



Supplementary Information

Distribution, source and ecological risk assessment of parent and alkylated PAHs in coastal environment of Dalian, China after oil spill

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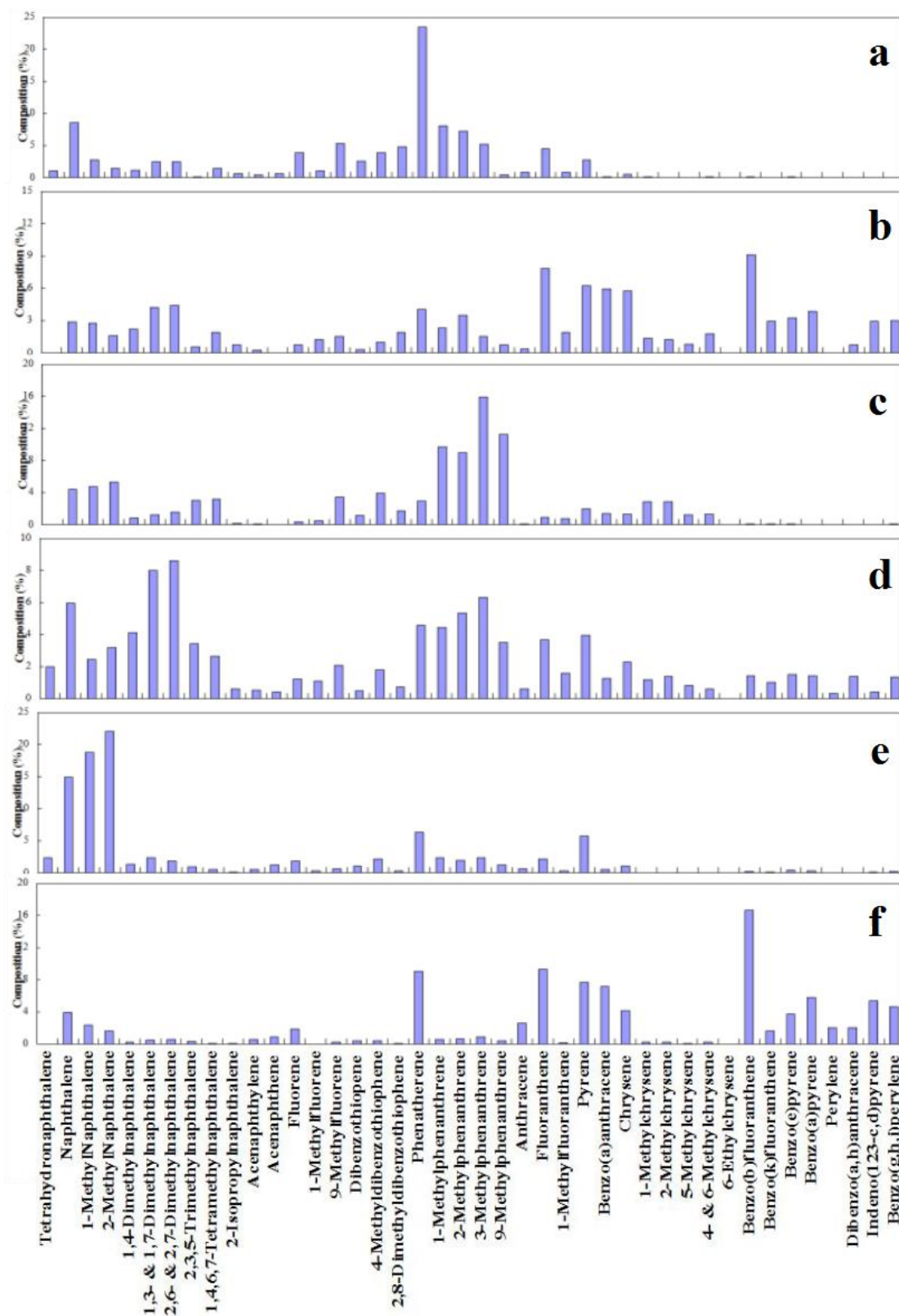


Figure S1. Compositions of 45 PAH congeners in multi-matrices in Dalian: a, gaseous phase; b, particle phase; c, crude oil; d, surface runoff; e, effluent of wastewater treatment plants; f, surface sediment.

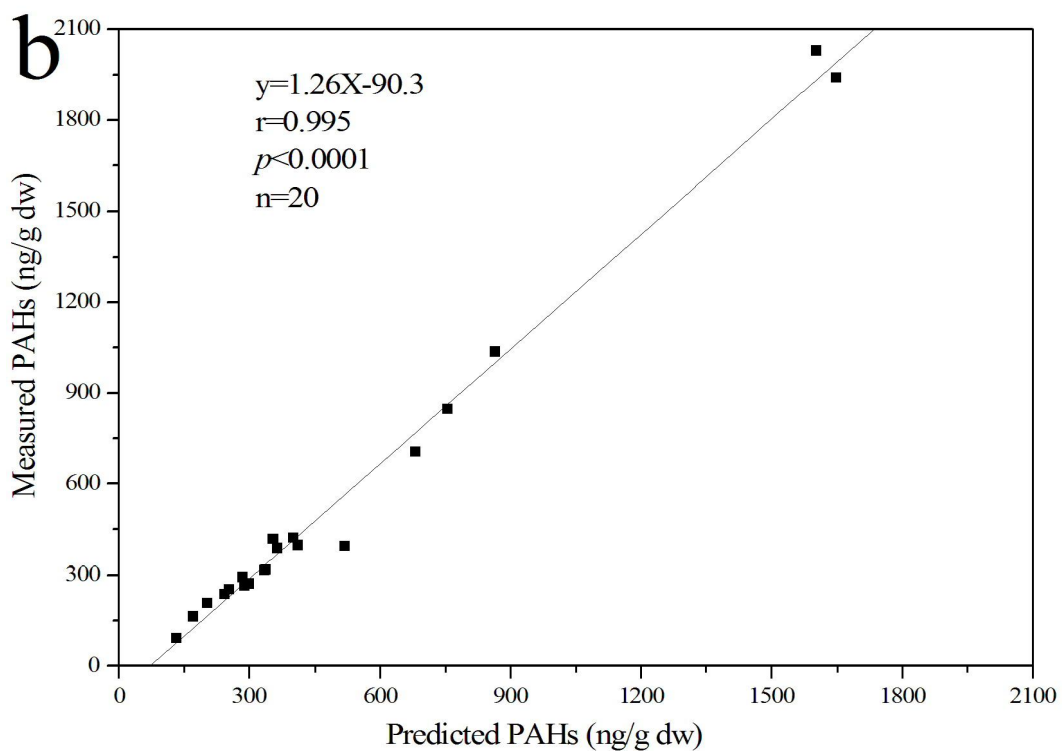
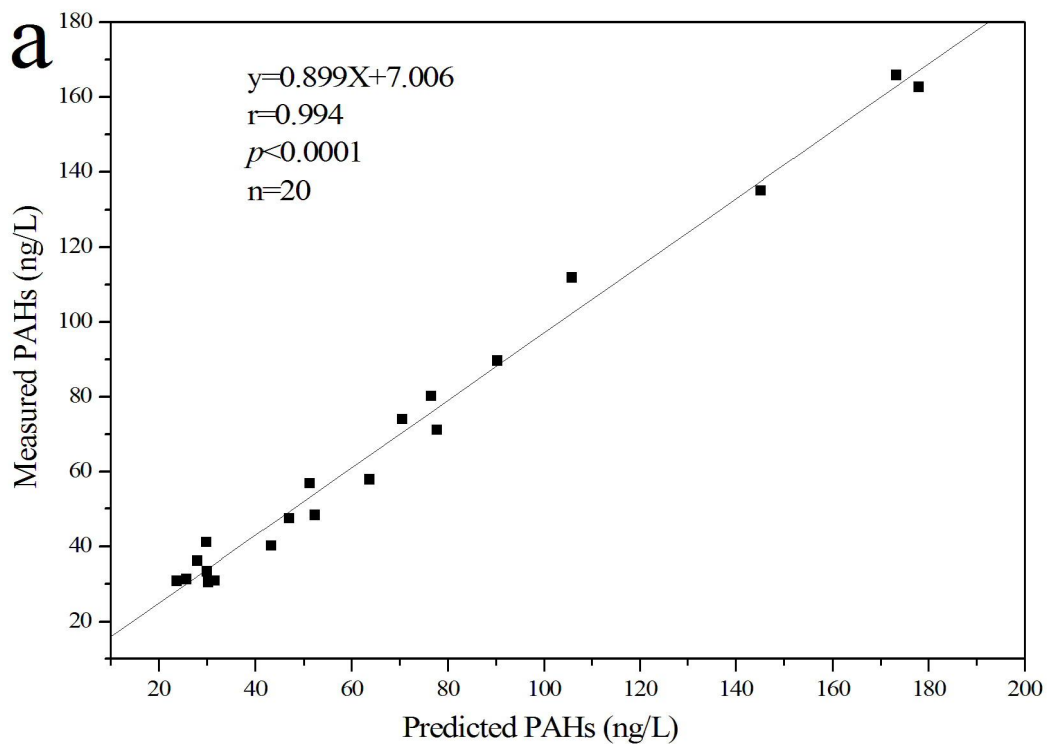


Figure S2. Comparison of measured and predicted PAHs concentrations obtained by PCA-MLR, (a) seawater, (b) sediment.

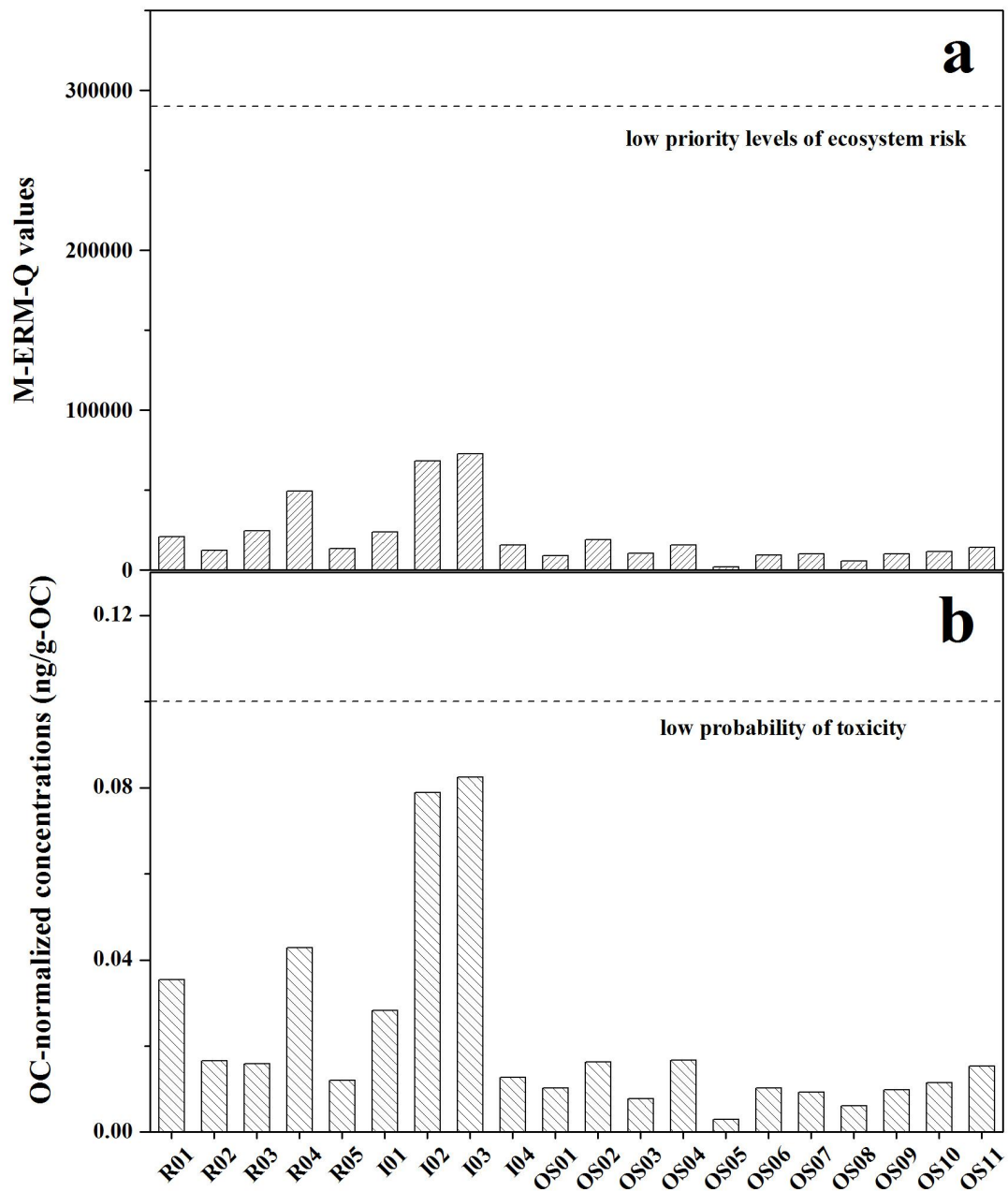


Figure S3. The OC-normalized concentrations (a) and M-ERM-Q values (b) of PAHs at different stations from coastal areas of Dalian.

Table S1 Some basic information for the five municipal sewage treatment plants.

Plant	Sewage treatment process	Sewage fate after treatment (Effluent fate)	Treatment capacity (10 ³ t/day)
Sew01	Cyclic Activated Sludge Technology (CAST)	Into the sea	70
Sew02	Biological Aerated Filter	Into the sea	120
Sew03	Biological Aerated Filter	Into the sea	120
Sew04	Anaerobic/Oxic	Into the sea	10
Sew05	Cyclic Activated Sludge Technology (CAST)	Into the sea	80

Table S2 Method detection limits values for parent- and alkyl-PAHs.

PAHs	Sample type	Water (pg/L)	Sediment (pg/g dry weight)	Gaseous phases (pg/m ³)	Particle phases (pg/m ³)
Tetrahydronaphthalene		155	31	0.15	0.13
Naphthalene		199	40	0.79	0.78
1-MethylNaphthalene		363	73	1.4	1.4
2-MethylNaphthalene		367	73	1.4	1.4
1,3- & 1,7-Dimethylnaphthalene		429	86	2.4	2.5
1,4-Dimethylnaphthalene		608	122	1.7	1.7
2,6- & 2,7-Dimethylnaphthalene		542	108	2.2	2.2
2,3,5-Trimethylnaphthalene		203	41	0.81	0.80
1,4,6,7-Tetramethylnaphthalene		181	36	0.73	0.71
2-Isopropylnaphthalene		257	51	1.0	1.0
Acenaphthylene		342	68	1.4	1.4
Acenaphthene		189	38	0.75	0.70
Fluorene		391	78	1.6	1.6
1-Methylfluorene		379	76	1.5	1.5
9-Methylfluorene		494	99	2.0	2.0
Dibenzothiopene		12	2	0.10	0.11
4-Methyldibenzothiophene		954	191	3.8	3.8
2,8-Dimethyldibenzothiophene		328	66	1.3	1.3
Phenanthrene		529	106	2.1	2.1
1-Methylphenanthrene		898	180	3.6	3.6
2-Methylphenanthrene		675	135	2.7	2.7
3-Methylphenanthrene		1047	209	4.0	4.0
9-Methylphenanthrene		582	116	2.4	2.4
Anthracene		365	73	1.5	1.5
Fluoranthene		167	33	0.67	0.69
1-Methylfluoranthene		353	71	1.4	1.4
Pyrene		196	39	0.78	0.75
Benzo(a)anthracene		139	28	0.56	0.51
Chrysene		105	21	0.42	0.40
1-Methylchrysene		1160	231	4.6	4.6
2-Methylchrysene		1020	204	4.1	4.1
4- & 6-Methylchrysene		997	199	6.3	6.3
5-Methylchrysene		1580	316	4.0	4.0
6-Ethylchrysene		1280	255	5.1	5.1
Benzo(b)fluoranthene		405	81	1.6	1.6
Benzo(k)fluoranthene		405	81	1.6	1.6
Benzo(e)pyrene		624	125	2.5	2.5
Benzo(a)pyrene		639	128	2.6	2.6
Perylene		525	105	2.1	2.0
Indeno(123-c,d)pyrene		89	18	3.5	3.5
Dibenzo(a,h)anthracene		89	18	3.5	3.5
Benzo(g,h,i)perylene		116	23	4.6	4.6

Table S3 Summary of PAHs concentrations in surface water (ng/L).

Chemical	Maximum	Minimum	Mean	SD	Detection rate (%)
1-Methylnaphthalene	31.7	2.27	9.85	8.72	100
2-Methylnaphthalene	30.1	3.84	11.9	8.13	100
1,4-Dimethylnaphthalene	6.74	1.36	3.18	1.80	45
1,3- & 1,7-Dimethylnaphthalene	5.83	1.06	2.40	1.39	100
2,6- & 2,7-Dimethylnaphthalene	1.73	BDL	0.430	0.565	100
2,3,5-Trimethylnaphthalene	4.32	0.258	1.01	0.859	100
1,4,6,7-Tetramethylnaphthalene	0.776	BDL	0.116	0.226	25
2-Isopropylnaphthalene	2.17	BDL	0.603	0.509	85
1-Methylfluorene	1.26	BDL	0.308	0.360	50
9-Methylfluorene	1.11	BDL	0.184	0.345	25
4-Methyldibenzothiophene	7.61	BDL	0.912	1.72	45
2,8-Dimethyldibenzothiophene	0.34	BDL	0.017	0.075	5
1-Methylphenanthrene	1.26	BDL	0.063	0.283	5
2-Methylphenanthrene	1.19	BDL	0.338	0.483	35
3-Methylphenanthrene	3.06	BDL	0.153	0.684	5
9-Methylphenanthrene	1.85	BDL	0.479	0.639	40
1-Methylfluoranthene	1.16	BDL	0.249	0.322	40
1-Methylchrysene	BDL	BDL	-	-	0
2-Methylchrysene	BDL	BDL	-	-	0
5-Methylchrysene	BDL	BDL	-	-	0
4- & 6-Methylchrysene	4.67	BDL	0.536	1.22	20
6-Ethylchrysene	3.45	BDL	0.173	0.772	5
Σ_{25} alkyl-PAHs	79.8	12.5	32.9	22.4	-

Table S4 Summary of PAHs concentrations in surface sediment (ng/g dw).

Chemical	Maximum	Minimum	Mean	SD	Detection rate (%)
1-Methylnaphthalene	37.1	2.74	13.2	10.2	100
2-Methylnaphthalene	21.4	3.35	8.99	5.02	100
1,4-Dimethylnaphthalene	6.77	1.64	3.36	1.43	100
1,3- & 1,7-Dimethylnaphthalene	6.08	1.22	2.67	1.40	100
2,6- & 2,7-Dimethylnaphthalene	4.68	0.538	1.41	0.939	100
2,3,5-Trimethylnaphthalene	4.02	0.818	1.62	0.906	100
1,4,6,7-Tetramethylnaphthalene	1.37	0.264	0.575	0.288	100
2-Isopropyl-naphthalene	0.470	0.100	0.256	0.114	100
1-Methylfluorene	0.576	0.0498	0.215	0.146	100
9-Methylfluorene	2.77	0.661	1.41	0.523	100
4-Methyldibenzothiophene	7.96	0.420	2.172	1.57	100
2,8-Dimethyldibenzothiophene	2.38	0.0900	0.270	0.500	100
1-Methylphenanthrene	10.7	1.06	3.01	2.13	100
2-Methylphenanthrene	11.7	1.56	3.73	2.52	100
3-Methylphenanthrene	15.5	2.16	5.10	3.99	100
9-Methylphenanthrene	6.59	0.794	2.13	1.36	100
1-Methylfluoranthene	2.98	0.272	0.905	0.771	100
1-Methylchrysene	5.26	0.291	1.35	1.36	100
2-Methylchrysene	5.40	0.440	1.53	1.26	100
5-Methylchrysene	3.09	0.000	0.610	0.715	80
4- & 6-Methylchrysene	4.27	0.297	1.19	1.11	100
6-Ethylchrysene	1.62	BDL	0.199	0.383	40
Σ_{25} alkyl-PAHs	156	22.2	55.9	34.8	-

Table S5 Concentrations of alkyl-PAHs in sediment (ng/g dw) from different locations around world.

Location	Year	Sampling number	Concentration	Reference
Spanish river	2003	36	13-13564 (1530)	Planas et al., 2003
Rural, Laos	2005	4	6-11 (8)	Saha et al., 2009
Urban, Laos	2005	5	187-1007 (439)	
Rural, Cambodia	2004	15	3-213 (38)	
Urban, Cambodia	2004	4	221-2382 (1311)	
Rural, Vientam	2004	6	8-122 (41)	
Urban, Vientam	2000-2004	13	25-2997 (786)	
Rural, Thailand	2003	18	2-34 (17)	
Urban, Thailand	2003	17	28-821 (306)	
Rural, Philippines	2002	4	1-98 (34)	
Urban, Philippines	2002	6	205-2287 (781)	
Rural, Indonesia	1998	6	3-109 (35)	
Urban, Indonesia	2000	4	514-2733 (1807)	
Rural, Malaysia	1998-1999	17	0-9 (3)	
Urban, Malaysia	1998-2004	17	6-217 (100)	
Rural, India	2002-2006	21	6-41 (17)	
Urban, India	2003-2006	17	12-6680 (1679)	
Urban, Japan	2005-2006	24	53-2538 (521)	
Osaka Bay, Japan	2000-2006	44	13.7-1700	
Mohang Harbor, South Korea	2009	8	10.5-331 (221)	Kim et al., 2014
The eastern coast of Qatar, Arabian Gulf	2012	16	0.6-988 (96.7)	Soliman et al., 2014

Table S6 Summary of PAHs concentrations in air (ng/m³).

Chemical	Gaseous phase (ng/m ³)			Particle phase (ng/m ³)		
	GP01	GP02	GP03	PP01	PP02	PP03
Tetrahydronaphthalene	0.238	0.246	0.237	BDL	BDL	BDL
Naphthalene	2.22	1.79	1.87	0.0574	0.0294	0.0498
1-Methylnaphthalene	1.02	0.418	0.486	0.0653	0.0339	0.0345
2-Methylnaphthalene	0.543	0.213	0.244	0.0297	0.0122	0.0339
1,4-Dimethylnaphthalene	0.531	0.107	0.155	0.0388	0.0279	0.0399
1,3- & 1,7-Dimethylnaphthalene	1.13	0.251	0.332	0.0736	0.0460	0.0828
2,6- & 2,7-Dimethylnaphthalene	1.14	0.228	0.294	0.0739	0.0504	0.0865
2,3,5-Trimethylnaphthalene	0.0381	0.0121	0.0153	0.00736	0.00672	0.0144
1,4,6,7-Tetramethylnaphthalene	0.571	0.137	0.257	0.0345	0.0225	0.0334
2-Isopropylnaphthalene	0.248	0.0753	0.125	0.0126	0.0109	0.0128
Acenaphthylene	0.198	0.0589	0.0625	0.00553	0.00132	0.00602
Acenaphthene	0.313	0.0408	0.0746	BDL	BDL	BDL
Fluorene	1.51	0.447	0.663	0.0121	0.0101	0.0145
1-Methylfluorene	0.451	0.111	0.159	0.0235	0.0167	0.0188
9-Methylfluorene	2.09	0.615	0.903	0.0384	0.0168	0.0198
Dibenzothiophene	0.971	0.381	0.415	0.00661	0.00404	0.00567
4-Methyldibenzothiophene	1.61	0.496	0.564	0.0223	0.0116	0.0125
2,8-Dimethyldibenzothiophene	1.83	0.670	0.786	0.0452	0.0240	0.0229
Phenathrene	8.51	3.66	3.82	0.0808	0.0454	0.0688
1-Methylphenanthrene	2.96	1.18	1.36	0.0637	0.0291	0.0198
2-Methylphenanthrene	2.74	1.05	1.18	0.101	0.0350	0.0326
3-Methylphenanthrene	1.99	0.737	0.841	0.0380	0.0151	0.0194
9-Methylphenanthrene	0.157	0.0739	0.0953	0.0125	0.0133	0.0114
Anthracene	0.203	0.192	0.202	0.00569	0.00662	0.00530
Fluoranthene	1.71	0.693	0.652	0.139	0.114	0.120
1-Methylfluoranthene	0.239	0.145	0.157	0.0419	0.0305	0.0180

Chemical	Gaseous phase (ng/m ³)			Particle phase (ng/m ³)		
	GP01	GP02	GP03	PP01	PP02	PP03
Pyrene	1.17	0.399	0.359	0.110	0.0971	0.0921
Benzo(a)anthracene	0.0328	0.0202	0.0363	0.107	0.0849	0.092
Chrysene	0.147	0.0748	0.111	0.109	0.0811	0.0859
1-Methylchrysene	0.0275	0.0277	0.0273	0.0334	0.0165	0.0161
2-Methylchrysene	0.0114	0.0138	0.0191	0.0162	0.00906	0.0354
5-Methylchrysene	0.0150	0.0135	0.0148	0.0141	0.0123	0.0124
4- & 6-Methylchrysene	0.0254	0.0218	0.0384	0.0559	0.0122	0.0124
6-Ethylchrysene	BDL	BDL	BDL	BDL	BDL	BDL
Benzo(b)flouranthene	0.0371	0.0182	0.0241	0.177	0.127	0.131
Benzo(k)flouranthene	0.0249	0.0119	0.0136	0.0586	0.0416	0.0397
Benzo(e)pyrene	0.0247	0.0128	0.0186	0.0624	0.0480	0.0460
Benzo(a)pyrene	0.0229	0.0132	0.0141	0.0691	0.0532	0.0632
Perylene	BDL	BDL	BDL	BDL	BDL	BDL
Dibenzo(a,h)anthracene	0.00754	0.00791	0.0173	0.0151	0.00851	0.0116
Indeno(123-c,d)pyrene	0.0312	0.0122	0.00656	0.0781	0.0309	0.0318
Benzo(g,h,i)perylene	0.0257	0.0115	0.0134	0.0816	0.0334	0.0296
Σ ₁₆ EPA-PAHs	16.2	7.45	7.94	1.11	0.764	0.841
Σ ₂₅ alkyl-PAHs	19.4	6.60	8.05	0.841	0.455	0.592
Σ ₄₅ PAHs	36.8	14.7	16.7	2.02	1.27	1.49

Table S7 Summary of PAHs concentrations (ng/g dw) in Crude oil and Oil dispersant (ng/mL).

Chemical	Crude oil (ng/g dw)			Oil dispersant (ng/mL)		
	CO01	CO02	CO03	OD01	OD02	OD03
Tetrahydronaphthalene	BDL	BDL	BDL	BDL	BDL	BDL
Naphthalene	490	486	490	BDL	BDL	BDL
1-Methylnaphthalene	496	494	578	3.29	3.56	3.25
2-Methylnaphthalene	622	520	624	9.96	11.1	10.6
1,4-Dimethylnaphthalene	92.6	77.6	96.0	14.4	14.5	14.6
1,3- & 1,7-Dimethylnaphthalene	138	131	151	15.5	16.2	16.1
2,6- & 2,7-Dimethylnaphthalene	179	159	172	14.4	13.9	14.0
2,3,5-Trimethylnaphthalene	286	346	378	5.19	4.48	5.42
1,4,6,7-Tetramethylnaphthalene	396	302	344	5.52	5.15	5.26
2-Isopropyl naphthalene	19.0	25.8	19.9	BDL	BDL	BDL
Acenaphthylene	10.5	10.4	11.2	BDL	BDL	BDL
Acenaphthene	BDL	BDL	BDL	BDL	BDL	BDL
Fluorene	41.6	39	41.6	0.276	0.377	0.276
1-Methylfluorene	58.6	45.8	55.2	4.51	4.02	4.16
9-Methylfluorene	412	356	380	4.60	3.99	4.36
Dibenzothiophene	129	121	127	0.501	0.389	0.448
4-Methyldibenzothiophene	448	424	432	BDL	BDL	BDL
2,8-Dimethyldibenzothiophene	202	191	185	5.54	5.89	5.57
Phenathrene	344	300	328	1.98	1.60	1.81
1-Methylphenanthrene	1060	1074	1060	2.10	1.81	2.05
2-Methylphenanthrene	1066	948	962	4.56	3.48	3.90
3-Methylphenanthrene	1770	1670	1800	3.12	3.03	3.13
9-Methylphenanthrene	1240	1210	1250	3.43	3.33	3.89
Anthracene	14.5	14.8	16.4	1.88	1.65	1.97
Fluoranthene	105	101	99.8	3.25	2.60	3.13

Chemical	Crude oil (ng/g dw)			Oil dispersant (ng/mL)		
	CO01	CO02	CO03	OD01	OD02	OD03
1-Methylfluoranthene	87.6	78.2	83.2	5.10	5.37	5.55
Pyrene	224	212	218	2.00	1.65	1.77
Benzo(a)anthracene	139	163	158	4.22	3.65	3.66
Chrysene	136	151	150	4.07	3.17	3.90
1-Methylchrysene	294	332	320	6.73	6.57	6.65
2-Methylchrysene	304	332	326	4.65	4.95	4.58
5-Methylchrysene	134	135	138	4.59	3.85	4.37
4- & 6-Methylchrysene	141	146	140	5.36	5.68	5.23
6-Ethylchrysene	BDL	BDL	BDL	BDL	BDL	BDL
Benzo(b)fluoranthene	8.08	10.6	10.2	2.11	1.92	1.97
Benzo(k)fluoranthene	8.76	4.74	5.34	BDL	BDL	BDL
Benzo(e)pyrene	7.9	7.96	7.12	BDL	BDL	BDL
Benzo(a)pyrene	5.16	7.04	4.98	BDL	BDL	BDL
Perylene	2.14	3.36	2.32	BDL	BDL	BDL
Dibenzo(a,h)anthracene	4.36	3.88	4.12	BDL	BDL	BDL
Indeno(123-c,d)pyrene	2.44	4.28	5.31	BDL	BDL	BDL
Benzo(g,h,i)perylene	6.84	9.00	6.32	BDL	BDL	BDL
Σ_{16} EPA-PAHs	1540	1520	1550	19.8	16.6	18.5
Σ_{25} alkyl-PAHs	9450	9000	9500	123	121	112
Σ_{45} PAHs	11100	10600	11200	143	138	131

Table S8 Factor pattern for PAHs in seawater in Dalian.

PAHs	PC1	PC2	PC3	PC4	PC5	PC6
Tetrahydronaphthalene	0.10	0.21	-0.09	-0.07	0.93	0.03
Naphthalene	0.21	-0.09	0.94	-0.11	0.15	0.00
1-Methylnaphthalene	-0.04	-0.11	0.98	0.01	-0.03	-0.09
2-Methylnaphthalene	-0.04	-0.11	0.98	0.01	-0.03	-0.09
1,4-Dimethylnaphthalene	0.67	0.70	-0.02	0.10	0.05	-0.06
1,3- & 1,7-Dimethylnaphthalene	0.24	0.92	-0.08	0.00	0.17	-0.04
2,6- & 2,7-Dimethylnaphthalene	0.13	0.94	-0.05	-0.18	0.16	-0.01
2,3,5-Trimethylnaphthalene	0.48	0.84	-0.04	-0.01	0.08	-0.07
1,4,6,7-Tetramethylnaphthalene	0.31	0.75	0.03	-0.13	0.26	0.14
2-Isopropylnaphthalene	0.96	-0.01	0.02	-0.11	-0.05	-0.04
Acenaphthene	-0.07	0.82	-0.10	0.01	-0.17	-0.18
Fluorene	0.14	0.94	-0.07	-0.04	0.24	-0.04
1-Methylfluorene	-0.02	0.26	-0.06	-0.01	0.20	-0.05
9-Methylfluorene	0.96	0.19	-0.04	0.03	-0.08	-0.06
4-Methyldibenzothiophene	0.88	0.03	0.35	0.11	0.13	0.03
Phenathrene	0.03	0.54	-0.10	-0.04	0.77	-0.16
1-Methylphenanthrene	0.78	0.31	-0.12	-0.05	0.31	-0.03
2-Methylphenanthrene	0.90	-0.30	0.02	0.04	-0.07	-0.10
3-Methylphenanthrene	0.98	-0.11	-0.04	0.01	-0.03	-0.09
9-Methylphenanthrene	0.79	-0.15	-0.01	-0.48	-0.01	0.27
Anthracene	0.29	0.88	-0.04	0.02	0.17	-0.12
Fluoranthene	0.08	-0.10	-0.04	0.31	0.21	0.66
1-Methylfluoranthene	0.06	0.46	-0.11	0.04	0.37	-0.02
Pyrene	0.13	0.17	0.04	0.35	0.33	0.77
Benzo(a)anthracene	0.00	0.18	-0.15	-0.18	-0.07	0.89
Chrysene	-0.16	-0.14	-0.15	0.05	0.11	0.91
1-Methylchrysene	0.96	0.15	-0.09	0.02	0.03	-0.05
2-Methylchrysene	0.01	0.16	-0.02	-0.02	0.97	0.02
4- & 6--Methylchrysene	0.07	0.39	0.00	0.03	0.83	0.13
6-Ethylchrysene	0.88	0.11	-0.05	0.11	0.06	-0.06
Benzo(b)fluoranthene	-0.09	-0.08	-0.22	0.87	-0.02	-0.09
Benzo(k)fluoranthene	-0.03	-0.06	0.05	0.89	-0.03	-0.10
Benzo(e)pyrene	-0.20	-0.28	-0.12	0.23	0.03	0.57
Benzo(a)pyrene	0.00	-0.18	0.23	0.79	-0.16	0.54
Perylene	0.14	0.00	0.19	0.80	0.09	0.31
Explained variance (%)	37.9	20.4	10.8	7.8	5.0	4.0

Table S9 Factor pattern for PAHs in sediment in Dalian.

PAHs	PC1	PC2
Tetrahydronaphthalene	0.82	0.29
Naphthalene	0.73	0.63
1-Methylnaphthalene	0.88	0.35
2-Methylnaphthalene	0.91	0.33
1,4-Dimethylnaphthalene	0.88	0.32
1,3- & 1,7-Dimethylnaphthalene	0.86	0.45
2,6- & 2,7-Dimethylnaphthalene	0.82	0.44
2,3,5-Trimethylnaphthalene	0.75	0.58
1,4,6,7-Tetramethylnaphthalene	0.67	0.64
2-Isopropyl naphthalene	0.45	0.47
Acenaphthylene	0.74	0.60
Acenaphthene	0.77	0.47
Fluorene	0.91	0.36
1-Methylfluorene	0.49	0.79
9-Methylfluorene	0.63	0.62
Dibenzothiopene	0.32	0.86
4-Methyldibenzothiophene	0.31	0.87
2,8-Dimethyldibenzothiophene	0.25	0.91
Phenanthrene	0.84	0.49
1-Methylphenanthrene	0.55	0.82
2-Methylphenanthrene	0.68	0.70
3-Methylphenanthrene	0.37	0.73
9-Methylphenanthrene	0.63	0.75
Anthracene	0.52	0.40
Fluoranthene	0.88	0.45
1-Methylfluoranthene	0.59	0.79
Pyrene	0.81	0.57
Benzo(a)anthracene	0.90	0.35
Chrysene	0.85	0.51
1-Methylchrysene	0.09	0.88
2-Methylchrysene	0.63	0.77
5-Methylchrysene	0.61	0.76
4- & 6--Methylchrysene	0.53	0.83
6-Ethylchrysene	0.09	0.16
Benzo(b)fluoranthene	0.87	0.47
Benzo(k)fluoranthene	0.52	0.38
Benzo(e)pyrene	0.81	0.57
Benzo(a)pyrene	0.85	0.52
Perylene	0.83	0.51
Dibenzo(a,h)anthracene	0.81	0.55
Indeno(123-c,d)pyrene	0.81	0.57
Benzo(g,h,i)perylene	0.73	0.66
Explained variance (%)	70.8	18.3

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