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Sources and geochemical background of potentially toxic metals in surface sediments from the Zhejiang coastal mud area of the East China Sea

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ABSTRACT

The geochemical background concentration (GBC) of potentially toxic metals in coastal surface sediments is a useful reference to assess the extent of sediment contamination caused by human activities. Using surface sediments collected from the Zhejiang coastal mud area of the East China Sea, a regional GBC function (GBCF) for potentially toxic metals in sediments was constructed based on statistical techniques. Principal component analysis (PCA) was utilized to identify potentially toxic metal sources. For potentially toxic metals of natural origin, the GBCF was developed by directly fitting concentration with Al_2O_3 in a linear regression model. For potentially toxic metals of anthropogenic origin, concentration data were normalized with Al_2O_3 to eliminate the influences of grain size and mineral, and cleaned by the cumulative distribution function before linear regression analyses. At each sampling station, the enrichment factor (EF) was modified by the corresponding GBC calculated from the GBCF, and was then applied to identify metal sources. The EF results were consistent with those of the PCA and correlation analyses, indicating that the GBCs of potentially toxic metals at each station were applicable. The approach used in this study will be useful for assessing potentially toxic metal pollution in sediment and managing sediment quality.

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1. Introduction

As one of our most important food sources, coastal ecosystems are significantly affected by human activities and often the ultimate receptacle of pollutants (Maanan et al., 2014; Magesh et al., 2011). With rapid industrialization and economic development in coastal regions, huge amounts of potentially toxic metals are discharged into coastal ecosystems every year via untreated industrial wastewater, municipal sewage, and surface run-off (Gao and Li, 2012). Enrichment of potentially toxic metals in coastal sediments can result from both anthropogenic activities and natural processes (Nriagu, 1989; Veena et al., 1997). High concentrations of potentially toxic metals from natural origins in sediments are often enriched by refractory minerals and do not exhibit potential toxicity to an ecosystem (Xu et al., 2009). Consequently, the clear identification of anthropogenic potentially toxic metals from natural sources is important for evaluating the extent of pollution, preventing further environmental damage, and planning remedial strategies.

The geo-accumulation index (Fu et al., 2013; Xu et al., 2014), pollution load index (Sun et al., 2010), enrichment factor (Magesh et al., 2011; Soltani et al., 2015), and potential ecological risk index (Yi et al., 2011; Liu et al., 2014) can quantitatively assess potentially toxic metal pollution, ecological risk, and pollution source in sediments, although reference levels of potentially toxic metals in sediments are firstly required. Average shale, upper crust, and preindustrial levels of potentially toxic metals have been used extensively as reference baseline concentrations (Zahra et al., 2014). However, regional background values of potentially toxic metals in sediments depend on local geological properties, which might differ from global reference levels (Jiang et al., 2013). Therefore, understanding the geochemical background concentrations (GBC) or baseline levels of potentially toxic metals in a studied area is essential for evaluating the extent of potentially toxic metal pollution and ecological risk.

There are three major methods for establishing GBC (Carballeira et al., 2000; Reimann and Caritat, 2000), which all require non-contaminated samples. The first approach relies on non-contaminated samples collected from pristine areas unaffected by human activities or by oceanic modification processes such as scavenging (Song et al., 2014; Xu et al., 2014). In the second approach, statistical methods are applied to

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infer GBC in surface sediments (Jiang et al., 2013; Wang et al., 2015; Karim et al., 2015). By making assumptions about the normal distribution of potentially toxic metals in non-contaminated samples, the GBC can be estimated by first removing data outliers, and then by applying several statistical techniques (Matschullat et al., 2000). The third method uses non-contaminated samples in core sediments, which can be defined by age in dated core sediments. This method is the most reliable for establishing GBC if post-depositional remobilization is negligible (Song et al., 2014). Although these three methods can establish a constant for GBC, using a single regional GBC alone has the disadvantage of excluding natural variability in potentially toxic metal concentrations (Loring, 1991; Daskalakis and O'Connor, 1995; Covelli and Fontolan, 1997). In particular, GBC may vary within a region and between regions due to the influences of sediment grain size as well as mineralogical and chemical composition (Reimann and Garrett, 2005; Singh et al., 2005; Jain et al., 2007).

Based on chemical sedimentation mechanisms, potentially toxic metal sources, and statistical techniques, we proposed a method to determine the GBC function (GBCF), and calculated the GBC of potentially toxic metals at each station along the Zhejiang coastal mud area of the East China Sea (ECS).

The ECS, which is adjacent to Zhejiang and Fujian provinces and the Yangtze River Delta (Fig. 1), is the largest agricultural production base in China (Wang et al., 2014). Its coastal zone includes many large and medium cities with dense human populations, such as Shanghai, Hangzhou, and Ningbo, as well as numerous concentrated industries. With the rapid economic development and intense industrialization in the area in recent decades, considerable amounts of anthropogenic potentially toxic metals and organochlorine pesticides have been delivered to the ECS by river conveyance (e.g., Yangtze and Qiantang rivers), and then transported via the Zhejiang-Fujian Coastal Current (ZFCC) (Lin et al., 2002; Zhou et al., 2013; Wang et al., 2014). Based on previous reports, water quality in the coastal areas around Hangzhou Bay and the Zhoushan islands is mostly below Grade I (Liu et al., 1991). Both organochlorine pesticide and potentially toxic metal pollution in the Yangtze River estuary and adjacent sea areas have been studied comprehensively (Chen et al., 2004; Yuan et al., 2004; Fang et al., 2009; Dong et al., 2014.). However, reports related to the GBC of potentially toxic metals in the ECS are scarce.

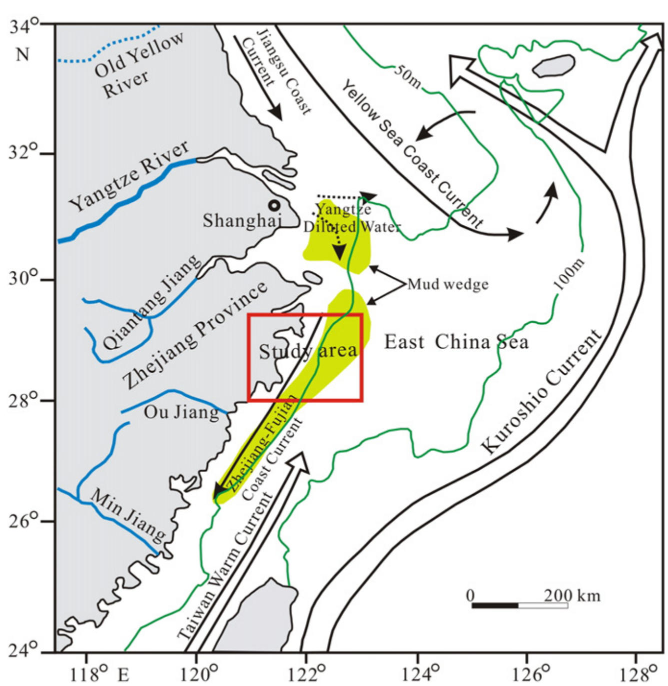

Fig. 1. Location of study area and current circulations in the East China Sea (ECS) (circulation systems are modified according to Zheng et al., 2010).

2. Materials and methods

2.1. Study area

The study area is located along the inner shelf of the ECS (Fig. 1). The ECS is one of the largest marginal seas in the western North Pacific Ocean, and is noted for its tremendous river runoff, notably from the Yangtze River. Sediments on the ECS shelf consist of sand and mud. Muddy sediment occurs along the near-shore to inner shelf environments off the Yangtze River on the south coast of China (Saito et al., 1998). Sandy sediment occurs in the middle to outer shelf areas (Alexander et al., 1991), and is considered to be transgressive sediment deposited since the last glacial maximum ~20–18 ka BP (Bartek and Wellner, 1995).

The Yangtze River is the dominant contributor to the inner shelf mud wedge in the ECS. Of the 482 Mt/year of sediment load measured at Datong gauging station, channel aggradation and delta progradation trap about 70% of the sediment before it reaches the ECS, leaving ~150 Mt/year of sediment to be transported southward (Milliman et al., 1985). The southward dispersal of Yangtze River sediment occurs mainly along the inner shelf, especially landward of the 50 m isobath, though reaching to the 100 m isobath (Xu et al., 2009). This southward dispersal has created an elongated mud wedge, the inner shelf mud wedge, extending from the Yangtze River mouth to 1000 km into the Taiwan Strait.

Coastal oceanography in the study area is dominated by the southward flowing ZFCC, a relatively cold and brackish counter current (Fig. 1). This current intensifies in the winter, carrying the brackish water and sediment of the Yangtze southward along the inner shelf (Liu et al., 2006). Offshore is a northward flow of warm and saline water, known as the Taiwan Warm Current (TWC, Fig. 1), which intensifies in summer in response to the prevailing southeast monsoon and as the southward ZFCC weakens (Lee and Chao, 2003).

2.2. Collection of surface sediment samples

In the summer of 2013, 350 surface sediment samples were collected along the Zhejiang coastal mud area of the ECS (Fig. 2). Sub-samples were taken from the top 2 cm of the box center. Each bulk sediment

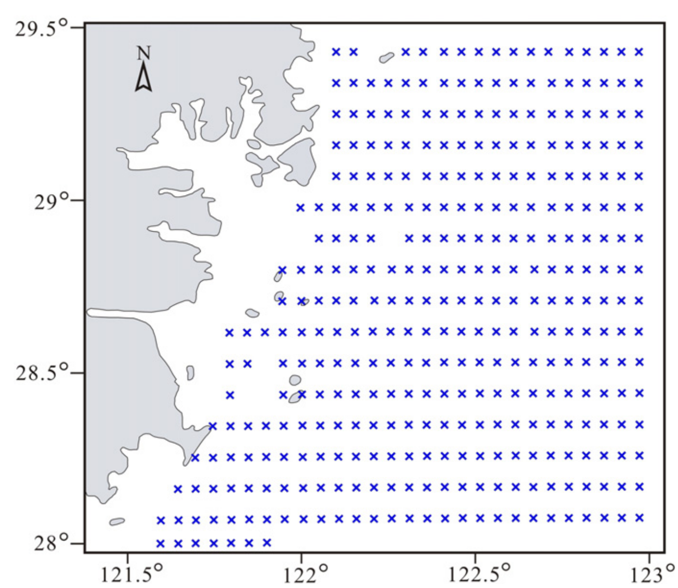

Fig. 2. Sampling stations of surface sediments in the Zhejiang coastal mud area of ECS.

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