

# Article Seasonal Variation and Ecological Risk Assessment of Heavy Metal in an Estuarine Mangrove Wetland

Hualong Hong <sup>1</sup>, Binghuang Zhang <sup>1</sup> and Haoliang Lu <sup>1,2,\*</sup>

- Key Laboratory of the Ministry of Education for Coastal and Wetland Ecosystems, Xiamen University, Xiamen 361102, China; honghl@xmu.edu.cn (H.H.); 33120191151719@stu.xmu.edu.cn (B.Z.)
- <sup>2</sup> Fujian Key Laboratory of Coastal Pollution Prevention and Control, College of the Environment and Ecology, Xiamen University, Xiamen 361102, China
- \* Correspondence: luhl@xmu.edu.cn; Tel.: +86-592-288-0230

**Abstract:** Potential toxic metal pollution in mangroves has attracted extensive attention globally; however, the seasonal variation of potential toxic metals in mangrove wetlands is still poorly understood. Herein, we investigated the variation of content as well as chemical speciation of typical metals (Pb, Cr, Zn and Cu) in the sediments from the Zhangjiang Estuary mangrove wetland, China. The potential risk of metal contamination was also investigated. Compared to the wet season, we found that sediment metal content was higher in the dry season. Mangrove sites show accumulated significant metals than does the mudflat both in wet and dry seasons. Geo-accumulation (Igeo) shows moderate pollution, probably because of the dilution as result of runoff and tidal hydrodynamics in the wet season. Increased concentrations of all metals in the acid-soluble fraction and decreased metal contents in the residue fraction were found in the dry season. Risk assessment indicated that the concentrations of Pb poses a higher environmental risk in the dry season. These results can increase awareness of metal pollution in the dry season and provide information for potential toxic metal management in mangrove wetlands.

Keywords: chemical speciation; trace metal; mangrove sediment; coastal management

# 1. Introduction

Mangroves are widely distributed in more than 100 countries in the tropical and subtropical regions of the world [1]. Mangrove forests are well known for their role in the protection of coastal ecosystems as well as improving water qualities [2]. However, because of increasing anthropogenic activities, mangrove ecosystems are suffering from a variety of contaminants [3]. For instance, mangrove sediments could serve as a reservoir of potential toxic metals, buffering their release and bioavailability in the land–sea interface [4]. The content of potential toxic metals in mangrove sediments is concerning. Potential toxic metals in mangroves could form complexes with particulate organic carbon, Fe-Mn hydroxides and sulphides [4–6]. However, when the wetlands are flooded by the tide, potential toxic metals can be released from sediments into pore-waters by dissolution of bearing minerals under anoxic conditions [7,8]. Thus, dissolved toxic metals can be exported to an adjacent marine environment and be transferred to the food chain in mangroves' ecosystems. Sequential extractions provide useful tools for the operational assessment of metals in different geochemical fractions, and have an excellent performance in revealing the influence of sediment characteristics in mangrove wetlands [9,10].

The increasing runoff and soil leaching during the monsoon season was reported to elevate the potential toxic metal input to the estuary [11]. Besides, hydrological conditions and input of terrestrial components are thought to influence the distribution, chemical fraction, bioavailability and fate of potential toxic metals in coastal mangroves [12]. However, there could be also a dilution effect by the increasing surface runoff and groundwater exchange in mangroves that were once polluted but are now under good management.



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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Seasonal variation of metals in such mangrove ecosystems as well as the risk of these metals on the sediment quality are still lacking. Thus, the aims of this research were: (1) to evaluate the distribution and geochemical fraction of potential toxic metals (Pb, Cr, Cu and Zn) in the mangrove sediment of the Zhangjiang Estuary with emphasis on seasonal variation; and (2) to assess the potential ecological risks of these metals in view of the dilution effect.

# 2. Materials and Methods

## 2.1. Site setting and Sediment Collection

Sediments were collected from representative stations in Yunxiao Mangrove National Nature Reserve (23°55′51.31″ N, 117°24′50.57″ E) located in the Zhangjiang Estuary, Fujian Provence, China, with a total area of 2360 hm<sup>2</sup> (Figure 1). A subtropical monsoon climate dominates Yunxiao with an annual precipitation of 1715 mm and annual temperature of 21.2 °C. Precipitation during spring and summer months accounts for about 80% of the annual rainfall. The flood season is from April to September. The particulate matter content of the water body ranged from 0.25–0.36 g L<sup>-1</sup>. The estuary is semi-diurnal with a large tidal difference (0.43 to 4.67 m). The sampling site is habitat for indigenous mangrove species *Aegiceras corniculatum* (Ac), *Kandelia obvata* (Ko), *Avicennia marina* (Am) and is invaded by salt marsh species *Spartina alterniflora* (Sa). Sampling sites were selected in these habitats to represent the dominant species in Zhangjiang Estuary. Sediment samples were collected in the rhizosphere and the bulk soil of the above plant communities as well as in the mudflat (Mf) in June and December of 2017 to represent the wet and dry seasons (Figure 1).



**Figure 1.** Map showing the sampling sites in the current study. Z J represents Zhangjiang Estuary, Am represents *A. marina* community, Ko represents *K. obovate* community, Ac represents *A. corniculatum* community, Sa represents *S. alterniflora* community and Mf1–Mf4 represent the corresponding mudflats of each plant community.

At each sampling site, three surface sediment samples were collected at the depth of 0–20 cm in order to avoid disturbance of fiddler crabs and plant roots in mangroves. All the samples were sealed in pre-washed plastic bags, stored at 4  $^{\circ}$ C and sent back to

the laboratory at soon as possible. The sediments were stored at -20 °C prior to the following analysis.

## 2.2. Heavy Metal Chemical Speciation Analysis

The total potential toxic metal contents were determined by microwave digestion of sediment samples in a mixture of 9 mL of nitric acid (HNO<sub>3</sub>), 3 mL of hydrofluoric acid (HF) and 1 mL of hydrochloric acid (HCl). The modified version of the BCR three step sequential extraction procedure was used according to Rauret et al. [13]. Briefly, the various single extraction methods are as follows: after the soil sample is dried to a constant weight at 105  $^{\circ}$ C, 1 g of the soil sample is placed in a 50 mL centrifuge tube. Each fraction is extracted sequentially through different forms of association and available mechanisms. The extraction of exchangeable components and soluble components (Exc) was carried out in this way. Extract 1 g of soil sample with 40 mL 0.11 mol  $L^{-1}$  acetic acid solution for 16 h. For the reducible component (Red): add 40 mL of 0.5 mol L<sup>-1</sup> hydroxyammonium chloride solution to the residue of the first step, and shake the mixture for 16 h. For the oxidizable component (Oxi): add 10 mL of 8.8 mol  $L^{-1}$  hydrogen peroxide solution to the residue of the second step. The residue was extracted with 50 mL of 1 mol  $L^{-1}$  ammonium acetate. For the residue fraction (Res): digest the residue from the third step with aqua regia. All metals were analyzed by inductively coupled plasma mass spectrometry (ICP-MS 7700X, Agilent, Santa Clara, CA, USA). All reagents used in this study were reagent grade or higher. All laboratory containers were soaked in 10% HNO<sub>3</sub> for more than 48 h, then rinsed with MQ water. Quality control was carried out by analysis repeats, method blanks and Chinese national standard materials (offshore marine sediments, GBW07314). Ouality control includes blank analysis procedures and certified materials for every 10 samples. Rh and In were used as internal standards. The average recovery rate of investigated metals (Cr, Cu, Pb, and Zn) is 90% to 110%.

#### 2.3. Risk Assessment

Geoaccumulation Index (I<sub>geo</sub>) was applied to assess metal contamination in mangrove sediments. It was a comparison between observed metal contents and unpolluted baseline [14].

$$I_{\text{geo}} = \log_2(C_n/1.5 \times B_n), \tag{1}$$

where  $I_{geo}$  was used to represent geoaccumulation index,  $C_n$  was metal content in sample, 1.5 acted as a correction threshold,  $B_n$  was the background content of the specific metal in baseline.  $I_{geo}$  could be categorized between 0 and 6, as shown in Table 1. The background values were selected according to previous mangrove studies in China (i.e., Cr: 38.7 mg kg<sup>-1</sup>, Zn: 57.2 mg kg<sup>-1</sup>, Cu: 13.7 mg kg<sup>-1</sup>, Pb: 24.1 mg kg<sup>-1</sup>) [15].

Table 1. Grading criteria for index of geoaccumulation.

Igeo	Class	Pollution Category
$\leq 0$	0	Uncontaminated/unpolluted
0–1	1	Unpolluted/moderately
1.00-2.00	2	moderately
2.00-3.00	3	moderately/heavily
3.00-4.00	4	heavily
4.00–5	5	heavily/extremely
$\geq 5$	6	extremely

Potential Ecological Risk was used to evaluate the potential toxic metal risk and the environmental effect of metal accumulation in the sediment (Table 2) [16].

$$C_f^i = C_s^i / C_n^i \tag{2}$$

$$E_i^r = T_r^i \times C_f^i \tag{3}$$

$$PERI = \sum E_i^r \tag{4}$$

where  $C_f^i$  was used to represent each element contamination factor,  $C_s^i$  was the content of element in samples,  $C_n^i$  was the value of the element,  $E_i^r$  was the potential ecological risk index of a single element and  $T_r^i$  was the biological toxicity of a single element.

Table 2. Grading criteria for potential ecological risk index.

Er	Monomial Potential Ecological Risk Factor	RI	Risk
$\leq 40$	Low	$\leq 150$	Low
40-79	Moderate	150-299	Moderate
80-159	Moderate to high	300-600	High
160-320	High	600	Very High
$\geq$ 320	Very high		

### 2.4. Statistical Data Analyses

Statistical analyses were performed using SPSS (16.0) software. All data were showed as the mean and standard deviation of three replications. Analysis of variance (ANOVA) followed by the Duncan-type post hoc test was employed to test the quantitative significance among plant type and season.

## 3. Results

### 3.1. Season Variation of Potential Toxic Metals in Zhangjiang Mangrove

The total amount of potential toxic metals in the sediment of the Zhangjiang Estuary mangrove wetland is shown in Figure 2. In wet season, the average potential toxic metal contents of the sediment showed the trend as: Zn (111.3 mg kg<sup>-1</sup>) > Pb (71.9 mg kg<sup>-1</sup>) > Cr (64.4 mg kg<sup>-1</sup>) > Cu (32.7 mg kg<sup>-1</sup>). The concentration of potential toxic metals in the sediment in the plant community, whether from rhizosphere or bulk sediment, was lower than that in the mudflat. The content of potential toxic metals in the surface sediments in the dry season, with the following trend Zn (183.0 mg kg<sup>-1</sup>) > Pb (97.5 mg kg<sup>-1</sup>) > Cr (88.9 mg kg<sup>-1</sup>) > Cu (30.2 mg kg<sup>-1</sup>), was found to be enriched compared to that in the wet season. In dry season, the accumulation of metals was observed to be significantly higher than that in mudflat only in the mangrove A. marina community. In general, the heavy metal content fluctuated more in the wet season than in the dry season. Cr, Pb and Zn contents were significantly higher (p < 0.01) in the dry season than in the wet season. Cr, Cu, Pb and Zn content were not significantly different in the rhizosphere and bulk soil in wet season. We found that there were also no significant differences in heavy metal contents between bulk soil and mud flat in most of the sampling sites during wet season. However, in the dry season, the heavy metals in bulk soil were significantly higher (p < 0.01) than those in the mudflats both in *Avicennia marina* and *Spartina alterniflora* sample sites. In the wet season, Cr, Cu and Pb were not significantly higher than the mudflats in the Spartina alterniflora dominated sites. However, in the dry season, the rhizosphere heavy metal content of *Spartina alterniflora* site was significantly greater than that of the mudflats. In the wet season, the four metals were significantly enriched in the mangrove Aegiceras corniculatum site, but in the dry season, there was no significant difference between inside and outside the forest.



**Figure 2.** Changes of total potential toxic metals in sediments. Different letters above the error bars (a, ab, b, bc, c) represent significant differences (p < 0.05) in potential toxic metal contents in rhizosphere, bulk soil and mud flat at the same sampling site.

## 3.2. Speciation of Potential Toxic Metals in Sediments

As shown in Figure 3, most fractionation of Pb, Cr, Cu and Zn were bound to the residual fraction in the Zhangjiang Estuary mangrove wetland. The exchangeable fraction of metals was higher in dry season. The proportion of Oxi fraction of heavy metals in the rhizosphere was significantly higher compared to bulk soil. In addition, the oxidizable fraction of Pb in rhizosphere sediment was increased compared to bulk sediment. The Cr and Pb partitioning were characterized by Res > Red > Oxi > Exc in the mudflat and the mangrove site both in dry and wet season. The Zn partitioning were Res > Red > Oxi > Exc, without any difference in sampling sites. The Cu partitioning differed between sites: Res > Exc > Oxi > Red in the Avicennia site, Res > Red > Oxi > Exc in the *S. alterniflora* site (Figure 3).



Figure 3. Speciation of potential toxic metals in sediments.

## 3.3. Risk Assessment of Potential Toxic Metal Pollution in Sediments

The potential toxic metal I<sub>geo</sub> for mangrove sediments showed the following trend: Zn > Cu > Pb > Cr (Table 3). In wet season, the I<sub>geo</sub> values suggested that sediments were not to moderately contaminated by Cr, Cu and Pb ( $0 < I_{geo} < 1$  in Table 3) and moderately contaminated by Zn (I<sub>geo</sub> = 2.16) in the *S. alterniflora* site. In dry season, overall, Cr, Cu and Zn showed no pollution in the Zhangjiang Estuary mangrove (I<sub>geo</sub> < 1 in Table 3). However, I<sub>geo</sub> values of Pb in most research areas were near or exceeding 1, indicating the sediment was moderately contaminated by Pb. The geo-accumulation index indicated that plant communities increased metal accumulation compared to mudflat. The potential ecological risks of investigated metals in sediments were assessed employing the biological toxicity factor  $T_r^i$ . The risk index showed that the potential ecological risks of heavy metals in wet season were less than that in dry season. For Pb, Cr, Cu and Zn,  $E_i^r < 40$  indicated low potential risk in both wet and dry season (Table 4).

Season	Туре	Am	Ко	Ac	Sa	Mf
Wet season	Cr	0.05	0.13	0.04	0.02	0.03
	Cu	0.29	0.3	0.03	0.35	0.48
	Zn	0.04	0.02	0.13	2.16	0.84
	Pb	0.57	0.6	0.57	0.24	0.2
Dry season	Cr	0.57	0.63	0.5	0.5	0.28
	Cu	0.08	0.05	0.1	0.14	0.36
	Zn	0.6	0.69	0.58	0.57	0.31
	Pb	0.95	1	0.91	0.89	0.57

Table 3. Geoaccumulation index of Pb, Cr, Cu and Zn in the wet and dry seasons.

Season	Туре	Am	Ко	Ac	Sa	Mf
Wet season	Cr	3.1	3.27	3.08	2.95	3.05
	Cu	9.2	9.2	7.34	5.9	5.36
	Zn	1.55	1.48	1.64	0.33	0.84
	Pb	11.12	11.36	11.14	8.83	8.62
	RI	24.97	25.31	23.21	18.01	17.87
Dry season	Cr	4.44	4.44	4.25	4.25	3.65
	Cu	7.1	7.23	7	6.81	5.84
	Zn	2.28	2.29	2.25	2.22	1.86
	Pb	14.46	15.06	14.07	13.93	11.14
	RI	28.28	29.02	27.57	27.22	22.49

Table 4. Potential ecological risk index of Pb, Cr, Cu and Zn in wet and dry seasons.

#### 4. Discussion

Potential toxic metal contents (Pb, Cr, Cu, and Zn) in the Zhangjiang Estuary mangrove sediments (Figure 2) both in wet and dry season are lower than of mangroves exposed to intense anthropogenic activities in Shenzhen (China) [17], Divar Island and South Gujarat Coast (India) [9,18]. Potential toxic metals in the mudflat were lower than those reported in the offshore mangrove sediments of Qinzhou [19] and Sundarban [20]. This indicates that metals in the Zhangjiang Estuary mangrove wetland were primarily derived from the physicochemical weathering processes of basaltic rocks in the Zhangjiang River basin. In addition, metals with dominant proportion in residue fraction (Figure 3) maybe support the natural origins of metals in this mangrove.

Sequential extraction results suggested that the residual fraction prevailed in the distribution of Pb, Cr, Cu and Zn in the Zhangjiang estuary mangrove sediments. In dry season, geochemical speciation data suggest a high amount of Cr and Pb in the exchangeable fraction, which could induce an adverse impact on aquatic lifeforms (Figure 3). A high percentage of heavy metals was associated with the residual fraction, indicating that the heavy metals had natural origins. Changes in heavy metal patterns in the rhizosphere region may be due to oxygen secretion by wetland plant roots [8]. The mangrove plant *A. marina* has a large number of respiratory roots and has a greater ability to intercept sediment than other plants, which may be responsible for the higher heavy metal content and risk in the forest floor than in other sampling sites (Figure 2). The non-residual fractions of Pb, Cr, Cu and Zn were much lower than their residual fractions. Thus, the potential toxic metals in the sediment of the Zhangjiang mangrove were greatly immobilized and their potential toxicities were reduced [5,11,21]. In wet season, the non-residual fractions of potential toxic metals reported in the current study were much lower than the observations from the contaminated mangrove wetlands [11,18,22], suggesting the potential toxic metals in the study area are at a relatively low risk to coastal lifeforms. However, due to poor seawater exchange capacity, the increasing biological toxicity of Pb and Cr in the exchangeable speciation should need to have attention paid in dry season.

It is important to determine whether the potential toxic metals in sediments pose a stress to the coastal lifeforms [11,23]; therefore, the potential toxic metal pollution and potential ecological risks were applied to assess possible risk arising from the potential toxic metal contamination in sediments of the Zhangjiang Estuary mangrove wetland. Both in the wet and dry seasons,  $E_i^r < 40$  indicated there were low potential risks throughout the mangrove habitats. It need to be noted that, the presenting research involved four periority metals because of their well-known association with human activity near coastal wetlands. Benthic exchange fluxes from anthropogenic sources, such as agriculture drainage and shrimp farming, might increase contamination levels in the dry season [24,25]. Some other metals, like Cd, Hg, Tl, were also toxic to the lifeforms habitating the coastal to offshore ecosystem. In this case, Hakanson's RI slightly increased to low-to-moderate risk after a correction based on the sum of  $T_r^i$  considering the metal inflation effect, which still calls for the further management of mangrove wetlands. The Igeo values showed that sediments in the Zhangjiang mangrove were non to moderately contaminated especially for Pb in the dry season. Our finding was different from the metal contamination pattern in Southeast Asia mangrove wetland, where enhanced water exchange during the monsoon season leads to elevated metal inputs to the estuary [9,26]. Our results indicated that water dilution was also critical, especially for mangroves under good protection. Their potential ecological impacts need to be considered for mangrove wetland management.

#### 5. Conclusions

Seasonal variation of content, speciation and ecological risks of potential toxic metals in the sediments from the Zhangjiang Estuary mangrove wetland were revealed. Mangrove sediments did not present high metals concentrations. Low ecological risks were observed for the investigated potential toxic metals at most sites in the dry and wet periods, suggesting that these metals would not be associated with biological toxic effects. The geoaccumulation Index, Igeo, indicated that Pb was the potential toxic metal that requires priority attention in the Zhangjiang Estuary mangrove. Hydrological conditions, such as the intensity and duration of floods and the groundwater level, will affect the speciation, migration and transformation of metals in the sediments of estuarine mangrove wetlands. In terms of coastal ecosystem restoration and management, further work is required on detailed investigation of the origins and variations of potential toxic metal in different seasons.

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