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Spatial variation and sources of polycyclic aromatic hydrocarbons influenced by intensive land use in an urbanized river network of East China

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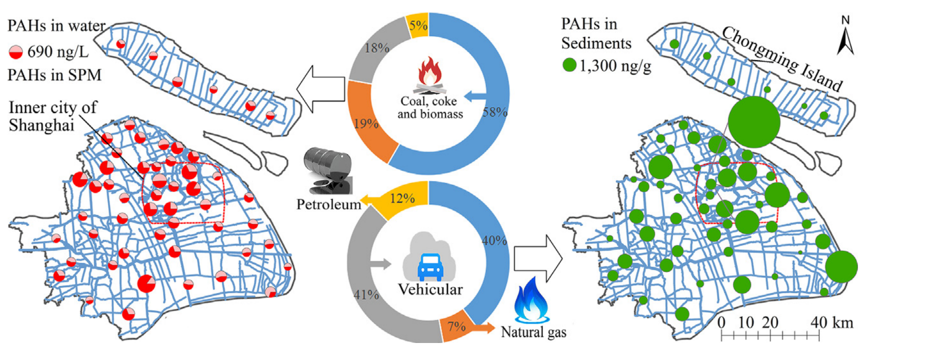
HIGHLIGHTS

- PAHs were analyzed in water, SPM and sediments of Shanghai river network.
- PAHs in SPM and sediments showed the highest concentrations in inner city.
- Main factors affecting the partitioning and spatial variations of PAHs were analyzed.
- Vehicle emissions in the urbanized and industrialized areas contributed strongly to PAHs.

GRAPHICAL ABSTRACT



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ABSTRACT

The concentrations and distribution of polycyclic aromatic hydrocarbons (PAHs) in urbanized river networks are strongly influenced by intensive land use, industrial activities and population density. The spatial variations and their influencing factors of 16 priority PAHs were investigated in surface water, suspended particulate matter (SPM) and sediments among areas under different intensive land uses (industrial areas, agricultural areas, inner city, suburban towns and island areas) in the Shanghai river network, East China. Source apportionment was carried out using isomer ratios of PAHs and Positive Matrix Factorization (PMF). Total concentrations of 16 PAHs ranged from 105.2 to 400.5 ng/L, 108.1 to 1058.8 ng/L and 104.4 to 19,480.0 ng/g in water, SPM and sediments, respectively. The concentrations of PAHs in SPM and sediments varied significantly among areas ($p <$ 0.05), with the highest concentrations in inner city characterized by highly intensive land use and high population density. The PAH concentrations in sediments were positively correlated with those in SPM and were more strongly correlated with black carbon than with total organic carbon, indicating a stronger influence of prolonged anthropogenic contamination than the recent surface input in sediments. Biomass and coal combustion contributed strongly to total PAHs, followed by natural gas combustion in water and SPM, and vehicular emissions in

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sediments. Vehicular emissions were the strongest contributors in SPM and sediments of the inner city, indicating the strong influence of vehicular transportation to PAHs pollution in the urbanized river network.

1. Introduction

PAHs are ubiquitous environmental contaminants and have received considerable attention because of their toxic, mutagenic and carcinogenic characteristics (Long et al., 1995; Menzie and Potokib, 1992). Anthropogenic PAH sources have been identified to mainly originate from vehicular emissions, industrial activities, domestic heating, waste incineration, petroleum spills, biomass and coal combustion (Guo et al., 2009; Harrison et al., 1996; Simcik et al., 1999; Zhang et al., 2017). The PAHs generated from these anthropogenic sources and natural processes such as volcanoes and forest fires can be transported to surface water through atmospheric deposition, road runoff, spillage of petroleum products and wastewater discharges (Herngren et al., 2010; Uher et al., 2016). As a result of their hydrophobic characteristics, PAHs in aquatic environments tend to become associated with suspended particles and are subsequently deposited to the sediment layers.

Intensive industries and urbanization activities have an important influence on the spatial distribution of PAHs in the aquatic system (Chen et al., 2007; Ligaray et al., 2016; Timoney and Lee, 2011). Sediment PAH concentrations in the Athabasca River Delta were related to industrial activities (Timoney and Lee, 2011). Urban areas have the highest PAH loadings compared to rural areas (Chen et al., 2007; Ligaray et al., 2016). Timoney and Lee (2011) found no significant correlations between river discharge variables and sediment PAH concentrations. Intensive urban use was identified as the most important factor influencing PAH distribution in river sediments, which not only led to highly PAH-polluted river sediments, but also resulted in significant fluctuation of PAH levels among locations (Liu, A. et al., 2017). Dissolved and labile phases of PAHs were the phases most related to the watershed population density, whereas particulate PAHs were more related to local pressure (Uher et al., 2016).

Numerous studies have reported the levels, spatial and temporal variations, partitioning characteristics, sources and ecological risks of PAHs in river systems (Montuori et al., 2016; Sarria-Villa et al., 2016; Zhang et al., 2017). Almost all related research has focused mainly on individual rivers or river sections; however, in some coastal regions, several rivers are interconnected with each other and form river networks. To date, few studies have been conducted on PAH pollution in these river networks (Liu et al., 2016). Given their location and interconnected nature, these river networks would be more influenced by intensive urban land use, industries and population density.

The Shanghai river network is a highly urbanized and industrialized region in China and is a typical river network with weak hydrodynamic conditions. The distribution of PAHs in three hierarchical rivers in this region and sources of PAHs in surface sediments of the Huangpu River were reported (Liu et al., 2009; Liu et al., 2016). The aims of this study were to investigate the concentrations and spatial variation of PAHs in water, SPM and sediments among different intensive land uses and to analyze the main factors affecting the partitioning and distribution of PAHs in multiple phases of the Shanghai river network to ultimately identify and quantitatively assess source contributions to PAHs. The results of this research will be useful for alleviating PAH contamination in urbanized river networks worldwide.

2. Materials and methods

2.1. Study area and samples collection

The study area is located in Shanghai (30°40′–31°53′ N, 120°52′–122°12′E), which is one of the largest and most important industrial cities in China, and sits on the Yangtze Estuary of East China. Shanghai belongs to the northern subtropical monsoon climate zone, with an annual average precipitation of 1435.8 mm, an annual rainy days of 151 days, and an annual mean temperature of 17.1 °C in 2012 (SMSB, 2013). The main land Shanghai involves a soft deltaic deposit with some isolated outcrops of bedrock. Under the upper clayey soil, there is a sand layer, which is a phreatic aquifer, where the groundwater level is 0.5–2.5 m below the surface (Shen et al., 2014). There are about 33,127 rivers or creeks in Shanghai, with a length of ~24,915 km and total water area of ~642.7 km^2 (SMWAB, 2016). These rivers are interconnected with each other, producing a river network. Huangpu River is the dominant river in this river network system and the last tributary of the Yangtze River before discharge to the ocean. Except for the Huangpu River, most of the rivers have been cut off by flood gates in this river network system, which results in weak hydrodynamic conditions. The length of drainage sewer pipes and the number of pumping stations were 18,191 km and 183 in 2012 (SMSB, 2013). The increased surface runoff due to land surface modification during urbanization has aggravated the burden of drainage network.

The population density in Shanghai reached 3754 persons/km^2 in 2012, and the maximum population density in the inner city was up to 36,014 persons/km^2 (SMSB, 2013). There are 104 small towns outside the inner city areas, the total area of which accounts for 87.08% of the total area of Shanghai, and the total population of the township accounts for 50.60% of the total resident population in Shanghai (Liu, H. et al., 2017).The average daily energy consumption in Shanghai in 2012 were 12.73×10^7 kg for coal, 1.84×10^7 kg for coke, 1.75×10^7 kg for fuel oil, 1.56×10^7 kg for diesel oil, 1.42×10^7 kg for gasoline, 1.10×10^7 kg for kerosene, 18 million m^3 for diesel oil and 371 million kwh for electricity (SMSB, 2013). Six key industries including electronic information product manufacturing, automobile manufacturing, petrochemical and fine chemical products manufacturing, fine steel manufacturing, equipment complex manufacturing and bio-medicine manufacturing contributed 66.0% to the total gross output value of industry in Shanghai in 2012 (SMSB, 2013). The total sown area of the crops was 3.9×10^5 hm^2, of which the grain crops and the economic crops contributed 48.1% and 51.9% respectively (SMSB, 2013).

According to the principle of gridded stationing, 53 sampling sites were selected for sample collection from areas under the following intensive land uses: inner city ($n = 11$), industrial areas ($n = 11$), agricultural areas ($n = 15$), suburban towns ($n = 10$) and island areas on Chongming Island ($n = 6$) (Fig.1). Detailed description of sampling areas was listed in Table 1. Chongming Island, with the population density of 590 persons/km^2 (SMSB, 2013), is located in the Yangtze estuary and aims to develop ecological agriculture. The rivers on the island are connected to the Yangtze River, so the river network on Chongming Island is separated into island areas. Sampling sites were selected from straight flowing river section in the Sampling grid. Sewage outlets and new dredged river sections without sediments were avoided.

Surface water, SPM and surface sediment samples were collected in April 2012. Samples were collected in the middle of the river cross section by means of a bridge or a boat. Bipartite samples of surface water (5–15 cm depth) were collected using a plexiglass sampler and stored in 1 L clean brown glass bottles; surface sediments (0–15 cm depth) were sampled using the Ekman-Birge bottom sampler (HYDRO-BIOS, Germany) and stored in sealed polyethylene bags. Both water and sediment samples were directly transported to the laboratory after sampling and stored at 4 °C and −18 °C, respectively. A global positioning system (GPS) was used to locate the sampling sites. Temperature, pH and dissolved oxygen (DO) contents of the river water were measured