- **1** Supporting Information:
- 2 Measurement and Modeling of Activated Carbon Performance for the
- 3 Sequestration of Parent- and Alkylated-Polycyclic Aromatic
- 4 Hydrocarbons in Petroleum-Impacted Sediments
- 5
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#### 13 Details on the experimental procedure for sediment characterization

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15 The concentrations of polycyclic aromatic hydrocarbons (PAHs) in the sediment samples 16 were determined by ultrasonic extraction following USEPA Method 3550B as described elsewhere<sup>1</sup>. The concentrations of 18 parent-PAHs and 20 groups of alkylated-PAH 17 18 isomers in the extracts were determined by an analytical method provided in the 19 following section. The oil and grease contents and total petroleum hydrocarbons of the 20 sediments were analyzed in ACZ Laboratories (Steamboat Springs, CO). The oil and 21 grease contents were determined as the *n*-hexane extractable material by Soxhlet 22 extraction following USEPA Method 9071B. The total petroleum hydrocarbons were 23 analyzed following USEPA Method 8015D after extracting the sample by Soxhlet 24 extraction (USEPA Method 3540). The total organic carbon content was analyzed in 25 Agvise Laboratories (Northwood, ND) by a carbon analyzer. The black carbon content 26 was analyzed in Dr. Upal Ghosh's lab at the University of Maryland Baltimore County 27 (Catonsville, MD) following the wet chemical oxidation method using sulfuric acid and potassium dichromate as described in Grossman and Ghosh<sup>2</sup>. 28

# 30 Details on the PAH analytical method

32	As described in the main text, the analysis of parent- and alkylated-PAHs was performed
33	using a Gas Chromatograph-Mass spectrometer (GC-MS; Agilent model 6890 and 5973N,
34	respectively). Firstly, identification of each compound was performed in a scan mode.
35	Retention times for 18 parent- and 22 alkylated-PAH compounds were determined using
36	commercially available standards. For alkylated-PAHs of which standards could not be
37	obtained, an extract of S-1 sediment was used for identification. Each peak of the extract
38	was identified referring to the literature <sup>3</sup> and the NIST mass spectral library.
39	Consequently, retention time windows were determined for following groups of
40	alkylated-PAH isomers: C1-C4 naphthalenes, C1-C4 fluorenes, C1-C4 phenanthrenes/
41	anthracenes, C1-C4 fluoranthenes/pyrenes, and C1-C4 benz[a]anthracenes/chrysenes, the
42	numbers referring to the numbers of aliphatic carbons attached to the corresponding
43	parent-PAHs.
44	Quantification of PAHs was performed in selected ion monitoring mode for better
45	sensitivity. For alkylated-PAHs, response factors determined for the 22 commercially
46	available standards were used to estimate the average response factor for each group of
47	isomers. When no standards were available, the response factor of the closest group was
48	used as suggested by Stout and Wang <sup>4</sup> and Hawthorne et al. <sup>5</sup> . The PAH analytical method
49	developed in this study allows quantitation of 18 parent-PAHs and 20 groups of
50	alkylated-PAH isomers. Additionally, 22 alkylated-PAH compounds with commercially
51	available standards can be analyzed individually using the response factor determined for
52	each compound.

#### 53 Details on mass transfer modeling

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55 According to Werner et al.<sup>6</sup>, the overall mass balance in a sediment slurry where

56 activated carbon (AC) and polyethylene (PE) passive samplers are added can be written

57 as

$$\frac{dC_{w}}{dt} = -\frac{V_{s}}{V_{w}} \cdot \frac{d}{dt} \left[ \frac{3}{R_{s}^{3}} \left\{ \int_{0}^{R_{s}} r^{2} S_{s,fast}(r) dr + \int_{0}^{R_{s}} r^{2} S_{s,slow}(r) dr \right\} \right] - \frac{V_{AC}}{V_{w}} \cdot \frac{d}{dt} \left[ \frac{3}{R_{AC}^{3}} \int_{0}^{R_{AC}} r^{2} S_{AC}(r) dr \right] - \frac{V_{PE}}{V_{w}} \frac{2k_{o}}{x_{PE}} \left( C_{w} - \frac{S_{PE}}{K_{PE}} \right)$$
(1)

58 where  $C_w$  (mol cm<sup>-3</sup>) is the aqueous concentration,  $S_{s,fast}$ ,  $S_{s,slow}$ ,  $S_{AC}$  and  $S_{PE}$  are the 59 volumetric concentrations (mol cm<sup>-3</sup>) in fast- and slow-releasing sediment fractions, in 60 AC, and in PE, respectively;  $V_w$ ,  $V_s$ ,  $V_{AC}$  and  $V_{PE}$  are the volumes (cm<sup>3</sup>) of water,

61 sediment, AC and PE, respectively;  $R_s$  and  $R_{AC}$  are the radii (cm) of sediment and AC,

62 respectively;  $k_o$  (cm s<sup>-1</sup>) is the overall mass transfer coefficient for the PE-water boundary

63 layer; and  $x_{PE}$  is the PE thickness (cm). The sediment domain is assumed to be divided

64 into two fractions, fast- and slow-releasing fractions, operationally defined by PAH

65 releasing kinetics observed in desorption tests. The boundary layer model is applied for

66 the mass transfer between PE and water as suggested by many researchers<sup>7-10</sup>. For each

67 particulate domain, i.e., the two sediment fractions and AC, an intra-particle diffusion

68 model is applied for the sorption-desorption kinetics as developed by Wu and

69 Gschwend<sup>11</sup> and described elsewhere<sup>6, 12</sup>. The boundary conditions for the particulates

70 were set by assuming no flux at the center of the aggregate and sorption equilibrium at

71 the aggregate surface. The Matlab code used in this study for the simulation of PAH mass

72 transfer can be found in Werner et al.<sup>6</sup>

### 73 Details on the laboratory experiments for mass parameter determination

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75 An aqueous equilibrium test was carried out for S-1 sediment to determine the aqueous equilibrium concentrations ( $C_w$ , ng g<sup>-1</sup>) and the sediment-water distribution coefficients 76  $(K_d, \text{cm}^3 \text{g}^{-1})$ . In triplicate 1-L bottles, 950 mL artificial seawater (20 parts per thousand; 77 ppt) with 1 g  $L^{-1}$  sodium azide and 50 g dry wt. sediment were added. After rolling for 14 78 79 days at 2 rpm, the bottle was taken and the aqueous phase was separated by coagulation followed by centrifugation as described in Ghosh et al.<sup>13</sup>. The water samples were 80 81 extracted for three times by 40 mL hexane each. The wet sediment samples were also 82 taken from the bottles and dried, crushed, and extracted by ultrasonic extraction 83 following USEPA Method 3550B. Assuming linear partitioning between sediment and 84 water, the sediment-water distribution coefficients are calculated by

$$K_d = \frac{C_{sed}}{C_w} \tag{2}$$

85 where  $C_{sed}$  is the concentration in sediment (mol g<sup>-1</sup>).

86 The parameters related to the release of the model compounds from S-1 sediment 87 were determined by sediment desorption tests following a previously described procedure<sup>14</sup>. In 40 mL vials, triplicate 1.0 g dry wt. sediment samples were contacted 88 with 1.0 g of Tenax<sup>®</sup> beads (60-80 mesh; Supelco, Bellefonte, PA) in a slurry phase up to 89 90 56 days. The beads were collected and replenished with new beads at time intervals (0.25,0.5, 1, 3, 7, 28, and 56 days). After the final sampling of the Tenax<sup>®</sup> beads, the remaining 91 92 sediment was collected and analyzed for residual PAH concentration by ultrasonic 93 extraction for mass balance purposes. The results were fitted to a sediment desorption kinetic model described in Grathwohl<sup>15</sup> assuming two contaminant releasing fractions in 94

sediment, i.e., fast- and slow-releasing fractions. The mass transfer parameters, fraction of the slow-releasing sediment particles ( $f_{slow}$ , -) and compound release rates from fastand slow-releasing sediment particles ( $k_{slow}$  and  $k_{fast}$ , respectively, s<sup>-1</sup>) were determined by fitting the model with the experimental results. The sediment desorption tests were also conducted for S-2 sediment for comparison of those parameters.

100 The kinetic and equilibrium parameters of AC sorption, i.e., AC-clean water partitioning coefficients ( $K_{AC \ clean}$ , cm<sup>3</sup> g<sup>-1</sup>) and effective intra-particle diffusivity of AC 101  $(D_{AC,eff}, \text{cm}^2 \text{ s}^{-1})$ , were measured using PE samplers 'preloaded' by the site-specific 102 103 distribution of contaminants. A similar experimental procedure has been employed by Hale and Werner<sup>12</sup>. At first, PE strips were preloaded with PAHs by mixing the strips 104 105 with S-1 sediment in a slurry phase for 20 weeks. The preloaded PE strips were then 106 collected, wiped clean, and stored at 4°C until use. Triplicate preloaded PE samples were 107 analyzed to determine PAH concentrations. As expected, the distribution patterns of 108 PAHs in the preloaded PE samplers followed the patterns of compound distribution in S-1 sediment as shown in Figure S5. The preloaded concentrations were  $89.9\pm1.0 \ \mu g \ g^{-1}$ 109 and 1910 $\pm$ 130 µg g<sup>-1</sup> as total parent-PAHs and alkylated-PAHs, respectively 110 (average $\pm$ standard deviation, n=3). Using the preloaded PEs, the AC sorption test was set 111 112 up with two different particle sizes of AC (75-150 and  $<45 \,\mu\text{m}$ ) in clean water. In a 250-113 mL amber bottle, a 2.5 cm  $\times$  10 cm strip of the preloaded PE and approximately 10 mg of AC were added, and the bottle was filled with 20 ppt artificial seawater containing 1 g  $L^{-1}$ 114 115 sodium azide. The bottles were placed on a horizontal shaker at 60 rpm for adequate 116 mixing. After 6 and 12 months, triplicate bottles were sacrificed to measure the residual 117 concentration in the PE. The PAH concentrations in AC is calculated by

$$C_{AC} = \frac{V_{PE} (S_{PE0} - S_{PE}) - V_w (S_{PE} / K_{PE})}{M_{AC}}$$
(3)

where  $C_{AC}$  is the concentration (mol g<sup>-1</sup>) in AC after the contact;  $V_{PE}$  and  $V_w$  are the 118 volumes (cm<sup>3</sup>) of PE and water, respectively;  $S_{PE0}$  and  $S_{PE}$  are the volumetric 119 concentrations (mol cm<sup>-3</sup>) in the preloaded PE before and after the contact, respectively; 120  $K_{PE}$  is the PE-water partitioning coefficient (cm water cm<sup>-1</sup> PE); and  $M_{AC}$  is the mass of 121 122 AC added in the bottle (g). For  $<45 \,\mu\text{m}$  AC, the concentrations in AC did not increase 123 after 6 months indicating that the system has reached (quasi-)equilibrium. Therefore, the 6- and 12-month data for  $<45 \mu m$  AC were used to determine the AC-clean water 124 partitioning coefficient ( $K_{AC \ clean}$ , cm<sup>3</sup> g<sup>-1</sup>) by 125

$$K_{AC\_clean} = \frac{C_{AC}}{S_{PE} / K_{PE}}$$
(4)

The linear sorption isotherm has been shown to be appropriate for the low aqueous concentrations as observed in these tests<sup>16</sup>. The data for 75-150  $\mu$ m AC were fitted with the mass transfer model shown in Eq. (1) substituting  $V_s=0$  (no sediment particles) to determine effective intra-particle diffusivity of AC ( $D_{eff,AC}$ , cm<sup>2</sup> s<sup>-1</sup>). Further discussion on the results of AC performance tests with S-2 sediment

133	For S-2 sediment with $<32$ um AC (5% AC dose), the reductions in the uptake in
134	PE for total PAHs showed a slight decrease from 98% at 1 month to 95% at 12 months
135	over time (Student's t-test, $p < 0.05$ ) as shown in Figure 2. A closer inspection of the data
136	for each compound suggests that the mass transfer processes in this setting was still in
137	progress towards a thermodynamic endpoint with time (Table S7 of the SI). The
138	reductions for the high molecular weight PAHs increased as the molecules diffused into
139	the AC particles over time. For example, the reductions for BaA and BghiP increased
140	from 98.5% and 96.9% at 1 month to 99.8% and >99.2% at 12 months, respectively. A
141	few low molecular weight PAHs such as naphthalene, acenaphthene, and
142	alkylnaphthalenes showed slight decrease in reduction over time, which contributed
143	significantly to the total amounts of PAH uptake in PE. The possible explanation for this
144	increase in uptake in PE over time for low molecular weight PAHs is that the sorption
145	sites initially occupied by these compounds were taken up by high molecular weight
146	PAHs, which diffuse slowly into the AC particles but have higher affinity to AC,
147	resulting in the release of the low molecular weight compounds back to the aqueous
148	phase and PE. The displacement of adsorbed molecules by compounds with higher
149	affinity to AC has been well described in many studies <sup>17-18</sup> .
150	

- **Table S1**. Physicochemical properties of activated carbon (Type TOG, 50×200, Calgon
- 153 Corporation, Pittsburg, PA) used in this study.

<b>Specific surface area</b> <sup>19-20</sup>	<b>Organic carbon content</b> <sup>20</sup>	<b>Specific micropore area</b> <sup>19</sup>
$(m^2 g^{-1})$	(%)	$(m^2 g^{-1})$
720-1030	0.87	430

- 156 detection limits (MDLs) for the determination of the concentrations in 40 mg (51 μm
- 157 thickness, cut in 2.5 cm  $\times$  2.5 cm) polyethylene.

Parent-PAHs		Alkylated-PAHs	
Compound	MDL	Isomer group	MDL
	(ng g <sup>-1</sup> )		(ng g <sup>-1</sup> )
Naphthalene (NAPH)	33	C1-NAPH	63
Acenaphthylene (ACENY)	3.5	C2-NAPH	230
Acenaphthene (ACEN)	15	C3-NAPH	100
Fluorene (FLUO)	23	C4-NAPH	34
Phenanthrene (PHEN)	23	C1-FLUO	110
Anthracene (ANTH)	12	C2-FLUO	91
Fluoranthene (FLUA)	140	C3-FLUO	790
Pyrene (PYR)	3.3	C4-FLUO	42
Benz[a]anthracene (B[a]A)	5.5	C1-PHEN/ANTH	40
Chrysene (CHRY)	6.3	C2-PHEN/ANTH	83
Benzo[b]fluoranthene (B[b]F)	12	C3-PHEN/ANTH	46
Benzo[k]fluoranthene (B[k]F)	14	C4-PHEN/ANTH	620
Benzo[e]pyrene (B[e]P)	17	C1-FLUA/PYR	30
Benzo[a]pyrene (B[a]P)	13	C2-FLUA/PYR	350
Perylene (PER)	2.9	C3-FLUA/PYR	350
Indeno[1,2,3-cd]pyrene (I[1,2,3-cd]P)	10	C4-FLUA/PYR	310
Dibenz[a,h]anthracene (D[ah]A)	15	C1-BaA/CHRY	120
Benzo[ghi]perylene (B[ghi]P)	18	C2-BaA/CHRY	450
		C3-BaA/CHRY	230
		C4-BaA/CHRY	240

<b>Table S3.</b> List of 22 alkylated-PAH compounds used for alkylated-PAH analysi	is in this	S
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159 study.

Group of isomers	Compound name
C1-naphthalenes	1-methylnaphthalene
	2-methylnaphthalene
C2-naphthalenes	1,2-dimethylnaphthalene
	1,3-dimethylnaphthalene
	1,4-dimethylnaphthalene
	1,8-dimethylnaphthalene
	2,6-dimethylnaphthalene
C3-naphthalenes	2,3,5-trimethylnaphthalene
	2,3,6-trimethylnaphthalene
C4-naphthalenes	1,4,6,7-tetramethylnaphthalene
C1-fluorenes	1-methylfluorene
C1-phenanthrenes/anthracenes	1-methylphenanthrene
	2-methylphenanthrene
	1-methylanthracene
	2-methylanthracene
C2-phenanthrenes/anthracenes	3,6-dimethylphenanthrene
	9,10-dimethylanthracene
	2-ethylanthracene
C1-fluoranthenes/pyrenes	1-methylpyrene
C1-benz[a]anthracenes/chrysenes	7-methylbenz[a]anthracene
	6-methylchrysene
C2-benz[a]anthracenes/chrysenes	7,12-dimethylbenz[a]anthracene

161 **Table S4.** List of input parameters for the mass transfer model related to mass transfer

Parameter **Determination method** Annotation (unit)  $C_w \pmod{\operatorname{cm}^{-3}}$ Aqueous equilibrium concentration Aqueous equilibrium test  $K_d \,({\rm cm}^3 {\rm g}^{-1})$ Sediment-water distribution coefficient Aqueous equilibrium test  $k_{fast}$  (s<sup>-1</sup>) Compound release rate from fast-releasing Sediment desorption test sediment particles  $k_{slow}$  (s<sup>-1</sup>) Compound release rate from slow-releasing Sediment desorption test sediment particles  $f_{slow}(-)$ Fraction of slow-releasing particles Sediment desorption test  $K_{AC}$  (cm<sup>3</sup> g<sup>-1</sup>) AC-water partitioning coefficient AC sorption test  $D_{eff,AC}$  (cm<sup>2</sup> s<sup>-1</sup>) AC sorption test Effective intra-particle diffusivity of AC PE-water partitioning coefficient  $K_{PE}(-)$ Unpublished data<sup>a</sup>  $k_o \,({\rm cm \, s^{-1}})$ Booij et al.<sup>7</sup> Overall mass transfer coefficient for PE and water

162 processes in the sediment-water-activated carbon (AC)-polyethylene (PE) system.

<sup>a</sup> Measured at 20°C with 20 ppt salinity, manuscript in review.

165 **Table S5.** Physicochemical properties and PAH concentrations for the sediment samples

166 used in this study. Values in parentheses represent standard deviations for triplicate

167 measurements.

Sediment sample	S-1	S-2 <sup>a</sup>
Total organic carbon content (%)	4.0 (±0.1)	-
Black carbon content (%)	0.73 (±0.07)	-
Total petroleum hydrocarbon (mg kg <sup>-1</sup> )	3740	23000
Oil and grease content (mg kg <sup>-1</sup> )	10800	20400
Total parent-PAHs <sup>b</sup> (mg kg <sup>-1</sup> )	11.6 (±4.3)	45.4 (±2.0)
Total alkylated-PAHs <sup>c</sup> (mg kg <sup>-1</sup> )	114 (±16)	2510 (±280)
Total PAHs (mg kg <sup>-1</sup> )	125 (±17)	2550 (±290)

<sup>a</sup>Total organic carbon and black carbon contents are expected to be similar for the two

169 sediments sampled in close locations and analyzed only for S-1.

<sup>b</sup>Sum of the 18 parent-PAHs.

<sup>c</sup>Sum of 20 groups of alkylated-PAHs ranging from C1- to C4-PAHs.

<sup>b,c</sup>List of the parent- and alkylated-PAHs can be found in Table S2.

Compound	S-2			S-1		
	$k_{fast}$ (s <sup>-1</sup> )	$k_{slow}(s^{-1})$	fslow (-)	$k_{fast}$ (s <sup>-1</sup> )	$k_{slow}(s^{-1})$	$f_{slow}(-)$
ANTH	1.1×10 <sup>-5</sup>	2.5×10 <sup>-8</sup>	0.07	1.9×10 <sup>-6</sup>	4.1×10 <sup>-8</sup>	0.31
PYR	8.8×10 <sup>-6</sup>	1.1×10 <sup>-8</sup>	0.08	4.1×10 <sup>-6</sup>	5.3×10 <sup>-8</sup>	0.18
BaA	6.8×10 <sup>-6</sup>	3.2×10 <sup>-9</sup>	0.20	3.2×10 <sup>-6</sup>	1.5×10 <sup>-7</sup>	0.35
BaP	2.5×10 <sup>-6</sup>	5.3×10 <sup>-9</sup>	0.32	5.3×10 <sup>-7</sup>	2.5×10 <sup>-8</sup>	0.25
BghiP	1.5×10 <sup>-6</sup>	1.9×10 <sup>-9</sup>	0.41	3.2×10 <sup>-7</sup>	3.2×10 <sup>-8</sup>	0.44
2-NAPH	8.8×10 <sup>-6</sup>	5.3×10 <sup>-9</sup>	0.23	1.9×10 <sup>-6</sup>	1.9×10 <sup>-8</sup>	0.39
1,4,6,7-NAPH	1.1×10 <sup>-5</sup>	8.8×10 <sup>-8</sup>	0.02	8.8×10 <sup>-6</sup>	2.5×10 <sup>-7</sup>	0.14
2-ANTH	1.1×10 <sup>-5</sup>	5.3×10 <sup>-8</sup>	0.06	1.9×10 <sup>-6</sup>	5.3×10 <sup>-8</sup>	0.37
7-BaA	5.3×10 <sup>-6</sup>	1.5×10 <sup>-8</sup>	0.15	1.1×10 <sup>-6</sup>	4.1×10 <sup>-8</sup>	0.20

**Table S6**. Desorption parameters for S-2 and S-1 sediments determined by sediment

175 desorption tests.

Table S7. Activated carbon performance test results using S-2 sediment for 18 parent-PAHs and 20 groups of alkylated-PAH isomers 

shown as relative uptake in PE (%) compared to controls (no AC addition). The data are shown as average ± one standard deviation of

triplicate measurements. 

Compound	75-150 μm AC, 5%				<32 µm AC,	5%		
	1 month	2 month	8 month	12 month	1 month	2 month	8 month	12 month
Parent-PAHs								
NAPH	1.9±1.3	0.7±0.3	3.6±2.1	2.5±1.2	4.2±1.5	3.8±1.4	8.2±4.4	9.1±5.5
ACENY	37.2±13.9	22.3±6.9	23.0±7.8	152.9±64.2	29.5±9.9	30.2±7.8	26.3±9.1	25.1±7.6
ACEN	1.6±0.8	0.9±0.3	$2.5 \pm 0.7$	3.1±1.1	2.7±0.5	3.3±1.1	4.5±0.9	4.6±1.3
FLUO	2.6±0.8	1.4±0.3	1.9±0.6	1.8±0.6	1.7±0.5	2.6±0.7	$2.6 \pm 0.7$	$2.5 \pm 0.8$
PHEN	0.7±0.2	0.3±0.1	$0.5 \pm 0.1$	0.2±0.1	0.4±0.1	0.5±0.2	0.5±0.1	$0.5 \pm 0.2$
ANTH	3.2±0.5	1.9±0.5	2.0±0.4	0.4±0.1	1.2±0.4	1.2±0.6	1.2±0.3	$1.1\pm0.4$
FLUA	3.1±0.4	3.0±0.9	<2.2	<1.5	<2.0	<2.3	<2.0	<1.5
PYR	3.5±0.8	2.5±0.9	$0.4{\pm}0.1$	0.2±0.0	0.4±0.1	0.4±0.2	$0.2{\pm}0.0$	$0.2{\pm}0.0$
B[a]A	4.3±1.3	2.9±1.0	$1.4{\pm}0.4$	0.6±0.3	1.5±1.1	1.0±0.4	$0.4 \pm 0.2$	0.2±0.1
CHRY	4.6±0.9	3.0±0.7	$0.9 \pm 0.2$	0.6±0.2	0.5±0.2	0.4±0.1	0.3±0.1	$0.2{\pm}0.1$
B[b]F	7.7±3.8	5.0±1.2	1.6±0.5	0.8±0.5	1.2±0.7	0.8±0.2	< 0.5	< 0.4
B[k]F	10.0±4.8	7.4±1.9	2.5±0.6	1.2±0.4	1.6±0.7	1.2±0.5	<0.9	<0.6
B[e]P	8.3±4.1	5.3±1.2	$1.8 \pm 0.5$	1.2±0.6	1.1±0.4	0.9±0.4	<1.0	<0.7
B[a]P	9.4±2.3	6.5±1.8	1.6±0.5	0.5±0.3	1.1±0.2	1.6±0.3	<0.6	<0.4
PER	38.5±7.4	20.2±7.7	9.8±3.8	35.4±22.4	5.2±1.0	5.9±1.9	<3.1	<1.6
I[1,2,3-cd]P	25.4±3.5	19.2±3.8	7.0±1.7	< 0.7	4.5±0.5	5.7±2.3	<1.1	<0.7
D[ah]A	18.8±2.7	12.0±2.6	<4.5	9.4±4.6	<4.5	<4.4	<3.9	<2.7
B[ghi]P	25.1±6.9	17.6±2.9	5.1±1.8	<0.8	3.1±0.5	3.9±2.1	<1.3	<0.8
Total Parent-PAHs	1.9±0.1	1.3±0.2	1.7±0.2	1.8±0.5	1.1±0.2	1.5±0.4	2.1±0.2	2.2±0.4

## 182 Table S7 (continued)

Compound	75-150 μm AC, 5%				<32 μm AC, 5%			
	1 month	2 month	8 month	12 month	1 month	2 month	8 month	12 month
Alkylated-PAHs								
C1-NAPH	$0.9 \pm 0.5$	$0.6\pm0.2$	3.1±1.3	3.3±3.1	$2.8 \pm 0.7$	3.9±1.5	6.9±2.3	7.0±3.0
C2-NAPH	$1.8 \pm 0.9$	1.0±0.3	$2.8 \pm 0.8$	3.5±1.8	2.4±0.6	3.3±1.2	$4.7 \pm 0.9$	4.9±1.4
C3-NAPH	2.6±1.1	$1.5 \pm 0.5$	$2.8 \pm 0.8$	3.7±1.5	1.7±0.3	$2.7 \pm 0.9$	$3.6 \pm 0.8$	3.8±1.0
C4-NAPH	5.0±1.9	3.4±1.1	4.3±1.2	5.5±1.8	2.3±0.5	3.6±1.2	4.7±1.2	4.9±1.3
C1-FLUO	$1.8 \pm 0.8$	$1.0\pm0.4$	1.3±0.4	1.7±0.5	$1.0\pm0.1$	$1.5 \pm 0.4$	$1.5 \pm 0.4$	1.6±0.3
C2-FLUO	4.4±1.2	$2.7 \pm 0.8$	1.7±0.5	$1.9 \pm 0.5$	$1.0\pm0.2$	$1.4 \pm 0.5$	$1.4{\pm}0.4$	$1.4 \pm 0.3$
C3-FLUO	9.2±8.1	7.5±1.2	3.4±1.0	$2.9{\pm}0.7$	1.7±0.2	1.9±1.0	2.1±0.5	$1.8 \pm 0.4$
C4-FLUO	32.0±5.3	$18.9 \pm 2.3$	3.5±1.0	$2.7 \pm 0.7$	$1.2 \pm 0.2$	$1.6\pm0.6$	$1.5 \pm 0.4$	$1.4{\pm}0.4$
C1-ANTH/PHEN	$1.4 \pm 0.2$	0.9±0.3	$0.7 \pm 0.1$	$0.7 \pm 0.2$	$0.4{\pm}0.1$	$0.7 \pm 0.2$	$0.5 \pm 0.1$	$0.5 \pm 0.1$
C2-ANTH/PHEN	4.3±0.8	2.5±0.6	$1.0\pm0.2$	$0.9{\pm}0.2$	$0.5 \pm 0.0$	0.7±0.3	$0.5 \pm 0.1$	$0.4{\pm}0.1$
C3-ANTH/PHEN	15.7±2.8	8.5±2.5	1.7±0.4	1.2±0.4	$0.6 \pm 0.0$	$0.8 \pm 0.3$	$0.5 \pm 0.1$	$0.4{\pm}0.1$
C4-ANTH/PHEN	40.0±3.3	23.4±6.8	4.7±1.3	2.5±1.0	<1.6	1.6±0.6	<1.3	<0.9
C1-FLUA/PYR	8.3±1.4	4.9±0.9	1.1±0.3	0.9±0.3	0.6±0.1	$0.9 \pm 0.5$	$0.4{\pm}0.1$	$0.4{\pm}0.1$
C2-FLUA/PYR	15.4±2.4	9.2±1.9	2.1±0.5	1.4±0.6	<1.6	<1.4	<1.3	<0.8
C3-FLUA/PYR	33.9±6.0	20.7±4.9	5.5±1.4	3.3±1.9	<5.4	<4.5	<3.7	<2.4
C4-FLUA/PYR	62.8±15.6	39.2±9.9	<10.6	<5.9	<13.2	<11.5	<9.3	<5.8
C1-B[a]A/CHRY	57.2±72.5	8.8±2.2	$2.9 \pm 0.7$	1.7±0.7	<2.1	2.0±0.7	<2.4	<1.6
C2-B[a]A/CHRY	30.5±10.1	16.8±3.9	<7.8	<4.4	<6.8	<6.2	<6.9	<4.3
C3-B[a]A/CHRY	50.7±19.3	41.2±9.4	<12.7	<7.1	<8.8	<10.3	<11.1	<7.0
C4-B[a]A/CHRY	67.9±0.0	49.3±5.9	<26.4	<14.0	<19.8	<20.1	<23.1	<13.7
Total alkylated-PAHs	3.3±0.4	2.5±0.5	3.6±0.4	4.2±1.6	1.8±0.3	3.1±1.0	4.3±0.3	4.7±0.7
Total PAHs	3.3±0.1	2.4±0.7	3.5±0.3	4.2±1.9	1.8±0.1	3.0±1.4	4.2±0.2	4.7±0.8

- Figure S1. Concentrations of (a) 18 parent-PAHs and (b) 20 groups of alkylated-PAH
- isomers in the two sediment samples (S-1 and S-2) used in this study. The error bars
- represent one standard deviation of the triplicate measurements.





Figure S2. Relative uptake of (a) parent-PAHs and (b) alkylated-PAHs in polyethylene (PE) samplers compared to controls (no AC addition) after contacting S-2 sediment and AC with different size ranges for 1, 2, 8, and 12 months in sediment slurry tests. The error bars represent one standard deviation of the triplicate measurements.





195Figure S3. Correlation between the AC-clean water partitioning coefficients ( $K_{AC\_clean}$ )196measured in this study and the octanol-water partitioning coefficients ( $K_{ow}$ ) for the nine197PAH model compounds. The error bars represent one standard deviation of the triplicate198measurements.



Figure S4. Simulation results versus experimental data for the AC performance tests using S-1 sediment and (a) 75-300  $\mu$ m AC at 5% and 10% dose and (b) 75-150  $\mu$ m and 32-75  $\mu$ m AC at 5% dose for nine model PAH compounds selected in this study. Data are shown as relative uptake in polyethylene (PE) samplers compared to controls (no AC addition). The error bars represent one standard deviation of the triplicate measurements.

207 (a) 75-300 µm AC, 5% and 10%



- 75-300 μm, 5%, experimental data
- Π 75-300 μm, 10%, experimental data



• 32-75 µm, 5%, experimental data

- Figure S5. Concentrations of (a) 18 parent-PAHs and (b) 20 groups of alkylated-PAH
- isomers in polyethylene (PE) samplers preloaded with S-1 sediment for 20 weeks. The



error bars represent one standard deviation of the triplicate measurements. 

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