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Comprehensive large-scale investigation and assessment of trace metal in the coastal sediments of Bohai Sea

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ABSTRACT

The Bohai Sea is characterized as a semi-closed sea with limited water exchange ability, which has been regarded as one of the most contaminated regions in China and has attracted public attention over the past decades. In recent years, the rapid industrialization and urbanization around the coastal region has resulted in a severe pollution pressure in the Bohai Sea. Although efforts from official government and scientific experts have been made to protect and restore the marine ecosystem, satisfactory achievements were not gained. Moreover, partial coastal areas in the Bohai Sea seemingly remain heavily polluted. In this study, we focused on five coastal regions around the Bohai Sea to study the spatial distribution pattern of trace elements in the sediments and their ecological risk. A total of 108 sediment samples were analyzed to determine the contamination degree of trace elements (Cu, Cd, As, Pb, Zn, Cr, and Hg). Contamination factor (*CF*), pollution load index (*PLI*), geoaccumulation index (I_{geo}), and potential ecological risk index (*PERI*) were utilized to assess the pollution extent of these metals. Spatial distribution patterns revealed that the sedimentary environments of coastal Bohai were in good condition, except Jinzhou Bay, according to the Marine Sediment Quality of China. The concentrations of Hg and Cd were considerably higher than the average upper crust value and presented high potential ecological risk and considerable potential ecological risk, respectively. The overall environment quality of the coastal Bohai Sea does not seem to pose an extremely serious threat in terms of metal pollution. Thus, the government should continue implementing pollution control programs in the Bohai Sea.

1. Introduction

The Bohai Sea is a semi-closed shallow marginal sea located in the northeast of China. Given its special geographical and oceanographic characteristics, the Bohai Sea is sensitive to the environmental impacts of human activities in the catchment and sea areas. The Bohai Sea is connected to the Yellow Sea and bounded by the Bohai Strait between the Laotieshan in the southern Liaodong Peninsula and in the northern Shandong Peninsula. The Bohai Sea is surrounded by the three provinces of Liaoning, Hebei, and Shandong, and one municipality of Tianjin. The Bohai Sea covers an area of approximately 77,000 km^2, with an average depth of 18 m. Three major bays form inside the Bohai Sea, namely, Liaodong Bay to the north, Bohai Bay to the west, and Laizhou Bay to the south. The surrounding area of the Bohai Sea experiences rapid economic development, accompanied by substantial amounts of land-based pollutants. The ecosystem health of the Bohai Sea has attracted attention in recent years due to intensive human activities along the coastline. The Bohai Sea suffers from rapid industrialization and urbanization of adjacent terrestrial areas and is considered one of the most contaminated coastal areas in China (Hu et al., 2013; Liang et al., 2004; Wang and Wang, 2007; Xu et al., 2016). The Bohai Sea was formerly a famous fishing ground with several fishery resources. High fishing intensity, environmental pollution and habitat degradation have resulted in the sharp fishery decline in recent years (Jin, 2004; Xu et al., 2010). An urgent concern is that the Bohai Sea will degrade into a dead sea if no effective protection action is implemented. Thus far, investigation and assessment of heavy metals covering the entire area of the Bohai Sea have been inadequate. Gao et al. (2014) conducted a comprehensive literature review of the pollution status of trace elements (Cu, Cd, As, Pb, Zn, Cr, Hg, and Ni) in the seawaters, sediments, and inhabitant organisms over the past decades in the Bohai Sea. The authors concluded that metal pollution is closely related to the fast economic growth of the local region and is high in the coastal areas. However, considerable work has concentrated on a few

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local areas. Comprehensive large-scale coastal investigation is lacking, which makes comparison a difficult task.

Although heavy metals are naturally originated elements that exist throughout the earth's crust, anthropogenic metal pollution that accompanies rapid economic development has attracted global attention (Cheng, 2003; Furness, 2018; Pan and Wang, 2012). Heavy metals are regarded as trace elements because of their occurrence in trace concentrations in various matrices. However, these metals notoriously cause serious contaminations because of their toxicological, ubiquitous, persistent, and irreversible characteristics in the environment. Reported sources of anthropogenic heavy metal include industrial, agricultural and domestic effluents, and atmospheric sources (Förstner and Wittmann, 2012). Excessive amounts of metals are released to estuarine and coastal environments through land-based sources as a result of rapid urbanization along the coastlines (Feng et al., 2004; Qiao et al., 2013). Heavy metal residues in polluted habitats accumulate through the dietary pathways of marine organisms, which may threaten the health of human beings (Zhang et al., 2012). Previous studies indicated that the coastal ecosystems in China are now facing increasing metal pollution (Gao et al., 2014; Wang et al., 2013).

Although contaminants in the marine environment derive from various sources, the majority of contaminants adsorb to particulate phases and inhabit the sediment. Coastal sediment has been regarded as an important sink and repository for trace elements. In addition, metal-contaminated sediments are frequent, with metal concentrations several orders of magnitude greater than pre-industrial or background values. Moreover, sediments will release other contaminants when the local environment changes (Eggleton and Thomas, 2004). Therefore, an investigation of metal concentrations in the sediment is important to evaluate the health status of coastal ecosystems and understand the possible environmental risks caused by anthropogenic activities.

In this study, we chose a few representative sites as the study area and conducted a systematic risk assessment of heavy metals in the coastal sediments of the Bohai Sea. Hence, this study mainly aimed to (1) investigate the spatial distribution of trace elements and assess the current situation of trace metal pollution in the coastal sediment of the Bohai Sea, (2) assess the ecological risk of heavy metals in the sediment, and (3) identify the main controlling factors in the distribution of metal contaminants and possible sources of pollution.

2. Materials and methods

2.1. Description of sample area

A total of 108 sampling sites were distributed over the coastal area of the Bohai Sea. These sites were mainly from five regions, including Shuangtaizi Estuary (27 sites), Jinzhou Bay (12 sites), Luanhe Estuary (24 sites), Bohai Bay (20 sites) and Laizhou Bay (25 sites) (Fig. 1). Sampling sites were selected to cover the important estuarine and coastal areas of the Bohai Sea. Shuangtaizi Estuary is located in the northeast of Bohai Sea, is an outlet of three main rivers, including Daliao River, Shuangtaizi (Liao) River and Daling River. Jinzhou Bay, located in northwest of Liaodong Gulf, is a semi-closed shallow water area. Six rivers including Lianshan River, Wuli River, Lao River, Cishan River, Zhouliu River and Tashan River flow into Jinzhou Bay (Wang et al., 2010). It is famous as an old industrial base, and become one of the most polluted coastal area in China. Luanhekou Estuary is located on the northwest coast of the Bohai Sea with water depths < 20 m. Freshwater and sediment discharges have decreased greatly since the 1980s due to large dams and reservoirs built along the Luanhe River (Liu et al., 2016). Bohai Bay is located in the west of the Bohai Sea, near the city of Tianjin. Bohai Bay is a typical semi-enclosed coastal area and has limited water exchange with the ocean. Large quantities of industrial and domestic wastewater discharges flow into Bohai Bay from rivers of Beijn-Tianjin. The western coast of Bohai Bay locates the Tianjin Ports, the 10th largest port in the cargo throughout in the world. Laizhou Bay is located in the southern part of Bohai Sea, accounting for up to 10% of the total area (Zhuang and Gao, 2014). It's a semi-closed shallow area with average water depth less than10 m. There are more than a dozen of rivers running into the Laizhou Bay, among which Yellow River, Xiaoqinghe River and Wei River are the most important.

2.2. Sample collection

Throughout the survey, a global positioning system (GPS) was used to locate all sampling sites. Surface sediment samples (0–5 cm) were collected using a grab sampler in August 2015. At each site, three surface sediments were collected and placed into dark-colored polyethylene bags and sealed. After sampling, the samples were transported to the laboratory and stored at 4 °C for further analysis.

2.3. Analytical methods

All digestive processes were referred to China National Standards (GB 17378.5-2007). Sediment samples (20 g) for element analysis were oven-dried (< 60 °C), and then ground to powder (< 63 μm). Briefly, the powered samples (0.5 g) were digested in 20 mL of a 5:4:1 guaranteed reagent HNO_3 + $HClO_4$ + HF on a heating plate for 10 h to dryness. Afterwards, the residue was extracted with HNO_3 and diluted to volume. The total organic carbon (TOC) was measured with an elemental analyzer (Vario EL-III). The concentrations of the dissolved heavy metals were determined by inductively coupled plasma mass spectrometry (ICP-MS, Thermo X series) for Cd, Pb, Zn, Cu, and Cr, whereas the contents of Hg and As were determined using an atomic fluorescence spectrometer (AFS-920). Grain size of the sediment samples was analyzed using a Mastersizer-2000 particle size analyzer (Malvern, UK). Prior to particle size analysis, the fresh subsamples were pretreated with 10% H_2O_2 and 1 mol/L HCl for 24 h to remove organic matter and biogenic carbonate. The samples were divided into four fraction on the basis of particle size: clay (< 2 μM), silt (2–40 μM), fine sand (40–200 μM) and coarse sand (> 200 μM) (Ip et al., 2007). About 20 g sediment samples for element analysis were oven-dried (< 60 °C), and then ground to powder (< 63 μm). About 0.1 g powered samples were totally digested in a 60-mL Teflon vessel with a mixed solution of HNO_3 and $HClO_4$. The solution was diluted with deionized water and filtered quantitatively in a 25-mL colorimetric tube. One blank was digested similarly for quality control.

2.4. Quality assurance and quality control (QA/QC)

The analytical data quality was guaranteed through the implementation of quality assurance and quality control, with method blanks and standard reference materials. Three replicates were conducted for the determination of the total content of the metals. All analyses were carried out in duplicate, and the results were expressed as the mean. The precision of the analytical procedures, expressed as the relative standard deviation (RSD), was > 5% for all tests. The quality of the analytical procedures was tested by the recovery measurements on the Chinese national geo-standard (GBW-07333 and GBW-07314). The results were consistent with the reference values, and the differences were within 10%.

2.5. Assessment of metal contamination

In order to best characterize heavy metal pollution extent in the coastal sediments of Bohai Sea, different empirically derived sediment quality guidelines were applied in this study.

2.5.1. Sediment quality guidelines

Sediment quality guidelines (SQGs) is a simple standard for assessing the risk of metal pollute in an aquatic ecosystem. In this study, two