



## The distribution, enrichment and source of potential harmful elements in surface sediments of Bohai Bay, North China

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### ABSTRACT

A geochemical study of Bohai Bay surface sediments was carried out to analyze the potential harmful element (PHE: Ge, Mo, In, Sn, Sb, Te, Tl, Bi and V) concentrations, transportation and deposition, enrichment factors and sources. Germanium, Mo, In, Sn, Sb, Te, Tl, Bi and V concentrations in the surface sediments were: 1.43–1.71, 0.52–1.43, 0.04–0.12, 2.77–4.14, 1.14–2.29, 0.027–0.085, 0.506–0.770, 0.27–0.63 and 70.35–115.90  $\mu\text{g/g}$ , respectively. The distributions of total PHE concentrations, together with sequential extraction analyses, showed that the PHEs were mainly due to natural inputs from the continental weathering delivered to the bay by rivers and atmospheric transportation and deposition. However, high Mo, Sb, Te, Bi and V occurred in non-residual fractions, suggesting some anthropogenic inputs in addition to the natural inputs. Besides sources, the distributions of PHEs were influenced by the coupling of physical, chemical and biological processes. Enrichment factor (EF) was computed for each site for each element in order to assess the polluting elements and the degree of pollution at each site. Results revealed that the EFs were generally lower than 1.0, particularly for Ge, Mo, In, Sn, Tl and V; however, the EFs were higher (>1.5), particularly for Sb, Te and Bi, revealing moderate contamination.

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### 1. Introduction

A good deal of research on biogeochemical processes in aquatic environment has been, and continues to be, devoted to trace elements, particularly transition metals. Rather less attention has been focused on the so-called metal and semi-metal elements, such as Ge, Mo, In, Sn, Sb, Te, Tl, Bi and V. Especially, the aquatic geochemistry of these elements is poorly known. Because of their very low abundance, these trace elements are particularly sensitive to surrounding environmental conditions, which influence their physical–chemical species and behaviors in the ecosystems. These elements are deemed serious pollutants because of toxicity, persistence and non-degradability in the environment. So, they are called “Potential Harmful Elements (PHEs)” [1]. Among them, germanium, Sn, V and Mo in trace amounts are beneficial to normal cell growth, being the so-called essential elements. However, toxicities arise when element concentrations increase to a higher level [2,3].

Antimony, Tl and their compounds are classified as priority pollutants by the Environmental Protection Agency of the United States (USEPA) [4,5]. Indium, Te and Bi are known to be non-essential and have potential toxic and harmful for living organisms [6–8]. In recent years, PHEs have been discharged into coastal oceans as a result of the rapid industrialization. Hence, although these elements except for V are found in relatively low abundances, it is possible that the high toxicities of these elements can induce environmental problems [9].

Sediments are important deposits of PHE accumulation in the aquatic environment. The large adsorption capability of sediment to accumulate compounds makes them a major repository of natural and anthropogenic PHEs and one of the most important tools to assess the contamination level of aquatic ecosystems [10]. Due to their stability within the sedimentary column, most of the contaminants can leave their fingerprint in sediments. Thus, the study of geochemical characteristics of sediment has shown to be an excellent tool for establishing the effects of anthropogenic and natural processes on depositional environments. On the other hand, the non-residual fractions of PHEs (i.e., exchangeable, bound to carbonates, bound to Fe–Mn oxides and bound to organic matter

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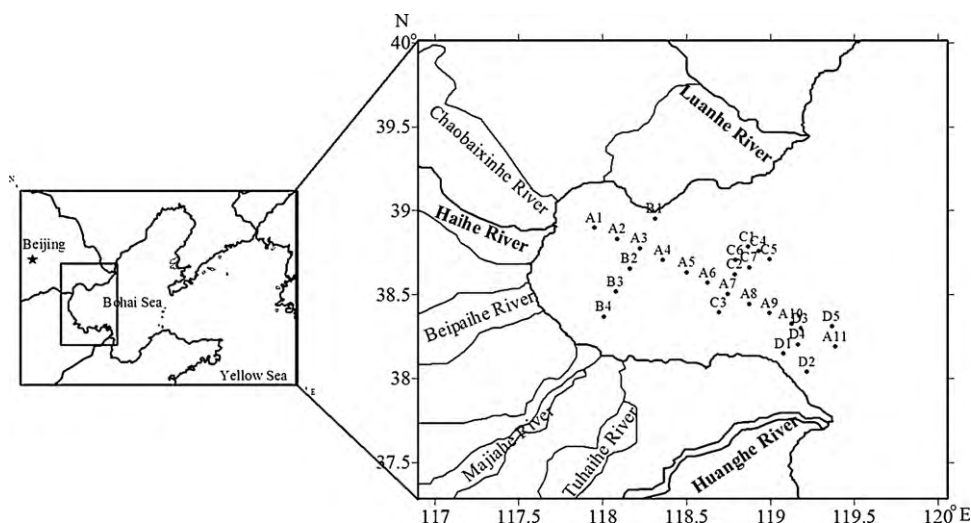


Fig. 1. Location of the stations in Bohai Bay, 2008.

fractions) can be easily released to water column through any disturbance, such as hydrological, physical, chemical and biological activities [11]. Thus, sediments are a potential source of pollution for surrounding water column. Consequently, the knowledge of the concentrations, distributions and chemical fractions of PHEs in sediments plays a key role in detecting sources of PHEs and assessing the ecological risks of PHEs in aquatic systems.

The PHEs transferred into ocean systems are associated with physical and chemical processes, such as weathering processes, subsequent riverine transportation of particulate matter, atmospheric transportation and deposition. Once entered the aquatic environment, PHEs would be adsorbed on sediment particles or precipitate into bottom sediments. The extent of PHE sorption and the factors controlling PHE distributions remarkably depend on the sediment characteristics (grain size, chemical components, organic carbon contents, mineral phases and texture), the sedimentary environmental characteristics (salinity, pH, temperature, oxidation–reduction potential (Eh), presence of ligands and ocean currents), sources whereby PHEs entered the environment and the physical, chemical and biological processes [12].

The PHEs are introduced into the oceans by natural processes including erosion of ore-bearing rocks, wind-blown dust, volcanic activity and forest fires, and processes derived from human activities by means of atmospheric deposition, rivers and direct discharges or dumping [13]. The major anthropogenic sources of PHEs are coal and fossil fuel combustion and steel manufacturing plants. So the riverine inputs and atmospheric deposition are the main carriers of PHEs to ocean. In particular, atmospheric deposition is now widely recognized as an important source of PHE contamination for surface sediments. Hence, surface sediment records provide the most compelling evidence of regional or long-range atmospheric transportation of PHEs [14].

Because of their bioavailability and toxicity, the PHEs should be of global interest and be much accounted of, but there is little information on geochemical characteristics and behaviors of PHEs in sediments. To understand behaviors and fates of PHEs and evaluate the ecological risk of PHEs, the total concentrations, distributions and chemical fractions of PHEs in the Bohai Bay surface sediments were determined. The objectives of this study were (1) to determine the PHE concentrations and their chemical fractions in the surface sediments of Bohai Bay; (2) to analyze the factors controlling the PHE distributions; (3) to evaluate the enrichments of PHEs in the surface sediments of Bohai Bay; and (4) finally to discuss the origins of PHEs in the surface sediments of Bohai Bay.

## 2. Materials and methods

### 2.1. Site description

The focus of the paper concentrated on Bohai Bay. It is a semi-enclosed bay located in the western region of the Bohai Sea in the North China. It covers an area of  $1.6 \times 10^4$  km<sup>2</sup> with a population of about 70 million. The average depth is 12.5 m with the maximum of 32 m. Bohai Bay receives both industrial and domestic wastewater discharges from Beijing, Tianjin and Hebei province. All the wastewater through rivers and channels drains into the near-shore waters of Bohai Bay directly [15]. Approximately 87% of pollutants in Bohai Bay are discharged from land, and 95% of pollutions generated in the area are transported by river into Bohai Bay. There are several rivers (e.g., the Haihe River, Jiyunhe River, Huanghe River, Tuhaihe River and Majiahe River) emptying into Bohai Bay with a total annual water discharge of  $6.5 \times 10^9$  m<sup>3</sup>/yr and annual suspended matter input of  $6.0 \times 10^6$  tons/yr [16,17].

### 2.2. Sampling

The present study was based on the surface sediments from Bohai Bay sampled during 21 to 28 April 2008. According to their latitude from 38°14'04" N to 38°58'00" N, a population of 27 sample stations were designated (Fig. 1). Position of sampling sites was recorded by the aid of a Global Positioning System (GPS) technology. Surface sediments were collected using a grab sampler. Sub-samples were taken from the center with a polyethylene spoon to avoid possible contamination by the metallic parts of the sampler. Immediately after collection, samples were placed in pre-cleaned polyethylene bags, sealed, refrigerated, and transported to the laboratory. Samples were dried in an oven at 60 °C for 72 h. Dried aliquots were ground using an agate mortar and pestle for homogenization, and prepared for analysis. Data reported in this study are calculated as dry weight.

### 2.3. Sediment analyses

About 0.1 g dry sample was digested in closed Teflon digestion vessel with a mixture of 0.6 ml HNO<sub>3</sub> and 2 ml HF, and then the mixture was heated on an electric heating plate at 150 °C for 24 h. Then 0.5 ml HClO<sub>4</sub> was added to the mixture, and it was evaporated to dryness. After cooling, the residue was digested again

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