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Petroleum hydrocarbons in a water-sediment system from Yellow River estuary and adjacent coastal area, China: Distribution pattern, risk assessment and sources

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ABSTRACT

Aliphatic hydrocarbons (AHs), biomarker and polycyclic aromatic hydrocarbons (PAHs) concentrations of surface water and sediment samples collected from Yellow River Estuary and adjacent coastal area in China were measured to determine their spatial distributions, analyze their sources and evaluate the ecological risk of PAHs in the water–sediment system. The spatial distributions of *n*-alkane in sediments are mainly controlled by the mixing inputs of terrigenous and marine components. In comparison with AHs, the total concentrations of $\Sigma 16$ PAHs in surface sediments from a transect of the offshore area were noticeably higher than that of the riverine and estuary areas. Additionally, the AHs and total PAHs concentrations all indicated an overall pattern of a seaward decrease. The PAHs concentrations during the dry season (mainly in the form of dissolved phase) were higher than that of PAHs (mainly dissolved phase and particulate phase form) in the flooding season. In comparison with global concentration levels of PAHs, the level of PAHs in suspended particulate matter and sediments from the Yellow River Estuary was lower than those from other countries, while the concentration of PAHs in the dissolved phase were in the middle range. Petroleum contamination, mainly from oil exploration and discharge of pollutants from rivers, was the main source of *n*-alkanes. The PAHs in the river were mostly of petrogenic origin, while those in the estuarial and marine areas originated mainly from pyrogenic sources. The results of the toxicology assessment suggested that the PAHs in sediments from Yellow River Estuary and adjacent coastal area exhibited a low potential eco-toxicological contamination level.

1. Introduction

Organic pollution from anthropogenic sources to aquatic environments is becoming increasingly concerning. Among the organic contaminants, aliphatic hydrocarbons (AHs) and polycyclic aromatic hydrocarbons (PAHs) have been widely recognized as common organic contaminants that may impact the health of ecosystems and humans. They can enter river or coastal zone environments via direct discharge, surface run-off, atmospheric deposition and biogenic inputs. More importantly, concern has focused on PAHs due to their toxicity, mutagenicity and carcinogenicity (Malik et al., 2011; Yuan et al., 2014). For the reasons above, 16 PAH compounds have been classified as priority pollutants by the United States Environmental Protection Agency (US EPA) and the European Union due to their potential threat to ecosystems and humans (Hu et al., 2010). Owing to their physicochemical properties, PAHs in aquatic systems exist in several phases, including the free dissolved phase, dissolved organic matter (DOM) bound,

adsorbed suspended particulate matter (SPM) and surface sediments. SPM may be generated from the re-suspension of the sediment. As a result, SPM plays an important role in the transportation of PAHs in aquatic multimedia. In general, PAHs come mainly from two sources: petrogenic and pyrogenic. Apart from the combustion source, another common anthropogenic source of PAHs is the accidental spillage of fossil fuels.

Estuaries and coastal zones are a transitional zone between land and ocean systems. Therefore, the physical and chemical changes are more extreme in estuary environments than in many other environments as a result of tides, river flow and dry-wet cycles. As the second largest river in China and sixth largest river in the world, the Yellow River carries a huge amount of sediments to the Bohai Sea when flowing through the Loess Plateau, China. The Yellow River Delta (YRD), one of the most active regions of land–ocean interaction among the large river deltas in the world, is a newly born wetland in China. Estuaries often act as sinks for sediments and the associated particle-reactive contaminants; thus,

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AHs and PAHs from the upper reaches of the Yellow River may have been transported to the estuaries by solids or water before accumulating at the estuary mouth. The YRD is the main sink of organic pollutants discharged from the Yellow River watershed. Rapid economic growth in YRD raises concerns of significant oil pollution to aquatic environment and sediment. Additionally, the increased exploitation of the nearby Shengli Oilfield, the second largest oilfield in China, probably resulted in local petroleum hydrocarbon contamination in the area. Thus, the investigation of AHs and PAHs in the water–sediment system of estuaries and coastal zone areas may help elucidate the balance between inputs and losses and further our understanding of the associated environmental processes and outcomes.

Protecting the delicate ecology in the YRD is of great importance. There have been several studies on AHs and PAHs in marine sediments, such as distribution, source apportionment of PAHs in estuarine sediments from the Yellow River (Liu et al., 2008), hydrocarbons in the surface sediments from the Huanghe Estuary and adjacent area (Zhang et al., 2010) or distribution, source apportionment and potential toxicological significance of AHs and PAHs in Yellow River Delta wetlands (Yang et al., 2009; Wang et al., 2011; Yuan et al., 2014) from the YRD, sources appointment and ecological risk assessment of PAHs in sediments of Erhai Lake, China (Hezhong et al., 2016). They indicate that sedimentary AHs and PAHs are at low or medium levels compared with the reported values of other deltas, and are derived from mixed pyrogenic and petrogenic sources. However, to the best of our knowledge, detailed information on the spatial-temporal distribution and ecological risk evaluation of the distribution of AHs and PAHs in the Yellow River Estuary (YRE) remains scarcely documented; especially with respect to the water–sediment system encompassing the marine, estuarine and lower estuarine regions. The aims of this study were to (1) determine the spatial–temporal distributions of AHs, biomarkers and PAHs in the water–sediment system from the Yellow River encompassing the riverine, estuarine and offshore areas; (2) examine the potential input sources of petroleum hydrocarbons and analyze the source apportionment of PAHs in the sediment–water system; and (3) assess the potential eco-toxicological consequences on organisms in the region.

2. Materials and methods

2.1. Sampling location

The surface sediments used were collected from the YRD and adjacent coastal area in November 2012 (Fig. 1). The collecting period avoided the high rainfall season. Lijin Hydrographic Station is the last hydrographic station of the Yellow River. The sampling stations were arranged along a 127.5 km stretch from the Lijin Hydrological Stations to the river mouth along the Yellow River, extending from the lower stretch estuary (R1–R8) to the main estuary (Est-1–Est-5) and the offshore area of Bohai Sea with the transect (T-B; Table 1). Est-1 station is Xintan Oilfield Floating Bridge, which is regarded as the Yellow River

Entrance. Sampling sites used in this study commenced from the freshwater end-member in the upper reaches of the river estuaries to the saltwater sea and encompassed the entire salinity gradient. The surface sediments (0–5 cm) were collected using a Van Veen style stainless steel grab sampler (24 L) manufactured by the Wildlife Supply Company. Following collection, the samples were homogenized, placed into sterile polyethylene bags, sealed and stored at $-4\text{ }^{\circ}\text{C}$ until further analysis. The concurrent sampling of the surface water (0.5 m depth) from the 4 sites (R6, R8, Est.-2, T-B5) were also collected in May (Flooding season) and November (Dry season) 2012. Twenty liters of water was collected from each sampling site. 100 mg/L HgCl_2 was added to each water sample container for the inhibition of bacterial growth during transportation and temporary storage. Then transported on ice to the laboratory, where they were stored at $-20\text{ }^{\circ}\text{C}$ until further analysis.

2.2. Extraction and fractionation

The procedures for eluting dissolved organics from XAD resins and for processing glass fiber filters have been described extensively elsewhere (Sun et al., 2009). Briefly, after back to the laboratory, SPMs were collected by filtering the water samples through GF/F glass fiber filters (diameter 0.7 mm, Whatman, UK). The SPM samples filtered through the glass fiber filter were freeze-dried and weighed using an analytical balance. The filtered water was then passed through a mixed resin (Sigma Amberlite XAD-4 and XAD-2 1:1 mixtures) glass column (400 mm \times 25 mm i.d.) to retain dissolved organics. The XAD resin was transferred into a glass flask and extracted 3 times with 2:1 dichloromethane/*n*-hexane (v/v) in an ultrasonic bath, after which the extracts were combined. The fraction drained with anhydrous Na_2SO_4 was then concentrated using a rotary evaporator and further evaporated under a nitrogen stream to approximately 1–2 mL. Lastly, the fraction was spiked with surrogate recovery standards (terphenyl-d14) for Gas Chromatography–Mass Spectrometer (GC–MS) analysis.

The sediment samples were freeze-dried, ground and separated with a 125- μm sieve to remove large particles and debris in April 2013. The freeze-dried SPM-loaded glass fiber filters and sediment samples were weighed and spiked with surrogate recovery standards (terphenyl-d14) and Soxhlet-extracted for 72 h, with *n*-hexane, toluene, chloroform and methanol as extraction solvent. Elemental sulfur was removed from the extracts using activated copper. The extracts were fractionated by column chromatography on alumina over silica gel. Silica gel and anhydrous sodium sulfate were baked in a furnace oven at $650\text{ }^{\circ}\text{C}$ for 6 h prior to use. The fractions of saturated hydrocarbons, aromatic hydrocarbons and non-hydrocarbons were obtained by successively eluting with *n*-hexane, toluene and chloroform/methanol (98:2), respectively. The saturated and aromatic hydrocarbon fractions were concentrated again in a rotary evaporator, transferred to a vial, and then the volume was evaporated to exactly 1 mL using a stream of filtered N_2 gas. An aliquot of 1 mL of each extract was subjected to analysis by GC–MS.

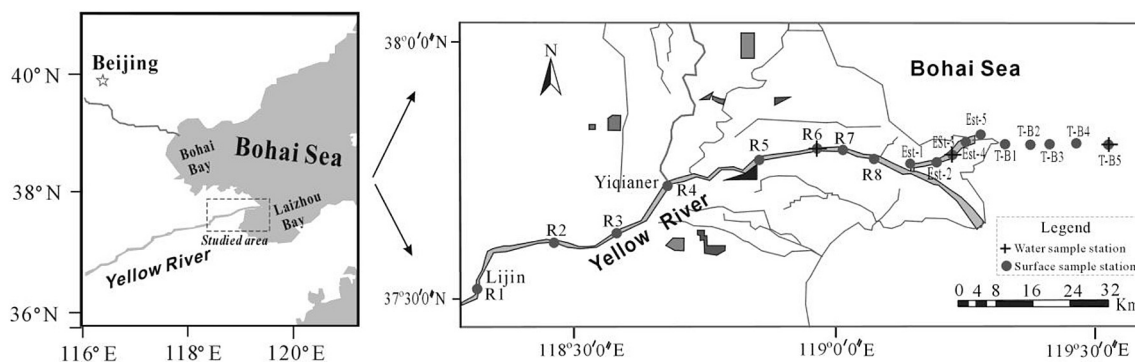


Fig. 1. Study areas and sampling location.

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