



Research papers

Distribution and mass inventory of mercury in sediment from the Yangtze River estuarine-inner shelf of the East China Sea



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ABSTRACT

Mercury (Hg) was measured in 70 sediment samples from the Yangtze River estuarine-inner shelf of the East China Sea (ECS) to evaluate its occurrence, distribution, and deposition flux. Its concentrations were 10–92 ng/g with a mean of 46 ± 17 ng/g. A decrease of Hg concentration with increasing distance offshore suggested a dominance of riverine input. The high levels of Hg observed at the southern inner shelf were partly due to the sorption affinity of fine-grained sediments. Hg concentration was significantly correlated with total organic carbon content and sediment grain size, implying that the nature of sedimentary organic matter and hydrodynamic forces could influence the Hg occurrence. A moderate correlation between Hg with high-molecular-weight polycyclic aromatic hydrocarbons in the YRE suggested that they shared a similar input pathway. The total deposition flux of Hg was estimated to be ~ 52 t/y with a deposition rate of 6–120 ng/cm² y, which indicated that the estuarine-inner shelf of the ECS was a major sink of Hg in the margins off China, and this area could play a significant role in the Hg biogeochemical cycle on a global scale.

1. Introduction

Mercury (Hg) is a highly toxic element that can adversely affect organisms; however, its toxicity depends mainly on its chemical form (Li et al., 2011). Approximately 95% of atmospheric Hg occurs as gaseous elemental Hg⁰ (Schroeder and Munthe, 1998), which has a long atmospheric residence time and can be transported globally due to its low reactivity and stability. The high vapor pressure and low oxidation potential make it uneasy to be scavenged and deposited via wet or dry deposition before oxidation to Hg²⁺ (Hylander and Goodsite, 2006; Lindberg et al., 2007). Elemental Hg can be released into the environment from natural and anthropogenic sources (Gustin et al., 2008). Global Hg emissions are dominated by anthropogenic sources, particularly the combustion of coal and petroleum products (Streets et al., 2005; Keeler et al., 2006; Lindberg et al., 2007).

In aquatic systems, sediments are usually considered as an ultimate sink of pollutants discharged from land-based sources, such as heavy metals and persistent organic pollutants either by atmospheric deposi-

tion or riverine input (Frignani et al., 2005; Elbaz-Poulichet et al., 2011; Li et al., 2012; Hu et al., 2014). Mercury can be accumulated in sediments and be released to surrounding media (Stein et al., 1996). It has potential toxic effects on biological resources, and indirectly, on human health (Peng et al., 2009; Chakraborty et al., 2010; Ernst, 2012; Pena-Fernandez et al., 2014).

Once Hg⁰ is released into the air, it undergoes a series of complex transportation and transformation processes. In the presence of atomic bromine or ozone and hydrogen, Hg⁰ can be oxidized to Hg²⁺ (Holmes et al., 2006). The resulting Hg²⁺ is deposited onto land and water, which can be absorbed by inorganic particles, biological particles, or organic matter (OM) (Ullrich et al., 2001). Inorganic Hg can be converted into organic Hg via biomethylation or abiotic methylation, and because organic forms of Hg, such as methylmercury, can be decomposed by solar ultraviolet radiation, methylation reactions are poor in water; therefore, sediments are considered as the main production site of methylmercury (Jonsson et al., 2012; Moreno et al., 2013; Zhang et al., 2014).

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The coast of the East China Sea (ECS) is one of the most developed regions in China, providing 42% of China's gross domestic product and housing 400 million inhabitants (Yang et al., 2006; Li et al., 2015). The origins of Hg in the regional-scale could be mainly derived from coal combustion and various industrial emissions; and recent rapid economic development and industrialization in this region has resulted in significant deposition of anthropogenic heavy metals, including Hg, into the coastal ECS from the Yangtze River (Streets et al., 2005; Lindberg et al., 2007; Chen et al., 2014; Wang et al., 2014, 2015). As the fifth largest river in water discharge and the fourth largest one in sediment discharge in the world, the Yangtze River has its drainage basin of $\sim 1.94 \times 10^6$ km², which accounts for about 20% of the area of Mainland China (Yang et al., 2006; Bianchi and Allison, 2009). Discharged sediments and associated pollutants are primarily trapped in the Yangtze River Estuary (YRE) and the inner shelf of the ECS due to the net effects of the shear forces of coastal currents (Milliman et al., 1985; Liu et al., 2006, 2007; Bianchi and Allison, 2009; Li et al., 2012).

The distribution and input of Hg into the coastal margin have been recently explored around the world, for examples, Amos et al. (2014) reported the global biogeochemical implications of Hg discharges from rivers and sediment burial, and indicated that the burial of Hg in ocean margin sediments represents a major sink in the global Hg biogeochemical cycle. Liu et al. (2016) simulated and calculated Hg export from mainland China to adjacent seas. There have been some efforts to study Hg contamination in the sediments from several estuaries and coastal regions in coastal China seas, including the Bohai Sea (Luo et al., 2010), the ECS (Fang and Chen, 2010; Meng et al., 2014), the Xiamen Bay (Yan et al., 2010), the Pearl River Estuary, and the coastal region of Hong Kong (Zhou et al., 2007). However, there has been no systematic study found on the distribution and mass inventory of Hg in sediments of the inner shelf in the ECS, and the literature is particularly lacking in source-to-sink processes and the fate of Hg in the river-dominated coastal margin. The estuarine-inner shelf region of the ECS is a typical alongshore sediment dispersal system due to the large river discharge and sedimentary dynamics and thus can serve as an ideal natural experiment area for the study of deposition and fate of anthropogenic pollutants. In this study, we examined the concentration, distribution and inventory of Hg in sediments of the estuarine-inner shelf of the ECS, and identified potential factors influencing the source-to-sink processes of Hg in this region.

2. Materials and methods

2.1. Sampling

In total, the selected 70 surface sediment samples (0–3 cm) from the Yangtze River estuarine-inner shelf of the ECS (Fig. 1) were sampled using a stainless steel box corer on two occasions, first by the *R/V Dong Fang Hong 2* research vessel of the Ocean University of China in 2006 and *R/V Kan 407* research vessel in 2007. The samples were packed in aluminum foil and stored at -20 °C. Most of the surface samples were collected from the muddy areas of the estuary and inner shelf (Fig. 1).

2.2. Mercury analysis

The analytical procedure of Hg followed that described by Leipe et al. (2013). A DMA-80 Hg analyzer (Milestone Scientific Inc., USA) was used to measure Hg concentrations in sediments as it has a very low detection limit (Leipe et al., 2013). To ensure analytical accuracy and precision, standard reference materials (SRMs) were used, including environmental standards (ESS-4, 0.021 µg/g Hg) from the China Environmental Monitoring Station, and certified reference material (CRM) soil (GSS-13) from the Institute of Geophysical and Geochemical Exploration in China. The calibration curve included eight concentrations of Hg, from 0.004 to 20 ng. Standard materials

were measured every 10 samples to control measurement quality and stability. For quality control, CRMs and sample replicates were included in the analytical procedure. The measured concentrations of SRMs were within their certified ranges and the recovery of Hg was $94.33 \pm 9.53\%$ (n=8).

2.3. Dataset consolidation and map generation

The total organic carbon (TOC), median diameter (MD), and ratio of carbon-to-nitrogen (C/N) datasets used in this study were from Hu et al. (2012), and the dataset of 16 polycyclic aromatic hydrocarbons (PAHs) cited in this study was from Lin et al. (2013). Detailed information on the original TOC, MD, and PAH datasets are summarized and abbreviated in Table S1. Contour maps were generated using isopleths based on the consolidation and standardization of these datasets using the Kriging gridding method in the surface mapping system (Surfer version 11.0, Golden Software, Inc).

2.4. Principal component analysis

Principal component analysis (PCA) is a multivariate analytical tool used to determine the sample distribution and study the relationships of measured parameters. Before the analysis, non-detectable values were replaced with concentration values equal to one-half the method detection limits. Then the raw data matrix was Z-score standardized and mid-range normalized to eliminate the influence of different units and ensure that each determined variable had equal weighting in the PCA. The PCA was performed using SPSS 13.0 for Windows (SPSS Inc., Chicago, IL, USA) (Hu et al., 2012). The Hg concentrations, individual compounds of 16 PAHs, MD, and TOC of the 70 sediment samples were used to perform the PCA analysis and SPSS was applied to extract the principal components (PCs) based on the correlation matrix. Principal components with eigenvalues >1 were considered to be factors for further examination.

3. Results and discussion

3.1. The concentration of Hg

The Hg concentration in the study area was 10–92 ng/g with a mean of 46 ± 17 ng/g, lower than those reported from the Northern Gulf of Mexico (10–200 ng/g) (Apeti et al., 2012), comparable to those from the Strait of Malacca, Malaysia (17–114 ng/g) (Zhuang and Gao., 2015), and higher than those in the Eastern Basin and Mediterranean Sea (< 40 ng/g) (Ogrinc et al., 2007) (Table 1). A much higher concentration of Hg (150–1500 ng/g) was observed in the Kaohsiung River mouth, where Hg was emitted from municipal solid waste (Chen et al., 2012). These comparisons indicate that Hg concentrations in marine environments can vary by orders of magnitude (Ogrinc et al., 2007; Abi-Ghanem et al., 2011; Jin et al., 2012; Deng et al., 2013) (Table 1).

Mercury is persistent, bioaccumulated and toxic in the environment (Jiang et al., 2006; David et al., 2009; Neville et al., 2014; Chakraborty et al., 2015). Sediment quality guidelines, effects range low (ER-L), and effects range median (ER-M) were used to assess the potential biological risks of Hg (Table S2). Concentrations lower than the ER-L represent a minimal effect; those between the ER-L and ER-M represent a possible effect; and those above the ER-M represent a probable effect (Long et al., 1995). All of the Hg concentrations in the estuarine-inner shelf of the ECS were lower than the ER-L (Table S2), indicating that the Hg in sediment of the coastal ECS represented low-risk towards organisms.

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