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Polycyclic aromatic hydrocarbons in sediments from the Old Yellow River Estuary, China: Occurrence, sources, characterization and correlation with the relocation history of the Yellow River

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ABSTRACT

The levels of 16 USEPA priority PAHs were determined in surface sediments and one dated sediment core from the abandoned Old Yellow River Estuary, China. Total PAH concentrations in the surface sediments ranged from 100.4 to 197.3 ng g^{-1} dry weight and the total toxic equivalent quantity (TEQ^{carc}) values of the carcinogenic PAHs were very low. An evaluation of PAH sources based on diagnostic ratios and principal component analysis suggested that PAHs in the surface sediments mainly derived from combustion sources. The total PAH concentrations altered significantly with year of deposition and showed quite different patterns of change compared with other studies: it is hypothesized that the principal cause of these changes is the relocation of the course of the Yellow River to the sea in 1976 and 1996.

1. Introduction

Reconstructing the history of contaminant inputs to an environment is helpful in revealing the socioeconomic development, historical energy consumption and historical events which have occurred nearby, thus improving our understanding of the relationship between anthropogenic activities and environmental quality (Santschi et al., 2001; Van Metre and Mahler, 2005; Denis et al., 2012; Liu et al., 2012a). The insights gained will, in turn, assist in the development of efficient strategies for reducing adverse human effects on the environment. One efficient way to reconstruct the chronology of contaminants is to investigate sediment cores and polycyclic aromatic hydrocarbons (PAHs) have been the focus of many pollution reconstruction studies (Lima et al., 2003; Kannan et al., 2005; Moriwaki et al., 2005; Barra et al., 2006; Martins et al., 2010, 2011; Machado et al., 2014).

PAHs are a group of ubiquitous environmental pollutants with two or more fused aromatic rings (Menzie et al., 1992), and are of environmental concern due to their toxic, mutagenic and carcinogenic potential (International Agency for Research on Cancer (IARC), 1987). They primarily originate from anthropogenic sources, such as incomplete combustion of fossil fuels and direct release of oil and oil products (Simpson et al., 1996). Since PAHs typically correlate well with anthropogenic activities, they are useful geochemical markers of anthropogenic impact (Liu et al., 2012a). Once PAHs from various sources reach an aquatic environment, owing to their strong hydrophobic character (low aqueous solubility and high octanol/water partition coefficients), they tend to associate with particulate material in the aquatic environment, and are then transported and accumulate with the underlying sediments (Cardellicchio et al., 2007; Martins et al., 2010; Patrolecco et al., 2010). As the deposition of sediments continues over time, sediments can act as geochronometers of PAH deposition in the environment (Kannan et al., 2005). Moreover, owing to the persistent nature of some PAHs, the resulting deposition may be preserved in the sediment cores for a long period (Elmquist et al., 2007; Lin et al., 2012). Therefore, sediment studies can be an effective approach to PAH contamination research and PAHs are good indicators of historical inputs of anthropogenic matter into the sedimentary environment (Viguri et al., 2002; Martins et al., 2010).

The Yellow River, the second longest river in China, has flowed into the Bohai Sea in North China since 1855, gradually forming the Yellow River Delta, which is the largest delta plain in China and one of the most biodiverse zones in the world. The course of the Yellow River to the sea was subject to constant changes for natural and anthropogenic reasons, before ultimately following the current course, which is called

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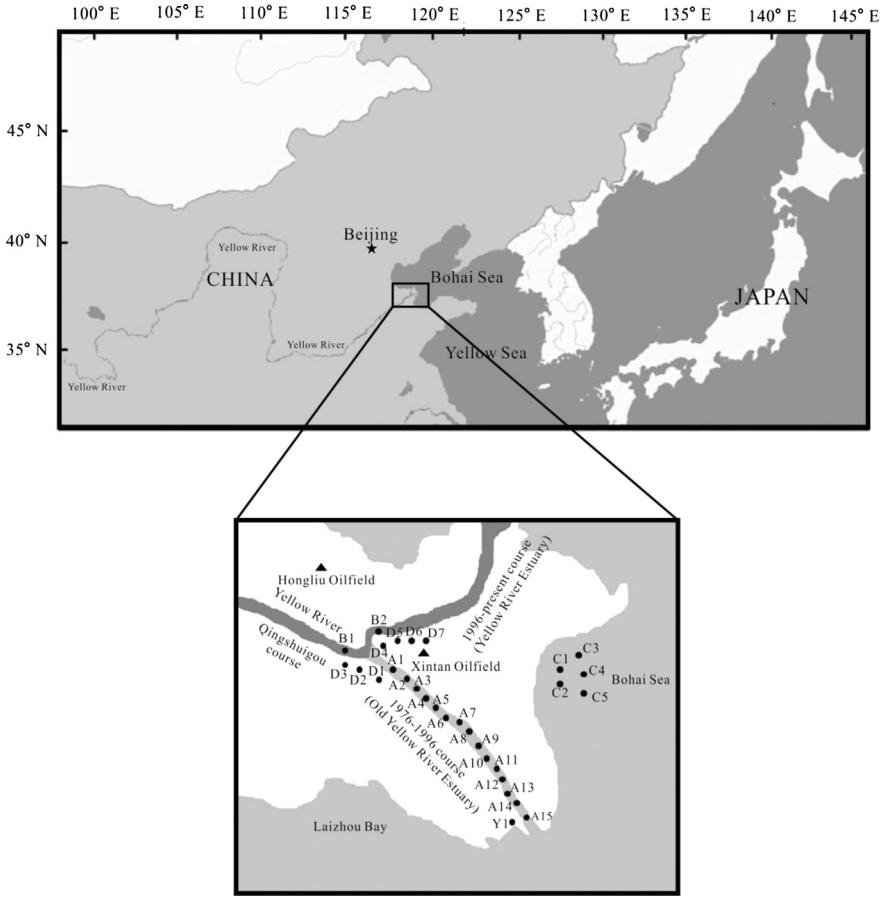

Fig. 1. Sampling locations in the Old Yellow River Estuary, China.

the Yellow River Estuary. However, from 1976 until the formation of the present estuary in 1996, the Yellow River flowed into the sea through the Old Yellow River Estuary (Fig. 1). Estuaries often act as sinks for fine-grain sediments and associated particle-reactive contaminants and may serve as important sources of organic contaminants to coastal marine environments (Feng et al., 1998), so PAHs from the upper reaches of the Yellow River may have been transported to the estuaries by solids and water and accumulated at the estuary mouth. Additionally, the exploitation of the nearby Shengli Oilfield, which is the second largest oilfield in China, probably caused local PAH contamination here (Wang et al., 2011). After relocation of the Yellow River to the present Yellow River Estuary and the blocking of the former entrance, the abandoned Old Yellow River Estuary received backwash from the Bohai Sea. Moreover, the fishery and shipping activities in the Old Yellow River Estuary are very common. Since toxic contaminants such as PAHs can affect human health through the food chain, an investigation into the level of threat of PAH contamination to the aquatic environment is urgently required. Although the Old Yellow River Estuary is an abandoned channel, studies of abandoned channels are still of great significance, because they provide important evidence of the historical evolution of river morphology and human civilization along the river. However, recent studies have concentrated on the current Yellow River Estuary and Yellow River Delta (Hui et al., 2009; Wang et al., 2009; Yang et al., 2009; Hu et al., 2014) and little research has specifically investigated PAH contamination in the abandoned Old Yellow River Estuary, though this was an important channel in the modern era of the Yellow River, and the course it followed to the sea for nearly twenty years.

The objectives of the present study include the following: (a) investigating PAH contamination in surface sediments and identifying possible anthropogenic sources; (b) reconstructing the history of PAH contamination and establishing a correlation between PAH concentrations and the socioeconomic development and historical evolution of river morphology. Since anthropogenic activities in the abandoned Old Yellow River Estuary have been intensive after the discovery of the nearby Shengli Oilfield, and the course of the Yellow River to the sea was subject to two major changes during the period studied, this area was considered suitable for our investigation.

2. Materials and methods

2.1. Sample collection

The study area and sampling locations are illustrated in Fig. 1. Fifteen surface sediment samples (A1 to A15) were collected in the Old Yellow River Estuary using a stainless steel grab in July 2012. For comparison, another fourteen surface sediment samples were collected in adjacent waters at the same time: specifically, from two locations in the Yellow River (B1 and B2), five locations in the Bohai Sea (C1 to C5) and seven locations in the adjacent wetland (D1 to D7). Moreover, an undisturbed sediment core (Y1) was collected using a gravity corer in an intertidal mud flat area, unaffected by any human interference, near the mouth of the Old Yellow River Estuary. The entire core, which was 41 cm long, was sectioned at 1 cm intervals using a stainless steel cutter. All the surface sediments and sectioned sediment samples were wrapped in aluminium foil, which was pre-combusted at 450 °C for 4 h to remove organic matter, and then transported on ice to the laboratory, where they were stored at −20 °C until further analysis.

2.2. ^{210}Pb and ^{137}Cs dating of the sediment core

The dating of the sediment core was conducted by an Ortec HPGe GWL series well-type coaxial low background intrinsic Ge detector manufactured by AMETEK. The system calibration was performed using three reference materials, namely RGU-1, RGTh-1 and RGK-1, obtained from the International Atomic Energy Agency. According to the methods described in prior studies (Wu et al., 2006; Guo et al., 2007),