Concentrations of Cu, Pb, Zn, Cr and Cd in Surficial Sediments from Mangrove Wetlands of China

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Abstract: Mangrove sediments were sampled from 14 main mangrove reserves of China. Concentrations of Cu, Pb, Zn, Cr and Cd were analyzed with a PE ICP-MS, and physical and physicochemical factors such as particle size distribution, pH, salinity and Total Organic Matter (TOM) were also determined. In the mangrove sediments of China, the average contents of heavy metals decreased in the order of $Zn > Pb > Cu > Cr > Cd$. The average contents of heavy metals were: $Cu = 22.8 \pm 16.46$ mg/kg, $Pb = 76.1 \pm 52.33$ mg/kg, $Zn = 86.3 \pm 42.65$ mg/kg, $Cr = 18.4 \pm 5.83$ mg/kg and $Cd = 0.21 \pm 0.25$ mg/kg. Compared with previous studies, levels of heavy metals increased greatly. Contents of Cd, Cu, Pb and Zn were higher than the Background Values (BV). However, the contents of Cr was under the BV. The results revealed that levels of heavy metals changes significantly with anthropogenic input, physical and physicochemical factors. Significant correlations were found between concentrations of Cu, Pb and pH, TOM.

Key words: Mangrove, surface sediments, heavy metal, China.

1. Introduction

Mangrove wetlands are important ecosystems. They provide breeding and feeding habitats for various organisms, and protect shorelines from erosion [1, 2]. In recent years, solid wastes and wastewaters metals have been discharged into mangrove wetland in southeastern China. Because of periodic water logging of tides, mangrove sediments are potential sinks for anthropogenic metal contaminants [3-5]. Heavy metals are commonly retained within mangrove sediments [3, 6, 7]. After reaching mangrove wetlands, heavy metals tend to form insoluble compounds, mainly as metal-sulfides and metal-organic matter complexes, and/or adsorb onto the surface of particles [3, 7, 8]. Due to biogeochemical cycle and exchange between sediment and water, heavy metals enter food chain and cause public health concern [9]. Zheng et al. [10-13] studied cycling of heavy metals in mangrove community in Shenzhen and Fugong. He et al. [14] investigated distribution of heavy metals in mangrove

sediment of Shankou. Ding et al. [15] summarized distribution of Hg in mangrove sediments of China. Li et al. [16] reported that distribution and mobility of copper, zinc and lead in plant-sediment systems of Quanzhou bay estuary. All these studies, however, only focused specifically on either particular heavy metal species or heavy metal pollution within a certain area, therefore, a current systematic information on heavy metal pollution in surficial sediments of major mangrove reserves in China is still absent. In this paper, authors studied the accumulation of Cu, Pb, Zn, Cr and Cd in sediments from 14 main mangrove reserves of China, and discussed influencing factors including pH, TOM, salinity, particle size distribution, levels of Fe and Mn.

2. Materials and Methods

Mangrove wetlands are mainly distributed in the coasts of southeastern China, from 18°09' N to 27°20' N. Over 90% mangrove wetlands in China occur in Hainan Province, Guangdong Province, Guangxi Autonomous Region and Fujian Province [2]. These 14 selected areas almost cover all important mangrove distribution of China (Fig. 1).

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Fig. 1 Study area.

Totally 91 sediment samples (0–20 cm) were collected with a plastic scoop during low tide. Samples were put in clean valve bags, and brought to the laboratory. Sediments were also subjected to granulometric measurements. A fraction of fresh sample (1.0–2.0 g) was separated for grain size analysis by laser diffraction using a Mastersizer 2000 (Malvern) with a range of 0.02–2,000 μm . The particles were classified as sand (0.05–1 mm), silt (0.001–0.05 mm) and clay (< 0.001 mm). Next, the samples were air dried and weighed to obtain dry masses. After drying, the sediment samples were ground with an agate mortar and sieved with a 100-mesh sieve.

The samples were digested with HNO_3 (AR)– HCl (AR)– HClO_4 (AR) [17], and then concentrations of Cu, Pb, Zn, Cr, Cd, Fe and Mn were analyzed with a PE ICP-MS. Moisture, pH and TOM were measured also according to ISSCAS [17], and salinity (S) was measured with a conductometer. The standard reference material (GSD1-3) and blind duplicates served as the quality control.

The mean, standard deviation and range of the heavy metal concentrations and parameters of sediments were calculated. All data were expressed in terms of air-dried masses. One-way ANOVA was applied to test the significant differences of sediment properties. All statistical analyses were carried out using the SPSS 19.0 for windows.

3 Results

3.1 Contents of Heavy Metals

A wide range of values for heavy metal

concentrations was observed for the surficial sediments from main mangrove reserves of China (Table 1) [18]. Levels of major heavy metals Cu, Pb, Zn, Cr and Cd were within the ranges of 5.69–69.80 mg/kg, 23.56–180.13 mg/kg, 34.87–193.72 mg/kg, 9.01–30.29 mg/kg and 0.05–0.98 mg/kg respectively. Average concentrations of metals in mangrove sediments were: $\text{Cu} = 22.8 \pm 16.46$ mg/kg, $\text{Pb} = 76.1 \pm 52.33$ mg/kg, $\text{Zn} = 86.3 \pm 42.65$ mg/kg, $\text{Cr} = 18.4 \pm 5.83$ mg/kg and $\text{Cd} = 0.21 \pm 0.25$ mg/kg. In the mangrove sediments of China, mean concentrations of heavy metal were in the order of: $\text{Zn} > \text{Pb} > \text{Cu} > \text{Cr} > \text{Cd}$. Each individual area displayed a wide variation in metal concentration of sediments as reflected by the large standard deviation values.

Compared with the Background Value (BV) of sediments from the China shelf sea, the contents of Cr for all samples were under the BV, and the contents of Cd for all samples were much higher than the BV. However, Cu, Pb, Zn contents fluctuated around the BV, and most of them were higher than the BV.

The correlation among individual elements can give information on the geochemical associations of elements, mobility of metals, and information on the sources and sink of trace metals [19]. The correlation matrix is given in Table 2. Significant correlations were found among concentrations of Cu, Zn, Cr and Cd (Table 2), which indicated that these heavy metals might be discharged from the similar pollution source and had similar depositional behavior.

3.2 Geochemical Characteristics of Sediments

Physical and physicochemical factors, such as particle size distribution, pH, organic content and salinity, affect the accumulation of heavy metals. The mean pH of sediments ranged from 5.16 to 7.19, and they were acidic in most mangrove wetlands (Table 3). Similar sediment pH values were reported in other mangrove wetlands around southeast China [4, 20, 21]. The acidic pH was partly due to active microbial decomposition of mangrove litter and hydrolysis of

Table 1 Average concentrations of heavy metals in surface sediments (mg/kg, dry weight).

Area	Cu	Pb	Zn	Cr	Cd
Sanya (8) ^a	12.44 ± 6.06	42.27 ± 34.22	52.76 ± 46.53	9.21 ± 6.95	0.09 ± 0.16
Dongzhaigang (14)	12.57 ± 3.87	24.37 ± 13.8	78.27 ± 51.75	17.79 ± 15.18	0.45 ± 0.35
Daguansha (13)	9.80 ± 3.18	29.17 ± 12.68	75.14 ± 34.67	14.42 ± 5.83	0.19 ± 0.23
Beilun estuary (4)	5.69 ± 0.55	23.55 ± 1.06	49.28 ± 10.91	12.15 ± 3.89	0.17 ± 0.14
Qinzhouwan (3)	17.08 ± 7.13	46.74 ± 16.57	65.08 ± 29.47	13.41 ± 8.35	0.07 ± 0.09
Shankou (6)	17.64 ± 4.32	81.79 ± 18.33	75.75 ± 20.56	15.16 ± 5.63	0.08 ± 0.14
Zhanjiang (5)	13.88 ± 5.78	64.42 ± 20.83	34.82 ± 13.39	19.93 ± 5.36	0.05 ± 0.06
Taishan (3)	30.94 ± 9.01	67.67 ± 10.13	79.93 ± 3.59	19.87 ± 8.96	0.12 ± 0.16
Shenzhen (5)	69.80 ± 17.56	133.34 ± 10.58	193.72 ± 71.81	30.28 ± 10.87	0.98 ± 1.41
Yunxiao (10)	18.06 ± 3.40	30.81 ± 15.96	84.18 ± 25.05	3.63 ± 2.27	0.10 ± 0.03
Fugong (6)	28.96 ± 4.11	180.12 ± 34.16	148.63 ± 22.88	29.92 ± 9.44	0.13 ± 0.28
Quanzhou (3)	34.00 ± 17.44	167.21 ± 89.79	106.77 ± 59.01	18.64 ± 18.48	0.06 ± 0.05
E bay (7)	21.48 ± 4.49	77.01 ± 12.99	80.15 ± 29.31	21.84 ± 12.75	0.26 ± 0.69
Yaojiayu (7)	22.79 ± 10.37	51.98 ± 19.10	82.01 ± 33.32	20.32 ± 10.72	0.12 ± 0.13
Average	22.80 ± 16.46	76.1 ± 52.33	86.3 ± 42.65	18.4 ± 5.83	0.21 ± 0.25
Background value ^b	15.0	20.0	65.0	60.0	0.065

^a a Number in parentheses is sample amount;

^b Background Value for sediments of the China shelf sea from Zhao et al. [18].

Table 2 Correlation matrix of heavy metals in sediments.

	Cr	Cu	Zn	Cd	Pb
Cr	1				
Cu	0.577**	1			
Zn	0.450**	0.533**	1		
Cd	0.416**	0.295*	0.570**	1	
Pb	0.151	0.595**	0.418**	0.096	1

** Correlation is significant at the 0.01 level (2-tailed);

* Correlation is significant at the 0.05 level (2-tailed).

Table 3 Physicochemical characteristics of sediments (% dw).

	Fe	Mn (mg/g, dw)	pH	TOM	Salinity	Sand	Silt	Clay
Sanya (8)	1.91	155.11	5.8	3.04	7.82	70.36	28.83	0.81
Dongzhaigang (14)	2.18	157.17	5.61	4.38	14.99	55.64	42.76	1.59
Daguansha (13)	1.03	114.50	5.64	1.86	6.08	87.96	12.04	0
Beilun estuary (4)	5.19	722.47	5.06	1.24	7.16	64.69	34.09	1.23
Qinzhouwan (3)	1.46	158.45	6.1	2.61	6.72	32.86	65.23	1.93
Shankou (6)	3.09	201.94	6.5	2.59	15.87	33.64	64.38	1.97
Zhanjiang (5)	3.96	329.02	5.79	4.49	16.98	22.13	75.34	2.53
Taishan (3)	6.28	1477.68	6.73	2.42	20.10	20.69	75.17	4.15
Shenzhen (5)	4.00	463.35	6.58	3.75	19.07	20.96	60.9	3.09
Yunxiao (10)	-	-	6.34	11.08	21.10	-	-	-
Fugong (6)	3.91	727.11	6.82	2.67	6.27	5.75	90.75	3.5
Quanzhou (3)	3.98	2017.31	7.19	1.5	8.28	4.57	91.45	3.98
E bay (7)	2.82	395.77	6.74	2.8	15.68	6.57	91.45	4.41
Yaojiayu (7)	3.78	554.75	6.37	2.15	11.57	19.65	76.76	3.59
Average	3.35 ± 1.48	574.9 ± 571.6	6.23 ± 0.61	2.73 ± 0.99	12.69 ± 5.58	34.27 ± 27.01	62.24 ± 25.64	2.52 ± 1.39

* “-” undetermined.

tannin in mangrove plants, and partly resulted from oxidation of FeS_2 and FeS to H_2SO_4 [21].

The particle size distribution of sediments changed greatly. Percentage of sand ranged from 4.57% to 87.96%, with a mean value of 34.27%, and percentage of silt from 12.04% to 91.45%, with a mean value of 62.24%, and percentage of clay from 0% to 4.41%, with a mean value of 2.52%. Sediments from Daguangsha had the highest percentage of sand (87.96%), followed by Sanya (70.36%), Beilun estuary (64.69%) and Dongzhaigang (55.64%). The percentages of sand were relatively lower (< 7%) in mangrove sediments from Quanzhou, Fugong and Ewan.

Concentrations of TOM, Fe, Mn and salinity in sediments also fluctuated in the 14 mangrove wetlands (Table 3). The concentration of Fe ranged from 1.03% to 6.28%, with a mean value of 3.35%, and the concentration of Mn ranged from 114.50 $\mu\text{g/g}$ to 2,017.31 $\mu\text{g/g}$, with a mean value of 574.97 $\mu\text{g/g}$. TOM was much higher in mangrove sediments from Yunxiao. Receiving sewage, aquaculture wastewater and solid wastes, Yunxiao mangrove wetland was strongly affected by human activities.

4. Discussion

Mangroves with a area of not less 1 hm^2 is protected by law in China, but none of the above measures refers to the discharge of wastewater and solid waste. Parts of mangrove wetland in Shenzhen are used to dispose of sewage [22], which lead to rising of contents of heavy metals in mangrove sediments [23]. In most areas, mangroves are embraced with lots of aquiculture ponds. Furthermore, waterway, port and highway are always close to mangroves, such as in Fugong, Yaojiayu, Ewan, Quanzhou, Taisan and Shenzhen.

Mangrove sediments are reservoirs of heavy metals which are affected by factors such as the distance to city, local economic level, terrestrial pollutants and geomorphic properties. The close proximity to residential areas and industry zones usually causes

higher pollution levels. For example, mangrove wetlands in Shenzhen, Fugong and Quanzhou are near big cities. Sewage and industrial wastewater are discharged into the mangroves, which causes serious heavy metal pollution. Economic level is another mark of pollution. Heavy metal pollution is usually more serious in developed districts. With rapid development of the electronics industry, a large number of outdated electronic products have been discarded into the environment, and these “e-wastes” have caused serious heavy metal pollution in mangrove wetlands in Shenzhen and Quanzhou. In addition, terrestrial pollutant is an important contamination source of mangrove wetlands. Besides these factors, different geomorphic and hydrological properties influence the enrichment of heavy metal in sediments. For example, Daguangsha belongs to the open seacoast and has a flat landform, so, it is more difficult to retain heavy metals.

Levels of heavy metals are substantially higher than past (Table 4). Except Cu, concentration of Pb, Zn, Cr and Cd increased dramatically in Dongzhaigang, Shankou, Shenzhen and Fugong.

Local anthropogenic metal input affected contents of heavy metals significantly. The highest contents of Cu, Zn, Cr and Cd were found in Shenzhen mangrove sediment. As the most flourishing city, Shenzhen has turned into one important industrial center of China from a small fishing village during 30 years. Much solid wastes and wastewaters were input into Shenzhen gulf where mangrove occurs. However, concentrations of one or several kinds of heavy metal were higher in other mangrove reserves. Fugong mangrove wetland is near a busy sea-route, and a highway runs through Quanzhou mangrove wetland, which may account for high levels of Pb in sediments from Fugong and Quanzhou.

The concentrations of heavy metals in the sediment depend not only on natural process and anthropogenic discharging, but also many factors, such as TOM, pH, particle size distribution, salinity and sedimentary

Table 4 Comparison of heavy metal contents of surface sediment with past(mg/kg) [10-14, 24].

Area	Cu	Pb	Zn	Cr	Cd
Dongzhaigang	5.04	10.36	20.06	-	0.06
	12.57	24.37	78.27	17.79	0.45
Shankou	18.9	10.0	47.0	9.27	0.08
	17.64	81.79	75.75	15.16	0.08
Shenzhen	38.3	28.7	114.0	7.79	0.14
	69.80	133.34	193.72	30.28	0.98
Fugong	29.7	18.3	111.0	4.73	0.01
	28.96	180.12	148.63	29.92	0.13

Table 5 Correlation coefficient between concentrations of heavy metals and environmental factors.

	Cu	Pb	Zn	Cr	Cd
Fe	0.262*	0.313**	0.042	0.086	-0.005
Mn	0.257*	0.483**	0.067	0.010	-0.104
pH	0.325**	0.384**	0.219*	0.061	0.068
TOM	0.474**	0.224*	0.199	0.560**	0.172
Salinity	0.284*	0.084	0.023	-0.020	-0.028
Sand (%)	-0.432**	-0.590**	-0.268*	-0.259*	0.121
Silt (%)	0.411**	0.598**	0.184	0.188	-0.262*
Clay (%)	0.419**	0.436**	0.253*	0.286*	-0.139

** $p < 0.01$ Correlation is significant at the 0.01 level (2-tailed);

* $p < 0.05$ Correlation is significant at the 0.05 level (2-tailed).

environment. These factors control concentration of heavy metal in the mangrove surficial sediment. Being ligand and sorbent of heavy metals, organic matter plays an important role in accumulation of heavy metal in surface sediment.

Contents of heavy metals in sediments were affected strongly by the particle size distribution. Generally, fine particle with large surface area is easy to absorb heavy metals. Significant negative correlations were observed between concentrations of Cu, Pb, Zn, Cr and percentage of sand in sediment, but significant positive correlations were observed between concentrations of Cu, Pb, Zn, Cr and percentage of clay. Singh et al. [25] found that concentrations of heavy metals in the fine-grained fraction ($< 37 \mu\text{m}$) of sediment were 1-4 times higher than that in coarse-grained fraction (200-600 μm). Clay absorbs heavy metals strongly. Significant positive correlations were found between concentrations of Cu, Pb, Zn, Cr and percentage of clay in sediments, but Cd made an exception, and the reason was unclear.

Concentrations of heavy metals in mangrove sediments may be related to the mobilization of metals, as well as the mixing of fluvial and marine particulate material. The present study shows that the mangrove sediments are capable of accumulating heavy metals, while the amounts of heavy metals retain variedly with sediment properties.

Statistical analysis of the sediment compositional data revealed the heavy metals in the sediments were related to certain components such as TOM, pH and particle size distribution [26]. Correlations among concentrations of heavy metals and physical and physicochemical factors of sediments are listed in Table 6. Heavy metals such as Cu and Pb in the surficial sediment showed significant correlations ($p < 0.05$) with TOM and concentrations of Fe, Mn throughout the study areas (Table 5). There were significant correlations between TOM and concentrations of Cu, Pb and Cr. Significant correlations were found between pH and concentrations of Cu, Pb and Zn. There were significant correlations

between concentrations of Cu and salinity.

5. Conclusion

In the mangrove sediments of China, mean heavy metal concentrations were: $Zn > Pb > Cu > Cr > Cd$. The average concentrations of metals in mangrove sediments were: $Cu = 22.8 \pm 16.46$ mg/kg, $Pb = 76.1 \pm 52.33$ mg/kg, $Zn = 86.3 \pm 42.65$ mg/kg, $Cr = 18.4 \pm 5.83$ mg/kg and $Cd = 0.21 \pm 0.25$ mg/kg.

Physical and physicochemical factors of sediments varied greatly in study areas, and influenced concentrations of heavy metals significantly. TOM and particle size distribution of sediments were important factors to concentrations of heavy metals. There was significant correlation between concentrations of Cu, Pb, Cr and TOM. Significant correlations were found between concentrations of Cu, Pb, Zn and pH. Significant negative correlations were observed between concentrations of Cu, Pb, Zn, Cr and percentage of sand, but significant positive correlations were observed between concentrations of Cu, Pb, Zn, Cr and percentage of clay and silt. There were significant correlations between concentrations of Cu, Pb and Fe, Mn contents.

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