

## **Bioaccumulation of Polycyclic Aromatic Hydrocarbons (PAHs) by the Marine Clam, *Macra veneriformis*, Chronically Exposed to Oil-Suspended Particulate Matter Aggregates**

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## Supporting Materials and Methods

**Stable Isotope Analysis.** Only soft tissues were analyzed for stable isotope ratios of carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ). Soft tissues were rinsed with distilled water, homogenized, and stored frozen at  $-20\text{ }^{\circ}\text{C}$  until analysis. Subsamples of OSAs were sieved through a glass-fiber filter and then stored at  $-20\text{ }^{\circ}\text{C}$  until isotopes were analyzed. To determine stable isotopic ratios of clams after feeding OSAs, a method slightly modified from that described previously was used<sup>1</sup>, wherein soft tissues sampled at Days 1, 2, 7, 14, 30, and 50 were lyophilized and then lipids were removed. The analysis consisted of first adding approximately 10 mL of dichloromethane (DCM)/methanol (2:1, v/v) to 10-20 mg of freeze-dried soft tissue. The mixture was then sonicated for 10 min and centrifuged at 4000 g for 15 min. Then organic solvents were pipetted off and discarded. This extraction procedure was repeated three times. Then, lipid-free tissues were evaporated under a gentle stream of  $\text{N}_2$  at room temperature until dry. Before quantifications of isotopes in OSAs, samples were freeze-dried and de-carbonated overnight by fuming with  $\text{HCl}$  in a desiccator<sup>2,3</sup>.

$\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  were measured with an Elemental Analyzer (EA)-Isotope Ratio Mass Spectrometer (IRMS) (Elementar, Hanau, Hesse). High purity  $\text{CO}_2$  and  $\text{N}_2$  were used as reference gases, while  $\text{He}$  and  $\text{O}_2$  were used as the carrier and combustion gases, respectively. Stable carbon and nitrogen isotopic ratios were expressed as ‰ delta notation (Equations 1 and 2)

$$\delta^{13}\text{C} (\text{‰}) = [\text{R}_{\text{sam}}/\text{R}_{\text{ref}} - 1] \times 1000 \quad (1)$$

$$\delta^{15}\text{N} (\text{‰}) = [\text{R}_{\text{sam}}/\text{R}_{\text{ref}} - 1] \times 1000 \quad (2)$$

where:  $\text{R}_{\text{sam}}$  and  $\text{R}_{\text{ref}}$  are the ratios ( $^{13}\text{C}/^{12}\text{C}$  or  $^{15}\text{N}/^{14}\text{N}$ ) of the sample and reference, respectively. Isotopic ratios were reported relative to the Vienna Pee Dee Belemnite (VPDB) standard. International isotope standards, IAEA-CH-3 and IAEA-N-2 to calibrate  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ , were used.

## Supporting Results

**Comparison of Stable Isotopic Ratios in Clams.** Based on carbon and nitrogen stable isotope ratios, determined for clams in our experiment and the typical fractionation pattern for a single trophic step (from food source to consumer), isotopic values for *Macraa veneriformis* averaged  $-17.2\text{‰}$  for  $\delta^{13}\text{C}$  and  $12.2\text{‰}$  for  $\delta^{15}\text{N}$  (SI-Figure S6). Stable isotopic ratios were lighter than values previously determined for soft clams, averaging  $-26.8$ ,  $-20.8$ , and  $-26.9\text{‰}$  for  $\delta^{13}\text{C}$  and  $9.7$ ,  $8.4$ , and  $7.3\text{‰}$ , for OSAs, sediment, and crude oil, respectively. By Day 50 the mean difference in stable isotope ratios varied little relative to ratios obtained on Day 1:  $0.9 \pm 0.1\text{‰}$  for carbon and  $0.3 \pm 0.2\text{‰}$  for nitrogen (SI-Figure S6).

**Table S1.** Mini-review of bioaccumulation of PAHs in marine organisms (fish and bivalves), data from in situ measurements.

Species	Region	Dominating Mol. Mass	ΣPAHs (ng/g)	Reference
<b>Fish</b>				
<i>Clupea pallasii</i>	Puget Sound, Washington, USA	LMM <sup>a</sup>	1.1 – 140	West et al., 2014 <sup>4</sup>
<i>Mullus barbatus</i>	Mediterranean Sea	LMM	14.7 – 49.6	Baumard et al., 1998 <sup>5</sup>
<i>Serranus scriba</i>	Mediterranean Sea	LMM	23.6 – 139	
<i>Micropogonias furnieri</i>	Bahía Blanca Estuary, Argentina	LMM	34.87	Oliva et al., 2017 <sup>6</sup>
<i>Cynoscion guatucupa</i>	Bahía Blanca Estuary, Argentina	LMM	98.25	
<i>Anguilla anguilla</i>	Biertze Lagoon, Tunisia	LMM	114.5 – 133.7	Barhoumi et al., 2016 <sup>7</sup>
<i>Scophthalmus maximus</i>	Laboratory	LMM	121	Baussant et al., 2001 <sup>8</sup>
<i>Cynoglossus senegalensis</i>	Gulf of Guinea, Ghana	LMM	67 – 163	Bandowe et al., 2014 <sup>9</sup>
<i>Drepane africana</i>	Gulf of Guinea, Ghana	LMM	136 – 453	
<i>Pomadasys perotai</i>	Gulf of Guinea, Ghana	LMM	163 – 252	
<i>Sarotherodon mossambicus</i>	Pearl River Estuary, Hong Kong	LMM	184 – 194	Liang et al., 2007 <sup>10</sup>
<i>Ramnogaster arcuata</i>	Bahía Blanca Estuary, Argentina	LMM	182.35	Oliva et al., 2017 <sup>6</sup>
<i>Mustelus schmitti</i>	Bahía Blanca Estuary, Argentina	LMM	308.96	
<i>Anguilla anguilla</i>	Amsterdam, Netherlands	LMM	11,000 – 39,000	van der Oost et al., 1994 <sup>11</sup>
<b>Bivalves</b>				
<i>Chlamys farreri</i>	Yellow Sea, China	NA <sup>c</sup>	ND <sup>d</sup>	Liu et al., 2008 <sup>12</sup>
<i>Cyclina sinensis</i>	Yellow Sea, China	NA	ND	
<i>Neptunea arthritica</i>	Yellow Sea, China	NA	ND	
<i>Neverita didyma</i>	Yellow Sea, China	NA	ND	
<i>Anadara kagoshimensis</i>	Yellow Sea, China	NA	ND	
<i>Siliqua pulchella</i>	Yellow Sea, China	NA	ND	
<i>Bullacta exarata</i>	Yellow Sea, China	HMM <sup>b</sup>	23.5	
<i>Euspira gilva</i>	Yellow Sea, China	HMM	23.5	
<i>Solen grandis</i>	Yellow Sea, China	HMM	23.5	
<i>Mytilus edulis</i>	Yellow Sea, China	HMM	ND – 29.3	
<i>Saccostrea cucullata</i>	Yellow Sea, China	HMM	29.7	
<i>Magallana gigas</i>	Yellow Sea, China	HMM	24.7 – 30.3	
	Pacific Coast, Japan	LMM	370	Onozato et al., 2016 <sup>13</sup>
<i>Leukoma jedomensis</i>	Yellow Sea, China	HMM	26.5 – 39.7	Liu et al., 2008 <sup>12</sup>
<i>Anadara broughtonii</i>	Yellow Sea, China	HMM	41.9	
<i>Mytilus galloprovincialis</i>	Gironde Estuary France	LMM, HMM	49.8 – 101.5	Bodin et al., 2004 <sup>14</sup>
	Pacific Coast, Japan	LMM	301	Onozato et al., 2016 <sup>13</sup>
	Arachon Bay, France	HMM	279 – 2420	Baumard et al., 1998 <sup>5</sup>
<i>Meretrix meretrix</i>	Yellow Sea, China	LMM	28.3 – 108.0	Liu et al., 2008 <sup>12</sup>
<i>Ruditapes philippinarum</i>	Pacific Coast, Japan	LMM	244	Onozato et al., 2016 <sup>13</sup>
	Yellow Sea, China	HMM	24.8 – 302.0	Liu et al., 2008 <sup>12</sup>
<i>Brachidontes rodriguezi</i>	Bahía Blanca Estuary, Argentina	LMM	ND – 482.4	Oliva et al., 2017 <sup>6</sup>
<i>Mytilus galloprovincialis</i>	Mediterranean Sea	LMM	25.1 – 337	Baumard et al., 2001 <sup>5</sup>
	Biertze Lagoon, Tunisia	LMM	107.4 – 430.7	Barhoumi et al., 2016 <sup>7</sup>
	Urdaibai Estuary, Spain	HMM	21 – 64	Orbea et al., 2002 <sup>15</sup>
<i>Magallana sp.</i>	Urdaibai Estuary, Spain	HMM	31 – 218	
<i>Mya truncata</i>	Svalbard Fjord	LMM	200,080*	Camus et al., 2003 <sup>16</sup>

\*Analyzed in lipid extracts from *Mya truncata*.

a = “Lower Molecular Mass”, b = “Higher Molecular Mass”, c = “Not Available”, and d = “Not Detected”.

**Table S2.** Baseline information of sediment properties. Sandy loam was used for bottom sediment in experimental tank.

Experimental treatment	Organic contents* (%)	Mud contents (%)	Grain size	Sediment source
Control	0.92 ± 0.08	0.79 ± 0.25	<1 mm	Samhangang INC., Incheon, KOR
OSA <sub>low</sub>	0.98 ± 0.20			
OSA <sub>high</sub>	0.81 ± 0.08			

\*Mean during the whole treatment period (Day 1, 2, 4, 7, 14, 30, 40, and 50)

**Table S3.** Experimental conditions of preparation of the OSAs.

<i>Preparation of OSAs</i>	
<div style="display: flex; justify-content: space-around; align-items: flex-start;"> <div style="text-align: center;"> <p><b>Schematic drawing</b></p> </div> <div style="text-align: center;"> <p><b>OSAs + Seawater mixture in carboy</b></p> </div> </div>	
<b>Reaction chamber</b>	SCHOTT-DURAN 20 Lit. Glass Aspiration Carboy (①)
Volume	2000 mL
Headspace	>10% by volume (②)
<b>Water column</b>	GF/F-filtered seawater (③)
Volume	1000 mL (1000 g)
Salinity	34 psu
<b>Oil</b>	Iranian heavy crude oil (④)
Concentration	0.6 g/L
<b>Sediment</b>	Dried, from a mudflat (④)
Concentration	0.2 g/L
Size	$\Phi < 20 \mu\text{m}$
Total organic matter contents	~6%
<b>Reciprocating shaker</b>	EYELA Shaker NTS-4000A
Shaking speed	170 rpm (⑤)
Shaking duration	24 h (⑥)
Shaking condition	Sealed vessel in dark room at test temperature (15 °C) (⑦)
<b>Subsampling of OSAs</b>	(only used for neutral and negative buoyancy)
Settling	5 min
Extraction (in carboy)	700 mL (OSAs + seawater mixture) (⑧)

This method was slightly modified from Sun et al., 2010<sup>17</sup>.

**Table S4.** List of target PAHs compounds measured by gas chromatograph equipped with a mass selective detector (GC/MSD) in OSAs experiment; Full chemical name, acronym, molecular mass grouping, carbon ring number, and Log K<sub>ow</sub> values with references given.

Full chemical name	Acronym	MMG*	Ring number	Log K <sub>ow</sub> **
Naphthalene	Nap	LMM	2	3.33 <sup>b</sup> , 3.35 <sup>a</sup>
Acenaphthylene	AcI	LMM	3	3.67 <sup>a</sup> , 4.2 <sup>b</sup>
Acenaphthene	Ace	LMM	3	3.92 <sup>a</sup> , 4.0 <sup>b</sup>
Fluorene	Flu	LMM	3	4.18 <sup>a</sup> , 4.32 <sup>b</sup>
Dibenzothiophene	Dbt	LMM	3	4.38 <sup>a</sup> , 4.52 <sup>c</sup>
Phenanthrene	Phe	LMM	3	4.52 <sup>b</sup> , 4.57 <sup>a</sup> , 4.90 <sup>c</sup>
Anthracene	Ant	LMM	3	4.50 <sup>b</sup> , 4.68 <sup>a</sup>
1-Methylnaphthalene	C1-Nap	LMM	2	3.89 <sup>d</sup>
2-Methylnaphthalene	C2-Nap	LMM	2	3.88 <sup>a</sup> , 3.96 <sup>d</sup>
1,4,5-Trimethylnaphthalene	C3-Nap	LMM	2	4.48 <sup>d</sup>
1,2,5,6-Tetramethylnaphthalene	C4-Nap	LMM	2	4.95 <sup>d</sup> , 5.53 <sup>a</sup>
9-Methylfluorene	C1-Flu	LMM	3	4.66 <sup>c</sup>
1,7-Dimethylfluorene	C2-Flu	LMM	3	5.08 <sup>c</sup>
9-n-Propylfluorene	C3-Flu	LMM	3	5.85 <sup>a</sup>
2-Methyldibenzothiophene	C1-Dbt	LMM	3	4.93 <sup>a</sup> , 5.04 <sup>c</sup>
2,4-Dimethyldibenzothiophene	C2-Dbt	LMM	3	5.47 <sup>c</sup>
2,4,7-Trimethyldibenzothiophene	C3-Dbt	LMM	3	6.03 <sup>a, c</sup>
3-Methylphenanthrene	C1-Phe	LMM	3	5.34 <sup>c</sup>
1,6-Dimethylphenanthrene	C2-Phe	LMM	3	5.64 <sup>c</sup>
1,2,9-Trimethylphenanthrene	C3-Phe	LMM	3	6.03 <sup>c</sup>
1,2,6,9-Tetramethylphenanthrene	C4-Phe	LMM	3	6.48 <sup>c</sup>
Fluoranthene	Fl	HMM	4	5.20 <sup>b</sup> , 5.23 <sup>a</sup>
Pyrene	Py	HMM	4	5.00 <sup>b</sup> , 5.13 <sup>a</sup>
Benzo[ <i>a</i> ]anthracene	BaA	HMM	4	5.91 <sup>a, b</sup>
Chrysene	Chr	HMM	4	5.81 <sup>a</sup> , 5.86 <sup>b</sup>
Benzo[ <i>b</i> ]fluoranthene	BbF	HMM	5	5.78 <sup>a</sup> , 5.8 <sup>b</sup>
Benzo[ <i>k</i> ]fluoranthene	BkF	HMM	5	6.11 <sup>b</sup>
Benzo[ <i>a</i> ]pyrene	BaP	HMM	5	6.13 <sup>a</sup> , 6.35 <sup>b</sup>
Perylene	Pery	HMM	5	6.25 <sup>a</sup>
Indeno[1,2,3- <i>cd</i> ]pyrene	IcdP	HMM	6	6.58 <sup>a</sup> , 6.72 <sup>b</sup>
Dibenzo[ <i>a,h</i> ]anthracene	DbahA	HMM	5	6.75 <sup>a, b</sup>
Benzo[ <i>g,h,i</i> ]perylene	BghiP	HMM	6	6.5 <sup>a</sup> , 6.9 <sup>b</sup>
3-Methylchrysene	C1-Chr	HMM	4	6.42 <sup>c</sup>
6-Ethylchrysene	C2-Chr	HMM	4	6.88 <sup>c</sup>
1,3,6-Trimethylchrysene	C3-Chr	HMM	4	7.44 <sup>c</sup>

\*Molecular mass groups (MMGs) were categorized based on ring number class as either lower molecular mass (LMM, 2-3 rings) or higher molecular mass (HMM, 3-4 rings).

\*\*Data for Log K<sub>ow</sub> values obtained from studies by: (a) ref 18, (b) ref 19, (c) ref 20, (d) ref 21, and (e) ref 22.

**Table S5.** GC/MSD conditions for the analyses of PAHs and alkylated PAHs.

GC/MSD system	Agilent 7890A GC and 5975C MSD
Column	DB-5MS (30 m long, 0.25 mm i.d., 0.25 µm film thickness)
Gas flow	1 mL/min He
Injection mode	Splitless
Injection volume	2 µL
MS temperature	180 °C
Detector temperature	230 °C
Oven temperature	60 °C hold for 2 min Increase 6 °C/min to 300 °C 300 °C hold for 13 min

**Table S6.** Mean concentrations of PAHs (ng/g, wm) in soft tissue of clams, *Mactra veneriformis*, under OSAs feeding experiments. Acronyms for PAHs given in SI-Table S4.

Treatment		OSA <sub>low</sub>								OSA <sub>high</sub>							
Sampling day		1	2	4	7	14	30	40	50	1	2	4	7	14	30	40	50
LMM PAHs	<b>Nap*</b>	341.9	97.1	365.9	79.0	120.1	114.5	389.5	375.9	336.1	87.8	255.6	69.5	94.8	120.3	506.1	376.0
	<b>C1-Nap</b>	57.8	67.3	58.9	69.6	74.4	78.0	32.2	47.8	78.1	62.0	50.3	70.5	81.4	78.7	59.3	49.9
	<b>C2-Nap</b>	177.4	127.0	185.9	206.4	162.1	124.6	60.7	94.2	369.5	153.7	320.8	311.8	151.8	131.9	176.0	146.8
	<b>C3-Nap</b>	129.1	77.0	147.4	196.1	177.9	96.1	78.1	90.0	252.6	125.7	228.6	213.0	138.0	125.6	237.4	215.5
	<b>C4-Nap</b>	106.2	108.7	138.3	290.8	330.2	264.1	106.9	128.9	68.1	203.2	174.6	471.5	214.8	370.7	273.5	307.6
	Flu	3.0	5.7	3.4	6.6	7.1	6.1	2.8	2.9	4.6	5.5	4.7	6.9	5.2	7.1	4.8	3.6
	<b>C1-Flu</b>	30.1	21.5	35.1	48.7	44.0	25.1	16.4	31.5	50.5	30.1	56.0	60.9	31.5	32.4	29.6	54.0
	<b>C2-Flu</b>	16.1	27.5	23.5	79.2	93.8	72.8	23.9	26.7	25.8	43.3	36.1	96.7	77.7	118.3	48.6	56.4
	<b>C3-Flu</b>	24.4	45.2	39.8	176.0	262.7	238.4	42.6	74.4	36.1	79.3	50.2	189.8	209.3	433.8	152.7	164.4
	Dbt	0.4	3.6	1.3	16.1	5.8	3.4	0.7	0.5	0.6	8.1	2.2	20.6	6.8	6.8	1.1	1.0
	<b>C1-Dbt</b>	2.8	16.2	8.1	98.0	52.0	63.9	5.5	5.2	4.2	32.8	13.3	85.9	64.4	122.6	5.8	12.3
	<b>C2-Dbt</b>	0.9	21.1	3.1	217.8	207.1	360.0	4.1	2.9	1.4	54.9	4.1	235.8	264.4	738.1	3.5	8.0
	<b>C3-Dbt</b>	3.7	11.3	10.2	126.1	190.2	317.7	19.4	18.8	5.3	27.8	12.8	128.4	223.9	629.2	21.1	50.0
	Phe	7.0	13.3	9.5	21.8	19.6	18.0	6.2	7.5	11.2	15.9	13.9	20.9	14.6	21.6	11.4	12.4
	<b>C1-Phe</b>	23.8	15.2	37.1	37.9	46.0	21.0	34.7	40.7	40.5	25.0	56.4	42.4	35.6	53.7	77.1	90.8
	<b>C2-Phe</b>	68.0	29.6	118.0	114.4	205.1	158.5	169.1	230.3	109.0	61.2	147.5	143.4	159.9	287.0	315.3	495.9
	<b>C3-Phe</b>	13.4	33.6	22.8	117.0	197.7	198.2	91.7	71.6	17.2	43.1	28.7	134.2	179.8	367.9	154.1	125.3
	<b>C4-Phe</b>	5.9	8.6	8.1	17.0	22.5	25.1	28.0	32.0	8.0	7.5	10.3	18.9	21.7	45.2	32.5	58.8
HMM PAHs	Fl	2.9	4.1	3.3	3.8	4.7	4.6	1.8	3.5	3.7	3.3	3.1	3.1	4.3	5.3	2.9	3.0
	Py	3.5	3.3	4.0	4.1	5.7	5.5	4.5	7.2	4.6	3.3	4.1	4.3	6.6	8.7	6.8	9.4
	<b>BaA</b>	2.8	1.2	1.4	1.6	9.3	1.9	6.7	7.5	1.7	1.3	1.7	2.0	7.9	5.3	15.1	8.1
	<b>Chr</b>	3.6	3.1	2.2	4.8	1.1	12.4	6.7	7.9	2.4	3.6	2.0	6.6	8.8	23.0	1.1	14.4
	<b>C1-Chr</b>	0.4	8.4	0.4	12.4	20.2	26.5	2.4	0.7	0.5	7.2	0.5	16.2	22.5	57.1	5.3	0.3
	<b>C2-Chr</b>	0.3	13.2	0.3	18.8	17.7	20.0	3.1	0.3	0.2	13.6	1.1	17.3	23.0	42.4	6.2	0.4
	<b>C3-Chr</b>	0.2	3.3	0.3	6.2	6.9	7.8	1.7	0.4	0.3	4.7	0.7	6.6	8.9	14.6	2.0	0.4

\*20 PAHs in bold indicate that accumulated PAHs concentrations higher than 30 ng/g wm for LMM PAHs and 10 ng/g wm for HMM PAHs over the period of 50 days.



**Table S7.** Result of Mann-Whitney U-test for comparison of replicates (n = 2) per treatment in total PAHs. The replications of Control, OSA<sub>low</sub>, and OSA<sub>high</sub> were tested.

	Experimental treatment		
	Control	OSA <sub>low</sub>	OSA <sub>high</sub>
Mann-Whitney U	18.0	29.0	28.0
p value	.491	.798	1.00

**Table S8.** Spearman rank correlation results for concentrations of 20 PAHs in soft tissue of clams, *Macra veneriformis*. Values in bold indicate correlation significance at the  $p < 0.01$  level (matched with ++). Acronyms for PAHs given in SI-Table S4.

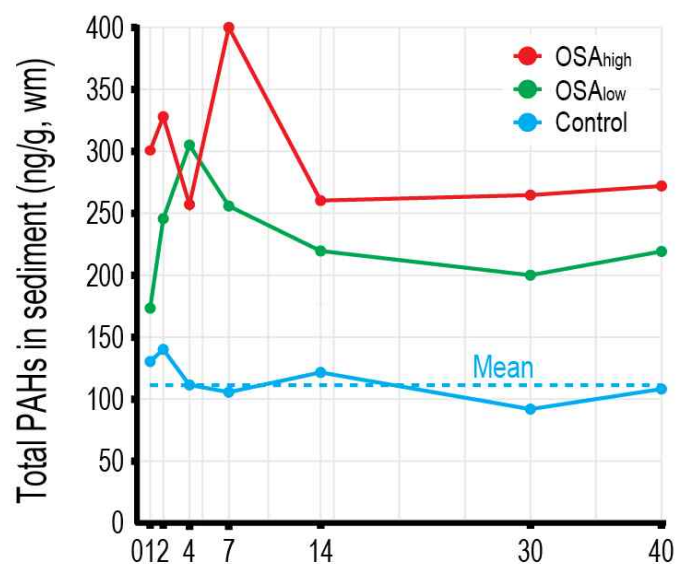
Compounds		Lower molecular mass (LMM) PAHs														Higher molecular mass (HMM) PAHs					
		Nap	C1-Nap	C2-Nap	C3-Nap	C4-Nap	C1-Flu	C2-Flu	C3-Flu	C1-Dbt	C2-Dbt	C3-Dbt	C1-Phe	C2-Phe	C3-Phe	C4-Phe	BaA	Chr	C1-Chr	C2-Chr	C3-Chr
LMM PAHs	Nap		<b>-.624</b>	-.229	.085	-.362	-.221	-.556	-.435	<b>-.753</b>	<b>-.659</b>	-.432	.335	.521	-.206	.379	.476	-.112	<b>-.624</b>	<b>-.624</b>	-.618
	C1-Nap	++		.194	.115	.365	.144	.609	.562	<b>.624</b>	<b>.624</b>	.544	-.106	-.129	.476	-.076	-.082	.115	<b>.747</b>	<b>.641</b>	<b>.688</b>
	C2-Nap				<b>.815</b>	.041	<b>.703</b>	.003	-.224	-.018	-.259	-.306	.297	-.371	-.394	-.544	-.268	-.571	-.268	-.244	-.256
	C3-Nap			++		.247	<b>.724</b>	.159	-.012	-.071	-.224	-.097	<b>.668</b>	.138	-.091	-.056	.186	-.421	-.309	-.229	-.200
	C4-Nap						.403	<b>.918</b>	<b>.879</b>	<b>.776</b>	<b>.726</b>	<b>.824</b>	.535	.491	<b>.794</b>	.535	.356	.250	.532	<b>.691</b>	<b>.688</b>
	C1-Flu			++	++			.341	.159	.224	.024	.068	<b>.632</b>	.085	-.053	-.047	.018	-.088	-.191	-.097	-.068
	C2-Flu	+	+			++			<b>.947</b>	<b>.882</b>	<b>.859</b>	<b>.903</b>	.388	.374	<b>.844</b>	.471	.279	.318	<b>.759</b>	<b>.835</b>	<b>.868</b>
	C3-Flu		+			++		++		<b>.832</b>	<b>.874</b>	<b>.971</b>	.312	.503	<b>.941</b>	.585	.391	.400	<b>.791</b>	<b>.865</b>	<b>.891</b>
	C1-Dbt	++	++			++		++	++		<b>.944</b>	<b>.832</b>	.062	.062	<b>.691</b>	.215	-.100	.344	<b>.788</b>	<b>.921</b>	<b>.888</b>
	C2-Dbt	++	++			++		++	++	++		<b>.918</b>	-.038	.168	<b>.809</b>	.347	.026	.497	<b>.859</b>	<b>.959</b>	<b>.953</b>
	C3-Dbt		+			++		++	++	++	++		.203	.482	<b>.947</b>	.585	.362	.526	<b>.806</b>	<b>.894</b>	<b>.912</b>
	C1-Phe				++	+	++								<b>.703</b>	.288	.562	-.085	-.168	-.059	-.006
	C2-Phe	+							+				++		<b>.647</b>	<b>.932</b>	<b>.859</b>	.321	.088	.168	.212
	C3-Phe					++		++	++	++	++	++		++		<b>.741</b>	.544	.438	<b>.774</b>	.821	<b>.853</b>
	C4-Phe			+		+			+			+	+	++	++		<b>.726</b>	.488	.253	.321	.362
HMM PAHs	BaA												+	++	+	++		.144	.079	.082	.147
	Chr			+							+								.300	.356	.356
	C1-Chr	++	++			+		++	++	++	++	++			++					<b>.912</b>	<b>.947</b>
	C2-Chr	++	++			++		++	++	++	++	++			++				++		<b>.976</b>
	C3-Chr	+	++			++		++	++	++	++	++			++				++	++	

++ Significantly correlated at the  $p < 0.01$  level (2-tailed).

+ Significantly correlated at the  $p < 0.05$  level (2-tailed).

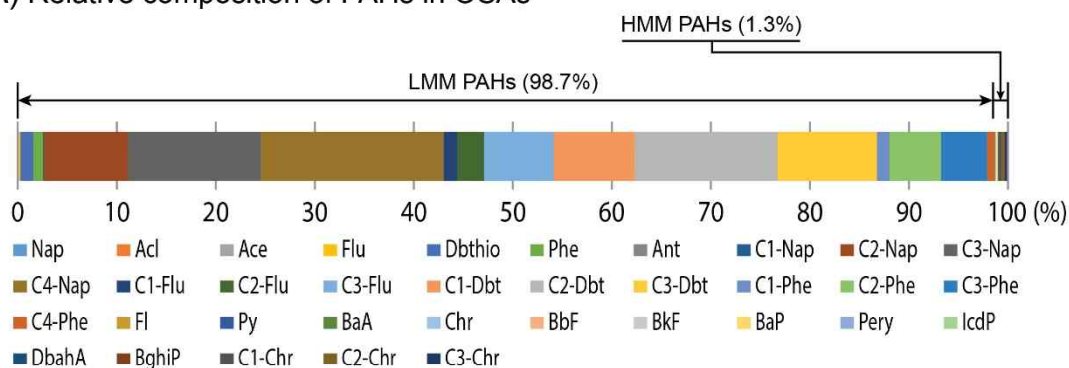
**Table S9.** Result of Kolmogorov-Smirnov normality test for concentrations of PAHs in soft tissue of clams, *Macra veneriformis*, over the period of 50 days in OSAs feeding treatments (OSA<sub>low</sub> and OSA<sub>high</sub>). The concentrations of 20 PAHs (refer to SI-Table S6) were standardized by total, and root transformed before these test.

Kolmogorov-Smirnov normality test		
Statistic	degree of freedom	p value
.048	320	.072

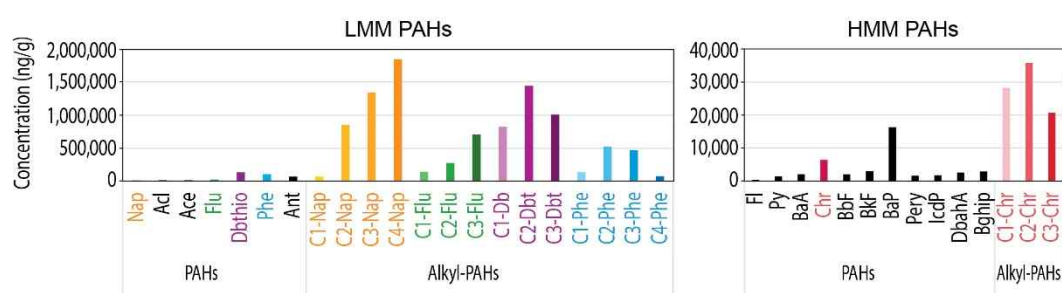


**Figure S1.** Total PAHs concentration in sediments. Blue line denotes Control treatment, green line denotes the OSA<sub>low</sub> treatment, and red line denotes the OSA<sub>high</sub> treatment, the PAHs concentrations are found to be fairly consistent after the 14 days and maintained at certain levels for the last period of experiments cross the treatments.

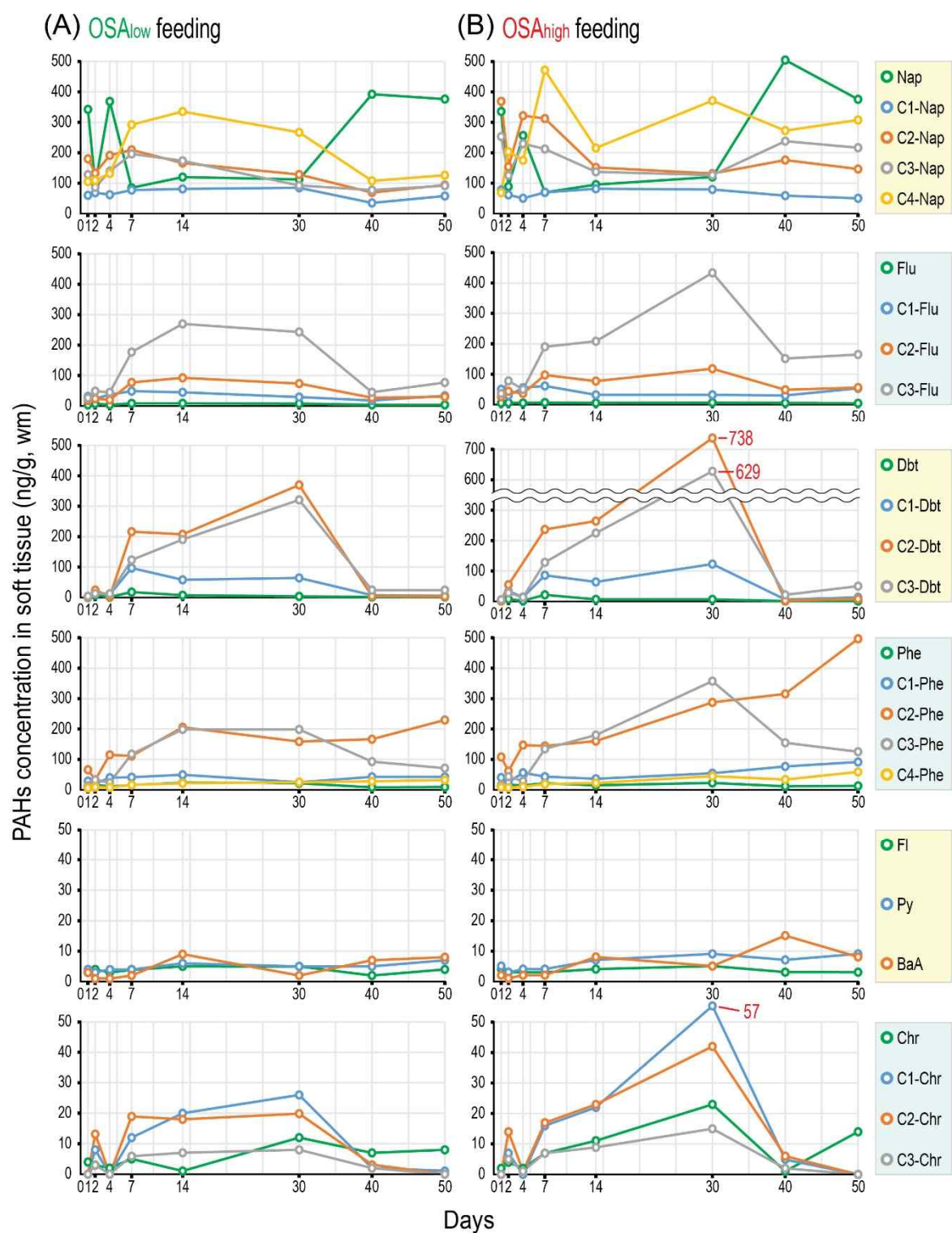
(A) Relative composition of PAHs in OSAs



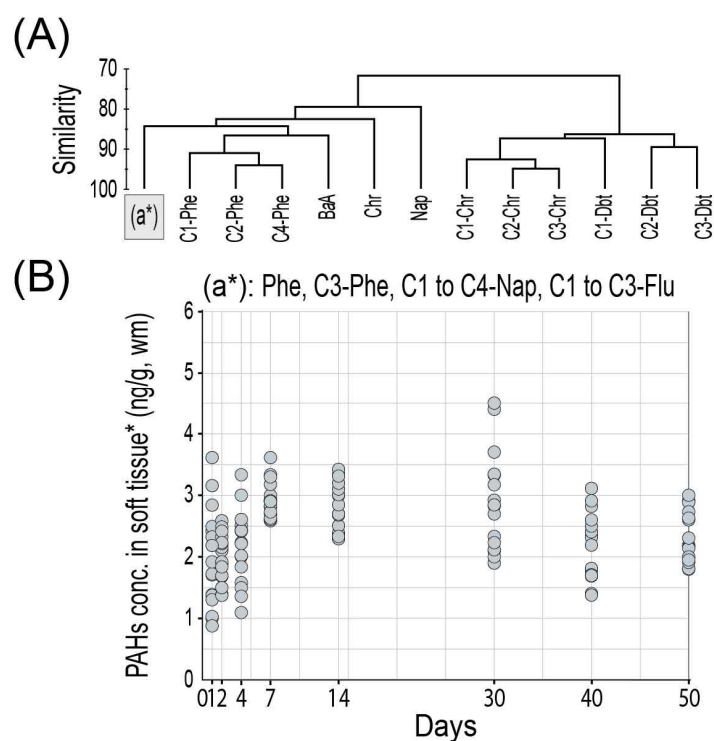
(B) PAHs concentrations in OSAs



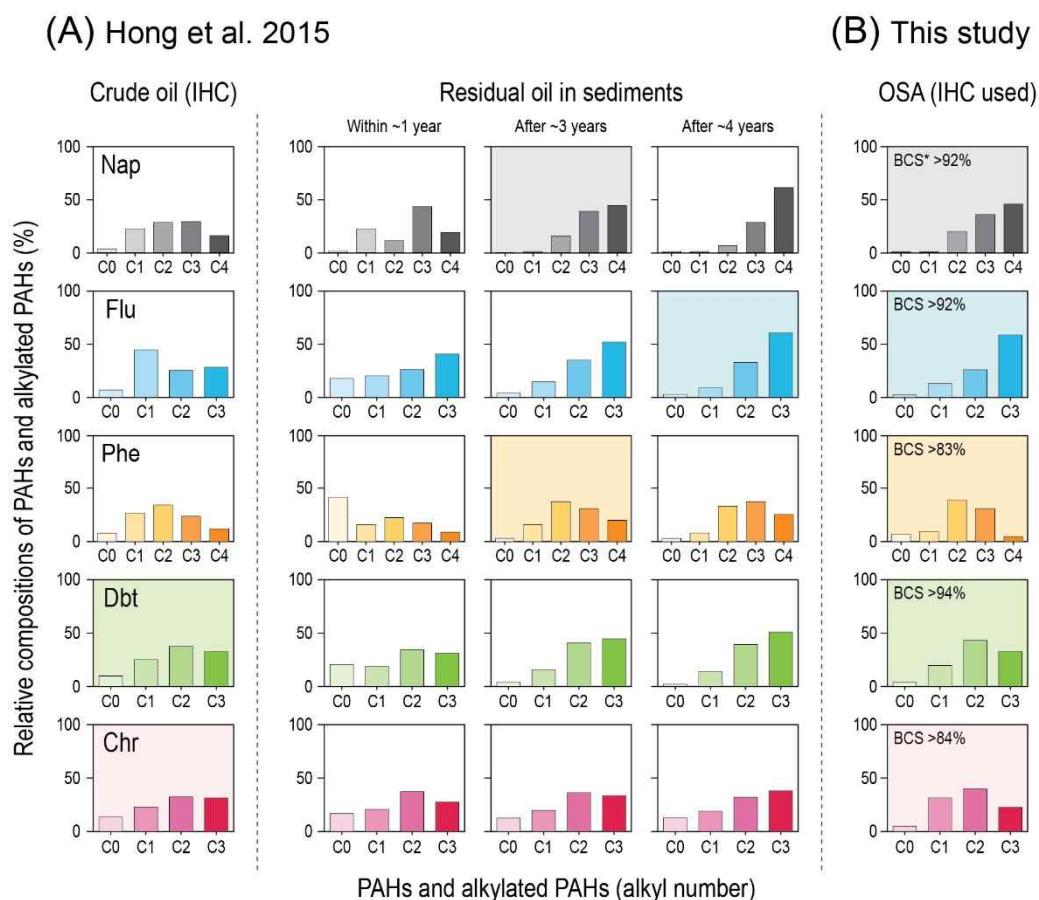
**Figure S2.** PAHs and alkylated PAHs in OSAs. (A) Relative composition of PAHs compounds in OSAs and (B) Concentrations of PAHs and alkylated PAHs in OSAs. Acronyms for the target PAHs given in SI-Table S4.



**Figure S3.** Concentrations of 25 PAHs in soft tissue of clams, *Mactra veneriformis*, under (A)  $OSA_{low}$  feeding treatment and (B)  $OSA_{high}$  feeding treatment. Acronyms for PAHs given in SI-Table S4.

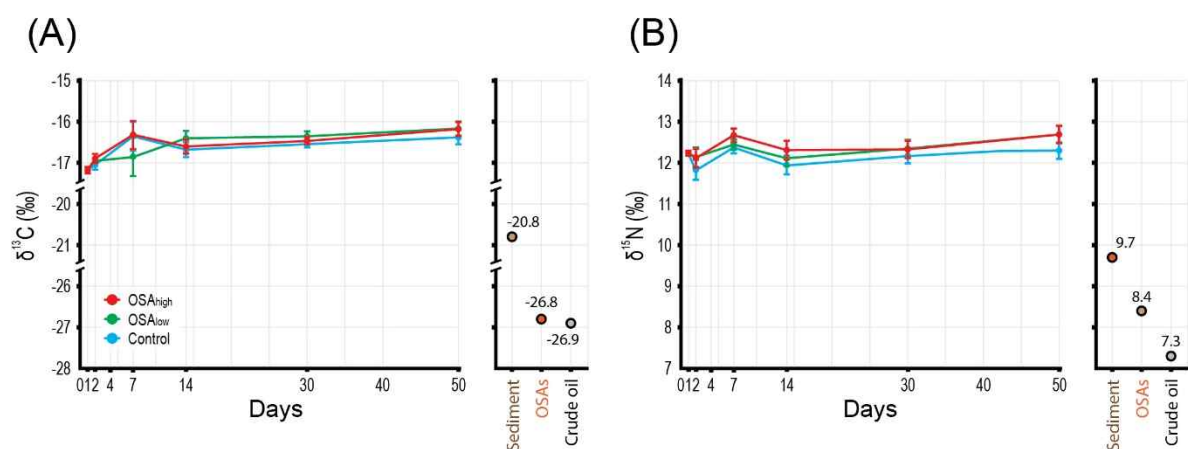


**Figure S4.** PAHs compounds in soft tissue of clams, *Macra veneriformis*, by the bioaccumulation of OSAs feeding experiments; (A) Dendrogram representing hierarchical clustering based on Bray-Curtis similarities (BCS) by group means, (B) Group (a\*) indicated group of PAHs in BCS <85% including Phe, C3-Phe, C1 to C4-Nap, and C1 to C3-Flu (\*Concentrations of PAHs in soft tissue were standardized by total, and square root transformed for normality). Acronyms for PAHs given in SI-Table S4.

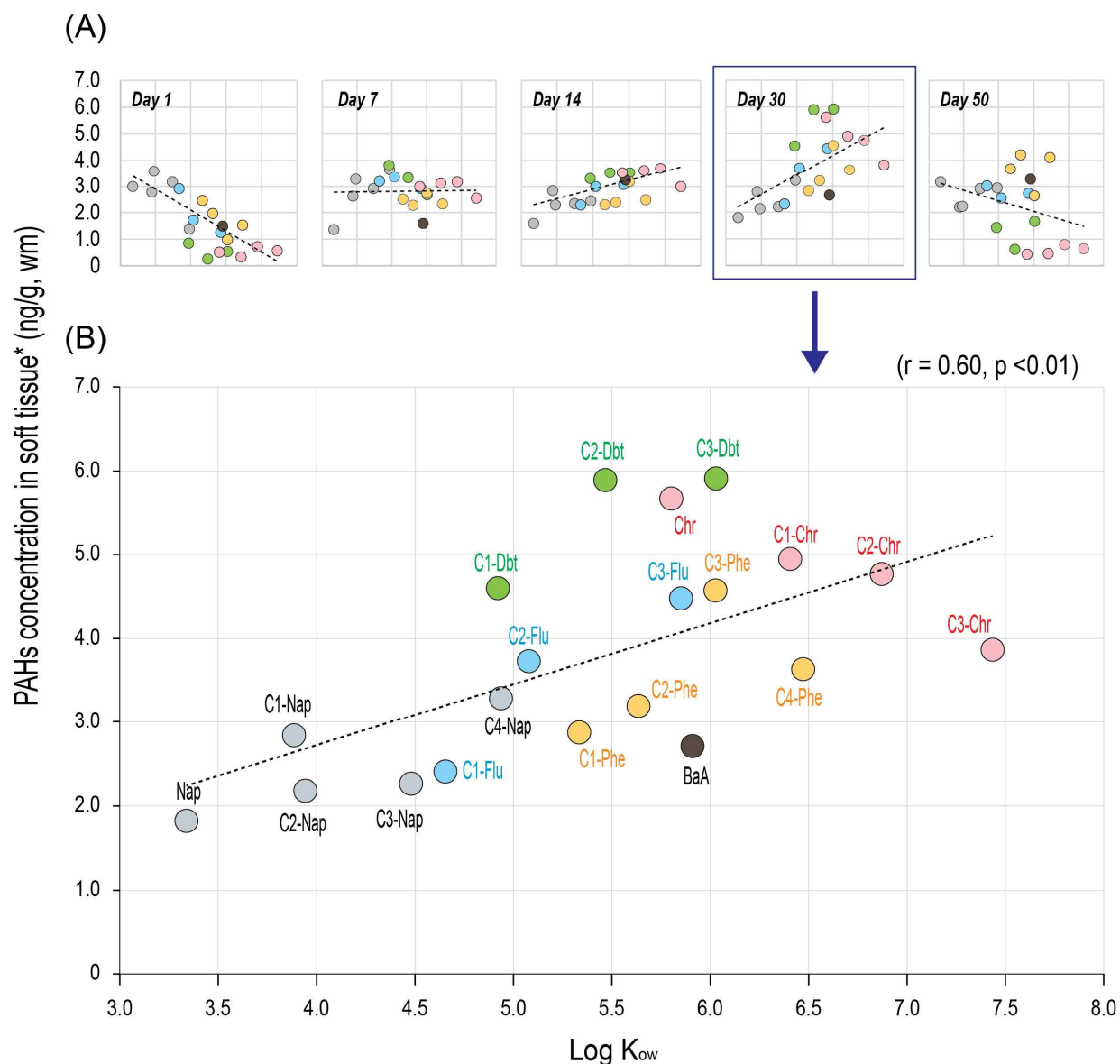


**Figure S5.** Relative compositions of PAHs and alkylated PAHs in crude oil (Iranian Heavy Crude, IHC), sediments contaminated by *Hebei Spirit* oil spill, and OSA made of IHC. (A) These results were referenced from a previous study (Hong et al. 2015) and (B) concentration of OSA quantified in this study (\*Comparisons of Bray-Curtis similarities in relative compositions of PAHs and their alkyl derivatives showed that the most resembled period in weathering of IHC compared to OSA). Acronyms for PAHs given in SI-Table S4.





**Figure S6.** (A) Stable isotopic ratios of carbon ( $\delta^{13}\text{C}$ ) and (B) nitrogen ( $\delta^{15}\text{N}$ ) in clams, OSAs, sediment, and crude oil. Blue, green, and red lines denote Control, OSA<sub>low</sub>, and OSA<sub>high</sub> treatment, respectively.



**Figure S7.** Relationship between Log  $K_{ow}$  and concentration of PAHs in soft tissue of clams, *Mactra veneriformis*. (A) Concentration of PAHs compounds in OSA<sub>high</sub> treatments at Days 1, 7, 14, 30, and 50. (B) The result of OSA<sub>high</sub> treatment at Day 30 as peak concentration of PAHs in soft tissue\* (\*The concentrations were standardized by total, and square root transformed for normality). Acronyms for PAHs given in SI-Table S4.

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