

Supplementary Material

Distribution and source estimation of polycyclic aromatic hydrocarbons in coastal sediments from Seto Inland Sea, Japan

Hiroaki Tsuji,^A Waqar Azeem Jadoon,^{A,B} Yoko Nunome,^C Hideo Yamazaki,^D Satoshi Asaoka,^E Kazuhiko Takeda^{A,C} and Hiroshi Sakugawa^{A,C,F}

^AGraduate School of Biosphere Science, Hiroshima University, 1-7-1 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8521, Japan.

^BDepartment of Environmental Sciences, Hazara University, Mansehra, Khyber Pakhtunkhwa, Pakistan.

^CGraduate School of Integrated Sciences for Life, Hiroshima University, 1-7-1 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8521, Japan.

^DGraduate School of Science and Engineering, Kindai University, 3-4-1 Kowakae, Higashi-Osaka, Osaka 577-8502, Japan.

^EResearch Center for Inland Seas, Kobe University, 5-1-1 Fukaeminami, Higashinada, Kobe, 658-0022, Japan.

^FCorresponding author. Email: hsakuga@hiroshima-u.ac.jp

Table S1. Description of the 2015 sampling sites.

Sampling Site	Latitude	Longitude	Water depth (m)	Sediment core length (cm)	Sediment grain size
Osaka Bay					
St. 7a	N 34.38.092	E 135.20.010	16.0	40	Clay / Silt
St. 8a	N 34.34.995	E 135.15.037	19.7	20	Clay / Silt
St. 9a	N 34.29.957	E 135.10.118	35.9	10	Sand / Silt
St. 10a	N 34.25.097	E 135.05.050	36.0	10	Clay / Silt

Table S2. Description of the 2016 sampling sites.

Sampling Site	Latitude	Longitude	Water depth (m)	Sediment core length (cm)	Sediment grain size	Water content (%)
Aki-Nada						
St. 1	N 34.12.010	E 132.36.121	13.8	10	Clay / Silt	60.7
St. 2	N 34.09.040	E 132.36.425	22.5	6	Clay / Silt	49.7
St. 3	N 34.05.471	E 132.37.232	46.6	6	Clay / Silt	45.4
Harima-Nada						
St. 4	N 34.35.075	E 134.39.033	27.0	18	Clay / Silt	74.0
St. 5	N 34.30.885	E 134.35.040	35.0	24	Clay / Silt	79.4
St. 6	N 34.24.969	E 134.35.046	40.0	22	Clay / Silt	78.7
Osaka Bay						
St. 7b	N 34.38.348	E 135.19.792	15.0	22	Clay / Silt	75.3
St. 7c	N 34.35.874	E 135.22.506	12.3	30	Clay / Silt	77.4
St. 8b	N 34.34.803	E 135.15.118	19.0	18	Clay / Silt	61.0
St. 9b	N 34.30.045	E 135.09.968	36.0	8	Sand / Silt	31.9
St. 10b	N 34.24.868	E 135.04.920	36.9	14	Clay / Silt	67.0
Kii Channel						
St. 11	N 34.09.765	E 134.55.117	64.0	10	Sand / Silt	44.0
St. 12	N 33.59.960	E 134.55.081	68.8	8	Sand / Silt	35.5

Table S3. Linear calibration curves and detection limits of 17 PAHs.

PAH	Correlation coefficient (n=17)	Detection Limit (pg g ⁻¹ dw)
Nap	0.9970	51.9
Acy	0.9963	37.9
Ace	0.9962	33.3
Flu	0.9956	78.7
Phe	0.9954	27.5
Ant	0.9949	64.5
Flt	0.9957	31.9
Pyr	0.9959	31.6
BaA	0.9840	19.7
Chr	0.9898	111
BbF	0.9881	40.2
BkF	0.9919	50.5
BeP	0.9937	49.8
BaP	0.9885	43.2
IncdP	0.9901	41.4
DahA	0.9740	31.0
BghiP	0.9910	49.5

Table S4. The concentrations of 17 PAHs in surface sediments at each sampling site in 2015 (ng g⁻¹ dw, nd: not detected).

	St. 8a	St. 9a	St. 10a
Nap	nd	1.22	1.17
Acy	2.18	0.90	1.72
Ace	2.40	0.47	0.62
Flu	6.18	20.0	4.80
Phe	19.3	3.70	10.1
Ant	6.77	1.21	3.00
Flt	38.6	6.00	21.1
Pyr	30.2	5.51	19.1
BaA	33.8	5.57	18.8
Chr	33.6	6.00	20.4
BbF	57.2	17.9	57.1
BkF	37.8	1.64	4.53
BeP	23.6	5.84	16.8
BaP	40.3	8.20	26.3
IcdP	70.0	13.0	52.7
DahA	19.6	3.74	6.26
BghiP	49.5	7.56	24.0
Σ17PAH	471	108	288

Table S5. The concentrations of 17 PAHs in the surface sediments at each sampling site in 2016 (ng g⁻¹ dw).

	St. 1	St. 2	St. 3	St. 4	St. 5	St. 6	St. 11	St. 12
Nap	4.50	5.38	3.44	6.74	3.43	4.11	2.36	1.11
Acy	3.24	2.15	1.41	3.73	2.26	2.89	1.16	0.52
Ace	2.01	1.53	1.28	2.59	1.41	2.27	1.19	0.61
Flu	2.85	3.48	3.13	3.78	2.56	2.95	2.82	1.03
Phe	26.2	15.6	11.1	15.6	14.7	14.7	10.3	7.04
Ant	8.72	4.36	2.74	5.49	4.52	3.99	2.08	1.30
Flt	59.7	20.7	11.5	32.0	26.7	32.7	7.96	7.06
Pyr	54.2	16.3	10.0	31.7	23.6	26.2	7.58	5.10
BaA	45.2	19.0	9.25	18.8	29.0	17.8	6.71	2.77
Chr	38.9	18.6	10.1	23.6	28.0	26.6	9.11	4.87
BbF	40.8	12.9	5.62	23.5	19.0	16.4	6.86	8.43
BkF	17.7	10.0	4.01	23.9	14.1	22.9	3.71	3.50
BeP	21.0	7.05	3.21	19.1	10.7	18.0	3.63	4.31
BaP	31.0	10.7	4.31	23.2	16.3	19.8	3.76	3.48
IcdP	35.7	16.3	6.20	21.8	25.5	31.6	5.95	5.67
DahA	4.93	4.53	2.01	12.5	6.14	6.22	2.08	2.30
BghiP	19.6	11.0	4.50	21.8	17.9	19.7	5.07	6.67
Σ17PAH	416	179	93.8	290	246	269	82.3	65.8

Table S6. The concentrations of 17 PAHs in the sediment core recovered from St. 7a in 2015 (ng g⁻¹ dw).

Depth (cm)	Year	Nap	Acy	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BeP	BaP	IncdP	DahA	BghiP	Σ17PAH
0–5	2009.7	10.5	6.06	20.8	66.1	112	47.0	116	113	97.1	107	194	116	102	141	161	36.5	143	1590
5–10	2001.0	12.7	5.45	9.68	25.5	62.2	20.9	97.2	99.3	110	116	212	136	111	162	189	44.1	152	1560
10–15	1995.3	9.12	4.85	5.66	16.3	49.4	15.1	78.1	88.7	92.8	102	183	126	100	152	170	40.5	134	1370
15–20	1989.4	13.2	6.26	3.77	8.65	46.9	14.9	83.5	83.4	63.6	111	198	114	99.0	167	232	60.9	186	1490
20–25	1983.6	9.94	6.46	4.50	11.0	44.1	15.4	80.3	78.5	82.9	82.9	178	119	93.2	151	220	58.1	176	1410
25–30	1977.8	21.7	11.6	7.25	16.3	95.6	29.6	141	138	179	159	348	232	160	306	470	140	358	2810
30–35	1971.9	9.86	8.31	4.21	8.94	47.0	25.7	97.5	95.9	115	106	228	157	114	210	292	83.4	224	1830
35–40	1966.0	18.6	8.98	6.17	14.4	55.5	21.0	99.0	102	123	112	258	166	125	224	316	89.6	243	1980

Table S7. The concentrations of 17 PAHs in the sediment core recovered from St. 7b in 2016 (ng g⁻¹ dw).

Depth (cm)	Nap	Acy	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BeP	BaP	IncdP	DahA	BghiP	Σ17PAH
0–2	4.16	2.64	1.39	2.58	13.9	4.79	22.7	23.1	30.7	29.0	24.3	14.8	11.6	20.3	28.1	8.12	19.3	261
2–4	6.96	4.07	2.14	4.18	21.5	7.78	33.7	34.6	37.6	37.3	29.0	17.7	14.9	24.8	28.3	7.73	21.3	334
4–6	5.42	3.40	1.85	3.40	17.2	6.18	28.2	28.3	30.9	30.8	21.2	13.1	11.2	18.6	21.7	6.03	16.2	264
6–8	6.83	4.40	2.34	4.31	21.8	7.74	31.8	32.0	34.5	34.3	26.0	15.7	14.2	23.5	27.1	7.57	20.3	314
8–10	9.64	6.22	3.26	6.07	30.2	10.9	49.6	49.7	51.8	52.8	35.6	23.1	19.4	32.1	37.3	10.4	27.8	456
10–12	6.44	4.11	2.21	4.09	20.6	7.52	29.7	29.8	32.6	32.1	23.1	15.1	13.0	21.6	25.5	7.15	18.8	293
12–14	6.45	5.85	2.68	6.26	62.3	25.1	120	103	81.0	61.6	113	50.9	55.2	61.0	87.1	21.3	77.9	941
14–16	6.19	4.08	2.27	4.18	32.2	8.30	60.2	58.5	47.7	40.1	78.6	35.3	38.9	47.9	64.8	15.7	58.9	604
16–18	8.51	5.33	3.26	4.99	48.6	12.9	87.5	87.4	67.2	52.1	105	47.2	52.4	67.6	87.3	20.8	78.4	836
18–20	6.52	4.18	2.08	4.08	32.3	8.13	59.1	62.1	52.0	42.7	84.7	38.1	41.8	52.7	53.6	19.5	64.6	628
20–22	6.87	4.20	2.66	4.37	36.6	8.02	69.1	69.3	54.7	46.5	91.3	41.1	45.5	56.3	60.1	17.8	66.7	681

Table S8. The concentrations of 17 PAHs in the sediment core recovered from St. 7c in 2016 (ng g⁻¹ dw).

Depth (cm)	Nap	Acy	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BeP	BaP	IncdP	DahA	BghiP	Σ17PAH
0–2	9.36	4.83	3.68	5.18	33.7	10.7	50.4	51.3	38.3	43.9	77.1	34.5	36.4	46.1	89.1	14.6	55.2	604
2–4	6.99	3.64	2.84	3.90	28.6	10.2	48.4	48.4	38.4	37.3	68.8	30.9	32.8	39.9	59.5	12.2	36.5	509
4–6	12.2	5.81	4.22	5.70	44.2	14.9	74.3	73.2	60.7	57.2	129	58.2	65.0	87.0	97.8	27.3	62.1	879
6–8	7.50	4.32	3.23	4.18	33.3	10.4	53.9	52.6	44.4	42.1	78.2	35.1	36.9	47.2	64.8	15.4	41.9	576
8–10	7.30	3.53	2.15	3.69	31.2	9.53	51.6	52.1	36.1	29.9	71.0	31.8	33.1	41.1	54.7	12.4	52.3	524
10–12	9.34	3.76	2.47	4.27	32.9	9.64	59.1	58.7	42.9	35.6	85.9	38.5	40.0	51.1	64.1	15.7	60.0	614
12–14	7.83	3.64	2.27	3.59	31.8	9.08	56.7	55.0	43.5	34.9	81.6	36.7	38.5	49.6	59.1	15.6	54.1	584
14–16	5.02	2.55	1.57	2.72	14.0	5.97	23.5	26.5	31.6	29.3	26.8	17.2	13.6	23.2	29.3	8.40	22.3	284
16–18	5.06	2.90	1.74	2.91	16.0	6.60	27.5	29.5	39.1	34.9	29.4	19.7	15.2	27.6	35.4	10.4	25.5	329
18–20	8.94	4.69	2.74	4.57	23.6	10.2	42.0	46.1	58.8	51.6	41.6	35.2	23.6	42.1	53.8	15.8	39.0	505
20–22	7.36	4.16	2.44	3.88	21.8	10.1	39.0	42.0	60.2	51.9	42.1	30.5	22.4	42.3	55.2	15.1	38.6	489
22–24	6.92	3.85	2.22	3.65	20.9	8.70	39.9	43.1	58.4	49.1	42.8	27.4	21.5	39.6	48.3	14.0	34.6	465
24–26	8.28	4.73	3.43	4.84	25.5	11.5	49.7	52.2	68.7	58.4	52.4	32.8	25.8	48.1	58.7	17.1	41.4	564
26–28	6.73	3.58	2.26	3.80	20.9	21.5	6.15	31.1	121	44.0	39.3	26.4	19.6	37.8	49.8	14.8	34.7	484
28–30	6.25	3.16	1.80	3.38	18.7	8.37	32.5	33.6	47.7	41.8	38.6	22.5	18.2	34.2	42.7	13.1	29.4	396

Table S9. The concentrations of 17 PAHs in the sediment core recovered from St. 8b in 2016 (ng g⁻¹ dw).

Depth (cm)	Nap	Acy	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BeP	BaP	IncdP	DahA	BghiP	Σ17PAH
0–2	3.20	3.44	1.27	2.66	18.2	6.35	43.9	42.5	42.2	38.1	22.0	20.1	14.8	26.3	26.8	7.27	20.1	339
2–4	4.53	2.60	1.79	3.32	15.9	4.62	26.3	24.5	27.2	28.0	22.0	16.1	13.0	19.3	26.5	6.94	17.2	260
4–6	3.66	2.48	1.52	3.07	14.8	4.65	23.2	21.6	26.4	26.2	22.0	13.4	11.8	18.9	23.1	6.49	17.3	240
6–8	4.41	2.45	1.69	3.22	15.4	4.81	24.1	23.3	29.3	27.8	21.1	14.6	11.1	18.1	21.3	6.62	15.9	245
8–10	4.00	2.58	1.87	3.48	16.4	6.01	27.3	25.1	31.7	29.8	24.4	14.7	12.2	19.3	23.8	7.09	18.1	268
10–12	3.84	2.46	1.45	2.92	13.5	4.28	21.1	20.1	23.5	23.2	18.3	15.7	11.2	16.6	20.1	5.95	15.8	220
12–14	4.50	3.14	1.85	3.23	17.4	5.52	30.4	29.1	33.9	33.9	25.7	21.7	15.5	23.6	27.3	8.42	20.8	306
14–16	4.66	3.59	1.90	3.58	19.0	6.56	37.0	37.1	48.8	44.8	33.7	26.6	19.5	30.7	37.0	10.7	26.9	392
16–18	4.56	3.56	2.08	3.46	18.2	6.39	34.2	33.7	43.9	42.1	31.1	25.5	18.5	29.8	34.8	10.2	25.2	367

Table S10. The concentrations of 17 PAHs in the sediment core recovered from St. 9b in 2016 (ng g⁻¹ dw).

Depth (cm)	Nap	Acy	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BeP	BaP	IncdP	DahA	BghiP	Σ17PAH
0–2	2.74	1.20	1.03	1.65	5.75	2.05	5.82	5.49	5.31	6.72	6.38	4.38	4.00	4.52	5.54	3.14	4.86	70.6
2–4	1.83	1.12	1.00	1.64	5.48	1.64	6.10	5.65	5.67	6.90	6.12	4.55	3.76	4.38	6.34	2.26	5.45	69.9
4–6	1.44	1.09	0.73	1.27	4.59	1.50	5.48	5.54	5.61	5.89	5.02	4.14	3.07	4.33	5.63	1.87	4.56	61.8
6–8	2.60	1.44	1.21	2.01	7.60	2.20	8.39	7.64	7.71	9.57	7.42	5.48	4.24	5.24	7.95	2.86	6.27	89.8

Table S11. The concentrations of 17 PAHs in the sediment core recovered from St. 10b in 2016 (ng g⁻¹ dw).

Depth (cm)	Nap	Acy	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BeP	BaP	IncdP	DahA	BghiP	Σ17PAH
0–2	3.51	2.44	1.10	2.98	23.6	5.69	30.8	25.1	19.0	21.1	33.1	11.5	15.7	18.0	30.5	4.56	18.1	267
2–4	3.20	1.88	1.58	2.55	17.2	2.79	19.5	16.8	11.8	14.2	26.1	10.1	12.3	12.5	23.0	3.66	13.8	193
4–6	3.32	1.93	1.32	2.59	16.8	4.03	22.2	19.5	13.8	14.7	24.0	9.93	12.1	14.5	20.8	3.48	12.4	198
6–8	2.95	1.80	1.39	2.46	15.1	2.60	17.5	15.1	11.2	13.0	24.4	6.36	10.2	12.1	16.1	2.80	9.50	164
8–10	3.45	2.18	1.85	2.80	19.6	3.68	29.7	24.6	20.1	20.2	29.6	13.1	14.1	18.6	25.0	4.19	15.1	248
10–12	3.12	2.04	1.32	2.60	16.6	3.01	19.7	17.3	12.8	14.8	28.8	9.98	13.2	15.2	25.5	4.06	14.8	205
12–14	3.75	2.38	1.67	3.02	19.7	3.49	30.3	22.9	17.5	18.4	33.3	12.8	15.5	16.9	33.4	4.68	19.0	259

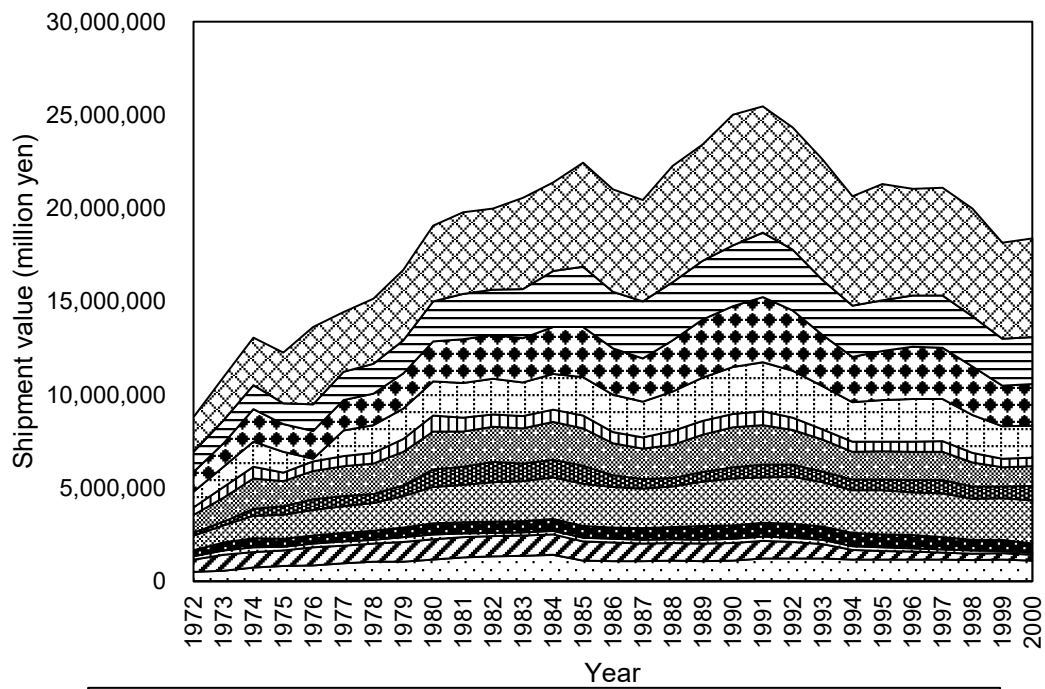


Fig. S1. Historical changes in shipment values of manufactured products in Osaka Prefecture.

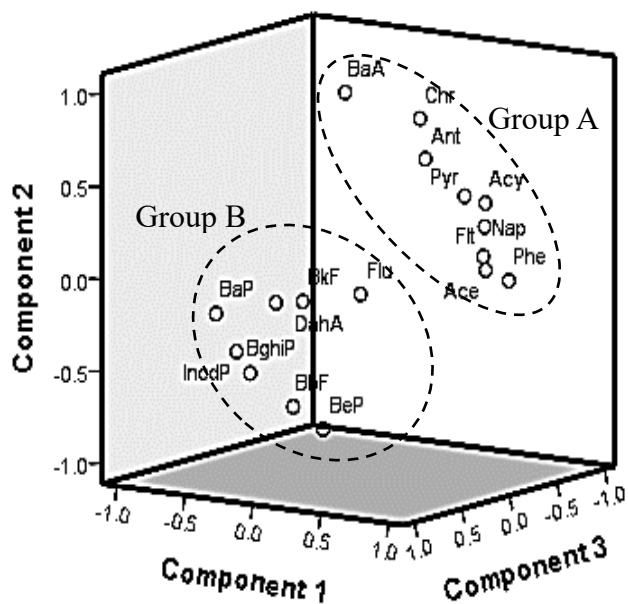


Fig. S2. Loading plot for the first three principal components of 17 PAHs from surface sediments and sediment cores of the Seto Inland Sea. Group A indicates the source is industry and biomass burning, and Group B indicates a vehicle source.