



An assessment of human influences on sources of polycyclic aromatic hydrocarbons in the estuarine and coastal sediments of China



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ABSTRACT

Sediments collected from the coastal area of China, embracing west coast of Bohai Sea, south coast of Shandong Peninsula, and the Changjiang estuary (listed in order of decreasing north latitude), were analyzed for polycyclic aromatic hydrocarbons (PAHs). Σ PAH (Sixteen US EPA priority PAHs) were 2.7–350.9 ng/g. Petroleum residue was the major contributor of PAHs in the coastal sediments of China due to oil leakage from ships and offshore oil fields. The contribution of vehicular emissions in coast of North China was significantly lower than that in the Changjiang Estuary, and the reverse was true for coal combustion. PAH concentrations in the sediment core of the Changjiang estuary steadily increased upward and the variation was primarily due to economic development and severe floods. The impact on PAHs by vehicular emissions (37.2%) and petrogenic sources (45.8%) overwhelmed combustion sources (17.0%).

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1. Introduction

China is one of the largest coastal countries in the world. The coastal and estuarine environments in China are under growing environmental pressure because pollutants released along with municipal and industrial wastes can enter the sea by river and atmosphere, and ultimately accumulate in the coastal areas, introducing long-term accumulative effects (Yuan et al., 2001; Cao and Wong, 2007; Pan and Wang, 2012). In 2014, 57360 km² of offshore areas of China were heavily polluted (NBO, 2014).

A large proportion of polycyclic aromatic hydrocarbons (PAHs) are formed naturally by incomplete combustion of coal, wood, and petroleum products (Yunker et al., 2002), and they may also be released from petrogenic and biogenic sources. Sedimentary PAH concentrations were reported to be correlated significantly with local socioeconomic activities (Liu et al., 2008a, 2013). Traffic and coal combustion have been considered to be the two most important sources of PAHs in many areas (Soliman et al., 2014). Therefore, PAHs can serve as geochemical markers for assessing anthropogenic influences within a specific

environmental medium because they are closely related to human activities (such as anthropogenic combustion processes). PAHs input to the coastal marine environment are primarily from atmospheric deposition from airsheds of the coastal ocean (precipitation and dry deposition) and the movement of water containing dissolved and particulate constituents derived from watersheds (Latimer and Zheng, 2003).

Coastal sediments are considered as both sinks and secondary sources of terrestrially derived pollutants (Blair and Aller, 2012). Many studies (Mai et al., 2001; Hui et al., 2009) have been done to investigate the contents of PAHs in estuaries and bays of China. Municipal wastewater facilities and industrial outfalls were the major point sources for most of those areas. Little current data are available on the differences between sources of PAHs in the whole coastal sediments of China (i.e. between north and south). Lin et al. (2011) and Liu et al. (2012) observed that the composition of PAH molecules in sediments of the northern area of China Sea differed from that in sediments of the southern area of China Sea. This discrepancy was probably resulted from higher PAH emissions from coke industry and domestic coal combustion in North China than in South China (Xu et al., 2006; Lin et al., 2011; Liu et al., 2012).

The interest of this work is thus to derive the concentrations and potential sources of sedimentary PAHs in the coastal

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sediments collected from different regions of China, to diagnose the input mechanisms of PAHs, and to examine whether there is any correlation between regional anthropogenic activities and the spatial distributions of PAHs.

2. Methods

2.1. Sampling area and sample collection

The natural environments and human activities of various regions of China were significant different. Thus, the quantities and compositions of pollutants in environments vary a lot among different regions. This study focused on the regional differences of the coastal environment. Geographically from north to south (Fig. 1), three major geographical regions, west coast of Bohai Sea (Region A), south coast of Shandong Peninsula (Region B) and the Changjiang estuary (Region C) were defined. Region A and B are far from cities and harbors.

Surface sediments (0–2 cm layer) were collected from Region A (sites A1–A19, August 2013) and B (sites B1–B23, October 2013) with a stainless steel grab. In March 2011, a sediment core was collected from Region C (Fig. 1) with a stainless steel box-corer. The core (YE1, 30 cm) was sectioned immediately at 1-cm intervals aboard the ship using a stainless steel blade. All the samples were wrapped in aluminum foil after collection and immediately stored at $-20\text{ }^{\circ}\text{C}$ until analysis. The sediment core was dated based on ^{210}Pb and ^{137}Cs activity-depth profiles at University of Science and Technology of China by Prof. Liguang Sun research team. The average depositional rate was 0.23 cm/a .

2.2. Sample preparation and GC–MS analysis

The sediments were freeze-dried and homogenized. Each sample (10.0 g) was spiked with perylene- d_{12} and phenanthrene- d_{10} as surrogate standards. The samples were then ultrasonic extracted with acetone and *n*-hexane (1:1, v/v). Activated copper powder was added into the extracted samples to remove element sulfur. The extract was rotary-evaporated to about 2 mL and then cleaned and fractionated on a multilayer silica–alumina composite column, which consisted of 7.0 g of 5% deactivated silica gel, 7.0 g of 5% deactivated aluminum oxide, and 2.0 g of anhydrous sodium sulfate from bottom to top. The column was pre-cleaned with 50 mL of *n*-hexane and the extract was eluted in sequence with 20 mL of hexane (first fraction) and 40 mL of dichloromethane/hexane (3:7, v/v) (second fraction). The second fraction was collected and concentrated to near dryness under a gentle N_2 stream and then reconstituted in 100 μL of isoocane.

Operating conditions of GC–MS were as described by Deng et al. (2013). The analysis of PAHs fraction was performed on an Agilent Technologies 6890 GC coupled with an Agilent Technologies 5975 mass spectrometer, and a DB-5MS (30 m, i.d. 0.25 mm, film thickness 0.25 μm) capillary column, using selective ion monitoring mode (SIM). The column temperature was started from $60\text{ }^{\circ}\text{C}$ (held for 2 min), increased to $300\text{ }^{\circ}\text{C}$ at $4\text{ }^{\circ}\text{C}/\text{min}$ and held for 10 min. The quantification of PAHs was conducted using response factors of phenanthrene- d_{10} (internal standard for PAHs with mass less than 252) and perylene- d_{12} (for higher molecular weight ones).

Sixteen USEPA priority PAHs were analyzed in terms of individual and total concentrations. The 16 PAHs measured were: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Acp),

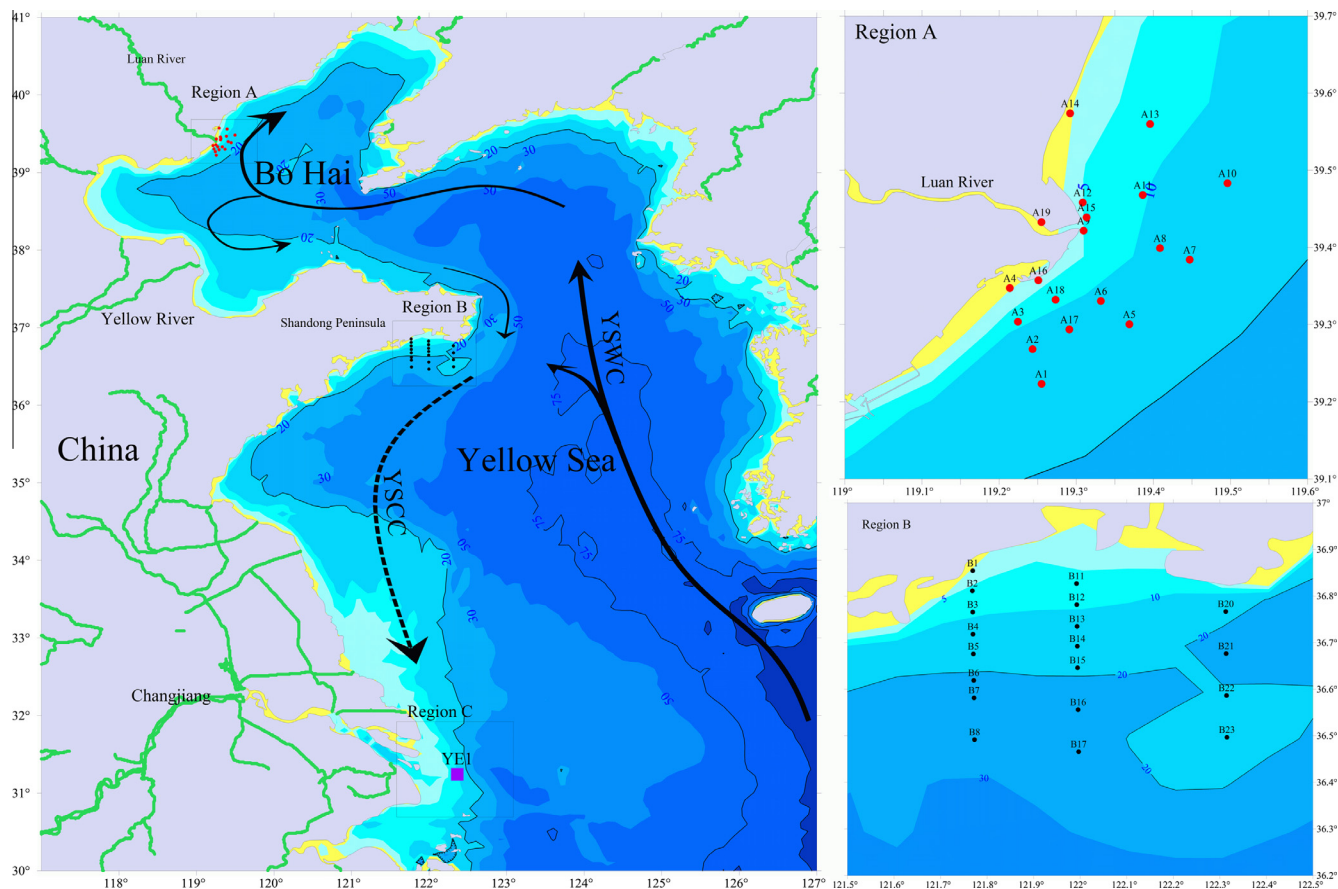


Fig. 1. Locations of sampling sites. General circulation system in the Yellow Sea and Bohai Sea (indicated by arrows) is after Liu et al. (2007). YSCC: Yellow Sea Coastal Current; YSWC: Yellow Sea Warm Current.

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