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Distribution and assessment of heavy metals off the Changjiang River mouth and adjacent area during the past century and the relationship of the heavy metals with anthropogenic activity

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ABSTRACT

Forty-three surface sediment samples and one gravity core obtained from the offshore area of the Changjiang River were analyzed for selected heavy metals (Cu, Pb, Zn, Cd, As, Hg) to evaluate the spatial distribution and potential ecological risk during the last century. The results indicated that the sediments are composed of silty sand, sandy silt and silt and were deposited in a relatively stable environment over the last century. The studied marine sediments are fine and easily adsorb heavy metals from aquatic systems. The heavy metal concentrations were found to be enriched in the sediments and were generally closely related to anthropogenic activities. However, the data analysis demonstrated that the levels of heavy metal contamination were below background values during the last century, indicating low ecological risk. Spatially, a higher concentration was found at the entrance to the Changjiang River, while it decreased to the northeast. The vertical distribution of contamination levels and ecological risk can be divided into four periods based on the downcore variation in heavy metals: pre-1940s, 1940s–1970s, 1970s–1990s and the late 1990s to the present. These conclusions form the basis for implementing appropriate policies to protect marine sediment quality.

The coastal and shelf zones, located between land and oceans, serve as a source of marine sediment and a sink for riverine sediments, which are produced from continuous weathering and leaching processes (Chen et al., 2000; Zhang and Liu, 2002). Fine sediments are major carriers of elements from drainage basins to oceans and have a significant impact on sedimentary and ecological systems in estuarine and coastal regions (Zhang, 1999; Zhang and Liu, 2002). Thus, the composition of suspended sediments can preserve information regarding geological and climate type within a drainage basin, as well as impacts from anthropogenic activities. Estuarine and coastal zones are usually polluted by different contaminants from industrial and agricultural processes (Mattiessen and Law, 2002; Ip et al., 2007; Bastami et al., 2012; Pan and Wang, 2012; Gan et al., 2013), with heavy metals comprising an important portion of these materials due to their persistence in the environment (Wang and Rainbow, 2008; Pan and Wang, 2012). The level of anthropogenic heavy metals has increased dramatically since the Industrial Revolution (Nriagu, 1979; Lee et al., 2008; Thevenon et al., 2011), especially after the reform and opening-up policy in China, which has led to a thriving economy accompanied by many environmental problems (Gao and Chen, 2012; Gu et al., 2012; Zhang et al., 2012; Zhao et al., 2013).

Accordingly, it is necessary to investigate the occurrence and spatial distribution of heavy metals in the expansive ocean area in China. Recent studies have mainly focused on the assessment of heavy metal pollution in different areas, particularly in the Bohai Sea (Zhong et al., 2011; Feng et al., 2011; Li et al., 2013), Yellow Sea (Gao et al., 2003; He et al., 2008; Sheng et al., 2013), East China Sea (Chen et al., 2004; Liu and Fan, 2010; Liu et al., 2011) and South China Sea (Zhu et al., 2011; Dou et al., 2013; Hu et al., 2013; Gan et al., 2013).

The Changjiang River, which is one of the five largest rivers in the world in terms of both sediment load and water discharge (Milliman et al., 1985), transports vast amounts of terrestrial materials to the East China Sea (ECS) (Huang et al., 2001). Changjiang River sediments are mainly fine-grained and therefore easily

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absorb heavy metals. In addition, as the earliest industrialized and most developed region in China, rapid economic development in the region has led to large amounts of heavy metals and industrial, agricultural and domestic wastes being delivered into the Changjiang Estuary and ECS. About 15,000 tons of heavy metals were discharged into the abovementioned area in 2002, and this value will continue to increase in the next decade (Lin et al., 2002). The sediments in the Changjiang Estuary and its subaqueous delta have a relatively fine grain size, resulting in increased heavy metal deposition (Lin et al., 2002; Guo et al., 2007). Therefore, this study area is ideal for investigating the heavy metal distribution and assessing the pollution level of the region. As an important economic area, the Changjiang Estuary and adjacent subaqueous delta play an important role in economic development and natural environmental evolution. The temporal–spatial distribution and assessment of heavy metal pollution over the past century are the basis for interpreting their mechanisms of transportation and accumulation and exploring the relationship between heavy metal pollution and anthropogenic activities. The results of this study would provide a useful aid for sustainable marine management in the region.

A total of 43 surface sediments were collected by grab sampler in the offshore area of the Changjiang Estuary. In addition, a 375-cm-long sediment core (CJ0702) was collected from the mud deposition area (31°N, 122.67°E) using a gravity corer (Fig. 1). Once transported to the Qingdao Institute of Marine Geology laboratory, the core was split, restored, described and subsampled. The entire core was divided into samples (about 1–2 cm thick) for high-resolution analysis. Grain size analysis, dating analysis and elemental analysis of these samples were then conducted in the laboratory.

Prior to grain size analysis, samples were pretreated with 10% H_2O_2 and 0.1 mol/L HCl to remove organic matter and biogenic carbonate, respectively. The samples were then analyzed using a Mastersizer 2000 laser particle size analyzer, which had a measurement range of 0.02–2000 μm, a resolution of 0.01Φ and a reproducibility >3%. A total of 31 samples were selected from intervals between 2 and 10 cm in the sediment core and were subjected to ^{210}Pb, ^{226}Ra and ^{137}Cs analysis to support calculation of sediment accumulation rates at the sampling site. ^{210}Pb activities were determined by a BE3830 gamma-ray spectrometer (Canberra Ltd., USA) at the Experiment Testing Center of the Qingdao Institute of Marine Geology.

The elemental geochemistry of these samples was measured at the Experiment and Testing Center of Qingdao Institute of Marine Geology, China Geological Survey. The major and trace elements were determined by an X-ray fluorescence spectrometer (XRF, Phillips PW 2440) and inductively coupled plasma mass spectrometry (ICP-MS, AFS-920) according to the method described by Xia et al. (2008). Blanks and China Steam Sediment Reference Materials (GBW07345, GSD4 and GSD9) were included in the analyses as part of quality assurance and quality control (QA/QC) (see Dou et al., 2013 and Hu et al., 2013 for details).

Based on sediment grain-size analysis, the sediments in the offshore area were composed of silty sand, sandy silt and silt, with occasional sand stations. These marine sediments are fine and easily adsorb heavy metals in aquatic systems. Core CJ0702 was generally uniform in color and sediment content, and the sediments were deposited in a relatively stable environment. The sediment was mainly composed of silt and clay, with occasional sand (Fig. 2). Variations in grain size and sedimentary structure (Fig. 2) enabled division of the core into three sections: 0–48 cm, 48–332 cm and 332–375 cm. For dating purposes, 31 samples were analyzed for ^{210}Pb and ^{137}Cs activities. The ^{210}Pb concentrations decreased steadily from the surface to a depth of 120 cm (Fig. 2), after which they maintained a constant value. A mean accumulation rate of 3.11 cm/yr was calculated from the ^{210}Pb data (standard deviation of $r^2 = 0.56$). Based on this accumulation rate, core CJ0702, which was 375 cm long, was found to cover a time span of about 120 years, from approximately 1887 to 2007.

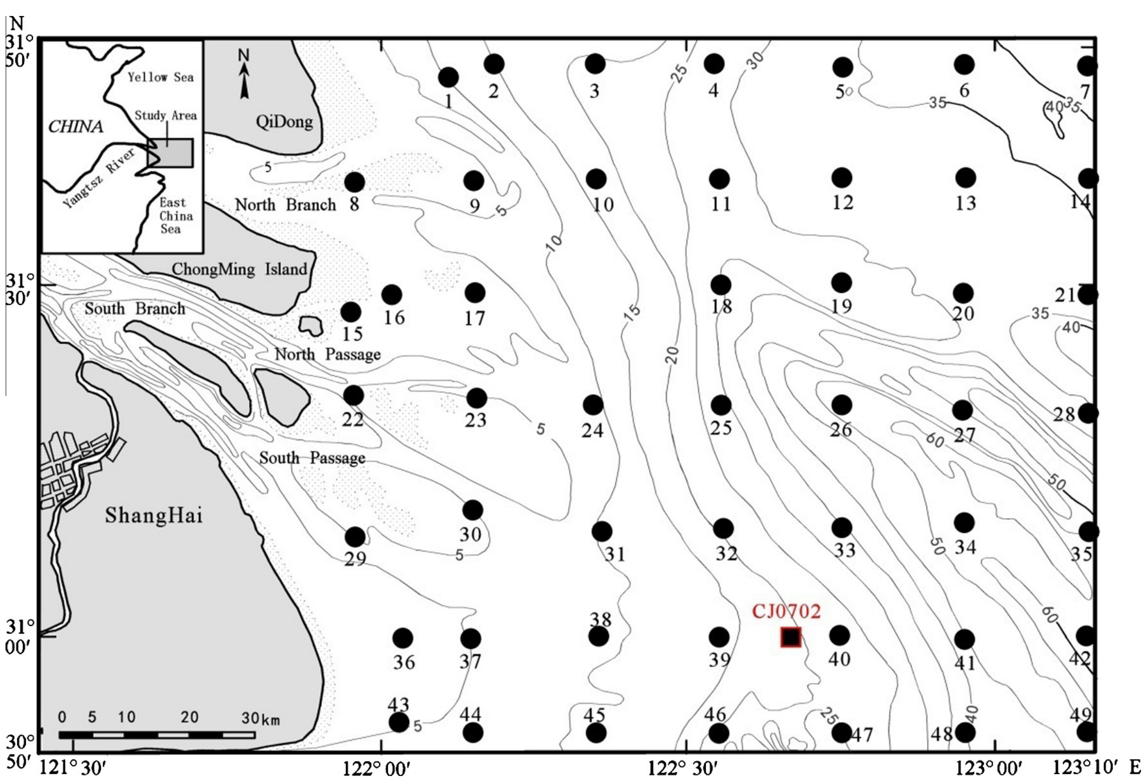

Fig. 1. Locations of surface sediment samples and core CJ0702 (no samples were collected from stations 7, 14, 19, 24, 29 or 48).

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