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Factors Controlling Alkylbenzene Sorption to Municipal Solid Waste

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The sorption of toluene and *o*-xylene to individual municipal solid waste (MSW) constituents [office paper, newsprint, model food and yard waste, high density polyethylene, and poly(vinyl chloride) (PVC)] was evaluated. Effects of sorbent decomposition and solvent composition on alkylbenzene sorption were studied by evaluating biodegradable sorbents in both fresh and anaerobically decomposed form and by complementing single-solute isotherm tests with experiments conducted in acidogenic and methanogenic leachate. Alkylbenzene sorption to plastics was greater than to biopolymer composites, and differences in sorbate/sorbent solubility parameter compatibility explained this observation. Alkylbenzene sorption to biopolymer composites yielded linear isotherms, and sorption capacities [log(K_{oc}/K_{ow})] decreased linearly with increasing sorbent polarity as expressed by the O-alkyl/alkyl ratio. Leachate composition had little effect on alkylbenzene sorption with one exception; volatile fatty acids in acidogenic leachate appeared to convert PVC from a glassy to a rubbery polymer. The results of this study showed that sorbent organic matter affinity for hydrophobic organic contaminants (HOCs) increases with increasing extent of MSW decomposition because of the recalcitrance of plastics and the preferential degradation of polar biopolymers. Furthermore, the plasticizing effect of volatile fatty acids in acidogenic leachate may enhance the bioavailability of HOCs sorbed to glassy organic matter in MSW or in soils contaminated with acidogenic leachate.

Introduction

Approximately 25% of the sites on the National Priority List (NPL) of Superfund are municipal landfills that accepted hazardous waste (*1*). Unlined landfills typically result in groundwater contamination, and priority pollutants such as alkylbenzenes are often present (*2*). Ultimately, strategies are needed to manage these landfills as well as thousands of sites not on the NPL in a manner that is both cost-effective and protective of the environment. To select cost-effective risk management alternatives, better information on factors controlling the fate of contaminants in landfills is required.

Sorption to organic municipal solid waste (MSW) components such as plastics and biopolymer composites (paper, food and yard waste, wood) is one factor that limits both the biodegradation (*3*) and transport (*4*−*7*) of hydrophobic organic contaminants (HOCs) in landfills. Biological processes in landfills lead to the degradation of biopolymer composites, whose principal constituents are cellulose, hemicellulose, lignin, proteins, and lipids (*8*, *9*). The resulting compositional changes affect HOC sorption because (i) the organic carbon content of the sorbents decreases (*9*, *10*) and (ii) the polarity of the remaining sorbent organic matter decreases as a result of the preferential degradation of more polar constituents such as cellulose and hemicellulose over less polar components such as lignin and lipids (*9*, *11*). As previously shown for sediments, biologically mediated increases in sorbent hydrophobicity can lead to enhanced HOC sequestration (*12*), and similar effects may be expected in landfills. Furthermore, contaminant aging can reduce bioavailability as contaminants diffuse into hard, glassy organic matter (*13*, *14*), a factor that is likely significant in landfills where HOCs are exposed to sorbents for long periods of time. Finally, dissolved organic matter in landfill leachate can affect HOC sorption to MSW. Leachate generated from MSW contains high concentrations of dissolved organic matter (*15*−*17*), and some dissolved organic compounds in landfill leachate can bind HOCs (*18*, *19*).

A mechanistic understanding of the factors controlling sorptive processes in landfills is limited. Landfills differ from more extensively studied environments involving geosorbents such as soils and sediments because MSW has a large organic carbon content (*20*), a large fraction of MSW consists of young organic matter that is readily biodegraded (*9*, *11*), and landfill leachates have a high dissolved organic carbon concentration (*15*−*17*). Consequently, the principal objective of this research was to elucidate factors that control the sorption of toluene and *o*-xylene in landfills. Specific objectives were to determine (i) the sorptive capacity of model MSW components, (ii) the effects of sorbent decomposition on sorbent hydrophobicity and contaminant sorption, and (iii) the effects of leachate composition on contaminant sorption.

Materials and Methods

MSW Components. MSW consists primarily of paper (38.6%), yard waste (12.8%), food waste (12.1%), plastics (9.9% with high- and low-density polyethylene as the major contributors), metals (7.7%), rubber/leather/textiles (6.8%), glass (5.5%), and wood (5.3%; *21*). Therefore, major components of MSW that are expected to sorb HOCs include paper, food and yard waste, and plastics. Newsprint and office paper were chosen to represent the range of paper types disposed in landfills. Newsprint is a mechanical pulp that contains nearly all of the lignin present in wood while office paper is a chemical pulp from which most of the lignin has been removed. Newsprint represents about 5.1% of MSW; and office paper represents 3.2% (*21*). Newsprint was collected from The News & Observer Recycling Division (News & Observer Publishing Co., Garner, NC). Office paper was collected from the North Carolina State University recycling center. Upon receipt, both office paper and newsprint were shredded into 2-cm squares. Rabbit food (Manna Pro Corporation, St. Louis, MO) was used to represent a combination of food and yard waste. Rabbit food contains both simple and complex carbohydrates as well as proteins in the form of alfalfa, wheat, soy, and oat products, and it has been used to represent food and yard waste previously (*22*). High-density polyethylene [HDPE] and poly(vinyl

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chloride) [PVC] (Catalog Nos. 42,799-3 and 18,958-8, Sigma-Aldrich Milwaukee, WI) were tested to represent rubbery and glassy plastic components of MSW, respectively.

Anaerobically degraded forms of office paper, newsprint, and rabbit food were prepared in laboratory reactors using leachate recycle and neutralization as described (*11*), except that substrates were seeded with anaerobic sewage sludge instead of well-decomposed refuse. Reactors containing shredded office paper and newsprint were operated for about 9 months to obtain materials that, based on their methane yield, were significantly decomposed relative to the starting materials. Reactors containing rabbit food were operated in semi-batch mode to control excessive volatile fatty acid (VFA) production that otherwise resulted from its rapid biodegradation. Over a 9-month period, rabbit food was added to reactors on a daily to weekly basis, depending on the stability of methane production in the system.

MSW Component Characterization. The analytical methods for cellulose, hemicellulose, and lignin (*23*) and lipophilic extractives (*24*) have been described. C, H, and N analyses were performed with a CHN analyzer (Perkin-Elmer PE2400 CHN Elemental Analyzer, Perkin-Elmer Corp., Norwalk, CT), and oxygen contents were measured according to ASTM D5622 (Huffman Laboratories, Golden, CO). To minimize interference from inorganic compounds, elemental analyses were performed on materials that had been washed with 1 N HCl until a constant pH was reached and rinsed with organic-free water until the pH returned to neutral. Crude protein content was calculated as 6.25 times the N content (*25*). Ash contents were determined by combusting 2 g of dry sample at 550 °C for 2 h. Brunauer, Emmett, and Teller (BET) surface areas were determined from N_2 adsorption data collected at 77 K (Autosorb 1-MP, Quantachrome Corp., Boynton Beach, FL). ^{13}C nuclear magnetic resonance spectra were obtained with a Chemagnetics CMX-II solid-state NMR spectrometer operating at 75.694 MHz and using a Chemagnetics 5-mm double resonance magic-angle spinning probe. A quasi-adiabatic sequence (*26*) using two-pulse phase modulation (TPPM) decoupling (*27*) at 75 kHz was employed. The ^{13}C cross polarization (CP) contact pulse of 1 ms length was divided into 11 steps of equal length with ascending radio frequency field strength, while the ^{1}H contact pulse had constant radio frequency field strength of 301.00 MHz. At least 720 scans were acquired with a delay of 15.0 s. The sample spinning frequency was 6.0 kHz, maintained to within a range of ±5 Hz or less with a Chemagnetics speed controller.

Sorbates. Toluene and *o*-xylene, two alkylbenzenes with different polarities, were selected because they are frequently detected in landfill leachates (*2*), sorb to refuse (*3*), and biodegrade (toluene; *3*) or have the potential to biodegrade (*o*-xylene; *28*) in the refuse ecosystem. Experiments were conducted with ^{14}C-labeled compounds (Sigma-Aldrich). Stock solutions were prepared by dissolving neat ^{14}C-labeled compounds in 6 mL of methanol (HPLC-grade, Fisher Scientific, Pittsburgh, PA) and stored at −10 °C. Toluene and *o*-xylene concentrations were assayed by liquid scintillation counting. Purity of ^{14}C-labeled compounds was assessed by gas chromatographic analysis that showed peaks corresponding to toluene and *o*-xylene only as well as sparging tests that showed that 99.85 and 99.4% of the ^{14}C was volatile in aqueous toluene and *o*-xylene solutions, respectively. The remaining nonvolatile ^{14}C counts were nonsorbable, and all data were corrected for this nonsorbing impurity.

Leachates. Acidogenic leachate was produced by recirculating water through fresh residential refuse (*29*). The leachate was not neutralized to maintain the refuse in the acid phase of decomposition, which was confirmed by a low leachate pH and the absence of gas production. Methanogenic leachate was generated by recirculating water through refuse in the methane phase of decomposition (*29*), which was confirmed by a leachate pH near neutral and methane production. Detailed leachate characteristics are provided with the results. Acidogenic leachate was filtered through 0.45-μm membrane filters prior to storage at 4 °C. Methanogenic leachate was stored in a 55-gal drum at 4 °C with minimum headspace. Leachate characterization included measurement of pH, dissolved organic carbon (DOC), UV absorbance at 254 nm, and chemical oxygen demand (COD) according to standard methods (*30*). Conductivity was measured using a conductivity meter (Fisher Scientific). High-pressure size exclusion chromatography was used to obtain both number- and weight-averaged molecular weights (*31*, *32*).

Isotherms. Isotherm experiments were conducted with individual MSW components after they had been ground in a Wiley mill to pass a 1-mm screen. PVC was obtained in powdered form (diameter = 140 μm) and used directly. All materials were oven-dried at 100−105 °C for 24 h and stored in a descicator until use. Flame-sealed glass ampules (20 mL) were used to collect batch isotherm data in phosphate-buffered (1 mM) organic-free water and in acidogenic and methanogenic leachates (*33*). Ampules were washed and baked in a muffle furnace at 325 °C overnight prior to use. Sodium azide (100 mg/L) was added as a microbial growth inhibitor to prevent aerobic sorbent and sorbate degradation. Isotherms experiments were conducted at initial sorbate concentrations of 30−1000 μg/L. Liquid:solid mass ratios ranged from 10:1 to 50:1 for fresh and degraded rabbit food, newsprint, and office paper, from 8:1 to 500:1 for HDPE, and from 10:1 to 1000:1 for PVC at each initial sorbate concentration. Using a gastight syringe, toluene and *o*-xylene were spiked into the suspension at the ampule bottom immediately prior to ampule sealing. Methanol contributions from toluene and *o*-xylene stock solutions were kept below 0.1% v/v in isotherm tests to avoid cosolvent effects (*34*). To reach sorption equilibria, ampules were tumbled end-over-end to ensure effective mixing. Once equilibrated, ampules were centrifuged at 2000 rpm for 20 min after which a 1-mL sample was removed for scintillation counting.

For isotherms conducted in leachates, all glassware was autoclaved prior to use. For isotherms in methanogenic leachate, ampules were filled with methanogenic leachate in an anaerobic hood to minimize oxygen effects. Upon removal from the anaerobic hood, samples were spiked immediately with toluene or *o*-xylene and flame-sealed.

Initially, tests were conducted to determine the time needed to reach sorption equilibria in buffered organic-free water and in leachates. Multiple replicate glass ampules were set up using one water:solid ratio and one initial sorbate concentration. Replicate ampules were sampled over 20 days for rabbit food, HDPE, newsprint, and office paper and up to 250 days for PVC. With the exception of PVC, no measurable changes in liquid-phase concentrations were detected after 5 days. Approximately 8 months were required to reach equilibrium with PVC.

Results and Discussion

Sorbent Characterization. Physical and chemical characteristics of the studied MSW components are summarized in Table 1. For biodegradable MSW components, the data indicate that biodegradation resulted in the preferential removal of cellulose and hemicellulose and the enrichment of lignin, consistent with the results of prior studies (*9*, *11*). Table 1 further shows that protein accumulated during degradation, suggesting that microbial cell components constituted a portion of the degraded materials. Relative to cellulose and hemicellulose, losses of lipophilic extractives were smaller, a result that can be explained by microbial resynthesis of lipids (cell walls, lipid bilayers of cell membranes) and the poor biodegradability of such lipids (*9*, *35*).

TABLE 1. Sorbent Characteristics[a]

sorbent	cellulose (%)	hemicellulose (%)	lignin (%)	lipophilic extractives (%)	crude protein (%)	ash (wt %)	f_{oc}	BET surface area (m^2/g)
poly(vinyl chloride)	na[b]	na	na	na	na	0.00	0.389	0.8
high-density polyethylene	na	na	na	na	na	0.01	0.876	0.6
fresh office paper	64.7	13.0	0.93	0.7	0.31	11.6	0.373	2.8
degraded office paper	36.2	6.9	4.8	3.3	4.99	38.4	0.278	6.0
fresh newsprint	48.3	18.1	22.1	1.6	0.44	2.0	0.451	2.6
degraded newsprint	35.1	16.0	32.3	1.4	3.74	6.4	0.455	3.4
fresh rabbit food	30.6	15.4	9.5	4.9	18.1	7.7	0.423	0.6
degraded rabbit food	7.1	5.7	25.2	4.5	20.6	34.5	0.329	0.5

[a] Values are averages of replicate analyses. [b] na, not analyzed.

TABLE 2. Elemental Composition of Acid-Washed Sorbents[a,b]

sorbent	(wt %) C	H	N	O	ash	total	(O+N)/C[c]
poly(vinyl chloride)[d]	38.9	4.7	0.1	1.7	0.0	102.1[e]	–[f]
high-density polyethylene[d]	87.6	14.5	0.08	0.1	0.01	102.3	–
fresh office paper	41.7	5.2	0.04	49.6	1.1	97.6	0.89
degraded office paper	41.7	5.0	0.8	43.4	7.6	98.5	0.80
fresh newsprint	45.0	4.9	0.07	47.5	2.3	99.8	0.79
degraded newsprint	47.6	5.0	0.4	43.7	2.0	98.7	0.70
fresh rabbit food	45.3	5.4	2.4	45.4	1.1	99.6	0.80
degraded rabbit food	48.8	5.3	3.2	39.0	2.9	99.2	0.66

[a] Values are averages of replicate analyses. [b] Sulfur content was not measured but should be 0.4% (wt/wt) or less for the tested materials (*75*). [c] Atomic ratio on a dry, ash-free basis. [d] HDPE and PVC were not acid-washed. [e] Includes theoretical chlorine content of 56.7%. [f] Ratios for PVC and HDPE are not meaningful because of very low oxygen and nitrogen contents.

Similar trends were recently observed during the anaerobic degradation of mixed waste (*9*). Ash contents increased upon biodegradation, a result that can be explained in part by the conversion of organic matter to CO_2 and CH_4 and in part by the addition of sodium carbonate for pH neutralization, especially during the preparation of degraded rabbit food. The organic carbon content (f_{oc}) of each biodegradable material was calculated from the carbon content of the acid-washed materials (Table 2) according to $f_{oc} = [C\ (wt\ \%)(1 - f_{ash,NAW})]/100 \times (1 - f_{ash,AW})$, where $f_{ash,NAW}$ is the ash content (wt %/100) of the nonacid-washed material (Table 1) and $f_{ash,AW}$ is the ash content of the acid-washed material (Table 2). For PVC and HDPE, materials with known organic carbon content, f_{oc} values in Table 1 closely correspond to the theoretical carbon contents of 0.384 and 0.857, respectively. For materials with low ash contents such as fresh and degraded newsprint, the difference between calculated f_{oc} values (Table 1) and directly measured f_{oc} values for nonacid-washed materials (not shown) was less than 1% ($\Delta f_{oc} \times 100\%$). Larger discrepancies were observed between calculated and directly measured f_{oc} values for nonacid-washed materials with higher ash contents, such as degraded office paper and degraded rabbit food, because the inorganic matrix contained a substantial amount of carbonates that affected the directly measured carbon content. Anaerobic degradation decreased the organic carbon contents of office paper and rabbit food by about 9.5% ($\Delta f_{oc} \times 100\%$) while that of newsprint remained approximately constant.

BET surface area data showed that the plastics as well as fresh and degraded rabbit food exhibited negligible internal porosity (Table 1). Nonetheless, glassy polymers such as PVC contain internal nanovoids that are inaccessible to N_2 at 77 K but that provide surfaces for HOC adsorption (*36*). BET surface areas of office paper and newsprint are similar to those of many soils and sediments (*37*, *38*), and they are in agreement with those previously observed for dry pulp fibers (*39*). While swelling of cellulose in water can increase the BET surface area of wet pulp fibers to 150 m^2/g (*39*), this surface area is unlikely to contribute significantly to HOC adsorption from aqueous solution because of the strong adsorption of water on polar surfaces.

TABLE 3. Distribution of Functional Groups in Biopolymer Composites Based on ^{13}C CP-MAS NMR Spectra

sorbent	carbonyl[a] (%)	aromatic[b] (%)	O-alkyl[c] (%)	alkyl[d] (%)	O-alkyl/alkyl
fresh office paper	0.9	3.4	94.0	1.7	55.3
degraded office paper	2.7	8.0	82.8	6.5	12.7
fresh newsprint	2.1	12.2	82.4	3.3	25.0
degraded newsprint	2.8	17.1	74.8	5.4	13.9
fresh rabbit food	7.8	3.4	79.6	9.3	8.6
degraded rabbit food	8.3	10.9	63.4	17.4	3.6

[a] Chemical shift: 200–160 ppm. [b] Chemical shift: 160–110 ppm. [c] Chemical shift: 110–50 ppm. [d] Chemical shift: 50–0 ppm.

The elemental composition of the sorbent organic matter is summarized in Table 2. The sum of the C, H, N, O, and ash contents in Table 2 resulted in recoveries of 97.6–102.3%, indicating that the analytical methods effectively characterized the sorbent composition. Elemental data in Table 2 were utilized to characterize sorbent polarity by means of the polarity index [(O+N)/C] (*40*). For biodegradable materials, the polarity index values illustrate that the sorbent organic matter became more hydrophobic during anaerobic decomposition, which is in agreement with the preferential degradation of cellulose and hemicellulose over lignin, protein, and lipophilic extractives as shown in Table 1 as well as with the ^{13}C CP-MAS NMR data presented in Table 3. Upon degradation, NMR signals in the carbonyl region (amides, carboxyl groups in fatty acids), in the aromatic region (lignin), and in the alkyl region (methylene groups in fatty acids, protein side chains) increased while the O-alkyl signals attributable to cellulose and hemicellulose decreased. As a result, the ratio of O-alkyl to alkyl groups, which has been

TABLE 4. Leachate Characteristics

leachate	COD (mg L^{-1})	DOC (mg L^{-1})	UV_{254} (m^{-1})	SUVA (L mg^{-1} m^{-1})	pH	conductivity ($\mu\Omega^{-1}$ cm^{-1})	av mol wt (g/mol) M_w	M_n
acidogenic	22400	6620	10.4	0.00157	4.9	6540	856	181
methanogenic	550	115	3.5	0.0304	7.8	1850	5849	293

TABLE 5. Isotherm Parameters Describing Toluene Sorption to MSW Components

sorbent	N^a	log K_F [b]	n^c	R^2	K_p [d]	R^2	K_{oc} [e]
poly(vinyl chloride)	41	3.221 (± 0.045)	0.864 (± 0.020)	0.994	546.6 (± 38.3)	0.912	4276[f]
high-density polyethylene	54	1.826 (± 0.047)	1.012 (± 0.023)	0.993	70.7 (± 3.7)	0.946	80.7
fresh office paper	27	0.642 (± 0.079)	0.923 (± 0.035)	0.991	2.7 (± 0.2)	0.957	7.2
degraded office paper	42	1.400 (± 0.020)	0.988 (± 0.010)	0.999	23.4 (± 0.3)	0.995	83.1
fresh newsprint	44	1.220 (± 0.027)	0.961 (± 0.015)	0.998	13.0 (± 0.2)	0.997	28.8
degraded newsprint	42	1.464 (± 0.029)	0.980 (± 0.016)	0.997	25.8 (± 0.4)	0.994	56.7
fresh rabbit food	46	1.450 (± 0.020)	0.996 (± 0.011)	0.997	27.5 (± 0.2)	0.999	65.0
degraded rabbit food	40	1.491 (± 0.067)	0.976 (± 0.027)	0.993	27.3 (± 0.9)	0.982	82.9

[a] Number of observations. [b] K_F units are (μg/kg)(L/μg)^n; 95% confidence interval of log K_F in parentheses. [c] Dimensionless Freundlich exponent; 95% confidence interval in parentheses. [d] Partition coefficient units are (μg/kg)(L/μg); 95% confidence interval in parentheses. [e] K_{oc} units are (μg/kg of organic carbon)(L/μg). [f] Based on K_F, units are (μg/kg of organic carbon)(L/μg)^n.

TABLE 6. Isotherm Parameters Describing *o*-Xylene Sorption to MSW Components

sorbent	N^a	log K_F [b]	n^c	R^2	K_p [d]	R^2	K_{oc} [e]
poly(vinyl chloride)	33	3.666 (± 0.056)	0.718 (± 0.034)	0.983	777.7 (± 58.4)	0.934	11910[f]
high-density polyethylene	48	2.386 (± 0.021)	1.004 (± 0.012)	0.998	244.1 (± 3.7)	0.996	278.7
fresh office paper	41	0.782 (± 0.055)	0.985 (± 0.027)	0.993	5.7 (± 0.1)	0.992	15.3
degraded office paper	62	1.621 (± 0.033)	1.014 (± 0.019)	0.995	44.5 (± 1.2)	0.976	160.1
fresh newsprint	35	1.522 (± 0.024)	0.988 (± 0.012)	0.999	31.1 (± 0.4)	0.998	69.0
degraded newsprint	39	1.851 (± 0.021)	0.944 (± 0.012)	0.999	52.7 (± 0.7)	0.997	115.8
fresh rabbit food	44	1.837 (± 0.033)	1.010 (± 0.017)	0.996	76.5 (± 1.4)	0.994	180.9
degraded rabbit food	39	1.856 (± 0.062)	1.009 (± 0.035)	0.989	79.3 (± 4.4)	0.954	241.0

[a] Number of observations. [b] K_F units are (μg/kg)(L/μg)^n; 95% confidence interval of log K_F in parentheses. [c] Dimensionless Freundlich exponent; 95% confidence interval in parentheses. [d] Partition coefficient units are (μg/kg)(L/μg); 95% confidence interval in parentheses. [e] K_{oc} units are (μg/kg of organic carbon)(L/μg). [f] Based on K_F, units are (μg/kg of organic carbon)(L/μg)^n.

employed as an indicator of the extent of natural organic matter degradation (*41*), decreased upon anaerobic decomposition of the biodegradable materials (Table 3).

Leachate Characterization. Leachate characteristics are presented in Table 4. The COD and pH data are consistent with previous reports on acidogenic and methanogenic leachate (*2*). In acidogenic leachate, VFAs constituted 63% of the COD. The dominant VFA was butyric acid (3300 mg/L), followed by acetic, propionic, and valeric acids (~1500 mg/L each). In methanogenic leachate, no humic acid was present, and 57% of the DOC was fulvic acid (retained by XAD-8 column with $k'_{0.5r} = 5$; *42*). Compared to the high DOC content of the leachates, UV_{254} absorbance values were small, suggesting that aromatic and unsaturated functionalities were largely absent in both acidogenic and methanogenic leachate organic matter. Consequently, specific UV absorbance values (SUVA = UV_{254}/DOC) were small and about 2−3 orders of magnitude lower than in natural waters with significant aquatic humic content (*43*). Number-averaged molecular weights (M_n) indicated that relatively low molecular weight organic matter dominated among UV_{254}-absorbing compounds in both leachates. In agreement with prior studies (*16*), M_n for acidogenic leachate was smaller than for methanogenic leachate. Weight-averaged molecular weights (M_w) illustrated that high molecular weight UV_{254}-absorbing organic matter was present in methanogenic leachate but not in acidogenic leachate.

Single-Solute Isotherms. Single-solute isotherms were obtained for toluene (Table 5) and *o*-xylene (Table 6) with each model MSW component, and *o*-xylene isotherm data for fresh materials are depicted in Supporting Information Figure S1. Equilibrium liquid-phase concentrations (C_e) for each isotherm spanned 2−3 orders of magnitude and relative to the aqueous solubility (S_w) of toluene (518 mg/L; *44*) and *o*-xylene (185 mg/L; *44*), the largest relative concentrations (C_e/S_w) corresponded to about 0.002 and 0.005 for toluene and *o*-xylene, respectively. The tested relative concentration ranges were sufficiently low to assess any isotherm nonlinearity arising from the presence of glassy polymeric domains or high surface area carbonaceous materials in the tested sorbents (*36*, *45*−*47*).

To mathematically describe the isotherm data, the nonlinear Freundlich model ($\log q = \log K_F + n \log C_e$) and the linear partition model ($q = K_p C_e$) were compared, and the resulting single-solute isotherm parameters for toluene and *o*-xylene are summarized in Tables 5 and 6, respectively. In these models, q (μg/kg) and C_e (μg/L) represent the equilibrium solid- and liquid-phase concentrations, respectively. In the two-parameter Freundlich isotherm model, K_F is indicative of sorption capacity at $C_e = 1$ μg/L (for the units chosen in this study), and n indicates the site energy distribution of a sorbent where sorbent heterogeneity increases as n decreases from 1 (*48*). The relatively high *o*-xylene sorption capacity of PVC (K_F values in Table 6; Figure S1) can be explained on the basis of solubility parameters (δ), where sorbate/sorbent systems with matching solubility parameters represent a compatible solute/solvent system (*49*). Thus, PVC [$\delta = 7.8-11$ (cal/cm^3)$^{1/2}$; *49*] represented a suitable sorbent for *o*-xylene [$\delta = 8.9$ (cal/cm^3)$^{1/2}$; *49*] because its solubility parameter range includes that of *o*-xylene. Furthermore, a nonlinear adsorption component on the surfaces of internal nanovoids contributed to *o*-xylene uptake

by the glassy PVC polymer (*50*). HDPE exhibited a smaller *o*-xylene sorption capacity than PVC because the solubility parameter for *o*-xylene falls outside of the range for HDPE [δ = 7.7−8.2 (cal/cm^3)$^{1/2}$; *49*] and because an adsorption component was absent for this rubbery polymer. For the biopolymer-containing sorbents, rabbit food exhibited the largest *o*-xylene sorption capacity, most likely because of its relatively high lipophilic extractives content (Table 1). The solubility parameter for soybean oil [δ = 8.4−8.9 (cal/cm^3)$^{1/2}$; *51*], which should be present in rabbit food based on its ingredients, is similar to that of *o*-xylene. Furthermore, the presence of lignin [δ = 9.8−14.3 (cal/cm^3)$^{1/2}$; *52*] and protein, which exhibits HOC sorption capacities similar to lignin (*53*), contributed to *o*-xylene sorption to rabbit food. Consistent with solubility parameters, newsprint with a 22% lignin content exhibited a larger sorption capacity than office paper that contained primarily cellulose [δ = 14.5−16.5 (cal/cm^3)$^{1/2}$; *49*] and hemicellulose. Overall, the biopolymer composites exhibited lower sorption capacities than plastics because (i) the differences in solubility parameters between *o*-xylene and lignin or cellulose were greater than those between *o*-xylene and PVC or HDPE, and (ii) the content of highly compatible sorbent fractions, such as lipophilic extractives, was small in the biopolymer composites. The order of sorbent affinity for toluene was the same as that for *o*-xylene, a result that can be expected given that the solubility parameter of toluene [δ = 8.8 (cal/cm^3)$^{1/2}$; *49*] is similar to that of *o*-xylene.

With the exception of the toluene and *o*-xylene isotherms for PVC and the toluene isotherm for fresh office paper, Freundlich exponents in Tables 5 and 6 varied between 0.94 and 1.01, indicating that the organic sorbents acted primarily as partitioning media. Because the *o*-xylene isotherm for fresh office paper exhibited a Freundlich exponent that did not differ significantly from 1 ($p < 0.05$), it is doubtful that the Freundlich exponent of 0.92 for toluene sorption to fresh office paper is indicative of true isotherm nonlinearity. Instead, a smaller number of data points and the greater scatter of the toluene isotherm data for this poorly sorbing material may have contributed to this observation. There are two additional reasons why linear toluene sorption to office paper would be expected. First, fresh office paper consists primarily of cellulose, and amorphous regions in cellulose are plasticized by water at room temperature (*54*). Consequently, wet cellulose is a partitioning medium and should exhibit linear isotherms (*55*). Second, the presence of small quantities of hydrophobic sizing agents on fresh office paper, such as alkyl ketene dimers of C_{14}−C_{18} unsaturated fatty acids (*39*), should lead to linear partitioning.

Because lignin constituted a large percentage of fresh and degraded newsprint and lipophilic extractives were low, it can be assumed that lignin controlled alkylbenzene sorption to newsprint. HOC sorption to lignin, a glassy polymer, should be nonlinear, and a Freundlich *n* value of about 0.80 was recently obtained for phenanthrene sorption to extracted lignin (*55*). The greater isotherm linearity observed in this study with newsprint may have resulted from (i) the topological or covalent bonding between hemicellulose and lignin in wood and wood pulp (*56*−*59*) that led to a new glass transition temperature for the intimately mixed lignin-carbohydrate complex (*60*) and/or (ii) the introduction of hydrophilic sulfone groups into the lignin structure during the preparation of newsprint (e.g., bisulfite addition during pulping) that enhanced the ability of water to plasticize lignin (*39*).

In fresh and degraded rabbit food, linear partitioning into phases containing lipophilic extractives was likely dominant and rendered the overall isotherms linear. Furthermore, proteins represented a partitioning medium because amorphous protein regions are plasticized by water at room temperature (*61*, *62*). Finally, the behavior of lignin−carbohydrate complexes in rabbit food may have deviated from that of glassy lignin extracts (*55*) as explained above. Both toluene and *o*-xylene sorption to PVC was nonlinear, consistent with the dual-mode partitioning and adsorption mechanism (*37*) that is typically observed for glassy polymers (*47*, *50*). For HDPE, the observation of linear toluene and *o*-xylene isotherms was consistent with previously observed linear partitioning into rubbery polymers (*63*).

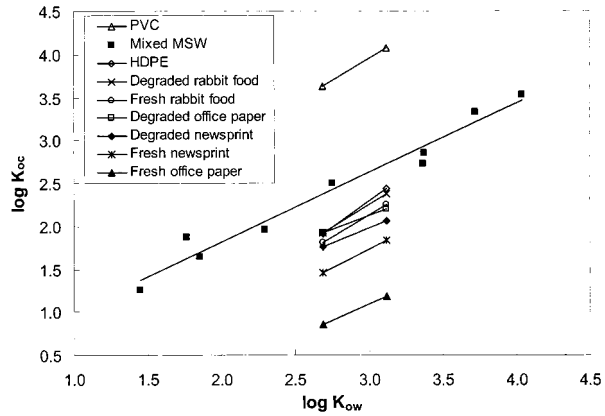

FIGURE 1. Dependence of log K_{oc} on log K_{ow} for individual MSW components and mixed MSW. PVC data represent Freundlich K_F values divided by f_{oc} and provide a valid comparison only at an equilibrium liquid-phase concentration of 1 μg/L. Data for mixed MSW are from ref *10*.

Tables 5 and 6 also summarize partition coefficients (K_p) for toluene and *o*-xylene, respectively. Except for PVC, for which the Freundlich isotherm model was more appropriate, the partition model described the isotherm data well over the entire liquid-phase concentration range (Figure S1). To compare the affinity of the sorbent organic matter for toluene and *o*-xylene, K_{oc} values ($K_{oc} = K_p/f_{oc}$) were therefore calculated from the partition coefficients for all materials but PVC, for which K_{oc} was estimated from K_F. Thus, K_{oc} comparisons involving PVC are valid only at an equilibrium liquid-phase concentration of 1 μg/L. Relating K_{oc} values to sorbate K_{ow} values, Figure 1 illustrates that K_{oc} values increased with increasing sorbate hydrophobicity (i.e., with increasing octanol/water partition coefficient, K_{ow}), a trend that has been established previously for mixed MSW (Figure 1; *10*) as well as for soils and sediments (*64*, *65*). For a given sorbate, the large differences in K_{oc} values between fresh MSW components as well as between fresh and degraded materials (Figure 1; Tables 5 and 6) illustrate that the chemical nature of the sorbent organic matter greatly influenced HOC sorption and that a unique relationship between log K_{oc} and log K_{ow} could therefore not be obtained. Figure 1 also shows that, with the exception of PVC, the individually tested MSW components exhibited a smaller sorption capacity than a mixed MSW sample from a prior study (*10*). The ethyl ether extractable lipid content of the mixed MSW sample was 4.6% (*10*) and thus similar to that of rabbit food evaluated in this study. Consequently, differences in the lipophilic extractives contents alone could not account for the observed differences in K_{oc} values. Furthermore, differences in equilibration time (24 h for mixed MSW samples (*10*), 5 d or longer in the current study) could not explain the comparatively large sorption capacity of mixed MSW. The results in Figure 1 therefore illustrate that sorbent organic matter characteristics need to be considered in addition to sorbate hydrophobicity to predict HOC sorption capacities of complex organic sorbents.

One sorbent characteristic that controls HOC sorption is the sorbent polarity, as measured by the polarity index [(O+N)/C] (*40*, *66*, *67*) or by the percentage of polar organic carbon determined from solid-state ^{13}C CP-MAS NMR spectra

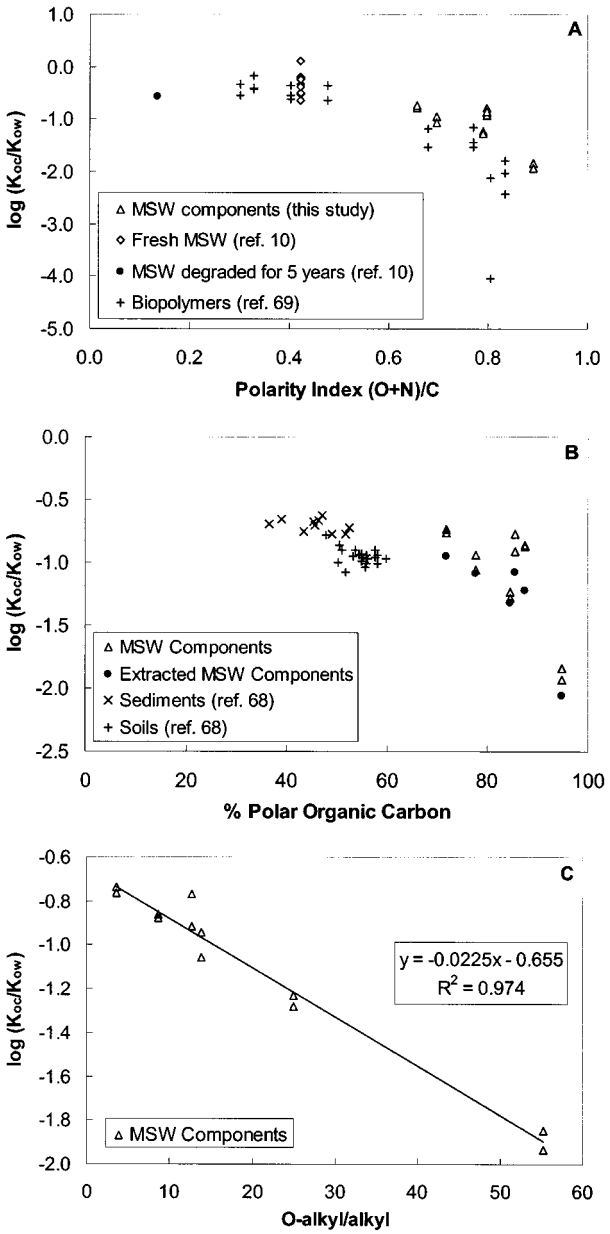

FIGURE 2. Dependence of normalized sorption parameter on sorbent polarity as expressed by (A) the polarity index [(O+N)/C], (B) % polar organic carbon (= O-alkyl + carbonyl from Table 3), and (C) O-alkyl/alkyl ratio (from Table 3).

(*68*). Figure 2A depicts the dependence of the normalized sorption parameter [$\log(K_{oc}/K_{ow})$] (*69, 70*) on the polarity of the sorbent organic matter as measured by the polarity index. In addition to the biodegradable MSW component data, Figure 2A depicts HOC sorption data for mixed MSW (*10*) and biopolymers (*40, 53, 66,* as summarized in *69*). The results in Figure 2A suggest that HOC sorption to individual MSW components and mixed MSW follows a trend similar to that established by individually tested biopolymers such as lignin, protein, and cellulose. Furthermore, the results in Figure 2A illustrate that the lower polarity of the mixed MSW sample (*10*) relative to that of the biodegradable MSW components evaluated in this study can explain the relatively large HOC sorption capacity of the mixed MSW sample (Figure 1).

Figure 2B depicts the dependence of $\log(K_{oc}/K_{ow})$ on the percentage of polar organic carbon in sorbent organic matter. For reference, Figure 2B also includes carbon tetrachloride sorption capacities for soils and sediments (*68*). The latter

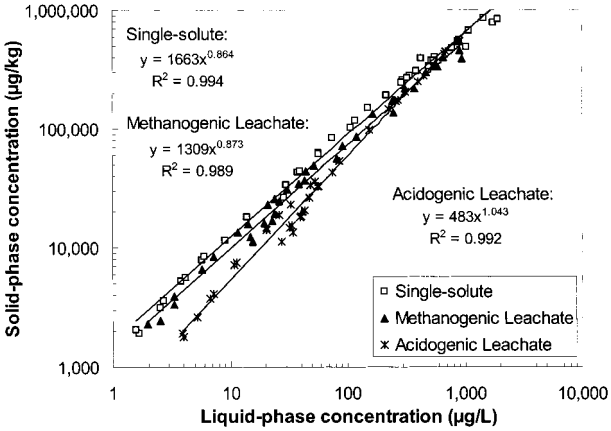

FIGURE 3. Effect of aqueous-phase composition on toluene sorption to PVC.

data suggest that sorbent polarity effectively describes HOC sorption trends for both relatively young sorbent organic matter, such as that in MSW, and more diagenetically altered sorbent organic matter such as that in soils and sediments. Furthermore, the similarity of panels A and B (Figure 2) illustrates that the polarity index and percent organic carbon are equivalent descriptors of sorbent polarity.

Although the overall dependence of $\log(K_{oc}/K_{ow})$ on sorbent polarity is clearly established in panels A and B (Figure 2), a closer look at the MSW component data illustrates a fair amount of variability. Two sorbents in particular, fresh rabbit food and degraded office paper, exhibited alkylbenzene sorption capacities that were larger than those expected from the trend established by the other biopolymer composites evaluated in this study. The toluene sorption capacities of solvent-extracted sorbents in Figure 2B indicate that relatively small amounts of lipophilic extractives (<5% of the sorbent mass) can cause large differences in sorption capacity, and these differences are not captured well by the polarity index or the percentage of polar organic carbon. To recognize the importance of both sorbent polarity and lipophilic extractives on HOC sorption, the ratio of O-alkyl to alkyl groups, determined from solid-state ^{13}C CP-MAS NMR spectra (Table 3), was therefore selected as a new sorbent organic matter descriptor. As shown in Figure 2C, an improved correlation resulted when relating $\log(K_{oc}/K_{ow})$ to the O-alkyl/alkyl ratio. It appears reasonable to focus on the O-alkyl and alkyl regions of the NMR spectra because these regions unambiguously represent polar and nonpolar organic matter, respectively. In contrast, carbonyl and aromatic moieties can be present in relatively hydrophilic and hydrophobic compounds. While the O-alkyl/alkyl ratio appears to be a good predictor for sorbents containing relatively young organic matter, i.e., materials with significant cellulose and hemicellulose content, it may not be sufficiently sensitive for geosorbents that have undergone more extensive diagenesis and thus contain few O-alkyl groups. In the limit, the correlation in Figure 2C predicts a $\log(K_{oc}/K_{ow})$ value of -0.66, which is quite similar to the plateau region [average $\log(K_{oc}/K_{ow}) = -0.44$] established by the eight least polar biopolymers in Figure 2A and the largest $\log(K_{oc}/K_{ow})$ value of -0.62 for the sediments shown in Figure 2B. It should be recognized, however, that HOC sorption to highly diagenetically altered geosorbents such as coals and shale has yielded positive $\log(K_{oc}/K_{ow})$ values (*69, 71*).

Leachate Effects. To evaluate the effects of leachate composition on alkylbenzene sorption to MSW components, isotherm data were collected for selected sorbents in acidogenic and methanogenic leachates. Figure 3 summarizes isotherm data describing toluene sorption to PVC in organic-free water and in the leachates. Toluene isotherms in organic-

free water and in methanogenic leachate were statistically similar ($p < 0.05$) with respect to isotherm nonlinearity and differed only slightly with respect to sorption capacity. Thus, the composition of methanogenic leachate had little effect on toluene sorption to PVC. In contrast, the toluene isotherm became linear in acidogenic leachate (Figure 3). This result suggests that organic compounds in acidogenic leachate served as plasticizers that converted PVC from the glassy to the rubbery state. The softening of glassy polymers by high concentrations of cosolutes in multisolute systems has been reported previously (*72*, *73*). In acidogenic leachate, propionic and butyric acid may have been the principal plasticizers of PVC because their solubility parameters [$\delta = 9.7$ and 9.1 $(cal/cm^3)^{1/2}$, respectively; *74*] fall into the range of δ values given above for PVC. Table S1 in the Supporting Information summarizes K_p values describing toluene and *o*-xylene sorption to sorbents that exhibited linear partitioning in single-solute systems. A comparison of single-solute K_p values (Tables 5 and 6) with those obtained in leachates (Table S1 in Supporting Information) illustrates that the effects of leachate on toluene and *o*-xylene sorption were small (compared to single-solute values, K_p values in leachates were 11% larger to 26% smaller).

Environmental Significance. The results of this study illustrate that toluene and *o*-xylene sorption to MSW is strongly influenced by the presence of plastics and lipophilic compounds. The recalcitrance of plastics combined with the microbial synthesis of poorly degradable lipids should therefore increase the affinity of sorbent organic matter for HOCs as biologically mediated sorbent degradation proceeds in landfills (i.e., K_{oc} increases over time). Consequently, HOC bioavailability may decrease as sorbents degrade and HOCs are sequestered in glassy (*50*) and/or paraffinic (*12*) sorbent organic matter. It is difficult to predict how HOC concentrations in landfill leachate may behave over time as increases in sorbent organic matter hydrophobicity are counterbalanced by losses of sorbent organic matter as a result of biogasification. A more definitive statement can be made for fresh MSW that is relatively nonpolar, e.g., the MSW evaluated by Reinhart et al. (*10*), for which increases in sorbent hydrophobicity as a result of biopolymer degradation had little effect on K_{oc}, as illustrated by the plateau region in Figure 2A. Consequently, the loss of sorbent organic matter due to biogasification decreased the overall HOC sorption capacity of MSW over time, suggesting that leachate HOC concentrations would increase.

The results obtained with acidogenic leachate further illustrate that, during the early stages of waste decomposition, elevated VFA levels can plasticize glassy sorbent organic matter and may thus increase HOC mass transfer rates from VFA-plasticized sorbents into leachate. To better address the environmental significance of sorptive processes, additional studies are needed to quantify desorption rates of aged alkylbenzenes from fresh and degraded sorbents into acidogenic and methanogenic leachate.

Acknowledgments

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Supporting Information Available

One figure depicting *o*-xylene isotherms and one table providing toluene and *o*-xylene partition coefficients for selected sorbents in acidogenic and methanogenic leachates. This material is available free of charge via the Internet at http://pubs.acs.org.

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Table S1. Partition coefficients describing toluene and *o*-xylene sorption to MSW components in acidogenic and methanogenic leachate.

Sorbent	toluene				*o*-xylene			
	acidogenic leachate		methanogenic leachate		acidogenic leachate		methanogenic leachate	
	K_p[a]	R^2	K_p[a]	R^2	K_p[a]	R^2	K_p[a]	R^2
High density polyethylene	76.1 (± 1.6)	0.992	72.4 (± 0.8)	0.998				
Fresh office paper	2.0 (± 0.1)	0.909						
Degraded office paper	19.9 (± 0.2)	0.997	20.8 (± 0.3)	0.996				
Fresh newsprint	13.4 (± 0.4)	0.986	13.3 (± 0.3)	0.991	34.0 (± 0.3)	0.999	30.1 (± 0.4)	0.998
Degraded newsprint	20.5 (± 0.5)	0.992	22.8 (± 0.2)	0.998	46.2 (± 0.6)	0.997	46.5 (± 0.6)	0.998
Fresh rabbit food	30.4 (± 0.4)	0.997	24.4 (± 0.4)	0.997	69.9 (± 0.6)	0.999	58.5 (± 1.6)	0.979

[a] Partition coefficient units are (μg/kg)(L/μg), 95% confidence interval in parentheses.

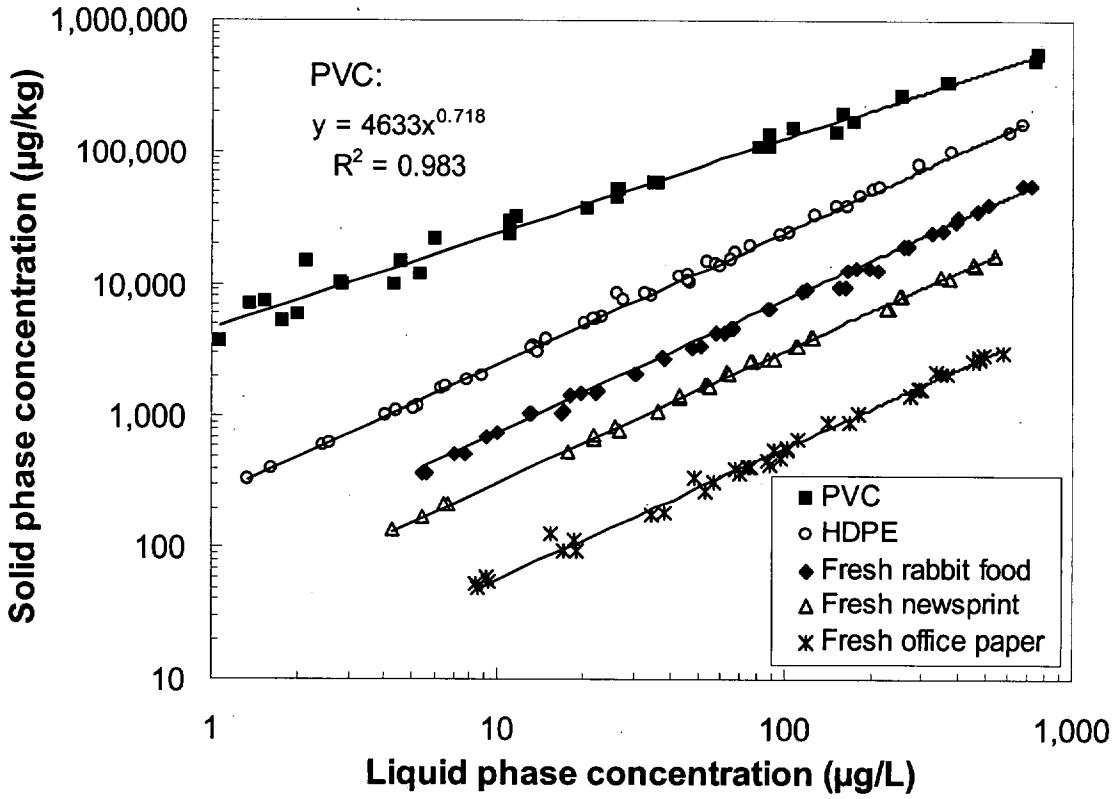

Figure S1. Single-solute *o*-xylene isotherms for model MSW components. Solid line through PVC data indicates the best fit of the Freundlich isotherm equation. Solid lines through all other data sets indicate best fits of the linear partition model; partition coefficients are shown in Table 6.