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RESEARCH ARTICLE

Distribution, sources, and ecological risk assessment of polycyclic aromatic hydrocarbons in the water and suspended sediments from the middle and lower reaches of the Yangtze River, China

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Abstract Samples were collected from the middle and lower reaches of the Yangtze River, China, to study the concentrations, distributions, and compositions of 16 US-EPA priority polycyclic aromatic hydrocarbons (PAHs) in water and suspended particulate matter (SPM). We also evaluated sources of the PAHs and their potential toxicity. Total concentrations of the PAHs (ΣPAHs) in water ranged from 17.33 to 77.12 ng L^{-1}, and in SPM, the levels ranged from 595.91 to 2473.74 ng g^{-1}. Total concentrations of seven carcinogenic PAHs (ΣCPAHs) ranged from 7.63 to 13.02 ng L^{-1} in water and 276.55 to 1216.89 ng g^{-1} in SPM. PAH levels in water samples were relatively low, and those in the lower reaches were higher than in the middle reaches. SPM samples had higher levels of PAHs, especially in the lower reaches and in Dongting Lake and Poyang Lake. Principal component analysis (PCA) with multiple linear regression analysis (MLR) was performed to quantitatively characterize the PAH sources. Two factors and their contributions were identified from water samples. Coal and wood combustion accounted for 74.1 % of the PAHs, and petroleum emissions explained 25.9 % of the PAHs. Three source factors were identified from SPM samples: these were vehicular emissions (46.3 % of PAHs), wood and coal combustion (40.4 % of PAHs), and petrogenic sources (13.3 %). Ecological risk assessment indicated that a moderate undesirable impact will be caused by PAHs, and some control measures and remedial actions should be conducted.

Keywords Polycyclic aromatic hydrocarbons · Surface water · Suspended particulate matter · Source apportionment · The Yangtze River

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are common and persistent organic pollutants which consist of two or more fused benzene rings. They have a range of physicochemical properties. Many PAHs are dangerous to ecosystems and human health due to their toxicity, mutagenicity, and carcinogenicity (Zhang and Tao 2009; Sun et al. 2012). The United States Environmental Protection Agency (USEPA) identified 16 PAHs as high priority pollutants (Qiao et al., 2006). PAHs are mainly produced by incomplete combustion of fossil fuels (coal, petroleum, and fuel oil), coke ovens, exhaust emissions, and oil leaks (Yunker et al. 1996; Simoneit 2002). In addition, forest fires, volcanic activities, and petroleum seeps can also produce PAHs (Yunker et al. 2002; Lima et al. 2005). PAHs in aquatic environments are mainly introduced by atmospheric fallout, surface runoff, municipal and industrial effluents, and shipping (Lima et al. 2005; Parinos et al. 2013). Due to their high hydrophobicity and lipid solubility, most PAHs are sorbed onto the surface of fine particles, and end up sinking to the bottom sediment in bodies of water or

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they accumulate in fatty tissues of organisms (Qiu et al. 2009; Marrucci et al. 2013). Rivers are important pathways for transport of PAHs to the sea (Lipiatou et al. 1997; Motelay-Massei et al. 2006; Wang et al. 2007). Studying the sources, concentrations, and fate of PAHs in rivers is significant for analysis of marine pollution. In addition, rivers collect the runoff from drainage basins, and their PAH levels are an indication of contamination in the entire drainage basin. River pollution with PAHs is a major public concern due to potential long-term adverse effects on human and river organism health.

The focus area of this study was the Yangtze River, which is the largest river in China and of great significance to the Chinese economy. Most of the major international cities such as Chengdu, Chongqing, Wuhan, Hefei, Nanjing, Suzhou, Wuxi, and Shanghai are located in the Yangtze River Basin (YRB) and more than 400 million humans live in this area (Zhao et al. 2015). Rapid economic development and urbanization in the whole basin has resulted in water pollution and an increase in organic contaminants such as PAHs and polychlorinated biphenyls (PCBs) (Feng et al. 2007; Wang et al. 2016). Two metropolitan regions, the Yangtze River Delta Urban Agglomerations and the Triangle of Central China, are located in the middle and lower reaches of the Yangtze River. This has resulted in a large amount of pollutant discharge. The Yangtze River is an important water resource for residential uses, industry, and agriculture so pollution levels of the water are important to monitor. Additionally, healthy aquatic ecosystems such as the Yangtze River play a critical role in the sustainable development of China (Wang et al., 2012; Tu, 2014).

Some pollution research on the Yangtze has previously been done, but most studies have been of limited scope, and evaluation of PAHs was mainly focused on sediment samples (Feng et al. 2007; Li et al. 2012; Wang et al. 2015a). Therefore, more attention should be paid to the distribution of PAHs in a large scale of the Yangtze River such as the middle and lower reaches of the Yangtze River. The distribution of dissolved and particulate PAHs in the Yangtze River, which directly influences the PAHs discharged into the East China Sea (ECS), has not been studied. As one of the largest contributors of terrestrial materials, including PAHs, to the western Pacific shore should be paid more attention. Therefore, the primary objective of this study was to (1) preliminary investigate the possible sources, concentration, and distribution of PAHs in the aquatic environments of the middle and lower reaches of Yangtze River and (2) evaluate the potential toxicity and ecological risk of PAHs using toxic equivalent quantity. To achieve these goals, 16 US-EPA priority PAHs were measured in water and suspended particulate matter (SPM) in the main and branch streams of the section between Yueyang City in Hunan Province and Changshu City in Jiangsu Province.

Materials and methods

Study area

The Yangtze River has the fifth largest water discharge and fourth largest sediment load in the world (Milliman and Meade 1983; Milliman and Syvitski 1992; Eisma 1998). The Yangtze River drainage area is 1.8×10^6 km^2, and the river has a total length of 6300 km. It originates in the Qinghai-Tibet Plateau at 5100 m, then flows east through the Sichuan Basin, the Jianghan Plain, and the Middle-Lower Reach Plain before discharging into the ECS (Fig. 1). On the basis of hydrologic and geographic criteria, the Yangtze River can be divided into three parts: the upper, middle, and lower reaches. In this study, we focused on the middle and lower reaches which originate in Yichang City in Hubei Province and have a combined length of 1900 km and a drainage basin of 0.8×10^6 km^2. Three tributaries in the basin, Dongting Lake, Han River, and Poyang Lake, supply large amounts of water and sediments to the main stream. The annual mean precipitation in the basin ranges from 1000 to 1400 mm (Ding and Chan 2005). Weather is controlled by monsoon, and there are two primary seasons: a dry season from November to April and a wet season from May to October. The annual mean temperature in most parts of the middle and lower reaches ranges from 16 to 18 °C and differs between the south and the north.

The middle and lower reaches of the Yangtze River is the most prosperous region in China, and it has two of China's five major metropolitan areas: the Yangtze River Delta metropolitan area and the metropolitan area in the Middle Reaches of Yangtze River. In 2014, the gross domestic product (GDP) of these two metropolitan areas was 20 trillion yuan accounting for 31.4 % of the national GDP, and more than 320 million people live in this region (NBSC (National Bureau of Statistics of China) 2014). Over the past decade, the explosive population increase coupled with rapid economic growth has generated significant environmental pressures. Substantial amounts of domestic sewage and industrial effluents are discharged into rivers. In addition, industrial development and transportation consumes large quantities of fossil fuel and produces significant amounts of air pollutants. These human activities can all release PAHs into the environment.

Sample collection

Eleven sample stations along the river and its tributaries were selected (all located at the important cities or the confluence of rivers), and all of these are illustrated in Fig. 1. Eight stations were located on the main stream and were the following: Three Gorge Dam (TGD), Yueyang (YY), Wuhan (WH), Jiujiang (JJ), Fanchang (FC), Nanjing (NJ), Zhenjiang (ZJ), and Changshu (CS). Three stations were located in the

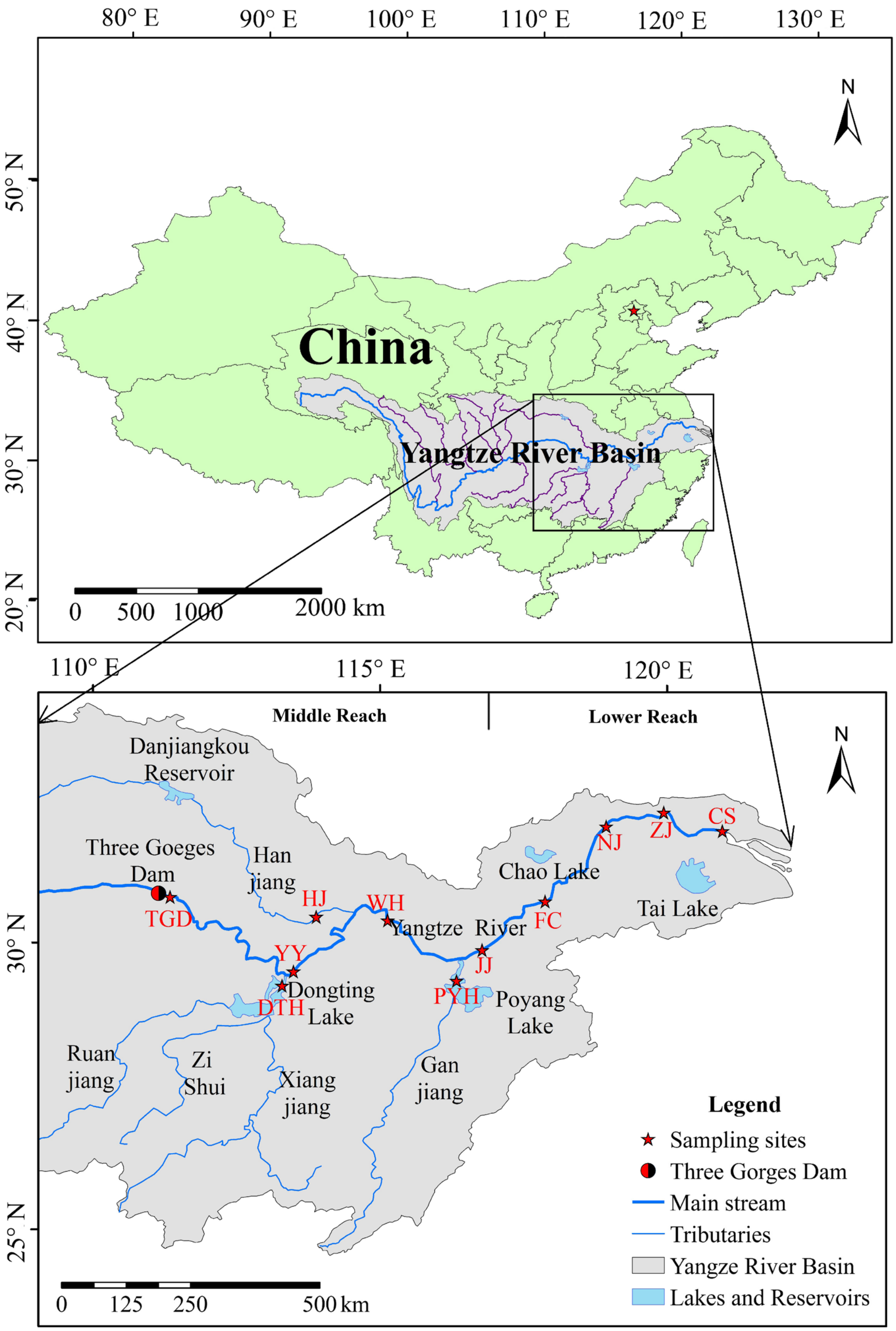

Fig. 1 **a** Map of the Yangtze River Basin in China showing the study sites. **b** The sampling sites with a *red star* were located within the middle and lower reaches of the Yangtze River (*TGD* Three Gorge Dam, *YY* Yueyang, *WH* Wuhan, *JJ* Jiujiang, *FC* Fanchang, *NJ* Nanjing, *ZJ* Zhenjiang, *CS* Changshu, *DTH* Dongting Lake, *HJ* Hanjiang, *PYH* Poyang Lake)

tributaries and were as follows: Dongting Lake (DTH), Han River (HJ), and Poyang Lake (PYH).

All the samples, including 11 water samples and 10 SPM samples, were collected in July and August 2015 (SPM in TGD was not acquired due to the lower suspended sediment concentration). Water samples of 100 L were collected from each sample site at the upper layer (0.5 m from the air-water surface) using a mental bucket and filtered immediately after sampling through precombusted (450 °C, 5 h) Whatman GF/F filters (nominal pore size 0.7 μm) to obtain sufficient SPM. All SPM samples were stored at −20 °C until determination of particulate PAH (PPAH). Additionally, other 2 L water samples were filtered, placed into precleaned brown glass jars, and kept frozen (−20 °C) until determination of dissolved PAH (DPAH).

Reagents and materials

A mixture standard of 16 US-EPA priority PAHs obtained from Supelco (Bellefonte, PA, USA) was used for external calibration, including naphthane (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), anthracene (Ant), phenanthrene (Phe), benzo[a]anthracene (BaA), chrysene (Chr), fluoranthene (Flo), pyrene (Pyr), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DahA), benzo[b]fluoracene (BbF), benzo[k]fluoracene (BkF), benzo[g,h,i]perylene (BghiP), and indeno[1,2,3-cd]pyrene (IcdP). Each component had a concentration of 2000 μg mL^{-1}. All solvents (dichloromethane, acetone, n-hexane, and methanol) used for sample processing were of HPLC grade from Tedia Co. (Tedia, USA). Anhydrous sodium sulfate was of analytical grade and activated at 450 °C for 5 h to remove impurities before using. Silica gel (100–200 mesh) was purchased from Qingdao Haiyang Chemical Co. (Shandong, China) and was extracted using acetone, dichloromethane, and n-hexane, then activated for approximately 16 at 130 °C.

Extraction and analysis of PAHs

Water samples (2 L) were solvent-extracted following standard method (Solid Phase Extraction, EPA Method 525) using solid-phase extraction cartridges (Supelco C18 cartridge, 500 mg/3 mL) at a flow rate of 5 mL min^{-1}. First, the cartridges were activated with 2 × 3 mL dichloromethane followed by 2 × 3 mL methanol and 2 × 3 mL ultrapure water. Extracts were eluted from the cartridge with 3 × 3 mL dichloromethane solvent. Then, the extracts were concentrated using a gentle stream of high purity nitrogen to 1 mL.

The SPM samples were processed by Soxhlet extraction which described by Wang et al. (2015b)). Samples were freeze-dried for 48 h, ground in a mortar to pass through a sieve with 200 mesh, and fully homogenized. Five grams (accuracy ±0.0001 g) of dry and homogenized samples was Soxhlet extracted with a mixture of n-hexane and acetone (100 mL, 1/1, *v*/*v*) for 24 h, to which 2 g of activated copper was added for desulfurization. The extracts were concentrated to 1 mL in a rotary evaporator, and then passed through an alumina/silica gel chromatography column (25 cm × 1 cm internal diameter) for cleanup and fractionation, and eluted with 50 mL of dichloromethane/n-hexane (2/3, *v*/*v*). Finally, the eluent was vacuum-evaporated and solvent-exchanged to n-hexane, and then concentrated under a gentle flow of nitrogen to 1 mL.

The concentrations of 16 US-EPA priority PAHs were analyzed by gas chromatography–mass spectrometry (GC-MS) using a Shimadzu QP2010 Ultra GC/MS equipped with a fused silica capillary Rtx-5MS column (30 m × 0.25 mm internal diameter, 0.25 μm film thickness). The carrier gas was helium at a flow rate of 1.0 mL min^{-1}. The GC temperature model was as follows: the initial temperature of 80 °C was held for 2 min and increased at a rate of 20 °C min^{-1} to 180 °C and held for 5 min. It was then increased at a rate of 10 °C min^{-1} to 290 °C and held for 15 min. The injection port was set at 290 °C. The interface and ion source were maintained at 280 and 230 °C, respectively. Ionization was carried out using the electron impact (EI) mode, and data were acquired using the selective ion monitoring (SIM) mode. Identification of PAHs was based on the selected ions and the relative retention time between samples and a standard solution containing individual PAHs.

Quality assurance/quality control

During the sample analyses, a procedural blank and a matrix spiked samples consisting of all analytes were analyzed in every six-sample set. The 16 US-EPA priority PAHs were quantified using the external standard method which was demonstrated effective by Wang et al. (2015a). Calibration curves based on a set of five standards of concentration 10, 50, 100, 250, and 500 ng L^{-1} were drawn. The standard curve was checked daily using the reference standard and ensured satisfactory linear regression coefficients ($R^2 > 0.999$) for all kinds of PAHs. The average recoveries of 16 PAHs based on matrix-spiked samples (PAH standards spiked into preextracted sample) ranged from 74.5 to 106.2 %. Limit of detection (LOD) was calculated as triple of the noise level of the blank sample (Wang et al. 2015b). LOD of DPAHs ranged between 0.12 and 0.63 ng L^{-1}, and PPAHs ranged between 0.16 and 0.77 ng g^{-1} in all samples. The average recoveries and LOD for 16 US-EPA PAHs are listed in Table S1 (supplementary material). Twenty percent of the samples (two water samples and two SPM samples) were duplicated, and the relative standard deviation (RSD) calculated to reflect the repeatability of the experiment was below 15 %.

Principal component analysis with multiple linear regression analysis (PCA-MLR)

The PCA-MLR model is a traditional and useful receptor model. It has been widely used to identify possible sources of PAHs (Sofowote et al. 2008; Li et al. 2012). To find a rational solution, we considered eigenvalues, factor loadings, and the accumulated variance contribution rate. The possible source categories can be identified based on these parameters. Then, we can obtain the principal component scores through the factor score matrix (Thurston and Spengler 1985). Finally, the contributions of the different sources to the 16 US-EPA priority PAHs in the water and SPM samples can be calculated based on the principal component scores, using multiple linear regression. A more detailed description of the PCA-MLR model can be found in literature (Li et al. 2012; Thurston and Spengler 1985; Shi et al. 2009). The PCA-MLR model was carried out using SPSS version 22.0.

The PCA model with varimax rotation was carried out to identify possible sources of PAHs in both water samples and SPM. Only factors from PCA analyses with eigenvalues >1 were taken into consideration, and chemical species were used to identify possible source categories only if these species factor loadings (absolute value) were greater than 0.7.

Results and discussion

PAHs distributions and composition in water and SPM samples

Spatial distribution of PAHs in water and SPM

Sixteen US-EPA PAHs were detected in the surface water and SPM samples from the middle and lower reaches of the Yangtze River. The surface water PAHs concentration among the 11 sampling sites ranged from 17.73 ng L^{-1} (DTH station) to 77.12 ng L^{-1} (ZJ station), with a mean value of 47.26 ng L^{-1} (Fig. 2). The SPM sample PAHs concentrations among the ten sampling sites ranged from 595.91 ng g^{-1} dry weight (dw) (JJ station) to 2473.74 ng g^{-1} dw (ZJ station), with a mean value of 1369.92 ng g^{-1} dw (Fig. 2). The ZJ station is located at Zhenjiang and near a harbor, where diesel leakage from ships may have contributed to the high PAH values. Figure 2 shows that PAHs in the lower reaches of the river were higher than middle reaches in water and SPM. However, the PPAH content in Dongting Lake (DTH station) and Poyang Lake (PYH station) was much higher than other stations in the middle reach and higher than some stations in the lower reach such as the FC and CS stations. Poyang Lake and Dongting Lake are the first and second largest freshwater lakes in China. These two lakes both have a large catchment and accumulate a large amount of pollutants. Zhi et al. (2015)) studied the PAHs in Poyang Lake and found that the level of PAHs can exceed 266 ng L^{-1}. Several PAHs, such as BaA, Chr, BaP, BkF, BbF, IcdP, and DahA, have been identified as potentially carcinogenic (Lee and Dong 2010; Tian et al. 2013). The total concentrations of seven carcinogenic PAHs (ΣCPAHs) in water and SPM ranged from 7.63 to 13.02 ng L^{-1} (with an average of 9.71 ng L^{-1}), and 276.55 to 1216.89 ng g^{-1} dw (with an average of 565.73 ng g^{-1} dw), respectively. BaP and DahA which have high toxicity were detected at all sampling stations. The lowest concentrations of ΣCPAHs in water and SPM samples were found at the YY and JJ stations, respectively. The highest concentrations of ΣCPAHs in water and SPM samples were found at the PYH and ZJ stations, respectively. The ΣCPAHs in water and SPM samples accounted for 21 and 41.3 % of total concentration of 16 US-EPA priority PAHs (ΣPAHs), respectively. The content of ΣCPAHs in SPM was much higher than water, which mainly resulted from the PAHs physicochemical properties

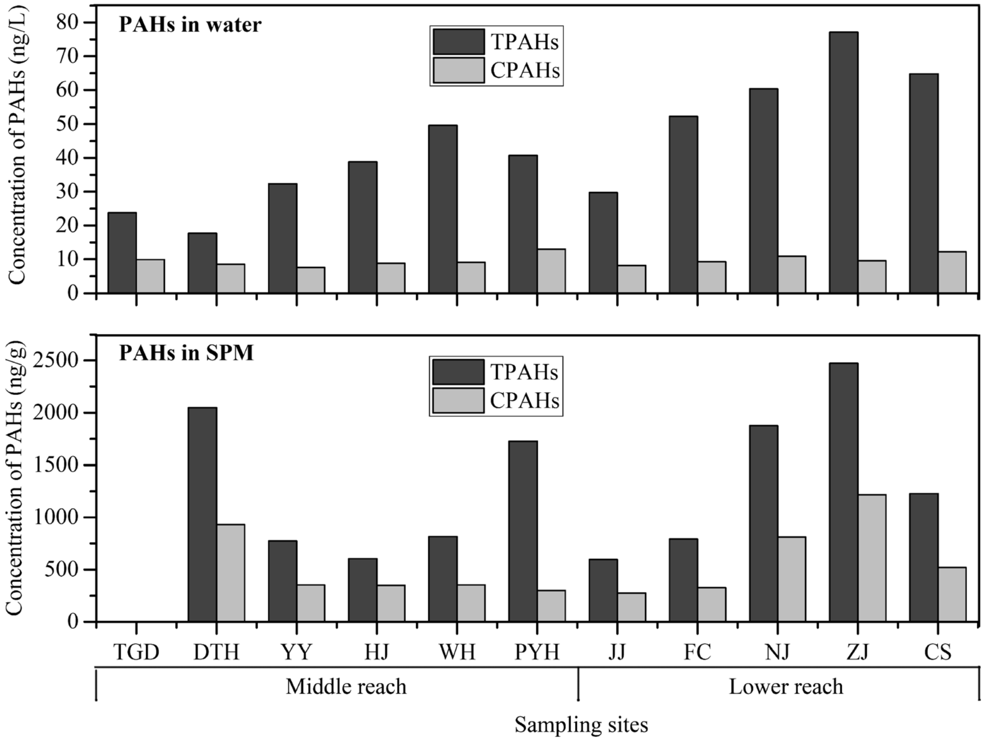

Fig. 2 Concentrations of total PAHs (TPAHs) and carcinogenic PAHs (CPAHs) in water and SPM samples at each sampling site from middle and lower reaches of Yangtze River (*TGD* Three Gorge Dam, *YY* Yueyang, *WH* Wuhan, *JJ* Jiujiang, *FC* Fanchang, *NJ* Nanjing, *ZJ* Zhenjiang, *CS* Changshu, *DTH* Dongting Lake, *HJ* Hanjiang, *PYH* Poyang Lake)

(lower water solubility and strong sorption) (Fig. 2). The levels of ΣCPAHs in ΣPAHs were relatively high, which may cause some environmental issues and human health problems.

Previous studies focused on the PAHs concentrations in water and SPM samples in the Wuhan section (Feng et al. 2007), Three Gorges Reservoir (Deyerling et al. 2014), and Poyang Lake (Zhi et al. 2015) (Table 1). We found that the concentrations of PAHs in different areas of the Yangtze River have relatively large differences (Table 1). The Wuhan section has a higher concentration of PAHs than other regions including those of this study. PAHs concentrations in the Three Gorges Reservoir and Poyang Lake are consistent with our results. Compared to other polluted rivers in the world the concentrations of PAHs in the water samples from the middle and lower reaches of Yangtze River are relatively low (Table 1). Comparison of middle and lower reaches of Yangtze River, middle and lower reaches of Yellow River, and the lower Mississippi River shows that the DPAH level is higher than the Mississippi River (12–25 ng L^{-1}) but lower than the Yellow River (179–2182 ng L^{-1}). PAHs in SPM samples from middle and lower reaches of Yangtze River were in the medium range. Concentrations of PAHs in SPM samples (595.9–2473.7 ng g^{-1} dw) were similar to those of the lower Mississippi River (1100–1700 ng g^{-1}) and much lower than levels in the middle and lower reaches of the Yellow River (54,000–155,000 ng g^{-1}). Concentrations of PAHs in water and SPM samples from the middle and lower reaches of Yangtze River were relatively low which may be attributed to the large amount of runoff and sediment load of the Yangtze River during the monsoon flood season.

Composition patterns of PAHs

The mean concentrations of individual PAHs in surface water and SPM samples are shown in Fig. 3. Phe, Flu, Pyr, and BbF predominated among the 16 US-EPA priority PAHs in SPM samples, and accounted for 50.3 % of ΣPAHs. BaA, Chr, BaP, IcdP, and BghiP also had high concentrations and accounted for 34.6 % of ΣPAHs. These results suggest that these compounds may accumulate in SPM and resist to degradation. The compositional pattern of PAHs in surface water samples is different from SPM and shows that Nap, Fl, and Phe are the most abundant PAHs in surface water samples among the 16 US-EPA priority PAHs (Fig. 3). These three individual PAHs accounted for 49.8 % of ΣPAHs. Therefore, these three PAHs are the predominant compounds and were more common than other PAHs, which was a function of their physicochemical

Table 1 PAH concentrations in water and SPM samples from various rivers in the world

Medium	Locations of sampling sites	PAHs	Concentration (ng g^{-1}, ng L^{-1})		Reference
			Mean	Range	
Water	Gao-ping River, Taiwan	16	430	10–9400	Doong and Lin 2004
	Middle and lower reaches of Yellow River, China	15	419	179–2182	Li et al. 2006
	Lower Mississippi River, USA	18		12–25	Mitra and Bianchi 2003
	Lower Brisbane River, Australia	15	8.2		Shaw et al. 2004
	Soan River, Pakistan	16	134	61–207	Aziz et al. 2014
	Yellow River Delta, China	14	121.3	64.8–334.6	Wang et al. 2009
	Middle and lower reaches of Yangtze River, China	16	47.26	17.73–77.12	This study
	Yangtze River, Wuhan section	11	1452	242–6235	Feng et al. 2007
	Poyang Lake, China	16		5.56–266.1	Zhi et al. 2015
	Three Gorges Dam	16	14.3		Deyerling et al. 2014
	Sarno, Italy	16	739	12.4–2321.1	Montuori and Triassi 2012
SPM	Middle and lower reaches of Yellow River, China	15		54,000–155,000	Li et al. 2006
	Lower Mississippi River, USA	18		1100–1700	Mitra and Bianchi 2003
	Yellow River Delta, China	14	209.1	65.6–675.4	Wang et al. 2009
	Middle and lower reaches of Yangtze River, China	16	1369	595.9–2473.7	This study
	Yangtze River, Wuhan section	11	3556	95.3–8798.1	Feng et al. 2007
	Sarno, Italy	16		833.1–8091.9	Montuori and Triassi 2012
	Three Gorges Dam	16	102.6		Deyerling et al. 2014

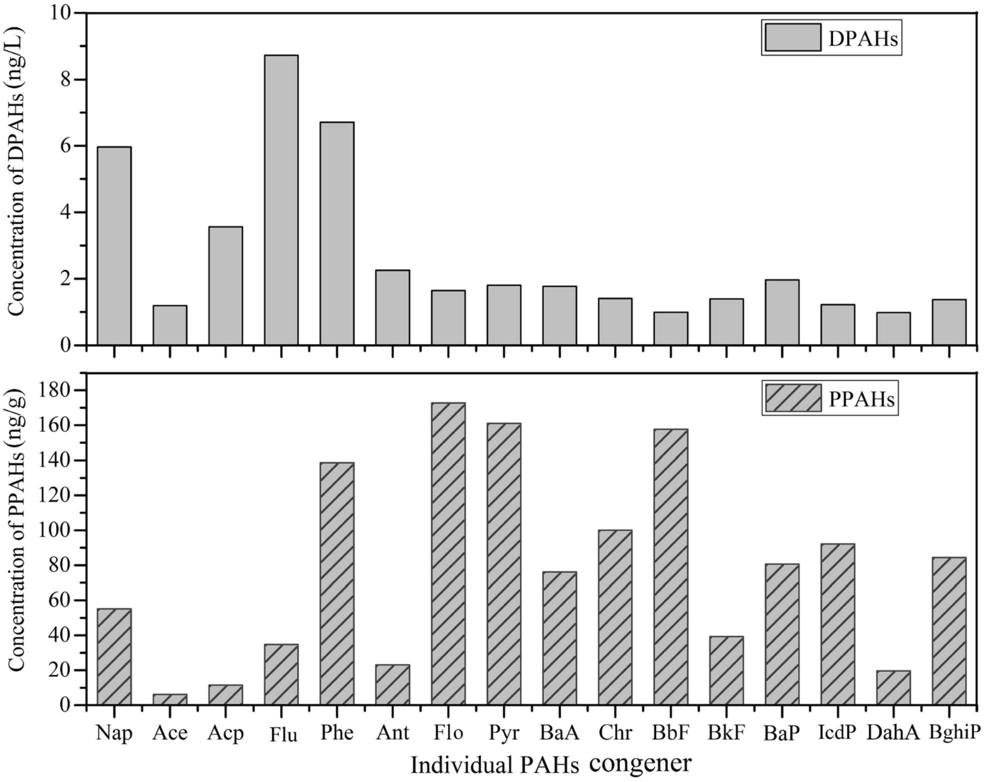

Fig. 3 Average concentrations of individual PAH compound in water and SPM samples from the middle and lower reaches of the Yangtze River

properties. Previous studies have been demonstrated that lower molecular weight PAHs (2–3 rings) have greater water solubility which may explain their relatively high concentrations in water (Bertilsson and Eidenfalk 2002; Liu et al. 2013).

According to the number of benzene rings, we can divide the 16 US-EPA priority PAHs into two groups: low-molecular-weight PAHs (LMW PAHs, with 2–3 rings) and high-molecular-weight PAHs (HMW PAHs, with 4–6 rings). Figure 4 shows the LPAH and HPAH percentages at each sampling site in the surface water and SPM samples. The compositional patterns of LMW PAHs and HMW PAHs are significantly different and show an opposite trend. HMW PAHs were predominant in SPM and LMW PAHs mainly found in water samples. Previous studies have demonstrated that the water solubility of PAHs decrease with the increasing number of aromatic rings and that HMW PAHs can exist for long periods in ambient environmental conditions (Sinaei and Mashinchian 2014; Li et al. 2015; Liu et al. 2015). HMW PAHs can persist in the environment for a long period due to their physicochemical properties and is easily absorbed in fine particle matrices, which resulted in a relatively high concentration of HMW PAHs in SPM samples. LMW PAHs have a relative high water solubility resulting in a high relative amounts of LMW PAHs in the water samples. LMW PAHs are mostly products of low-temperature combustion processes whereas HMW PAHs are produced by high-temperature combustion, and the LMW PAH/HMW PAH ratio can be used to qualitatively identify possible sources of PAHs (Wang et al. 2015a; Doong and Lin 2004). In this study, the ratios of LMW PAH/HMW PAH in water and SPM samples ranged from 0.55 to 3.1 and 0.36 to 0.97, with means of 2.13 and 0.57, respectively, According to these ratios of LMW PAH/HMW PAH, we qualitatively suggest that the main sources of PAHs found in water samples and SPM originated from coal, wood combustion, and vehicular emissions (Doong and Lin 2004; Wang et al. 2015b).

Source apportionment

Two source apportionment methods, molecular diagnostic ratios (MDRs) and principal component analysis (PCA), were used in this study to identify the possible sources of PAHs in water and SPM samples from the middle and lower reaches of the Yangtze River.

Molecular diagnostic ratios (MDRs)

The MDR method has been extensively used as a qualitative source apportionment method to identify possible PAHs sources (Yunker et al. 2002; Yan et al. 2005). Several molecular ratios, such as Flo/(Flo+Pyr), Ant/(Ant+Phe), IcdP/(IcdP+BghiP), and BaA/(BaA+Chr) have been developed for interpreting possible sources of PAHs (Yunker et al. 2002; Yan et al. 2006; Zhang et al. 2005; Wang et al. 2009). Previous studies have demonstrated that the Ant/(Ant+Phe) ratio should be used with caution, primarily because of

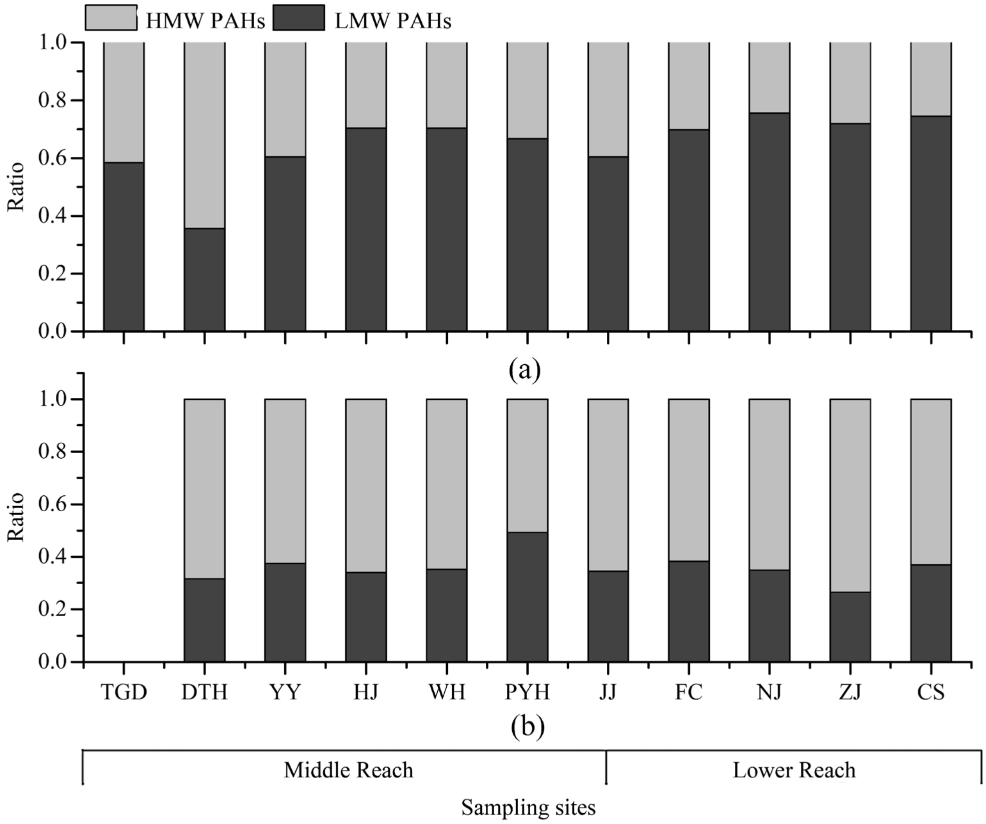

Fig. 4 The percentage variation of low-molecular-weight PAHs (LPAHs) and high-molecular-weight PAHs (HPAHs) in water (**a**) and SPM samples (**b**) at each sampling sites from the middle and lower reaches of Yangtze River (*TGD* Three Gorge Dam, *YY* Yueyang, *WH* Wuhan, *JJ* Jiujiang, *FC* Fanchang, *NJ* Nanjing, *ZJ* Zhenjiang, *CS* Changshu, *DTH* Dongting Lake, *HJ* Hanjiang, *PYH* Poyang Lake)

differences between the environmental behavior of Phe and Ant (Yan et al. 2006). Rogge et al. (1993) and Yan et al. (2005) demonstrated that Ant is more susceptible to degradation than Phe. In this study, we used Flo/(Flo+Pyr), IcdP/(IcdP+BghiP), and BaA/(BaA+Chr) as indicators to identify the possible sources of PAHs (Fig. 5).

Most of the stations (nine of the ten stations) had high values of Flo/(Flo+Pyr) ratio (>0.5) in SPM samples, which indicated that wood and coal combustion could be the possible source of PAHs in the middle and lower reaches of Yangtze River in SPM samples (Yunker et al. 2002). The ratio of Flo/(Flo+Pyr) in water sample shown an obvious difference between middle and lower reach, which is high than 0.5 in middle reach and between 0.4 and 0.5 in lower reach. Therefore, the PAHs in water samples originated from difference sources, and wood and coal combustion is the main source in middle reach, and liquid fossil fuel combustion is the main source in lower reach (Yunker et al. 2002). For ratio of IcdP/(IcdP+

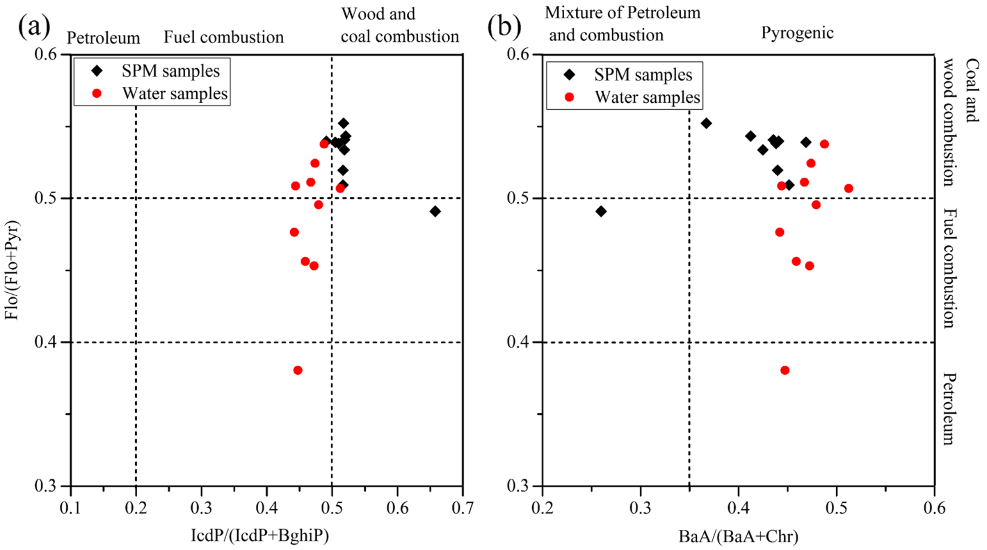

Fig. 5 Cross plots of **a** IcdP/(IcdP+BP) versus Flu/(Flu+Pyr) and **b** BaA/(BaA) versus Flu/(Flu+Pyr) in water and SPM samples from the middle and lower reaches of Yangtze River

BghiP), the water and SPM samples also shown an obvious difference, which showed that the ratio in water samples is between 0.2 and 0.5, and the SPM samples are almost larger than 0.5. Therefore, the source of PAHs in water samples mainly originated from fuel combustion and PAHs in SPM samples are mainly from wood and coal combustion (Yunker et al. 2002). In addition, the ratio of BaA/(BaA+Chr) are almost larger than 0.35, which means that the sources of PAHs in water and APM samples are all originated from pyrogenic source. Overall, the PAHs in water samples mainly originated from fuel combustion, and wood and coal combustion are the main sources of SPM samples.

Principal component analysis with multiple linear regression analysis (PCA-MLR)

The results of the PCA-MLR model are shown in Table 2 which identified two and three specific factors for water samples and SPM, respectively. In water samples, two principal components (PC1 and PC2) were extracted by the PCA-MLR model and accounted for the majority (72.19 %) of the total variance. First, PC1 with 44.14 % of the total variance was characterized by high loadings of Phe, Ant, Pyr, and BaP and moderate loading of Nap. BaP, Flu, and Pyr which are typical markers for coal combustion sources (Kavouras et al. 2001; Sofowote et al. 2008), and Ant was considered to be derived from wood combustion sources (Harrison et al. 1996). PC1 is therefore considered as a mixed contribution of coal and wood combustion sources (Zhang et al. 2012). Second, PC2 explained 28.05 % of the total variance and was dominated by BghiP, IcdP, and BbF and moderate loadings of DahA and BkF. BbF and BkF were identified as components of fossil fuels associated with their combustion (Kavouras et al. 2001). In addition, DahA and BghiP are characteristic emissions form gasoline combustion (Simcik et al. 1999), and IcdP was associated with diesel combustion (Sofowote et al. 2008). PC2 was therefore interpreted to represent loadings from vehicular emission sources including gasoline and diesel combustion. Multiple linear regression analysis (MLR) was performed on the PCA scores to obtain contribution rate to ΣPAHs of the two sources. The values of source contributions are presented in Table 2. We see that the highest contribution to the TPAHs in water samples was 74.1 % from the mixed wood and coal combustion which was mainly from industrial and agricultural activities (Zhang et al. 2012). The second major contribution to the TPAHs was 25.9 % from vehicular emission sources was mainly from shipping, fishing activity, and vehicle exhaust.

In the SPM, three principal components (eigenvalues >1) were extracted, which accounted for the majority (96.27 %) of the total variance. The first factor explained 69.8 % of total variance and had high loadings for the PAHs including BaA, Chr, BbF, BaP, and BghiP, and moderate loadings for DahA, IcdP, and BkF. BbF, BaA, and BghiP were associated with gasoline combustion, and IcdP was associated with diesel combustion (Sofowote et al. 2008). Consequently, factor 1 indicates

Table 2 Rotated component loadings of principal components for PAHs compositions in water and SPM samples from the middle and lower reaches of Yangtze River

	Water samples		SPM		
	PC1	PC2	PC1	PC2	PC3
Nap	**0.86**	−0.402	0.316	−0.044	**0.916**
Ace	0.056	0.009	0.614	0.502	0.57
Acp	0.026	**0.785**	0.689	0.099	0.701
Flu	0.552	**0.727**	0.03	**0.955**	0.256
Phe	**0.866**	0.267	0.395	**0.859**	0.283
Ant	**0.893**	−0.016	0.753	0.314	0.494
Flo	0.616	0.182	0.174	**0.978**	−0.091
Pyr	**0.893**	−0.037	0.133	**0.976**	−0.14
BaA	0.679	0.349	**0.928**	0.063	0.335
Chr	0.615	0.565	**0.938**	0.162	0.282
BbF	−0.144	**0.869**	**0.919**	0.318	0.2
BkF	0.152	**0.762**	**0.875**	0.06	−0.109
BaP	**0.892**	0.035	**0.942**	0.138	0.288
IcdP	−0.024	**0.891**	**0.897**	0.288	0.318
DahA	−0.201	**0.784**	**0.887**	0.245	0.383
BghiP	0.232	**0.927**	**0.917**	0.134	0.36
Contributions	74.1 %	25.9 %	46.3 %	40.4 %	13.3 %
Possible sources	Wood and coal combustion	Vehicular emission	Vehicular emission	Wood and coal combustion	Petrogenic source

contributions of PAHs from vehicular emission sources (a combination of gasoline and diesel emissions). Factor 2 accounts for 19.48 % of the total variance with high loadings of Flu, Phe, Flo, and Pyr. Phe associated with wood combustion (Simcik et al. 1999). Pyr and Flu are important indicators of biomass burning (Jenkins et al. 1996). Factor 2 can be identified as a wood/biomass combustion source. The third rotated component contributed 6.99 % of the total variance and was characterized by high loading of Nap, which is consistent with sources related to generic petroleum such as crude oil (Dahle et al. 2003; Saha et al. 2009). The contribution rate of three sources was evaluated by MLR, and the values of source contributions are given in Table 2. We found that the highest contribution to TPAHs in sediments was 46.3 % from vehicular emission sources which were mainly from shipping, fishing activities, and vehicle exhaust. The second major contribution to TPAHs was 40.4 % from a mix of coal and wood combustion sources generated by industrial, agricultural, and habitat activities (Zhang et al. 2012). In addition, petroleum sources contributed 13.3 % to PAHs in this area.

Overall, two primary sources of PAHs (wood and coal combustion and vehicular emission) were identified by the MDR and PCA-MLR methods. However, the source apportionment of PAHs in SPM samples through the PCA-MLR method identified additional sources, which provide more details and validate source information. The PCA-MLR method could also evaluate the contributions of every source to the TPAHs, and this could be used to quantitatively characterize the PAH sources. Therefore, the combination of MDR and PCA-MLR methods can verify source apportionment.

Potential toxicity based on carcinogenic PAHs (CPAHs)

Assessment of potential toxicity was performed using the total concentration of CPAHs (BaA, Chr, BaP, BbF, BkF, IcdP, and DahA) (Fig. 6). BaP, IcdP, BkF, Chr, and BaA were the predominant compounds, and DahA was also present at a relatively high concentration. However, the composition patterns in SPM differed from water samples. BkF, Chr, and BaA accounted for a relatively larger proportion and BaP and IcdP only predominated at the DTH, NJ, ZJ, and CS stations. The potential toxicity of sediment was calculated using the total toxic BaP equivalent (TEQ^{care}) as per the following equation:

$$TEQ^{care} = \sum Ci \times TEF_i \tag{1}$$

C_i is the CPAHs concentration (ng g^{-1} dw) and toxic equivalency factors (TEF_i) is the toxic factor of CPAHs relative to BaP. Among seven CPAHs, BaP is the only compound for which toxicological data allow derivation of a carcinogenic potency factor (Peters et al. 1999). The TEF of BaP was therefore regarded as the reference and assigned the value 1. The other six species—BaA, Chr, BbF, BkF, IcdP, and DahA—were assigned values 0.1, 0.001, 0.1, 0.1, 0.1, and 1, respectively (Cao et al., 2010). The TEQ^{care} values of water and SPM

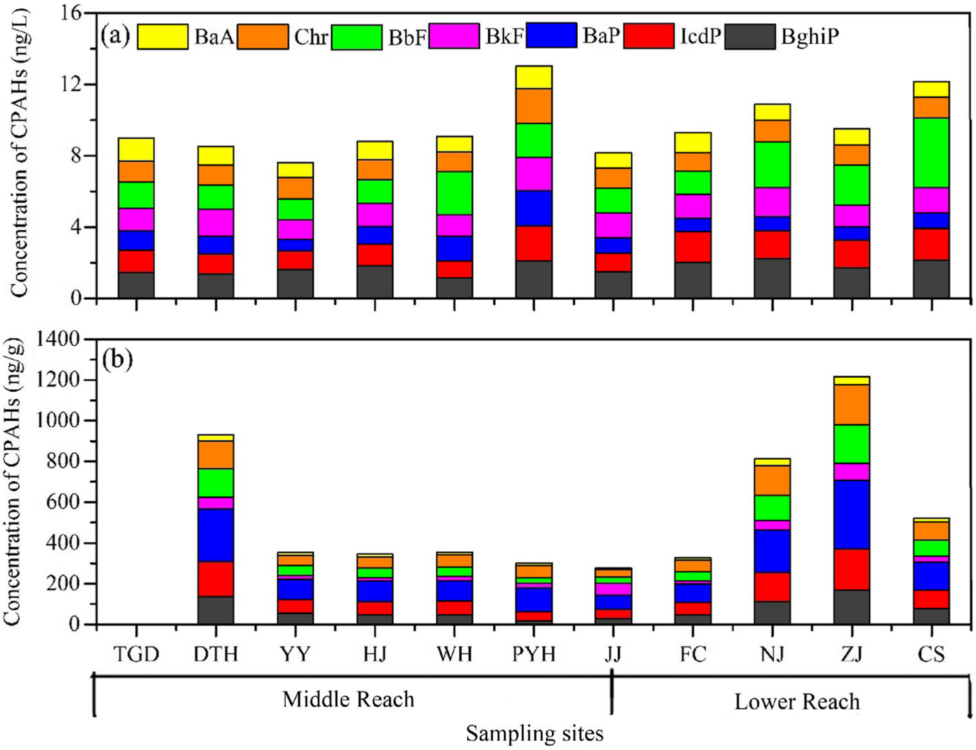

Fig. 6 Concentrations of carcinogenic PAHs (CPAHs) in water (a) and SPM (b) samples at sampling sites from the middle and lower reaches of the Yangtze River (*TGD* Three Gorge Dam, *YY* Yueyang, *WH* Wuhan, *JJ* Jiujiang, *FC* Fanchang, *NJ* Nanjing, *ZJ* Zhenjiang, *CS* Changshu, *DTH* Dongting Lake, *HJ* Hanjiang, *PYH* Poyang Lake)

samples ranged from 2.4 to 5.2 ng TEQ L^{-1} and 49.98 to 303.34 ng TEQ g^{-1} dw, respectively. In water samples, the concentration of TEQ^{care} in all sampling stations has a similar value. On the contrary, an obvious increasing trend of TEQ^{care} occurred in the SPM samples along the river. Economic development, especially industrial activity, plays an important role in the discharge of CPAHs into the environment. Among the seven CPAHs, contributions to the total CPAHs in water samples were, in ascending order, as follows: BkF (0.3 %), Chr (0.7 %), BaA (5.7 %), DahA (6.9 %), BbF (11.8 %), BP (14.6 %), and BaP (60 %). In SPM samples, contributions were Chr (0.4 %), BkF (0.4 %), BbF (3.2 %), DahA (3.7 %), BaA (5.3 %), BghiP (28.6 %), and BaP (58.4 %).

Ecological risk assessment

In order to assess potential ecological risk of PAHs in the Yangtze River, risk quotients (RQs) were used to evaluate the levels of risk caused by PAHs and showed as follows (Cao et al., 2010):

$$RQ = C_{PAH}/C_{QV} \quad (2)$$

where C_{PAH} was the concentration of certain PAH in the water and SPM samples; C_{QV} was the corresponding quality values of certain PAH in the samples. In this study, two critical values (negligible concentrations (NCs) and maximum permissible concentrations (MPCs)) were referenced by Kalf et al. (1997) and Cao et al. (2010) to use as the quality values in the samples. The "NCs" means negligible concentration, and if the PAH concentration in the study area are lower than this value, no risk will occurr. The "MPCs" means maximum permission concentration which will not exceed, and serious harm will happen if the PAH concentration in the study area is higher than this value. RQ_{NCs} and RQ_{MPCs} were defined as follows:

$$RQ_{NCs} = C_{PAH}/C_{QV(NCs)} \quad (3)$$

$$RQ_{MPCs} = C_{PAH}/C_{QV(MPCs)} \quad (4)$$

where $C_{QV(NCs)}$ and $C_{QV(MPCs)}$ was the quality values of the NCs and MPCs of PAH in the samples. The RQ_{NCs} and RQ_{MPCs} in different samples are listed in Table 3. Previous studies have demonstrated that $RQ_{NCs} < 1$ indicated that PAH might be probably negligible concern, while both RQ_{NCs} and RQ_{MPCs} are >1 for individual PAH indicated that risk is high and remedial actions must be undertaken at once. When $RQ_{NCs} > 1$ and $RQ_{MPCs} < 1$ indicated that the contamination of PAH might list as midline. In Table 3, RQ_{MPCs} of individual PAH were all less than 1 except for Pyr in SPM, and most of the RQ_{NCs} of individual PAH was >1, which showed that the ecological risk of PAHs in aquatic environment of the middle and lower reaches of Yangtze River was moderate. Therefore, some ecological risk might be caused to aquatic organism and some control measures and remedial actions should be conducted to decrease the contamination of PAHs.

Table 3 Mean values of $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ of individual PAH in water and SPM samples in the middle and lower reaches of Yangtze River

	Water (ng L^{-1})		SPM (ng g^{-1})		Water			SPM		
	NCs	MPCs	NCs	MPCs	Mean (ng L^{-1})	$RQ_{(NCs)}$	$RQ_{(MPCs)}$	Mean (ng g^{-1})	$RQ_{(NCs)}$	$RQ_{(MPCs)}$
Nap	12	1200	1.4	140	5.56	0.46	0	55.19	**39.42**	0.39
Ace*	0.7	70	1.2	120	1.19	**1.71**	0.02	6.13	**5.10**	0.05
Acp*	0.7	70	1.2	120	3.44	**4.91**	0.05	11.47	**9.56**	0.10
Flu*	0.7	70	1.2	120	7.88	**11.26**	0.11	34.75	**28.95**	0.29
Phe	3	300	5.1	510	6.39	**2.13**	0.02	138.64	**27.18**	0.27
Ant	0.7	70	1.2	120	2.16	**3.09**	0.03	23.08	**19.24**	0.19
Flo	3	300	26	2600	1.65	0.55	0	172.73	**6.64**	0.07
Pyr*	0.7	70	1.2	120	1.80	**2.57**	0.03	161.10	**134.25**	**1.34**
BaA	0.1	10	3.6	360	1.74	**17.41**	0.17	76.18	**21.16**	0.21
Chr	3.4	340	107	10700	1.39	0.41	0	100.04	0.93	0.01
BbF*	0.1	10	3.6	360	1	**10**	0.10	157.77	**43.82**	0.44
BkF	0.4	40	24	2400	1.38	**3.46**	0.03	39.30	**1.64**	0.02
BaP	0.5	50	27	2700	1.92	**3.84**	0.04	80.67	**2.99**	0.03
IcdP	0.4	40	59	5900	1.21	**3.03**	0.03	92.17	**1.56**	0.02
DahA*	0.5	50	27	2700	0.97	**1.93**	0.02	19.62	0.73	0.01
BghiP	0.3	30	75	7500	1.37	**4.55**	0.05	84.36	**1.12**	0.01

The quality value of individual PAH with "*" was referenced Cao et al. (2010)

Conclusions

This study systematically evaluated the characteristics of 16 US-EPA priority PAHs in water and SPM samples from the middle and lower reaches of the Yangtze River. Water samples had relatively low levels of PAHs (mean concentration = 47.26 ng L^{-1}) compared to other reported studies. The concentration of 16 US-EPA priority PAHs in SPM were higher (mean concentration = 1369.92 ng g^{-1} dw). The results of source apportionment indicated that mixed coal and wood combustion sources and vehicular emissions are the main sources of PAHs in the study area. Contribution rates of every source were evaluated by the PCA-MLR method, which showed that PAHs in water samples mainly originated from mixed coal and wood combustion sources (74.1 %) followed by vehicular emissions (25.9 %), whereas vehicular emissions accounted for 46.3 % of ΣPAHs in SPM samples and the mixed coal and wood combustion contributed 40.4 % of PAHs. The potential toxicity and risk quotients calculated indicated that some ecological risk might be caused to aquatic organism in the middle and lower reaches of Yangtze River.

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