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Distribution of polycyclic aromatic hydrocarbons in sediments from Yellow River Estuary and Yangtze River Estuary, China

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Abstract

Surface sediment samples collected from twenty-one sites of Yellow River Estuary and Yangtze River Estuary were determined for sixteen priority polycyclic aromatic hydrocarbons (PAHs) by isotope dilution GC-MS method. The total PAH contents varied from 10.8 to 252 ng/g in Yellow River Estuary sediment, and from 84.6 to 620 ng/g in Yangtze River Estuary sediment. The mean total PAH content of Yangtze River Estuary was approximately twofold higher than that of Yellow River Estuary. The main reasons for the difference may be the rapid industrial development and high population along Yangtze River and high silt content of Yellow River Estuary. The evaluation of PAH sources suggested that PAHs in two estuaries sediments estuaries were derived primarily from combustion sources, but minor amounts of PAHs were derived from petroleum source in Yellow River Estuary. PAHs may be primary introduced to Yellow River Estuary via dry/wet deposition, wastewater effluents, and accidental oil spills, and Yangtze River Estuary is more prone to be affected by wastewater discharge.

Key words: PAHs; sediment; Yellow River Estuary; Yangtze River Estuary
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Introduction

Polycyclic aromatic hydrocarbons (PAHs), an important class of organic pollutants, are ubiquitous in the environment and known as carcinogens and mutagens. PAHs originated mainly from anthropogenic sources, such as combustion of fossil fuels and direct release of oil products (Simpson *et al*., 1996). Anthropogenic PAHs can be introduced to estuaries via urban runoff (McCready *et al*., 2000), industrial processes (Simpson *et al*., 1996), vehicle exhausts and spillage of fossil fuel (Pettersen *et al*., 1997; Wang *et al*., 1999) in the upper and middle reach areas. Due to their hydrophobic nature ($\log K_{ow}$, 3–8), once PAHs are introduced into the estuarine environment, they were rapidly adsorbed onto suspended particles and hence bottom sediments (Tam *et al*., 2001; Nogami *et al*., 2002). Estuarine sediment which contains large pools of organic matter can be a significant repository for PAHs (Pereira *et al*., 1999).

As two largest rivers in China, Yellow River and Yangtze River drain about 1.8×10^6 and 0.75×10^6 km^2 area, respectively, and totally cover about 25% of the territory of the country. Contaminants from the Yellow River Basin (Northern China) and Yangtze River Basin (Southern China) are ultimately transported by solid and water to estuaries. Yellow River Estuary and Yangtze River Estuary are major commercial arteries and industrial centers because of Shengli Oilfield (the second largest oil production base in China) and Shanghai City (one of the biggest metropolitan cities of China). Therefore, estuarine sediments of these two rivers can serve as field archives not only for ambient environmental conditions of estuary but also for the pollution status of PAHs in the upper and middle reach areas. The Yellow River and Yangtze River discharge 1.1×10^9 and 0.48×10^9 tons of fine-grained sediments annually to oceans, respectively, totally accounting for 10% of the world's annual sediment discharge (Milliman and Syvitski, 1992). This huge amount of sediment is one of the major factors affecting the balance of sedimentary and ecological environments in the Bohai Sea, Yellow Sea and East China Sea (Zhang, 1999). Although there were a few investigations on levels of PAHs in surface sediments from Yangtze River Estuary (Liu *et al*., 2001, 2008), little information about PAHs in surface sediments in Yellow River Estuary was reported.

The present study aimed to assess the levels of selected parent PAHs (sixteen priority pollutants) and examine the spatial distributions and potential sources in Yellow River Estuary and Yangtze River Estuary. The pollution status of PAHs in those two estuaries was also compared.

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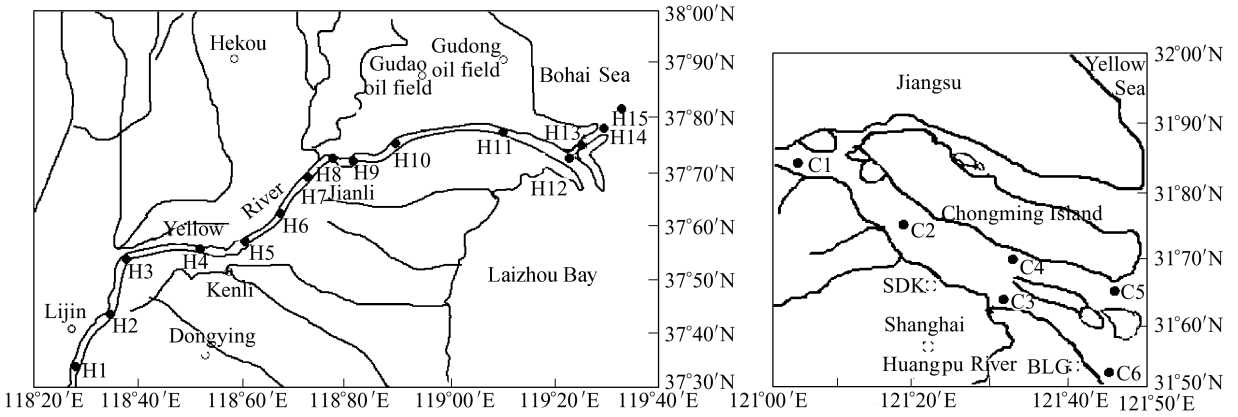

Fig. 1 Map of sampling sites. H1–H15: sediment sampling sites in Yellow River Estuary; C1–C6: sediment sampling sites in Yangtze River Estuary.

1 Materials and methods

1.1 Surface sediment samples

In Yangtze River Estuary, six sampling sites were selected adjacent to Shanghai City; and in Yellow River Estuary, fifteen sampling sites distributed in Dongying City were selected (Fig. 1). Surface sediment samples were collected using a stainless steel grab sampler in April 2004 in Yangtze River Estuary, and August 2004 in Yellow River Estuary. All sediment samples were stored frozen at –20°C until laboratory analysis.

1.2 Sample extraction and cleanup

The sample was freeze-dried and homogenized, and then 5 g subsamples were weighed for extraction. Surrogate standards (predeuterated PAHs) were added to the sample aliquot, and sample was Soxhlet-extracted for 24 h with 250 mL dichloromethane/hexane solvent (1:1, *V/V*). The extract was then concentrated to about 2 mL by rotary evaporator for the following cleanup. The concentrated extract was fractioned with a silica gel column (30 cm × 10 mm i.d.) packed with 10 g of silica gel, which was pre-eluted with 40 mL of hexane before loading sample. After sample was transferred into the column, it was eluted with 25 mL of hexane and followed by 50 mL 50% dichloromethane in hexane. The second fraction was collected and then concentrated to 0.2 mL by evaporation and the gentle nitrogen stream.

1.3 PAH analysis

The determination of PAHs was performed on an Agilent 6890 gas chromatography-5973 mass selective detector (GC-MS, Agilent, USA) system equipped with a fused silica capillary DB-5 column (30 m × 0.25 mm i.d., 0.25 μm film thickness, Agilent, USA). The oven temperature program was as follows: start 50°C held for 2 min, 50–200°C at 19.5°C/min held for 2 min, 200–240°C at 4.5°C/mim held for 2 min, 240–290°C at 2.5°C/min held for 5 min. An aliquot sample (1 μL) was injected in the splitless mode and helium was used as the carrier gas (1.0 mL/min). The mass spectrometer was operated in the selective ion monitoring (SIM) mode using positive ion electron impact ionization (EI). In this study, sixteen priority pollutants were analyzed. The identification of individual compound was achieved by comparing GC retention time and ion abundance ratio of two exacts (*m/z*) with the corresponding retention time of an authentic standard and the acquired ion-abundance ratio of the two exact (*m/z*). Quantification of individual compound was based on the comparison of peak areas with surrogate standards.

1.4 Quality control and quality assurance

Seven surrogate standards (naphthalene-d_8, acenaphthylene-d_8, phenanthrene-d_{10}, fluoranthene-d_{10}, pyrene-d_{10}, benzo(a)pyrene-d_{12} and benzo(g,h,i)perylene-d_{12}) were added to all samples to monitor matrix effects. The average recoveries of surrogate standards varied from 81.6% to 112%. In addition, the detection limit ranged from 0.15 to 0.7 ng/g dry weight (dw).

2 Results and discussion

2.1 PAH concentration and distribution

The total PAH contents in sediments of Yellow River Estuary ranged from 10.8 ng/g dw at site H7 to 252 ng/g dw at site H5 (Table 1), with a mean content of 90.7 ng/g dw. In order to determine PAH spatial distributions, fifteen samples in Yellow River Estuary were grouped into three segments averagely along the lower stretch of estuary: segment 1, segment 2, and segment 3. Every segment has five sampling sites. Segments were selected based on hydrographical information, river characteristics, discharge of water and silt content, especially socio-economy. The mean total PAH content in sediments is spatially distributed as segment 1 (146.8 ng/g), segment 2 (26.4 ng/g) and segment 3 (99 ng/g). Overall, the mean total PAH content is obviously higher in segment 1 and segment 3 than segment 2. There are big industrial bases (Dongying and Hekou) in south of segment 1 and north of segment 3 but a rural setting and large-scale farms along segment 2, therefore, there is the great potential for PAHs loading into segment 1 and segment 3 from urban runoff, municipal and industrial wastewater effluents, atmospheric deposition of industrial processes and vehicular emissions.

Table 1 Concentrations of PAHs in surface sediments from Yellow River Estuary (ng/g dw)

	Segment 1					Segment 2					Segment 3				
	H1	H2	H3	H4	H5	H6	H7	H8	H9	H10	H11	H12	H13	H14	H15
Naphthalene	< 0.2	< 0.2	35.9	11	17.2	< 0.3	< 0.3	<0.2	< 0.2	9.2	21.2	< 0.2	11.3	102	< 0.2
Acenaphthylene	0.2	1.1	2.1	1.4	1.8	< 0.2	< 0.1	0.3	< 0.1	< 0.1	1.6	0.7	1.5	2.3	0.5
Acenaphthene	<0.2	0.7	8.8	2.0	3.6	< 0.5	< 0.3	0.4	< 0.2	< 0.5	3.0	<0.3	0.7	2.1	0.9
Fluorene	2.5	2.8	11.7	5.3	11.2	4.2	< 0.2	1.9	4.0	3.3	8.7	3.2	3.2	20.9	6.6
Phenanthrene	6.2	9.2	37.2	21.5	34.7	9.9	< 0.1	7.5	11.3	8.7	27.8	11	11.7	10.7	6.4
Anthracene	0.5	1.5	10.4	5.3	6.3	1.2	0.3	0.9	0.7	1.0	6.5	0.7	2.0	0.5	0.3
Fluoranthene	1.7	16.8	19.6	20.5	28.0	3.9	1.9	3.2	2.7	6.4	24	7.3	8.3	4.6	1.7
Pyrene	1.8	12.4	21.6	21.1	28.2	2.9	2	2.9	2.6	4.4	25	6.5	9.2	4.4	2
Benzo(a)anthracene	0.5	6.7	8.2	13.3	12.5	0.9	0.9	1.1	1.3	1.2	0.5	3.3	4.3	2.1	0.7
Chrysene	0.8	18.6	9.9	16.0	15.1	1.9	1.4	1.7	1.7	1.9	1.2	6.0	5.9	3.7	1.1
Benzo(b)fluoranthene	0.7	11.8	6.1	9.9	23.6	2.0	1.5	1.4	1.2	1.1	13.9	4.5	4.0	3.1	0.8
Benzo(k)fluoranthene	0.2	4.6	2.7	3.5	8.9	0.5	0.5	0.5	0.4	<0.3	4.9	1.4	1.4	1.0	<0.4
Benzo(a)pyrene	0.2	3.8	7.2	8.7	16.4	0.8	0.6	0.3	0.8	< 0.5	12.5	2.2	1.8	1.4	<0.4
Ideno(1,2,3-cd)pyrene	0.4	8.5	5.2	9.5	18.2	0.9	0.7	1.1	0.7	< 0.5	16.1	3.3	3.0	1.9	0.5
Dibenzo(a,h)anthracene	<0.4	3.1	1.8	4.4	7.7	<0.6	<0.7	<0.5	<0.5	<0.4	7.0	< 0.7	1.3	< 0.6	< 0.4
Benzo(ghi)perylene	0.5	7.8	5.7	9.7	18.4	1.5	1.0	1.0	0.8	0.9	15.0	3.5	3.1	2.3	0.6
∑PAHs	16.2	109	194	163	252	30.6	10.8	24.2	28.2	38.1	189	53.6	72.7	163	22.1

∑PAHs: the sum of sixteen priority PAHs; LOD: limit of detection; assuming that all values of different congeners less than LOD are equal to 0.

Moreover, the mean total PAH content in segment 1 is obviously higher than that in segment 3, that likely owing to the rapid urbanization and industrialization in segment 1 region and high content of silt in segment 3 region. The total PAH content also shows a great difference in segment 1 and segment 3 (Table 1). Although segment 1 is the most seriously contaminated area, in site H1 the PAH content is significantly low (16.2 ng/g). The phenomenon may be due to that for site H1 located in the upper stream of Dongying, the level of PAHs affected by oil industry can be neglected. In segment 3, total PAH contents are significantly high at sites H11 (189 ng/g) and H14 (163 ng/g) than other sites and vary low at site H15 (22.1 ng/g). Site H11 is close to two oil fields (Gudao and Gudong) and site H14 is located at the end of the estuary, PAHs may enter waters through shipping activities, offshore oil production and transport as well as accidental oil spills. While site H15 is near the mouth of Yellow River and terrestrial inputs of PAHs is diminished. No significant difference in the total PAH content is found in segment 2.

The total PAH contents in sediments of Yangtze River Estuary ranged from 84.6 ng/g at site C2 to 620 ng/g at site C6 (Table 2), with a mean value of 259 ng/g. No significant difference was observed in sediments from the different sampling sites. However, levels of all compounds are slightly high in sites C1, C3 and C6. At site C1, the pollution from upstream and the high alluvial rate result in a slightly high total PAHs content. Moreover, because site C3 located at the lower stream of Shidongkou sewage outlets and at the influx of Huangpu tributary into the mainstream of Yangtze River, and site C6 is proximity to Bailonggang sewage outlets, the pollutants from sewage discharge of sewage outlets may result in a higher total PAH content.

Liu *et al*. (2001) analyzed intertidal flat surface sediment samples collected in August 2000 in the Yangtze River Estuary and discussed the distribution and sources of PAHs. For the purpose of studying the trend of PAHs in the environment, some of sampling sites studied by Liu *et al*. (2001) were also selected in the present study, in which the total PAH contents were from 556 to 6372 ng/g in sediments with a mean value of 2095 ng/g. A significant decrease of contamination was observed between samples collected during 2000 and 2004. There are two reasons for the descent: first, sedimentation has been declined since the construction of Three-Gorge Reservoir in the Yangtze River (Chen *et al*., 2008), which results in a less input of pollutants carried by suspended sediments from upper and middle reach areas. Second, many of thermal power generating plants with capacity below 50000 kW along Yangtze River had been closed since 1999 according to State Economic and Trade Commission.

Table 2 Content of PAHs in surface sediments from Yangtze River Estuary (ng/g dw)

	C1	C2	C3	C4	C5	C6
Naphthalene	18	33.3	28.9	22.5	19.4	43.5
Acenaphthylene	2.1	< 0.2	2.3	1.6	1.2	5.7
Acenaphthene	3.1	2.4	5.4	3.4	5.5	17.7
Fluorene	12.7	9.3	14.7	12.7	7.7	24.7
Phenanthrene	27.8	14.3	43	26.1	17.2	76.3
Anthracene	4.9	1.7	9.2	3.1	2.6	20.6
Fluoranthene	25.2	4.9	41.1	11.3	8.8	73.4
Pyrene	24.4	5.4	39.7	11.6	8.9	75.4
Benzo(a)anthracene	16.7	1.8	27.3	3.5	4.4	47.1
Chrysene	22.9	3.3	38.1	5.1	6.9	48.8
Benzo(b)fluoranthene	22.6	3.4	29.5	4.6	6.2	37.6
Benzo(k)fluoranthene	8.5	0.6	10.3	1.1	1.2	14.2
Benzo(a)pyrene	15.1	<0.5	22.3	8.2	4.5	40.5
Ideno(1,2,3-cd)pyrene	18.3	1.7	26.9	3.0	4.0	39.2
Dibenzo(a,h)anthracene	7.2	<0.6	12.4	<0.6	1.7	16.1
Benzo(ghi)perylene	16.9	2.5	24.5	4.8	5.1	39.4
∑PAHs	246	84.6	376	123	105	620

∑PAHs are the same meaning as that in Table 1.

The mean total PAHs content in Yangtze River Estuary was approximately twofold higher than that in Yellow River Estuary. PAHs are considered to be produced mainly from incomplete combustion of fossil fuels, and are good indicators of anthropogenic activities. Therefore, the high total PAH content in Yangtze River Estuary can be explained by that Shanghai City is a highly industrialized city and its GDP and population are evidently higher than

Table 3 Comparison of PAH concentrations in surface sediments of rivers

Location	Range (ng/g)	Mean ± S.D. (ng/g)	Reference
Beiluohe River, China	17.7– 407.7	106.8 ± 150.6	Zhang *et al*., 2007
Chao Phraya Estuary, Thailand	30–724	179 ± 222	Boonyatumanond *et al*., 2006
Daliao River watershed, China	61.9–840.5	287.3 ± 222	Guo *et al*., 2007
Gao-ping River, Taiwan, China	8–356	80.6 ± 31.8	Doong and Lin, 2004
Masan Bay, Korea	41.5–1100	353 ±252	Khim *et al*., 1999
Milwaukee Harbor Estuary, USA	200–390800	57360	Li *et al*., 1998
Pearl River Delta, China	156–10811	2057 ± 3063	Mai *et al*., 2002
San Francisco Bay, USA	2653–27680	7457 ± 6908	Pereira *et al*., 1996
Todos Santos Bay, BC, Mexico	7.6–813	96	Macias-Zamora *et al*., 2002
Tonghui River, Beijing, China	127–928	540 ± 292	Zhang *et al*., 2004
Whole Thai Coast, Thailand	9–228	50 ± 56	Boonyatumanond *et al*., 2006
Yangtze River Estuary, China	84.6–620	259.3 ± 190.1	This study
Yellow River Estuary, China	10.8–252	91.16 ± 77.6	This study

that of Dongying City. Another explanation is that Yellow River is the most turbid large river in the world (Zhao *et al*., 1998) and the biodegradation rates of PAHs increased with the sediment content in the water (Xia *et al*., 2006).

The levels of PAHs in Yellow River Estuary and Yangtze River Estuary are low to moderate in comparison with many other aquatic systems (Table 3). The total PAH content in surface sediments found in Yellow River Estuary are similar to the levels in Whole Thai Coast and Todos Santos Bay in Mexico. The levels of PAHs in Yangtze River Estuary are similar to that in Masan Bay, Korea. However, the levels of PAHs in both studied estuaries are substantially less than those in some highly urbanized and contaminated region such as Pearl River Delta, China and San Francisco Bay, USA. The total PAH contents in sediments of two studied estuaries are significantly lower than the Effects Range Low (ERL) level of 4000 ng/g dw, proposed as the Sediment Quality Guidelines (SQGs) to marine ecosystems by Long *et al*. (1995). Moreover, for individual PAHs, the contents do not exceed their respective ERL values. It may be concluded that the PAHs alone would not cause immediate biological effect in the sedimentary environment of Yellow River Estuary and Yangtze River Estuary.

2.2 PAH sources identification

PAH isomer ratios, such as anthracene/anthracene + phenanthrene (Ant/Ant+Phe), benzo(a)anthracene/ benzo(a)anthracene+chrysene (BaA/BaA+CHR), fluoranthene/fluoran thene+pyrene (Fl/Fl+Pyr) and ideno(1,2,3-cd)pyrene/ideno(1,2,3-cd)pyrene+benzo(ghi)pery-lene (IP/(IP+BghiP)), have been used as distinct chemical tracers to infer possible sources of PAHs (Budzinski *et al*., 1997; Tsapakis *et al*., 2003; Yunker *et al*., 2002; Zhang *et al*., 2004). Based on the PAH isomer ratio measurements compiled by Yunker *et al*. (2002), an Ant/Ant+Phe ratio of 0.1 is usually defined as the petroleum/combustion transition point. Fl/Fl+Pyr < 0.4 implies petroleum, 0.4–0.5 implies petroleum combustion, and > 0.5 implies combustion of coal and biomass. Ratio of BaA/BaA+CHR < 0.2 implies petroleum, 0.2–0.35 indicates either petroleum or combustion, and > 0.35 implies combustion. IP/IP+BghiP < 0.2 indicates petroleum, 0.2–0.5 petroleum combustion, > 0.5 combustion of coal or biomass.

The PAH isomer ratios shows that PAHs in Yellow River Estuary sediments are derived primarily from combustion sources and minor amounts of PAHs are derived from direct input of unburned fossil fuels (Figs. 2 and 3). The Ant/(Ant+Phe) and BaA/(BaA+CHR) isomer ratios show that PAHs are derived primarily from combustion (> 66% frequency) with lower occurrence of PAHs from petroleum (< 34% frequency) (Figs. 3a and 3b) and the frequency is greatly different among segments (Fig. 3a). In segment 1 and segment 2, PAHs may be greatly derived from combustion (80% frequency). But in segment 3, PAHs may be derived primarily from petroleum (60% frequency). That is, sediments exhibited more evidence of petroleum-derived pollution in segment 3. The status is likely owing to the influence of adjacent oil fields (Gudong and Gudao). The isomer ratios of Ant/(Ant+Phe) and BaA/(BaA+CHR) show that PAHs are mainly originated from combustion and the petroleum-derived pollution should not be ignored. Moreover, other two ratios of IP/(IP+BghiP) and Fl/(Fl+Pyr) used to identify biomass/coal and petroleum combustion showed that PAHs are derived primarily from petroleum combustion (80% and 53.3% frequency) with a lower occurrence of PAHs derived from biomass/coal combustion (20% and 46.7% frequency) (Figs. 3c and 3d) and the frequencies are greatly different among segments (Fig. 3d). In segment 1 and segment 3, PAHs may be greatly derived from petroleum combustion (80% and 60% frequency); and in segment 2, PAHs may be derived primarily from biomass and coal combustion (80% frequency). The combustion of crude oil (primarily segment 1 and segment 3) may be a major source of PAHs in Yellow River Estuary sediment, and rude oil may be also a source for PAHs because Shengli Oilfield is around Yellow River Estuary where crude oil refineries could contribute PAH from the refining process through air emission and wastewater effluents.

The PAH isomer ratios show that PAHs in Yangtze River Estuary sediments are also derived primarily from combustion sources (Fig. 2). However, compared with Yellow River Estuary, there are fewer amounts of PAHs that are possibly derived from direct input of unburned fossil fuels. The Ant/(Ant+Phe) and BaA/(BaA+CHR) ratios show that PAHs are derived primarily from combustion with significantly lower occurrence of PAHs from petroleum because of one or two spots at the source boundary (Fig. 2). In contrast, isomer ratios IP/(IP+BghiP) and Fl/(Fl+Pyr)

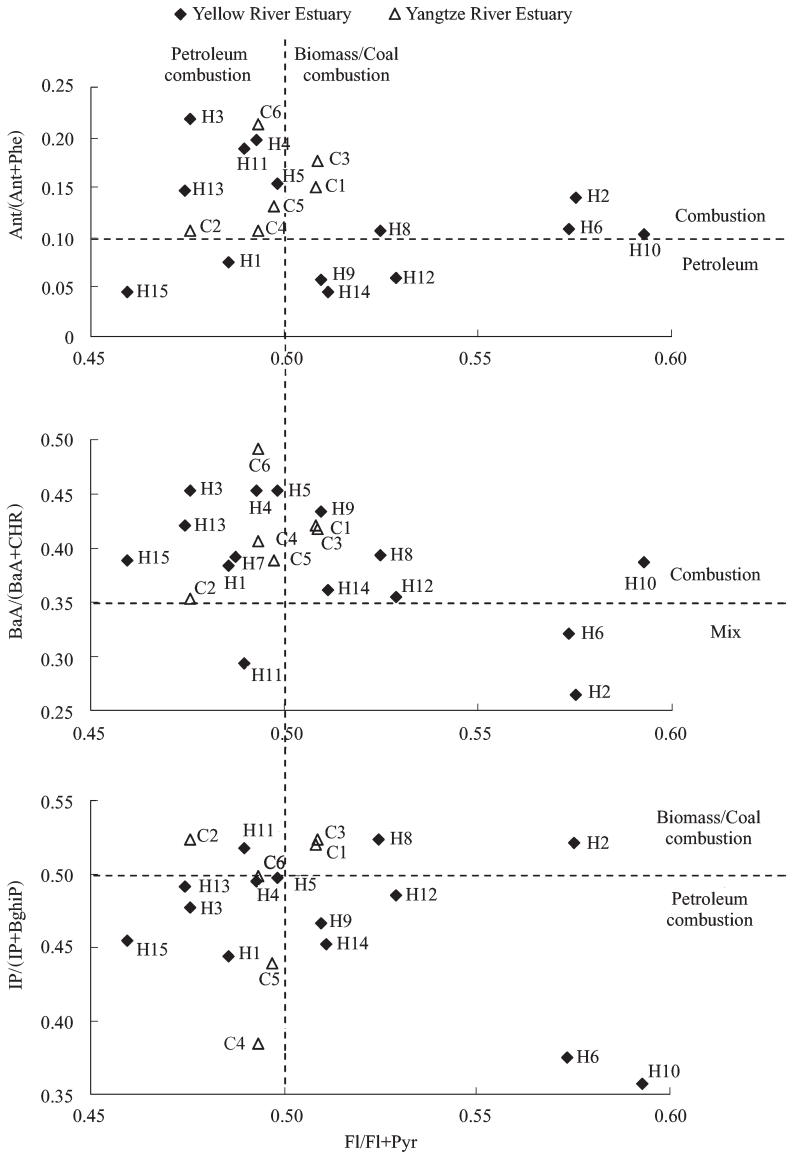

Fig. 2 Plots for the ratios of Ant/(Ant+Phe) vs. Fl/(Fl+Pyr), BaA/(BaA+CHR) vs. Fl/(Fl+Pyr), IP/(IP+BghiP) vs. Fl/(Fl+Pyr).

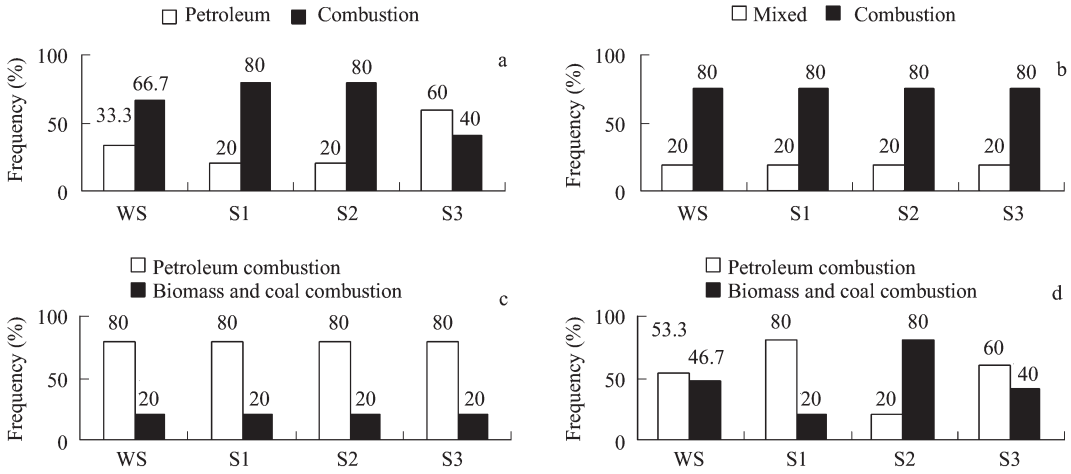

Fig. 3 Frequency of PAHs derived from various sources in Yellow River Estuary. (a) Ant/(Ant+Phe) ratio showing percentage of PAHs from petroleum and combustion sources; (b) BaA/(BaA+CHR) showing percentage of PAHs from mixed and combustion sources; (c) IP/(IP+BghiP) ratio in this figure only showing percentage of PAHs from petroleum combustion, and biomass and coal combustion sources; (d) Fl/(Fl+Pyr) ratio in this figure only showing percentage of PAHs from petroleum combustion, and biomass and coal combustion sources. S1: segment 1, S2: segment 2, S3: segment 3; WS: all samples.

in sediments are near transition point of two kinds of combustion sources (Fig. 2). Xu *et al*. (2006) reported that coke production and combustion of petroleum were two major PAH sources in Shanghai City and the former accounted for 68% of the total emission, while biomass burning could almost be ignored.

3 Conclusions

The concentration ranges of PAHs in sediments in Yellow River Estuary and Yangtze River Estuary were demonstrated. The concentrations of total PAHs in studied samples are similar to or substantially lower than those found in many other aquatic systems and significantly less than current sediment quality criteria (ERL). Diagnostic ratios analysis indicated that anthropogenic sources of PAHs in sediments are obvious. PAHs in two studied estuaries sediments are both derived primarily from combustion sources, and oil industry has a great influence on Yellow River Estuary than Yangtze River Estuary. The time trend analysis suggested that there is a significant decrease of PAHs contamination in Yangtze River Estuary in recent years.

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