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Spatial distribution, sources and ecological risk assessment of polycyclic aromatic hydrocarbons in surface seawater from Yangpu Bay, China

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ABSTRACT

The occurrence of 14 polycyclic aromatic hydrocarbons (PAHs) listed by the United States Environmental Protection Agency was investigated in surface seawater of Yangpu Bay, China in November 2013 (winter) and May 2014 (summer). Seventy-two samples were collected from 12 sampling sites. The total concentrations of PAHs in Yangpu Bay showed obvious variations in different seasons, which varied from 582.8 to 2208.3 ng L⁻¹ in winter and 952.4 to 1201.7 ng L⁻¹ in summer, respectively. Two-ring PAHs accounted for more than 91.6% of total PAHs in winter, and three-ring PAHs were dominant with 81.6% of total PAHs in summer. Molecular indices analyses indicated that the main source of PAHs in Yangpu Bay could be petrogenic contamination. The ecological risk assessment by Risk quotients (RQ_{NCS} and RQ_{MPCs}) showed a potential ecological risk of PAHs in Yangpu Bay, indicating a close attention should be paid to pollution of PAHs in the coastal area of Yangpu.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are an important class of ubiquitous coastal environmental persistent organic pollutants (POPs) with toxic, mutagenic and carcinogenic potentials (IARC, 1987). The contaminations of PAHs in the marine environment can be derived from three main sources, including pyrogenic, petrogenic and diagenetic origin (Lafamme and Hites, 1978; Mostafa et al., 2009; Yim et al., 2014). Because of their persistence, long-range transport and high octanol/air partition coefficients (K_{OA}), PAHs can be retained for a long time in the environment and accumulate in the organisms through the food chains (Huang et al., 2012). There are sixteen PAHs placed in the priority-pollutant list by the United States Environmental Protection Agency (USEPA) (Keith and Telliard, 1979).

As PAHs are preferentially adsorbed onto particulate matter, bottom sediments are considered to be a reservoir for these

hydrophobic pollutants in aquatic environment (Budzinski et al., 1997). Therefore the investigations of PAHs in particles and sediments were widely studied around the world (Huang et al., 2012; Darilmaz et al., 2013; Retnam et al., 2013; Miki et al., 2014). However, few data on the PAHs dissolved in water, especially in the marine environment are available (Cao et al., 2010; Ya et al., 2014), although more attentions should have been also paid to PAHs in water, as they may produce direct toxic effects on organisms and transport with water current (McGrath and Di Toro, 2009).

Yangpu Bay is located in the northwestern part of Hainan Island, China with an area of 220 km². An important industrial development zone and several large ports are located along the coast. Municipal wastewater, industrial sewage, oil spillage and atmospheric deposition are potential sources of pollution introduced into Yangpu Bay because of petroleum industry, shipping, fishing and human activities (Wei et al., 2010). Due to the potential impact of the pollution from anthropogenic sources on marine organisms, it is important to identify and assess the risk caused by anthropogenic pollution on the ecosystem of Yangpu Bay. However, there has been little data about PAH contaminations at this area.

The present study is the first investigation of the current status of PAHs contamination in the surface water of the Yangpu coastal

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area. The aims of this study were to identify spatial distribution, concentration determination, potential sources and risk assessment of PAHs in the surface seawater so as to provide a valuable reference data for PAHs contamination control and coast environmental management.

2. Materials and methods

2.1. Study area and sampling

Yangpu Bay locates between 19°11'N and 109°43'E. It has a tropical monsoon climate with clear rainy and dry seasons. The annual average rainfall is 1113.8 mm. The Beibu Gulf's circulation driven by monsoon winds, the tidal current entering the gulf from southeast to northwest form as clockwise circulation in summer; the direction of tidal currents in winter is opposite to that in summer (Bauer et al., 2013).

Twelve sampling sites were studied in the Yangpu coastal area based on their different potential levels and sources of contamination (Fig. 1). Seventy-two surface seawater samples (50 cm below the seawater) in Yangpu Bay were collected in November 2013 (water temperature of 22 °C) and May 2014 (water temperature of 32 °C). Global positioning system (GPS) was used for locating the sampling positions. All of the samples were placed in pre-cleaned brown glass bottles and stored at 4 °C until analysis. The surface seawater sampling was carried out according to the Off-shore Marine Areas Monitoring Technical Specifications (HJ422-2008).

2.2. Chemicals and reagents

Sixteen priority PAHs listed by USEPA used in the study are: naphthalene (Nap); acenaphthylene (Acy); acenaphthene (Ace); fluorene (Flu); phenanthrene (Phe); anthracene (Ant); fluoranthene (Flua); pyrene (Pyr); benzo[*a*]anthracene (BaA); chrysene (Chy); benzo[*b*]fluoranthene (BbF); benzo[*k*]fluoranthene (BkF); benzo[*a*]pyrene (BaP); benzo[*ghi*]perylene (BghiP); indeno[1,2,3-

cd]pyrene (InP); dibenzo[*a,h*]anthracene (DahA). However, Acy and InP could not be quantified because of very low recovery rate and poor resolution in the chromatogram. The standards of the sixteen PAHs (purity > 99%) were obtained from AccuStandard (Connecticut, USA). The organic solvents of dichloromethane (DCM) and methanol ($\geq 99.9\%$, HPLC grade, Fisher Scientific Company, USA) were used for samples processing. Ultrapure water (Cascade Lab Water Systems, final resistivity: $18.2 \text{ M}\Omega \text{ cm}^{-1}$) was used in the analysis process. Anhydrous sodium sulfate (analytical grade) were heated at 450 °C for 6 h and stored in sealed containers. SPE Solution Classic C18 cartridges (500 mg/6 ml with capacity for 45 μm particle size) were purchased from Simon Aldrich, German.

2.3. Sample preparation and extraction

PAHs in surface seawater were extracted by following procedures. Each water sample (500 mL) with the addition of 5 mL methanol was filtered and preconcentrated by solid-phase extraction (SPE) with a C18 cartridge at a flow rate of 5 mL min^{-1} . Each C18 SPE cartridge was preconditioned with 10 mL dichloromethane followed by 10 mL methanol and 10 mL ultrapure water. After the percolation, the cartridge was dried under vacuum condition. Then PAHs were eluted from the cartridge with 12 mL dichloromethane, and Na_2SO_4 was added for removing residual water. The extract was concentrated to 1 mL by dry nitrogen ($\geq 99.99\%$) in a water bath (40 °C). The concentrated solution was then solvent-exchanged into 3 mL methanol and reduced to nearly dry at 40 °C under nitrogen purge. The resulting residue was re-dissolved with 1 mL methanol for ACQUITY Ultra Performance Liquid Chromatography (UPLC) analysis.

2.4. Analysis methods

The quantification of PAHs was performed by an UPLC system coupled with photo diode array (PDA) and fluorescence detectors (FLD) (Waters, Massachusetts, USA). The separation was carried

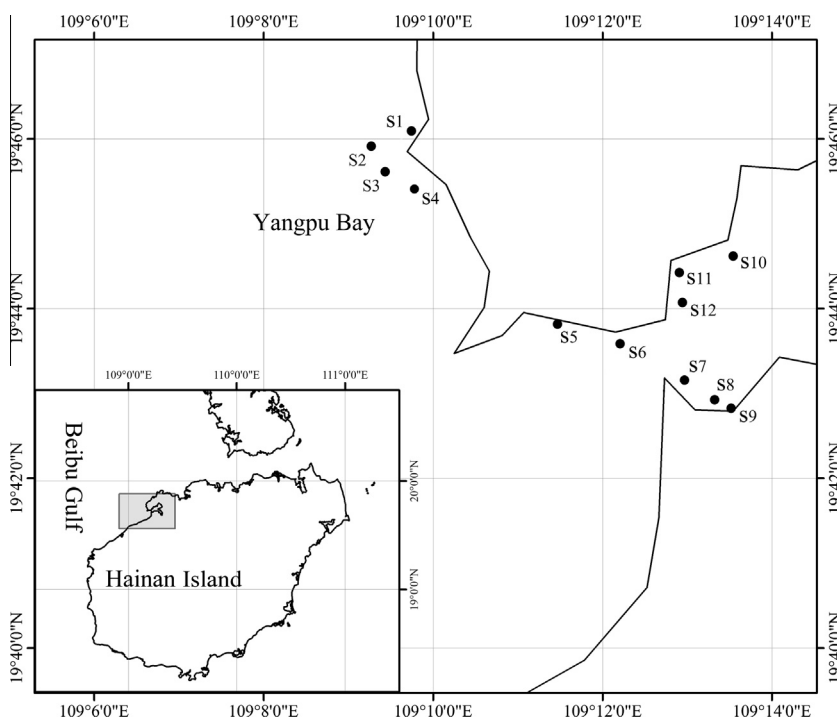


Fig. 1. Location of the sampling sites.

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