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Geochemical analysis of sediments from a semi-enclosed bay (Dongshan Bay, southeast China) to determine the anthropogenic impact and source

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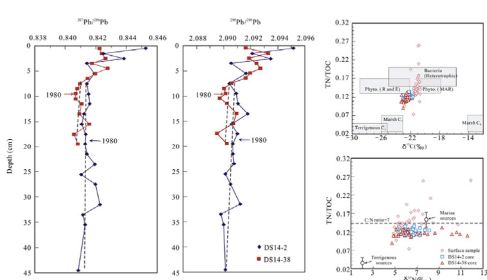
HIGHLIGHTS

- Heavy metals, Pb isotopes and biogenic elements from a semi-enclosed bay were analysed.
- EFs and Pb isotopes indicated minor pollution in the past 30 years.
- OM was primarily derived from aquatic plants, terrigenous erosion, fertilizers and sewage.
- The TOC and TN fluxes had increased since the 1980s, reflecting the use of N fertilizer.

GRAPHICAL ABSTRACT



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ABSTRACT

The geochemical compositions of sediments in the Dongshan Bay, a semi-enclosed bay on the southeast coast of China, were obtained to identify pollutant sources and evaluate the anthropogenic impacts over the last 100 years. The results indicated that the metal flux had been increasing since the 1980s. Enrichment factor values (Pb, Zn and Cu) suggested only slight enrichment. The proportion of anthropogenic Pb changed from 9% to 15% during 2000–2014. Coal combustion might be an important contamination source in the Dongshan Bay. The historical variation in the metal flux reflected the economic development and urbanization in the Zhangjiang drainage area in the past 30 years. According to the Landsat satellite remote sensing data, the urbanization area expanded approximately three times from 1995 to 2010. The $\delta^{13}C$ values (−21‰ to −23‰) of the organic matter (OM) in the sediments indicated that the OM was primarily sourced from aquatic, terrigenous and marsh C_3 plants. Nitrogen was mainly derived from aquatic plants and terrigenous erosion before the 1980s. However, the total organic carbon (TOC) contents, total nitrogen (TN) contents and $\delta^{15}N$ had been increasing since the 1980s, which suggested that the sources of nitrogen were soil erosion, fertilizer and sewage. In addition, the TOC and TN fluxes in the Dongshan Bay had significantly increased since the 1980s, which reflected the use of N fertilizer. However, the TOC and TN fluxes significantly decreased in the past decade because environmental awareness increased and environmental protection policies were implemented.

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1. Introduction

The economic development in China has led to the production of many pollutants that are critical to the environment (Cheng, 2003; Zhu et al., 2007; Pan and Wang, 2012; Li et al., 2012a). Estuarine and coastal areas are the most important regions for human inhabitants and are usually the catchments for anthropogenic pollutants (Ip et al., 2004, 2007; Hao et al., 2008; Pan and Wang, 2012; Xu et al., 2014; Wang et al., 2014; Guo and Yang, 2016). Approximately 17.6 million tons of pollutants were discharged into the sea in China in 2014 (NBO, 2015). The main pollutants were inorganic nitrogen, active phosphates and petroleum in estuaries and bay, and eutrophication was notably present in some areas (NBO, 2015). Furthermore, metal pollution may also have a serious impact on the environment (Li et al., 2012a; Pan and Wang, 2012). More than 56,000 tons of anthropogenic lead is discharged to atmosphere in China per year (Niisoe et al., 2010).

Large amounts of chemical fertilizers are used within a limited area in China, which may cause water quality deterioration and accelerate eutrophication in watersheds (Guo et al., 2010). Identifying the sources of pollutants is important in environmental investigation and ecological impact assessment. Isotopes, which are remarkably more sensitive tracers than elemental concentrations, could be used to trace pollutant sources. The carbon and nitrogen isotopes in marine sediments can be used to identify whether the organic matter (OM) and nitrogen are natural or anthropogenic in origin (Dai et al., 2007; Goñi et al., 2014; Xia et al., 2015). Lead is characterized as insoluble and as having a conservative behaviour in the secondary environment. Moreover, Pb isotopes do not fractionate during industrial and environmental processes. Therefore, Pb isotopes have been used to track the sources of metal pollutants (Choi et al., 2007; Komárek et al., 2008; Cheng and Hu, 2010; Bird, 2011; Li et al., 2012b; Xu et al., 2014).

Metal pollution and eutrophication in the coastal region of Fujian in China has been a matter of concern for the last decades (Li, 2000; Zhang and Liu, 2002; Zhang et al., 2007; Xu et al., 2014; Wang and Wang, 2016). The Dongshan Bay is a semi-enclosed bay on the southeast coast and is an important aquaculture base in Fujian Province. The aquaculture area in the Dongshan Bay decupled from 1999 to 2012 (Gao et al., 2014). Moreover, some Pb pollution occurred in the coast surrounding the Dongshan Island (Chen, 2016). The bay has a relatively stable sedimentary environment and could adequately record the evolution of the sedimentary environment. On the basis of the grain size, heavy metals, Pb isotopes and biogenic elements, the major objective of this study was to identify the pollutant sources and evaluate the anthropogenic impacts on the Dongshan Bay over the last 100 years.

2. Materials and methods

2.1. Study region

The Dongshan Bay is a semi-enclosed bay in the northern part of South China Sea (Fig. 1). The bay's mouth opens to the south and is only 4 km wide. The Zhangjiang River flows into the sea at the top of the bay. The Zhangjiang River has an annual average discharge of water and sediments of approximately 1.0×10^9 m^3 and 39.1×10^4 tons, respectively. The marsh at the estuary is one of the largest mangrove reserves in China, with an area of 21 km^2. In recent years, with the development of industry and agriculture in the areas surrounding the Dongshan Bay, petrochemical, iron and steel industries have been established on the Gulei Peninsula. In addition, the aquaculture area reached approximately 3.4 km^2 in 2012 (Gao et al., 2014).

2.2. Sample collection

Fifty-one surface sediments were collected using a grab sampler and three cores were obtained in PVC pipes (inner diameter, 11 cm) using a gravity corer in the Dongshan Bay in April 2014 (Fig. 1). The lengths of the sediment cores were 1.90, 1.70 and 1.65 m for DS14-2, DS14-16 and DS14-38, respectively. Care was taken to minimize disturbance while obtaining the cores. The upper portions of the cores were sub-sampled at 1-cm intervals. The sediments were dried and ground to 200 mesh (74 μm) using an agate grinder. Analyses of grain size, chemical composition and biogenic elements were measured in the Third Institute of Oceanography, SOA. Pb isotopes were measured in the Guangzhou Institute of Geochemistry, CAS.

2.3. Radioactive dating

The total counts of ^{210}Po and tracer ^{209}Po were measured by 7200-8 Alpha spectrometry (Canberra, USA). From the total counts of ^{210}Po and ^{209}Po, the total ^{210}Pb activities were calculated (Nittrouer et al., 1979). The excess $^{210}Pb_{ex}$ activity (dpm/g) was calculated by deducting the background ^{210}Pb activity from the total activity (DeMaster et al., 1985). $^{210}Pb_{ex}$ decays exponentially following the radioactive decay law $A_t = A_0e^{-\lambda t}$, where λ is the ^{210}Pb decay constant (0.0311 yr^{-1}).

$^{210}Pb_{ex}$ generally showed an exponential decrease distribution in the core, which indicated the probability for using the constant initial concentration (CIC) model (Robbins, 1978) for this study. The sedimentation rate was calculated according to the equation $S = -\lambda/K$, where S_R is the sedimentation rate (cm yr^{-1}) and K is the slope of the linear regression of depth plotted between the natural logarithm of $^{210}Pb_{ex}$ and depth (Fig. A1).

2.4. Grain size

The samples were soaked in 15 mL of 30% H_2O_2 and 5 mL of 3 mol L^{-1} HCl for 24 h to remove OM carbonates. The grain size was measured using the Malvern Mastersizer 2000 laser diffraction particle size analyzer. The measurement range of Mastersizer 2000 is 0.02–2000 μm and the resolution is 0.01φ. The relative error of duplicated measurements is ≤3%.

2.5. Trace elements and Pb isotope analyses

The samples were digested by Multiwave 3000 microwave digestion system. The elements were detected by ICP-AES (OPTIMA 7300DV). Detailed procedures and equipment parameters were previously described (Xu et al., 2014). The relative standard deviation of the offshore marine sediment (GBW-07314) was ≤5%.

The Pb isotopic compositions were analysed using the Isoprobe MC-ICPMS (Micromass). The average $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$ ratios of BHVO-2 were 0.8327 ± 0.0002 (2SD) (n = 7) and 18.651 ± 0.004 (2SD) (n = 7), respectively, and the average $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$ ratios of JB-3 were 0.8494 ± 0.0001 (2SD) (n = 8) and 18.294 ± 0.005 (2SD) (n = 8), respectively.

2.6. Biogenic elements

The total organic carbon (TOC) and total nitrogen (TN) contents were determined using Element Analyzer (Elementar vario EL-III) after removing carbonate. Precision of replicate analyses of acetanilide standard was approximately 0.3%. The $\delta^{13}C$ and $\delta^{15}N$ values were analysed using the stable isotope mass spectrometer (Thermo Delta V Advantages). The $\delta^{13}C$ and $\delta^{15}N$ values were calculated

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