



# Controlling factors for the distribution of typical organic pollutants in the surface sediment of a macrotidal bay

Ning Wang<sup>1</sup> · Ya Ping Wang<sup>1,2</sup> · Xiaoyong Duan<sup>3</sup> · Jianqiang Wang<sup>4</sup> · Yongqing Xie<sup>4</sup> · Chao Dong<sup>4</sup> · Jianhua Gao<sup>1</sup> · Ping Yin<sup>3</sup>

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## Abstract

Marine sediment is the final sink of polycyclic aromatic hydrocarbons (PAHs) from river input and atmospheric deposition. Such kind of pollutant cycles in the marine environment is usually controlled by hydrodynamic conditions. Many previous studies have explored how ocean currents influence pollutant distribution, but very few studies have focused on the relationships between the distribution patterns of pollutant and residual currents or sediment transport. In this study, 16 monomers of PAHs from 135 surface sediment samples collected in the Hangzhou Bay, a typical macrotidal bay, were systematically identified and their sources were analyzed. The sediment characteristics and distribution pattern were also comprehensively analyzed. The results showed the seabed sediments were moderately polluted by PAHs with a level of 38.58–1371.06 ng/g (median 186.70 ng/g). Most of the PAHs are composed of three to five rings, originated from combustion of coal and firewood. The combustion of oil also contributes to PAHs in seabed sediments for some areas. The PAHs are found to be concentrated within the estuary and the offshore areas, as well in coastal ocean. Fine-grained sediment transport is controlled by residual currents, which leads to PAH accumulation in the bay and the offshore areas, forming a high-value distribution pattern. Hence, we conclude that residual current is the main factors that control the long-term distribution of PAHs in the seabed sediments of the macrotidal bay. Temporal and spatial varying models of PAHs were needed in a further study to explore further the mechanisms how PAHs are transported in coastal areas.

**Keywords** PAHs · PAH sources · Hangzhou Bay · Residual currents · Sediment transport

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✉ Ya Ping Wang  
ypwang@nju.edu.cn

✉ Xiaoyong Duan  
duan-x@foxmail.com

- <sup>1</sup> Ministry of Education Key Laboratory for Coast and Island Development, School of Geographic and Oceanographic Sciences, Nanjing University, Nanjing 210093, China
- <sup>2</sup> State Key Laboratory of Estuarine and Coastal Research, School of Marine Sciences, East China Normal University, Shanghai 200241, China
- <sup>3</sup> Qingdao Institute of Marine Geology, China Geological Survey, Qingdao 266071, China
- <sup>4</sup> Zhejiang Institute of Hydrogeology and Engineering Geology, Zhejiang 315012, China

## Introduction

Polycyclic aromatic hydrocarbons (PAHs) mainly originate from anthropogenic activities such as vehicle emissions and incomplete combustion of fossil fuels and biomass (Sverdrup et al. 2002). They are known for their toxic, carcinogenic, mutagenic, bio-accumulative, and persistent characteristics (Khairy et al. 2014). Marine sediments are the final sink of PAHs from river input and atmospheric deposition (Nellier et al. 2015). Because of their low solubility and hydrophobic characteristics, PAHs can easily combine with suspended substances and settle to the sea bottom. Photochemical degradation or microbial oxidation decomposition is generally weak for PAHs after they attach to sediments, causing them to accumulate on sediments (Qiu et al. 2004; Dai and Liu 2011); therefore, the PAH concentrations in sediments can reflect the pollution of the seawater in a long period (Yang et al. 2019).

Hydrodynamic conditions are the main dynamic factors shaping coastal topography and regulating the evolution of

estuaries. Since water quality models were introduced in the early twentieth century, a large number of research results have been obtained by using tidal current as the main influencing factor in pollutant transport behavior and diffusion in coastal ocean and steeply sloped terrain. For example, because of the net effect of shear of coastal flows, most of the discharged sediments and related pollutants are trapped in the inland shelves of the Yangtze estuary (YRE) and East China Sea (ECS) (Milliman et al. 1985; Liu et al. 2006a, b, 2007; Bianchi and Allison 2009). Although PAHs have hydrophobicity and low solubility, strong hydrodynamics are generally considered to be one of the important factors affecting their distributions by influencing sediment transport. It is considered that the concentration of PAHs attached to sediments is relatively high in the muddy sedimentary area as sediments are transported and deposited with the current (Lin et al. 2013; Yu et al. 2015). Because of the seasonality and periodicity of ocean currents, the correlation between grain size and PAHs is not significant in some regions, and high PAH concentrations vary in argillaceous regions. Residual currents describe the net flow of water body after removing the periodic variations (e.g., tidal cycles), which show the average flow pattern in a long time, and has a long-distance transport effect on pollutants. Moreover, residual currents determine the long-term transport trend of fine sediment (Hamrick 1987; Li et al. 2005; Shou 2009). It is of great significance to study residual currents for the diffusion of pollutants (Hu and Sun 2009). Presently, however, the influence of residual currents on PAH distribution is rarely discussed.

Hangzhou Bay and the Zhoushan archipelago (ZA) are selected as our study area (Fig. 1). This area is a typical semi-closed bay with strong tides (i.e., the maximum tidal range of 8.94 m). It belongs to a wide shallow estuary with large residual current velocities, which is generally above

3.0 cm/s and the maximum is 8.4 cm/s. This area is also one of the most developed regions in China, with a large amount of pollutant emissions (Duan et al. 2019). At the same time, there are plenty of observation and simulation data for the flow field. Therefore, it is a unique and ideal place to study the role of hydrodynamic deposition process on the distribution and fate of polycyclic aromatic hydrocarbons in sediments. In this paper, the PAH concentrations in surface sediments were measured, and the distribution characteristics, composition structure, and source were analyzed. In particular, this contribution enhances the understanding of the mechanism on how residual currents and sediment transport influence the PAH pollution distribution patterns in the sea, so as to further evaluate the environmental pollution status of coastal sea and formulates pollution control measures.

## Materials and methods

### Sediment samples collection and pretreatment

In May 2016 and June 2017, a total of 135 surface sediment samples were collected by using a grab sampler from the Hangzhou Bay and surrounding waters (Fig. 1). The sample weight at each station was more than 1000 g (according to the GB 17378.3-2007, the *Specification for marine monitoring—part 3: sample collection, storage and transportation*, National Standards of P. R. China, 2007). Samples were placed in a sealed bag after being wrapped with aluminum foil and then stored at  $-20\text{ }^{\circ}\text{C}$  until analysis. The collected samples are mixed evenly according to the standard (the HJ 805-2016, *Soil and sediment—determination of polycyclic aromatic hydrocarbons-high performance liquid chromatography*, the Environmental Protection Industry Standards of P. R. China, 2016).

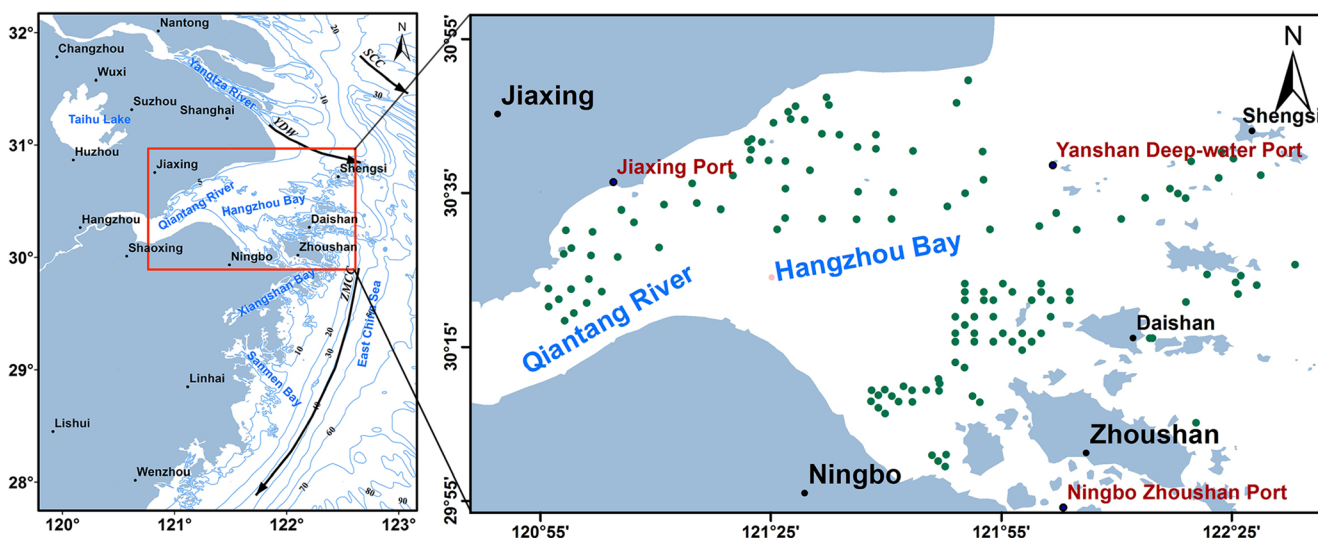


Fig. 1 Location of Hangzhou Bay (left) and magnified figure (right) showing sediment sampling stations

## Methods of sample extraction and analysis

The samples were analyzed at the Marine Geology Experimental Testing Center of the Ministry of Natural Resources. Ten grams of sediment samples was weighed in the accelerated solvent extraction (ASE) tank; D-PAHs and deuterium tetradecane were added as internal standards of aromatics and alkanes. Total extractable lipids (TEL) were determined by the ASE extraction. TEL was collected in a collection bottle. Copper was added for overnight desulfurization. The solution was evaporated under a flow of nitrogen to about 1 mL and then used for the solid-phase extraction (SPE). After extraction, the alkanes and aromatics were separately eluted from the SPE cartridge with *n*-hexane and *n*-hexane/dichloromethane (1/1, *v/v*). The elution was concentrated with nitrogen to 1 mL before analysis. The separation and detection of the EPA's 16 priority pollutant PAHs were performed on the Agilent 7890A-5975C GC-MSD system.

The total organic carbon and grain size analyses were conducted following GB 12763.8-2007 (*Specifications for oceanographic survey—part 8: marine geology and geophysics survey*, National standards of P. R. China, 2007).

## Grain size trend analysis

Based on the three grain size parameters of surface sediment, namely, mean grain size, sorting coefficient, and skewness, grain size trend analysis models can be used to calculate and obtain the net transport trend of sediment in marine environment (McLaren and Bowles 1985; Gao and Collins 1991). The 2-D Gao-Collins model has been successfully applied in many marine environments around the world, such as the Izmir Bay in Chile (Duman et al. 2004), Lirquen Bay in the Aegean Sea (Roux et al. 2002), Jiaozhou Bay and the adjacent ocean (Wang et al. 2014), North Branch of the Yangtze River Estuary (Yang and Liu 2002), etc., and its performance has been validated by other evidences of sediment transport (Ma et al. 2008). In this study, the grain size trend model proposed by Gao and Collins (1992) and Gao et al. (1994) was used to analyze the grain size trend in Hangzhou Bay. Based on the grain size analysis data of 135 PAH sampling stations and 375 surrounding stations, two kinds of interrelationships were identified from the possible eight grain size trends to meet the maximum frequency in the sediment transport direction, that is, (1) the sediment sorting becomes better, the mean grain size becomes finer, and the skewed becomes more and more negative and (2) the sediment sorting becomes better, the mean grain size becomes coarser, and skewness is more and more positive. A dimensionless unit grain size trend vector is defined between two adjacent stations when any of the two conditions is satisfied. Then the trend vector of each station and its adjacent stations is summed up to get the trend vector of the point. The transport pattern of sediment was obtained by

eliminating the influence of high-order noise using the vector smoothing synthesis.

## Residual current data

Residual currents are usually described as Eulerian and Lagrangian residual currents (Wang 2009). Euler residual current refers to the average current for a fixed location within a certain period of time; Lagrange residual current tracks the spatial displacement of a water particle within a certain period of time (representative of the periodic significance). The velocity of Lagrange residual current is calculated as the distance from the starting point to the end point divided by the corresponding time period, and the direction of Lagrange residual current is calculated as the vector from the starting point to the end point. Lagrange's long-term material transport theory has been proposed and well applied in the Bohai Sea and East China Sea since the 1980s and 1990s (Feng et al. 1986a, b; Wei et al. 2001), as well as the southwest of Vancouver Island in Canada (Foreman 1988), the English Channel (Delhez 1996), the Chesapeake Bay (Dortch et al. 1992; Cerco 1995), and many other areas. The Lagrangian residual current can better reflect the characteristics of long-term circulation and material transport than Euler residual current.

Residual current was caused by the interactions of wind-driven current, density current, tide-induced residual current and runoff into the sea. In situ measurements and numerical simulations are the main approaches to obtain the residual current field of the Hangzhou Bay in previous studies (Li and Hu 1987; Chen et al. 2003; Ni et al. 2003; Zou 2008; Xie et al. 2012). However, the results from both approaches were inconclusive because the calculation methods of residual current were different, which would be affected by topography, wind force, and non-linear interactions of tide and wave. In this paper, we obtained consistent dataset by collecting and comparing the flow field data in Hangzhou Bay in recent years. The overall characteristics of the flow field in the Hangzhou Bay were shown by the Lagrange residual current field.

## Results and discussion

### PAH concentrations

The results showed that the EPA's 16 priority pollutant PAHs were detected in all surface sediment samples from the 135 stations in the ZA area and Hangzhou Bay, suggesting that PAHs were widely distributed in the Hangzhou Bay. The concentration level (dry weight) was 35.38–1371.06 ng/g, with a median concentration of 186.70 ng/g (mean 254.22 ng/g), which was close to the results of Li et al. (2015; Li and Duan 2015) found in the East China Sea. The PAH concentrations of

surface sediments in the ZA sea area were much lower than that recorded by Jiang et al. (2014). According to Baumard et al.’s (2009) grading standard for PAH contamination in sediments, concentrations 0–100 ng/g indicate light pollution, 100–1000 ng/g indicate moderate pollution, 1000–5000 ng/g indicate high pollution, and greater than 5000 ng/g indicates severe pollution. Among the collected sediment samples in the study area, 21% have light PAH pollution, 76% have medium pollution, and 3% have severe pollution (Fig. 2a).

The concentrations of PAHs in surface sediments of Hangzhou Bay are compared with other coastal areas. Dalian Bay (157–20,855 ng/g, mean 1152.08 ng/g; Liu et al. 2011), Laizhou Bay (mean 554.24 ng/g, Zhang et al. 2016), and Qingdao offshore (255.77–1884.07, Wang et al. 2010), which are located in the north of China, were found to have

much higher concentrations of PAHs. Xiamen Bay (203.98–1590.47, Li et al. 2009) and Shenzhen offshore (227.51–3897.42 ng/g, Tang et al. 2017), which are located in the south of China, also showed much higher concentrations of PAHs. On the other hand, the less economic-developed Meizhou Bay in Fujian (196.7–299.7 ng/g, Lin et al. 2003) showed a similar concentration level of PAHs comparing with the Hangzhou Bay, and the less-developed coastal areas of Leizhou (42.0–109.0 ng/g, with an average of 74 ng/g) and Liusha Bay in Guangxi (108.3–370.4 ng/g, with an average of 158.9 ng/g, Sun 2012) showed a lower concentration level of PAHs comparing with the Hangzhou Bay. Compared with other economically developed regions, the concentrations of PAHs in the Hangzhou Bay are much lower than that in the Kaohsiung port (472–16,201 ng/g, Chen and Chen 2011), Casco Bay (16–

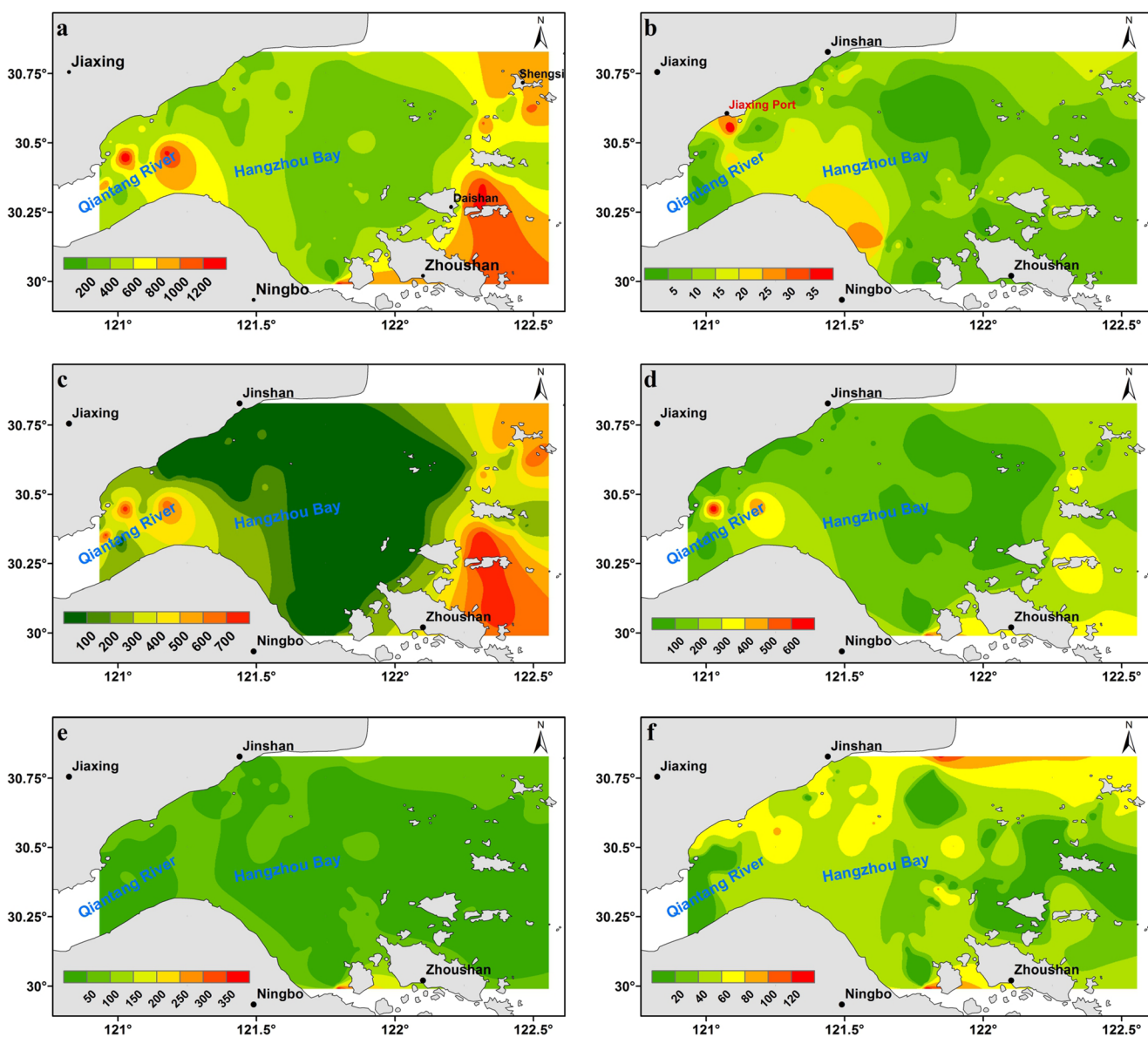


Fig. 2 Distribution patterns of PAHs in the Hangzhou Bay: a  $\Sigma$ PAHs, b two rings, c three rings, d four rings, e five ring, and f six rings

20,748 ng/g, Kennicutt 1994), Masan Bay (207–2670 ng/g, Yim et al. 2005) and Tokyo Bay (109–1170 ng/g, Horii et al. 2009). The Hangzhou Bay is one of the most economically developed regions in China. It was estimated that the annual flux of the 16 USEPA priority PAHs (16 PAHs) from the Yangtze River to the sea was 232 t, significantly higher than that from the Yellow River (70.5 t) and the Pearl River (33.9 t) (Wang and Gao 2007). River discharge (carrying dissolved and particulate associated pollutants) and atmospheric deposition from coastal ocean (both in precipitation and dry deposition) were primary sources of PAH inputs to coastal marine environments (Guo et al. 2007; Lohmann et al. 2007). Assuming the amount of PAHs from atmospheric deposition in this region is close to that in North and South China (Wang et al. 2017), the medium-low levels of PAH concentrations in this region is probably caused by the strong diluting effect of large river runoff and strong tidal forces (Chen et al. 2006; Lin et al. 2013).

### Spatial distribution and composition of PAHs

Figure 2a shows the spatial distribution of total PAHs ( $\Sigma$ PAHs). The high concentrations of PAHs in the sediments were observed near the Qiantang River Estuary, Daishan (in the north of ZA) and Shengsi Islands. The maximum value occurred near Jiaying, at the mouth of Qiantang River. The minimum value appeared near the mouth of the Yongjiang River (near Ningbo), which is experiencing coastal erosion. Overall,  $\Sigma$ PAHs were high near the estuary and in coastal areas, and low near the central gulf and open waters. PAH concentrations in sediments were closely related to nearby terrestrial environments. For instance, the distance from a city may be an important factor affecting the PAH concentrations in surface sediments (Li et al. 2015; Li and Duan 2015). Figure 2a shows that the concentration distribution pattern of PAHs within the study area can be roughly divided into three regions, i.e., the Qiantang River estuary area, central Hangzhou Bay area, and ZA inshore area.

PAHs can be divided into two groups based on the number of benzene rings: low molecular weight PAHs (PAHs with two to three rings) and high molecular weight PAHs (PAHs with four to six rings). The single PAHs in the sediment samples from this region were predominantly with three, four, and five rings (with mean values of 34.3, 27.6, and 19.3%, respectively), but the distributions of different types of PAHs varied among regions (Fig. 2b–f). Among the 16 PAHs, the average content of phenanthrene (tricyclic ring) is the highest, similar to the results of other researches (Ou et al. 2009; Hu et al. 2017). The concentration range of low molecular weight (LMW) PAHs was 10.55–746.42 ng/g. LMWs accounted for 12.0–88.8%, with an average of 39.7%, of the total PAHs. High molecular weight (HMW) PAHs were highly concentrated, ranging from 28.03 to 1043.76 ng/g and accounted for 11.2–88.0%, with an average of 60.3% of the total PAHs. It

should be noted that, over the entire study area, the concentration of LMW PAHs on the land is higher than that of HMW PAHs (Li et al. 2016), while the HMW PAHs are higher in the sea, possibly because of their different sources and physico-chemical characteristics. Previous studies have shown that HMW PAHs can exist for long periods of time in the sea and are easily adsorbed to fine particle surface (Lin et al. 2013; Li et al. 2015; Li and Duan 2015).

The samples from coastal areas in the northwest and southwest of the study area had high concentrations of two-ring PAHs (Fig. 2b). According to previous researches (Yamada et al. 2003; Stogiannidis and Laane 2015), two-ring PAHs are closely associated with oil pollution. One of China's busiest ports (Ningbo–Zhoushan Port) was located in this area. Water transportation may have played an important role on the environment (Li et al. 2010; Lin et al. 2011). Further, used crankcase oil may be an important source of PAHs in coastal sediments (Zakaria et al. 2002).

Three-ring low molecular weight PAHs originated mainly from petroleum products (Mai et al. 2003). The high concentrations of three-ring PAHs were observed in the Zhoushan archipelago and the coastal waters of Jiaying (Fig. 2c). A large iron ore transfer terminal, Shulanghu Transfer Terminal, is under construction in Zhoushan. The oil leakage generated by busy ships may be the cause of the high concentrations of 3-ring aromatic hydrocarbons in this area.

High concentrations of 4-cyclic aromatic hydrocarbons were found in the coastal ocean of Jiaying, and Ningbo (Fig. 2d). The largest refinery Sinopec is located in Ningbo. It has the capacity to process 23 million t/yr crude oil, produce 1 million t/yr ethylene and 2 million t/yr aromatics, and has a 45 million t/yr deep marine terminal throughput capacity and a storage capacity of more than 3.3 million m<sup>3</sup>.

The high concentrations of five cyclic aromatic hydrocarbons appeared in the southwest (Fig. 2e), and the high concentrations of six-ring PAHs occurred in the south and the Yangtze estuary and nearshore areas of Jiaying (Fig. 2f). In the southern region, there were a large number of chemical enterprises such as Ningbo Qingzhi Chemical Wharf Co., Ltd., Hai-Neng Oil Co., Ltd., and Ningbo Gulf Heavy Industries Co., Ltd., which may release PAHs to the coastal ocean. In addition, atmospheric concentration of benzo[a]pyrene (five rings) in China was much higher than that reported elsewhere in the world. It may due to the dominance of coal as a fuel source (Douben 2003). However, six-ring PAHs, such as benzo[g,h,i]perylene and indeno[1,2,3-cd]pyrene, were only detected in the samples of highway tunnel, diesel and gasoline engine rather than coke or wood combustion samples (Motelay-Massei et al. 2007), which indicated that six-ring species were mainly released by vehicles. Liu et al. (2015) reported that PAHs that were detected in dry and wet deposition samples in Shanghai, were from vehicle exhausts, and it contributed over 40% of the total PAHs. In

addition, PAHs detected in soil samples from Shanghai also originated from vehicular emissions (Wang et al. 2015). Therefore, the main source of PAHs detected in riverine inputs and surface runoff was vehicular emissions. This is determined by atmospheric physical processes and physicochemical properties of PAHs: PAHs from vehicle-exhaust emission are close to the earth surface, resulting in easy absorption onto soil particles. The relatively higher density of vehicle exhausts than normal air, and thus unfavored to be transported to the upper air and consequently over longer distances, also made the vehicle exhaust one of the most important source for PAHs in soils and rivers.

The composition and concentration of PAHs in the Qiantang River estuary, Hangzhou Bay, and ZA coastal area have distinct characteristics (Table 1). In the Qiantang River estuary and nearshore area of ZA, most of the LMW/HMW ratios were higher than 1 (more than 83%). The proportion of low molecular weight PAHs was around 65%, with a low content of five-ring aromatic hydrocarbons. On the other hand, the LMW/HMW ratios at most stations in Hangzhou Bay were lower than 1, with an approximately 30.6% LMW PAHs. The proportions of four- and five-ring PAHs were relatively high, indicating heat sources are dominant in the bay.

**PAH sources**

PAHs are mainly produced by human activities, including incomplete combustion of fossil fuel (coal, oil, etc.) and biomass fuel, automobile exhaust, and oil spill. The component of PAHs vary with their generation processes, thus the qualitative and quantitative analysis of the component of PAHs can be used to determine the sources of PAHs (Deng 2013).

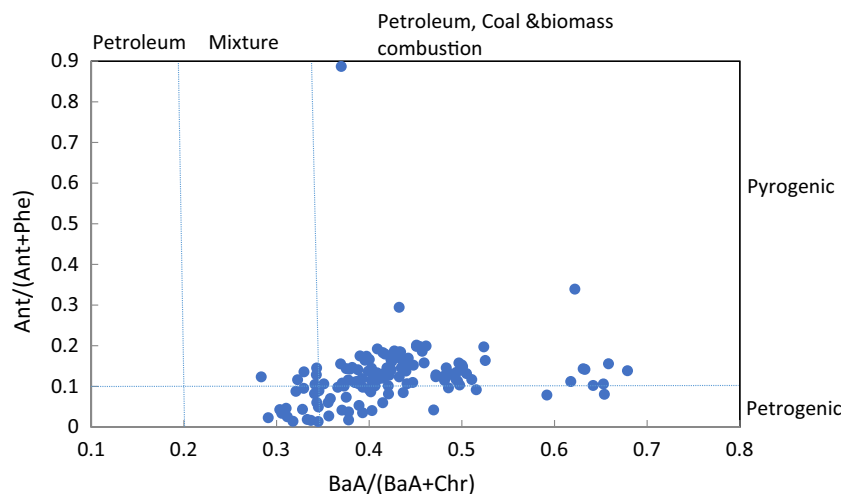
Diagnostic ratio is a widely used method to reveal the origin and sources of PAHs (Yunker et al. 2002; Jiang and Dong 2008; Mostert et al. 2010; Tobiszewski and Namieśnik 2012; Katsoyiannis and Breivik 2014). Yunker et al. (2002) integrated four isomer ratios from a large number of environmental samples or single source and stated that Ant/(Ant+Phe), Fla./(Fla+Pyr), BaA/(BaA+Chr), and InP/(InP+BgP) ratios of 0.1, 0.4, 0.2, and 0.2, respectively, indicated petrogenic and pyrogenic sources. The pyrogenic sources include combusted coal, grass, wood, and petroleum. Previous studies reported that BaA/(BaA+Chr) < 0.2 indicated a petroleum source. When the ratio is above 0.35, PAHs mainly came from petroleum, coal, and biomass combustion. When the ratio is between 0.2 and 0.35, PAHs were from both sources. The calculated diagnostic ratios of sediment samples in the Hangzhou Bay suggest that pollution sources were mainly coal, biomass, and petroleum combustion, and some sites were also affected by oil leakage (Fig. 3).

Principal component analysis (PCA) was further used to analyze PAH sources in this region, and three principal components with characteristic values greater than 1 were

**Table 1** Mean/median and contributions of PAHs (ng/g) in sediments from different regions

Area	ΣPAHs (ng/g)	Mean (median) ΣPAHs (ng/g)	LMW (ng/g)	Mean (median) LMW (ng/g)	LMW Contribution (%)	HMW (ng/g)	Mean (median) HMW (ng/g)	HMW Contribution (%)
Qiantang Estuary	113.45–1371.06	445.99 (318.04)	57.16–704.7	284.07 (229.19)	65.2	28.44–666.36	161.92 (82.79)	34.8
Hangzhou Bay	36.58–1186.24	173.40 (176.49)	10.55–142.48	48.94 (44.51)	30.6	28.03–1043.76	124.46 (122.56)	69.4
Zhoushan Archipelago	183.03–1044.78	506.96 (421.12)	95.79–746.42	332.42 (275.32)	64.6	61.55–388.93	174.54 (144.57)	35.4

**Fig. 3** Diagnostic ratio charts for Ant/(Ant+Phe) and BaA/(BaA+Chr) at the sampling sites



extracted. The cumulative variance contribution rate was 83.6%, and the variance contribution rates were 50.39, 24.87, and 8.36%, respectively (Table 2).

In factor 1 (F1), PAHs with high molecular weights, including BaA, Chr, BbF, BkF, BaP, InP, and BgP, had very high loads. The high molecular weight PAHs were mainly formed during high-temperature combustion processes. For example, Pyr, BaA, and Chr were usually related to coal combustion, while BbF and BkF were indexes of diesel combustion. InP and BgP were often used as tracers of automobile exhaust (Yunker et al. 2002; Deng 2013). Therefore, F1 demonstrated

high-temperature combustion sources (fossil fuel combustion) of PHAs, which can be further categorized as a mix of combustion and vehicle exhaust. This is consistent with the energy (mainly coal and coke) consumption structures of Zhejiang, Shanghai, and other places (China National Bureau of Statistics 2017).

In factor 2 (F2), the components with large loads were mainly low molecular weight PAHs, including Flu, Phe, and Ant. The PAHs with low molecular weight were mostly from crude oil and petroleum product leakage, incomplete combustion of fossil fuels, and low-temperature combustion or diagenetic biomass processes. Therefore, F2 mainly demonstrated petroleum sources.

In factor 3 (F3), Nap load was relatively high. The very little information in F3 made it difficult to infer reasonable sources. Principal component analysis showed that high-temperature combustion processes were the main source of PAH pollution in surface sediments of this region, occupying the first principal component (50.39%), while the second principal component with low molecular weight loads can be considered a petroleum source, accounting for 24.87%. The results of PCA were consistent with the diagnostic ratio.

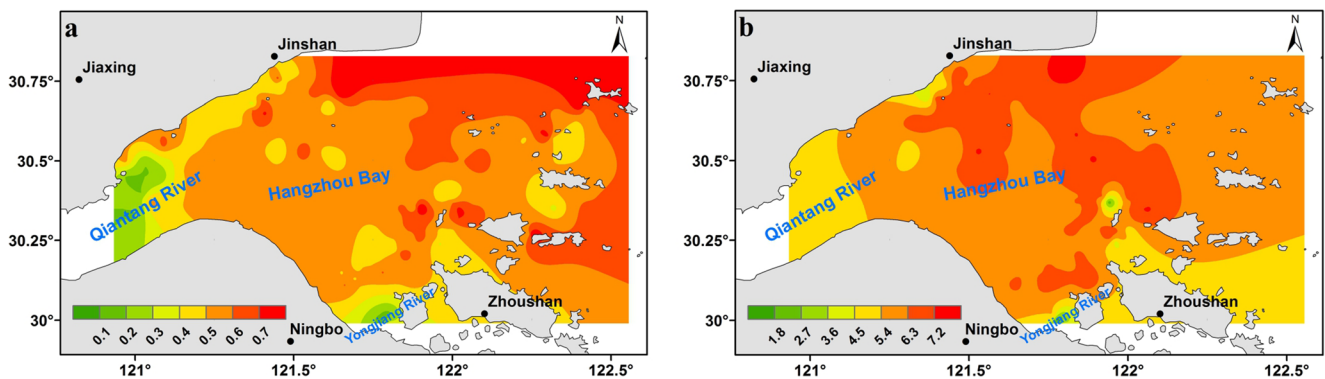
**Table 2** Results of principal component analysis of PAHs in Hangzhou Bay

PAHs	Component		
	F1	F2	F3
Naphthalene (Nap)	0.353	-0.053	0.827
Acenaphthylene (Acy)	0.879	-0.120	0.265
Acenaphthene (Ace)	0.688	0.047	0.463
Fluorene (Flu)	0.276	0.857	0.211
Phenanthrene (Phe)	0.196	0.909	0.012
Anthracene (Ant)	0.366	0.794	-0.141
Fluoranthene (Fla)	0.399	0.848	-0.100
Pyrene (Pyr)	0.576	0.700	-0.139
Benzo(a)anthracene (BaA)	0.886	-0.212	-0.311
Chrysene (Chr)	0.909	-0.203	-0.232
Benzo(b)fluoranthene (BbF)	0.946	-0.196	-0.052
Benzo(k)fluoranthene (BkF)	0.907	-0.211	-0.186
Benzo(a)pyrene (BaP)	0.920	-0.255	-0.231
Indeno(1,2,3-cd)pyrene (InP)	0.548	-0.502	0.149
Dibenzo(a,h)anthracene (DbA)	0.784	0.149	0.102
Benzo(g,h,i)perylene (BgP)	0.927	-0.230	0.024
Variance (%)	50.39	24.87	8.36

## Factors controlling the spatial distribution patterns of PAHs

### Relationship between PAHs and TOC or grain size

PAHs have generally low concentrations in seawater, but they can be adsorbed by particles and then settled in the ocean. Most scientists believe that the organic pollutants are absorbed by both the mineral components and organic matter in the sediments. However, because of the polarity of the mineral surface, the dipole effect occurs in seawater, which makes the polar water molecules and the mineral surface combine to occupy the adsorption position on the surface. Therefore,



**Fig. 4** Distributions of **a** TOC (%) and **b** median grain size ( $\Phi$ ) in the Hangzhou Bay

the adsorption of organic pollutants by mineral components is secondary, and the substances that play a major role in adsorption are organic matter (Luo et al. 2004). Generally speaking, the fine-grain size components of sediment have high TOC contents and adsorption surface areas, so the PAH concentrations of the fine-grain size components are usually high (Upal and Jeffrey 2001; Jiao et al. 2010). Therefore, the total organic carbon content and grain size are generally considered two important factors to control the distribution of PAHs. Figure 4 shows the spatial distributions of TOC and median grain size in the study area.

The TOC content in the study area varied widely from 0.02 to 0.75%, with an average of 0.42%. The TOC content also showed distinct regional characteristics (Fig. 4a). In the Qiantang River and Yongjiang estuary, the TOC content was low, while the TOC content was high near the Yangtze River estuary. The average TOC content increased from west to east, from 0.20% in the Qiantang River estuary to 0.44% in the Hangzhou Bay and 0.50% in the ZA area. No significant correlation was found between the TOC and PAHs (correlation coefficient  $< 0.4$ ) in the study area. Possible reasons for the complex relationships between PAHs and TOC include: (1) as PAH concentrations in the environment are low, the adsorption of PAHs by TOC does not reach the saturation state, hence the PAH content is mainly affected by the distance and conditions of pollution sources (e.g., degree of pollution and time of pollution), and hydrological conditions; (2) input, diversity of sources, difference in degradation degree, and PAHs released by disturbed sediments (Deng 2013).

Figure 4b shows the grain size ( $\Phi$ ) composition of sediments in ZA, Hangzhou Bay. Sediments became finer from west to east. The sediments in estuaries, such as the Qiantang River estuary, the Yongjiang estuary, and the Zhoushan coastal sea area, are relatively coarse, while sediments near the Yangtze River estuary are relatively fine (the mean value of TOC is also relatively high). The sediment along the Zhoushan coasts became coarser southward. The coarsest grain size of 1.38  $\Phi$  was identified near the Yongjiang River mouth, which had the minimum concentration of PAHs. Over

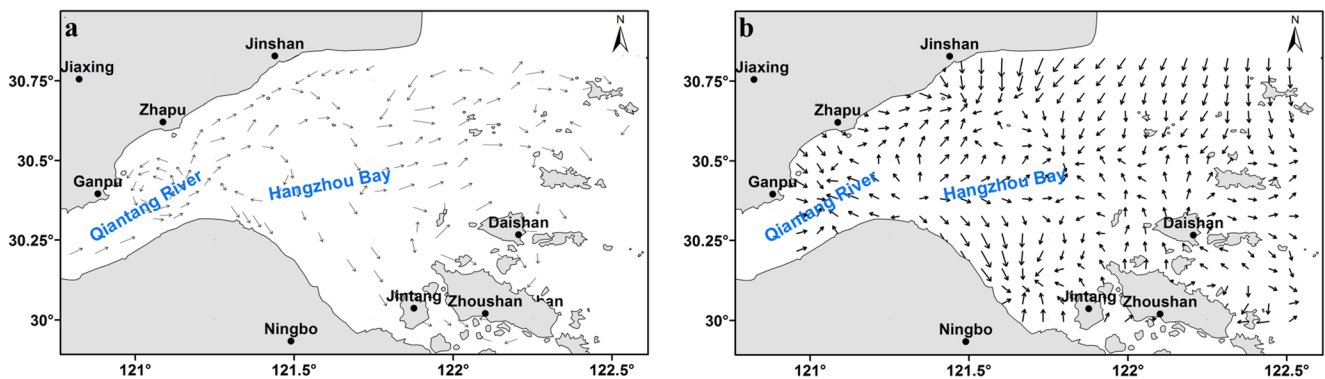
the entire study area, no significant linear correlations were found between grain size and PAH concentration or TOC, which is consistent with the results of previous studies around this area (Zhang 2011; Huang and Zhang 2015). This may be related to the distance of PAH pollution sources and the erosion/deposition caused by the strong hydrodynamic conditions (Kucklick et al. 1997; Wang et al. 2017).

#### Relationships between PAH distribution pattern and residual current or sediment transport

Previous studies have shown that PAH distribution pattern is related to regional hydrodynamic conditions, e.g., highly concentrated in the Yangtze River Estuary (YRE) and inland shelf of east China Sea (ECS) (Lin et al. 2013; Yu et al. 2015; Wang et al. 2016). The simulation results in Bohai Bay and other places show that the temporal and spatial distribution patterns of pollutants are very similar to that of the Lagrange residual current (Hu and Sun 2009). The residual current plays an important role in the accumulation of pollutants in Bohai Bay (Wang et al. 2006). The clockwise circulation in the northwest and the counterclockwise circulation in the central bay lead to the accumulation of pollutants and form a high concentration area (Wang et al. 2006; Hu and Sun 2009). High correlation was found between the pollutant transport and Lagrange residual current, as well as the distribution of suspended sediment in Zhoushan area (Shou 2009). In the macrotidal estuary, the fine-grained sediment may experience entrainment, resuspension, and sedimentation due to hydrodynamics, but the net sediment transport is controlled by the residual current (Yang 1993). Similarly, the net sediment transport in the coastal area is also determined by the near-shore residual current (Li et al. 2005; Wang and Gao 2007).

Hangzhou Bay is a macrotidal estuary with a maximum tidal range of 8.94 m. The residual current has the following characteristics (Fig. 5a). There were no obvious seasonal variations in flow direction. The residual current at the bottom was much smaller than that at the surface which was strengthened by the spreading of the Qiantang River plume. Northeast





**Fig. 5** The schematic residual current pattern (a) and the grain size trends showing the net sediment transport pattern (b)

flow was the dominate flow direction for the residual current in the Hangzhou Bay. Along the north coast, the Changjiang fresh water spreads into Hangzhou Bay (Li and Sun 1995). The magnitudes and directions of residual current were highly affected by the spread intensity of the Changjiang-diluted water (Chen et al. 2003). Although Hangzhou Bay had a general pattern of seaward directional residual current (Liu et al. 2006a, b), two circulations exist among Ganpu, Zhapu, and Jinshan where the high PAH concentrations exit (Fig. 5a). For the former circulation, residual current flowed in a counterclockwise direction, and for the latter circulation, residual current flowed in a clockwise direction. The residual currents converged at Jintang Island in ZA (Ni et al. 2003; Liu et al. 2006a, b), where the high PAH concentrations occurred. In the ZA area, the Lagrange residual current converged near Daishan (Zhu et al. 2000) that was consistent with the high PAH concentrations. The distributions of two, five, and six rings were different from that of residual current, which may be related to the PAH sources. The high concentrations of two, five, and six rings were all observed close to emission sources.

Figure 5b shows the grain size trend in the Hangzhou Bay. The vectors indicated the net transport directions of sediments, and the vector length showed the significance of grain size trend, rather than the magnitude of transport rate. The comparisons of Fig. 5a, b demonstrated that the spatial pattern of net sediment transport was consistent with that of residual current. For instance, the counterclockwise circulation pattern which was identified in the spatial pattern of residual current between Ganpu and Zhapu was also found in the spatial pattern of grain size trend in the same area, demonstrating that the residual flow in this region controlled the net transport of suspended sediment (Wang and Gao 2007). The sediment transport direction is consistent with the distribution of high PAH concentrations. The direction of residual current and sediment transport differed between Daishan and Zhoushan Islands, which may be caused by the complicated topography in the gorge (Jiang et al. 1998). In general, the sediment transport, as well as the residual current, is consistent with that of PAH distribution pattern.

## Conclusions

- (1). Sixteen of priority control PAHs were detected in the surface sediments in Hangzhou Bay, with the total PAH concentrations ranging from 35.38 to 1371.06 ng/g and the median value being 186.70 ng/g (mean 254.22 ng/g). PAH concentrations in the sediments indicated medium and low pollution levels in the study area.
- (2).  $\Sigma$ PAHs had distinct spatial distribution patterns in the study area, which could be divided into three regions. The high values mostly occurred in the Qiantang River estuary and the offshore of Zhoushan Shengsi and Daishan Island; the PAH concentrations were higher in the nearshore area than those in the offshore area. The content of phenanthrene in monomer PAH was the highest, and this area was dominated by three- to five-ring PHAs, mainly composed of high molecular weight PAHs.
- (3). The results showed that the main source of PAH pollution were coal, wood, biomass, and oil combustion, although some sites were also affected by oil leakage.
- (4). The residual current circulations in the Hangzhou Bay were consistent with the distribution of high values of PAH concentrations, e.g., the converge of residual currents was consistent with the high value of PAH concentrations in the ZA area. Using the Gao-Collins grain size trend model, it was found that the fine sediment transport was related to the residual currents, which was consistent with the PAH distribution patterns. This contribution can provide in-depth understandings of the pollutant transport processes, thus supporting the practices of pollution prevention and control in the global offshore areas.

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