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## Baseline

## Spatial distribution and ecological risk assessment of heavy metals in coastal surface sediments in the Hebei Province offshore area, Bohai Sea, China

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## ABSTRACT

Seven hundred and nine surface sediment samples, along with deeper sediment samples, were collected from Hebei Province along the coastal section of the Bohai Sea, China, and analyzed for grain size, concentrations of organic carbon (Corg) and heavy metals (Cu, Pb, Zn, Cr, Cd, As, and Hg). Results indicated that the average concentrations in the sediments were 16.1 mg/kg (Cu), 19.4 mg/kg (Pb), 50 mg/kg (Zn), 48.8 mg/kg (Cr), 0.1 mg/kg (Cd), 8.4 mg/kg (As), and 20.3 μg/kg (Hg). These concentrations generally met the China Marine Sediment Quality criteria. However, both pollution assessments indicated moderate to strong Cd and Hg contamination in the study area. The potential ecological risk index suggested that the combined ecological risk of the seven studied metals may be low, but that 24.5% of the sites, where sediments were finer and higher in Corg concentration, had high ecological risk in Hg and Cd pollution.

Coastal areas often serve as major sinks for sediments and associated contaminants from enhanced economic and industrial activity (Hu et al., 2013; Pan and Wang, 2012). With rapid coastal development, increasingly more anthropogenic heavy metals are discharged into estuarine and coastal areas through rivers, runoff, and land-based point sources (Bastami et al., 2015; Suresh et al., 2015). Yet, only a small portion of these metals remains dissolved in the water long enough to be transported into the deep sea; the greatest majority of heavy metals are deposited in coastal sediments (Aa et al., 2016), posing an unknown but persistent risk.

Accumulated heavy metals in sediments can become entrained in the food web as contaminants are subsequently bio-accumulated and bio-magnified, and taken up by humans directly or indirectly through consumption of or contact with seafood, respectively. Ingestion of heavy metals in this fashion can result in potential long-term implications for human and ecosystem health from heavy metal persistence and toxicity (Long et al., 1995; Pan and Wang, 2012; Praveena et al., 2007; Qiu et al., 2011). Therefore, it is important to investigate the spatial distribution of heavy metal concentrations in coastal sediments and evaluate their ecotoxicological risk by presence along contaminated coast lines versus non-polluted coast lines or areas. In this study, we contribute to the understanding of the geochemical distribution of heavy metals in coastal sediments and provide basic distributional information for environmental protection and pollution

control along a particularly important area of the Chinese coast (Long et al., 1995; Simpson and Batley, 2007; Simpson et al., 2005).

Hebei Province is located along the Western Bank of the Bohai Sea. Hebei Province borders Liaoning Province to the north, Shandong Province to the south, and is divided by the City of Tianjin toward the Bohai Sea, providing considerable contamination risk to coastal sediments from this major Chinese city. In addition, Qinhuangdao, Tangshan, and Cangzhou cities are also located along the coast of Hebei Province, and there are > 60 rivers flowing into the Bohai sea along this coast facilitating major industrial connections to the sea and nearshore sediments. Hebei Province is one of the four most densely populated and industrialized zones of the Bohai Sea, possessing oil, chemical, iron, and steel industrial bases and large ports. A large number of domestic sewage, industrial wastewater, and aquacultural wastewater facilities discharge into the Bohai Sea, contributing to a deteriorating ecological environment. In fact, the Hebei Province coast has become one of the most polluted areas in China (Liu et al., 2011; Sun et al., 2016). This is a major concern, as a huge amount of contaminants, including heavy metals, are deposited in the sediments as potentially long-term sinks but with unknown fate.

Heavy metals in sediments directly affect the benthos (Pan and Wang, 2012), but with tide and storm agitation, they can become re-dissolved, re-suspended, and either re-deposited or transported to affect additional nearshore and oceanic environments. Those heavy metals

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not transported offshore will cause the secondary pollution of coastal water bodies (Islam et al., 2015), and we know very little about this process. Consequently, heavy metal pollution status of Hebei Province offshore areas has attracted considerable attention from both scientific and regulatory communities (Fang et al., 2016; Han, 2011; Liu et al., 2015; Luyi, 2010; Ma et al., 2016; Zhi-Wei et al., 2008). Coastal sediments play a major role in how heavy metals are stored, conveyed to humans and ecosystems, or transported offshore.

Recent sediment bio-accumulation and bio-magnification studies are limited in quantity of samples, and are often focused to assess the heavy metal pollution risk from specific river outlets, with limited larger scale environmental assessment for Hebei, or any Chinese province. As a result, we analyzed 709 surface sediment samples obtained from the coastal areas of Hebei Province in a single month (June 2013), to understand the spatial distribution of selected heavy metals in Hebei Province. Our specific objectives are to: (1) analyze the spatial distribution of heavy metals (As, Cd, Cr, Cu, Hg, Pb, and Zn) in the coastal surface sediments of Hebei Province; (2) assess the heavy metal contamination using a geoaccumulation index ( $I_{geo}$ ) (Muller, 1969) and Enrichment Factor ( $EF$ ) process (Buat-Menard and Chesselet, 1979); and (3) evaluate the potential ecological risks of heavy metals using an ecological risk index ( $RI$ ) (Hakanson, 1980).

Seven hundred and nine surface sediment samples (0–5 cm depth) and 188 deep layer samples (150–200 cm) were collected from offshore areas associated with Hebei Province in June 2013 (Fig. 1). Samples were distributed evenly across the study area and within the depth of 20 m, which represented ~484 km of coastline and a sediment surface area of ~3000 km<sup>2</sup>. Most of the surface samples were obtained by a stainless steel grab sampler lowered from a boat, but some surface samples were collected using a diver in situations when the water depth was < 1 m. The average soil sampling depth was 5 cm. Deep layer samples were subsampled at 1.5–2 m below some of the surface samples using a gravity piston corer. The sedimentation rate in this area was generally < 2 cm/year. Therefore, the deep layer samples are representative sediments deposited before 1938, when there was not much industrial pollution. All sediments were air dried at room temperature for several days. When the samples were completely dry, they were broken and ground into a powder, sieved through a #10 mesh (< 2 mm) nylon screen, re-sealed into new polyethylene bags, and sent to a laboratory for chemical analysis.

All 709 surface sediment samples and 188 deep layer samples were analyzed for heavy metal concentration, but only the 188 surface samples where the deep layer sediments were sampled were analyzed for grain size. Sediment grain sizes were measured at the Experimental & Testing Center of Qingdao Institute of Marine Geology, China Geological Survey, Qingdao, China. Approximately 1 g of sample was treated with 10 ml of 5% hydrogen peroxide to decompose organic matter (OM) and then dispersed and homogenized via ultrasonic sonication for 30 s prior to instrumental analysis. Grain size was determined using a laser particle size analyzer (Mastersizer 3000, Malvern Instruments Ltd., UK) that could distinguish particles in the size range 0.02–2000 μm; the resolution ratio was 0.01 φ, and the analytical error was better than ± 2%. Organic carbon (Corg) was determined by wet oxidation in an acid dichromate solution followed by back titration of the remaining dichromate using a ferrous ammonium sulfate solution.

Heavy metals and organic carbon (Corg) in sediment samples were also analyzed at the Qingdao Institute of Marine Geology following the detailed methods described by Zhao et al. (2016), but with study-specific analytical methods listed and detection limits described specifically in Table 1.

The metal enrichment factor ( $EF$ ) was first proposed by Buat-Menard and Chesselet (1979), and is now commonly used to estimate metal sources and contamination levels in sediments of riverine, estuarine, and coastal environments. For data normalization purposes, the sample metal concentrations were initially normalized by using conservative elements in sediment samples, such as Al, Fe, Co, and Sc,

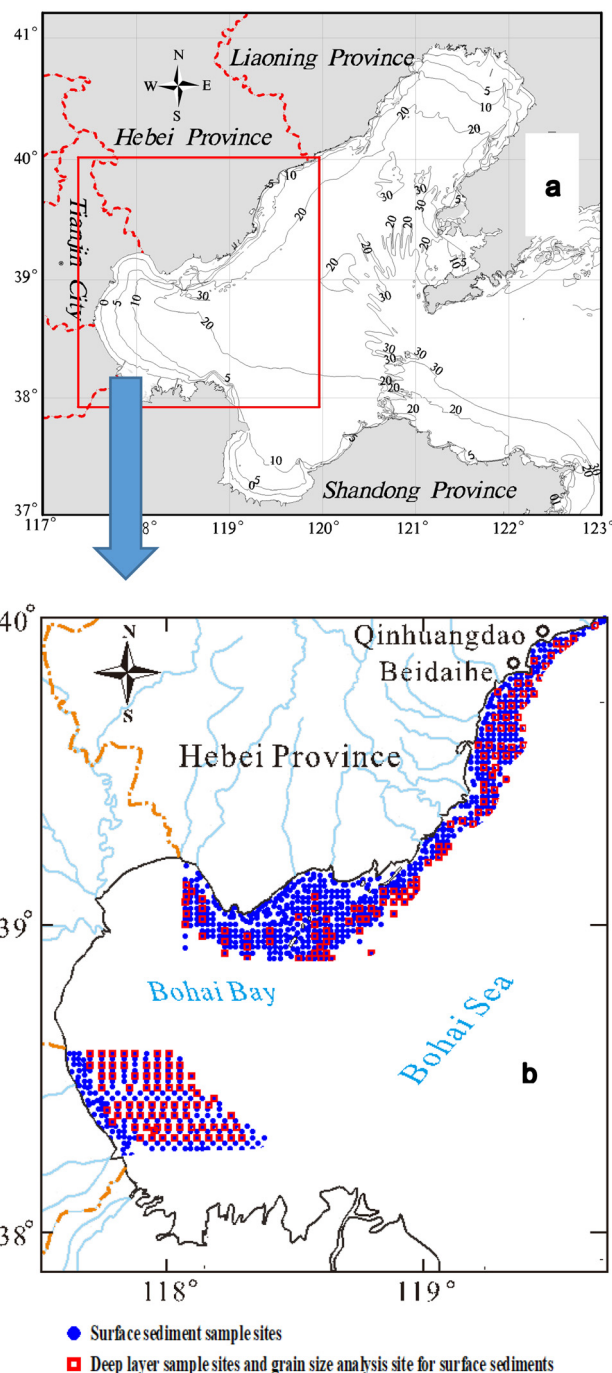


Fig. 1. Map showing the Bohai Sea in Northeast China including major provinces and associated 10 m contour depths offshore (a), and the location of surface sediment and deep layer samples taken from the Western Bank of the Bohai Sea, Hebei Province, China. (b).

to determine concentration ranges for specific areas sampled in order to alleviate the constituent variability produced by granulometric divergence. In this study, we used aluminum (Al) as the normalized element. The enrichment factor ( $EF$ ) is defined as,

$$EF = \frac{\left( \frac{X_{sample}}{Al_{sample}} \right)}{\left( \frac{X_{baseline}}{Al_{baseline}} \right)} \quad (1)$$

where  $X_{sample}$  ( $X_{baseline}$ ) and  $Al_{sample}$  ( $Al_{baseline}$ ) are heavy metal concentrations and aluminum concentrations of a sample and baseline

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