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Transport pathway and depocenter of anthropogenic heavy metals off the Shandong Peninsula, China

Gang Xu [a, b, c, d], Jian Liu [a, b, c, d, *], Shaofeng Pei [a, b, c, d], Maosheng Gao [a, b, c], Xianghuai Kong [a, b, c]

[a] Key Laboratory of Marine Hydrocarbon Resources and Environmental Geology, Ministry of Land and Resources, Qingdao Institute of Marine Geology, Qingdao, 266071, China
[b] Laboratory for Marine Geology, Qingdao National Laboratory for Marine Science and Technology, Qingdao 266061, China
[c] Key Laboratory of Coastal Wetland Biogeosciences, China Geological Survey, Qingdao 266071, China
[d] Qingdao Institute of Marine Geology, Qingdao, 266071, China

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ABSTRACT

Surface sediment grain size as well as the spatial distribution, sources and geochemical baseline levels of heavy metals in the south Shandong Peninsula clinoform were analyzed to determine the transport pathway and main depocenter of anthropogenic heavy metals off the peninsula. Results showed that the surface sediments were primarily silt-sized components, and the fine grain matter mainly originated from the Yellow River and rivers around Laizhou Bay. Heavy metals Cu, Pb, Zn, Cr, and Cd were predominantly from natural sources and their spatial distributions were controlled by grain size; conversely, anthropogenic As (concentration above geochemical baseline level 10.9 mg/kg) was principally derived from human activities, and its transportation from the Yellow River and Laizhou Bay was controlled by the Shandong Coastal Current off the Shandong Peninsula. Furthermore, the anthropogenic As was deposited in three main areas, that is, the Yellow River estuary, Laizhou Bay, and south Shandong Peninsula clinoform.

1. Introduction

Heavy metal contamination in aquatic ecosystems is of considerable concern due to its high toxicity, easy bioaccumulation and non-degradation (Bozkurt et al., 2000; Varol, 2011; Waheed et al., 2013). With rapid urbanization, industrialization and agriculturalization, huge amounts of heavy metals are delivered into aquatic ecosystems by human activities, and include industrial effluent, mining and refining waste, agricultural drainage, and domestic discharge (Romic and Romic, 2003; Iqbal and Shah, 2014; Palma et al., 2015; Bo et al., 2015). Moreover, heavy metal input into aquatic environments via agricultural activities has continued to increase over the past few decades in China (Li et al., 2008). Anthropogenic heavy metals transported into natural water systems are mainly incorporated into bottom sediments through adsorption, flocculation, and precipitation in the water column (Botte et al., 2007). However, anthropogenic metals that settle out of the water column are more likely to be resuspended and redissolved into pore water, from which sediment-associated heavy metals can be released into the overlying water by diffusive fluxes (De Mora et al., 2004). Therefore, bottom sediments not only act as the main sink for anthropogenic heavy metals, but are also potential secondary sources of pollutants in aquatic environments (Singh et al., 2005; Shipley et al., 2011). As bottom sediments show relative stability over space and time, consistent assessment of heavy metal contamination is possible (Pekey, 2006; Tuncer et al., 2001). Thus, sediment could be an effective indicator for evaluating pollution conditions and causes.

As part of the Bohai Economic Circle, Laizhou Bay contains three port industrial zones (Dongying Port Industrial Zone, Weifang Port Industrial Zone, and Laizhou Port Industrial Zone) (Xu et al., 2015a). In the Weifang Port Industrial Zone, more than 400 chemical enterprises are located along the southwestern coast of Laizhou Bay due to its abundant seawater and underground brine resources. Consequently, many point pollution sources have been created by

* Corresponding author. Key Laboratory of Marine Hydrocarbon Resources and Environmental Geology, Ministry of Land and Resources, Qingdao Institute of Marine Geology, Qingdao, 266071, China.
E-mail address: liujian0550@vip.sina.com (J. Liu).

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industrial wastewater, agricultural discharge, and non-purified or insufficiently purified sewage, which are frequently discharged into the surrounding rivers and bay with considerable anthropogenic heavy metal contamination. In addition, large amounts of heavy metals and organic pollutants from industrial and sewage wastewater of estuarine cities continue to be discharged directly into Xiaoqing River, which has deteriorated the local aquatic environment (Zhuang and Gao, 2014). Most anthropogenic heavy metals mainly deposited in the Laizhou Bay, and some were carried out the Laizhou Bay by the ebb tide and deposited in the other areas (Xu et al., 2015a).

The Yellow River plays a critical role in the economic development of China, supporting a population of 107 million, irrigating 15% of the agricultural land, and contributing to 9% of China's gross domestic product (Miao et al., 2010). As reported in the 2006 Bulletin of Yellow River Water Resources, ~4.3 Gt/yr of wastewater is discharged into the river, with 74% from secondary industry followed by domestic sewage (20%) and tertiary industry (6%) (Hu et al., 2015). In addition, fertilizer application on agricultural lands has elevated the heavy metal content in the river (Tang et al., 2010). Anthropogenic heavy metals from the Yellow River were mainly deposited in the subaqueous delta of the Yellow River, and some of them were might transported to the Shandong Peninsula clinoform by the Shandong Coastal Current (SDCC).

Three dimensional bathymetric morphology (Liu et al., 2004) has shown a subaqueous clinoform wrapping around the eastern Shandong Peninsula, called the Shandong mud wedge (Liu et al., 2002) or the Shandong clinoform (Liu et al., 2004, 2007). It is more than 40 m thick north of Shandong Peninsula and thins seaward, where it extends at least 150 km east of 123.5 °E (Liu et al., 2002, 2007). The Shandong Peninsula clinoform represents a direct escape route of Yellow River sediment into the South Yellow Sea (Alexander et al., 1991), and may also indicate the escape route of river sediment surrounding Laizhou Bay. In aquatic environments, heavy metals are predominantly transported in association with particulate matter (Li et al., 2000; Ridgway and Shimmield, 2005). Therefore, the Shandong Peninsula clinoform might be the depocenter of anthropogenic heavy metals from the Yellow River and rivers surrounding Laizhou Bay.

The contamination, sources, and transport of heavy metals in the surface sediments from Laizhou Bay were systematically studied and reported by Xu et al. (2015a), contamination and sources of heavy metals in surface sediments from the north Shandong Peninsula were described by Xu et al. (2015b), and the spatial distribution and ecological risk of heavy metals in surface sediment from the east Shandong Peninsula were reported by Liang et al. (2008). In addition to our research group, many other studies have been conducted on heavy metal pollution and toxicity in the surface sediments of Laizhou Bay (Zhuang and Gao, 2014; L. Li et al., 2014), the Yellow River (Hu et al., 2015), and Shandong Peninsula (Li et al., 2013). However, systematical research on the sources and transport pathway of heavy metals off the entire Shandong Peninsula is rare, and the transportation and depocenter of anthropogenic heavy metals from the Yellow River and rivers surrounding Laizhou Bay remain unclear. To clarify these questions, heavy metal sources were identified based on principal component analysis (PCA), and the geochemical baseline levels (GBL) of heavy metals were determined by the cumulative distribution function (CDF) in the surface sediments collected in the south Shandong Peninsula clinoform. The transport pathway and depocenter of anthropogenic heavy metals off the Shandong Peninsular were systematically studied based on the surface sediment samples collected in 2006, 2012, 2013, and 2014 (Fig. 1a).

2. Regional setting

The Yellow Sea is a broad, relatively shallow epicontinental sea above a flat, tectonically stable shelf. The Shandong Peninsula separates the South and North Yellow Sea (Fig. 1a). Water depths in the North Yellow Sea are generally less than 60 m, but deepen progressively southward and southeastward in the South Yellow Sea, where a SE-NW oriented trough with a maximum water depth of 100 m is defined by the 80 m isobath. Another marked depression in the topography of the Yellow Sea floor is located east of the eastern end of the Shandong Peninsula, where water depths locally exceed 70 m.

The modern Yellow River, which presently discharges into the western Bohai Sea, is known for its considerable annual sediment load (Milliman and Syvitski, 1992; Saito et al., 2001), although the annual load has decreased sharply to 0.15 Gt/yr between 2000 and 2005 (Wang et al., 2007; Kong et al., 2015), and now represents only 14% of the widely cited estimate of 1.08 Gt/yr (Wang et al., 2007). Most sediment discharged from the Yellow River is trapped in the subaqueous delta or within 30 km of the delta front by gravity-driven underflow (Wright et al., 1988), with only 1%–15% of total discharge being transported out of Bohai Sea and into the Yellow Sea (Martin et al., 1993; Alexander et al., 1991).

The circulation patterns in the Bohai and Yellow seas are dominated by the Yellow Sea Warm Current (YSWC) and coastal currents (Fig. 1a). The YSWC is a branch of the Tsushima Current that flows northwestward into the South Yellow Sea and carries warm, salty water into the Yellow Sea, roughly along the Yellow Sea Trough. In winter, when the shelf water column is nearly homogenous, the YSWC can intrude into the North Yellow Sea (Lan et al., 1986) and even into the Bohai Sea (Liu et al., 1998). The coastal currents along the southern coast of Bohai Sea and the western coast of the Yellow Sea flow persistently southward due to the strong inflow from the Yellow River.

3. Materials and methods

3.1. Collection of surface sediments

In the summer of 2006, 2012, and 2013, 240 surface sediment samples off the east Shandong Peninsula (Liang et al., 2008), 150 surface sediment samples in Laizhou Bay (Xu et al., 2015a), and 102 surface sediment samples off the north Shandong Peninsula were collected (Xu et al., 2015b), respectively (Fig. 1a). In the summer of 2014, 197 surface sediment samples were collected in the south Shandong Peninsula clinoform of China (Fig. 1b). The sampling stations in 2006, 2013, and 2014 were located in the Shandong Peninsula clinoform. The sub-samples for analyses were taken from the top 2 cm of the box center. Each bulk sample was divided into two parts for determination of elements and grain-size analysis, respectively. Sediment samples for element analysis were stored frozen to render them more stable.

3.2. Grain-size analysis

Sediment samples were pretreated with 10% H_2O_2 to digest the organic matter. Excessive H_2O_2 solution was removed by heating and evaporation, and then 0.5% of sodium hexameta-phosphate was added to disperse the sample completely. The mixture was analyzed with a laser particle size analyzer (Mastersize-2000, Malven Instruments Ltd., UK) at the Qingdao Institute of Marine Geology, China Geological Survey. Grain size parameters were calculated following the formula of Folk and Ward (1957).