

# A Comparison of Bituminous Coal-Based and Coconut Shell-Based Activated Carbon for Removal of Trace Hazardous Air Pollutants in Landfill Gas

Annaprabha Athappan, Melanie L. Sattler\*

Department of Civil Engineering, University of Texas at Arlington, Box 19308, Arlington, TX 76019

\*Corresponding author's Email address: Sattler@uta.edu

**ABSTRACT:** This study explored adsorption as a method of controlling hazardous air pollutant emissions from landfill gas (LFG). In particular, the study compared bituminous coal-based (BPL) and coconut shell-based (OVC) activated carbons as adsorbents for the hazardous landfill gas constituents para-, ortho-, and meta-xylene; ethylbenzene; and methyl ethyl ketone (MEK). Experiments were designed to replicate typical field usage conditions, using ambient temperature and relative humidity values, and testing the carbon as is, without pretreatment to remove moisture. BPL and OVC carbons from Calgon Carbon were tested in vials, with initial headspace concentrations ranging from 2130 to 5020 ppm<sub>v</sub> (86 to 120 mg/m<sup>3</sup>). Pollutant concentration remaining in the headspace at equilibrium was measured using an SRI 8610 gas chromatograph with flame ionization detector. Tests were conducted at 45-70% relative humidity and room temperature. Adsorption curve fits of Langmuir and Freundlich isotherms (LI and FI) were determined for each compound and each type of activated carbon. For MEK, BPL and OVC adsorption capacities were similar for all concentrations. For ethylbenzene and the xylene isomers, however, OVC coconut shell-based carbon had a higher adsorption capacity at lower concentrations, but BPL coal-based carbon had a higher capacity at higher concentrations. This is likely due to the greater external surface area for OVC carbon, and the greater internal surface area for BPL carbon. For both carbons, at low concentrations adsorption capacity was highest for ethylbenzene, at intermediate concentrations highest for para-xylene, and at high concentrations highest for ortho-xylene. For ethylbenzene and MEK, the Freundlich isotherm fit the data better; for the xylenes, the Langmuir isotherm generally fit the data better. Manufacturer isotherms, developed for ideal conditions, predicted significantly higher maximum adsorption capacities than those determined in this research. Both bituminous coal-based and coconut shell-based activated carbons showed promise for removing hazardous air pollutants from landfill gas, for different concentration levels.

**Keywords:** Adsorption, Landfill Gas, Hazardous Air Pollutants, Xylene, Ethylbenzene, MEK

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## INTRODUCTION

Refuse disposal is an issue that must be addressed by every community worldwide. As microbes degrade the organic fraction of waste in landfills, methane (CH<sub>4</sub>) is generated, along with carbon dioxide (CO<sub>2</sub>), water, and other trace landfill gas (LFG) constituents. The trace LFG constituents include many volatile organic compounds (VOCs), which contribute to ground-level ozone formation, and many of which are air pollutants (HAPs). According to the U.S. Environmental Protection Agency's AP-42, nearly 30 compounds found in LFG are hazardous air pollutants; exposure to these pollutants can lead to adverse health effects (U.S. EPA, 1998 and 2008).

Adsorption is a viable control option for landfill trace constituents. Adsorption captures a pollutant known as adsorbate on the surface of a solid adsorbent, in a process that can be either physical or chemical. Advantages of adsorption include high removal efficiency, low space requirements, and ability to regenerate and reuse the adsorbent. Although flares are also often used as a control technique for landfill gas, disadvantages of flaring include loss of methane as an energy source, removal of VOCs/HAPs perhaps significantly lower than 98%, and generation of other

pollutants as combustion by-products (Chen et al., 2007).

This study explored adsorption as a method of controlling hazardous air pollutant emissions from landfill gas. In particular, the study compared bituminous coal-based (BPL) and coconut shell-based (OVC) activated carbons as adsorbents for the hazardous landfill gas constituents para-, ortho-, and meta-xylene; ethylbenzene; and methyl ethyl ketone (MEK). Experiments were designed to replicate typical field usage conditions, using ambient temperature and relative humidity values, and testing the carbon as is, without pre-treatment to remove moisture. Although lab studies may heat carbon to remove initial moisture, this is not common practice before installing carbon for use in a column in the field.

## Adsorbents and adsorbates tested in this research

Of the HAPs identified in landfill gas in AP-42, xylene, ethylbenzene, and methyl ethyl ketone (MEK) were selected for this research because of their high concentrations in LFG compared with other HAPs (U.S. EPA, 1998 and 2008). In addition to being hazardous air pollutants, xylene, ethylbenzene, and MEK also contribute to ground-level ozone formation, with

incremental reactivities of 0.5, 0.3, and 0.6 ppm O<sub>3</sub>/ppm C, respectively (Carter, 1991).

Activated carbon has long been recognized as one of the most versatile adsorbents due to its high porosity and resulting high surface area (Prakash, 1994; Yang, 2003). Activated carbon is manufactured by using raw materials such as wood, coal, or coconut shell, heated to 600-900°C in an inert atmosphere, followed by heating to 600-1200°C in an oxidizing atmosphere. Depending on the raw material, different types of activated carbon have different physical/chemical characteristics, such as surface area, porosity, and surface functional groups. Bituminous-coal based and coconut-shell based activated carbons were selected because reactivation is good for both, compared to other types of carbon (Carbochem, 2008).

Several previous studies have compared bituminous-coal and coconut-shell based activated carbon, but not for adsorption of xylenes or ethylbenzene. Chiang et al. (2001) compared benzene adsorption onto bituminous-coal based carbon (Calgon Fitratorb 400) and coconut-shell based activated carbon (provided by Liangchien). Although the bituminous-coal based carbon had a higher specific surface area, the coconut-based carbon had a higher percent of micropores, which led to greater benzene adsorption. Kim et al. (2006) compared bituminous-coal based and coconut-shell based activated carbon for adsorption of toluene, MEK, and methyl isobutyl ketone (MIK) over a range of concentrations. For toluene, BPL had a slightly higher adsorption capacity; for MEK, OVC had a slightly higher adsorption capacity; for MIBK, OVC had a higher adsorption capacity at low concentrations, but BPL had a higher adsorption capacity at higher concentrations (>0.2 mmol/m<sup>3</sup>). In Kim's study, however, carbon was pre-treated to remove moisture.

## MATERIALS AND METHODS

### Adsorbents

Two types of activated carbon, bituminous coal-based BPL and coconut-based OVC (Calgon Carbon Corporation, Pittsburgh, PA) were used for this study. Table 1 shows characteristics of these carbons, according to manufacturer data. To replicate typical field usage conditions, the carbon was tested as is, without pre-treatment to remove moisture.

### Adsorbates

Para-xylene, meta-xylene, ortho-xylene, ethylbenzene, and MEK were purchased from Fisher Scientific in liquid form. To convert the liquids to gas phase, various known amounts of liquid were injected into 1-L air-filled Tedlar bags. A week was allowed for evaporation to occur. The known pollutant concentrations were then used for the experiments. Multiple gas-phase concentrations for each pollutant produced multiple isotherm points.

### Isotherm experimental procedure

Laboratory relative humidity, which was measured using a Testo 605-H1 humidity stick, ranged from 45-70%; this air was filled the adsorption vials. Room temperature was maintained around 70-80°F.

The experimental method for determining isotherms was based on that reported by Lavancy et al. (1996) and Himeno and Urano (2006). A carbon granule of 0.011 – 0.012 g was weighed using a digital weighing scale and dropped inside a 4 ml vial, which was sealed with a Teflon-coated septum and an aluminum cap. The pollutant compound was injected into the vial from the Tedlar bag using a gas-tight syringe. The vial was shaken intermediately, and 1 hour was allowed for equilibrium to be reached. Initial trials were conducted to determine that the 1-hour equilibrium time was sufficient.

After the equilibrium time had passed, the headspace concentration was measured using an SRI 8610C gas chromatograph (GC) with flame ionization detector (FID) and 60 m capillary column. The mass of adsorbed pollutant was determined by the difference between the total pollutant mass injected and that of the pollutant in the gas-phase in the vial at equilibrium. Adsorption equilibrium concentrations were determined for enough points to enable an isotherm to be plotted for each pollutant/adsorbent combination.

U.S. EPA AP-42 landfill concentrations for xylene, ethylbenzene, and MEK are 12.1, 4.61, and 7.09 ppm, respectively (U.S. EPA, 1998 and 2008). These low initial concentration values were tested with BPL and OVC but the equilibrium headspace concentration value was found to be zero. The GC was not sensitive enough to measure the very low concentrations remaining in the headspace. Hence, higher initial concentration values were used for developing the isotherms.

### Isotherm data analysis

Adsorption isotherms show equilibrium adsorption capacity (mass of pollutant adsorbed per unit mass of adsorbent) over a range of equilibrium pollutant partial pressures at a given temperature. Isotherms provide critical information for designing adsorption systems, and can also be used to find the maximum adsorption capacity. Adsorption isotherm results for BPL and OVC carbon were analyzed using the Freundlich and Langmuir models, which assume monolayer coverage of adsorbate molecules on the adsorbent. The Freundlich model has the form:

$$q_e = K_f C_e^{1/n}$$

where

$q_e$  = equilibrium amount of pollutant adsorbed per unit mass of adsorbent, (g/g);  $C_e$  = equilibrium gas-phase pollutant concentration, (mg/L);  $K_f$ ,  $n$  = Freundlich coefficients.

The linearized form of the Freundlich equation is:

$$\log q_e = \log K_f + 1/n \log C_e$$

$K_f$  and  $1/n$  can be determined via linear regression by plotting  $\log q_e$  vs  $\log C_e$ .

The Langmuir isotherm model has the form:

$$q_e = \frac{q_{\max} C_e}{1 + b C_e}$$

where  $q_{\max}$ ,  $b$  = Langmuir coefficients.  $q_{\max}$  also represents the maximum adsorption capacity. Linearized forms of the Langmuir equation include:

$$1/q_e = 1/q_{\max} + (1/(b q_{\max})) \times (1/C_e) \quad \text{(Form 1)}$$

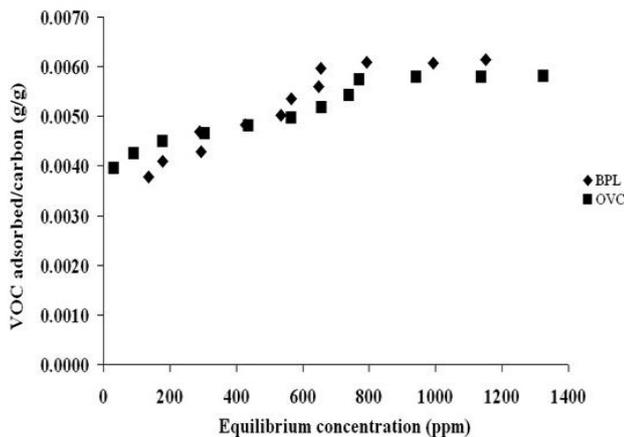
$$C_e/q_e = 1/(q_{\max} b) + (C_e/q_{\max}) \quad \text{(Form 2)}$$

In this research, Freundlich and Langmuir isotherm coefficients were determined using the non-linearized forms of the equations and non-linear regression (using Oakdale Engineering Datafit Software Version 9), and also using the linearized forms of the equations and linear regression. Coefficients determined using the various approaches were compared.

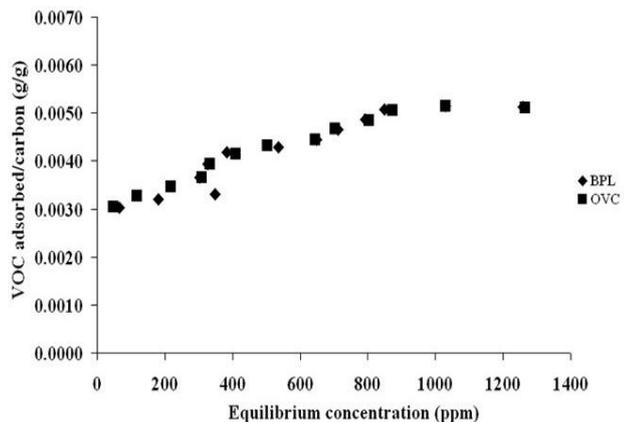
## RESULTS AND DISCUSSION

### Adsorption Isotherms for BPL and OVC Carbons

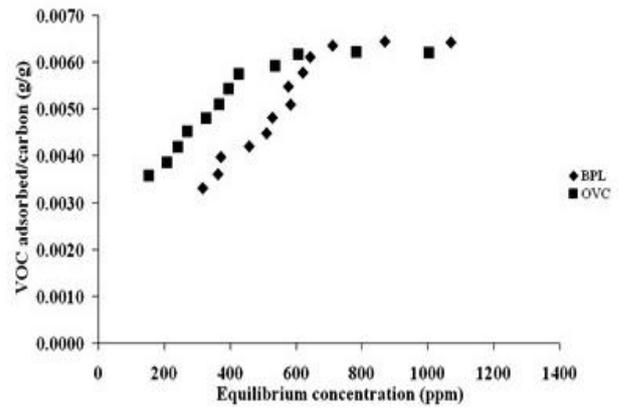
Graphs 1 - 5 show the experimental isotherm data collected for the 5 pollutants tested (including 3 isomers of xylene) onto the 2 adsorbents. For MEK, OVC and BPL adsorption capacities are similar for all concentrations, as shown in Graph 2. For ethylbenzene, ortho-xylene, para-xylene, and meta-xylene, however, OVC has a higher adsorption capacity for lower concentrations, but BPL has a higher adsorption capacity for higher concentrations, with the crossover occurring at concentrations around 400 ppm, 600 ppm, 400 ppm, and 300 ppm, respectively. OVC likely adsorbs more pollutant than BPL at low concentrations due to OVC's greater external surface area, which pollutant molecules can easily access. At higher concentrations, external adsorption sites are likely full, requiring pollutant molecules to access inner adsorption sites. Since BPL's internal macropore surface area is greater than OVC, as shown in Table 1, it adsorbs more pollutant at higher concentrations.



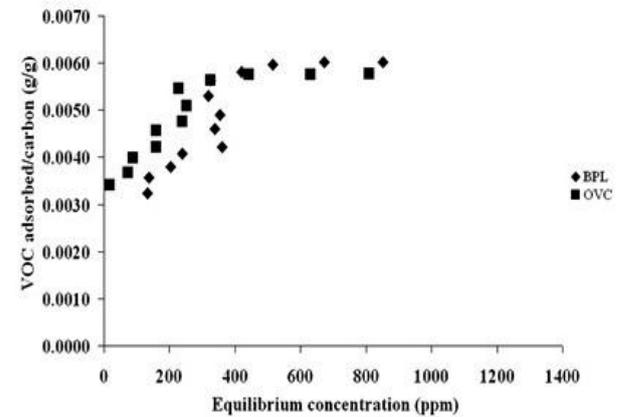
Graph 1. Ethylbenzene adsorption isotherm data for BPL and OVC activated carbons.



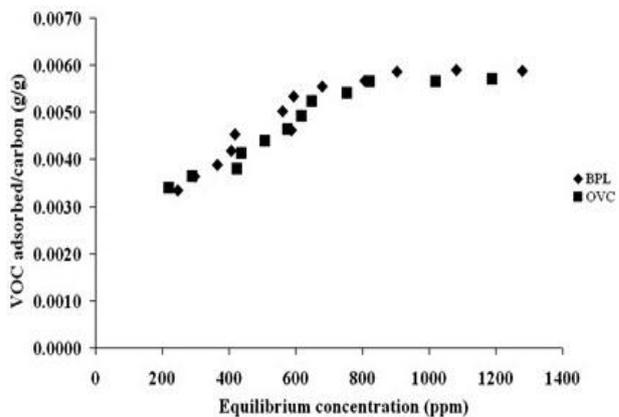
Graph 2. MEK adsorption isotherm data for BPL and OVC activated carbons.



Graph 3. Ortho-xylene adsorption isotherm data for BPL and OVC activated carbons



Graph 4. Para-xylene adsorption isotherm data for BPL and OVC activated carbons



Graph 5. Meta-xylene adsorption isotherm data for BPL and OVC activated carbons

Table 1. Comparison of properties of bituminous coal-based (BPL) and coconut shell-based (OVC) activated carbon

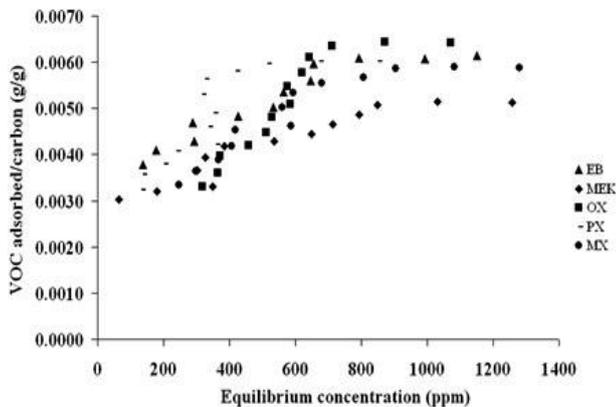
Properties	BPL	OVC
Micropores	High	High
Macropores	Medium	Low
Ash	10%	5%
Hardness	High	High
Density	0.48 g/cc	0.48 g/cc
Reactivation	Good	Good
Iodine no.	1000	1100
Dust	Medium	Low
Water soluble ash	Low	High
Mesh size	4x6	4x6

Graphs 6 and 7 show BPL and OVC data from Graphs 1-5 replotted with all 5 pollutants on one graph. It can be seen that for both BPL and OVC, at low concentrations adsorption capacity was highest for ethylbenzene, at intermediate concentrations highest for para-xylene, and at high concentrations highest for ortho-xylene. For both BPL and OVC, adsorption capacity for MEK is low compared to other compounds over most concentration ranges. Among the 3 xylenes, BPL and OVC adsorption capacities at low concentrations are highest for para-xylene and at high concentrations are highest for ortho-xylene; adsorption capacities are lowest for meta-xylene across most concentrations.

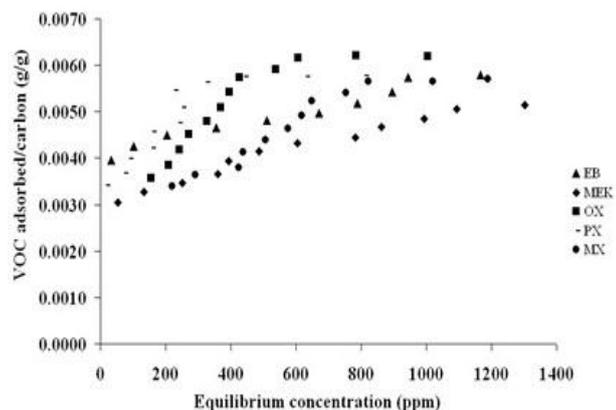
Adsorption capacity depends on VOC characteristics such as molecular weight, volatility, and size of the molecule. Table 2 compares adsorbate molecular weights and boiling points (the lower a compound's boiling point, the higher its volatility). MEK, with the lowest molecular weight and highest volatility of the compounds studied, had the lowest adsorption capacity, as expected. Ortho-xylene, with a high molecular weight and comparatively low volatility, had the highest adsorption capacity at high concentrations.

**Table 2.** Adsorbate molecular weights and boiling points

Adsorbate	Molecular Weight	Boiling Point (°C)
Ethylbenzene	106.16	140
MEK	72.11	80
Xylene (all isomers)	106.16	136



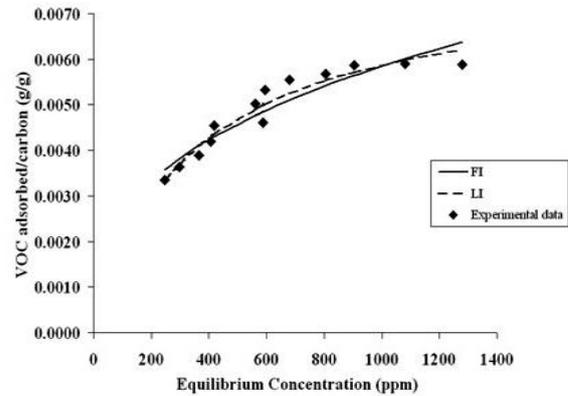
**Graph 6.** VOC adsorption isotherm data for BPL activated carbon.



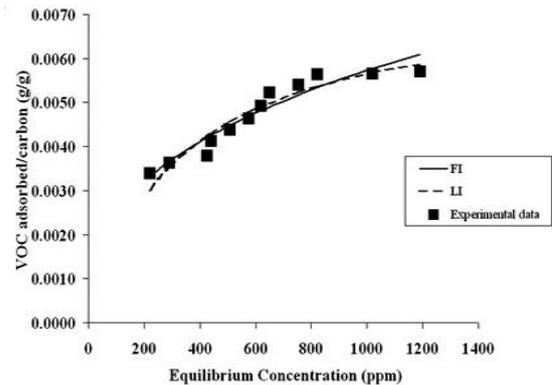
**Graph 7.** VOC adsorption isotherm data for OVC activated carbon.

### Freundlich and langmuir model coefficients:

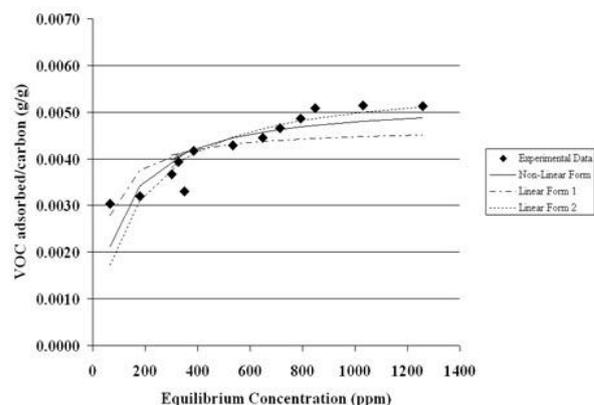
Graphs 8 and 9 show the experimental isotherm data for m-xylene onto both activated carbons, along with Freundlich and Langmuir model isotherms, with coefficients obtained from non-linear regression. Graphs for the rest of the adsorbates are available from the authors. Coefficients and associated  $R^2$  values obtained from linear and non-linear curve-fits of the Freundlich and Langmuir models are compared in Table 3.



**Graph 8.** Freundlich and Langmuir isotherm curve-fits for meta-xylene adsorption onto BPL activated carbon.



**Graph 9.** Freundlich and Langmuir isotherm curve-fits for meta-xylene adsorption onto OVC activated carbon.



**Graph 10.** Comparison of linear and non-linear Langmuir curve-fits for MEK adsorption onto BPL activated carbon

For ethylbenzene and MEK, the Freundlich isotherm produced higher  $R^2$  values; for the xylenes, the Langmuir isotherm gave higher  $R^2$  values. For the Freundlich model, coefficients from the non-linearized form produced  $R^2$  values greater than the linearized form in all cases except for para-xylene onto OVC. Similarly, for the Langmuir model, coefficients from the non-

linearized form gave  $R^2$  values greater than the linearized form in all cases except for ortho-xylene and meta-xylene onto BPL. Graph 10 compares curve-fits of the 3 forms of the Langmuir model for MEK onto BPL

as an example. As would be expected, Linearized Form 1 gives more importance to low pollutant concentration values, whereas Linearized Form 2 gives weight to high concentration values.

**Table 3.** Freundlich and Langmuir isotherm coefficients from linear and non-linear regression

Pollutant	Adsorbent	Isotherm	Coefficient	Coefficient Value		
				Linearized Form 1	Linearized Form 2	Non-linear form
Ethylbenzene	BPL	Freundlich	$K_f$	0.00112	--	0.00114
			n	4.05	--	4.10
			$R^2$	0.9985	--	0.9985
		Langmuir	$q_{max}$	0.00645	0.00697	0.00674
			b	0.00943	0.00662	0.00777
			$R^2$	0.9972	0.9974	0.9976
Ethylbenzene	OVC	Freundlich	$K_f$	0.00258	--	0.00254
			n	9.00	--	8.75
			$R^2$	0.9986	--	0.9986
		Langmuir	$q_{max}$	0.00533	0.00604	0.00545
			b	0.0800	0.0149	0.0616
			$R^2$	0.9937	0.9823	0.9941
MEK	BPL	Freundlich	$K_f$	0.00112	--	0.00101
			n	4.66	--	4.33
			$R^2$	0.9968	--	0.9969
		Langmuir	$q_{max}$	0.00467	0.00574	0.00525
			b	0.0228	0.0066	0.0104
			$R^2$	0.9884	0.9900	0.9915
MEK	OVC	Freundlich	$K_f$	0.00138	--	0.00132
			n	5.43	--	5.22
			$R^2$	0.9985	--	0.9986
		Langmuir	$q_{max}$	0.00462	0.00550	0.00493
			b	0.0326	0.0087	0.0189
			$R^2$	0.9906	0.9891	0.9924
ortho-Xylene	BPL	Freundlich	$K_f$	9.53E-05	--	1.58E-04
			n	1.60	--	1.83
			$R^2$	0.9930	--	0.9936
		Langmuir	$q_{max}$	0.0150	0.0121	0.0119
			b	0.00090	0.00128	0.00133
			$R^2$	0.9944	0.9962	0.9953
ortho-Xylene	OVC	Freundlich	$K_f$	6.82E-04	--	8.46E-04
			n	2.97	--	3.32
			$R^2$	0.9961	--	0.9964
		Langmuir	$q_{max}$	0.00788	0.00751	0.00775
			b	0.00504	0.00589	0.00533
			$R^2$	0.9985	0.9984	0.9985
para-Xylene	BPL	Freundlich	$K_f$	6.01E-04	--	7.25E-04
			n	2.80	--	3.07
			$R^2$	0.9913	--	0.9916
		Langmuir	$q_{max}$	0.00734	0.00743	0.00753
			b	0.00607	0.00589	0.00576
			$R^2$	0.9930	0.9931	0.9931
para-Xylene	OVC	Freundlich	$K_f$	0.00196	--	0.00193
			n	5.94	--	5.70
			$R^2$	0.9962	--	0.9958
		Langmuir	$q_{max}$	0.00518	0.00613	0.00567
			b	0.0918	0.0225	0.0415
			$R^2$	0.9899	0.9876	0.9977
meta-Xylene	BPL	Freundlich	$K_f$	4.41E-04	--	5.14E-04
			n	2.65	--	2.84
			$R^2$	0.9964	--	0.9966
		Langmuir	$q_{max}$	0.00792	0.00756	0.00782
			b	0.00290	0.00330	0.00300
			$R^2$	0.9981	0.9983	0.9982
meta-Xylene	OVC	Freundlich	$K_f$	4.65E-04	--	4.73E-04
			n	2.75	--	2.77
			$R^2$	0.9976	--	0.9976
		Langmuir	$q_{max}$	0.00685	0.00738	0.00747
			b	3.98E-03	3.19E-03	3.09E-03
			$R^2$	0.9972	0.9977	0.9977

Table 4 shows the experimentally-determined maximum adsorption capacities obtained for the 5 pollutants tested onto the 2 adsorbents. Maximum adsorption capacities are slightly higher for BPL carbon compared with OVC. Table 4 also shows maximum adsorption capacities determined from curve-fits of Langmuir isotherms to the experimental data (using coefficients from the non-linearized form). For BPL, the Langmuir model consistently overpredicts maximum

adsorption capacities compared with the experimental maximum capacities, particularly for ortho-xylene. The S shape of the ortho-xylene/BPL isotherm resembles a Brunauer type IV isotherm, which would not be fit particularly well by the Langmuir model. When isotherms do not have the right shape and only low concentration data is used, the Langmuir equation can provide estimates of adsorption maxima that are in error by 50% or more (Harter, 1984).

**Table 4.** Maximum adsorption capacities: comparison of experimentally determined values with Langmuir coefficient values and manufacturer values

VOC	Maximum adsorption capacity, g VOC adsorbed/g carbon					
	BPL			OVC		
	Exp. Data	Langmuir $q_{max}$	Manufacturer	Exp. Data	Langmuir $q_{max}$	Manufacturer
Ethyl benzene	0.0061	0.0067	0.41	0.0058	0.0055	0.39
Methyl ethyl ketone	0.0051	0.0053	0.38	0.0051	0.0049	0.38
Ortho-Xylene	0.0064	0.0119	0.42	0.0062	0.0078	0.40
Para-Xylene	0.0060	0.0075	0.44	0.0058	0.0057	0.44
Meta-Xylene	0.0059	0.0078	0.44	0.0057	0.0075	0.42

#### Comparison of adsorption capacities under simulated real-world conditions with capacities under ideal conditions

Isotherms for BPL and OVC were obtained from the manufacturer for comparison. Unlike the isotherms determined in this study, manufacturer isotherms were developed for ideal conditions, which included pre-treating the carbon by drying it in an oven and removing humidity from the gas stream. Experimentally-determined maximum adsorption capacities are compared with values from manufacturer isotherms in Table 4.

Isotherms developed in this research were similar to the manufacturer's isotherms in several respects:

- In both, OVC has a higher adsorption capacity at low concentrations but BPL has a slightly higher adsorption capacity at high concentrations.
- For both carbons, MEK has the lowest maximum adsorption capacity, and the other compounds have higher capacities, which are comparable in value.

However, the manufacturer's maximum adsorption capacities were significantly higher than the values determined in this research, by factors typically ranging from 50 to 70. This is likely due to the fact that experiments in this research were conducted to reflect real-world conditions, using ambient humidity and without pre-treating the carbon via heating to remove moisture.

Designing a system using manufacturer isotherms, rather than isotherms that reflect anticipated operating conditions, can lead to substantially overestimating the adsorption capacity of the system.

#### CONCLUSION

For MEK, BPL and OVC adsorption capacities were similar for all concentrations. For ethylbenzene and the xylene isomers, however, OVC coconut shell-based

carbon had a higher adsorption capacity at lower concentrations, but BPL coal-based carbon had a higher capacity at higher concentrations. This is likely due to the greater external surface area for OVC carbon, and the greater internal surface area for BPL carbon. For both carbons, at low concentrations adsorption capacity was highest for ethylbenzene, at intermediate concentrations highest for para-xylene, and at high concentrations highest for ortho-xylene. For ethylbenzene and MEK, the Freundlich isotherm fit the data better; for the xylenes, the Langmuir isotherm generally fit the data better. Manufacturer isotherms, developed for ideal conditions, predicted significantly higher maximum adsorption capacities than those determined in this research.

Other HAPs emitted from landfills should be tested, and combinations of HAPs should be tested to evaluate potential competitive adsorption. Testing adsorption of HAPs in the presence of carbon dioxide and methane, which are the principal constituents of landfill gas, should be conducted to determine whether competitive adsorption by methane or carbon dioxide poses a concern.

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