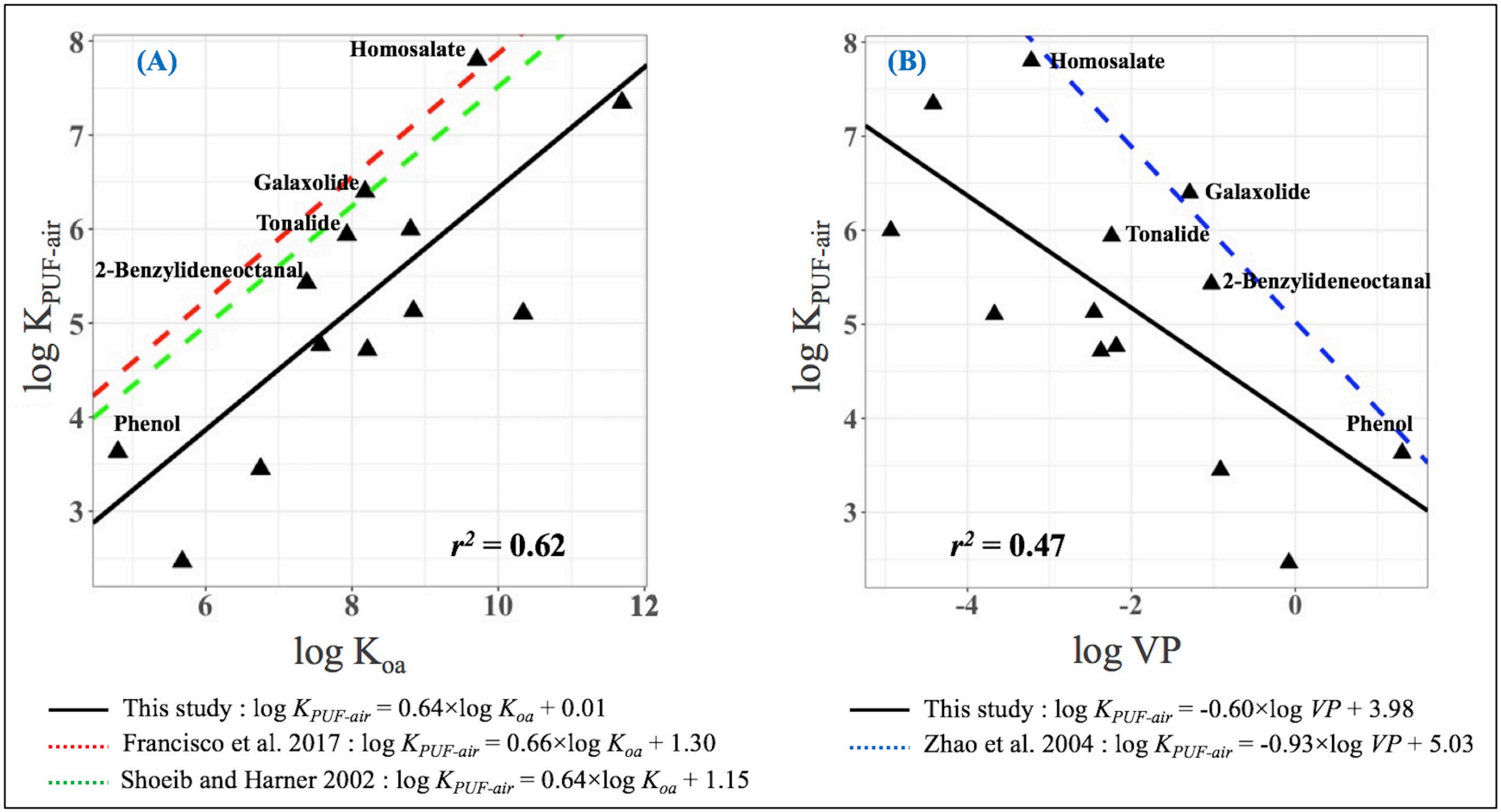

Figure 4.4. Calculated median log $K_{PUF-air}$ versus log K_{oa} (A) or vapor pressure (B) for 13 compounds detected in more than 50% of the top PUF samples and dust samples. Regression equations from other three studies are shown in dotted lines. Compound names shown in the plots are fragrance ingredients, UV filters or phenol. All $K_{PUF-air}$ values are expressed in the unit of m^3 of air/m^3 of PUF.

4.3.5. Holding capacity of indoor compartments for SVOCs

Among 13 compounds for which $K_{PUF\text{-}air}$ values were calculated in this study, the fugacity capacity of couch PUF (mol/m^3·Pa) ranged from 1.2×10^{-1} (DMP) to 2.6×10^4 mol/m^3·Pa (Homosalate) (Table S4.6). The fugacity capacity of couch PUF was relatively high for 4 skin-applied compounds (3 fragrance ingredients and 1 UV filter), compared to other chemical classes or use categories. The fugacity capacity of couch PUF was 3 to 8 orders of magnitude higher than that of air for all compounds and was up to 3 orders of magnitude lower than that of carpet and vinyl flooring for most compounds, except for galaxolide and homosalate. However, when comparing the mass distribution of SVOCs among five indoor compartments (i.e., couch PUF, air, carpet, wall, vinyl flooring), more than 20% of the total mass is distributed to couch PUF for phenol, 2-benzylideoneoctanal, galaxolide, and homosalate while less than 10% of the total mass is distributed to couch PUF for the other compounds (Figure 4.5).

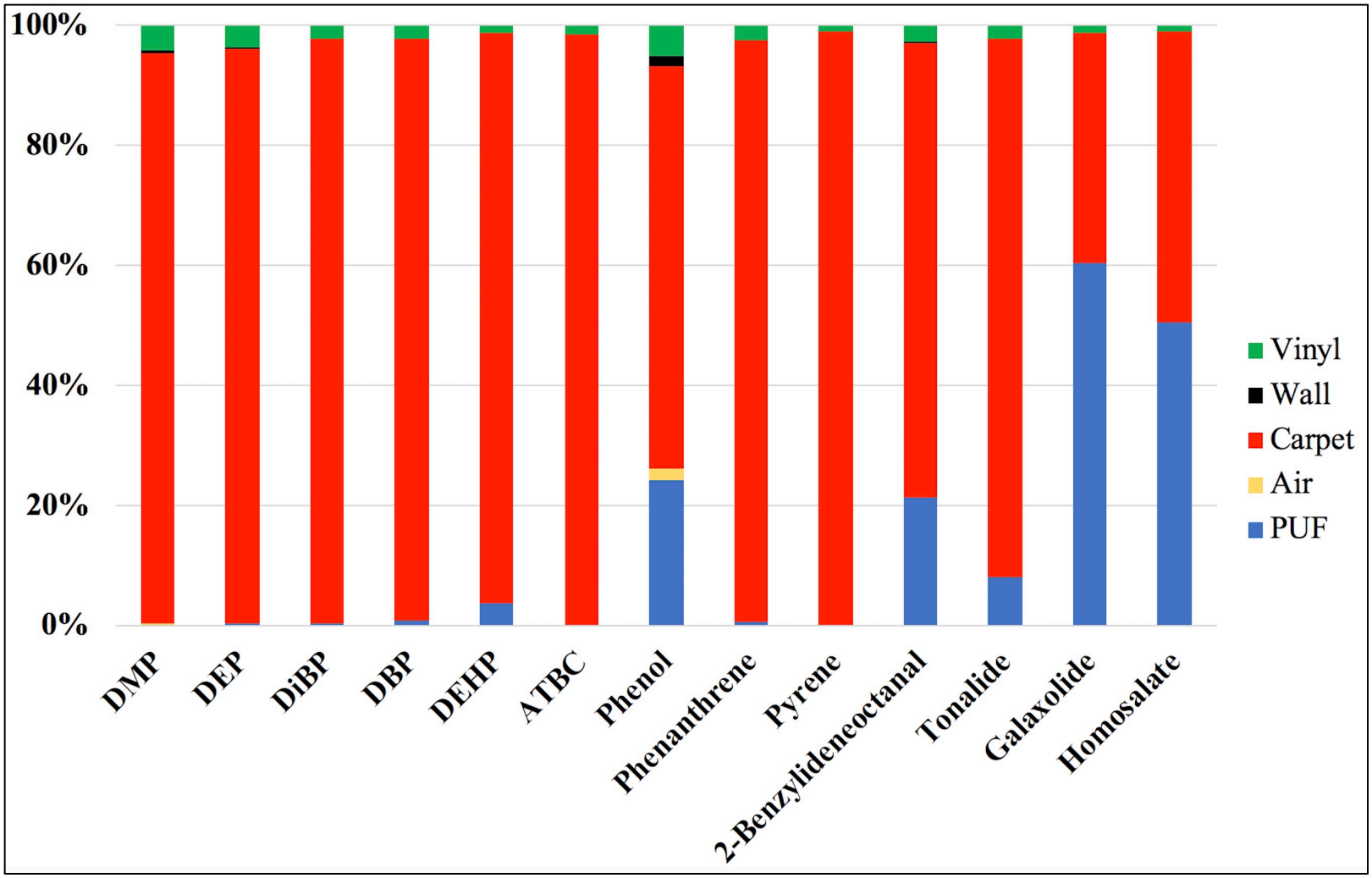

Figure 4.5. Mass distribution (%) of compounds among five indoor compartments.

4.4. Discussion

To examine whether couch foam can be used as a passive sampler for SVOCs commonly detected in the home and to evaluate the potential for the couch to serve as a sink for compounds in the home, we measured their concentrations in couch PUF samples at varying depths and examined concentration changes with depth. Among 64 studied compounds, 29 compounds were detected in at least one of the 39 samples, and 11 compounds were detected in more than 50% of the samples at all three depths. Among the 11 compounds, we observed that concentrations of 5 compounds decreased with depth. Among the 13 compounds detected in more than 50% of both the top PUF samples and dust samples, concentrations of 4 compounds showed positive correlation coefficients between the two media (p-value < 0.05). We observed that our calculated $K_{PUF\text{-}air}$ increased with K_{oa} and decreased with VP, and that K_{oa} (r^2 = 0.62) was a stronger predictor of $K_{PUF\text{-}air}$ than VP (r^2 = 0.47). Couch PUF had relatively higher holding capacity than air for all compounds but lower holding capacity than carpet and vinyl flooring for most compounds. However, for four compounds (i.e., phenol, 2-benzylideoneoctanal, galaxolide, and homosalate) whose predicted $K_{PUF\text{-}air}$ values in our study were similar to those in other chamber studies and that were measured in relatively high concentrations in couch PUF (GM > 7 μg/g of PUF, Table 1), more than 20% of the total mass in five indoor compartments was distributed to couch PUF.

We evaluated the potential utility of couch PUF as a passive sampling medium for indoor SVOCs by comparing our $K_{PUF\text{-}air}$ values calculated with direct concentration measurements in couch PUF with those measured or calculated in two chamber studies under controlled laboratory conditions (Francisco et al. 2017; Zhao et al. 2004) or in an indoor field study with purposefully-designed PUF air samplers (Shoeib and Harner 2002a). We observed that for most compounds, our calculated $K_{PUF\text{-}air}$ values were approximately 1 to 3 orders of magnitude

smaller than those estimated using predictive relationships developed in other three studies. One potential reason for these discrepancies is that equilibrium might not be reached between PUF and air in our system. The increasing or decreasing concentration profiles in the cushion (Figure 4.1) are a sign that for many of the compounds, equilibrium was not reached between PUF and air or inside the cushion. Interestingly, for four compounds (3 fragrance ingredients and 1 UV filter) that people "wear" or apply on the skin, our calculated $K_{PUF\text{-}air}$ values were on the same order of magnitude of those of other three studies (Figure 4.1). It is likely that the PUF-air system of these skin-applied compounds were closer to equilibrium than that of other compounds that people mostly do not wear or apply on the skin such as phthalates or PAHs. Other possible reasons include the effect of upholstery and other indoor sinks or removals. For example, mass-transfer of SVOCs from indoor air to couch PUF may be limited by couch upholstery because it acts as resistance to mass-transfer between air and PUF (Klopffer and Flaconneche 2001). In addition, compared to chamber studies where PUF competes for SVOCs primarily with air, couch PUF in our study competes with other indoor sinks (e.g., airborne particles, dust, surface film, and other indoor surfaces) (Little et al. 2012; Weschler and Nazaroff 2008, 2010, 2017) and removals (e.g., ventilation, vacuuming and surface cleaning) (Shin et al. 2012; Shin et al. 2013).

Among the 13 compounds detected in more than 50% of both the top PUF samples and dust samples, we observed positive correlation coefficients between the two media for DMP, DiBP, galaxolide, and homosalate (p-value < 0.05). DMP, with the lowest log K_{oa} value (= 5.68) among the five phthalates (i.e., DEHP, DiBP, di-n-butyl phthalate, DEP, DMP), had the highest correlation coefficient (r = 0.78). Its low K_{oa} value may accelerate mass-transfer between air and dust and between air and PUF (Dodson et al. 2015; Schripp et al. 2010). Because two fragrance ingredients (galaxolide and homosalate) are likely to be transferred (1) to couch PUF via direct

skin contact with couch surfaces and (2) to carpet dust via skin flaking (Weschler et al. 2011), it is likely that we observed positive correlation coefficients of these two compounds. For four skin applied compounds (3 fragrance ingredients and 1 UV filter), we observed relatively high ratios of C_{PUF} to C_{dust} from 3.4 to 5.7, implying high transfer rates of these compounds from skin to couch PUF. On the other hand, the other 9 compounds did not show statistically significant correlation coefficients, potentially due to high within-home variability of dust concentrations of these compounds or a small sample size. For example, the correlation coefficients were relatively small for phenanthrene and pyrene (r = 0.05, -0.10, respectively), presumably because of the seasonal variations in source rates of indoor PAHs including increased infiltration of outdoor air during summer and increased residential heating such as wood burning during winter.

Our study showed that PUF in upholstered home couches can absorb SVOCs because a wide range of SVOCs which are not known to be added to couch PUF were detected in our samples, including phthalates, other plasticizers, insecticide ingredients, PAHs, fragrance ingredients, UV filters, and skin oils (Table 4.1). We also observed the highest detection frequency in the top section for most compounds, supporting that home couches absorb SVOCs from air. Among the 11 compounds detected in more than 50% of the samples at all depths, concentrations of phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate decreased with depth (Figures 4.1 and 4.2), showing potential inward diffusion of these compounds from air to couch PUF. We also observed that phenol and three skin-applied compounds were distributed to couch PUF with relatively high percentages (21.2 – 60.5%) among five indoor compartments (Figure 4.5). As these compounds were also measured in relatively high concentrations in couch PUF, it may act as an important holding reservoir for these compounds. Moreover, we found that SVOC concentrations in PUF were different between

two couches placed in the same home (Figure 4.3). The reason for this difference is less clear but includes the differences in age of couches or materials of upholstery (Boor et al. 2015; Keimowitz et al. 2016). The age of couches in the home was not available because residents obtained used ones, but upholstery materials for two couches were microfiber and velvet, respectively. However, the role of couch PUF as a sink for SVOCs should be interpreted with caution because the fugacity capacity of couch PUF is several orders of magnitude lower than that of carpet and dust for most compounds (Table 4.3). We also observed that seven compounds, detected in more than 50% of the dust samples collected in the same home, were not detected or detected in less than 50% of the PUF samples. Because these seven compounds have relatively high K_{oa} values (e.g., log $K_{oa} > 9.5$), it is likely that compounds with high K_{oa} values may not be absorbed by the PUF well enough to be detected in the PUF.

Our study also showed that PUF in home couches may release some phenolic compounds including phenol, 4-chloro-3-methylphenol, and cresol (o, m). For example, among the same 11 compounds above, phenol had the highest geometric mean PUF concentration (32,832 ng/g of foam) and a relatively low geometric mean dust concentration (910 ng/g of dust) (Table 4.1), resulting in the highest ratio of C_{PUF} to C_{dust} (= 36.1). The geometric mean PUF concentration of 4-chloro-3-methylphenol (7,186 ng/g of foam) was the fifth highest, whereas it was detected in none of our dust samples. Furthermore, we observed a decreasing trend in PUF concentrations of cresol (o, m) from the middle section to the top section (p-value = 0.06, Figure 4.2). These showed potential outward diffusion of these phenolic compounds from PUF to indoor air.

We noted two major strengths of this study. Because upholstery may act as resistance between air and PUF, direct concentration measurements in upholstered couch PUF allowed us to observe differences in PUF's holding capacity between upholstered couch PUF and a bare

PUF. In addition, because a wide range of SVOCs are detected in indoor environmental media (e.g., airborne particles, dust, indoor surface), our novel sampling of upholstered couch PUF and subsequent chemical analysis helped understand its role on the absorption and release of SVOCs. However, some limitations should be noted. First, we did not differentiate the effect of different upholstery materials (e.g., leather, fabric) on couch PUF's holding capacity due to the limited sample size. Second, our predictive equations for $K_{PUF\text{-}air}$ versus K_{oa} may not be applicable to all SVOCs, because when regressing by individual use categories or chemical classes, the slope of $K_{PUF\text{-}air}$ against K_{oa} was approximately 30% steeper for skin-applied compounds than plasticizers. It is likely that these skin-applied compounds were primarily transferred to couch PUF via direct skin contact or skin flaking rather than molecular diffusion or deposition from air to PUF. Third, because only 4 out of the 11 compounds were correlated between the top PUF concentrations and dust concentrations, our calculated $K_{PUF\text{-}air}$ using C_{PUF} and C_{dust} may not be valid for those that were not correlated each other. Last, because of a small sample size and low detections in the current study, our predictive equations should be interpreted with caution.

CHAPTER 5: SUMMARY AND CONCLUSIONS

5.1. Summary of findings

In Chapter 2, I examined temporal trends and determinants of PFAS serum concentrations among mothers with a young child. Geometric mean concentrations of perfluorooctanoate (PFOA), perfluorooctane sulfonate (PFOS), and perfluorohexane sulfonate (PFHxS) decreased over the study period [percent change (95% confidence interval): −10.7% (−12.7%, −8.7%); −10.8% (−12.9%, −8.5%); −8.0% (−10.5%, −5.5%), respectively]. On the other hand, perfluorononanoate (PFNA) and perfluorodecanoate (PFDA) showed mixed time trends. Among the selected covariates, longer breastfeeding duration was associated with decreased maternal serum concentrations of PFOA, PFOS, PFHxS, PFNA and PFDA.

In Chapter 3, I examined temporal variability of dust concentrations and factors affecting dust concentrations for a wide range of SVOCs. Among 26 compounds that were detected in more than 50% of the samples at all three visits, 20 compounds had ICCs above 0.50 and 6 compounds had ICCs below 0.50. For 19 out of 26 compounds, correlation coefficients between spring and fall (r = 0.48-0.98) were higher than those between summer and winter (r = 0.09-0.92), implying seasonal effects on dust concentrations.

In Chapter 4, I evaluated the potential utility of couch PUF as a passive sampler for SVOCs. Among 29 detected compounds, 11 compounds were detected in more than 50% of the samples at all depths. Among the 11 compounds, concentrations of phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate decreased with depth. Among the studied SVOCs, calculated log $K_{PUF\text{-}air}$ values varied from 2.46 (dimethyl phthalate) to 7.80 (homosalate), and K_{oa} (r^2 = 0.62) was a stronger predictor of $K_{PUF\text{-}air}$ than VP (r^2 = 0.47).

5.2. Conclusions

Chapter 2 reported time trends in PFAS serum concentrations among 450 Northern California women with a young child between 2009 and 2016. Maternal serum concentrations of PFOA, PFOS, and PFHxS monotonically decreased during the study period, while those of PFNA and PFDA exhibited mixed trends. The current study also found that longer breastfeeding duration was associated with decreased maternal serum concentrations of PFOA, PFOS, PFHxS, PFNA, and PFDA, adding to the existing evidence that lactation is a major excretion route of the common long-chain PFAS for nursing mothers.

In Chapter 3, I observed relatively low temporal variability of dust concentrations for most SVOCs with higher temporal variability for some specific SVOCs, implying that dust concentrations may vary over time for SVOCs with seasonal variations in product use, or for those with significant outdoor sources that change by ventilation rates.

Chapter 4 showed that couch PUF can absorb many SVOCs. However, because most SVOCs were less frequently detected in couch PUF and had low correlation coefficients between concentrations in dust and PUF, couch PUF may not be an effective passive sampling medium for SVOCs. I also found that couch PUF is an important holding reservoir for compounds whose predicted $K_{PUF\text{-}air}$ values in our study were similar to those in other chamber studies and that were measured in relatively high concentrations in couch PUF.

5.3. Implications of the research

In Chapter 2, the decreasing trends of PFOA, PFOS, and PFHxS observed in my study are consistent with those of other studies reported for other U.S. populations. The results of these studies and my study suggest that serum concentrations of PFOA, PFOS, and PFHxS are

decreasing in the U.S. general population. This reflects regulatory and voluntary efforts in restricting the production, use, and emissions of these compounds.

In Chapter 3, I expect that my findings may support future work that includes dust collection because understanding temporal variability of dust concentrations is an important step for developing optimal sampling strategies. For example, for compounds with low temporal variability in dust concentrations over time, dust concentrations collected from a single visit may represent average exposure. On the other hand, for compounds with high temporal variability in dust concentrations, future studies may need to collect multiple dust samples to account for temporal variability to minimize bias in estimating average exposure from dust concentrations.

In Chapter 4, because upholstery may act as resistance between air and PUF, direct concentration measurements in upholstered couch PUF allowed me to observe differences in PUF's holding capacity between upholstered couch PUF and a bare PUF. I also observed that phenolic compounds and skin-applied compounds were detected higher in couch PUF than in dust (i.e., a ratio of C_{PUF}/C_{dust} was larger than 3.0), implying that direct skin contact with couch surfaces or inhalation while sitting on couches might be an important exposure route of these compounds, particularly for infants and young children.

5.4. Recommendations for future studies

In Chapter 2, in addition to the compounds detected in our serum samples, other 8 PFAS were detected in California residential indoor dust (Shin et al. 2020). It is known that thousands of PFAS are, or have been, on the global market (Wang et al. 2017). Since the common long-chain PFAS, including PFOA, PFOS, and PFHxS, were phased out, there must be increasing trends in the production of replacement to these long-chain PFAS or novel PFAS. In addition, there are many overlooked compounds that are structurally similar to PFOS, PFOA, or their

precursors, and are produced in high volumes (e.g., >10 tones per year, see references 6 to 9 in Wang et al. 2017). Because of structural similarity among many PFAS, future biomonitoring studies may need to include a large number of PFAS that have been detected in environmental samples, food, or consumer products.

In Chapter 3, further studies are needed to confirm my findings of temporal variability in SVOC dust concentrations by comparing exposures reconstructed from dust concentrations and those inferred from biomarkers. In addition to sampling seasons, because I observed that dust concentrations of some SVOCs were associated with household characteristics, future studies need to control relevant household characteristics when examining temporal variability of dust concentrations for these SVOCs.

In Chapter 4, additional studies are needed to examine the effect of various upholstery materials on couch PUF's holding capacity as different upholstery materials (e.g., fabric, leather) may have different degrees of resistance of mass-transfer between air and PUF. More studies are also recommended to analyze SVOCs on the surface of PUF-based home furniture with frequent skin contact, especially for compounds to which skin contact with furniture surface could be a significant exposure route.

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APPENDIX: SUPPLEMENTAL MATERIAL

Table S2.1. Estimated average annual percent changes (aAPC) in PFAS maternal serum concentrations (2009-2016) and results from unadjusted and adjusted regression models.

Compound	Time variable	Unadjusted					Adjusted				
		β [a]	SE_β [b]	p-value [c]	r^2	aAPC [d]	β [a]	SE_β [b]	p-value [c]	r^2	aAPC [d]
PFOA	year	-0.110	0.013	< 0.01	0.15	-10.5	-0.114	0.012	< 0.01	0.28	-10.7
PFOS	year	-0.105	0.013	< 0.01	0.13	-10.0	-0.114	0.013	< 0.01	0.22	-10.8
PFHxS	year	-0.075	0.014	< 0.01	0.06	-7.0	-0.084	0.014	< 0.01	0.15	-8.0
PFNA	year	-0.039	0.017	0.02			-0.037	0.017	0.03		
	$year^2$	-0.033	0.008	< 0.01	0.03	-0.6	-0.034	0.008	< 0.01	0.07	-0.2
PFDA	year	0.029	0.018	0.10			0.022	0.018	0.21		
	$year^2$	0.018	0.008	0.03	0.01	1.1	0.013	0.008	0.12	0.06	0.9

[a] β is the regression coefficient of the time variables (year and $year^2$) in the regression models.

[b] SE_β is the standard error of β.

[c] p-value of the null hypothesis that β equals to zero.

[d] aAPC: average annual percent change [%/year].

Table S3.1. Chemical properties of 47 detected compounds and summary of quality control.

Chemical class	Compounds	Chemical properties [a]					1st visit		2nd and 3rd visits	
		MW (g/mole)	H ($atm \cdot m^3$/mole)	VP (mmHg)	log K_{oa} [b]	log K_{ow} [b]	Absolute Recovery (%)	Precision (%)	Absolute Recovery (%)	Precision (%)
Phthalates	Benzyl butyl phthalate	312	2.8E-08	4.3E-06	9.83	4.73	116	4	-	13
	Di(2-ethylhexyl) phthalate	391	2.6E-07	2.9E-07	11.69	7.60	-	-	-	6
	Di-isobutyl phthalate	278	3.2E-07	3.2E-05	8.21	4.11	93	6	44	9
	Di-n-butyl phthalate	278	1.2E-06	2.6E-05	8.84	4.51	97	2	-	5
	Di-n-octyl phthalate	391	2.6E-07	4.0E-07	11.74	8.10	57	1	50	3
	Diethyl phthalate	222	2.4E-08	9.2E-04	6.75	2.42	73	7	45	3
	Dimethyl phthalate	194	9.4E-08	6.3E-03	5.68	1.60	86	15	48	5
Other plasticizers	Acetyl tributyl citrate	402	3.0E-08	1.6E-06	10.34	2.49	-	8	-	9
	Bis(2-ethylhexyl) adipate	371	4.1E-07	2.6E-06	10.86	6.85	89	13	61	-
	Dioctyl terephthalate	391	2.6E-07	4.3E-07	11.69	7.70	102	4	-	14
Insecticides	Bifenthrin	423	4.9E-08	1.2E-07	10.47	6.79	90	-	53	-
	Chlorpyrifos	351	2.1E-06	3.1E-05	10.57	4.96	85	14	48	22
	Cyhalothrin	450	1.2E-08	2.1E-09	11.69	6.80	118	-	52	-
	Cypermethrin	416	2.0E-09	1.1E-09	11.70	6.60	136	-	62	-
	Deltamethrin	505	1.8E-09	1.6E-09	11.69	6.20	72	-	37	-
	Esfenvalerate	420	8.0E-08	1.3E-09	11.69	6.20	78	-	42	-
	Etofenprox	376	2.2E-06	3.0E-08	11.72	7.05	91	-	47	-
	Permethrin	391	4.1E-09	1.0E-08	11.68	6.50	121	6	56	4
	Phenothrin	350	2.8E-08	1.0E-07	10.13	6.01	93	5	54	5
Phenols	2-Chlorophenol	129	5.2E-06	1.0E-01	4.97	2.15	55	9	33	-
	2-Nitrophenol	139	1.2E-05	1.9E-02	5.70	1.79	40	-	33	-
	4-Chloro-3-methylphenol	143	1.1E-06	3.8E-02	6.63	3.10	77	-	41	-
	para-Cresol	108	8.1E-07	2.2E-01	5.05	1.94	53	-	17	-
	Phenol	94	5.0E-07	1.5E-01	4.80	1.46	85	-	42	-
PAHs	Anthracene	188	5.5E-05	2.1E-05	7.50	4.53	82	25	45	-
	Benzo(a)anthracene	228	8.6E-06	1.1E-08	9.37	5.76	88	8	46	7
	Benzo(a)pyrene	252	5.7E-07	3.5E-09	10.34	6.13	24	7	56	4
	Benzo(b)fluoranthene	252	7.1E-07	1.1E-08	10.34	5.78	92	6	49	5
	Benzo(g,h,i)perylene	276	3.6E-07	4.6E-10	11.72	6.63	89	3	55	-

	Crysene	228	8.1E-06	1.1E-08	9.37	5.81	89	8	39	6
	Dibenzo(a,h)anthracene + Indeno(1,2,3-cd)pyrene [c]	-	-	-	-	-	84	2	54	14
	Fluorene	166	9.2E-05	1.6E-04	6.79	4.18	72	5	39	-
	Phenanthrene	178	5.2E-05	4.9E-05	7.57	4.46	87	6	41	24
	Pyrene	202	1.2E-05	8.7E-08	8.80	4.88	94	8	42	21
Cosmetic ingredients	Benzyl benzoate	212	3.3E-08	8.3E-04	7.46	3.97	72	-	57	-
	Glycerol tricaprylate	471	3.0E-07	5.9E-08	11.69	7.36	71	-	65	-
	Isopropyl myristate	270	7.6E-07	1.6E-04	8.21	6.90	90	-	53	-
	Lilial	204	4.8E-04	2.8E-03	6.90	3.93	106	-	29	-
	Triethyl citrate	276	2.5E-09	5.6E-05	9.07	0.51	67	-	45	-
Fragrance ingredients	2-Benzylideneoctanal	216	1.2E-04	7.1E-04	7.38	3.99	90	-	44	3
	Galaxolide	258	2.2E-05	3.9E-04	8.18	5.90	94	7	40	3
	Tonalide	258	1.3E-04	4.3E-05	7.93	5.70	94	7	49	26
UV filters	Benzophenone	182	1.1E-06	3.5E-04	7.25	3.18	61	-	48	-
	Homosalate	262	1.6E-09	4.5E-06	9.71	3.99	85	-	45	-
	Octocrylene	361	2.5E-08	1.9E-09	11.74	5.78	92	-	56	-
Skin oils	Cholesta-3,5-diene [d]	369	-	-	7.93	-	124	-	94	-
	Squalene	411	1.3E-06	1.2E-07	11.69	6.08	-	-	-	15

[a] Chemical properties were obtained from the U.S. Environmental Protection Agency (EPA)'s CompTox Chemicals Dashboard.
[b] When experimental values are available, we selected them over predicted values.
[c] Structures of these compounds were too close to discriminate one from the other.
[d] The K_{oa} value was estimated using the U.S. EPA's Estimation Program Interface (EPI) Suite.

Abbreviation: molecular weight (**MW**), Henry's law constant (**H**), vapor pressure (**VP**), octanol-air partition coefficient ($\mathbf{K_{oa}}$), octanol-water partition coefficient ($\mathbf{K_{ow}}$)

Table S3.2. Geometric mean (GM) dust concentrations (in ng/g of dust) and 95% confidence intervals (CI) for 6 phthalates and 1 fragrance ingredient that are both included in the current study and Mitro et al. (2016).

Compounds	Current study			Mitro et al.		
	GM	95% CI		GM	95% CI	
		Lower	Upper		Lower	Upper
BBP	7,786	6,108	9,927	44,294	22,075	88,876
DEP	940	802	1,101	2,033	1,148	3,601
DBP	5,060	4,233	6,048	13,643	9,780	19,031
DiBP	6,284	5,232	7,548	3,588	1,968	6,541
DEHP	79,546	72,556	87,343	237,542	168,030	335,843
DnOP	225	179	284	1437	1,020	2,024
Galaxolide	432	317	591	1977	551	7,101

Table S4.1. Chemical properties of 65 compounds.

Chemical class	Compounds	Chemical properties [a]				
		MW (g/mole)	H ($atm \cdot m^3$/mole)	VP (mmHg)	log K_{oa} [b]	log K_{ow} [b]
Phthalates	Benzyl butyl phthalate	312	2.8E-08	4.3E-06	9.83	4.73
	Di(2-ethylhexyl) phthalate	391	2.6E-07	2.9E-07	11.69	7.60
	Di-isobutyl phthalate	278	3.2E-07	3.2E-05	8.21	4.11
	Di-n-butyl phthalate	278	1.2E-06	2.6E-05	8.84	4.51
	Di-n-octyl phthalate	391	2.6E-07	4.0E-07	11.74	8.10
	Diethyl phthalate	222	2.4E-08	9.2E-04	6.75	2.42
	Dimethyl phthalate	194	9.4E-08	6.3E-03	5.68	1.60
Other plasticizers	Acetyl tributyl citrate	402	3.0E-08	1.6E-06	10.34	2.49
	Bis(2-ethylhexyl) adipate	371	4.1E-07	2.6E-06	10.86	6.85
	Dioctyl terephthalate	391	2.6E-07	4.3E-07	11.69	7.70
Insecticides	Bifenthrin	423	4.9E-08	1.2E-07	10.47	6.79
	Bioallethrin	274	2.6E-05	4.8E-06	7.93	3.74
	Chlorothalonil	266	2.8E-06	2.2E-04	8.49	3.10
	Chlorpyrifos	351	2.1E-06	3.1E-05	10.57	4.96
	Cyhalothrin	450	1.2E-08	2.1E-09	11.69	6.80
	Cypermethrin	416	2.0E-09	1.1E-09	11.70	6.60
	Deltamethrin	505	1.8E-09	1.6E-09	11.69	6.20
	Esfenvalerate	420	8.0E-08	1.3E-09	11.69	6.20
	Etofenprox	376	2.2E-06	3.0E-08	11.72	7.05
	Fipronil	437	9.1E-09	2.2E-09	10.54	3.94
	Fipronil-desulfinyl	389	1.6E-07	1.3E-08	10.63	3.55
	Fipronil-sulfide	421	1.5E-07	3.1E-09	11.25	3.76
	Fipronil-sulfone	453	9.2E-09	3.9E-09	10.96	3.45
	Imiprothrin	318	1.7E-09	9.8E-09	10.77	2.76
	Pentachlorophenol	266	3.4E-08	2.3E-04	8.46	5.02
	Permethrin	391	4.1E-09	1.0E-08	11.68	6.50
	Phenothrin	350	2.8E-08	1.0E-07	10.13	6.01
	Prallethrin	300	1.2E-04	7.6E-07	8.90	4.47
	Pyriproxyfen	321	6.1E-06	1.6E-07	10.33	4.20
	Resmethrin	338	3.2E-08	4.2E-08	10.19	6.12
	Tetramethrin	331	2.4E-06	4.2E-09	11.57	4.51
Phenols	2-Chlorophenol	129	5.2E-06	1.0E-01	4.97	2.15
	2-Nitrophenol	139	1.2E-05	1.9E-02	5.70	1.79
	2,3,4-Trichlorophenol	197	7.3E-07	6.8E-03	7.54	3.78
	2,3,5-Trichlorophenol	197	7.4E-07	7.1E-03	7.50	3.81
	2,3,6-Trichlorophenol	197	1.3E-06	8.5E-03	6.83	3.75
	2,4-Dichlorophenol	163	1.3E-06	2.8E-02	6.31	3.04
	2,4-Dimethylphenol	122	7.5E-07	5.6E-02	6.07	2.34
	2,4-Dinitrophenol	184	9.8E-08	5.9E-05	7.34	1.81
	2,4,5-Trichlorophenol	197	2.7E-07	2.6E-03	7.49	3.79
	2,4,6-Trichlorophenol	197	1.3E-06	5.1E-03	7.00	3.75
	2,6-Dichlorophenol	163	1.6E-06	2.5E-02	5.82	2.78
	3,4,5-Trichlorophenol	197	2.4E-07	5.9E-03	7.73	3.88

	4-Chloro-3-methylphenol	143	1.1E-06	3.8E-02	6.63	3.10
	4-Nitrophenol	139	9.2E-10	1.1E-02	6.11	2.01
	ortho-Cresol + meta-Cresol [c]	108	-	-	-	-
	para-Cresol	108	8.1E-07	2.2E-01	5.05	1.94
	Phenol	94	5.0E-07	1.5E-01	4.80	1.46
	Tetrachlorophenols [c]	232	-	-	-	-
PAHs	Anthracene	188	5.5E-05	2.1E-05	7.50	4.53
	Benzo(a)anthracene	228	8.6E-06	1.1E-08	9.37	5.76
	Benzo(a)pyrene	252	5.7E-07	3.5E-09	10.34	6.13
	Benzo(b)fluoranthene	252	7.1E-07	1.1E-08	10.34	5.78
	Benzo(g,h,i)perylene	276	3.6E-07	4.6E-10	11.72	6.63
	Crysene	228	8.1E-06	1.1E-08	9.37	5.81
	Dibenzo(a,h)anthracene + Indeno(1,2,3-cd)pyrene [d]	-	-	-	-	-
	Fluorene	166	9.2E-05	1.6E-04	6.79	4.18
	Phenanthrene	178	5.2E-05	4.9E-05	7.57	4.46
	Pyrene	202	1.2E-05	8.7E-08	8.80	4.88
Fragrance ingredients	2-Benzylideneoctanal	216	1.2E-04	7.1E-04	7.38	3.99
	Galaxolide	258	2.2E-05	3.9E-04	8.18	5.90
	Tonalide	258	1.3E-04	4.3E-05	7.93	5.70
UV filters	Homosalate	262	1.6E-09	4.5E-06	9.71	3.99
	Octocrylene	361	2.5E-08	1.9E-09	11.74	5.78
Skin oils	Squalene	411	1.3E-06	1.2E-07	11.69	6.08

[a] Chemical properties were obtained from the U.S. Environmental Protection Agency (EPA)'s CompTox Chemicals Dashboard.
[b] When experimental values are available, we selected them over predicted values.
[c] Isomers.
[d] Structures of these compounds were too close to discriminate one from the other.

Abbreviation: molecular weight (**MW**), Henry's law constant (**H**), vapor pressure (**VP**), octanol-air partition coefficient ($\mathbf{K_{oa}}$), octanol-water partition coefficient ($\mathbf{K_{ow}}$), polycyclic aromatic hydrocarbon (**PAH**), ultraviolet (**UV**)

Table S4.2. Median PUF concentrations (ng/g of foam) of 65 analyzed compounds in couch foam samples.

Chemical class	Compounds	LOD	LOQ	Top (*n*=13)		Top-middle (*n*=13)		Middle (*n*=13)		Total (*n*=39)	
				DF (%)	Median	DF (%)	Median	DF (%)	Median	DF (%)	Median
Phthalates	Benzyl butyl phthalate	500	1000	31	-	8	-	8	-	15	-
	Di(2-ethylhexyl) phthalate	1000	1000	92	1454	92	1611	85	1132	90	1456
	Di-isobutyl phthalate	250	1000	100	1285	69	422	31	-	67	500
	Di-n-butyl phthalate	500	1000	100	715	54	354	23	-	59	500
	Di-n-octyl phthalate	100	500	8	-	8	-	8	-	8	-
	Diethyl phthalate	50	100	100	419	100	410	100	308	100	414
	Dimethyl phthalate	10	25	100	28	100	34	100	32	100	33
Other plasticizers	Acetyl tributyl citrate	10000	25000	100	38665	23	-	8	-	44	-
	Bis(2-ethylhexyl) adipate	1000	5000	8	-	8	-	8	-	8	-
	Dioctyl terephthalate	2500	5000	0	-	0	-	0	-	0	-
Insecticides	Bifenthrin	10	25	0	-	0	-	0	-	0	-
	Bioallethrin	100	500	46	-	8	-	0	-	18	-
	Chlorothalonil	100	500	0	-	0	-	0	-	0	-
	Chlorpyrifos	10	25	0	-	0	-	0	-	0	-
	Cyhalothrin	50	250	0	-	0	-	0	-	0	-
	Cypermethrin	250	1000	0	-	0	-	0	-	0	-
	Deltamethrin	250	500	0	-	0	-	0	-	0	-
	Esfenvalerate	100	250	23	-	15	-	15	-	18	-
	Etofenprox	250	500	0	-	0	-	0	-	0	-
	Fipronil	10	50	0	-	0	-	0	-	0	-
	Fipronil-desulfinyl	10	25	0	-	0	-	0	-	0	-
	Fipronil-sulfide	10	10	0	-	0	-	0	-	0	-
	Fipronil-sulfone	50	100	0	-	0	-	0	-	0	-
	Imiprothrin	250	500	0	-	0	-	8	-	3	-
	Pentachlorophenol	1000	2500	0	-	0	-	0	-	0	-
	Permethrin	250	250	31	-	31	-	31	-	31	-
	Phenothrin	50	100	8	-	0	-	0	-	3	-
	Prallethrin	100	500	0	-	0	-	0	-	0	-
	Pyriproxyfen	10	50	0	-	0	-	0	-	0	-

	Resmethrin	100	250	0	-	0	-	0	-	0	-
	Tetramethrin	25	50	0	-	0	-	0	-	0	-
Phenols	2-Chlorophenol	25	100	0	-	0	-	0	-	0	-
	2-Nitrophenol	100	100	0	-	8	-	8	-	5	-
	2,3,4-Trichlorophenol	500	1000	0	-	0	-	0	-	0	-
	2,3,5-Trichlorophenol	100	500	0	-	0	-	0	-	0	-
	2,3,6-Trichlorophenol	100	500	0	-	0	-	0	-	0	-
	2,4-Dichlorophenol	50	250	0	-	0	-	0	-	0	-
	2,4-Dimethylphenol	50	100	0	-	0	-	0	-	0	-
	2,4-Dinitrophenol	2500	10000	0	-	0	-	0	-	0	-
	2,4,5-Trichlorophenol	500	1000	0	-	0	-	0	-	0	-
	2,4,6-Trichlorophenol	100	500	0	-	0	-	0	-	0	-
	2,6-Dichlorophenol	250	500	0	-	0	-	0	-	0	-
	3,4,5-Trichlorophenol	100	250	0	-	0	-	0	-	0	-
	4-Chloro-3-methylphenol	100	250	100	12656	100	10236	85	8229	95	10236
	4-Nitrophenol	2500	5000	0	-	0	-	0	-	0	-
	ortho-Cresol + meta-Cresol [c]	100	100	92	167	92	217	100	255	95	222
	para-Cresol	1000	1000	23	-	15	-	15	-	18	-
	Phenol	250	250	100	33134	100	37907	100	39560	100	35727
	Tetrachlorophenols [c]	100	500	0	-	0	-	0	-	0	-
PAHs	Anthracene	10	50	23	-	8	-	8	-	13	-
	Benzo(a)anthracene	10	25	0	-	8	-	8	-	5	-
	Benzo(a)pyrene	5	25	0	-	0	-	0	-	0	-
	Benzo(b)fluoranthene	10	25	0	-	0	-	0	-	0	-
	Benzo(g,h,i)perylene	25	100	0	-	0	-	0	-	0	-
	Crysene	50	50	0	-	0	-	0	-	0	-
	Dibenzo(a,h)anthracene + Indeno(1,2,3-cd)pyrene [d]	10	50	0	-	0	-	0	-	0	-
	Fluorene	10	50	46	-	46	-	46	-	46	-
	Phenanthrene	10	50	100	235	85	126	54	54	79	110
	Pyrene	5	25	100	94	46	-	38	-	62	39
Fragrance ingredients	2-Benzylideneoctanal	100	250	100	14314	100	6827	85	1558	95	5213
	Galaxolide	5	25	100	8588	100	6098	100	3155	100	4864
	Tonalide	10	50	100	1540	92	659	77	202	90	659
UV filters	Homosalate	25	50	100	13523	100	7073	92	1623	97	5173

	Octocrylene	100	250	23	-	0	-	0	-	8	-
Skin oils	Squalene	1000	1000	46	-	15	-	0	-	21	-

Abbreviation: limit of detection (**LOD**), limit of quantification (**LOQ**), detection frequency (**DF**), polycyclic aromatic hydrocarbons (**PAH**), ultraviolet (**UV**)

Table S4.3. Input parameters for calculating the fugacity capacity for indoor compartments.

Property name (unit)	Symbol	Value
Temperature (K)	T	298
Ideal gas constant ($Pa \cdot m^3/mol \cdot K$)	R	8.314
Dust loading on carpet (kg/m^2)	ρ_c	3.8×10^{-4}
Dust particle density (kg/m^3)	ρ_d	1500
Density of film (kg/m^3)	ρ_f	826
Dust loading on vinyl (kg/m^2)	ρ_v	5.0×10^{-4}
Thickness of carpet (m)	δ_c	1.6×10^{-2}
Thickness of organic film (m)	δ_{film}	4.8×10^{-9}
Thickness of vinyl (m)	δ_v	1.8×10^{-3}
Thickness of wall (m)	δ_w	1.5×10^{-2}
Fraction of organic carbon material in film (unitless)	$f_{oc,f}$	0.2
Area of house (m^2)	A	153
Height of ceiling (m)	h	2.44
Fraction of floor that is carpet (unitless)	ff_c	0.84
Fraction of floor that is vinyl (unitless)	ff_v	0.16

Table S4.4. Properties of particles (three size fraction) used for calculating the fugacity capacity for indoor compartments.

Particle size fraction (μm)	Fraction of organic carbon, $f_{oc,i}$ (unitless)	Particulate mass in air, $\rho_{p,i}$ (μg/m3)	Fraction in size fraction	
			Carpet, $f_{c,i}$ (unitless)	Vinyl, $f_{v,i}$ (unitless)
0-2.5	0.65	13	0.02	0.04
2.5-10	0.28	12	0.06	0.09
10-150	0.06	4.3	0.35	0.77

Table S4.5. Equations used to compute the fugacity capacity for indoor compartments.

Compartment	Phase	Equation	Description
Air	pure air	$Z_{air} = \frac{1}{R \times T}$	R is the ideal gas constant (Pa·m^3/mol·K) and T is the ambient temperature (K).
	air particles	$Z_{ap,i} = \frac{K_{p,i} \times \rho_d \times 10^9}{R \times T}$	K_p is the partition coefficient between particles and air (m^3/μg) and ρ_d is the dust particle density (1500 kg/m^3). $\log K_{p,i} = \log K_{oa} + \log\left(\frac{f_{oc,i}}{0.74}\right) - 11.91$, where K_{oa} is the octanol-air partition coefficient (unitless) and $f_{oc,i}$ the fraction of organic carbon for particles in a specific size fraction (unitless). 10^9 is included to convert the units from kilograms (kg) to micrograms (μg).
	total air	$Z_a = \frac{\sum_{i=1}^{3}(Z_{ap,i} \times \rho_{p,i})}{\rho_d \times 10^9} + Z_{air}$	$\rho_{p,i}$ is the particle mass concentration in the air for a given size fraction (μg/m^3).
Carpet	carpet	$Z_{carpet} = \frac{K_{ca}}{R \times T}$	K_{ca} is the carpet-air partition coefficient (unitless). $\log K_{ca} = 4.83 - 0.82 \log VP$ and VP is the vapor pressure (Pa).
	carpet dust	$Z_{cp} = \sum_{i=1}^{3}(Z_{ap,i} \times f_{c,i})$	$f_{c,i}$ is the volume fraction on carpet (unitless).
	total carpet	$Z_c = \frac{Z_{carpet}\delta_c + Z_{cp}\rho_c/\rho_d}{\delta_c + \rho_c/\rho_d}$	δ_c is the thickness of carpet (0.016 m) and ρ_c is the particle mass concentration on carpet (0.00038 kg/m^2).
Vinyl	vinyl	$Z_{vinyl} = \frac{K_{va}}{R \times T}$	K_{va} is the vinyl-air partition coefficient (unitless). $\log K_{va} = 5.20 - 0.68 \log VP$
	film	$Z_{film} = \frac{0.48 \times K_{ow} \times f_{oc,f} \times \rho_f}{H \times 1000}$	K_{ow} is the partition coefficient between octanol and water (unitless). $f_{oc,f}$ is the fraction of organic carbon material in the film (unitless). ρ_f is the density of film (kg/m^3). H is the Henry's law constant (Pa·m^3/mol). 1000 is a conversion factor with units of L/m^3.
	vinyl dust	$Z_{vp} = \sum_{i=1}^{3}(Z_{ap,i} \times f_{v,i})$	$f_{v,i}$ is the volume fraction on vinyl (unitless).

	total vinyl	$Z_v = \frac{Z_{film}\delta_{film} + Z_{vinyl}\delta_v + Z_{vp}\rho_v/\rho_d}{\delta_{film} + \delta_v + \rho_v/\rho_d}$	δ_{film} is the thickness of film (m), δ_v is the thickness of vinyl (m), and ρ_v is the dust loading on vinyl (kg/m^2).
Wall	wall	$Z_{wall} = \frac{K_{wa}}{R \times T}$	K_{wa} is the wall-air partition coefficient (unitless). $\log K_{wa} = 2.93 - 0.31 \log VP$

Table S4.6. Calculated fugacity capacities (mol/m^3·Pa) of five indoor compartments for studied compounds.

Compounds	PUF	Air	Carpet	Wall	Vinyl
Phthalates and other plasticizers					
Dimethyl phthalate	1.16×10^{-1}	4.04×10^{-4}	5.78×10^{1}	4.56×10^{-1}	1.19×10^{2}
Diethyl phthalate	1.14×10^{0}	4.04×10^{-4}	1.53×10^{2}	6.59×10^{-1}	2.67×10^{2}
Di-isobutyl phthalate	2.12×10^{1}	4.05×10^{-4}	2.40×10^{3}	1.87×10^{0}	2.63×10^{3}
Di-n-butyl phthalate	5.44×10^{1}	4.09×10^{-4}	2.81×10^{3}	1.98×10^{0}	3.00×10^{3}
Acetyl tributyl citrate	5.08×10^{1}	5.81×10^{-4}	2.80×10^{4}	4.72×10^{0}	2.04×10^{4}
Di(2-ethylhexyl) phthalate	9.04×10^{3}	4.37×10^{-3}	1.15×10^{5}	8.05×10^{0}	7.39×10^{4}
Phenols and Polycyclic aromatic hydrocarbons					
Phenol	1.72×10^{0}	4.04×10^{-4}	2.32×10^{0}	1.35×10^{-1}	8.29×10^{0}
Phenanthrene	2.38×10^{1}	4.04×10^{-4}	1.69×10^{3}	1.63×10^{0}	1.96×10^{3}
Pyrene	4.04×10^{2}	4.09×10^{-4}	3.04×10^{5}	1.16×10^{1}	1.45×10^{5}
Fragrance ingredients and Ultraviolet filters					
2-Benzylideneoctanal	1.09×10^{2}	4.04×10^{-4}	1.89×10^{2}	7.14×10^{-1}	3.18×10^{2}
Tonalide	3.52×10^{2}	4.04×10^{-4}	1.88×10^{3}	1.70×10^{0}	2.14×10^{3}
Galaxolide	1.01×10^{3}	4.05×10^{-4}	3.11×10^{2}	8.62×10^{-1}	4.84×10^{2}
Homosalate	2.55×10^{4}	4.45×10^{-4}	1.20×10^{4}	3.42×10^{0}	1.00×10^{4}

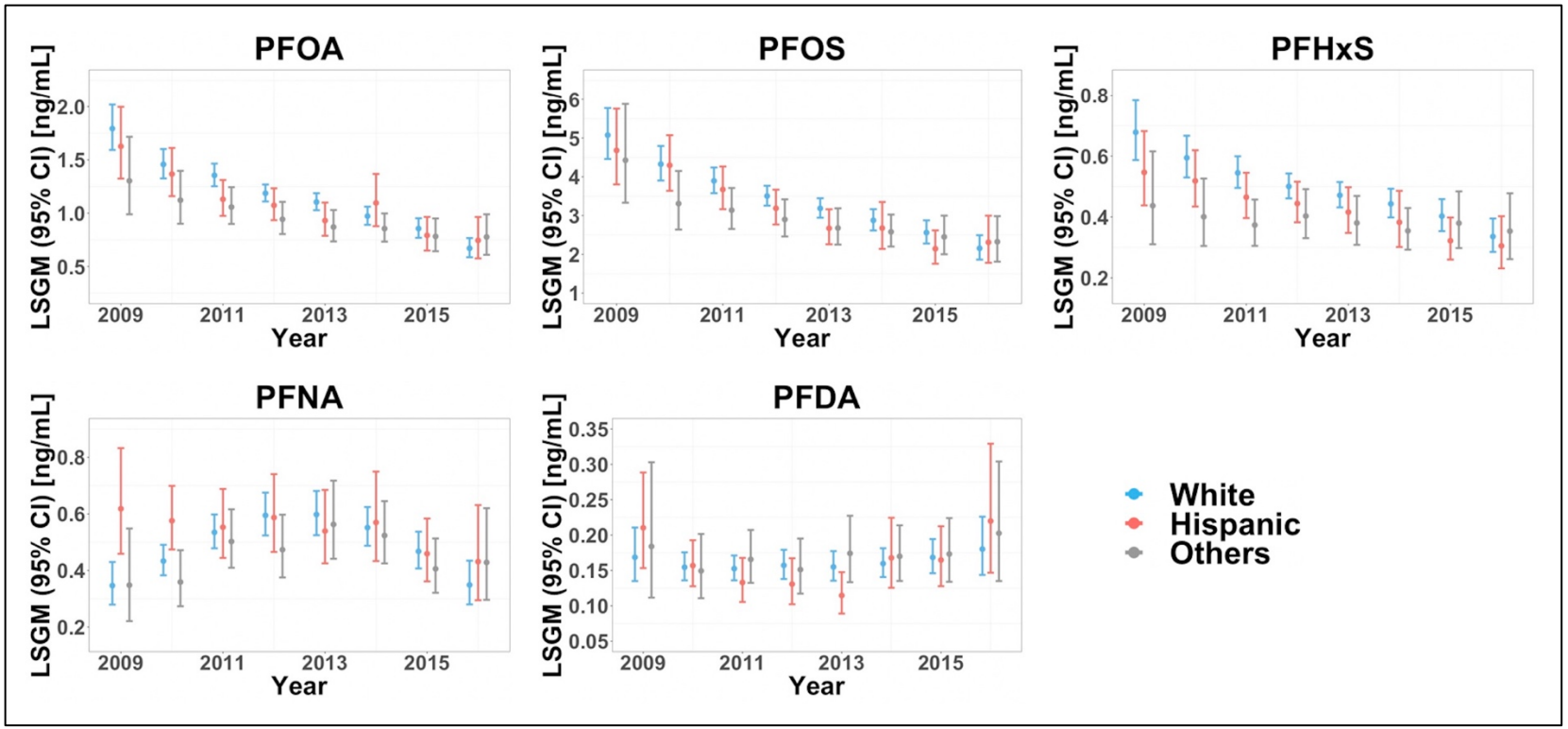

Figure S2.1. Least square geometric means (LSGMs) of PFAS maternal serum concentrations by maternal race/ethnicity.

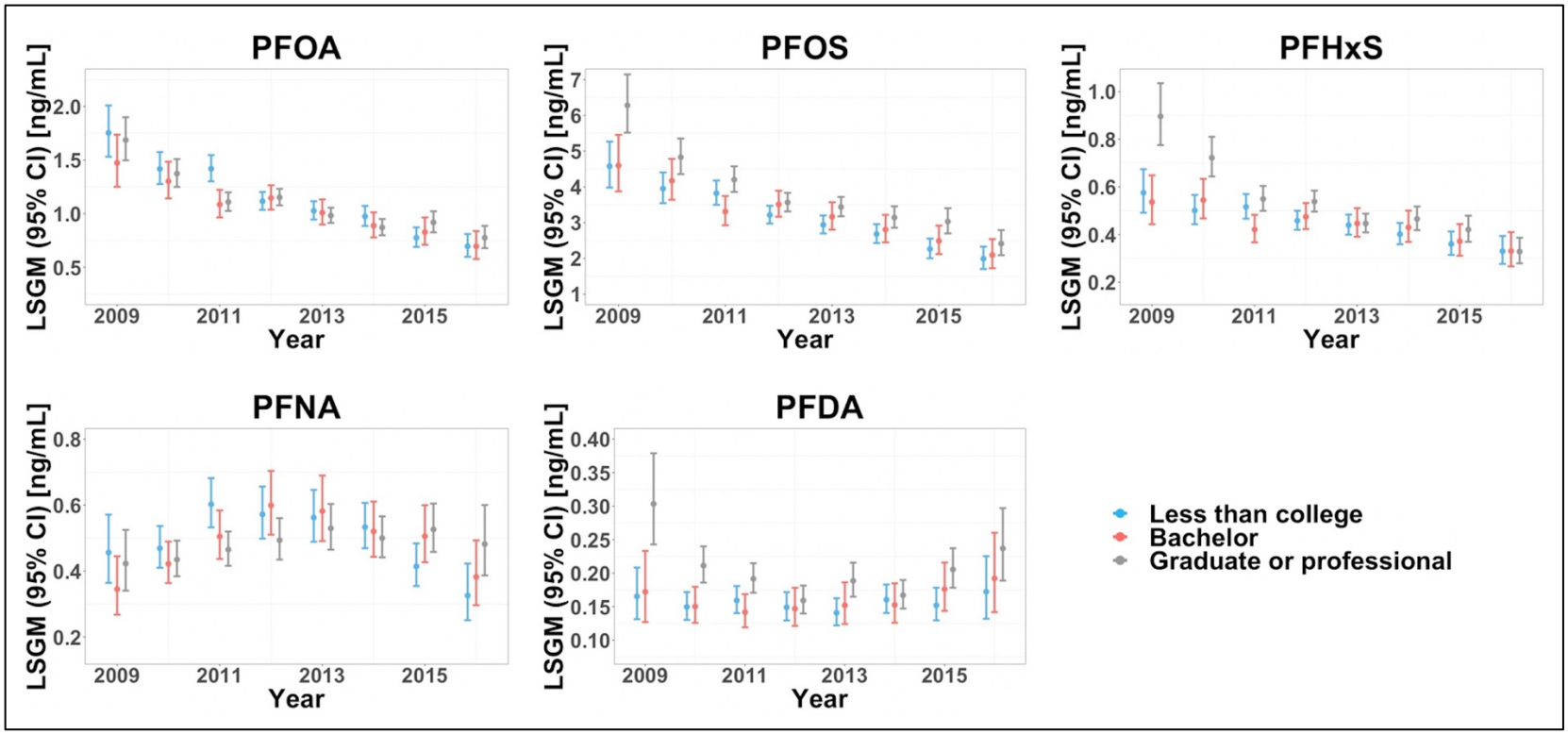

Figure S2.2. Least square geometric means (LSGMs) of PFAS maternal serum concentrations by maternal education.

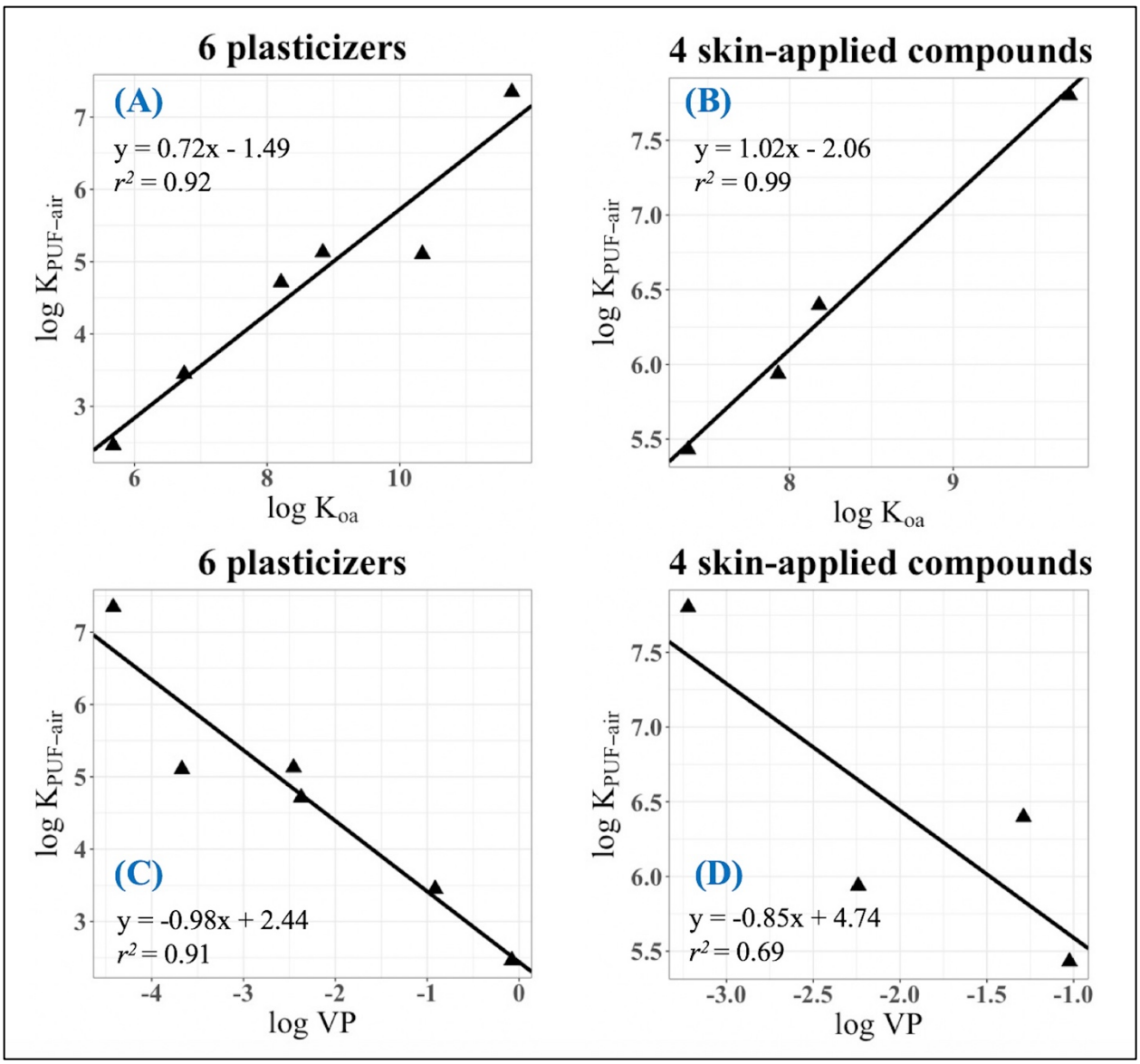

Figure S4.1. Calculated median log $K_{PUF\text{-}air}$ versus log K_{oa} (A and B) or log VP (C and D) for two individual use categories (i.e., plasticizers and skin-applied compounds).