



Distribution, enrichment and source of heavy metals in surface sediments of the eastern Beibu Bay, South China Sea

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ABSTRACT

Sixty-nine samples of surface sediments (0–5 cm) recovered from the eastern Beibu Bay were analyzed for TOC and heavy metals to examine the element distribution pattern and potential pollutant sources. The sediments in the study area are characterized by variable heavy metal concentrations that are comparable with those of the surrounding regions. Obvious positive correlations were observed amongst the concentrations of Zn, Cr, Pb, Cd, Cu and the clay contents, suggesting that fine clay particles are important carriers of trace metals in the sediments. Hg and As show a considerable/low positive correlation with TOC, indicating that organic matter may play a discernible role in the sediment chemistry. Cd contamination was detected in the north of the study area primarily due to the input of phosphate fertilizers carried by rivers. In comparison, Zn, Cr, Pb, and partly Cu are predominantly sourced from lithogenic components, and Hg and As are mainly from organic matter related to anthropogenic input.

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1. Introduction

Heavy metals have become a global problem in recent years due to their toxicity, wide sources, non-biodegradable properties, and accumulative behaviors (Irabien and Velasco, 1999; Yu et al., 2008). Sediments are the main sink for various pollutants and have thus attracted much attention from researchers (Dassenakis et al., 1997; Bettinetti et al., 2003; Singh et al., 2005). Heavy metal contamination in sediments can affect water quality and thus the bio-assimilation and bioaccumulation of metals in aquatic organisms, with potential long-term implications for human health and the ecosystem (Radha et al., 1997; Li et al., 2004; Ip et al., 2007). It is therefore important to understand the distribution of heavy metals in sediments to develop strategies and approaches for pollution control.

The semi-enclosed Beibu Bay lies in the west of Hainan Island, at the south of the Fujian Province, with its mouth opening towards South China Sea (Fig. 1). Many rivers discharge into the bay, such as the Red River from Vietnam and the Beilun, Maoling, and Dafeng Rivers from China. These rivers can be potential and significant sources of heavy metal pollution in the bay, with rapid

industrial and economic development in the catchments. Various studies on heavy metal contamination in the Beibu Bay have been conducted in recent years (Lian et al., 2001; Xia et al., 2008, 2011). However, these studies have mainly focused on the intertidal sediments deposited in the estuary and surrounding coastal areas (Xia et al., 2008, 2011), and few have touched upon the central part of the bay. For a better understanding of heavy metal contamination and sediment quality in the whole of Beibu Bay, high-resolution sampling and analytical works are urgently needed.

Quantitative geochemical methods, such as the enrichment factor (EF) and geoaccumulation index (I_{geo}), have been successfully used to estimate the impact of human activities on sediment quality (Cuadrado and Perillo, 1997; Villaescusa-Celaya et al., 2000; Cevik et al., 2009). Furthermore, appropriate statistical approaches, such as principal component analysis (PCA) and factor analysis (FA), are increasingly applied for environmental studies, including the measurement and monitoring of heavy metals in various environmental media (Huang et al., 1994; Facchinelli et al., 2001; Loska and Wiechula, 2003; Filgueiras et al., 2004; Boruvka et al., 2005; Tariq et al., 2006). In this work, the methods above have been applied to examine the sources of heavy metals in the eastern Beibu Bay and the further assess its contamination status. The main objectives of the study are (1) to quantify and investigate the spatial distribution of heavy metals in modern surface sediments; (2) to assess the state of heavy metal contamination using the EF and I_{geo} proxies; and (3) to analyze the sources and transport pathways of heavy metals in the eastern Beibu Bay.

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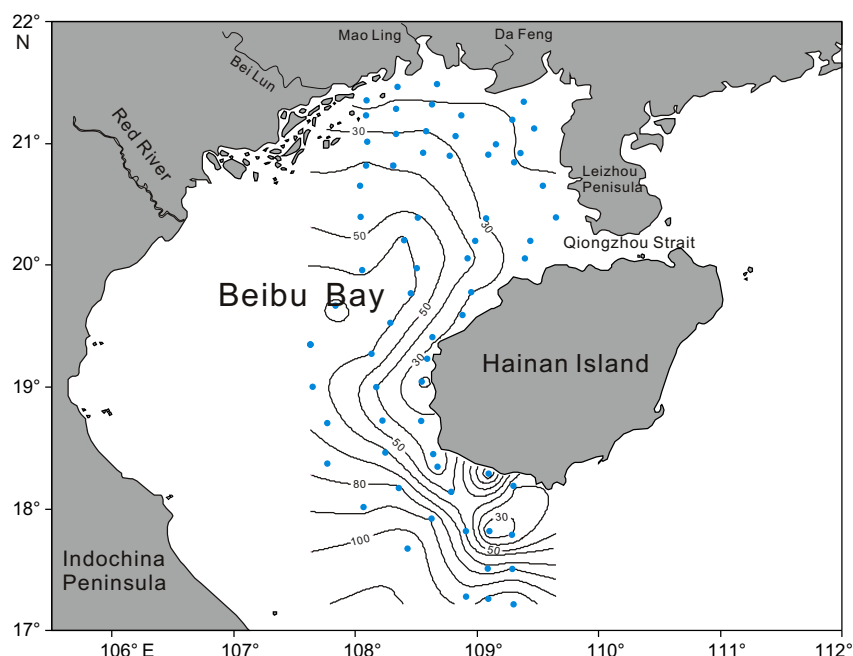


Fig. 1. A schematic map showing the study area with water depths and locations for the sediments taken from the eastern Beibu Bay.

2. Materials and methods

2.1. Sampling

Sixty-nine surface sediment (0–5 cm) samples were collected from the eastern Beibu Bay using grab samplers in July and August 2006 (Fig. 1). The sampling stations were chosen carefully to provide good area coverage. After sampling, the sediments were stored in a plastic vessel and frozen at -20°C . In the laboratory, the sediments were defrosted at room temperature, dried at 40°C to constant weight, and ground and homogenized in a mortar to a fine powder. In previous studies, the samples were mainly composed of fine-grained sediments (Xu et al., 2010), so the samples for this study were not separated by being passed through a nylon mesh sieve.

2.2. Metals analysis

For acid digestion, 0.25 g of powered sample was put in a Teflon bomb with an acid mixture (5:4:1 $\text{HNO}_3 + \text{HCl} + \text{HF}$, in volume) (Loring and Rantala, 1992) and then heated to 120°C for 12 h on a heating plate. The acid digestion was repeated until only a negligible amount of white residue remained. Afterwards, the solution was evaporated to dryness and extracted with HNO_3 . Heavy metals (Cd, Cu, Cr, Pb, Sc, and Zn) were determined by ICP-MS, as previously reported by Xia et al. (2011). Arsenic (As) and Hg were analyzed using Atomic Fluorescence Spectrometry (AFS). All the sediment samples were analyzed in duplicates and the quality assurance was controlled by standard reference materials (GSD-9 and GSD-4, GBW07345). The differences of the concentrations between the determined and certified values were less than 5%, and the analytical precision for replicate samples was within $\pm 10\%$. Overall, the analytical values of the reference material are within the range of the certified values (Table 1). Total organic carbon (TOC) was measured using a Carlo-Erba™ element analyzer. Grain size measurements were performed using a laser granulometer (Malvern Mastersizer 2000) according to Xu et al. (2010). All the experiments were performed at the Key Laboratory of Marine

Table 1

Analytical accuracy of Chinese National Reference Materials GSD-9, GSD-4 and GBW07345. Concentrations are in mg/kg.

Standard materials	Element	Measured values	Certified reference values
GSD-9	Sc	12.1	11.1 ± 0.8
	Cr	78.89	85 ± 10
	Cd	0.23	0.26 ± 0.05
GSD-4	Hg	0.056	0.044 ± 0.009
	As	21.58	19.7 ± 2.6
GBW07345	Cu	15.50	15.1 ± 0.7
	Zn	47.10	45 ± 2
	Pb	31.90	28 ± 2

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3. Results and discussion

3.1. Distribution patterns of TOC, heavy metals and As in the sediments

The surface sediments in the eastern Beibu Bay are primarily composed of silts with a mean size of 6.06Φ (Xu et al., 2010). The percentage of mud (silt and clay fractions) varies between 26.1% and 99.8%, with a mean value of 77.6%. The mean values, ranges and standard deviations of the heavy metal concentrations (Hg, Cd, Cu, Cr, Pb, Zn), As, and TOC are presented in Table 2. To assess the heavy metal contamination in this region, the background abundances of the trace metals in the intertidal sediments of the Beibu Bay are also listed in Table 2 (Xia et al., 2011). The concentrations of TOC in the eastern Beibu Bay range from 0.04% to 0.84% with a mean value of 0.49%, which is much lower than that of the nearby Quanzhou Bay (0.75%; Yu et al., 2008). Low concentrations of TOC are found in the central and southern parts of the study area, where the sediments are relatively coarse, suggesting the enrichment control of fine clay on organic matter (Carvalho et al., 2005; Uluturhan et al., 2011). Notably, the TOC contents in

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