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Pollution levels and risks of polycyclic aromatic hydrocarbons in surface sediments from two typical estuaries in China

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ABSTRACT

To assess the environmental risks of polycyclic aromatic hydrocarbons (PAHs), 48 and 45 sediments were collected from the Yangtze River Estuary (YRE) and Pearl River Estuary (PRE), respectively. The toxicity equivalency concentration (TEQ) in the YRE and PRE were ranged from 1.68 to 76.13 and 9.28 to 129.24 ng TEQ g^{-1}, respectively. Results of risk quotient suggest that ecological risks of two estuaries are at a moderate level, but are higher in the PRE than YRE. The increment lifetime cancer risks (ILCR) from the YRE via ingestion and dermal contact were 1×10^{-6} to 5.6×10^{-5} and 4×10^{-6} to 1.6×10^{-4}, and ranged from 7×10^{-6} to 9.4×10^{-5} and 2×10^{-5} to 2.8×10^{-4} in the PRE. ILCR results suggest that some low and moderate cancer risk exists in the YRE and PRE. Therefore, monitoring and control measures should be carried out immediately to reduce or eliminate the risks to human health from environmental exposure.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are typical persistent organic pollutants that occur in coastal marine sediments (Dudhagara et al., 2016). They have attracted much scientific and regulatory attention due to their toxic, carcinogenic, and mutagenic properties, and tendency towards bioaccumulation (Zhang and Tao, 2009; Khairy et al., 2014). Generally, PAHs may be released into the environment by anthropogenic activities such as fuel combustion, waste incineration, biomass burning, power generation plants, and other industrial processes (Chen et al., 2013; Khairy and Lohmann, 2013). Once released into the environment, PAHs can persist for a long time, undergo long-range transportation (Sun et al., 2009), and remain dangerous due to the persistent nature of their aromatic bonds. Sixteen priority PAHs have been regulated by the US EPA due to their potential adverse health effects, and seven of these have been identified as having potential carcinogenicity by the International Agency for Research on Cancer (Yang et al., 2014; Yu et al., 2015). All seven of these PAHs have molecules with 4–6 rings, which adsorb easily to the surfaces of fine particles (Li et al., 2015; Wang et al., 2016). Sediment is usually considered a major sink for PAHs due to their high hydrophobicity and lipid solubility. Therefore, coastal oceans, and especially estuaries, play an important role in the fate of PAHs because they are major reservoirs of sediments. PAHs are transported to marine ecosystems through direct and indirect pathways including atmospheric deposition and riverine inputs (Wang et al., 2007; Lin et al., 2013). Following release into the marine environment, most PAHs, particularly those with larger molecular weights, are easily adsorbed onto the surface of fine particles and are eventually deposited and accumulate in sediments. Therefore, PAHs in sediments have always been of great concern, and it is very important to routinely monitor the pollution status of sediments and to assess their potential risk to the environment.

Estuarine regions are important components of rivers and of marginal seas, which exhibit complex hydrodynamics and rich biodiversity (Yu et al., 2015). Therefore, estuarine aquatic ecosystems have been identified as the primary ecosystem resource category for study by the US EPA (Telesh, 2004). With industrial and urban development and population growth in estuarine regions, large amounts of pollutants, including PAHs, are typically discharged into estuaries and trapped with complex hydrodynamics, particularly large-river delta-front estuaries (LDEs) (Bianchi and Allison, 2009; Hung et al., 2011; Liu et al., 2012; Lin et al., 2013). LDEs are important interfaces between land and oceans for material fluxes that have global impacts on marine biogeochemistry (Bianchi and Allison, 2009). Therefore, LDEs are important sinks of PAHs from land and the atmosphere, and sediments play key roles in the transformation, migration, and accumulation of PAHs in LDEs. So far, however, studies on LDEs have mainly focused on global climate change, the marine biogeochemistry cycle, marine sedimentary

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dynamics, and estuarine ecology (Bianchi and Allison, 2009; Wu et al., 2013; Hu et al., 2012; Wang et al., 2013). In contrast, limited attention has been paid to the ecological and human health risks associated with organic pollution in LDEs. The concentration and distribution of PAHs in sediments reflects the extent of marine environmental pollution, and provides positive information for evaluating environmental health risks associated with PAHs. The Yangtze River Estuary (YRE) and Pearl River Estuary (PRE) are two typical LDEs on Chinese marginal seas. They are also two prosperous regions that receive large volumes of runoff, sediment loads, and associated PAHs from river basins. Previous studies have demonstrated that large amounts of PAHs are discharged into the YRE and PRE, making them two of the largest contributors of PAHs to the western Pacific shore. This pollutant load of PAHs will pose an increasing burden on water quality and the health of the marine coastal ecosystem (He et al., 2011; Qi et al., 2014), creating a potential adverse environmental risk to the ecosystem and to human health. It is necessary to pay more attention to comprehensive studies that integrate the ecological and human health risks from PAHs in the YRE and PRE. In addition, this study could be used as a reference for other studies about environmental risks in LDEs, and can serve as a point of comparison.

To investigate the ecological risk and carcinogenicity of PAHs, several methods have been utilized in ecosystem and human health studies. The scientifically justifiable Sediment Quality Guidelines were developed by the US National Oceanic and Atmospheric Administration to assess potential ecological risk (Long and MacDonald, 1998). Kalf et al. (1997) provided the risk quotient (RQ) method for evaluating ecological risk from sedimentary organic pollutants. The incremental lifetime cancer risk (ILCR) model has frequently been used to calculate the cancer risk to humans exposed to PAHs (Man et al., 2013; Yu et al., 2015). To date, numerous studies have investigated PAHs in the YRE and PRE (Li et al., 2012; Yu et al., 2015; Chen et al., 2006; Yuan et al., 2015), but they have focused on the sources and distributions of PAHs. The characteristics of ecological and human health risks of PAHs have not yet been systematically discussed.

In this study, we focused on two typical anthropogenically impacted LDEs in China (the YRE and PRE) to explore the ecological and human health risks in LDE systems. Surface sediments were collected from the YRE and PRE to help us understand this scientific issue. The key objectives of this study were: 1) to assess the ecological and human health risk levels of sedimentary PAHs from the YRE and PRE; 2) to explore the spatial variation of the risks in the YRE and PRE; and 3) to explore the factors (marine dynamics, sedimentary properties, and human activities) influencing this spatial variation.

2. Materials and methods

2.1. Study area and data collection

The YRE and PRE are two typical estuaries in China, which are also among the most developed and populated areas in China. In recent years, intensive industrialization and urbanization within their drainage basins has resulted in various pollutants, including PAHs, being discharged into the estuaries and adjacent coastal ocean. The Yangtze River is the longest river in Asia (6300 km), ranking fifth globally for water discharge levels and fourth for sediment loads (920 $km^3\ a^{-1}$ and 480 million t a^{-1}, respectively) (Milliman and Syvitski, 1992; Yang et al., 2006). The Yangtze River Basin is populated by 400 million inhabitants and has a drainage area of 1.8×10^6 km^2 (Yang et al., 2006). The Pearl River is the third longest river in China, and transports an average of 350 $km^3\ a^{-1}$ freshwater and 54 million t a^{-1} sediment load into the South China Sea (SCS) (Liu et al., 2014; Yuan et al., 2015). These two estuaries are key shipping hubs connecting inland areas and adjacent coastal ocean, and are also considered the most urbanized and industrialized regions in China. During the past three decades, rapid socio-economic development in the Yangtze River Delta (YRD) and Pearl River Delta (PRD) has resulted in significant environmental pollution. Contaminants discharged into estuaries and the adjacent coastal ocean through surface runoff include polychlorinated biphenyls (PCBs) (Yang et al., 2012; Sun et al., 2015), polybrominated diphenyl ethers (PBDEs) (Gao et al., 2013; Guan et al., 2009),organochlorine pesticides (OCPs) (Zhou et al., 2014; Guan et al., 2009), and PAHs (Yu et al., 2015; Wang et al., 2007); all of which may have long-term adverse effects on estuarine and marine organisms (Zhou et al., 2014; Gui et al., 2014). Previous studies have estimated that the annual input of Σ_{15}PAHs from the PRE is 33.9 t (Wang et al., 2007), and the annual input of Σ_{16}PAHs from the YRE is 369 t (Qi et al., 2014).

In this study, the YRE and PRE were selected to evaluate the ecological and human health risks posed by PAHs. Surface sediments were collected from the YRE (48 samples) and the PRE (45 samples). Sampling sites were uniformly distributed (Fig. 1). Surface sediment samples (at 0–2 cm depth) in the YRE were collected during December 2013 using stainless steel grab samplers. All samples were placed in pre-cleaned aluminum foil and stored at −20 °C prior to analysis. Data on the PRE were collected from published literature (Yuan et al., 2015).

Sixteen priority PAHs were determined. Seven of these (BaA, Chr, BaP, BbF, BkF, IcdP, and BghiP) were identified as carcinogenic (CPAHs). Detailed procedures are described by Wang et al. (2016). The concentrations of PAHs in surface sediments of the PRE were adapted from Yuan et al. (2015).

2.2. Ecological risk assessment

PAHs accumulated in marine sediments can be ingested by benthic organisms and thereby enter the food web, posing a potential risk to aquatic ecosystems. Ecological risk assessment is a useful tool for characterization of PAH risks to organisms and ecosystems, and has been widely used. The risk quotient (RQ) is a popular ecological risk assessment method, proposed by Kalf et al. (1997) and modified by Cao et al. (2010). In order to assess the potential ecological risk of PAHs in the YRE, RQ were used to evaluate the levels of risk posed by PAHs, as follows (Cao et al., 2010).

$$RQ = C_{PAH}/C_{QV} \tag{1}$$

where C_{PAH} is the concentration of a certain PAH in the sediment sample; and C_{QV} is the corresponding quality value of the PAH. The negligible concentrations (NCs) and the maximum permissible concentrations (MPCs) of PAHs (Table 2) in sediment referenced by Kalf et al. (1997) and Cao et al. (2010) were used as quality values for the samples. RQ_{NCs} and RQ_{MPCs} were defined as follows:

$$RQ_{NCs} = C_{PAH}/C_{QV(NCs)} \tag{2}$$

$$RQ_{MPCs} = C_{PAH}/C_{QV(MPCs)} \tag{3}$$

where $C_{QV(NCs)}$ and $C_{QV(MPCs)}$ are the quality values of NCs and MPCs of the PAHs in the samples. Furthermore, $RQ_{\sum PAHs}$, $RQ_{\sum PAHs(NCs)}$, and $RQ_{\sum PAHs(MPCs)}$ for ΣPAHs have been defined as follows:

$$RQ_{\sum PAHs} = \sum_{i=1}^{16} RQ_i \quad (RQ_i \geq 1) \tag{4}$$

$$RQ_{\sum PAHs(NCs)} = \sum_{i=1}^{16} RQ_{(NCs)} \quad \left(RQ_{(NCs)} \geq 1\right) \tag{5}$$

$$RQ_{\sum PAHs(MPCs)} = \sum_{i=1}^{16} RQ_{(MPCs)} \quad \left(RQ_{(MPCs)} \geq 1\right) \tag{6}$$

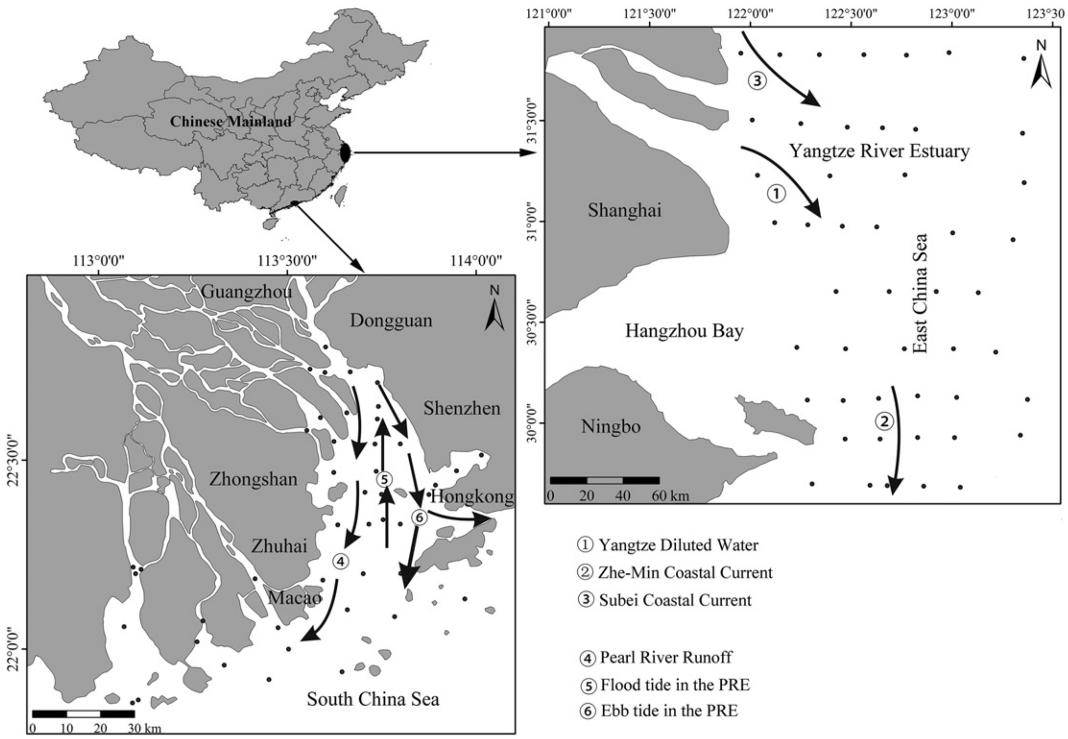

Fig. 1. Map of sampling sites in the YRE and PRE.

The RQ_{NCs} and RQ_{MPCs} of individual PAHs that were not <1 were summed to calculate the $RQ_{\sum PAHs(NCs)}$ and $RQ_{\sum PAHs(MPCs)}$ of PAHs (Cao et al., 2010; Dudhagara et al., 2016).

2.3. Cancer risk assessment

Two major exposure pathways that could impose cancer risk on fishermen or other coastal residents as a result of coming into contact with contaminated sediment were taken into consideration: accidental ingestion of sediment particles, and dermal absorption of PAHs via contact with sediment particles. The incremental lifetime cancer risk (ILCR) of PAHs is evaluated according to the toxic concentration of total PAHs. The USEPA (2004) proposed relevant equations to calculate exposure through these two pathways as follows:

$$ILCR_{ingest} = \frac{C_{sed} \times IngR \times EF \times ED}{BW \times AT} \times CF \times SF \quad (7)$$

$$ILCR_{dermal} = \frac{C_{sed} \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF \times SF \quad (8)$$

where $ILCR_{ingest}$ and $ILCR_{dermal}$ are the cancer risk from ingestion of, and dermal contact with, sediments; C_{sed} is the concentration equivalent to BaP toxicity in the sediments; IngR is the ingestion rate (100 mg day^{-1}); CF is the unit conversion factor (10^{-6} kg mg^{-1}); EF is the exposure frequency (350 days a^{-1}); ED is the exposure duration (70 years for adults); BW is body weight (70 kg for adults); AT is the average day (25,550 days); SF is the cancer slope factor of BaP (7.3 mg kg^{-1} day^{-1} for ingestion exposure and 25 mg kg^{-1} day^{-1} for dermal contact) (Knafla et al., 2006; Yu et al., 2015); and SA is the exposed skin surface area (3300 cm^2 for adults) (Man et al., 2013). Here, AF is the adherence factor of the sediment to skin (0.2 mg cm^{-2}) (US EPA, 1997); and *ABS* is the dermal absorption from the sediment (0.13).

The potential toxicity of sediments was determined based on their toxic equivalency factor (TEF), which is the most popular and widely accepted method (Dudhagara et al., 2016). Toxicity equivalency concentration (TEQ) is the product of the concentration of PAHs and their corresponding TEF values (Chen et al., 2013). The TEQ of all PAHs was evaluated using the following equation:

$$TEQ = \sum C_i \times TEF_i \quad (9)$$

where C_i is the concentration of an individual PAH, and TEF_i is the toxic equivalency factor of the PAH. According to the above, TEFs are based on the potency of individual PAHs relative to that of BaP, considered to be one of the most potent carcinogens in the PAH group (Peng et al., 2011; Yu et al., 2015). A relative value of 1 was assigned to BaP. We assigned TEF reference values for the 16 PAHs referred to by Peng et al. (2011) and Chen et al. (2013).

3. Results and discussion

3.1. Levels of ΣPAHs, CPAHs, and potential toxicity in the YRE and PRE

Detailed discussions on the concentration and distribution of PAHs in surface sediments from the YRE and PRE can be found in previous studies (Wang et al., 2016; Yuan et al., 2015). Briefly, the total concentrations of sixteen PAHs (ΣPAHs) in the YRE range from 27.20 to 620.94 ng g^{-1}, with an average value of 158.18 ng g^{-1}. Spatially, the ΣPAHs exhibit an increasing trend from north to south. The concentration of low molecular weight PAHs (LMW PAHs, with 2–3 rings) ranges from 7.71 to 185.16 ng g^{-1} (Fig. 2a) and accounts for 9.2–69.6% of ΣPAHs, with an average value of 37.7%. High molecular weight PAHs (HMW PAHs, with 4–6 rings) have a relatively high concentration (12.97–435.85 ng g^{-1}) (Fig. 2c) compared to LMW PAHs, and account for 30.3–89% of ΣPAHs, with an average value of 62%. Comparatively, the PRE has relatively higher ΣPAHs concentration than the YRE; ranging from 164.18 to 1202.21 ng g^{-1} with a mean value of 432.50 ng g^{-1}. The spatial distribution of ΣPAHs in the PRE displays a clear decreasing trend from the estuary to the coastal ocean. In addition, ΣPAHs concentration near the coast is higher than in the central area of the estuary, especially for the Dongguan coast and Shenzhen Bay. The concentration of LMW and HMW PAHs in the PRE ranges from 104.75 to 641.29 ng g^{-1} and 46.44 to 721.75 ng g^{-1}, respectively. The

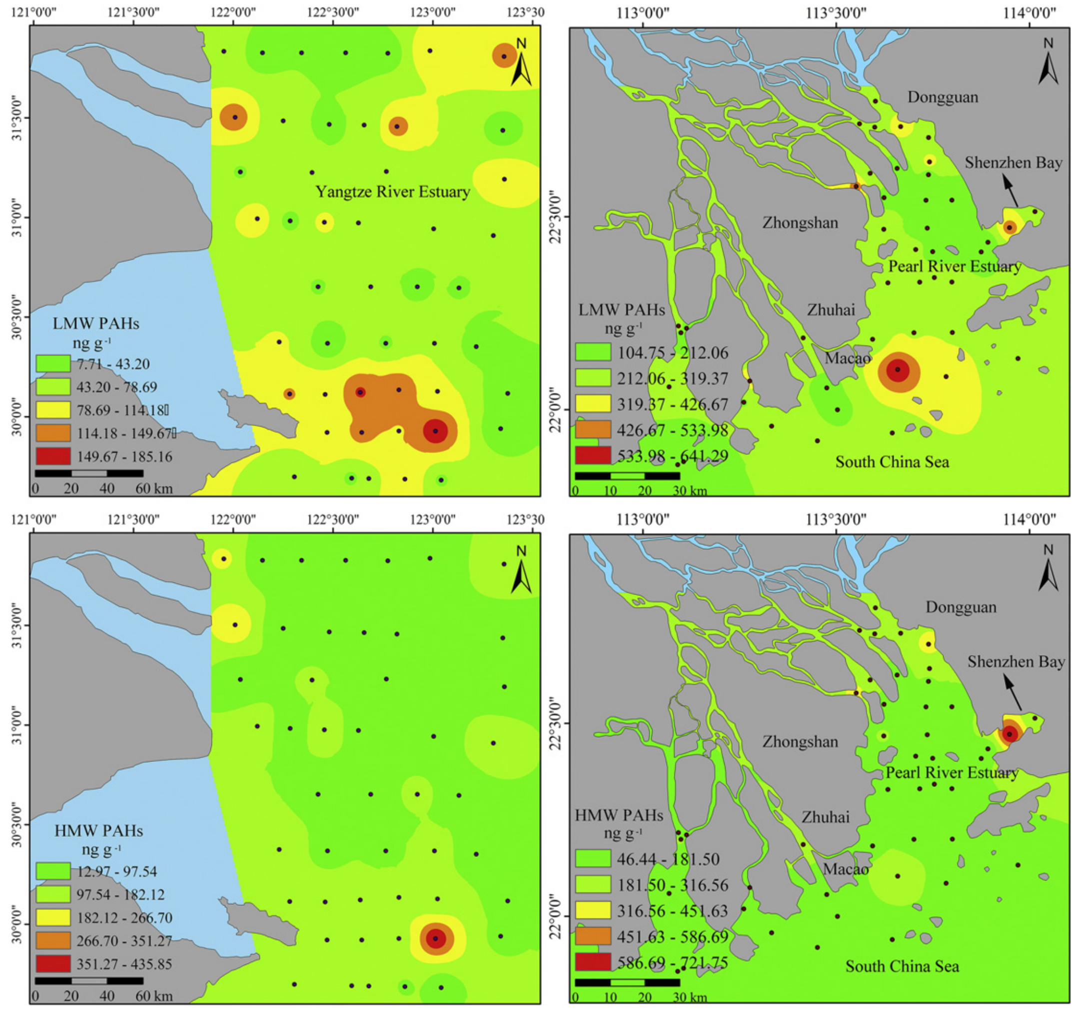

Fig. 2. Distribution patterns of LMW PAHs and HMW PAHs in the YRE and PRE.

percentage composition of LMW and HMW PAHs in the PRE contrasts with their concentrations in the YRE. LMW PAHs in the PRE account for 37.29–80.99% of ΣPAHs, with an average value of 59.64%. In comparison, the content of HMW PAHs ranges from 19.01 to 62.71% of ΣPAHs, with a mean value of 40.36%. The content of LMW PAHs is higher than HMW PAHs in the PRE, showing a different distribution pattern to that found in the YRE. Fig. 2b and d shows the distribution of LMW and HMW PAHs in the PRE, and indicates that both types of PAHs have a relatively high concentration in the inner estuary, but LMW PAHs also have high levels in the sediment below open water.

Previous studies have tended to focus on CPAHs that have been demonstrated to have higher potential carcinogenicity (Chen et al.,

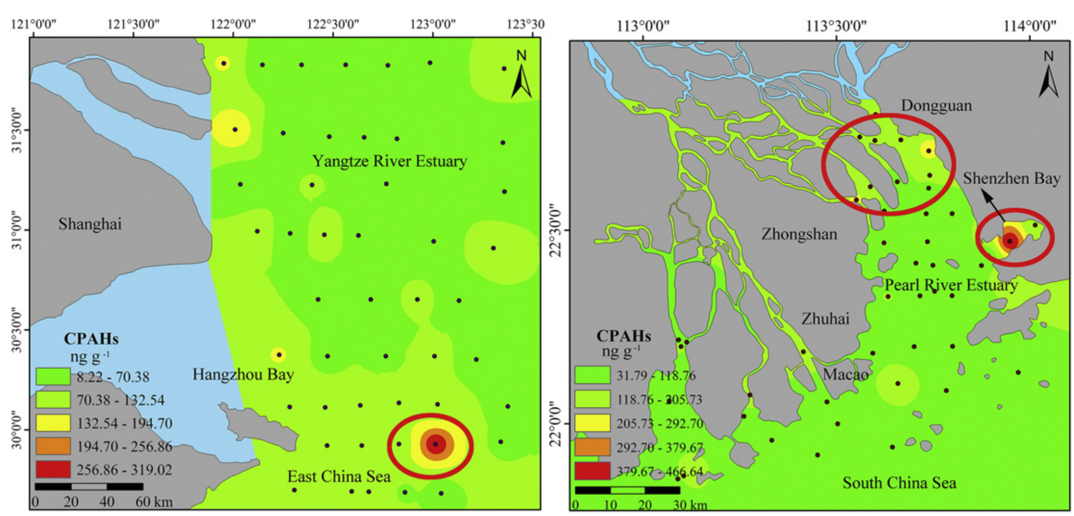

Fig. 3. Distribution patterns of CPAHs in the YRE and PRE.

Table 1
Mean and standard deviations of concentrations and TEQ of individual PAHs in the YRE and PRE.

PAH	Concentration (YRE) / ng g^{-1}	Concentration (PRE) / ng g^{-1}	TEF	TEQ (YRE) / ng g^{-1}	TEQ (PRE) / ng g^{-1}
Nap	17.31 ± 18.25	115.10 ± 52.74	0.001	0.02 ± 0.02	0.12 ± 0.05
Acy	1.12 ± 1.39	5.37 ± 6.94	0.001	NA	0.01 ± 0.01
Ace	1.77 ± 3.62	6.52 ± 4.44	0.001	NA	NA
Flu	6.44 ± 4.97	15.27 ± 9.06	0.001	NA	0.02 ± 0.01
Phe	17.45 ± 15.42	53.21 ± 27.36	0.001	0.02 ± 0.01	0.05 ± 0.03
Ant	2.39 ± 1.74	17.23 ± 9.97	0.01	0.02 ± 0.02	0.017 ± 0.10
Flo	13.05 ± 9.47	39.31 ± 30.68	0.001	0.01 ± 0.01	0.04 ± 0.03
Pyr	12.04 ± 8.92	48.44 ± 32.37	0.001	0.01 ± 0.01	0.05 ± 0.03
BaA	5.06 ± 5.87	9.41 ± 12.29	0.1	0.48 ± 0.51	0.94 ± 1.22
Chr	11.31 ± 7.58	27.46 ± 19.18	0.001	0.01 ± 0.01	0.03 ± 0.02
BbF	18.98 ± 15.06	23.09 ± 16.73	0.1	1.84 ± 1.35	2.31 ± 1.69
BkF	5.42 ± 4.88	14.37 ± 13.06	0.01	0.05 ± 0.05	0.14 ± 0.13
BaP	9.32 ± 12.16	20.00 ± 16.19	1	8.62 ± 10.07	20.00 ± 16.25
IcdP	18.60 ± 17.21	17.31 ± 14.31	0.1	1.78 ± 1.51	1.73 ± 1.44
DahA	3.32 ± 6.82	7.89 ± 4.25	1	2.94 ± 5.81	7.89 ± 4.33
BghiP	16.28 ± 13.55	12.53 ± 13.13	0.01	0.16 ± 0.12	0.13 ± 0.13
ΣPAHs	158.2 ± 101.11	432.50 ± 198.84			
ΣTEQ	15.98 ± 14.52	32.95 ± 21.18			

NA: not analyzed due to extremely low TEQ values.

2013; Devi et al., 2016). Therefore, the concentrations of seven CPAHs (ΣCPAHs) in surface sediments from the YRE and PRE are also discussed in this study. The ΣCPAHs concentration in the YRE varies from 8.22 to 319.02 ng g^{-1} with a mean concentration of 71.30 ng g^{-1} (Fig. 3a) and displays an increasing trend from north to south, consistent with the distribution patterns of ΣPAHs. The ΣCPAHs account for 19.6 to 65.8% of ΣPAHs in the surface sediment of the YRE. Fig. 3b shows the distribution of ΣCPAHs in the PRE, the concentration of which ranges from 31.79 to 466.64 ng g^{-1} with a mean value of 125.06 ng g^{-1}. ΣCPAHs account for 13.01 to 55.53% of ΣPAHs, a little lower than in the YRE. The distribution of CPAHs in the PRE shows a decreasing trend with increasing distance from the estuary towards the open sea, which is consistent with the distribution patterns of HMW PAHs. The CPAHs were mainly distributed in the inner estuary and Shenzhen Bay, and may have originated mainly from surface runoff. Their distribution is likely to be controlled by regional marine dynamics (runoff and tides).

TEQ was calculated for 16 PAHs to assess the potential toxicological risk of PAHs in surface sediments from the YRE and PRE (Table 1). The TEQ values of sediment collected from the YRE ranged from 1.68 to 76.13 ng TEQ g^{-1} with an average of 15.98 ng TEQ g^{-1}. This is consistent with results obtained by Yu et al. (2015). The TEQ values of sediment in the PRE ranged from 9.28 to 129.24 ng TEQ g^{-1} with an average of 32.95 ng TEQ g^{-1}. Table 1 shows that BaP is considered the most carcinogenic of the PAHs, followed by DahA, BbF, and IcdP. The distribution of TEQ in the surface sediments of the YRE and PRE are shown in Fig. 4. Overall, the distribution of TEQ in the YRE displays an increasing trend from north to south, with higher TEQ values occurring outside Hangzhou Bay, which is the center of sediment deposition from the YRE (Liu et al., 2007). The TEQ value of sediment from the PRE was high near the coast, especially in the inner estuary and Shenzhen Bay. To a certain extent, human activities could be the controlling factor in the pollution status of this area. The PRD is one of the most prosperous regions in China, and the Pearl River is the third largest river in China and discharges into the northern part of the South China Sea (Liu et al., 2014). Intensive urbanization and industrialization in the Pearl River Delta has led to the discharge of large amounts of PAHs into the estuary (Wang et al., 2007). The semi-enclosed nature of the bay has resulted in the trapping of large amounts of PAHs in the inner estuary and bay, causing high TEQ levels.

3.2. Environmental risks of PAHs from the YRE and PRE

3.2.1. Ecological risk assessment

We evaluated the ecological risks of PAHs in the YRE and PRE using the RQ method. The NCs and MPCs of individual PAHs, and the mean values of RQ_{NCs} and RQ_{MPCs} of each chemical, are listed in Table 2,

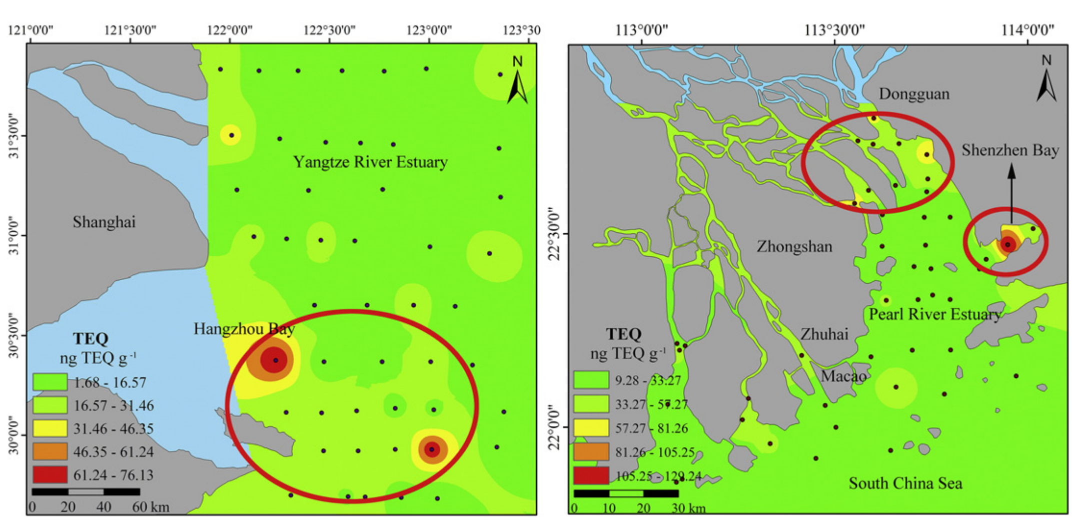

Fig. 4. Distribution patterns of TEQ value in the YRE and PRE.

Table 2
Mean values of RQ_{NCs} and RQ_{MPCs} of individual PAHs in surface sediments of the YRE and PRE.

PAHs	NCs	MPCs	YRE		PRE	
			RQ_{NCs}	RQ_{MPCs}	RQ_{NCs}	RQ_{MPCs}
Nap	1.4	140	12.01	0.12	82.21	0.82
Ace	1.2	120	0.89	0.01	4.48	0.04
Acy	1.2	120	1.26	0.01	5.44	0.05
Flo	1.2	120	5.31	0.05	12.73	0.13
Phe	5.1	510	3.36	0.03	10.43	0.10
Ant	1.2	120	1.96	0.02	14.35	0.14
Flu	26	2600	0.49	0.00	1.51	0.02
Pyr	1.2	120	9.80	0.10	40.36	0.40
BaA	3.6	360	1.33	0.01	2.61	0.03
Chr	107	10,700	0.10	0.00	0.26	0.00
BbF	3.6	360	5.12	0.05	6.41	0.06
BkF	24	2400	0.22	0.00	0.60	0.01
BaP	27	2700	0.32	0.00	0.74	0.01
IcdP	59	5900	0.30	0.00	0.29	0.00
DahA	27	2700	0.11	0.00	0.29	0.00
B[*ghi*]P	75	7500	0.21	0.00	0.17	0.00

which presents the overall ecological risk posed by individual PAHs in the YRE and PRE. According to the equations shown previously, $RQ_{NCs} < 1$ indicates a PAH that is probably of negligible concern. For both RQ_{NCs} and RQ_{MPCs}, values >1 for an individual PAH indicate that risk is high and remedial action must be undertaken immediately. When RQ_{NCs} is >1 and RQ_{MPCs} is <1, contamination by a PAH might be considered intermediate. In Table 2, we see that almost all the LMW PAHs (except Ace) in the YRE and PRE have high RQ_{NCs} values, as do some HMW PAHs (such as Pyr, BaA, and BbF). However, the mean RQ_{MPCs} value for all chemicals was less than one, which indicates no severe risk in the study areas. Therefore, LMW PAHs contributed to the burden of risk for the main ecosystem, and monitoring and control of LMW PAHs should be taken into consideration in the study areas. In addition, Table 2 also shows that the mean value of RQ_{NCs} in the PRE is much higher than that in the YRE, which means the ecological risk of PAHs in the PRE is higher. The mean RQ_{MPCs} values in the YRE and PRE are both less than one, but the RQ_{MPCs} value of Nap (0.82) in the PRE is closer to one, which may indicate a potentially severe risk.

We also calculated the percentage of stations falling within each ecological risk classification (Table 3). Generally, moderate risk was the most common risk classification in the YRE and PRE. Table 3 shows that the RQ_{NCs} values for Nap, Flo, Phe, Ant, Pyr, and BbF from the YRE exceed 1 at >60% of sampling stations, which indicates that most of these stations present a moderate ecological risk. Comparatively, the

Table 3
Percentage of stations in each ecological risk classification for individual PAHs.

Individual PAHs	$RQ_{NCs} < 1$		$RQ_{NCs} > 1; RQ_{MPCs} < 1$		$RQ_{MPCs} > 1$	
	YRE	PRE	YRE	PRE	YRE	PRE
Nap	32.47%	0.00%	67.53%	73.33%	0.00%	26.67%
Ace	74.03%	0.00%	25.97%	100.00%	0.00%	0.00%
Acy	62.34%	2.30%	37.66%	97.70%	0.00%	0.00%
Flo	11.69%	0.00%	88.31%	100.00%	0.00%	0.00%
Phe	22.08%	0.00%	77.92%	100.00%	0.00%	0.00%
Ant	32.47%	0.00%	67.53%	100.00%	0.00%	0.00%
Flu	96.10%	26.67%	3.90%	73.33%	0.00%	0.00%
Pyr	0.00%	0.00%	100%	100.00%	0.00%	0.00%
BaA	51.95%	26.67%	48.05%	73.33%	0.00%	0.00%
Chr	100.00%	100.00%	0%	0.00%	0.00%	0.00%
BbF	6.49%	0.00%	93.51%	100.00%	0.00%	0.00%
BkF	100.00%	86.36%	0%	13.64%	0.00%	0.00%
BaP	97.40%	75.56%	2.60%	24.44%	0.00%	0.00%
IcdP	98.70%	97.70%	1.30%	2.30%	0.00%	0.00%
DahA	97.40%	100.00%	2.60%	0.00%	0.00%	0.00%
B[*ghi*]P	100.00%	100.00%	0.00%	0.00%	0.00%	0.00%

Graphical abstract

situation is more severe in the PRE because most individual PAHs have RQ_{NCs} values that exceed 1, except Chr, BkF, BaP, IcdP, DahA, and B[*ghi*]P. In addition, the RQ_{NCs} values for Nap, Ace, Flo, Phe, Ant, Pyr, and BbF from the PRE exceed 1 at all sampling stations, which indicates an obvious ecological risk in this area. Table 3 also shows that approximately 26.67% of sampling stations have RQ_{MPCs} values that exceed 1, which indicates a severe risk at these stations. Emergency measures should be carried out to control the ecological risk. Combining the contents of Tables 2 and 3 shows that almost all of the LMW PAHs present relatively high ecological risks in the YRE and PRE.

The values of $RQ_{\sum NCs}$ and $RQ_{\sum MPCs}$ were also used to assess the ecological risk of ΣPAHs, which was demonstrated to be more accurate and scientific (Cao et al., 2010). In addition, a new ecological risk classification of ΣPAHs was proposed based on this method. In principle, if $RQ_{\sum NCs} = 0$, then no ecological risk exists in this area. If $1 \le RQ_{\sum NCs} < 800$ indicates a low-level risk in the study area, then there are two moderate risk levels via this method (Cao et al., 2010). These are "moderate risk" with $RQ_{\sum NCs} \ge 800$, $RQ_{\sum MPCs} = 0$; and "moderate risk" with $RQ_{\sum NCs} < 800$, $RQ_{\sum MPCs} \ge 1$. If $RQ_{\sum NCs} \ge 800$ and $RQ_{\sum MPCs} \ge 1$, there is a high risk in the study area. The results of $RQ_{\sum NCs}$ in surface sediments from the YRE and PRE are shown in Fig. 5. The $RQ_{\sum NCs}$ values in the YRE and PRE range from 6.83 to 138.30, and 71.94 to 490.99, respectively, which indicates that the ecological risk in the PRE is higher than that in the YRE. Spatially, the ecological risk in the YRE shows an increasing trend from north to south, which may be caused by regional hydrodynamics (Wang et al., 2016). Previous studies have demonstrated that a muddy area of finer-grained sediments has formed beyond Hangzhou Bay (Liu et al., 2007; Lin et al., 2013). Therefore, the concentration of PAHs is very high in this area (Wang et al., 2016), resulting in high $RQ_{\sum NCs}$ values. The value of $RQ_{\sum NCs}$ in the PRE is relatively high, which may lead to a series of ecological risks. The high $RQ_{\sum NCs}$ values were distributed near the coast, especially in Shenzhen Bay, the water quality of which may be controlled by surface runoff in this area. Overall, the $RQ_{\sum NCs}$ values in the YRE and PRE are all <800, which indicates that the ecological risks in these two estuaries are at a moderate level. Combining information in Tables 2 and 3, and Fig. 5, we found that all RQ_{NCs} and $RQ_{\sum NCs}$ in the YRE and PRE indicated ecological risk to a certain extent. In addition, the distribution patterns of ecological risk are more similar to the distribution patterns of PAHs, demonstrating that high levels of ecological risk occur in the PAH deposition center (Fig. 5). Therefore, monitoring and control measures should be carried out immediately to reduce or eliminate these ecological risks.

3.2.2. Human health risk assessment

USEPA (2004) explained that a one in a million chance of additional cancer over a lifetime (70 years) is the level of risk considered acceptable or inconsequential, a risk similar to that from diagnostic X-rays or fishing (Asante-Duah, 2002). Nevertheless, additional cancer risk of one in ten thousand or greater should be considered serious (Xia et al., 2013), and requires attention. The objective of this risk assessment was to explore the potential for cancer occurrence among local residents in the YRE and PRE as a result of exposure to PAHs via ingestion and dermal contact. The ILCR indices for ingestion and dermal exposure were calculated to indicate carcinogenic risk intensity. Qualitative descriptions of cancer risks via ILCR are as follows: $ILCR \le 10^{-6}$ indicates a very low risk; $10^{-6} < ILCR < 10^{-4}$ indicates low risk; moderate risk is indicated by an ILCR ranging from 10^{-4} to 10^{-3}; while $10^{-3} \le ILCR < 10^{-1}$ shows high risk. If the ILCR is $>10^{-1}$, the risk level is very high (Man et al., 2013).

The $ILCR_{ingest}$ and $ILCR_{dermal}$ values are presented in Fig. 6, which shows the carcinogenic risk distribution of sediments in the YRE and PRE. In Fig. 6, the $ILCR_{dermal}$ is larger than $ILCR_{ingest}$ in both the YRE and PRE, which means the cancer risk via dermal contact is more serious than via ingestion. The values of $ILCR_{ingest}$ and $ILCR_{dermal}$ in the YRE range from 1×10^{-6} to 5.6×10^{-5} and 4×10^{-6} to 1.6×10^{-4},

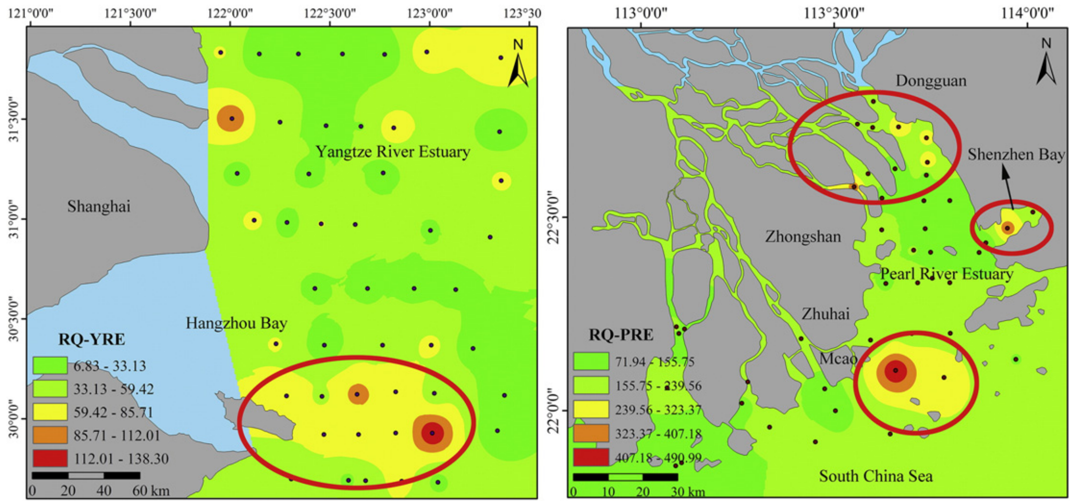

Fig. 5. Distribution patterns of RQ value in the YRE and PRE.

respectively. Therefore, cancer risk via ingestion is low at all sampling sites in the YRE, but at some sites low or even moderate risk exists if dermal contact occurs. Spatially, both $ILCR_{ingest}$ and $ILCR_{dermal}$ exhibit an increasing trend from north to south, which is consistent with the distribution of CPAHs and TEQ. The values of $ILCR_{ingest}$ and $ILCR_{dermal}$ in the PRE range from 7×10^{-6} to 9.4×10^{-5} and 2×10^{-5} to 2.8×10^{-4}, respectively. Overall, the cancer risks via ingestion and dermal contact in the PRE are relatively higher than in the YRE. A moderate risk level via dermal contact is present in Shenzhen Bay ($ILCR_{dermal}$ value of 2.8×10^{-4}). Fig. 6 shows a distinct difference in ILCR between the east and west parts of the PRE; ILCR in the east is much higher than in the west. The east estuary is the concourse of the Beijing, Dongjiang,

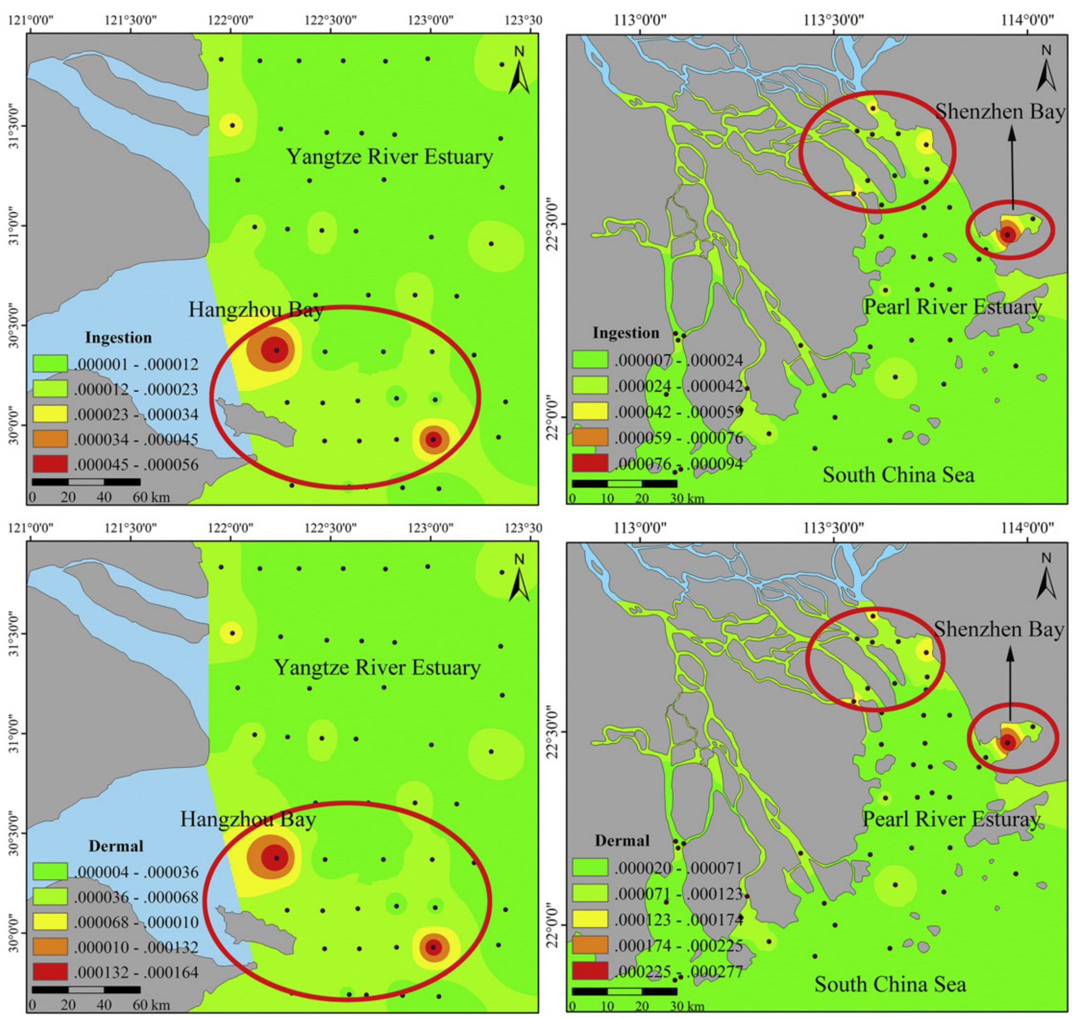

Fig. 6. Distribution patterns of ILCR value ($ILCR_{ingest}$ and $ILCR_{dermal}$) in the YRE and PRE.

and Pearl Rivers, and its catchment includes prosperous cities such as Guangzhou, Dongguan, and Foshan. In comparison with the east estuary, the catchment of Xijiang is largely underdeveloped, and therefore contains limited PAHs.

3.3. Factors influencing environmental risks of PAHs

In conclusion, ΣPAHs, CPAHs, TEQ, RQ, and ILCR show similar distributions in the YRE and PRE. In the YRE, all five indices display increasing trends from north to south and high values are observed outside Hangzhou Bay. This distribution pattern is controlled by relatively complicated factors that are a result of the synergy of marine dynamics, sedimentary properties, and anthropogenic activities. Fig. 1b shows the regional hydrodynamics in the YRE, indicating that Yangtze dilute water (YDW, Yangtze River runoff) and the Zhe-Min Coastal Current (ZMCC) are the main marine dynamics controlling the distribution patterns of PAHs. Yangtze-derived sediments and associated PAHs are discharged into the East China Sea with YDW and are deposited in the estuary. Previous studies have demonstrated that sediments in the YRE are re-suspended and transported southward under the influence of ZMCC (Liu et al., 2007; Lin et al., 2013). Therefore, PAHs associated with sediments are transported southward; this results in an increasing trend of PAHs from north to south. In addition, the distribution patterns of PAHs are also controlled by the grain-size of sediments due to the physicochemical properties of PAHs (Li et al., 2015). Our previous study demonstrated that the distribution patterns of sediment grain-size in the study area showed a trend towards increasingly fine from north to south, which was consistent with the distribution patterns of PAHs (Wang et al., 2016). The results of our study show that the highest environmental risk of PAHs in the YRE occurred outside Hangzhou Bay. Previous studies on sedimentary properties in the YRE have demonstrated that this area is a deposition center, with the fastest deposition rate in the estuary (Liu et al., 2007; Wang et al., 2016). Therefore, large amounts of sediment-associated PAHs are deposited in this area, which results in relatively higher environmental risks.

In the PRE, TEQ, RQ, and ILCR are mainly concentrated in the inner estuary and Shenzhen Bay. The distribution patterns of PAH concentrations and environmental risks show a similar decreasing trend with distance from the inner estuary to the open sea. Therefore, human activities and regional hydrodynamics are the main influencing factors and have resulted in the observed distribution patterns of PAHs and their environmental risks. In addition, all of these indexes were calculated based on ΣPAHs, which indicated the internal relations between each index and ΣPAHs. The Pearl River Delta is one of the most prosperous regions in China, and the Pearl River is the third largest river in China and discharges into the north part of the South China Sea (Liu et al., 2014). Intensive urbanization and industrialization in the Pearl River Delta led to the discharge of large amounts of PAHs into the estuary (Wang et al., 2007). In addition, Zhang et al. (2013) determined the net surficial sediment transport pattern in the PRE through the grain size trend analysis model. The results show that the surficial sediment of the PRE is controlled by complicated factors including complex delta formation, bottom topography, and the interaction between runoff and the tide (Zhang et al., 2013). In the inner estuary, the transport of surficial sediment is mainly controlled by river runoff, and the vectors are dominated by the southeastward transport trend, which results in decreased PAHs and environmental risks from the inner estuary to the open sea. However, in the central and outer part of the PRE, PAHs and associated environmental risks show relatively lower levels, which may be the result of a dilution effect. Once the higher concentrations of PAHs are discharged into open water, they are diminished by dilution. Therefore, the concentrations and environmental risk levels of PAHs are relatively lower in the outer than the inner estuary. In addition, high PAH concentrations and risk levels also occur in Shenzhen Bay, which may be related to anthropogenic activities and regional topography. First, Shenzhen Bay is a semi-enclosed bay surrounded by the major cities of Shenzhen and Hong Kong, leading to irregular water exchange. Second, Shenzhen and Hong Kong are two of the most prosperous cities in China, and they discharge large amounts of industrial effluent and domestic sewage into Shenzhen Bay. This has led to serious ecological and human health issues. In addition, there are millions of cars in this area that emit large amounts of exhaust, the particulates of which are washed into the sea by precipitation and eventually deposited in the sediments. Therefore, Shenzhen Bay is a convergent center of PAHs from Shenzhen and Hong Kong, and high levels of environmental risk are expected to occur in this area. Overall, the distribution patterns of PAHs and environmental risks in the YRE and PRE are mainly controlled by regional marine dynamics, sedimentary properties, human activities, and regional topography.

4. Conclusions

A comprehensive study of 16 high-priority PAHs in surface sediments from the YRE and PRE was conducted to assess the related ecological and human health risks. Information about 16 priority PAHs of the YRE and PRE were collected from previous studies. TEQ, RQ, and ILCR methods were used to evaluate ecological and human health risks. The levels of TEQ indicate that the main pollutants in the YRE and PRE are HMW PAHs. BaP was considered the worst of the carcinogenic PAHs, followed by DahA, BbF, and IcdP. The value of TEQ in the PRE (ranging from 9.28 to 129.24 ng TEQ g^{-1}) is relatively higher than in the YRE (ranging from 1.68 to 76.13 ng TEQ g^{-1}). The RQ method was used to evaluate the ecological risk of PAHs in the YRE and PRE. The results of the RQ suggest that the main ecological risk is caused by LMW PAHs such as Nap, Ace, Flo, Phe, Ant, Pyr, and BbF. All the sampling stations in the YRE and PRE were categorized as low risk and even most of them were located in a moderate risk level. In addition, the ILCR method was carried out to assess the level of risk to human health. The ILCR results indicate that the overall situation regarding cancer risk levels in the YRE and PRE is relatively optimistic. Most stations displayed low risk levels, with ILCR values within the limits of 10^{-6} to 10^{-4}. However, some stations had a relatively high ILCR value exceeding 10^{-4}, which suggests a moderate risk level. The distribution patterns of ecological and human health risks show very similar trends; all had high levels in the centers of PAH deposition. Regional hydrodynamics, sedimentary features, and regional topography are the main factors influencing the distribution patterns of ecological and human health risks. Overall, the TEQ, RQ, and ILCR results suggest that a certain degree of ecological and human health risks (low and moderate risk levels) exist in the YRE and PRE. Therefore, monitoring and control measures should be carried out immediately to reduce or eliminate the ecological risks.

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