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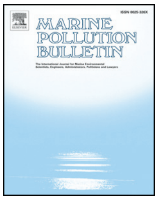

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Risk assessment and uncertainty analysis of PAHs in the sediments of the Yangtze River Estuary, China

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ABSTRACT

To better explore the concentration of polycyclic aromatic hydrocarbons (PAHs) in the sediments of the Yangtze River Estuary (YRE), 16 priority PAHs were analyzed based on sampling data obtained in February 2011. The results showed that the total concentrations of PAHs in sediments of the YRE varied from 65.07 to 668.98 $ng \cdot g^{-1}$. The results of toxic equivalent quantities of benzo[*a*]pyrene and the sediment quality guideline quotient suggested that PAHs had little or no adverse effects on the environment. The cancer risk results showed that the cancer risk at all sites exceeded 10^{-6}, with 73% of sites exceeding 10^{-4}, suggesting that people remain at risk of cancer as a result of their exposure to carcinogenic PAHs. However, the result of hazard index results showed that the non-cancer risks were substantially lower than one, indicating that PAHs in these sediments likely pose little or no adverse health threats to local inhabitants.

Polycyclic aromatic hydrocarbons (PAHs) are a typical group of organic compounds with two or more benzene rings. PAHs can be introduced into the environment from both natural and anthropogenic sources (Li et al., 2015). Because of their carcinogenicity, teratogenicity, mutagenicity and toxicity, when PAHs are released into the environment they pose a threat to local inhabitants (Bixian et al., 2001). In addition, PAHs can be bio-accumulated through the food chain, and the exposure of humans to PAHs may enhance the risk of cancer and cause other adverse health effects (Wan et al., 2007; Gu et al., 2013). Of the 129 types of “priority pollutants” reported by the United States Environmental Protection Agency (US EPA) in 1976, 16 types of PAHs have been listed. For decades, a high concentration of PAHs has been present in the environment, especially in some lakes, rivers and estuaries, and also in the atmosphere (Jiang et al., 2007; Hiller et al., 2011; Keshavarzifard et al., 2014). The distribution and toxicity of PAHs have caused wide concern.

Suspended solids and atmospheric aerosols are the main carriers of PAHs that enter waterbodies. Owing to their low water solubility, low volatility, and high persistence, PAHs in water systems tend to accumulate in sediments, resulting in long-term effects on benthic organisms (Liu et al., 2013). After being gathered in sediments, it is difficult for PAHs to decompose via photochemical degradation or microbial oxidation (Liu et al., 2000). As a result, sediments serve as a major reservoir for PAH contamination (Scheibye et al., 2014; Liu et al., 2015). Humans can be directly or indirectly exposed to PAHs in sediments, and so studies of PAH distributions in sediments are urgently needed (Boonyatumanond et al., 2006; Wang et al., 2015).

Estuaries, as the key interface between the land and ocean, represent a filter of materials from river inputs to coastal waters (Zhao et al., 2015). Approximately 60% of the world's population lives along estuaries and the coast (Kathiresan and Rajendran, 2005). Many pollutants created by human activities are deposited in estuaries, leading to the environmental degradation of many of these estuaries. Moreover, because pollutants can easily enter the water, are prone to enrichment and are retained for a long time in sediments, these sediments generally retain most of the pollutants that enter an estuary (Spasojević et al., 2015; Yu et al., 2015). In addition, estuary environments are highly dynamic, with freshwater and seawater entering from opposite directions, making the distribution of pollutants unpredictable (Zhao et al., 2015).

In order to obtain evidence in support of PAH pollution control in the sediments of estuaries, some methods have been developed to assess the level of PAH pollution. The toxic equivalent quantities of benzo[*a*]pyrene (TEQ_{Bap}) have been widely used to evaluate the potential toxicity of sedimentary PAHs (Dong et al., 2014). The carcinogenic potency of a sample can be calculated in terms of the BaP equivalent concentration (Liu et al., 2014a). The sediment quality guidelines (SQGs) provide two target values (ERL and ERM) to estimate the potential biological effects. Based on the SQG method, the sediment quality guideline quotient (SQGQ) and the mean ERM quotient (m-ERM-q) were developed to assess the potential biological effects of contaminant mixtures (Yang et al., 2013). Moreover, the human health risk assessment (HHRA) model was used to calculate the risks of residents exposed to PAHs (Yu et al., 2014; Liu et al., 2014c). All of these methods provide a scientific basis for PAH pollution control.

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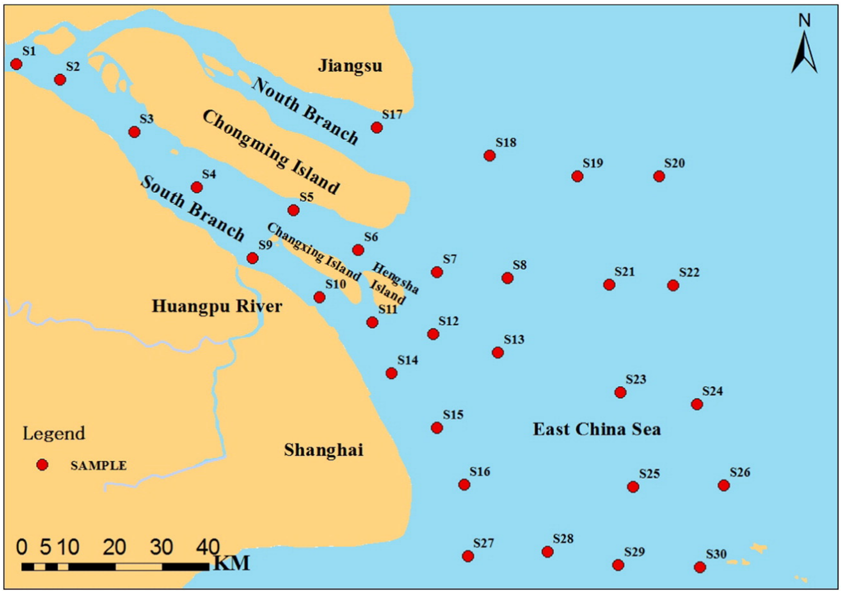

Fig. 1. Location of sampling sites in Yangtze River Estuary.

In the process of pollution evaluation, the pollution level was calculated for the whole area; however, this cannot show the spatial distribution of PAHs in large-scale regions. Many studies have confirmed that PAHs are not uniformly distributed, but their levels vary spatially and temporally (Zhao et al., 2012; Gu et al., 2012). Wang et al. found that the ΣPAH levels and compositions varied in different estuarine zones owing to different sources (Wang et al., 2012). He et al. found that PAH concentrations were positively correlated with clay content and negatively correlated with sediment grain size (He et al., 2014). As the spatial distribution of PAHs in sediments is very important when clarifying the pollution process in aquatic systems and identifying the sources of pollutant inputs (Bai et al., 2014), it is essential to study the spatial distributions of pollutants. Geographic information system (GIS), a popular technology used to display spatial distributions, is widely used in these areas (Ho et al., 2013). Combining pollution assessment and GIS can enable a better understanding of the pollution status of PAHs.

Based on 30 samples of surface sediments from the YRE in February 2011, the pollution-causing characteristics of PAHs were studied. The main purpose of this paper is to study the spatial distribution of PAHs using a GIS approach and to assess the risks of PAHs to the environment and also to human health. The TEQ_{Bap} and SQGQ methods were used to assess the risk to the environment, and the HHRA model was used to evaluate the health risk to humans.

The YRE, which is one of the world's largest estuaries, is located in China's fast-developing area on the boundary between the Yellow Sea and the East China Sea (Chen et al., 2012). The estuary is divided into four branches by three islands: Chongming Island, Changxing Island and Hengsha Island (Fig. 1). Chongming Island splits the estuary into the South Branch and the North Branch (Li et al., 2012).

As the main shipping route in China, the Yangtze River has historically transported 480 Mt $year^{-1}$ of fine sediments to the sea (Yang et al., 2006). Over half of these materials are deposited in the Yangtze Estuary, resulting in pollutant accumulation in the estuarine sediments (Li et al., 2012). Currently, the geographical conditions allow numerous enterprises to flourish in this area. With an increasing population and rapid growth of human activities near the YRE, the YRE has become an ecologically sensitive region (Feng et al., 2008; Liu et al., 2014b). Approximately 106 chemicals have been found in YRE sediments. Among these chemicals, 17 are listed as priority controlled pollutants in the United States of America (Liu et al., 2014c).

In this study, 30 samples were collected in February 2011. Surface sediment samples were collected with a grab sampler (Van Veen bodemhappe 2 L). All of the collected sediments were placed in aluminum containers and kept in a refrigerator at −20 °C for further analysis.

Sixteen priority PAHs were analyzed, including naphthane (Nap), acenaphthylene (Any), acenaphthene (Ace), fluorene (Flu), anthracene (Ant), phenanthrene (Phe), benzo[*a*]anthracene (BaA), chrysene (Chr), fluoranthene (Fla), pyrene (Pyr), benzo[*a*]pyrene (BaP), dibenzo[*a*,*h*] anthracene (DahA), benzo[*b*]fluoracene (BbF), benzo[*k*]fluoracene (BkF), benzo[*g*,*h*,*i*]perylene (BgP), and ndeno[1,2,3-*cd*]pyrene (I1P). High-performance liquid chromatography (HPLC) with a UV and fluorescence detector was used for analyses.

Quality assurance and quality control were performed using duplicates, method blanks, and standard reference materials. Standard reference materials (GSF) were used to examine the accuracy of the determination method. Three replicates were used to determine the total contents of the metals. The contents of the standard reference materials were found to be within 86–102% of the certified values.

This study analyzed sediment samples to assess the potential toxicological and biological impact of PAHs on both the environment and humans.

The toxic equivalent quantity of benzo[*a*]pyrene (TEQ_{Bap}) is a common method used to assess the toxicity of PAHs. The BaP toxicity coefficient is defined as one, and other PAH compounds are converted to

Table 1
The parameter values of the cancer risk (CR) and non-cancer hazard index (HI).

PAHs	Cancer risks ($mg^{-1} \cdot kg \cdot day$)		PAHs	Non-cancer risks ($mg^{-1} \cdot kg \cdot day$)	
	SF_d	SF_i		RfD_d	RfD_i
Baa	1.46E+00	7.30E-01	Nap	2.00E-02	4.00E-02
Chr	1.46E-02	7.30E-03	Any	3.00E-02	6.00E-02
Bbf	1.46E+00	7.30E-01	Ane	3.00E-02	6.00E-02
Bkf	1.46E-01	7.30E-02	Fle	2.00E-02	4.00E-02
Bap	1.46E+01	7.30E+00	Phe	1.50E-02	3.00E-02
Daa	1.46E+00	7.30E-01	Ant	1.50E-01	3.00E-01
Bgp	1.46E+01	7.30E+00	Fla	2.00E-02	4.00E-02
			Pyr	1.50E-02	3.00E-02
			Ilp	1.50E-02	3.00E-02

SF_d: the dermal absorption slope factor; SF_i: the ingestion slope factor; RfD_d: the reference dose of the toxicant through dermal; RfD_i: the reference dose of the toxicant through ingestion.

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