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Assessment of the spatial and temporal distribution of legacy persistent organic pollutants and recommendations for sample collection from the surficial sediments of estuaries and seas in China

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ABSTRACT

With the rapid economic development in China, environmental pollution has become a major concern, particularly pollution by persistent organic pollutants (POPs). Thus, these pollutants must be monitored over the long term. In this study, we analyze the distribution levels and sources of POPs in the surficial sediments of Chinese estuaries and seas. Results showed that POPs in sediments significantly distribute spatially and temporally. Furthermore, POPs not only concentrate in densely populated cities, bays, and industrial areas, but also follow the natural distribution of and temporal changes in local industrial structures. Hence, we recommend sampling sites and frequencies to monitor POPs in China over the long term and to defer their analysis.

1. Introduction

China has developed rapidly in terms of economy, industry, and agriculture in the past 20 years. Consequently, the total amount of chemical pesticides and fertilizers used has significantly increased. These substances are considered persistent organic pollutants (POPs). POPs are chemicals that are diverse, synthetic, toxic, and highly resistant to degradation in the environment. These pollutants can also be transported over long ranges. POPs tend to accumulate in food chains (Jones and de Voogt, 1999), thus affecting human health adversely (Harmens et al., 2013). At the Stockholm Convention (UNEP, 2001), the United Nations Environment Program (UNEP) highlighted 12 POPs (dirty dozen), namely, polychlorinated dibenzo-*para*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), hexachlorobenzene, polychlorinated biphenyls (PCBs), aldrin, chlordane, dichlorodiphenyltrichloroethane (DDT), dieldrin, endrins, heptachlor, mirex, and toxaphene. Additional substances and substance groups, are covered by the protocol of the United Nations Economic Commission for Europe (UNECE) on POPs, including PCDDs, PCDFs, and polycyclic aromatic hydrocarbons (PAHs) (UNECE, 1998).

The distribution of POPs has been widely investigated worldwide (Iwata et al., 1993; Kannan et al., 1997; Tanabe et al., 2000; Tanabe, 2006; Minh et al., 2008; Augusto et al., 2013; Zhang et al., 2013; Li et al., 2014). In water systems, POPs are hydrophobic and bind easily to particles before subsiding to the bottom through sedimentation. Therefore, POPs in sediments can be tracked over long term (Rawn et al., 2001). Global studies indicated that the sources of POP emission (such as DDTs and hexachlorocyclohexanes (HCHs)) have shifted from industrialized to developing countries (Loganathan and Kannan, 1994; Fu et al., 2003). In China, POPs have mainly been monitored and analyzed in economically developed cities and regions in the central east, particularly PAHs, PCBs, and organic chlorinated pesticides (OCPs, such as HCHs and DDTs) (Zhou et al., 2001; Yuan et al., 2001; Jiao et al., 2012). Most of these studies are recent; thus, POPs must be tracked long-term to determine their sources and years of entry into environment. Long-term monitoring also verifies the novelty of pollutants, as well as their accumulation or degradation in the environment. Therefore, target samples should be collected from appropriate sites at a certain frequency. By assessing the spatial and temporal distribution of POPs, we can determine their major types, sources, and accumulation

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levels in the study areas. This information can then guide long-term monitoring.

This study analyzes the spatial and temporal distribution of legacy POPs in the surficial sediments of Chinese estuaries and seas. The results indicate that POPs not only concentrate in densely populated cities, bays, and industrial areas in China, but also follow the natural distribution of and temporal changes in the local industrial structure. Prompted by these findings, we propose a rational process of sample collection to facilitate the long-term monitoring and deferred analysis of POPs in sediments of Chinese water bodies. These procedures may guide the formulation of policies and precautionary measures in the country significantly.

2. Methodology

2.1. Targeted areas

To monitor POPs in China over the long term and defer their analysis, we compiled data regarding the concentrations of POPs in well-known estuaries and seas along various parts of the Chinese coastline (Fig. 1 and Table S1). The study areas reflect most industrial structures; therefore, we can obtain statistically significant data regarding the spatial distribution of POPs in China.

To determine the temporal distribution of POPs, we analyzed changes in POP concentrations in Xiamen Bay over the last 20 years. Xiamen Bay is located in southeast China (Fig. 2) within the Xiamen Economic Special Zone. For the past 28 years, the rapid industrial, commercial, and urban development in this zone has greatly stressed the environment of Xiamen Bay and its adjacent areas. These regions represent a microcosm of the progression of China in the last two decades. Thus, Xiamen Bay is a representative sample for the temporal distribution of POPs in China.

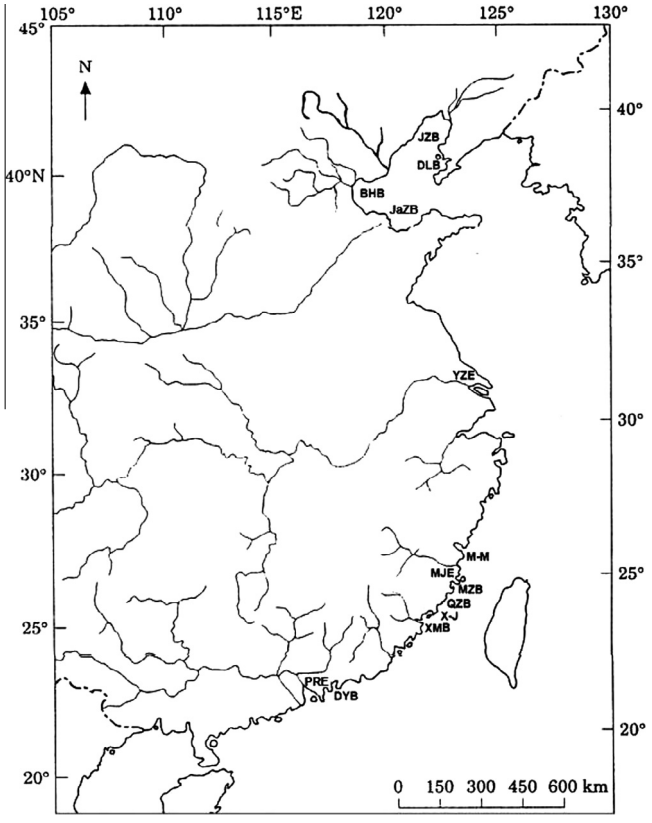

Fig. 1. Monitoring points for this study.

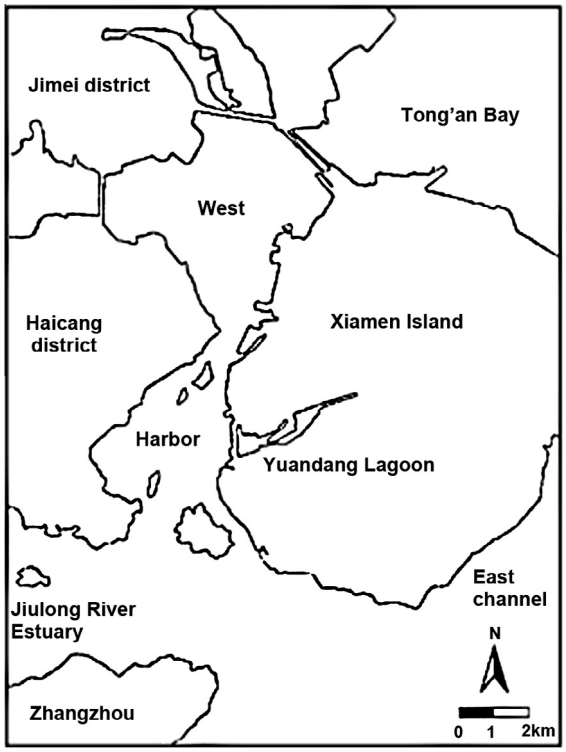

Fig. 2. Location of Xiamen Bay.

3. Results and discussion

3.1. Comprehensive assessment of the spatial distribution of legacy POPs in Chinese sediments

3.1.1. PAHs in sediments

PAHs have many human sources, such as the incomplete combustion of fossil fuels (oil, coal, and natural gas), the combustion of plants and other hydrocarbon compounds, and oil spills (Notar et al., 2001). The main sources of PAHs in offshore environments include urban sewage, industrial wastes, and surface runoff (Tsai et al., 2002), thus implying that regions with highly developed industries have high PAH concentrations (Van Metre et al., 2000).

Several studies have previously determined the distribution of PAHs in sediments of coastal and estuarine regions in China. As shown in Table 1 and Fig. 3, estuarine regions have higher PAH concentrations than other regions, because rivers can deliver PAHs from riparian areas to estuarine regions. In the Pearl River Estuary, the high average level of PAH (1863.0 ng g^{-1}) is primarily ascribed to the high emission of PAHs by the Pearl River Delta (PRD). This area is among the most agriculturally developed and economically thriving regions in China. With respect to Chinese bays, Jinzhou Bay records the highest PAH level (1000.0 ng g^{-1}) as a result of the high level of discharge from the Liao River, Daling River, and Xiaoling River.

3.1.2. PCBs in sediments

PCBs are composed of 209 individual chemical compounds, which are produced through various industrial mixtures by introducing different concentrations of elementary chlorine into biphenyl. The primary source of PCBs is industrial production, including discharged industrial wastewater and slag. Hence, PCB pollution may be prevalent in industrially active areas. As with the spatial distribution of PAHs in sediments, overall PCB distribution follows this pattern: estuaries > bays > seas (Table 2 and Fig. 4). The PCB