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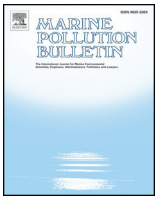

Trace metals in a sediment core from the largest mariculture base of the eastern Guangdong coast, South China: Vertical distribution, speciation, and biological risk

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ABSTRACT

A sediment core collected from Zhelin Bay, the biggest mariculture base of the eastern Guangdong Province, was analyzed for trace metal concentrations and chemical fractions. Average total concentrations (mg/kg) were 20.7 ± 15.4 (Pb), 74.6 ± 11.6 (Cr), 40.7 ± 6.0 (Ni), 55.9 ± 13.0 (Cu), and 169.0 ± 11.9 (Zn), with the concentrations of Cr, Ni, Cu and Zn being significantly higher than their corresponding background values. We identified two vertical distribution patterns of the trace metals in the sediment core. In all sub-samples, Pb was mainly associated with the reducible fraction, whereas a major portion (62.2 to 95.2%) of Cr, Ni, Cu, and Zn was strongly associated with the residual fractions. Biological risk assessment based on the mean effects range-median quotient suggests that the Zhelin Bay sediment core has a 21% probability of being toxic.

Costal and estuarine ecosystems are important reservoirs for many persistent contaminants, including metals, which may accumulate in organisms and bottom sediments (Liu et al., 2011; Gao et al., 2014; Alyazichi et al., 2015). Metals entering into aquatic systems are usually rapidly transported into sediments. (Gu et al., 2012a; Gu et al., 2016). However, with changing environmental conditions, e.g. pH and Eh; they can be released and represent a secondary contamination source, affecting ecosystems of nearby seas and estuaries (Ip et al., 2007; Nielsen et al., 2010; Yu et al., 2010; Kalantzi et al., 2013). Consequently, sediments are both carriers of metals and potential pollution sources in aquatic systems. Toxicity and mobility of metals in the sediments strongly depend on their specific chemical forms and binding state (Gleyzes et al., 2002). However, studies on trace metal speciation and biological risk using sediment cores from mariculture areas are scarce.

China is by far the world's largest fishery producer, consumer, and exporter. In 2012, It's aquaculture production was 36.73 million tons, accounting for 61.69% of the world's total production (Cao et al., 2015; FAO, 2015). Thus, China occupies a leading and decisive role in promoting aquaculture development. The quality of Chinese aquatic products, which is directly affected by the production environment, has important implications for consumers' health safety, and should be sufficiently evaluated.

Guangdong province has the most developed economy and the largest aquaculture production in China. Zhelin Bay, situated in the northeastern part of Guangdong Province, is the largest mariculture base of eastern Guangdong (Fig. 1). This bay, an area of about 70 km^2, is one of the most intensively managed coastal areas (Gu et al., 2014b), with two rivers, Huanggang and Nanxi, opening into this bay (Fig. 1.). Mariculture of Zhelin Bay has considerably boosted the local economy; however, over the past years, rapid deterioration of the aquatic ecosystem has occurred (Gu et al., 2014b; Yang et al., 2006). Apart from the negative impacts of aquacultures, the area is also experiencing pressures from increased industrial pollution, agriculture activities, and sewage discharge, issues which are related to the rapid economic development and urbanization during recent decades (Gu et al., 2014b).

To date, there are no studies evaluated metal speciation and biological risk in sediment core of Zhelin Bay. This study was therefore designed to (1) investigate trace metal concentrations and metal fractions in sediment core of Zhelin Bay and to (2) identify potential sources and evaluate biological risk assessment.

Prior to the collection of sediment cores, we scanned the topography of Zhelin Bay using a sub-bottom profiler system (RS-QP0116, Hangzhou Resound Marine Instruments Co., Ltd. China) and identified areas with good sediment stratification. A 108 cm long sediment core was obtained using a Piston corer (UWITEC Piston corer platform, UWITEC Co. Ltd. Austria) in October 2012 (Fig. S1); the sampling location is shown in Fig. 1. The sediment core was immediately sliced into 2 cm thick intervals; the slices were placed in plastic zip-lock bags, preserved with ice, transferred to the laboratory and kept at −20 °C until analysis.

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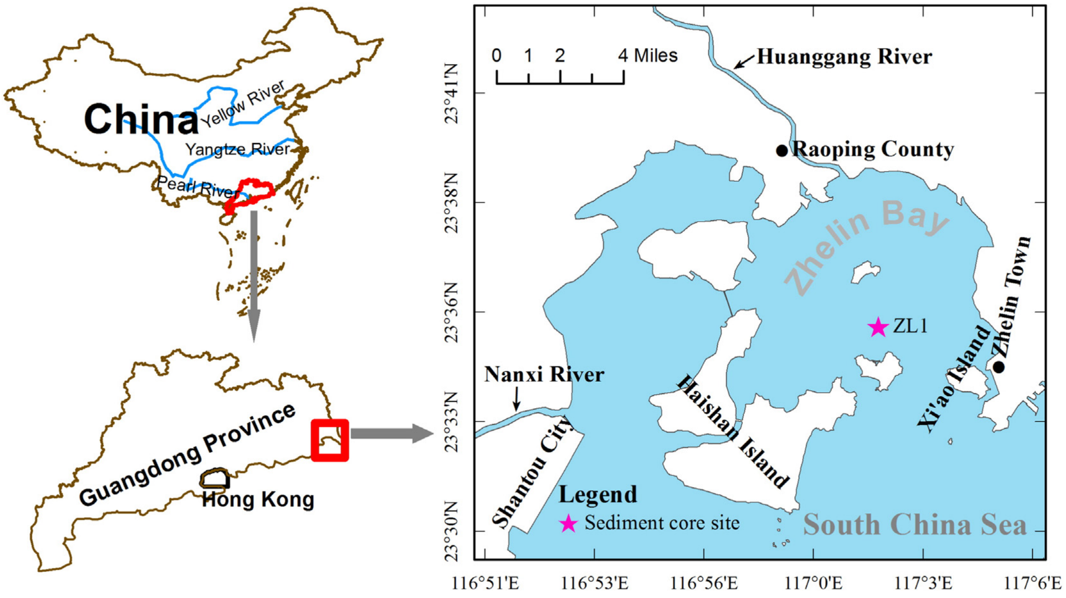

Fig. 1. Maps of the study are and the sampling location in Zhelin Bay, South China.

The frozen samples were over-dried at 40 °C until constant weight, gently ground using an agate pestle and mortar, passed through a 63 μm mesh sieve for homogenization, and stored in glass bottles for metal analyses. Total metal concentrations were measured using X-ray fluorescence spectroscopy (XRF). Briefly, pressed powder tablets were prepared from homogenized samples (<63 μm), followed by direct elemental determination of the samples and of China National Standard material (Offshore Marine sediment, GBW 07314). The sequential extraction protocol described in Rauret et al. (1999) was conducted to obtain information about metal fractions. This scheme splits the metals into four operationally defined chemical fractions: acid soluble, reducible, oxidizable, and residual. The detailed procedure for the sequential extraction applied in this study is described in Gu et al. (2012b). Metal concentrations of the four chemical fractions were measured through atomic absorption spectrophotometry (AAS, Hitachi Z2000).

To verify the accuracy of the sequential extraction procedure and to monitor the performance of the analytical method, the Chinese national standard sediment sample GBW07436 was analyzed; the ranges of the recoveries of Pb, Cr, Ni, Cu, and Zn in the acid soluble, reducible, and oxidizable fractions were 92 to 109%, 89 to 102%, 96 to 102%, and 98 to 110%, respectively. Another China National Standard material (Offshore Marine sediment, GBW 07314) was analyzed to verify XRF recovery. Recoveries for the nine metals were between 98 and 110%.

Mean, standard deviation (SD), median, and range of trace metal concentration in the sediment core at Zhelin Bay are summarized in Table 1; concentrations of major elements in the sediment core are shown in Table S1. Generally, metal concentrations followed the order: Zn > Cr > Cu > Ni > Pb. Concentrations were highest for Zn, which average Ni concentration was 40.7 mg/kg and Pb concentration below 20.7 mg/kg. Concentrations of Cr, Ni, Cu, and Zn were significantly higher than their corresponding background values based on the result of one-sample *t*-test, indicating pollution of anthropogenic origin.

Mean total concentrations of Cr, Ni, Cu, and Zn in the sediment core were considerably higher compared to the average crust values; mean total concentration of Pb was comparable to the average crust value (Table 1). The results from this study were also compared with those from other bays/estuaries in the China. In Jiaozhou Bay and Daya Bay, mean concentrations of Pb and Cr were higher and Ni, Cu, and Zn concentrations were lower than in our study (Table 1). However, the average concentrations of most metals were lower in Zhelin Bay than in the Pearl River Estuary (Table 1).

Vertical distribution of trace metals (Pb, Cr, Ni, Cu, and Zn) in the sediment core from Zhelin Bay is illustrated in Fig.2 and the profiles of major metals (Al, Fe, Ti and Mn) are shown in Fig. S2. For most metals, we found higher concentrations in the top layer of the sediment core. Two vertical distribution patterns (1 and 2) of metals in the core occurred in the depth profiles. *Pattern* 1 (gradual increase distribution): concentrations for Pb, Cr, Ni, and Zn increased gradually from the bottom to the top layer. *Pattern* 2 (uniform distribution): concentration of Cu varied in a relatively narrow range with increasing depth. Sediment cores can reflect historical inputs of contaminants into aquatic systems (Mudroch and Azcue, 1995). A previous study conducted on the western Guangond coast has shown an average sedimentation rate of 1.86 cm/year in a typical marine culture base (Yu et al., 2011). Accordingly, our sediment core from the Zhelin Bay may reflect changes of metal composition for over the last 58 years, quite possibly resulting

Table 1
Summary of trace metal concentrations in the sediment core from Zhelin Bay compared with the average metal concentration in sediments of other bays/estuary (mg/kg, dry weight).

		Pb	Cr	Ni	Cu	Zn
This study	Mean,	20.7	74.6[a]	40.7[a]	55.9[a]	169.0[a]
	SD	±15.4	±11.6	±6.0	±13.0	±11.9
	Median	15.7	74.2	39.6	54.9	167.9
	Range	3.2–59.7	54.3–102.0	32.0–58.3	36.5–81.5	148.0–195.5
BV[b]	Mean	40.3	28.6	14.9	11.8	38.0
UCC[c]	Mean	17	35	18.6	14.3	52
Jiaozhou Bay[d]	Mean	24.2	79.3	29.2	29.7	71.2
Daya Bay[e,f]	Mean	45.7[e]	75.6[f]	31.2[e]	20.8[e]	113[e]
PRE[g]	Mean	88.5	138	76.6	105	274
ERL[h]		47	81	20.9	34	150
ERM[h]		218	370	51.6	270	410

[a] $p < 0.01$ significant level
[b] Background Values (BVs) (Wang et al., 2013)
[c] Upper Continental Crust (Hans Wedepohl, 1995)
[d] (Deng et al., 2010)
[e] (Gao et al., 2010)
[f] (Yu et al., 2010)
[g] (Liu et al., 2011)
[h] ERL (Effects Range Low) guideline values indicate concentrations below which adverse effects on biota are rarely observed and ERM (Effects Range Low) guideline values indicate concentrations above which adverse effects on biota are frequently observed (Long et al., 1995).

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