Supplementary material

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Reagents and Materials

Hexane, methylene chloride, acetone and methanol (all of they are pesticide residue analysis grade) ere obtained from Honeywell International (Morristown, NJ, USA). A stock mixture solution containing 2000 μg/mL of each nathphalene (Nap), 2-methylnaphthalene (2-MNap), 1-methylnaphthalene (1-MNap), biphenyl (Bp), 2,6-dimethylnaphthalene (DNap), acenaphthylene (Ace), acenaphthene (Ac), 2,3,5-trimethylnaphthalene (TNap), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), 2-methylphenanthrene (2-MPhe), 1-methylphenanthrene (1-MPhe), 2,6-dimethylphenanthrene (DMPhe), fluoranthene (Flu), pyrene (Pyr), 11H-benzo[b]fluorine (11-BbF), benzo[a]anthracene (BaA), chrysene, benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), perylene (Per) and 9,10-diphenylanthracene (DPh), and three PAHs including indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthrancene (DahA), and benzo[g,h,i]perylene (BghiP) was purchased from AccuStandard (New Haven, CT, USA). The stock mixture solution containing five deuterated PAHs (naphthalene-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, and perylene-d_{12} at 100 μg/mL each) used as surrogate standards and the stock mixture solution containing 2-fluoro-1,1-biphenyl, *p*-terbenyl-d_{14}, and dibenzo(a,h)anthracene-d_{14} at 100 μg/mL used as internal standards were supplied by Cambridge Isotope Laboratories (Andover, MA, USA). Neutral silica gel (80–100 mesh) and alumina (100–200 mesh) were obtained from Guangzhou Xinshi Chemistry Experimental Equipment (Guangzhou, China) and were pre-cleaned by Soxhlet extraction with methanol and methylene chloride, respectively, for 48 hours prior to use. Sodium sulfate from Sigma-Aldrich (St Louis, MO, USA) was baked at 450 °C for 4 hours and stored in sealed containers. All glasswares were cleaned with chromic acid and tap water followed by rinsing with deionized water, and then baked at 450 °C for 4 hours. The cleaned glasswares were covered with aluminum foil and stored at room temperature before use.

Comparison with other lakes

To better understand the state of pollution by PAHs in Chaohu Lake, a comparison of the results from the present study and those found in other lakes worldwide was conducted. Because of different target PAHs monitored in various studies, it would be very difficult to compare their levels individually for each PAHs. It would be reasonable to use the maximum, minimum and mean levels of total PAHs for the comparison instead of comparing them one by one so as to achieve a likely consistent comparison result (Table S3). By comparing the levels in Chaohu Lake to other lakes in China, it was found that the total PAHs in the present study were obviously higher than those found in Baiyangdian [1], East Lake [2], and lakes in Nanling Mountain [3], and Qinghai Lake, Bosten Lake, Erhai Lake, Chenghai Lake and Sugan Lake in western China region [4]. However, the levels in Chaohu Lake were close to that in Yuandan Lake and Taihu Lake [5, 6], but lower than in Nasi Lake [7] and Hongfeng Lake [8]. This result is not surprising because the regions in Nasi Lake and Hongfeng Lake have been well industrialized and urbanized. On the contrary, the industrialization and urbanization in the regions of Yuandan Lake and Taihu Lake are very close to the region in Chaohu Lake.

Comparing to other lakes worldwide, Table S3 shows that the levels of PAHs in Chaohu Lake were much higher than those found in those lakes in Asia [9, 10], South America [11, 12], Europe[13-15], North America [16-18], but were similar to those found in Erie Lake, Canada [19] and Higgins Lake, USA [16]. Interestingly, the levels in Chaohu Lake were lower than several lakes in North America [16, 20, 21], Africa [22, 23], and Europe [24].

Table S1. CAS number, molecular weight (g/mol), equilibrium sediment benchmark (µg/g OC) and RLs (ng/g)r the individual target PAHs

Compound	Abb[a]	CAS	MW[b]	ESB[c]	RLs [d]
naphthalene[e]	Nap	91–20–3	128	385	3.02
2-methylnaphthalene	2_Mnap	91–57–6	142	444	1.28
1-methylnaphthalene	1_Mnap	90–12–0	142	444	0.54
biphenyl	Bp	92–52–4	154	NA	0.52
2,6-dimethylnaphthalene	2,6_DMNap	581–42–0	156	513	0.55
acenaphthylene[e]	Ace	208–96–8	152	452	0.21
acenaphthene[e]	Ac	83–32–9	154	491	0.27
2,3,5-trinaphthalene	TMNap	2245–38–7	170	584	0.37
fluorene[e]	Fl	86–73–7	166	538	0.39
phenanthrene[e]	Phe	85–01–8	178	596	3.08
anthracene[e]	Ant	120–12–7	178	594	0.17
2-methylphenanthrene	2_Mphe	2531–84–2	192	669	1.12
1-methylphenathrene	1_Mphe	832–69–9	192	670	0.55
2,6-dimethlyphenanthrene	2,6_Dphe	17980–16–4	206	749	0.30
fluranthrene[e]	Flu	206–44–0	202	707	1.25
pyrene[e]	Pyr	129–00–0	202	697	0.53
11H-benzo[b]fluorene	11H_BbF	243–17–4	216	787	0.17
benzo(a)anthracene[ef]	BaA	56–55–3	228	841	0.17
chrysene[e]	Chr	218–01–9	228	844	0.17
benzo(b)fluoranthene[e,f]	BbF	205–99–2	252	979	0.18
benzo(k)fluoranthene[e,f]	BkF	207–08–9	252	981	0.17
benzo(e)pyrene	BeP	192–97–2	252	967	0.17
benzo(a)pyrene[e,f]	BaP	50–32–8	252	965	0.17
perylene	Per	198–55–0	252	967	0.30
9,10-diphenylanthracene	DPA	1499–10–1	330	1021	0.17
Diden(123-cd)pyrene[e,f]	IcdP	193–39–5	276	1115	0.34
dibenzo(a,h)anthracene[e,f]	DahA	53–70–3	278	1123	0.34
benzo(ghi)perylene[e,f]	BghiP	191–24–2	276	1094	0.34

[a], abbreviation; [b], molecular weight (g/mol); [c], equilibrium sediment benchmark (µg/g OC), the data was obtained from reference [25]; [d], report limits; [e] in the list identified as the priority pollutants by the United States Environmental Protection Agency; [f], the contaminant considered as carcinogens.

Table S2. The target compounds, their abbreviation used in the present study and individual concentration (ng/g)

Compound	N	Mean	Median	Min[a]	Max[b]	Percentiles (%)					
						5	10	25	75	90	95
Nap	61	24.6	14.2	2.55	239	3.20	4.32	8.24	25.5	52.7	96.2
2_Mnap	61	34.2	14.8	0.70	282	1.54	3.02	6.07	47.1	94.6	148
1_Mnap	61	12.6	6.57	0.35	93	0.76	1.43	2.74	17.1	34.1	44.9
Bp	61	11.7	4.79	0.99	117	1.85	2.50	3.11	9.19	31.9	61.6
2,6_DMNap	61	16.7	8.28	1.05	142	1.59	2.09	3.58	17.3	40.3	85.5
Ace	61	6.01	1.81	0.18	62	0.42	0.60	0.94	3.23	22.9	32.3
Ac	61	14.5	2.32	0.28	180	0.65	0.83	1.33	7.74	45.2	108
TMNap	61	4.02	2.04	0.24	25.1	0.34	0.48	0.76	5.05	13.8	17.7
Fl	61	27.6	10.0	1.41	221	2.61	3.37	5.08	18.5	105	162
Phe	61	61.3	26.1	4.69	438	7.22	8.26	14.7	46.8	194	335
Ant	61	22.7	3.61	0.30	251	0.64	0.84	2.02	9.31	86.3	170
2_Mphe	61	15.6	8.12	0.93	121	1.65	1.98	3.51	12.9	48.7	79.3
1_Mphe	61	8.54	4.12	0.04	65	0.89	1.06	2.02	7.27	28.0	43.9
2,6_Dphe	61	4.30	1.87	0.22	37	0.41	0.49	0.89	3.15	14.1	23.5
Flu	61	119	48.1	6.01	927	9.21	12.4	28.5	90.7	387	639
Pyr	61	93.4	36.1	3.95	748	5.93	8.80	18.8	74.0	311	517
11H_BbF	61	29.4	9.36	0.95	280	1.41	2.36	4.78	18.4	105	160
BaA	61	78.3	21.3	2.45	742	2.88	6.37	11.0	60.7	285	465
Chr	61	75.4	26.5	3.18	694	4.07	7.73	15.2	63.5	249	407
BbF	61	157	59.7	5.01	1460	8.72	13.6	28.6	110	534	864
BkF	61	56.9	16.9	1.48	594	2.55	4.19	8.02	36.9	202	341
BeP	61	109	40.0	3.39	1050	5.31	7.65	16.8	74.3	374	614
BaP	61	92.4	22.1	2.03	1010	3.11	4.83	10.1	59.5	370	587
Per	61	193	138	5.01	792	10.6	16.2	53.8	280	463	559
IcdP	61	74.7	29.4	2.58	464	4.18	7.57	15.0	52.3	295	421

DahA	61	60.4	23.7	1.92	464	3.16	5.43	11.8	45.3	221	318
BghiP	61	269	100.3	8.37	1680	14.8	25.3	52.7	183	1040	1540
$\sum_7$PAH	61	595	205	23.8	5420	30.4	50.7	91.3	461	2110	3320
$\sum_{16}$PAH	61	1230	451	60.8	10200	102	116	231	1000	4210	6520
$\sum_{28}$PAH	61	1670	738	82.4	12900	161	249	403	1570	5410	8190

Note: [a] min is the minma concentration; [b] max is the maximum concentration value.

Table S3. The total concentration of PAHs found in the sediment of lakes around the world (with unit of ng/g).

Name	Min	Max	Mean	Reference
China				
Sugan Lake (China)[a]			55	4
Bosten Lake (China)[a]			74	4
East Lake (China)[b]	83	324		2
Chenghai Lake (China)a			247	4
Qinghai Lake (China)a			276	4
Erhai Lake (China)[a]			475	4
Lake in Nanling (China)[b]	86	778	667	3
Yuandan Lake (China)[c]	983	1.4×10^3	1.2×10^3	26
Taihu (China)[d]	858	5.3×10^3	1.4×10^3	5
Chaohu Lake (China)	82.4	1.3×10^4	1.7×10^3	This study
Meiliang Bay of Taihu (China)[b]		4.8×10^3	2.6×10^3	6
Nansi (China)[b]	160	3.3×10^4	6.1×10^3	7
Lake Hongfeng (China)[b]	2.9×10^3	5.3×10^3		8
Globe				
Gratiot Lake (USA)[b]			<50	16
Mullet Lake (USA)[b]			<50	16
Kentucky Lake (USA)[e]	0.01	90.4		27
Lake Balaton (Hungary)[f]	11	1.7×10^3	132	13
Lake in the Mackenzie Delta (Canada)[b]	20	200		17
Ibirité Reservoir, MG (Brazil)[b]	104	181		12
Crystal Lake (Benzie, USA)[b]			200	16
Laja Lake (Chile)[g]	77	398	226	11
Elk Lake (USA)[b]			235	16
Lake Galletue (Chile)[g]	32	862	279	11
Remote lake from Svalbard (Norwegian)[h]	11	1.1×10^3	282	14
Bolgoda and Beira Lakes (Sri Lanka)[i]	152	569	329	9
Whitmore Lake (USA)[b]			490	16
Littlefield Lake (USA)[b]			665	16
Crystal Lake (Montcalm, USA)[b]			767	16
Gull Lake (USA)[b]			800	16
Laguna Lake (Philippines)[j]	510	2.0×10^3	968	28
Lake Athabasca (Canada)[k]	1.0×10^3	1.4×10^3		18
Chaohu Lake (China)	82.4	1.3×10^4	1.7×10^3	This study
Lake Erie (Canada)[l]	224	5.3×10^3	2.1×10^3	19
Higgins Lake (USA)[b]			2.4×10^3	16
Lake Victoria (Kenya)[b]	40	3.2×10^4		22
Lake Icalma (Chile)[g]	39	343	3.2×10^3	11
Mecoacán Lake (Mexico)[b]	120	3.6×10^4	6.4×10^3	20
Cadillac Lake (USA)[b]			6.6×10^3	16
Lake Maryut (Egypt)[m]	106	5.8×10^4	7.0×10^3	23
Cass Lake (USA)[b]			1.69×10^4	16
Lake Jämsännvesi (Finland)[b]	8.0×10^3	3.3×10^6		24
Urban lake in the northwest USA[r]	6.6×10^4	1.65×10^7	2.59×10^6	21

[a], Seven PAHs including B*a*A, Chr, BbF, BkF, BaP, DahA and IcdP;

[b], sixteen USEPA priority PAHs,

[c], twenty-five PAHs including Nap, 2-MNap, 1-MNap, Ace, Ac, Fl, Phe, Ant, 9-methyl Ant, 2-MPhe, 1-MPhe, Flu, Pyr, BaA,1-methyl Flu, 1-methyl Pyr, Chr, 1-methyl Chr, 1-methyl BaA, BbF, BkF, BaP, IcdP, dibenzo[1,2,5,6]perylene, and BghiP;

[d], thirty-five PAHs including NaP, 1-MNaP, 2-MNaP, 3-MNap, 4-MNaP, Ac, Ace, Flu, 1-methyl-Fl, 2-methyl-Fl, Phe, 1-MPhe, 2-MPhe, 3-MPhe, Ant, 1-methy-Ant, dibenzothiophene, 1-methyl dibenzothiophene, 2-methyl dibenzothiophene, Flu, 1-methyl Flu, 2-methyl Flu, Pyr, 1-methyl Pyr, BaA, 1-methyl BaA, Chr, BbF, BkF, BeP, BaP, Per, DahA, IcdP, BghiP;

[e], seven PAHs including Nap, Phe, Ant, BbF, BkF, BaP, BghiP;

[f], thirteen PAHs including Fl, Phe, Ant, Flu, Pyr, BaA, Chr, BbF, BkF, BaP, DahA, BghiP and IcdP;

[g], NaP, Phe, Ant, Flu, Ace, Fl, Pyr, BaA, Chr, BaP, DahA, BkF, BghiP, and IcdP;

[h], fifteen PAHs including 16 USEPA priority PAHs minus BaA;

[i], eleven PAHs including NaP, Ace, Phe, Ant, Flu, Fl, Pyr, Chr, BaP, BkF and DahA;

[j], four PAHs including Fl, Phe, Pyr and Per;

[k], nine PAHs including NaP, Ace, Flu, Phe, Pyr, Chr, BaP, BaA, and DahA;

[l,] sixteen PAHs including Ace, Fl, Phe, Ac, Flu, Pyr, BaA, Chr, BbF, BkF, BeP, BaP, Per, IcdP, DahA, and BghiP;

[m], thirty-nine PAHs including NaP, 1-MNaP, 2-MNaP, 3-MNaP, 4-MNaP, BP, Ace, Ac, Fl, 1-methyl Fl, 2-methyl Fl, 3-methyl Fl, Phe, Ant, 1-MPhe, 2-MPhe, 3-MPhe, 4-MPhe, dibenzothiophene, 1-methyl dibenzothiophene, 2-methyl dibenzothiophene, 3-methyl dibenzothiophene, Flu, Pye, 1-methyl Flu, BaA, Chr, 1-methyl Chr, 2-methyl Chr, 3-methyl Chr, 4-methyl Chr, BbF, BkF, BeP, BaP, Per, IcdP, DahA, and

BghiP.

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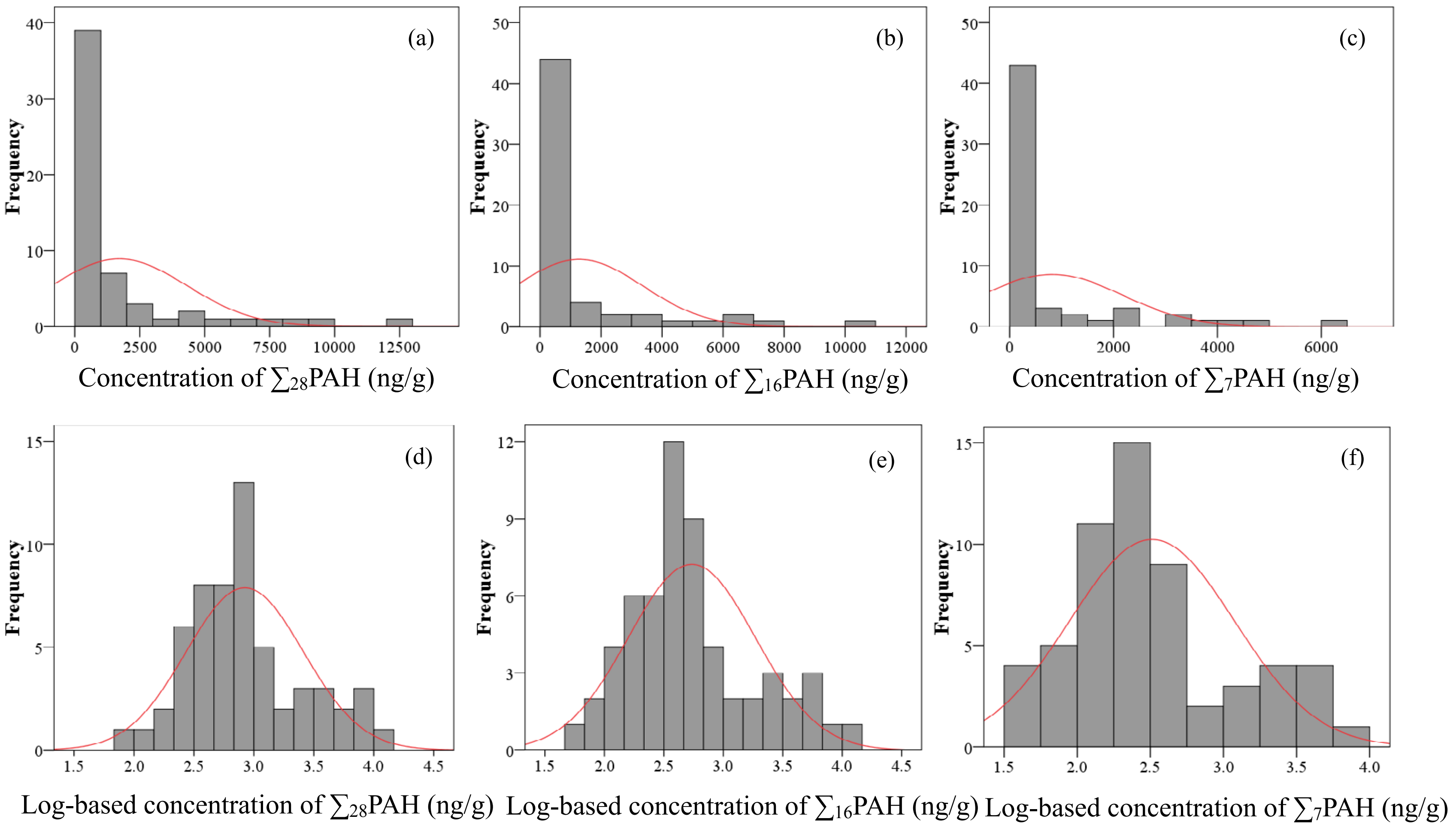

Fig. S1. The frequency of concentration of Σ_{28}PAH (ng/g) (a), Σ_{16}PAH (ng/g) (b) and Σ_{7}PAH (c); log-based concentration of Σ_{28}PAH (ng/g) (d), Σ_{16}PAH (ng/g) (e) and Σ_{7}PAH (f).

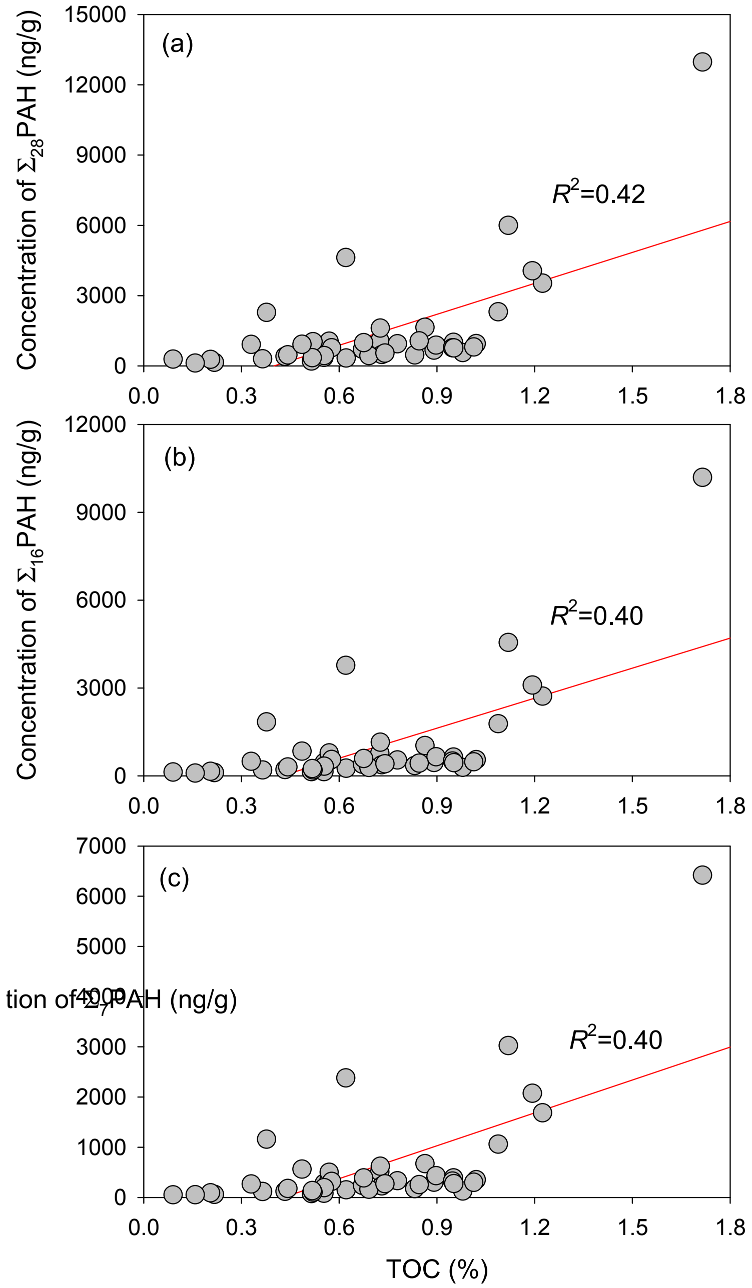

Fig. S2. The correlation relationship between the concentration of $\sum_{28}$PAH (a), $\sum_{16}$PAH (b) and $\sum_{7}$PAH with TOC

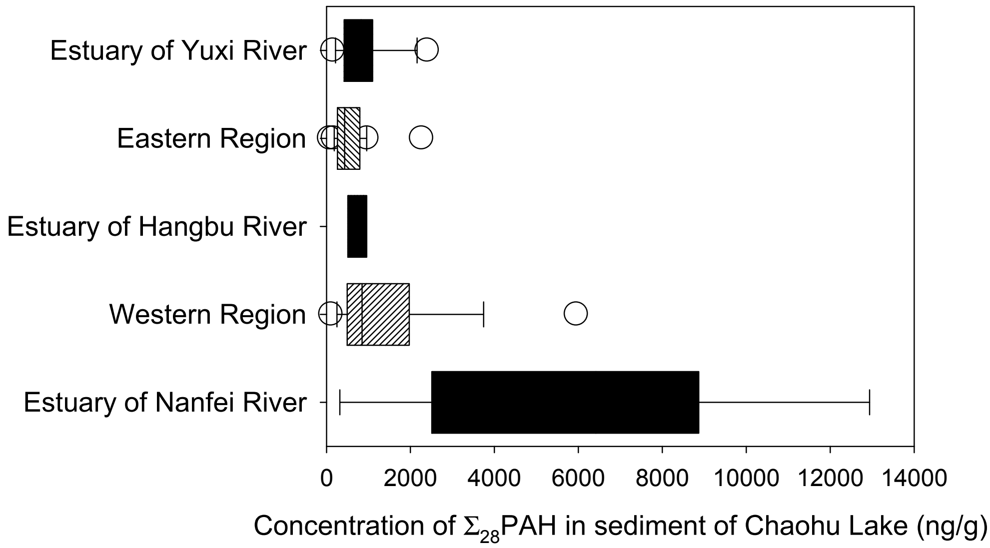

Fig. S3. The concentrations of Σ_{28}PAH in surface sediments from Western and Eastern region of Chaohu, Lake, China.

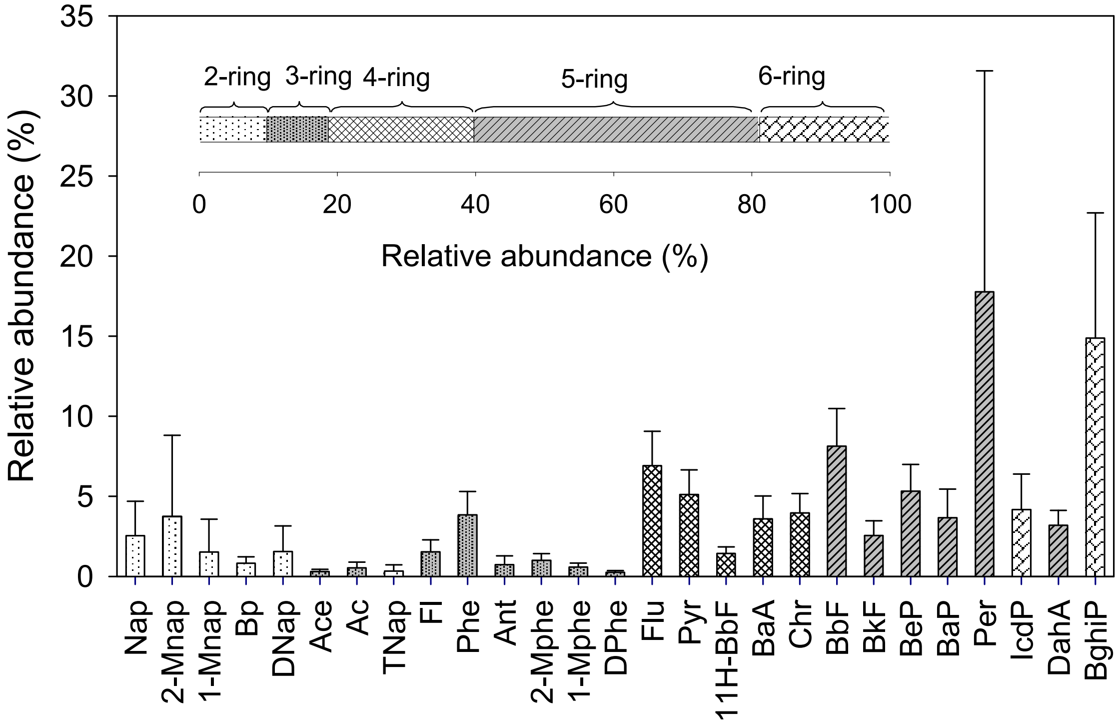

Fig. S4. Average relative percentages (%) of individual PAH in sediments of Chaohu Lake, China.

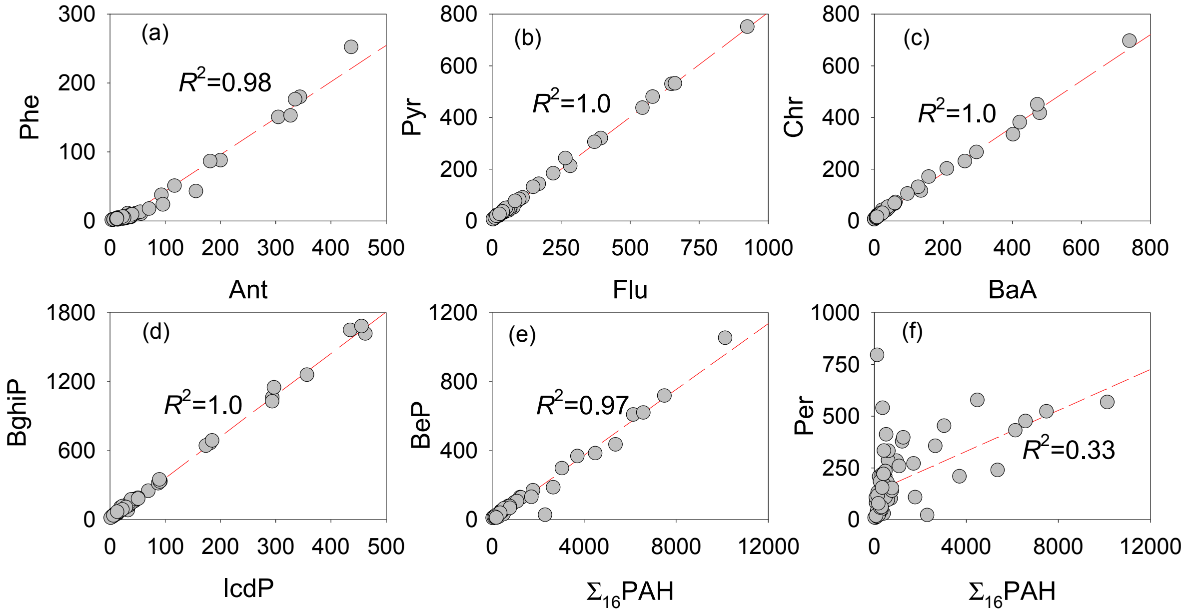

Fig. S5 Correlation relationships between concentration of anthrance (Ant) with phenthrene (Phe) (a), fluranthrene (Flu) with pyrene (Pyr) (b), benzo[a]anthrance (BaA) with chrysene (Chr) (c), indeno[1,2,3-cd]pyrene (IcdP) with benzo[g,h,i]perylene(BghiP) (d), Σ_{16}PAH with benzo[e]pyrene (BeP) (e) Σ_{16}PAH and with perylene (Per) (f).

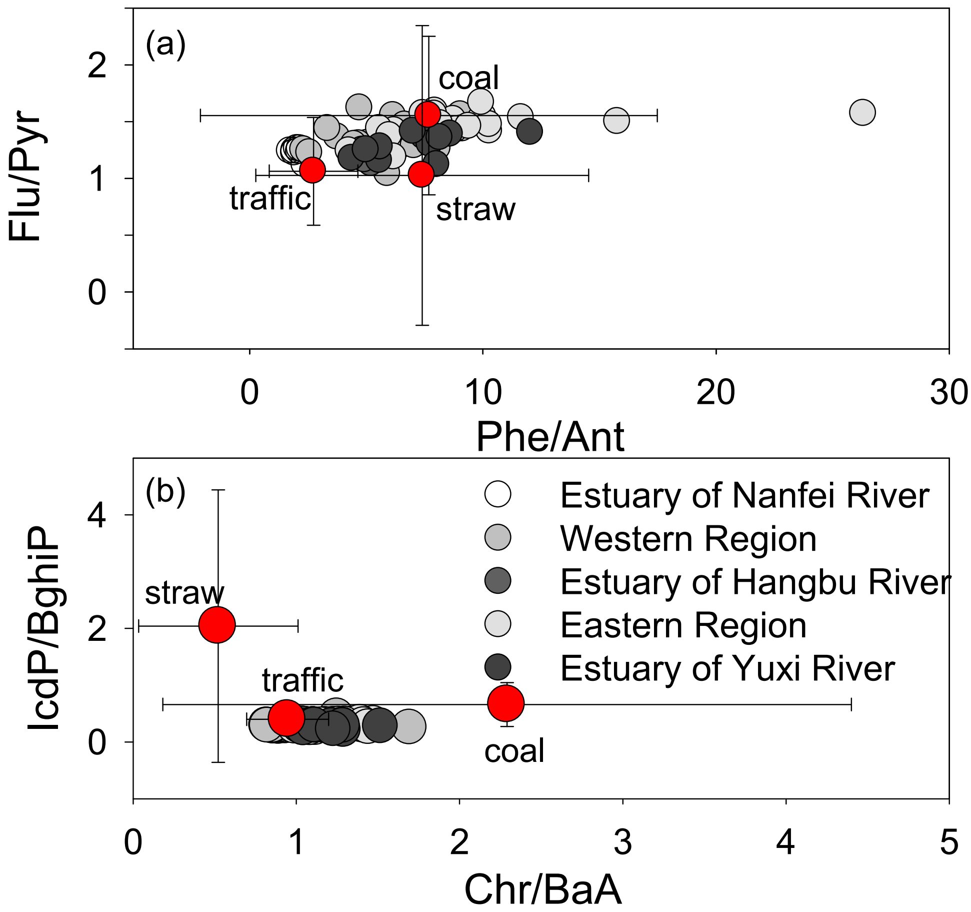

Fig. S6. The ratio of Phe/Ant (phenthrene /anthrance) vs the ratio of Flu/Pyr (flurenence/pyrenen) (a), and the ratio of Chr/BaA (chryene/benzo[a]anthrance) vs the ratio of IcdP/BghiP (indeno[1,2,3-cd]pyrene/benzo[g,h,i]pyrene). The ratios for straw burning, petroleum burning and coal combustion were obtained from references [29]; [30] and [31], respectively.