

Distribution, source, and ecological risk assessment of polycyclic aromatic hydrocarbons (PAHs) in surface sediments from the Hun River, northeast China

Zhiyang Liu · Lixiong He · Yingzhuan Lu · Jiao Su · Han Song · Xiangying Zeng · Zhiqiang Yu

Received: 20 January 2015 / Accepted: 9 April 2015 / Published online: 24 April 2015
© Springer International Publishing Switzerland 2015

Abstract In this study, the occurrence and distribution of 16 polycyclic aromatic hydrocarbons (PAHs), listed by the United States Environmental Protection Agency (US EPA), were investigated in surface sediment samples from the Hun River, northeast China. The data was then used to assess the potential ecological risk. The results indicated 15 PAHs were detected in these sediments, and the total concentrations of the 15 PAHs (not including naphthalene) ranged from 82.96 to 39,292.95 ng g⁻¹ dry weight (dw), with an average value of 3705.54 ng g⁻¹ dw, and 4-ring PAHs were the dominant compounds at most sites. The diagnostic parameters such as anthracene/(anthracene+phenanthrene), fluoranthene/(fluoranthene+pyrene), and indeno[1,2,3-cd]pyrene/(indeno[1,2,3-cd]pyrene+benzo[g,h,i]perylene) showed that they had been emitted from a number of different sources, especially the pyrolytic emissions. The results of the ecological risk assessment, which compared the PAH concentrations with the effect range low (ERL) and the effect

range median (ERM) values, indicated that several individual PAH concentrations at four sites in the downstream section of the Hun River were higher than the ERM, suggesting that there was a potential ecological risk in these areas.

Keywords PAHs · Occurrence · Sediment · Ecological risk assessment

Introduction

Polycyclic aromatic hydrocarbons (PAHs), a group of persistent organic pollutants with two or more fused aromatic rings, are widely distributed in the environment. Most have been found to be mutagenic, carcinogenic, and teratogenic, and 16 PAHs have been listed as priority pollutants by the US EPA (Haritash and Kaushik 2009; Hiller et al. 2011). PAHs are generally introduced into the environment via anthropogenic activities, including incomplete combustion of organic matter, oil leakages, traffic-related emissions, sewage discharges, etc. (Essien et al. 2011; Luo et al. 2006; Mirza et al. 2014; Martins et al. 2010). Owing to their hydrophobic and persistent nature, PAHs can be strongly adsorbed to particles present in aquatic ecosystems and accumulate in sediments, which act as a long-term reservoir for PAHs (Patrolecco et al. 2010; Scheibye et al. 2014). These PAHs may pose a direct potential threat to benthic organisms and indirectly to aquatic organisms (Feng et al. 2012; Gaspare et al. 2009; Kannan et al. 2005).

Z. Liu · Y. Lu · J. Su · H. Song · X. Zeng · Z. Yu (✉)
State Key Laboratory of Organic Geochemistry, Guangdong
Key Laboratory of Environment and Resources, Guangzhou
Institute of Geochemistry, Chinese Academy of Sciences,
Guangzhou 510640, China
e-mail: zhiqiang@gig.ac.cn

Z. Liu · Y. Lu · J. Su · H. Song
University of Chinese Academy of Sciences, Beijing 100049,
China

L. He
Guangzhou Hexin Analytical Instrument Co., Ltd.,
Guangzhou 510530, China

The Hun River, located on the southwestern part of northeast China, belongs to the Daliao River water system, which drains across important, heavy industry regions in northeast China. They contain a variety of industries, such as machinery, metal refining, chemical and petrochemical industries, which discharge large amounts of pollutants into the Hun River via industrial wastewater. Municipal sewage discharge is also a major pollutant source (Guo et al. 2007).

Several studies have concentrated on PAH contamination in the rivers of the Daliao watershed (Guo et al. 2007; Zhang et al. 2013a). For example, Zhang et al. (2013a) investigated seasonal variations in PAH contamination of surface water from the Hun River and Lin et al. (2013) elucidated the source apportionment for PAHs in the Dahuofang Reservoir. However, limited information is available about PAH contamination and resultant ecological risk in surface sediments from upstream sections of the Hun River and Dahuofang Reservoir and downstream sections of the Hun River. This study aimed to (1) investigate the occurrence and distribution of PAHs in sediments from the abovementioned areas, (2) identify their possible sources, and (3) to assess the potential ecological risk posed by PAHs to aquatic organisms.

Materials and methods

Sample collection

The Hun River, which has its source on Gunma Mountain in Qinyuan County, is about 415 km long and flows through several cities, such as Fushun and Shenyang, and has a catchment area of 11,500 km² (Zhang et al. 2013a). The Pu and Xi Rivers are two of the seven main tributaries flowing into Hun River. Dahuofang Reservoir, situated on the upstream section of the Hun River near Fushun City, is the largest man-made lake in Liaoning Province and has a water volume of 21.87 billion m³. It acts as primary source of drinking water for inhabitants in surrounding cities, such as Fushun, Shenyang, Liaoyang, and Anshan.

A total of 40 surface sediment samples were collected from the Hun River in July 2010. These consisted of 12 from the upstream section of the Hun River (H1–12), 7 from the downstream part of the Hun River (H13–19), 16 from Dahuofang Reservoir (D1–16), and 5 from the abovementioned tributaries (P1–2 and X1–3). The

sampling sites are shown in Fig. 1. Each surface sediment sample (0–20 cm) was collected using a pre-cleaned stainless steel grab sampler, packed into an aluminum container, and kept in a freezer at –20 °C. All samples were freeze-dried, ground, and homogenized after the removal of pebbles and sticks. After screening through an 80-mesh (0.2 mm) sieve, all the samples were stored in pre-cleaned brown bottles at –20 °C until analysis.

Chemicals and materials

The 16 mixed PAH standards specified in EPA methods 610, which are naphthalene (NaP), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DBA), indeno[1,2,3-cd]pyrene (InP), and benzo[g,h,i]perylene (BgP), together with the deuterated surrogate standards containing naphthalene-d₈, acenaphthene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, and perylene-d₁₂, were obtained from Supelco (Bellefonte, PA, USA). The internal standard, hexamethylbenzene (HMB), was purchased from Dr. Ehrenstorfer-Schäfer Bgm-Schlosser (Augsburg, Germany). All solvents and reagents used were of pesticide grade. Silica gel (80–100 mesh) and neutral alumina (100–200 mesh) were Soxhlet-extracted with dichloromethane (DCM) for 72 h, activated at 180 and 250 °C for 12 h, respectively, and then deactivated with 3 % re-distilled water and kept in *n*-hexane before use. Anhydrous sodium sulfate was baked at 450 °C for 4 h before use.

Sample cleanup and analysis

The method used for extraction and cleanup has been published previously (Zeng et al. 2010). Briefly, approximately 20 g of sediment samples were spiked with deuterated PAHs as surrogate standards and then Soxhlet-extracted with 200 mL DCM for 72 h. Activated copper was added to remove elemental sulfur. The extracts were concentrated and solvent-exchanged into *n*-hexane and then concentrated to approximately 1 mL with a rotary evaporator. Following this, the extracts were loaded on a combined column of neutral alumina/silica gel (1:2) and eluted with *n*-hexane and a

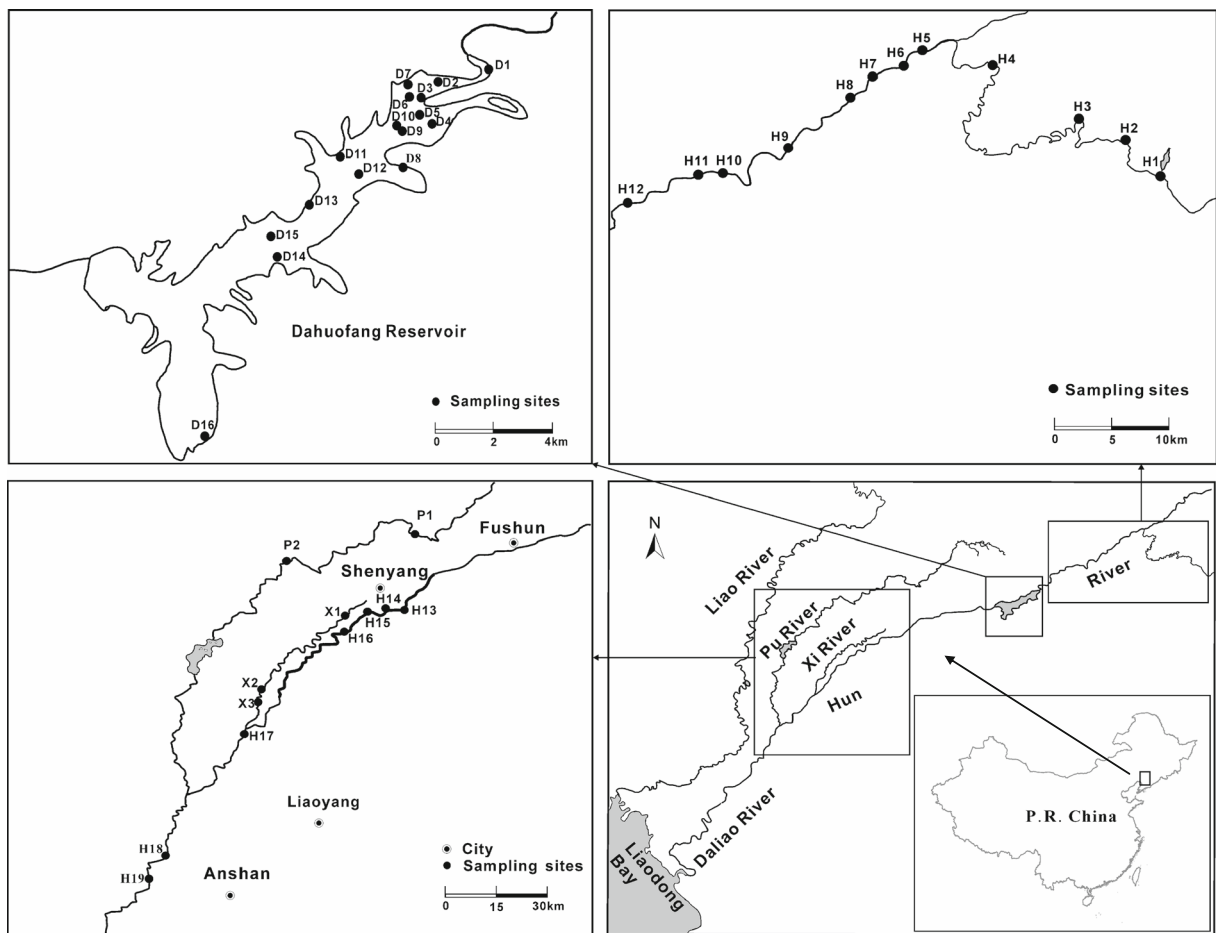


Fig. 1 Sampling location

mixture of *n*-hexane and DCM (3:1, v/v). The fraction containing the PAHs was concentrated to 1 mL, solvent-exchanged into *n*-hexane and further concentrated to 200 μL under a gentle N₂ stream. A known quantity of internal standard (HMB) was added before instrumental analysis.

Instrumental analysis

PAHs analysis was performed using a Shimadzu QP-2010 gas chromatograph equipped with a mass spectrometer in electron impact (EI) and selected ion monitoring (SIM) mode. A DB-5MS silica fused capillary column (30 m×0.25 mm i.d.×0.25 μm film) (Agilent Technology) was used. The oven temperature program was initially isothermally set at 70 °C for 2 min and then increased at a rate of 3 °C min⁻¹ to 295 °C, where it was held for 5 min. The temperatures of the injection port, the interface line, and the ion source were maintained at

290, 300, and 220 °C, respectively. A 1-μL sample was injected manually in splitless mode, and helium was used as the carrier gas at a flow of 1 mL min⁻¹.

Target analytes were identified and verified by comparing the retention times and mass spectra of the samples with the standards and were quantified using the internal standard method based on seven-point calibration curves ranging from 50 to 5000 μg L⁻¹ for individual compounds. The correlation coefficients (*R*²) were between 0.997 and 0.999, which was good. The detection limits for PAHs were 0.091–0.36 ng g⁻¹ for 20 g of sediment.

Quality assurance/quality control

Procedural blanks (solvent, *n*=4), spiked blanks (spiked standards into solvent, *n*=4), spiked matrixes (spiked standards into pre-extracted sediment, *n*=4), and duplicate samples were analyzed in each batch of samples.

The instruments were calibrated daily, and the relative difference between the seven-point calibration and the daily calibration was found to be <15 %. The surrogate recoveries were 67 ± 8 % for acenaphthene- d_{10} , 92 ± 10 % for phenanthrene- d_{10} , 94 ± 13 % for chrysene- d_{12} , and 83 ± 12 % for perylene- d_{12} . The relative standard deviations (RSD) of the duplicate samples were <15 %. The procedural blanks contained no detectable amounts of the target analytes. The Nap concentration was not included in this study due to its relatively low recovery in our method.

Results and discussion

PAH levels in the sediments

The occurrence and distribution of 15 EPA-listed PAHs (excluding Nap) in the sediments are shown in Table 1. The upstream section of the Hun River drains through an extensive agricultural area. Low levels of the 15 PAHs (Σ PAHs), ranging from 82.96 to 1268.73 ng g^{-1}

dw (with an average value at 436.11 ng g^{-1} dw), were found in the sediments. The highest concentration was found at H11, a sampling site near the inlet to the Dahuofang Reservoir. This showed that there were large PAH inputs from the upstream section of the Hun River into the Dahuofang Reservoir. Comparable Σ PAH concentrations, ranging from 292.93 to 778.14 ng g^{-1} dw (with an average of 451.38 ng g^{-1} dw), were detected in the sediments taken from the Dahuofang Reservoir, which is an important water-storage reservoir located in Liaoning Province. It provides industrial and agricultural water for the drainage basin and serves as an important drinking water for residents in the surrounding cities, such as Fushun City, Shenyang City, and Anshan City. Therefore, much more attention should be given to PAH pollution in Dahuofang Reservoir. In addition, much higher levels of Σ PAHs were detected (863.87–39,292.95 ng g^{-1} dw, with an average value of 11,313.84 ng g^{-1} dw) in the sediments from the downstream section of the Hun River. Very high PAH concentrations were found at H14 (39,292.92 ng g^{-1}) and X1 (34,023.58 ng g^{-1}). X1 was located in the Xi River,

Table 1 Concentrations of PAHs (ng g^{-1} dw) in the sediments from the Hun River and Dahuofang Reservoir and the sediment quality guidelines

Analyte	Upstream section of the Hun River		Dahuofang Reservoir		Downstream section of the Hun River		Guidelines	
	Mean	Range	Mean	Range	Mean	Range	ERL	ERM
Acy	3.27	0.28–15.47	2.28	0.81–3.87	75.87	2.61–337.72	44	640
Ace	1.96	LOD–7.54	2.57	0.29–4.66	226.92	4.60–1831.39	16	500
Fl	10.14	0.78–34.82	14.57	2.88–23.72	363.60	20.45–2255.45	19	540
Phe	84.14	9.88–275.04	76.81	38.63–141.97	1786.09	90.38–7192.33	240	1500
Ant	10.08	1.49–30.23	7.86	3.51–13.64	341.88	16.18–1434.62	863	1100
Flu	91.58	15.25–290.95	87.66	43.86–171.92	1949.63	196.08–7176.85	600	5100
Pyr	76.21	12.68–230.88	62.33	33.13–122.68	1748.47	183.74–5948.62	665	2600
BaA	21.22	5.54–53.16	21.08	14.18–30.01	802.21	37.66–2887.27	261	1600
Chr	30.61	6.33–106.03	38.88	27.91–55.67	786.14	46.02–2553.60	384	2800
BbF	47.88	9.20–163.90	53.39	35.23–118.86	1123.33	51.33–3903.15	–	–
BkF	10.55	2.74–21.82	14.89	10.96–18.89	299.66	16.42–1131.58	–	–
BaP	18.11	4.92–42.96	21.86	9.37–30.62	776.46	30.48–3109.80	430	1600
DBA	3.18	0.91–8.85	4.72	3.34–6.28	118.45	4.84–386.10	64.3	260
InP	13.33	4.02–36.59	20.30	15.98–24.32	445.55	19.12–1924.33	–	–
BgP	13.86	4.05–37.60	22.19	15.30–27.59	469.60	16.71–1814.15	–	–
Σ PAHs	436.11	82.96–1268.73	451.38	292.93–778.14	11,313.84	863.87–39,292.95	4000	44,792

Long et al. (1995)

LOD limit of detection, ERL the effects range low, ERM the effects range median

one of the important tributaries of the Hun River. It receives industrial effluents from Tiexi Economic Development Area, which is the most important industrial base in Shenyang. H14 was located in the section of the Hun River running through Shenyang City. The high concentrations suggested that these industrial wastewaters, as well as domestic sewage from the dense population, made important contributions to PAH levels in the sediments. Total organic carbon (TOC) is also an important factor that controls PAH levels in the sediments (Luo et al. 2006). A positive correlation ($R=0.631$, $P<0.01$) existed between the total PAH concentrations and TOC (Fig. 2), which indicated that TOC played an important role in controlling the distribution of PAHs in the Hun River sediments. Similar results have been observed in the sediments from other areas (Luo et al. 2006; Liu et al. 2013; Wang et al. 2015).

Baumard et al. (1998) proposed that PAH contamination levels could be classified into four groups: low ($0\text{--}100\text{ ng g}^{-1}$), moderate ($100\text{--}1000\text{ ng g}^{-1}$), high ($1000\text{--}5000\text{ ng g}^{-1}$), and very high ($>5000\text{ ng g}^{-1}$). If this classification is used, then the sediments from the Dahuofang Reservoir were slightly polluted; most sample sites in the upstream section were severely contaminated, and sediments from the downstream section of the Hun River were very significantly polluted with PAHs.

Many studies have investigated PAHs in the sediments worldwide. Some of the results are listed in Table 2. The table shows that the levels of ΣPAHs in the sediments from the upstream section of the Hun River and the Dahuofang Reservoir in this study were comparable to those detected in most of the major rivers in China, such as the Pearl River Estuary (Luo et al.

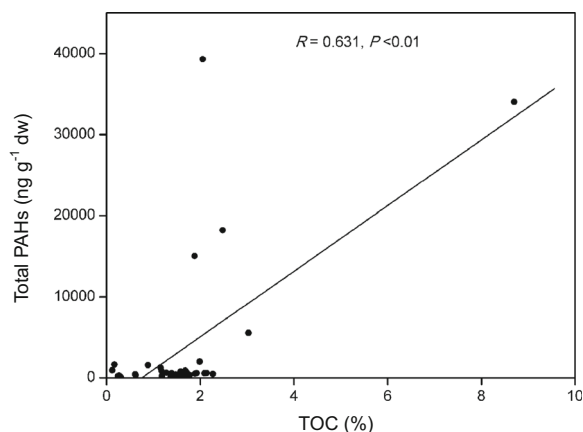


Fig. 2 Correlation plot of the total PAHs vs. TOC

2006), Qiantang River (Zhu et al. 2008), Huaihe River (Feng et al. 2012), Xiangjiang River (Zhang et al. 2013b), and Lijiang River (Xue et al. 2013). They were also comparable to reported results for other rivers and reservoirs around the world, such as Zemplinska sirava Reservoir in Slovakia (Hiller et al. 2011) and Ibirité Reservoir in Brazil (Mozeto et al. 2014) but were significantly lower than those found in contaminated and industrialized regions, such as the Wuhan section of the Yangtze River (Feng et al. 2007), Haihe River (Jiang et al. 2007), Liangtan River (Liu et al. 2013), Beijiing River (Song et al. 2014), Iko River estuary in Nigeria (Essien et al. 2011), Ruzin Reservoir and Velke kozmalovce Reservoir in Slovakia (Hiller et al. 2011), Ammer River in Germany (Liu et al. 2013), and Huveaune River in France (Kanzari et al. 2014). In contrast, the PAH concentrations in the downstream section of the Hun River were higher than for most of the abovementioned sites (Table 2).

PAH composition in the sediments

The 15 PAHs reported in this study were divided into four groups according to the numbers of rings: 3-rings (Acy, Ace, Fl, and Phe), 4-rings (Flu, Pyr, BaA, and Chr), 5-rings (BbF, BkF, BaP, and DBA), and 6-rings (InP and BgP). The distribution pattern by ring size for the PAHs is shown in Fig. 3. Generally, the high molecular weight (HMW) PAHs, i.e., 4- to 6-ring PAHs, predominated in all the sediment samples and ranged from 58.2 to 88.1 % of the total PAHs. This may be because the HMW PAHs are more hydrophobic than low molecular weight (LMW) ones, which means that they preferentially accumulate in the sediments, and secondly, the HMW PAHs are more resistant to degradation than LMW PAHs in the sediments and soil (Haritash and Kaushik 2009).

In the upstream section of the Hun River, the 4-ring PAHs were the most abundant components, ranging from 43.3 to 56.5 % of the total PAHs. A similar distribution pattern was found in the Dahuofang Reservoir where the 4-ring PAHs accounted for 40.7–51.7 % of the total PAHs. Most of the sediments from the downstream section of the Hun River were also dominated by 4-ring PAHs, making up 41.2–60.3 % of the total PAHs. At sampling site H17, the 3-ring PAHs accounted for 41.8 % of the total 15 PAHs; among which, Phe was the most abundant compound (accounting for 36.4 %). The differences in PAH composition patterns were

Table 2 Concentrations of Σ PAHs in the sediments from a number of different locations

Location	Number of PAHs	Σ PAHs (ng g ⁻¹ dw)		References
		Mean	Range	
Pearl River Estuary, China	16	189–637	362	Luo et al. (2006)
Yangtze River (Wuhan), China	16	72.4–3995.2	1334.5	Feng et al. (2007)
Haihe River, China	16	774.81–255,371.91	29,256.37	Jiang et al. (2007)
Qiantang River, China	16	91.3–1835.2	326.6	Zhu et al. (2008)
The upstream of Huaihe River, China	16	85.7–935.2	407.0	Feng et al. (2012)
Liangtan River, China	16	69–6250	2040	Liu et al. (2013)
Lijiang River, China	16	160–602	337	Xue et al. (2013)
Xiangjiang River, China	16	190–983	452	Zhang et al. (2013b)
Beijiang River, China	16	44–8921	1132.3	Song et al. (2014)
Iko River Estuary, Nigeria	16	610–35,270	16,340	Essien et al. (2011)
Ruzin Reservoir, Slovakia	15	2697–7561	4719	Hiller et al. (2011)
Velke kozmalovce Reservoir, Slovakia	15	7910–29,538	19,034	Hiller et al. (2011)
Zemplinska sirava Reservoir, Slovakia	15	84–631	434	Hiller et al. (2011)
Ammer River, Germany	16	112–22,900	8770	Liu et al. (2013)
Ibirité Reservoir, Brazil	16	79.8–219.9	129.5	Mozeto et al. (2014)
Huveaune River, France	16	572–4235	1966	Kanzari et al. (2014)
The upstream section of the Hun River	15	82.96–1268.73	436.11	This study
The downstream section of the Hun River	15	863.87–39,292.95	11,313.84	This study
Dahuofang Reservoir	15	292.93–778.14	451.38	This study

probably due to different PAH inputs. H17 was adjacent to an oil field in Liaoning Province. During the sampling period, oil exploration activity was still ongoing, so it was probably the main source of PAHs in the nearby sediments because PAHs derived from petroleum and its products are dominated by LMW PAHs (Hiller et al. 2011; Keshavarzifard et al. 2014; Scheibye et al. 2014).

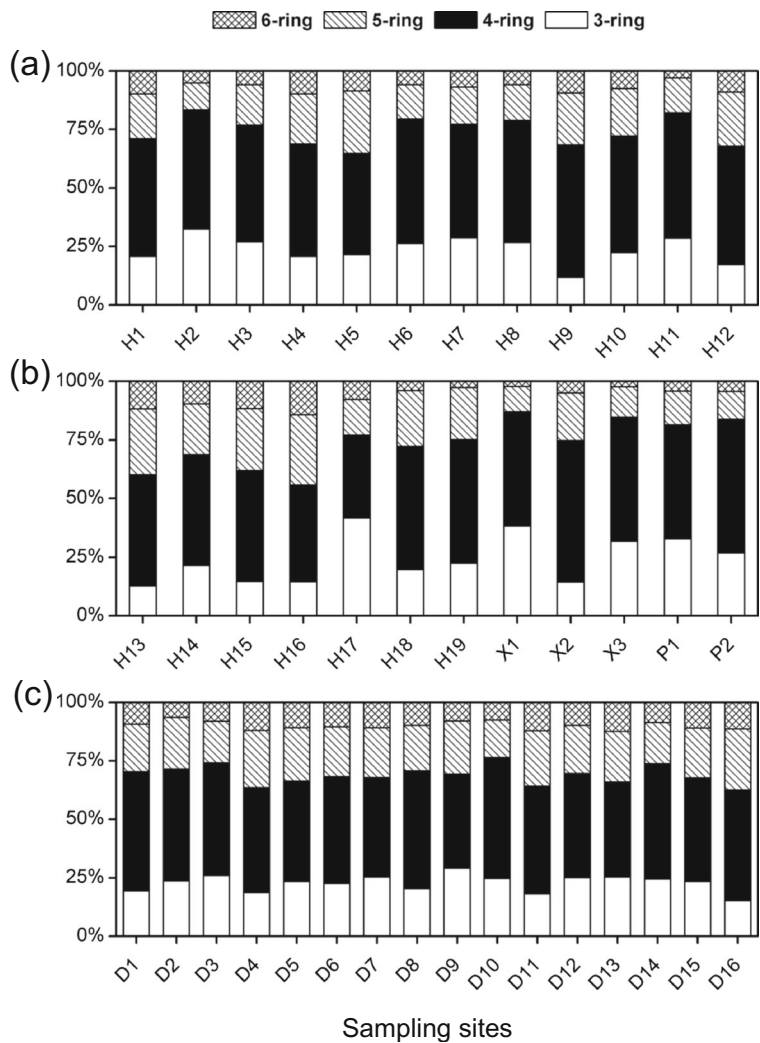
PAH sources diagnosis

According to the process by which PAHs are formed, PAH sources can be classified into two main categories: petrogenic source (mainly originating from petroleum) and pyrolytic source (mainly originating from incomplete combustion of petroleum and biomass). Several diagnostic parameters are usually used as distinct tracers to identify possible sources of PAHs, such as Ant/(Ant+Phe), Flu/(Flu+Pyr), and InP/(InP+BgP) (Tobiszewski and Namiesnik 2012; Yunker et al. 2002).

In this study, Ant/(Ant+Phe) and InP/(InP+BgP) were plotted against Flu/(Flu+Pyr) to trace the possible

sources of PAHs in sediment samples from the Hun River (Fig. 4). The Ant/(Ant+Phe) ratio of 0.1 is defined as the petroleum/combustion transition point, where below 0.1 is characteristic of petroleum contamination, and above 0.1 indicates a combustion source (Yunker et al. 2002). In this study, Ant/(Ant+Phe) ratios in the sediments from the upstream section of the Hun River, the Dahuofang Reservoir, and the downstream section of the Hun River ranged from 0.09 to 0.20, 0.07 to 0.12, and 0.03 to 0.24, respectively. The results suggested that the PAHs in the sediments were the results of a mixture of petroleum contamination and pyrolytic sources, and pyrolytic sources were dominant in the upstream and downstream section of the Hun River. A Flu/(Flu+Pyr) ratio <0.4 suggests petroleum origins, >0.5 implies biomass combustion (grass, wood, or coal), and between 0.4 and 0.5 is associated with petroleum combustion. A InP/(InP+BgP) ratio <0.2 is characteristic of petroleum pollution, >0.5 indicates biomass combustion, and between 0.2 and 0.5 corresponds to petroleum combustion (Yunker et al. 2002). All InP/(InP+BgP) and Flu/(Flu+

Fig. 3 The composition of PAHs: **a** the upstream section of the Hun River, **b** the downstream section of the Hun River, and **c** Dahuofang Reservoir



Pyr) ratios for the sediments in this study were higher than 0.4, which suggested that they were pyrolytic in origin. The predominance of pyrolytic sources have been previously noted by Lin et al. (2013), who concluded that coal combustion, residential emissions, wood burning, and traffic-related emissions were the main sources of combustion-derived PAHs in Dahuofang Reservoir. In particular, H17 had a very low Ant/(Ant+Phe) ratio at 0.03, which suggested that the primary source was a petroleum spillage.

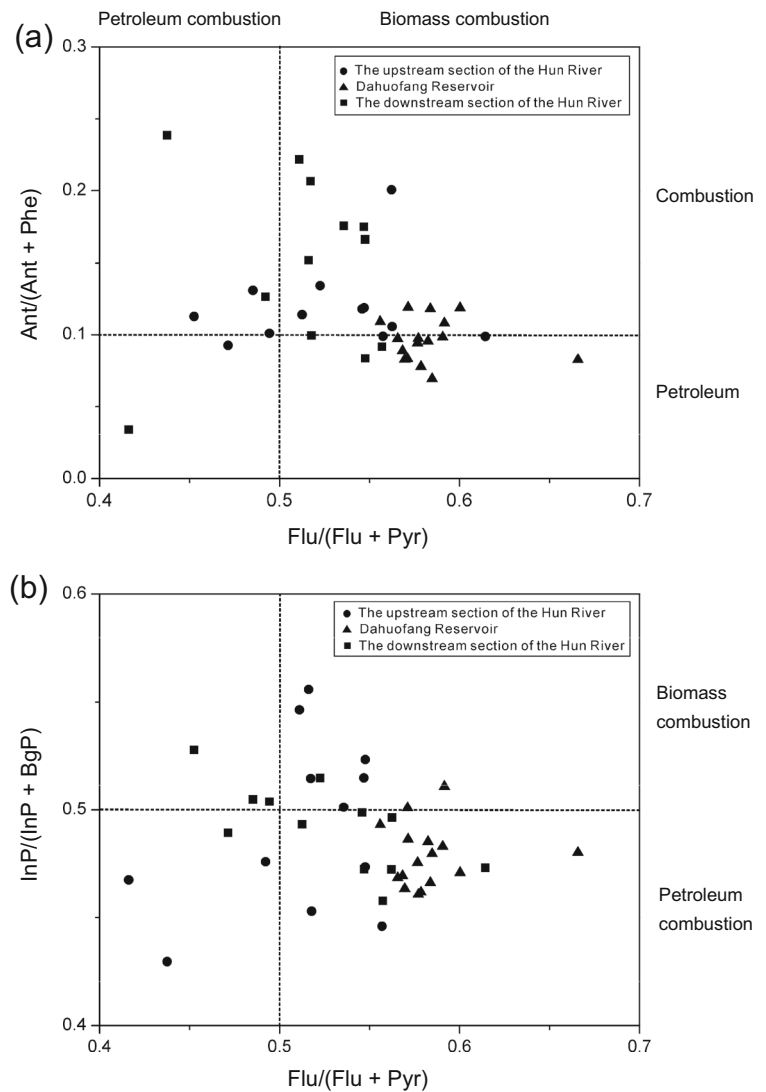
Ecological risk assessment of PAHs

Two effects-based guideline values, i.e., the effect range low (ERL) and the effect range median (ERM), are

widely used to evaluate the ecological risk caused by PAHs in aquatic sediments (Long et al. 1995). PAHs concentrations less than the ERL indicate that an adverse effect is rare, values exceeding the ERM suggest that adverse effect would occur frequently, while values between the ERL and ERM imply that there will be an occasional adverse effect.

We assessed the potential ecological risk posed by PAHs in the sediments by comparing the measured concentrations with the ERL and ERM values (Table 1). The \sum PAH concentrations in all the sediment samples from the upstream section of the Hun River and the Dahuofang Reservoir were significantly lower than the ERL, which suggested that there was a limited ecological risk posed by PAHs in these areas. However, the \sum PAH concentrations at most sites in the

Fig. 4 Plots of PAH isomeric ratios: **a** Flu/(Flu+Pyr) vs. Ant/(Ant+Phe), and **b** Flu/(Flu+Pyr) vs. InP/(InP+BgP)



downstream section of the Hun River were above the ERL, but below the ERM, which indicated that there may occasionally be a potential ecological risk.

At most sites in the upstream section of the Hun River and Dahuofang Reservoir, the concentrations of individual PAHs were below the ERL. Only Fl (H11, D2, and D9) and Phe (H11) posed possible ecological risks. In the downstream section of the Hun River, individual PAH levels at most sites were above the ERL but below the ERM, which indicated that an adverse ecological effect would occur occasionally. Several individual PAH concentrations were higher than the ERM at sites H14 (Fl, Phe, Ant, Flu, Pyr, BaP, and DBA), H16 (BaP and DBA), H17 (Phe), and X1 (Fl, Phe, Ant, Flu, and Pyr). The results indicated that at

these locations, there were high possibilities of potential ecological risks by these individual PAHs.

Conclusions

In this study, the PAHs on the US EPA priority list were measured in the sediments taken from the Hun River and the Dahuofang Reservoir. The total PAH concentrations (except for Nap) ranged from 82.96 to 39,292.95 ng g⁻¹ dw, with an average value of 3705.54 ng g⁻¹ dw. The data also showed that 4-ring PAHs were the dominant species at most sites in the study area. The values for the diagnostic parameters (Ant/(Ant+Phe), Flu/(Flu+Pyr) and InP/(InP+BgP))

indicated that the PAHs in the sediments had come from a number of sources, with pyrolytic origin being the dominant source. The results of the ecological risk assessment, which compared the PAH concentrations with the ERL and the ERM values, indicated that there was a potential ecological risk in the downstream section of the Hun River, and a limited risk in the upstream section of the Hun River and the Dahuofang Reservoir.

Acknowledgments This study was supported by National Natural Scientific Foundation of China (41130752, 41225013) and Integration of Production and Research projects of Guangdong province (2012B090500014).

References

- Baumard, P., Budzinski, H., & Garrigues, P. (1998). Polycyclic aromatic hydrocarbons (PAHs) in sediments and mussels of the western Mediterranean Sea. *Environmental Toxicology and Chemistry*, *17*, 765–776.
- Essien, J. P., Eduok, S. I., & Olajire, A. A. (2011). Distribution and ecotoxicological significance of polycyclic aromatic hydrocarbons in sediments from Iko River estuary mangrove ecosystem. *Environmental Monitoring and Assessment*, *176*, 99–107.
- Feng, C., Xia, X., Shen, Z., & Zhou, Z. (2007). Distribution and sources of polycyclic aromatic hydrocarbons in Wuhan section of the Yangtze River, China. *Environmental Monitoring and Assessment*, *133*, 447–458.
- Feng, J., Zhai, M., Sun, J., & Liu, Q. (2012). Distribution and sources of polycyclic aromatic hydrocarbons (PAHs) in sediment from the upper reach of Huaihe River, East China. *Environmental Science and Pollution Research*, *19*, 1097–1106.
- Gaspere, L., Machiwa, J. F., Mdachi, S. J. M., Streck, G., & Brack, W. (2009). Polycyclic aromatic hydrocarbon (PAH) contamination of surface sediments and oysters from the inter-tidal areas of Dar es Salaam, Tanzania. *Environmental Pollution*, *157*, 24–34.
- Guo, W., He, M., Yang, Z., Lin, C., Quan, X., & Wang, H. (2007). Distribution of polycyclic aromatic hydrocarbons in water, suspended particulate matter and sediment from Daliao River watershed, China. *Chemosphere*, *68*, 93–104.
- Haritash, A. K., & Kaushik, C. P. (2009). Biodegradation aspects of polycyclic aromatic hydrocarbons (PAHs): a review. *Journal of Hazardous Materials*, *169*, 1–15.
- Hiller, E., Zemanova, L., Sirotiak, M., & Jurkovic, L. (2011). Concentrations, distributions, and sources of polychlorinated biphenyls and polycyclic aromatic hydrocarbons in bed sediments of the water reservoirs in Slovakia. *Environmental Monitoring and Assessment*, *173*, 883–897.
- Jiang, B., Zheng, H.-L., Huang, G.-Q., Ding, H., Li, X.-G., Suo, H.-T., & Li, R. (2007). Characterization and distribution of polycyclic aromatic hydrocarbon in sediments of Haihe River, Tianjin, China. *Journal of Environmental Sciences*, *19*, 306–311.
- Kannan, K., Johnson-Restrepo, B., Yohn, S. S., Giesy, J. P., & Long, D. T. (2005). Spatial and temporal distribution of polycyclic aromatic hydrocarbons in sediments from Michigan inland lakes. *Environmental Science and Technology*, *39*, 4700–4706.
- Kanzari, F., Syakti, A. D., Asia, L., Malleret, L., Piram, A., Mille, G., & Doumenq, P. (2014). Distributions and sources of persistent organic pollutants (aliphatic hydrocarbons, PAHs, PCBs and pesticides) in surface sediments of an industrialized urban river (Huveaune), France. *Science of the Total Environment*, *478*, 141–151.
- Keshavarzifard, M., Zakaria, M. P., Hwai, T. S., Yusuff, F. M., Mustafa, S., Vaezzadeh, V., Magam, S. M., Masood, N., Alkhadher, S. A. A., & Abootalebi-Jahromi, F. (2014). Baseline distributions and sources of polycyclic aromatic hydrocarbons (PAHs) in the surface sediments from the Prai and Malacca Rivers, Peninsular Malaysia. *Marine Pollution Bulletin*, *88*, 366–372.
- Lin, T., Qin, Y., Zheng, B., Li, Y., Chen, Y., & Guo, Z. (2013). Source apportionment of polycyclic aromatic hydrocarbons in the Dahuofang Reservoir, Northeast China. *Environmental Monitoring and Assessment*, *185*, 945–953.
- Liu, Y., Beckingham, B., Ruegner, H., Li, Z., Ma, L., Schwientek, M., Xie, H., Zhao, J., & Grathwohl, P. (2013). Comparison of sedimentary PAHs in the rivers of Ammer (Germany) and Liangtan (China): differences between early- and newly-industrialized countries. *Environmental Science and Technology*, *47*, 701–709.
- Long, E. R., MacDonald, D. D., Smith, S. L., & Calder, F. D. (1995). Incidence of adverse biological effects within ranges of chemical concentrations in marine and estuary sediments. *Environmental Management*, *19*, 81–97.
- Luo, X. J., Chen, S. J., Mai, B. X., Yang, Q. S., Sheng, G. Y., & Fu, J. M. (2006). Polycyclic aromatic hydrocarbons in suspended particulate matter and sediments from the Pearl River Estuary and adjacent coastal areas, China. *Environmental Pollution*, *139*, 9–20.
- Martins, C. C., Bicego, M. C., Rose, N. L., Taniguchi, S., Lourenco, R. A., Figueira, R. C. L., Mahiques, M. M., & Montone, R. C. (2010). Historical record of polycyclic aromatic hydrocarbons (PAHs) and spheroidal carbonaceous particles (SCPs) in marine sediment cores from Admiralty Bay, King George Island, Antarctica. *Environmental Pollution*, *158*, 192–200.
- Mirza, R., Mohammadi, M., Faghiri, I., Abedi, E., Fakhri, A., Azimi, A., & Zahed, M. A. (2014). Source identification of polycyclic aromatic hydrocarbons (PAHs) in sediment samples from the northern part of the Persian Gulf, Iran. *Environmental Monitoring and Assessment*, *186*, 7387–7398.
- Mozeto, A. A., Yamada, T. M., de Morais, C. R., do Nascimento, M. R. L., Fadini, P. S., Torres, R. J., Sueitt, A. P. E., & de Faria, B. M. (2014). Assessment of organic and inorganic contaminants in sediments of an urban tropical eutrophic reservoir. *Environmental Monitoring and Assessment*, *186*, 815–834.
- Patrolecco, L., Ademollo, N., Capri, S., Pagnotta, R., & Polesello, S. (2010). Occurrence of priority hazardous PAHs in water,

- suspended particulate matter, sediment and common eels (*Anguilla anguilla*) in the urban stretch of the River Tiber (Italy). *Chemosphere*, *81*, 1386–1392.
- Scheibye, K., Weisser, J., Borggaard, O. K., Larsen, M. M., Holm, P. E., Vammen, K., & Christensen, J. H. (2014). Sediment baseline study of levels and sources of polycyclic aromatic hydrocarbons and heavy metals in Lake Nicaragua. *Chemosphere*, *95*, 556–565.
- Song, M. W., Gao, M., Wang, P. F., Xie, K. Z., & Zhang, H. (2014). Multivariate assessment of polycyclic aromatic hydrocarbons in surface sediments of the Beijiang, a tributary of the Pearl River in Southern China. *Environmental Monitoring and Assessment*, *186*, 907–918.
- Tobiszewski, M., & Namiesnik, J. (2012). PAH diagnostic ratios for the identification of pollution emission sources. *Environmental Pollution*, *162*, 110–119.
- Wang, Z. C., Liu, M., & Yang, Y. (2015). Characterization and sources analysis of polycyclic aromatic hydrocarbons in surface sediments in the Yangtze River Estuary. *Environmental Earth Sciences*, *73*, 2453–2462.
- Xue, B. M., Wang, Y. H., Zhang, D., Zhang, J. L., Leng, B., Huang, W. Y., & Chen, Z. H. (2013). Concentration, distribution and sources of polycyclic aromatic hydrocarbons (PAHs) in surface sediments from Lijiang River, South China. *Bulletin of Environmental Contamination and Toxicology*, *90*, 446–450.
- Yunker, M. B., Macdonald, R. W., Vingarzan, R., Mitchell, R. H., Goyette, D., & Sylvestre, S. (2002). PAHs in the Fraser River basin: a critical appraisal of PAH ratios as indicators of PAH source and composition. *Organic Geochemistry*, *33*, 489–515.
- Zeng, X., Lin, Z., Gui, H., Shao, W., Sheng, G., Fu, J., & Yu, Z. (2010). Occurrence and distribution of polycyclic aromatic carbons in sludges from wastewater treatment plants in Guangdong, China. *Environmental Monitoring and Assessment*, *169*, 89–100.
- Zhang, H. L., Sun, L. N., Sun, T. H., Li, H. Y., & Luo, Q. (2013a). Spatial distribution and seasonal variation of polycyclic aromatic hydrocarbons (PAHs) contaminations in surface water from the Hun River, Northeast China. *Environmental Monitoring and Assessment*, *185*, 1451–1462.
- Zhang, L., Qin, Y. W., Zheng, B. H., Lin, T., & Li, Y. Y. (2013b). Polycyclic aromatic hydrocarbons in the sediments of Xiangjiang River in south-central China: occurrence and sources. *Environmental Earth Sciences*, *69*, 119–125.
- Zhu, L. Z., Chen, Y. Y., & Zhou, R. B. (2008). Distribution of polycyclic aromatic hydrocarbons in water, sediment and soil in drinking water resource of Zhejiang Province, China. *Journal of Hazardous Materials*, *150*, 308–316.