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Distribution and sources of heavy metals in the sediment of Xiangshan Bay

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Abstract

Heavy metals in the surface sediments and sediment core from the Xiangshan Bay, a mariculture base on the coast of the East China Sea, were determined by inductively coupled plasma mass spectrometry (ICP–MS) in order to evaluate their levels and sources. The results showed that the levels of Cu, Pb, Zn, and Cr in the sediments of the bay were generally influenced by anthropogenic inputs since the founding of the People's Republic of China. In particular, Cu and Zn were polluted to some extent, as evidenced by high enrichment factors. Organic matter, grain size, wastewater discharge, and a low energy hydrodynamic environment played dominant roles in the heavy metal enrichment in the sediments. The ratio of terrigenous source to marine biogenic deposit of trace metals in the sediments was calculated, revealing that terrigenous inputs were the main source of Cu, Pb, and Zn, while biological pellets contributed much more to the enrichment of Cr and Cd. Considering the influence of biological sources on the enrichment of Cd and Cr, and the fact that the sediment has been polluted by Cu and Zn, the development of mariculture and

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discharge of wastewater into the Bay should be restricted.

Key words: anthropogenic effects, biogenic deposits, heavy metals, sediments, enrichment factor

1 Introduction

There has been an increasing awareness that heavy metals in the marine environment cause considerable problems due to their toxicity to marine organisms and harmfulness to the human body through food chains. There are diverse sources of heavy metals, varying from natural inputs (e.g., crustal detritus and biological pellets of organisms) to anthropogenic ones (e.g., wastewater discharge and atmospheric-borne particles). Once introduced into the sea water they are redistributed and eventually deposited in the sediments, directly influencing the benthic organisms living on the sea floor and other marine organisms through re-dissolution of heavy metals into the water column. Various factors (e.g., sources of heavy metals, metal speciation, sediment characteristics, and hydrodynamic conditions) have shown significant impacts on the interaction procedure between heavy metals and sediment. [Duan et al. \(2012\)](#) reported the influence of chemical speciation on metal behaviors; the relationships between heavy metal content and grain size, organic matter, and redox potential in the marine sediments have also been well studied by many authors ([Kalloul et al., 2012](#); [Durán et al., 2012](#); [Cai et al., 2011](#); [Ianni et al., 2010](#); [Point et al., 2007](#); [Santos et al., 2005](#)). However, few studies have investigated the impact of heavy metal sources on their distribution and behaviors in marine sediments, which is an important basis and premise for control and management of heavy metal pollution in the marine environment. In order to provide sufficient evidence for better marine environment management and support rational decision-making, it is thus essential to quantify the contents and sources of heavy metals in the

sediments.

The Xiangshan Bay is one of the most important mariculture bases located on the coast of the East China Sea. It supplies more than 50 000 t of fishes, shrimps, molluscs, and crabs to dining tables every year (Huang et al., 2008). The marine environment and ecosystem of the bay have suffered from industrialization and urbanization since the end of the 1970s. The sea water is polluted by heavy metals due to annual industrial wastewater discharge. It is urgent to assess the heavy metal levels in the sediments of the bay and their potential harmful effects to avoid uncontrolled expansion of mariculture and reduce public skepticism over the safety of seafood.

The aim of this work was to determine the levels of heavy metals, estimate their different sources, identify the main factors controlling their distribution, and discuss the anthropogenic effects on their enrichment in the sediments over the past 80 years.

2 Materials and methods

2.1 Sampling

Figure 1 shows the sampling stations in the study area, where the water depth ranges from 4–19 m. The surface sediments were collected at eight stations with a stainless steel grab. A sediment core with the length of 58 cm was then taken by a multi-corer and sectioned in thicknesses ranging from 2–4 cm. The samples were placed into plastic bags immediately after collection and section. They were stored at a temperature of $-20\text{ }^{\circ}\text{C}$ until freeze-dried before chemical and physical analysis.

Fig. 1. The map of the study area and the locations of the sampling stations.

2.2 Analytical procedures

Metals were determined by inductively coupled plasma mass spectrometry (ICP–MS) (Agilent 7000) after microwave-assisted digestion (CEM Mars) with a HNO_3 -HF mixture according to the US EPA Method 3052. The analytical accuracy was verified by reference material MESS-3 from National Research Council Canada and the repeatability of the data was verified by relative standard deviation (RSD). The recovery was 109% for Cu, 104% for Pb, 102% for Zn, 103% for Cr, 110% for Cd, and 99% for Al. The RSDs were generally less than 5.6%. All plastic and glass laboratory ware used for heavy metal sampling and analysis were soaked with 30% (v/v) HNO_3 (analytical grade, 69% w/w) for 24 h and triple rinsed with MilliQ water. All reagents used for analysis were guaranteed grade.

Total organic carbon (TOC) content was determined by Element Analyzer (Elementar, Germany). In brief, about 1.0 g of dried sample was treated with dilute HCl (1:3 v/v) and rinsed with MilliQ water. This procedure was carried out three times in order to remove carbonate, and then dried overnight at 60 °C before analysis by Element Analyzer. The grain size analysis was carried out by a Sympactec Laser Diffraction Helos System (H2250). The accumulation rate of the sediment was determined by ^{210}Pb dating and the average sedimentation rate was calculated with

the following equation:

$$s = -\lambda / a , \quad (1)$$

where s is the sedimentation rate (cm/a); λ is the decay constant of ^{210}Pb (0.031 a^{-1}); and a is the exponential index of the exponential regression between excess ^{210}Pb activity and core depth.

3 Results and discussion

3.1 Heavy metals in the sediment core

3.1.1 Sedimentation rate determined by ^{210}Pb dating

The average sedimentation rate was 0.75 cm/a calculated from the ^{210}Pb radioactivity attenuation with the least square method (Fig. 2). According to the sedimentation rate, the bottom layer (56–58 cm) of the core represented the years 1934–1936, and the surface layer (0–2 cm) was from the years 2008–2011. In such a way, the changes of heavy metal levels in the sediment during the recent 77 years could be interpreted from the whole core.

Fig. 2. Profile of the excess ^{210}Pb activity.

3.1.2 Vertical distribution of heavy metals

The contents of heavy metals in the sediment core varied from 43.1–50.7 mg/kg for Cu, 31.6–36.6 mg/kg for Pb, 94.6–129.0 mg/kg for Zn, 94.6–111.9 mg/kg for Cr, and 0.33–0.41 mg/kg

for Cd. As demonstrated in Fig. 3a, the contents of Cu, Pb, Zn, and Cr generally increased towards the surface, exhibiting a similar downcore profile. According to the ^{210}Pb dating, the levels of Cu, Pb, Zn, and Cr before the 1950s were the lowest, with average contents of 43.9 mg/kg for Cu, 32.1 mg/kg for Pb, 113.9 mg/kg for Zn, and 97.3 mg/kg for Cr. The heavy metal contents then increased rapidly and the first peak appeared at the beginning of the 1960s. After then, the heavy metal contents increased slightly and peaked again in the early 1980s. Before the founding of the People's Republic of China in 1949, the contents of heavy metals in the sediments were relatively low with low anthropogenic effects. However, the fast and over-emphasized economic development after the founding of the People's Republic of China led to rapid increasing of wastewater discharge into the marine environment and thus heavy metal deposition on the sea floor, especially during the Great Steel-making period at the end of the 1950s. The first peak of heavy metal content is most likely due to this "Great Steel-making" effect. After a short break in economic development during the Cultural Revolution of 1966–1976, the fast industrialization and urbanization also led to the increased burden of heavy metals on the marine sediments, especially since the Reform and Open Policy of 1978.

The influence of industrialization and urbanization on the deposition of heavy metals in the sediments of the Xiangshan Bay was also observed, although it is in a slight manner, which was possibly due to the position of the sampling station. Since heavy metals in the sea water tend to be absorbed by suspended particles and deposited on the sea floor with the sedimentation of the particles, a great portion of heavy metals from wastewater was directly deposited near the outlets and only a small part of the heavy metals was transported with the sea water. This small part of heavy metals diffused in a small area because of the low energy hydrodynamic conditions in the

Bay. In addition, the sediment core was located in the mariculture base and far away from wastewater discharge. It could have been influenced by wastewater only in a slight manner, as were the anthropogenic effects on the heavy metal levels.

The downcore profile of Cd contents (Fig. 3b) did not indicate any significant trend except the two peaks at the early 1960s and 1980s. If the two peaks were directly caused by anthropogenic input of Cd, the increasing trend of Cd content along the core should also be observed just like other heavy metals. Therefore, it is suggested that the content of Cd in the core was not significantly influenced by direct anthropogenic input. It is most likely controlled by marine environmental properties, which are related to the reservoir of Cd in the sediments. These properties changed remarkably during the peak periods and favored the preservation of Cd in the sediments, leading to the two peaks of Cd content in the core.

Fig. 3. The downcore profiles of heavy metals.

3.2 Heavy metals in the surface sediments

3.2.1 Contents and distribution

The contents of metals in the surface sediments varied from 33.0–56.7 mg/kg for Cu,

26.0–37.9 mg/kg for Pb, 84.1–151.8 mg/kg for Zn, 59.0–93.1 mg/kg for Cr, 0.36–0.41 mg/kg for Cd, and 3.31%–5.37% for Al. The heavy metal contents showed comparable results to reports of the same bay in different times ([Compilation Committee of *Gulf Topography of China*, 1993](#); [Zhou, 2005](#)) and with other important domestic estuary and coastal sediments ([Liu et al., 2002](#); [Guo et al., 2004](#); [Fang, 2007](#); [Dong et al., 2009](#)). The temporal and spatial comparisons suggested that the heavy metal levels in the sediments of the Xiangshan Bay changed slightly in the past 10 years, which was verified by the stable downcore profile of the heavy metals in the sediment core since the end of the 1990s (Fig. 3). The increasing of environmental awareness and active measures against marine environmental pollution in the 1990s led to the strict control of industrial wastewater discharge and waste gas and smoke emissions in China, however, this did not contribute to significant changes of heavy metal contents in the sediments in recent years.

With the exception of extremely high contents of Cu and Zn at Sta. X1, the spatial distributions of metals were generally similar, with higher contents at Stas X7 and X9, and lower content at Sta. X17 (Fig. 4). Station X1 was suspected to be contaminated by Cu and Zn, which was confirmed by the results from enrichment factors in the following text. Stations X7 and X9 are located in the narrow channel close to the shore. Long-term suffering from terrigenous inputs combined with less water exchange and slow diffusion has led to high enrichment of heavy metals in the sediments. However, Sta. X17 is on the open mouth of the bay with a long distance to the shore, leading to less terrigenous inputs and rapid diffusion of heavy metals in the water column. In this way, the deposition of heavy metals at Sta. X17 was relatively low.

Fig. 4. Distribution of heavy metals in the surface sediments.

Cluster analysis was conducted using SPSS 17.0 software in order to categorize the studied stations according to the contents of metals and TOC, proportion of clay fraction, and water depth. In this study, the stations were classified into two or three clusters (Table 1). Stations in the same cluster exhibited similar behavior of heavy metals in the surface sediments. In other words, the heavy metals should have similar sources or are governed by similar biogeochemical process. Table 1 shows the results of cluster analysis, revealing that Sta. X1 belonged to its own category, or together with Stas X7 and X9 in the same category. Based on the observation of the highest heavy metal contents at these stations, we suspected that the heavy metals at Sta. X1, or together with Stas X7 and X9, were significantly influenced by human activities, while others were not.

Table 1. Cluster membership of the sampling stations

Case	Station	3 Clusters	2 Clusters
1	X1	1	1
2	X3	2	2
3	X5	2	2
4	X7	3	1
5	X9	3	1
6	X10	2	2
7	X13	2	2
8	X17	2	2

The analysis of principal components was performed on the metal concentrations. The result showed that the first two components accounted for 87.2% of the variance of the phenomenon. In particular, the first component accounted for 61.0% and the second for 26.2%. The associated component plot and matrix are shown in Fig. 5. Zn, Cu, Pb, and Cr were heavily weighted by Component 1 and this aspect reflected the correlations evidenced by the matrix of Pearson coefficients (Table 2). Al and Cd were weighted by Component 2. Al is generally considered to originate from the crustal source and without anthropogenic influence in the marine sediments, so the highly significant difference for Al and Cd suggested that Cd was not significantly influenced by anthropogenic inputs. The elements in Component 1 were noticeably different from those in Component 2, which means that Cu, Pb, Zn, and Cr took part in the phenomenon in a different manner than Cd. Since Cd is mainly from natural sources, the distributions of Cu, Pb, Zn, and Cr were most likely influenced by human activities, which confirmed the results from the sediment core study in the above text.

Fig. 5. Component plot in rotated space and rotated component matrix. Extraction method: Principal Component Analysis. Rotation method: Varimax with Kaiser Normalization.

3.2.2 Interaction between metals and other components

Grain size and organic matter are the most important sediment properties that influence the capacity of sediments to concentrate heavy metals (Giordano et al., 1999; Cai et al., 2011). Generally, heavy metal contents increase with decreasing grain size (Förstner and Wittmann, 1983), and the mud fraction ($< 63 \mu\text{m}$, F63) is usually chosen for environmental investigation for its well-known capability to bind elements by complex mechanisms of absorption and/or adsorption (Tessier et al., 1984). However, F63 comprised more than 90% of all fractions and did not differ significantly for each station in the Xiangshan Bay. Thus, the clay fraction ($< 4 \mu\text{m}$) was chosen to investigate the relationship between heavy metal contents and grain size. The clay fraction contents in the bay were relatively high with the range varying from 26.8%–47.1%, reflecting high organic matter inputs and low energy hydrodynamic environment, which were prone to the absorption and/or adsorption and retention of chemicals from anthropogenic sources in the sediments. Table 2 lists the Pearson coefficients between metals and sediment properties.

The results showed that the contents of Cu, Zn, and Cr increased with the clay proportions, indicating considerable association of Cu, Zn, and Cr with the clay fraction. In contrast, the contents of Cd and Pb were not significantly correlated with the clay proportion, nor with silt, mud, and sand proportions. It implied that grain size was not the dominant factor controlling the distribution of Cd and Pb in the surface sediment of the Bay. The contents of TOC in the surface sediments ranged from 0.42%–0.74%. TOC generally co-varied with heavy metal contents (Table 2) and demonstrated similar results as those reported by other authors. Heavy metals tend to be absorbed by biological materials (Ianni et al., 2010), thus organic matter is a good metallic scavenger (Wasserman et al., 1998) and played a major role in the metal reserve in the sediments of the bay.

Pearson coefficients showed that Cu, Pb, and Zn were strongly correlated ($p < 0.01$), as were Cr and Cd ($p < 0.05$). Good correlation between Cr and Cd suggested that they had similar sources or were controlled by similar biogeochemical process. As mentioned above, Cr as well as Cu, Pb, and Zn were influenced by anthropogenic inputs, however, Cd was not, which meant that the hypothesis that Cd and Cr came from similar sources was not supported. Therefore, the contents of Cr and Cd in the sediment were both governed by a similar process, such as organic matter, which had good correlation with both Cr and Cd. However, Cu, Pb, Zn, and Cr were influenced by human activities, and the source composition controlled their distributions.

Table 2. Pearson coefficients between heavy metal contents and sediment properties

	Cu	Pb	Zn	Cr	Cd	Clay	TOC	Al
Cu	1							
Pb	0.87**	1						
Zn	0.94**	0.88**	1					
Cr	0.61	0.51	0.66	1				
Cd	0.38	0.48	0.34	0.75*	1			

Clay	0.64	0.41	0.75*	0.73*	0.12	1		
TOC	0.63	0.47	0.64	0.97**	0.72*	0.72*	1	
Al	-0.01	0.27	0.02	0.27	0.71*	-0.36	0.14	1

Notes: ** $p < 0.01$; * $p < 0.05$.

3.3 Enrichment factor and metal sources estimate

The enrichment factor (EF) is a well known indicator for revealing the natural or anthropogenic inputs of heavy metals (Fernandes et al., 2011). It is a ratio of heavy metal concentration to the reported natural abundance of metal in soils or crustal rocks, by normalizing against a geochemical marker (e.g., Al, Ti, or Li), represented as follows:

$$EF = (M/Al)_{\text{sample}} / (M/Al)_{\text{nature}}, \quad (2)$$

where $(M/Al)_{\text{sample}}$ is the metal to Al ratio in the samples, and $(M/Al)_{\text{nature}}$ is the natural abundance of metal to Al, which usually refers to the M/Al ratio in crustal rocks, the background values of local soils or sediments without “pollution”. A value of EF less than 2 reflects insignificant contamination. In this study, the average contents of heavy metals and Al in the bottom of the sediment core were selected as the background values of sediments, in brief, 43.9 mg/kg for Cu, 31.6 mg/kg for Pb, 113.2 mg/kg for Zn, 95.7 mg/kg for Cr, 0.37 mg/kg for Cd, and 5.69% for Al. The calculated enrichment factors are listed in Table 3. The results indicated that the surface sediments were generally not polluted by heavy metals ($EF < 2$). However, it should be noted that the EF at Sta. X1 were much higher than those at the other stations. In particular, the EF of Cu and Zn were higher than 2. Station X1 is located in the semi-enclosed Tie Port, where long water exchange period and low hydrodynamics are expected, leading to slow diffusion of heavy metals in the sea water and effective deposition on the sea floor. Furthermore, this station received the heaviest burden from industrial wastewater discharge in comparison with the other sea areas in the Xiangshan Bay (Huang et al., 2008). It could be concluded to some extent that the trace metal

levels in the Tie Port were greatly influenced by anthropogenic effects.

Based on the bottom sediment composition of the bay, which is regarded to have natural sediment composition, one can estimate the quantity of heavy metals present from pollution. The natural source of Cu and Zn in the sediment at Sta. X1 was calculated as follows:

$$M_{\text{nat, sed}} = Al_{\text{sample}} \times (M/Al)_{\text{nat, sed}} . \quad (3)$$

The contaminated portion of Cu and Zn was then obtained as:

$$M_{\text{pollut}} = M - M_{\text{nat, sed}} , \quad (4)$$

where $(M/Al)_{\text{nat, sed}}$ is the ratio of metal to Al in the natural sediment, $M_{\text{nat, sed}}$ is the natural source of heavy metals, and M_{pollut} is the contaminated portion of heavy metals. The result showed that the natural contents at Sta. X1 should be 25.5 mg/kg for Cu and 65.8 mg/kg for Zn. Thus, the contaminated portion contributed 31.2 mg/kg for Cu and 86.0 mg/kg for Zn.

Based on the average unpolluted soil composition of Zhejiang Province (Liao, 1992), one can also estimate how much of the heavy metal in unpolluted sediment is present from terrigenous materials, as follows:

$$M_{\text{terr}} = Al_{\text{sample}} \times (M/Al)_{\text{soil}} . \quad (5)$$

The heavy metal from marine biological pellets was then obtained as:

$$M_{\text{bio}} = M - M_{\text{terr}} , \quad (6)$$

where $(M/Al)_{\text{soil}}$ is the ratio of metals to Al in the background soil, M_{terr} is the terrigenous source of heavy metals, M_{bio} is the part from marine biological inputs, and M is the metal content of the unpolluted sediment. Since $EF < 2$ suggests no contamination, M is the determined total content of heavy metal except the contents of Cu and Zn at Sta. X1, which were influenced by human activities. For Sta. X1, the calculated $M_{\text{nat, sed}}$ of Cu and Zn were used as M in Eq. (6). Following

this procedure, one can estimate the value of $M_{\text{terr}}/M_{\text{bio}}$. The result (Table 3) showed that Cu, Pb, and Zn were mainly from terrigenous inputs, but biological inputs contributed much more to Cr and Cd in the sediment, especially to Cd, which was almost three times higher from biological inputs than from terrigenous ones.

Table 3. Calculated enrichment factors and ratios of terrigenous input to biogenic input (terr/bio) of heavy metals

Station	Cu		Pb		Zn		Cr		Cd	
	EF	terr/bio	EF	terr/bio	EF	terr/bio	EF	terr/bio	EF	terr/bio
X1	2.2	2.49 ¹⁾	1.9	0.75	2.3	3.63 ¹⁾	1.4	0.59	1.7	0.27
X3	0.9	3.37	1.2	2.09	1.1	2.74	0.7	2.28	1.2	0.41
X5	1.3	1.27	1.4	1.37	1.6	0.91	1.2	0.74	1.5	0.30
X7	1.3	1.30	1.3	1.88	1.3	1.39	1.0	0.99	1.4	0.34
X9	1.4	1.07	1.4	1.45	1.5	1.06	1.1	0.82	1.4	0.33
X10	1.0	2.15	1.1	2.63	1.2	1.86	1.2	0.76	1.5	0.32
X13	0.9	3.24	1.0	5.11	1.1	2.89	0.8	1.56	1.2	0.42
X17	1.1	1.88	1.2	2.22	1.1	2.88	1.0	1.04	1.6	0.29

Notes: ¹⁾ Calculated from natural contents, 25.5 mg/kg for Cu and 65.8 mg/kg for Zn.

Furthermore, negative correlations were observed between EF and the terr/bio ratio of Cd and Cr (Fig. 6). The results showed that the greater the proportion of biological input, the higher the enrichment of Cd and Cr in the sediments. Biogenic deposition played a dominant role in the enrichment of Cd and Cr in the sediments and should be controlled in order to protect the mariculture environment of the sea floor, particularly at Sta. X1, where the EFs for Cd and Cr were much higher. It was reported by [Huang et al. \(2008\)](#) that Tie Port bred much more marine products than the other sea areas in the Xiangshan Bay and the annual feeding materials put into this port were also the highest (22 487 t/a), which led to the high biogenic deposition and consequently to the high enrichment of Cd and Cr in the sediments. Therefore, we suggest that the

development of mariculture and discharge of wastewater in this port should be restricted, considering the influence of biological inputs on the enrichment of Cd and Cr, and the fact that the sediments in the port have been contaminated by Cu and Zn.

Fig. 6. Correlations between *EF* and terr/bio ratio of Cd and Cr in the sediment.

4 Conclusions

The heavy metal levels of Cu, Pb, Zn, and Cr in the sediment of the bay were generally influenced by anthropogenic effects. In particular, Cu and Zn were polluted to some extent, as evidenced by high enrichment factors. However, the direct effect of anthropogenic input on the levels of Cd in the sediments was not observed. Grain size, organic matter, wastewater discharge, and hydrodynamics played dominant roles in the enrichment of heavy metals in the sediments of the Bay. Terrigenous inputs were the main source of Cu, Pb, and Zn, while biological pellets contributed much more to the enrichment of Cr and Cd in the sediments. Considering the influence of biological sources on the enrichment of Cd and Cr, and the fact that the sediment in the Bay has been polluted by Cu and Zn, the development of mariculture and discharge of wastewater should be restricted.

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