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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).

$$RQ = C_{PAH}/C_{QV} \quad (1)$$

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:

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

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

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:

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

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

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