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Contamination of Heavy Metals in Sediments from an Estuarine Bay, South China: Comparison with Previous Data and Ecological Risk Assessment

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Abstract: Heavy metal contamination is an elevating threat to the aquatic environment of estuarine bays. In the present study, surface sediments from Shantou Bay in South China were collected and detected for the concentrations of heavy metals including copper (Cu), zinc (Zn), chromium (Cr), nickel (Ni), cadmium (Cd), and lead (Pb) using ICP-MS. Spatial distribution, temporal trend, and potential ecological risks for the metal pollution were discussed. The results showed that levels of metals generally decreased by the order of Zn > Pb > Cr > Cu > Ni > Cd. Spatial variation of metal contents was observed with an order of Rongjiang River > Niutian Bay > Shanthou Harbor for Zn and Cd. Sediments of the Niutian Bay were observed with higher levels of Cu, Cr, Ni, and Pb. Compared with reported data from the same region at different sampling periods, a low–high–low trend was observed for the concentrations of the six elements, suggesting a great improvement of sediment quality in Shantou Bay. The average I_{geo} values suggested moderate pollution of Cu, Zn, Pb, and Cd. The potential ecological risks of Cu, Pb, Zn, Ni, and Cr were in low levels. More attention should be paid to the Cd pollution, considering its great values of potential ecological risk index. Our findings provided better understanding of heavy metal pollution in estuarine environments.

Keywords: heavy metal; sediment; ecological risk; Shantou Bay

1. Introduction

Heavy metals are dangerous environmental pollutants known for their toxic, bioaccumulative, nonbiodegradable, and persistent natures [1,2]. They may come from some natural processes such as weathering and volcanic eruptions [3]. However, the major sources of the extra heavy metals that are continuously introduced to the environment could be ascribed to human activities such as mining, industrial processes, coal burning, electroplating, pesticide use, automobile exhaust, and so on [4,5]. Although some trace elements such as copper (Cu) and zinc (Zn) are indispensable for organisms, they can harm living beings when exceeding certain thresholds [2]. The other dispensable ones, such as cadmium (Cd) and mercury (Hg), are notorious for causing some tragic poisoning events that have occurred in Japan. It is obvious that various aquatic environments such as oceans, rivers, lakes, estuaries, and wetlands around the world are facing heavy metal problems [6–10].

Heavy metals that enter the aquatic environment may accumulate in the sediments, due to some processes such as adsorption and precipitation, and result in the concentrations of these elements in sediments several times higher than those in the water body [11,12]. Thus, sediments are generally considered as one of the most important sinks of heavy metals in aquatic ecosystems [10,11]. On the other hand, those trace elements stored in the sediments could be leaked from dirty tanks under some conditions and may enter into the food chain, posing health risks to aquatic organisms and humans [2,13]. Sediment has been proven as an effective indicator of metal pollution in rivers and estuaries [7–9]. Therefore, investigations on the levels, distribution, and risk assessment of heavy metals in sediment matrices are of practical meaning to evaluate the pollution status of these pollutants in water environment.

Shantou Bay is an estuarine bay that plays an important role in aquaculture, tourism, and navigation of the South China area due to its favorable geographical condition [14]. A famous harbor of China, Shantou Harbor, is located here. With the economic construction and development of the Shantou Special Economic Zone, large amounts of waste and sewage were released into this bay and pose great pressure on the aquatic environment [15]. Some investigations were conducted focusing on the metal pollution in sediments from this special semiclosed bay in the past few years [14,15], and it is important to take into account the rapid development of urbanization and environmental protection efforts of Shantou City. Thus, it is desirable to study the metal pollution status of Shantou Bay, and those results provide some possibilities to present continuous change of the sediment qualities.

In the present study, surface sediments from Shantou Bay were collected and detected for the concentrations of Cu, Zn, Cd, Cr, Ni, and Pb. The main objectives of this research were to (1) measure the levels and distributions of typical heavy metals in sediments from Shantou Bay; (2) reveal a temporal trend of metal contents compared with data reported in previous studies; and (3) evaluate the potential ecological risks of the heavy metals.

2. Materials and Methods

2.1. Study Areas and Sampling

Shantou Bay, whose flows pass through the Shantou City in Guangdong Province, is an important estuarine bay marked as an important fishery base, international harbor, and metropolis, with great populations in South China (Figure 1). It can be divided into two parts, named Niutian Bay and Shantou Harbor, respectively [16]. Two rivers, Rongjiang River and Meijing River, contribute about 66.25×10^9 m^3 of fresh water per year into this bay [16].

The whole research area in the present study was divided into three groups, which are Rongjiang River (S1–S6), Niutian Bay (S7–S14), and Shantou Harbor (S15–S18). Sampling was conducted in October 2016, and 18 surface sediment samples (mixed by three sub-samples) were randomly collected using a stainless steel grab sampler (Figure 1). Each sample was placed in a clean PE package and stored at −20 °C until further analysis.

2.2. Sample Analysis

In the lab, sediment samples were freeze-dried, ground, and passed through a 0.5 mm sieve. Approximately 0.2 g sediment samples were transferred into the digestion vessel, then a mixture of 5 mL nitric acid, 3 mL hydrochloric acid, and 2 mL hydrofluoric acid was added. The digestion was conducted using a microwave digestion instrument at 190 °C for 30 min. Upon completion of the procedure, evaporation was conducted by using electrothermal treatment. The digested samples were diluted to a specified volume by adding double-distilled water. After filtration, heavy metals were determined for Cu, Zn, Pb, Cd, Ni, and Cr by using an inductively coupled plasma mass spectrometer (iCAP RQ, Thermo Scientific, Germany). Contents of metal elements are presented in mg/kg dry weight (dw).

Quality control included analysis of sample blank, reference materials (ESS4), and duplicate samples. The recoveries of reference materials ranged from 80–136%. All glassware and equipment were acid-washed with 10% HNO_3 and rinsed with double-distilled water for the purpose of avoiding possible contamination.

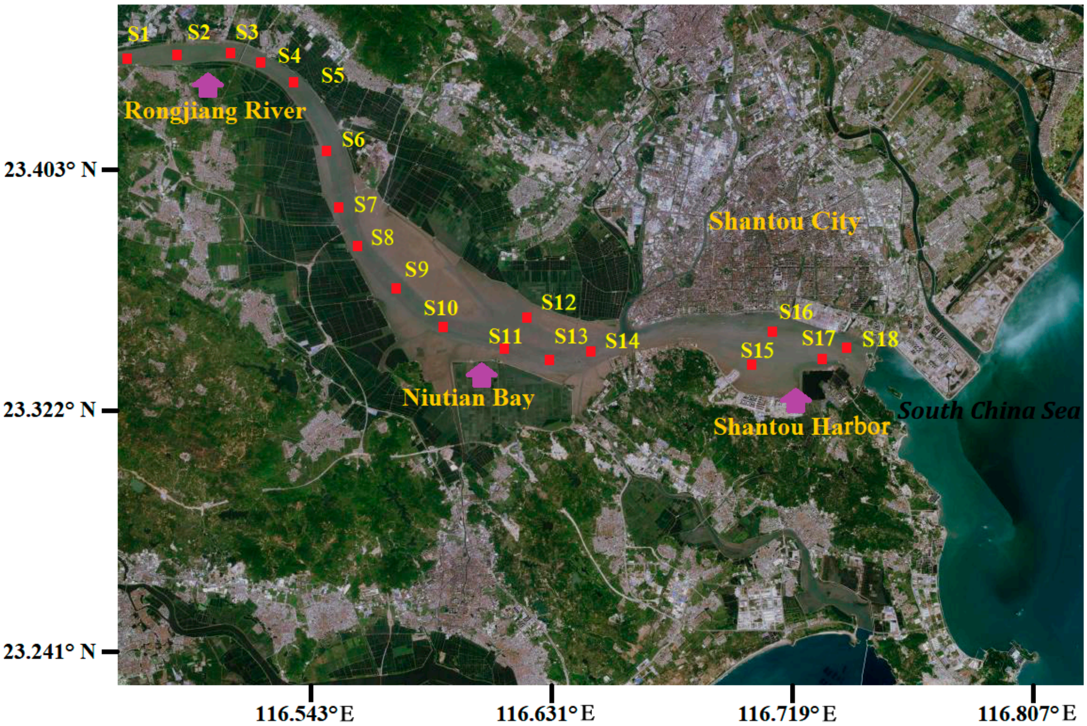

Figure 1. Distribution of sampling sites in Shantou Bay, South China.

2.3. Ecological Risk Assessment Methods

To describe the metal accumulation in the sediments, geoaccumulation indexes (I_{geo}) were calculated using the following equation [17]:

$$I_{geo} = \log_2 (C_i/1.5B_i) \tag{1}$$

where C_i is the concentration of metal i detected in sediment of each sampling site. B_i is the background value of metal i. In the present study, background values of Cu, Zn, Pb, Cd, and Cr in sediments of South China Sea were chosen as the associated references [18]. Background value of Ni in sediments was from that reported in Zhelin Bay, Guangdong Province [19]. A total of 1.5 was a correlation factor given to reflect the natural and anthropogenic effects. Based on the I_{geo} values, the contamination degrees could be divided into non-pollution ($\leq$0), slight pollution (0–1), moderate pollution (1–2), moderate to high pollution (2–3), high pollution (3–4), high to very high pollution (4–5), and very high pollution (>5) [3].

Potential ecological risk index (*RI*) is a common and efficient tool to evaluate the ecological risk degree of heavy metals in sediments, giving sufficient consideration to toxicological impacts [20]. *RI* was calculated using the following equations:

$$E_r^i = T_r^i \times (C_s^i/C_n^i) \tag{2}$$

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \cdot (C_s^i/C_n^i) \tag{3}$$

where C_s^i is the concentration of element i in the sediment sample and C_n^i is the geochemical background value of element i. E_r^i is the potential ecological risk factor of the heavy metal i, T_r^i is the toxic response factor of the element i. The T_r^i values for Cd, Pb, Cu, Ni, Cr, and Zn are 30, 5, 5, 5, 2, and 1, respectively [21]. The grade of ecological risk of a single metal (E_r^i)

could be classified into five categories: low (<40), moderate (40–80), considerable (80–160), high (160–320), and very high (>320). The synergistic hazards of multiple metals (*RI*) were divided into four classes: low (<150), moderate (150–300), high (300–600), and very high (>600).

2.4. Statistical Analysis

Statistical analysis was performed with SPSS 19.0 (SPSS Inc., Chicago, IL, USA). One-way analysis of variance (ANOVA) was used to determine the spatial differences of metal concentrations among the three groups (Rongjiang River, Niutian Bay, and Shantou Harbor). Cluster analysis based on Euclidian distances was used to compare the relationships among different sampling stations.

3. Results and Discussion

3.1. Levels of Heavy Metals in Sediments

The statistical parameters of metal elements in sediments from the Shantou Bay are presented in Table 1. Concentrations of Cr, Ni, Cu, Zn, Cd, and Pb were in the ranges of 23.7–80, 8.2–48, 19–64.5, 44.4–963, 0.2–3.6, and 21.9–103 mg/kg dw, respectively. Levels of metals varied among the sampling sites (Figure 2), with 39% of the samples showing the order of Zn > Pb > Cr > Cu > Ni > Cd. Similar sequence was also reported in a previous study [15]. The standard deviations of metal concentrations (especially for Zn) seemed considerable and this might be due to some natural geological reasons. In addition, point discharge might occur in some stations leading to some big values [15].

Table 1. Heavy metal concentrations in sediments from Shantou Bay (mg/kg).

	Cr	Ni	Cu	Zn	Cd	Pb	TOC (‰)
minimum	23.7	8.2	19.0	44.4	0.2	21.9	5.6
maximum	80.0	48.0	64.5	962.6	3.6	102.9	21.8
average	47.5	21.3	39.7	205.9	1.1	50.3	16.6
median	42.0	19.6	36.7	98.2	0.9	41.1	18.3
SD	14.9	9.0	12.9	250.7	1.0	24.0	4.4

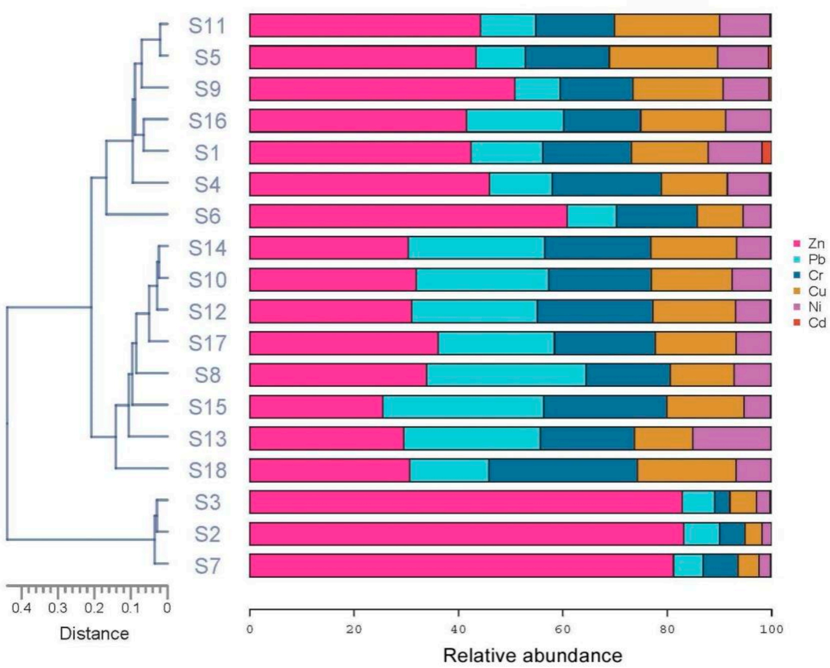

Figure 2. Composition profiles and cluster analyses of heavy metals in sediments of different sampling stations in Shantou Bay.

Compared with the background values of heavy metals in sediments of the South China Sea (39.3, 7.4, 54.5, 0.18, and 15.6 mg/kg for Cr, Cu, Zn, Cd, and Pb), 61%, 100%, 94%, 100%, and 100% of the samples were found with contents above the reference value for Cr, Cu, Zn, Cd, and Pb, respectively [18]. In addition, if compared with the background values for heavy metals in sediments from another bay nearby the Guangdong Province (28, 15, 16, 58, 0.15, and 32 mg/kg for Cr, Ni, Cu, Zn, Cd, and Pb, respectively) [19], high frequencies over the reference doses for each element were also observed. These observations suggest that the sediments in Shantou Bay were polluted by those heavy metals.

Table 2 summarizes contents of related elements in sediments of some other bays or estuaries. Concentrations of Cr in sediments of Shantou Bay were at moderate levels in comparison with those reported in some other regions. Much higher contents for Cr were reported in sediments from Palk Bay in India whose coast received large number of untreated solids and effluents [3]. Concentrations of Ni in Shantou Bay were almost equal to those reported in other references, while Cu levels in Shantou Bay seem in a low to medium grade. It was noteworthy that the Zn, Cd, and Pb concentrations in our study were, relatively, much higher. Pollution of Zn, Cd, and Pb in the Shantou Bay might be ascribed to the following reasons. Firstly, there were several mines in the upper region of Rongjiang River, and the metal elements in the mine solid waste and acid wastewater might enter the river through the surface runoff [15]. Secondly, fertilizer and pesticide use in the agriculture of the surrounding farmlands probably resulted in input of Cd in the bay [14]. Furthermore, Pb possibly came from the combustion of fossil fuels from automobiles and industries. In addition, industries such as toy production, equipment manufacturing, and electrical and electronic appliances manufacture of Shantou City are vigorously developed. Wastewater originating from these industries contained varieties of metals such as Cd, Zn, and Cu. The continuous discharge of treated and untreated effluent should not be ignored.

Table 2. Levels of heavy metals in sediments from different bays (mg/kg).

	Cr	Ni	Cu	Zn	Cd	Pb	Reference
Shantou Bay, China	47.5	21.3	39.7	205.9	1.1	50.3	This study
Laizhou Bay, China	17	-	7	-	0.08	14	[1]
Palk Bay, India	290	28	55	253	1.2	14	[3]
Haizhou Bay, China	44	-	14	42	0.08	22	[12]
Fudu Estuary, China	25	-	8	28	0.1	20	[21]
Meishan Bay, China	68	-	22	53	0.3	19	[22]
Laucala Bay, Fiji	24–48	23–35	44–170	15.5–157	1.7–6	50–80	[23]
Laizhou Bay, China	60	-	19	56	0.1	20	[24]
Hailing Bay, China	45	-	7.4	36	0.07	14.5	[25]
Pear River Estuary, China	-	33	55	139	-	36	[26]

3.2. Spatial Distribution and Temporal Trends of Metals

Spatial variations of trace element levels in sediments from the Shantou Bay were shown in Figure 3. It was obvious that concentrations of Zn and Cd decreased as a sequence of Rongjiang River (S1–S6) > Niutian Bay (S7–S14) > Shantou Harbor (S15–S18) ($p < 0.05$). The samples of Niutian Bay were observed with greater levels of other elements. Much higher contents of Zn and Cd in the Rongjiang River might be ascribed to the mining activities from the upper regions, while the relative greater amounts of other elements in Niutian Bay might be due to its special torose shape available for the sedimentation of pollutants both from the upper stream of Rongjiang River and Shantou City. The dilution effect of seawater could be the major explanation for the low contents of metals in Shantou Harbor.

A comparison with historical data [14,15] reported in the same region (Niutian Bay and Shantou Harbor) of Shantou Bay was shown in Figure 4. An obvious low–high–low trend could be observed for all the trace elements. This observation might correspond to the socioeconomic development of Shantou City. In the early phase (2002–2011) of the figure, the elevating trend for the metal concentrations could reflect a vigorous economic development with lack of attention on environmental protection. Accompanying the ecological civilization construction process in China, many more efforts have been taken to protect the environment by local sectors. The concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in sediments decreased by 76%, 60%, 61%, 46%, 64%, and 45%, respectively, from 2012 to 2016. Thus, the observed decreasing pollution tendency during 2012–2016 could reflect some achievements of the environmental protection steps of the local governments.

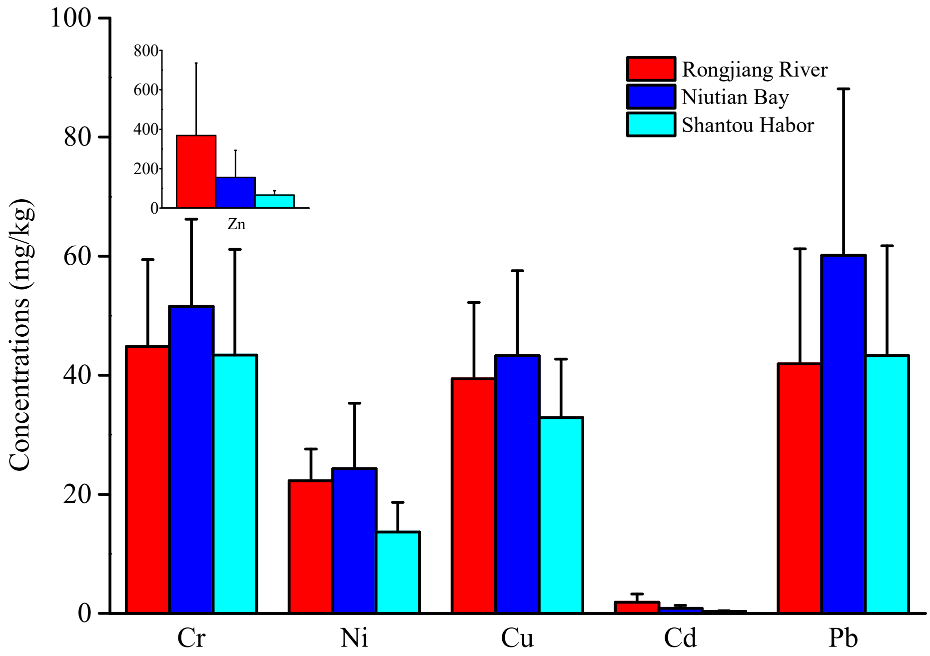

Figure 3. Spatial variations for metal concentrations in sediments of Shantou Bay.

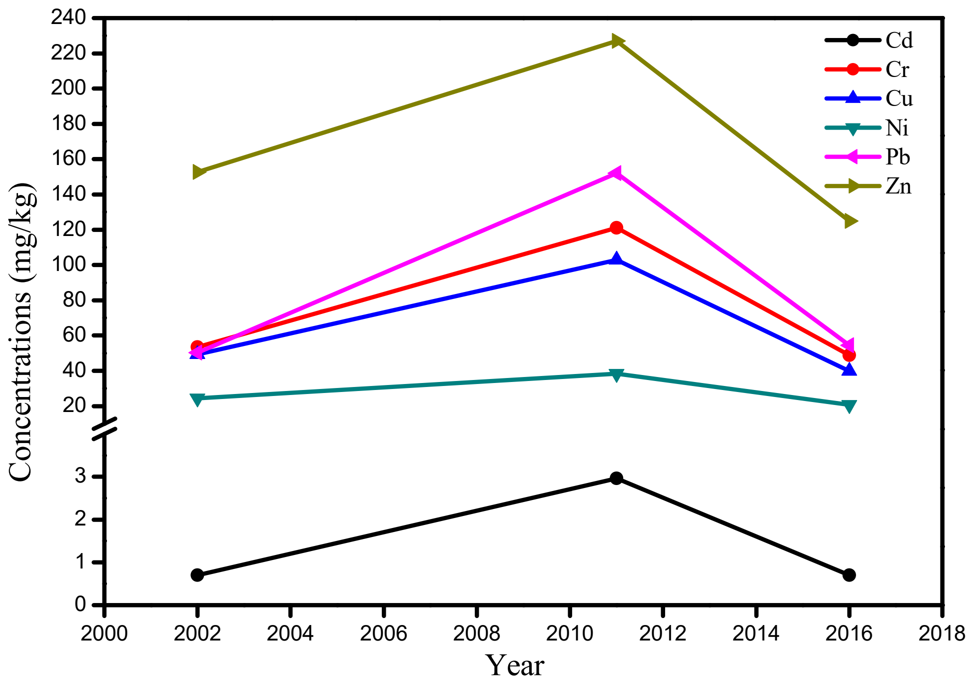

Figure 4. Temporal trend for metal concentrations in sediments of Shantou Bay.

3.3. Risk Assessment

The values for the I_{geo} of heavy metals in sediments from Shantou Bay are presented in Figure 5. The average I_{geo} values revealed an order of Cu (1.76) > Cd (1.53) > Pb (0.95) > Zn (0.71) > Ni (-0.19) > Cr (−0.38). Specifically, I_{geo} values for Cr ranged from −1.3 to 0.44, suggesting an unpolluted to slightly polluted degree of Cr. I_{geo} values for Ni were from −1.46 to 1.09, indicating an unpolluted to slight contamination of Cr. Therefore, the pollution of Ni and Cr in surface sediments Shantou Bay tended to be negligible, and their pollution degrees were significantly lower than those of Cu, Zn, Cd, and Pb ($p < 0.05$).

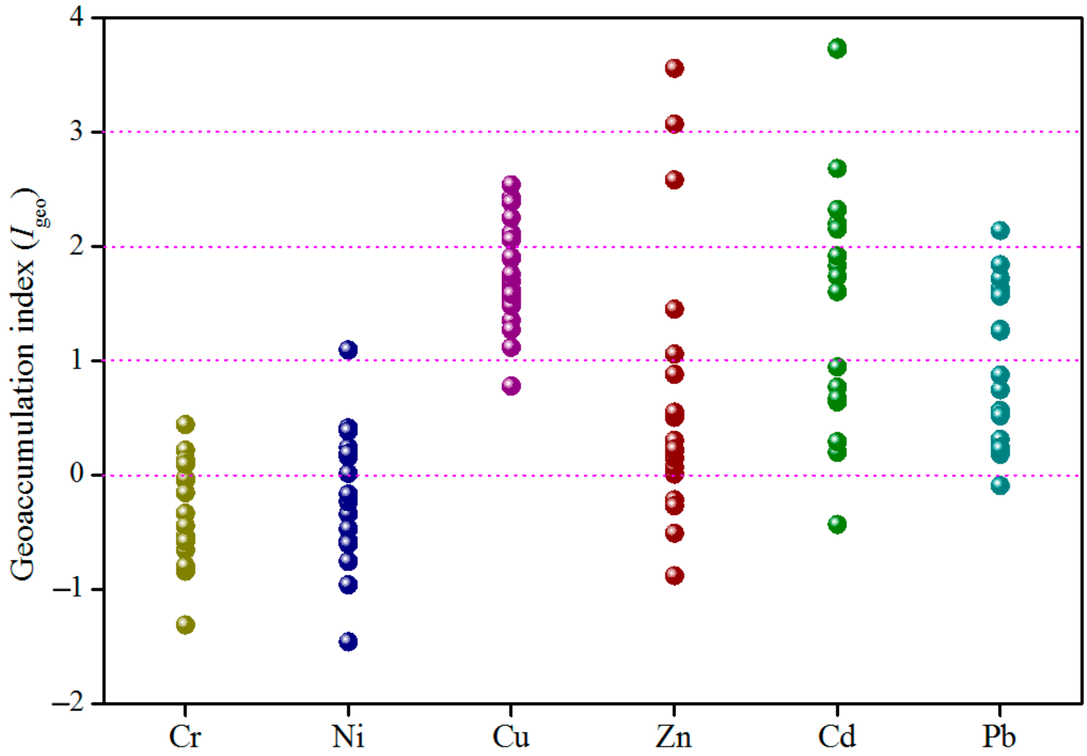

Figure 5. Geoaccumulation index for elements in sediments of Shantou Bay.

As for the other four elements, their contamination in the sediments should not be ignored by considering their relatively high I_{geo} values. I_{geo} values for Cu (0.78–2.54) showed a moderate to high pollution of Cu in a few of the sampling sites (S3, S5, S9, S11, S12, and S14). Pb with I_{geo} values of −0.09–2.14 tended to be in a slight to moderate level in most of the sampling sites. It is worth mentioning that the Zn and Cd pollution grades varied sharply among different stations. Greatest I_{geo} values for Zn and Cd were above 3.5, which suggested high pollution status in a few sampling stations of Rongjiang River (S2 and S3 for Zn; S1 and S3 for Cd). Therefore, attention should be given to the Zn and Cd pollution in sediments of Rongjiang River, which might be caused by the mining from the upper areas. In summary, based on the I_{geo} indexes, sediments in Shantou Bay could be considered moderately polluted by Cu, Zn, Pb, and Cd.

In the present study, E_r^i and *RI* were used as indicators to assess the potential ecological risk of the six heavy metals (Figure 6). The E_r^i values for Cr, Ni, Cu, Zn, Cd, and Pb were in the ranges of 3.02–10.2, 1.09–6.41, 12.9–43.6, 0.81–17.7, 33.3–600.3, and 7.02–33.0, respectively, generally showing an order of Cd > Cu > Pb > Cr > Zn > Ni. This sequence is in line with that reported by Li et al. [15] but slightly different from that reported by Qiao et al. (Cd > Cu > Pb > Ni > Zn > Cr) [14].

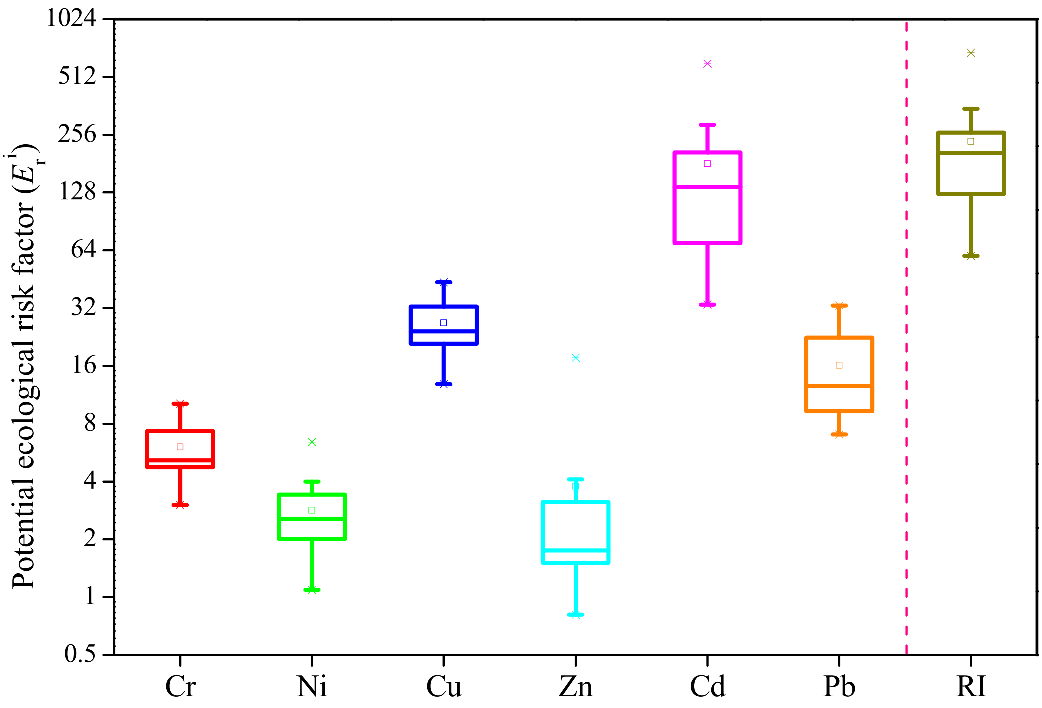

Figure 6. Box plot of potential ecological risk indexes for heavy metals in Shantou Bay.

Cr, Ni, Zn, and Pb in sediments from Shantou Bay had low ecological risk (E_r^i < 40) at each sampling station, and in most of the sampling stations, the potential ecological risks of Cu were low, too. E_r^i values of Cu in sediments of S3 and S14 were above 40, indicating moderate risk. Much higher values of E_r^i for Cd were observed in sediments of S1 (600), S3 (595), and S9 (350), suggesting extremely high risks. The average E_r^i of Cd was 236, representing a high risk. These observations suggested severe Cd pollution in sediments of Shantou Bay, especially for Rongjiang River. Cd is very toxic, and highly bioaccumulative and great amounts of Cd might pose critical threat to aquatic species and human health [11,15].

The values of *RI* ranged from 60 to 685 (60–350 when only considering Niutian Bay and Shantou Harbor), which indicated that different ecological risks were posed to the aquatic environment of Shantou Bay by the six metal elements. Two stations (S1 and S3) were observed with very high risk and one station (S5) had high risk due to the great contribution of Cd. It should be noted that in all the sampling stations, *RI* values were mainly contributed by great E_r^i of Cd (51–94%), leading to a spatial order of Rongjiang River > Niutian Bay > Shantou Harbor. The *RI* values reported in the previous study ranged from 115 to 603 in 2002 [27], while they reached >2000 (253 when excluding Cd) in 2011 [15]. Although the *RI* values showed an decreasing trend, the risk grades were still high. There was no doubt that the severe pollution of Cd was a key factor that might cause harm to the aquatic ecosystem of Shantou Bay.

4. Conclusions

Accompanying the urbanization and industrialization of the coastal cities, environmental problems caused by heavy metals should be paid enough attention. Thus, we performed this study to evaluate the concentration levels, tempo–spatial distribution pattern, and ecological risks of six selected heavy metals in the sediments of Shantou Bay. Our results demonstrated that Shantou Bay was polluted by heavy metals. Geographical, social, and economic reasons lead to the spatial variabilities of the concentrations of different elements. A comparison with historical data of heavy metal contents revealed that the sediment qualities tended to improve with the increasing regard to environment protection. Overall, the potential ecological risks of the six metals could not be taken lightly since the indicators such as I_{geo}, E_r^i and RI tend to give a warning. Among the six metals, Cd should be given special concern because it poses a major threat to the aquatic environment for its high toxicity and relatively large amounts over the background values. Therefore, it is of importance and necessity to constantly monitor the metal pollution in this unique estuarine bay.

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