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Occurrence and transfer of heavy metals in sediments and plants of *Aegiceras corniculatum* community in the Qinzhou Bay, southwestern China

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Abstract
Mangrove wetlands can reduce heavy metal pollution by trapping heavy metals. In this study, the concentration, transport and bioaccumulation of Cr, Cd, Cu, Zn and Pb in the sediments and different parts of *Aegiceras corniculatum* at four different sites in the Qinzhou Bay in southwestern China were investigated. The results showed that although the potential ecological risk of all five heavy metals was slight, the concentration of Cr was at a moderate pollution level due to the emissions of industries and aquaculture waste water. Core sediment records indicated that the concentrations of heavy metals at the depth of 0–20 cm were relatively high, showing an increasing trend of heavy metals over the past 20–30 years. Cr, Cu, Pb and Cd accumulated mainly in the roots of *A. corniculatum*, while Zn accumulated mainly in the stems. *Aegiceras corniculatum* showed the strongest transport capacity for Zn and Cu and the strongest bioaccumulation ability for Cd. Compared with other mangrove communities, *A. corniculatum* can be chosen as a restoration species in tropical and subtropical coastal zones polluted by Zn, Cu and Cd.

Key words: *Aegiceras corniculatum*, sediment, heavy metals, accumulation

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1 Introduction

Mangroves are woody plant communities in the intertidal zone of tropical and subtropical land-sea intersections. Mangroves play multiple roles in coastal ecosystems, such as protection against storm surges, promoting siltation and land reclamation, embankment protection, offering habitats for birds, fish and other species, and improving the ecological environment of the bay (Lau, 2000; MacFarlane et al., 2007; Nagelkerken et al., 2008; Wang et al., 2019). Due to the special and open habitats, developed roots, and the rich organic matter in sediments, mangroves receive a variety of contaminants from different channels and can act as important sources and sinks of pollutants in coastal and estuarine environments (Tam and Wong, 1995, 2000; MacFarlane and Burchett, 1999, 2002; Ray et al., 2006).

In mangrove ecosystems, due to the unique physiochemical conditions of the sediment and root structure of mangrove trees, heavy metals, including Cu, Zn, Cd, Cr and Pb, usually accumulate to considerable levels and may accumulate via the food chain to hazardous levels due to toxicity and persistence (Pan and Wang, 2012; Zhang et al., 2014b).

In mangrove wetlands, sediments are the main storage media of heavy metals, with small amounts absorbed by plants (Lau, 2000; Machado et al., 2002). Among different parts of mangrove plants, heavy metals mainly accumulate in the roots, and only a small fraction is transported to leaves (Cheng et al., 2010). Mangrove plants can excrete excessive non-essential elements, such as Pb, and control the concentrations of basic elements *in vivo*, such as Cu and Zn (MacFarlane et al., 2007; Yan and Tam, 2013). Mangrove wetlands may mitigate various pollutants through a series of physical, chemical, and biological effects, resulting in purification through absorption, accumulation and translocation (Tam and Wong, 1995, 1997; Tam et al., 2009).

It is worth noting that when the concentrations of heavy metals are above the upper tolerance limit of mangrove species, the toxic effects may lead to plant death (MacFarlane and Burchett, 2002; Qin et al., 2007). In cultivation experiments on *Avicennia marina*, MacFarlane and Burchett (2002) found that when the concentration of Cu in soils was 400 μg/g, the total biomass decreased and root growth was inhibited, while the levels of overall growth were suppressed when the concentration of Cu

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reached 800 μg/g. Liu et al. (2009) studied the tolerance of mangroves to mixed heavy metals and found that the heavy metals could inhibit the growth of mangrove seedlings. In cultivation experiments, Cheng et al. (2010) found that with an increase in Zn, only the seedling height and total biomass of *Bruguiera gymnorrhiza* decreased, but the seedling height, the number of stems and leaves and the total biomass decreased in *Rhizophora stylosa* and *Aegiceras corniculatum*. According to previous studies, a mangrove ecosystem is capable of accumulating toxic metals, thus reducing heavy metal pollution in adjacent estuarine and marine systems (Ramos e Silva et al., 2006; MacFarlane et al., 2007), and the translocation and bioaccumulation factors of heavy metals by mangroves showed a variety of trends (Analuddin et al., 2017). Thus, maintaining high diversity of mangroves is crucial to ensure the health and productivity of coastal zones. Therefore, it is necessary to investigate the distribution of heavy metals in mangrove soils and organs and to then choose appropriate species to prevent poor growth of restoration plants at sites polluted by heavy metals.

The Qinzhou Bay is an important international trade port in southwestern China, where mangroves are widely distributed along the coast. In recent decades, many heavy metals have been discharged into the bay with the rapid development of harbours, maritime industries, and tourism, which has resulted in the pollution of mangrove ecosystems (Zhang et al., 2010, 2014; Li et al., 2014; Li and Dai, 2015; Xu et al., 2017). Many studies (Zhang et al., 2010, 2014; Xu et al., 2017) have indicated that heavy metal pollution is a serious problem in the Qinzhou Bay, especially the Maowei Sea, which was established as the National Ocean Park of China in 2011. Previous studies focused on the distributions and ecological risks of heavy metals in marine sediments, but few studies have focused on mangrove sediments and the plant parts of mangrove species in the Qinzhou Bay. Therefore, the specific objectives of this study were to investigate the concentrations of Cu, Zn, Pb, Cd and Cr in the sediments of different soil layers and different parts of *A. corniculatum*, the most widespread species in the Qinzhou Bay. In addition, the potential ecological risks caused by the translocation and bioconcentration of heavy metals were evaluated to provide knowledge for the pollution control and rehabilitation of the mangrove coastal system in this area.

2 Materials and methods

2.1 *Study area*

The Qinzhou Bay is located at the top of the Beibu Gulf along the coast of Guangxi, China. It is a semi-enclosed natural estuary composed of an inner bay (Maowei Sea), neck bay, and outer bay. It has a subtropical monsoon climate with an average annual temperature between 21°C and 23°C and an annual precipitation of 1 500–1 800 mm. The tide is a regular diurnal tide with a range of –0.69–5.83 m and an average tidal height of 2.40 m. The major mangrove species in the Qinzhou Bay include *A. marina*, *A. corniculatum*, *Kandelia obovata*, and *R. stylosa*, with *A. corniculatum* being the most widely distributed (Yan and Liu, 2006; He et al., 2011).

2.2 *Sample collection*

Sampling was performed from July 25 to 28, 2014, at four sites: Kangxiling (KXL), Haixialou (HXL), Jianxinwei (JXW), and Qinzhougang (QZG) (Fig. 1). Leaves, stems, roots and fruits of *A. corniculatum* and surface sediments at depths of 0–10 cm were collected, and each sample was collected at least in triplicate. Core sediment samples from a depth of 0–100 cm, except at KXL (0–60 cm), were collected using a self-made polyvinyl chloride (PVC) corer and were divided into layers, i.e., 0–10 cm, 10–20 cm, 20–40 cm, 40–60 cm, 60–80 cm and 80–100 cm. All samples were sealed in polyethylene plastic bags and transported to the laboratory for analysis.

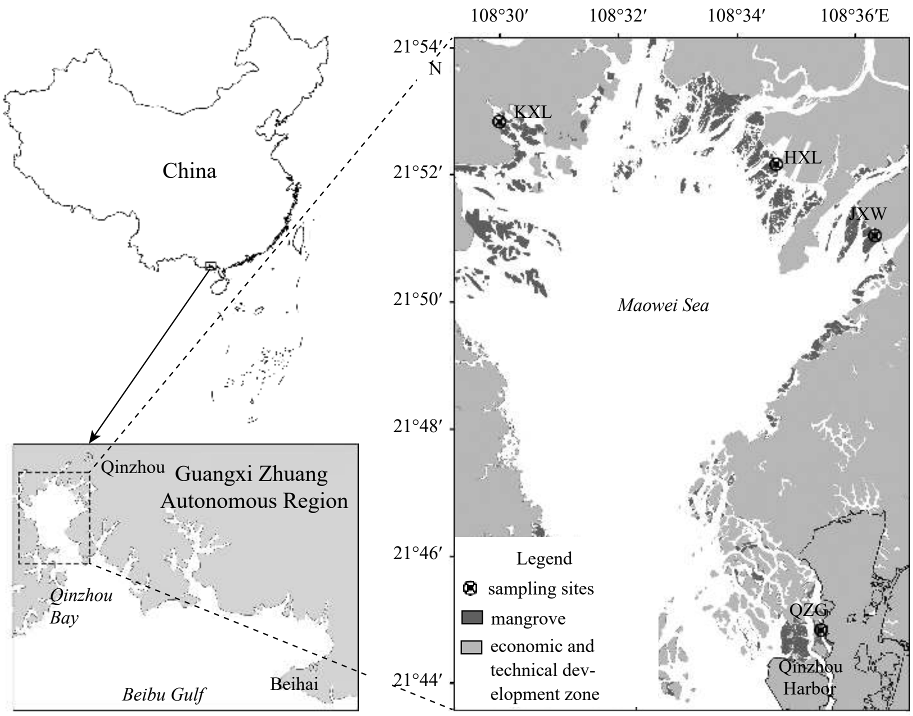

Fig. 1. Locations of sampling sites in the *A. corniculatum* forest in the Qinzhou Bay, China.

Table 1. Statistics of heavy metal concentrations (μg/g, dw) in sediments and mangrove parts in the mangrove areas of the Qinzhou Bay, China (values are the mean±SD)

	n	Cr	Cd	Cu	Zn	Pb
Surface sediments	12	113.66±13.42	0.28±0.13	34.95±9.90	108.69±35.75	37.90±6.00
Core sediments	4	68.65±20.85	0.14±0.09	19.16±8.67	72.68±21.2	20.32±6.71
Fruit	19	1.32±1.07	0.01±0.003	1.87±0.73	7.58±4.14	0.24±0.19
Root	16	32.12±18.67	0.17±0.10	5.17±1.37	27.51±5.37	10.75±4.01
Stem	18	2.43±1.96	0.1±0.06	3.29±0.53	30.43±6.71	8.16±4.42
Leaf	19	2.50±5.78	0.06±0.02	2.58±1.03	18.43±6.83	2.82±0.92
All plant parts	72	8.75±15.55	0.08±0.08	3.15±1.52	20.59±10.72	5.24±5.06

2.3 *Sample pretreatment and heavy metal determination*

Sediment samples and plant samples were dried at room temperature. After mixing, all the samples were sieved through a nylon sieve (140 mesh). First, a 0.1 g soil sample and a 0.5 g plant sample were weighed in a 25 mL Teflon vessel, and then 3 mL of nitric acid, 3 mL of hydrofluoric acid, and 1 mL of perchloric acid were added to the tube and allowed to stand until the samples were fully soaked. Then, these Teflon vessels were placed in a digestion high-pressure tank at 160°C for 9 h. After cooling to room temperature, the digestion tanks were placed on a hot plate to evaporate the acid, and the solutions were then transferred to tubes, diluted with 2% nitric acid solution to 50 mL, and analysed (Wang et al., 2008). Given the differences in *A. corniculatum* among sample plots, all of the samples of the plant parts were divided into sub-samples of 0.5 g after drying and mixing. The numbers of the sub-samples are shown in Table 1.

Cu, Zn, Pb, Cr and Cd were determined with flame atomic absorption spectrometry (AAS) (AAnalyst 800, USA); Cr, Cu and Zn were analysed with the flame atomic absorption method, while Cd and Pb were subjected to single-element analysis using the graphite furnace atomic absorption method.

2.4 *Quality assurance (QA) and quality control (QC)*

The quality control standard materials for the sediment samples and plant samples were GBW07603 (GSV-2) and GBW07406 (GSS-6), respectively. One blank sample was added to the experimental sequence every seven samples, and one sample was selected randomly to assess the reproducibility of the method. The recoveries of heavy metals in sediment samples and plant samples were 91.8%±10.4% and 95.4%±14.3%, respectively, with corresponding relative standard deviations of 2.75%–3.59% and 2.23%–4.01%, respectively.

2.5 *Data processing*

Pollution level and ecological risk assessments of heavy metals in the sediments were performed according to Hakanson (1980), who proposed the single factor index (E_r^i) and potential ecological risk index (*RI*). These calculations are as follows:

$$C_f^i = C_s^i / C_n^i, \tag{1}$$

$$C_d = \sum C_f^i, \tag{2}$$

$$E_r^i = T_r^i \times C_f^i, \tag{3}$$

$$RI = \sum E_r^i, \tag{4}$$

where C_f^i is the pollution index of heavy metal *i*; C_s^i is the measured concentration of heavy metal *i* in the sediment; C_n^i is the background value of heavy metal *i*; C_d is the comprehensive pollution index adopted from Wu et al. (2014). The background values of Cr, Cd, Cu, Zn and Pb were the concentrations of heavy metals in the sediments of pre-industrial times, which were 90, 1, 50, 175 and 170 μg/g, respectively (Hakanson, 1980). The toxic–response factor (T_r^i) for Cr, Cd, Cu, Zn and Pb was 2, 30, 5, 1 and 5, respectively (Hakanson, 1980). C_f^i, C_d and *RI* can be classified into four levels: low risk (C_f^i<1, C_d<5, *RI*<150), moderate risk (1≤C_f^i<3, 5≤C_d<10, 150≤*RI*<300), considerable risk (3≤C_f^i<6, 10≤C_d<20, 300≤*RI*<600), and very high risk (C_f^i≥6, C_d≥20, *RI*≥600). The degree of E_r^i is as follows: low risk (E_r^i<40), moderate risk (40≤E_r^i<80), considerable risk (80≤E_r^i<160), high risk (160≤E_r^i<320), and very high risk (E_r^i≥320).

The bioconcentration factor (BCF) and translocation factor (TF) of heavy metals in different parts of *A. corniculatum* were calculated with the formulas proposed by Agoramoorthy et al. (2008) and Usman et al. (2012, 2013) as follows:

$$BCF_{\text{parts}} = C_{\text{parts}} / C_{\text{sediment}}, \tag{5}$$

$$TF_{\text{leaf/stem/fruit}} = C_{\text{leaf/stem/fruit}} / C_{\text{root}}, \tag{6}$$

where C_{parts} are the metal concentrations in the leaf, stem, fruit and root; C_{sediment} is the metal concentration in the sediment; $C_{\text{leaf/stem/fruit}}$ represents the metal concentrations in the leaf, stem and fruit, respectively; and C_{root} is the metal concentration in the root.

2.6 *Statistical analyses*

The concentrations of heavy metals, BCF and TF in different sites and plant parts were subjected to one-way analysis of variance (ANOVA) with the least significant difference (LSD) test to understand the difference between each statistical indicator using SPSS 19.0 (SPSS Inc. USA). The significance was based on the *p*-value, which was less than 0.05.

3 Results

3.1 *Heavy metals in mangrove sediments*

The concentrations of Cr, Cd, Cu, Zn and Pb in the mangrove surface sediments of the Qinzhou Bay are shown in Table 1. Cr showed the highest concentration of (113.66±13.42) μg/g (mean±SD), followed by Zn ((108.69±35.75) μg/g), Pb ((37.69±6.00) μg/g), Cu ((34.95±9.90) μg/g), and Cd ((0.28±0.13) μg/g). Compared with the marine sediment quality (GB 18668—2002), Cr in all surface sediments was above level I (80 μg/g), while Cd and Pb were below level I (0.5 and 60 μg/g for Cd and Pb, respectively). Figure 2a illustrates the distribution of heavy metals at the four different sampling sites. The concentrations of Cd, Cu, Zn and Pb in the surface sediments of QZG were significantly higher than those at the three other sites. Moreover, the concentrations of Cu, Zn, Pb and Cd in the mangrove surface sediments of the Qinzhou Bay were at the medium level compared with other mangrove areas worldwide (Table 2).

According to the calculation results of C_f^i, the pollution index of Cr in the mangrove surface sediments of the Qinzhou Bay reached a moderate pollution level (1.002–1.58), but the pollution indexes of Cd, Cu, Zn and Pb were all less than 1, while the comprehensive pollution index (C_d) of these five heavy metals was less than 5. Potential ecological risk results showed that the individual potential ecological risk indexes (E_r^i) of Cr, Cu, Zn, Pb and Cd were less than 40, and the *RI* values were between 13.72 and 26.21, which are considerably lower than 150. The pollution indexes or the *RI* of Cr, Cu, Zn, Pb and Cd in mangrove surface sediments showed no significant difference among the four sampling sites of the Qinzhou Bay. In general, the concentrations of Cr in mangrove surface sediments in the Qinzhou Bay were relatively high and at the moderate pollution level, but the concentrations of Cu, Zn, Pb and Cd were low and were at the low pollution level, and the potential ecological risk results indicate that these five heavy metals have a slight ecological risk.

The concentrations of Cr, Cd, Cu, Zn and Pb in mangrove core sediments of the Qinzhou Bay are also shown in Table 1, and the distribution of those heavy metals at different depths is shown in Fig. 3. The concentrations of Cd, Cu, Zn, Pb and Cr generally decreased from the surface to the bottom of core sediments at QZG, KXL, JXW and HXL, and the concentrations of the five heavy metals in the 0–20 cm layer at QZG were the highest. Specifically, as shown in Fig. 3f, the mean concentrations of Cd, Cu and Pb in the core sediments of all sites decreased with core depth, although the concentrations of Cr (Fig. 3a) and Zn (Fig. 3d) at depths of 40–60 cm and 80–100 cm increased slightly.

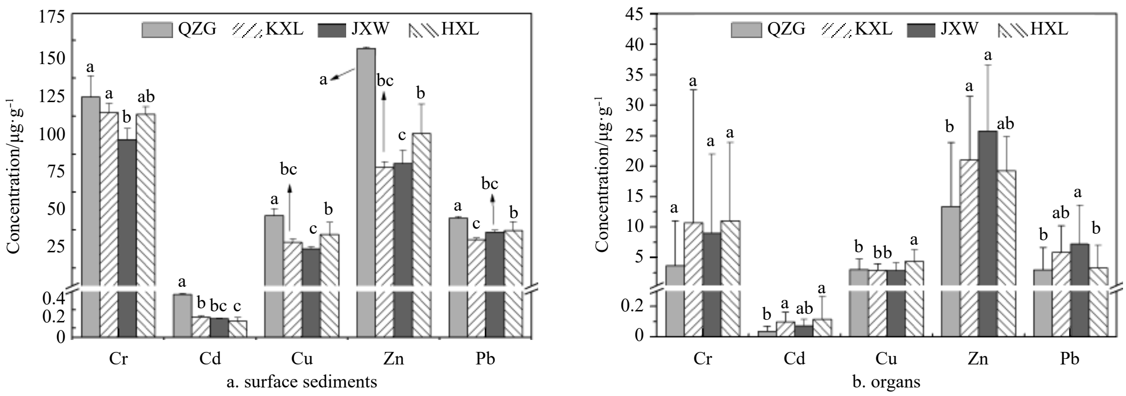

Fig. 2. Distributions of heavy metals in the mangrove surface sediments and different parts of *A. corniculatum* from four sites in the Qinzhou Bay, China. The letters a, b and c were used to indicate the ANOVA (p<0.05) results; and the different letters indicate significant differences.

Table 2. Concentrations (mean or range, μg/g, dw) of heavy metals in mangrove surface sediments of mangrove wetlands around the world

Location	Sampling year	Cu	Zn	Pb	Cd	Cr	References
Qinzhou Bay, China	2014	24.11–50.65	73.17–158.24	30.19–46.3	0.14–0.48	90.20–142.22	this study
Xiandao Park, Qinzhou Bay, China	2013	41.06	104.59	111.90	0.25	55.04	Li et al. (2014)
Qinzhou Bay, China	2006	17.08	65.08	46.74	0.07	13.41	Li et al. (2008)
Mai Po, Hong Kong, China	—	78.50	240.00	79.20	2.62	39.20	Tam and Wong (2000)
Dongzhai Harbor, Hainan, China	2008	18.00	57.00	19.00	0.11	40.00	Qiu et al. (2011)
Sanya Bay, Hainan, China	2008	9.00	53.00	18.00	0.13	12.00	Qiu et al. (2011)
Yalong Bay, Hainan, China	2008	5.00	26.00	15.00	0.12	11.00	Qiu et al. (2011)
Changle, Fujian, China	2009, 2011	30.90	77.20	86.40	0.23	84.30	Liu et al. (2014)
Quanzhou, Fujian, China	2009, 2011	14.30	50.90	59.50	0.11	59.80	Liu et al. (2014)
Yunxiao, Fujian, China	2009, 2011	25.20	83.70	65.80	0.29	70.00	Liu et al. (2014)
Zhanjiang, Guangdong, China	2009, 2011	14.00	33.20	25.30	0.16	36.30	Liu et al. (2014)
the United Arab Emirates, Arabian Gulf	—	7.21	11.30	28.10	4.82	11.90	Shriadah (1999)
Jequia'mangrove, Brazil	—	98.60	483.00	160.80	1.32	42.40	Kehrig et al. (2003)
Punta Mala Bay, Panama	2003	56.30	105.00	78.20	<10	23.30	Defew et al. (2005)
Sungei Buloh, Singapore	2004	7.06	51.24	12.28	0.18	16.61	Cuong et al. (2005)
Sungei Khatib Bongsu, Singapore	2004	32.00	120.33	30.98	0.27	32.07	Cuong et al. (2005)
Coast of Red Sea, Saudi Arabia	—	112.00	57.20	45.20	1.23	9.61	Usman et al. (2013)
Pichavaram, India	1995	43.40	93.00	11.20	6.60	141.20	Ramanathan et al. (1999)
Cienaga Grande, Colombia	1995	23.30	91.00	12.60	ND	13.20	Perdomo et al. (1999)
Queensland, Australia	1995	1.00–12.00	23.00–56.00	36.00	0.60	1.00–72.00	Preda and Cox (2002)
Newington, Australia	1996	71.30	229.90	121.90	ND	ND	MacFarlane et al. (2003)

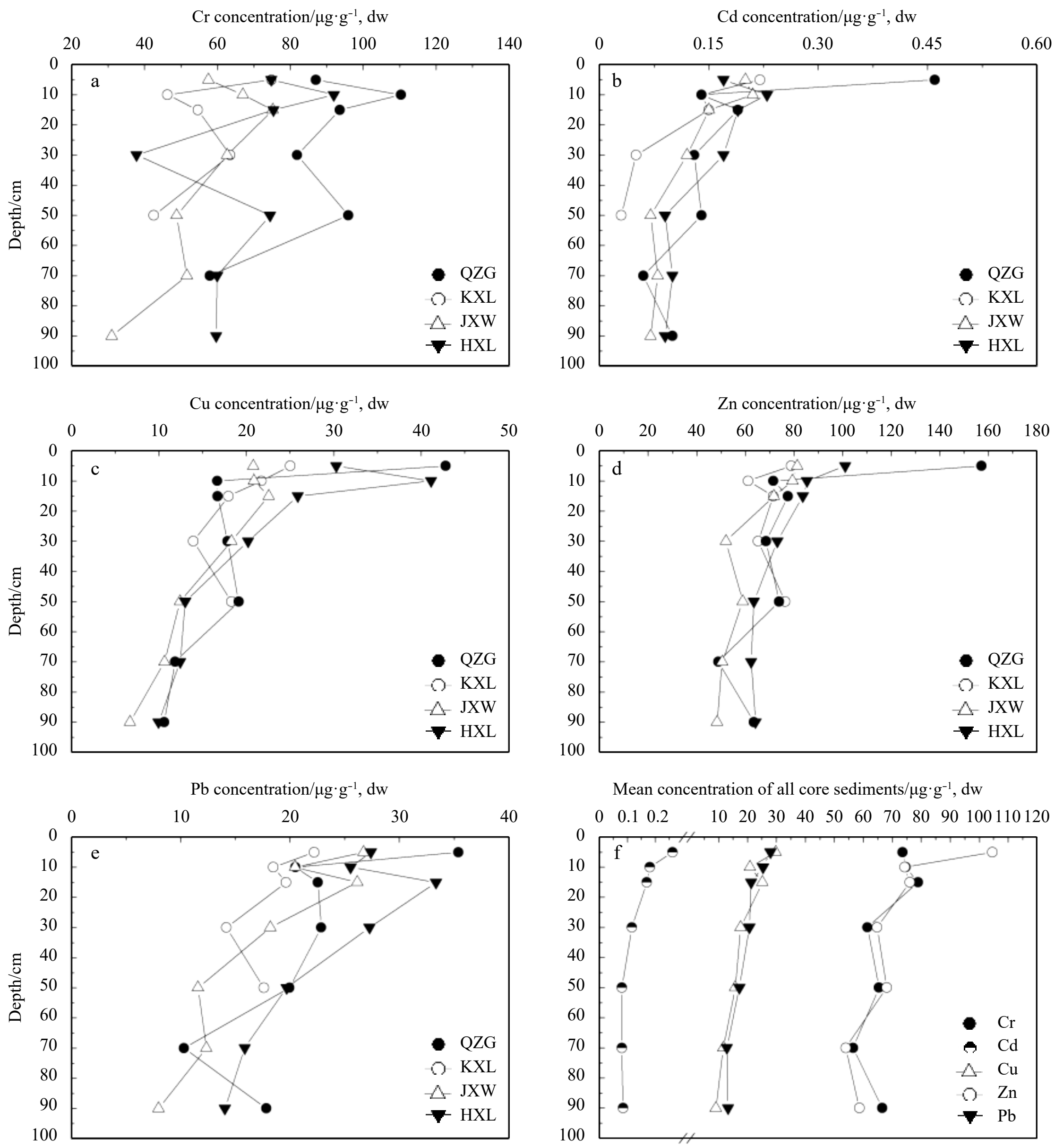

Fig. 3. Vertical profiles of heavy metal concentrations in mangrove core sediments in the Qinzhou Bay.

3.2 *Heavy metals in different plant parts*

The concentrations of Cr, Cd, Cu, Zn and Pb in fruits, roots, stems and leaves of *A. corniculatum* in the Qinzhou Bay are shown in Table 1. In all plant parts, Zn showed the highest concentration, followed by Zn, Pb, Cu, and Cd. In the different mangrove parts, the concentrations of Cr, Cd, Cu, and Pb were highest in roots but lowest in fruits. The accumulation of Zn was highest in the stem, followed by the root, leaf and fruit. The spatial distributions of heavy metals in all parts of *A. corniculatum* varied across site (Fig. 2b). The concentrations of Zn, Pb, Cr and Cd in the plants were the lowest at QZG (Fig. 2b), although they were the highest in the surface sediments at QZG (Fig. 2a). Among different parts (Fig. 4), the concentrations of Cr, Cd, Cu and Pb in roots were significantly higher than those in the fruits, stems and leaves of *A. corniculatum* at the four different sites, except that the concentration of Zn in the stem of *A. corniculatum* at JXW and KXL was the highest among different parts. In addition, the concentrations of the five heavy metals in the fruits were significantly lower than those in other parts.

3.3 *Bioconcentration and translocation of heavy metals*

The BCFs of Cr, Cd, Cu, Zn and Pb in parts of *A. corniculatum* in the Qinzhou Bay and other mangrove plants worldwide are shown in Table 3. In general, the BCFs of different parts showed no significant difference among the four sites in this study, and the BCFs of Cr, Cu, Pb and Cd in the roots of *A. corniculatum* in the Qinzhou Bay were significantly higher than those in the stems, leaves and fruits, while the BCFs of Cu, Cd, Zn and Pb in the fruits were significantly lower than those in the stems, roots and leaves.

The TFs of Cr, Cd, Cu, Zn and Pb in stems, leaves, and fruits of *A. corniculatum* in the Qinzhou Bay are shown in Table 3. The TFs showed no significant difference among the four sites in the Qinzhou Bay, and the TFs of Cd, Cu, Zn and Pb in different parts of *A. corniculatum* decreased in the following order: stem>leaf> fruit. The TFs of Cd, Cu, Zn and Pb in the stems of *A. cornicu-*

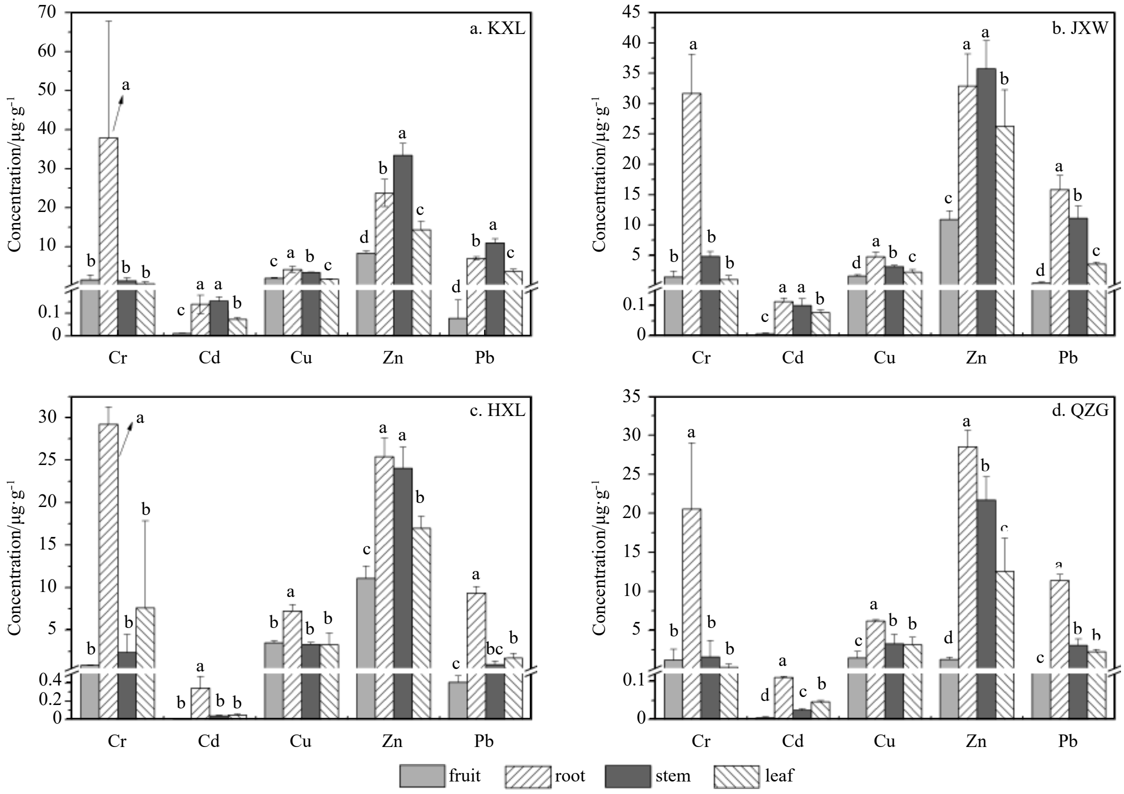

Fig. 4. Concentrations of heavy metals in different parts of *A. corniculatum* in the Qinzhou Bay. Bars with different letters are significantly different at p<0.05.

Table 3. Bioconcentration factors (BCFs) and translocation factors (TFs) of heavy metals in plant parts of *A. corniculatum* in the Qinzhou Bay and other mangrove wetlands

Element	Location	BCF				TF			References
		Fruit	Root	Stem	Leaf	Fruit	Stem	Leaf	
Cr	Qinzhou Bay, China	0.01±0.01	0.29±0.17	0.02±0.02	0.02±0.05	0.04±0.04	0.08±0.07	0.08±0.2	this study
	Shantou, China		0.58	0.06	0.04		0.10	0.08	Zhou et al. (2011)
Cd	Qinzhou Bay, China	0.03±0.02	0.8±0.64	0.44±0.27	0.28±0.11	0.06±0.03	0.74±0.43	0.45±0.22	this study
	Shantou, China		1.85	0.33	0.15		0.18	0.08	Zhou et al. (2011)
Cu	Qinzhou Bay, China	0.06±0.02	0.17±0.04	0.11±0.02	0.08±0.03	0.37±0.12	0.67±0.16	0.46±0.12	this study
	Ting Kok, Hong Kong, China		1.62		1.66			1.03	Chen et al. (2003)
	Wynnum site, Northern Australia				0.28				Saenger et al. (1991)
	Shantou, China		0.93	0.32	0.27		0.35	0.29	Zhou et al. (2011)
	Shenzhen, China				0.1				Tam et al. (1995)
	Mai Po, Hong Kong, China		0.71						Che (1999)
Zn	Qinzhou Bay, China	0.09±0.05	0.31±0.09	0.34±0.13	0.2±0.1	0.27±0.15	1.12±0.29	0.65±0.18	this study
	Ting Kok, Hong Kong, China		0.63		0.31			0.5	Chen et al. (2003)
	Shenzhen, China				0.58				Tam et al. (1995)
	Mai Po, Hong Kong, China		0.29						Che (1999)
	Wynnum site, Northern Australia				0.31				Saenger et al. (1991)
	Shantou, China		0.28	0.19	0.07		0.68	0.26	Zhou et al. (2011)
Pb	Qinzhou Bay, China	0.01±0.01	0.3±0.1	0.24±0.14	0.08±0.03	0.02±0.01	0.87±0.61	0.27±0.14	this study
	Ting Kok, Hong Kong, China		0.94		0.28			0.29	Chen et al. (2003)
	Wynnum site, Northern Australia				0.13				Saenger et al. (1991)
	Mai Po, Hong Kong, China		0.14						Che (1999)
	Shantou, China		1.30	0.08	0.01		0.06	0.01	Zhou et al. (2011)

latum were significantly higher than those in the leaves and fruits. Specifically, the TF of Zn in the stems of *A. corniculatum* was the highest (1.12). In addition, the TF of Pb in the stems was 0.87±0.61 (mean±SD), which was higher than that in the leaves and fruits. Therefore, the translocation ability of stems of *A. corniculatum* with respect to Zn and Pb in the Qinzhou Bay was relatively high.

4 Discussion

4.1 *Trend of heavy metal pollution in sediments in the Qinzhou Bay*

The past decade has been an important development and the fastest growing period of Qinzhou. Due to the rapid development of industrialization, urbanization, and fisheries, undesirable ecological and environmental problems have occurred in this area, including the Qinzhou Harbour, constructed as one of the most important harbours of the Maritime Silk Road in the Beibu Gulf. In this study, the sampling site QZG, located in the Qinzhou Harbour, had the highest concentrations of heavy metals (Fig. 2a). Compared with previous investigations in the Xiandao Park (near the Qinzhou Harbour), the concentrations of Cr, Cd, Cu and Zn in the mangrove surface sediments increased during the last ten years (Li et al., 2008, 2014), especially with respect to Cr (Table 2), which indicated that an external source of Cr existed outside of the mangrove swamp in the Qinzhou Bay. The spatial and temporal distributions of Cr in the Qinzhou Bay might be related to emissions from the chemical industry and improper discharge of aquaculture waste water through surface runoff (Mokrzycki et al., 2003; Liang and Wong, 2003; Bayen, 2012). It also caused higher concentrations of Cr in the Qinzhou Bay (present study) than those in the Dongzhai Harbour, Sanya Bay, and Yanglong Bay in Hainan Province (Qiu et al., 2011), Quanzhou in Fujian Province and Zhanjiang in Guangdong Province (Liu et al., 2014), as shown in Table 2.

The trend of increasingly serious pollution of heavy metals in surface sediments was verified by the investigation of core sediments in this study. In general, heavy metals accumulated in the 0–20 cm layer, which is consistent with previous studies (Tam and Wong, 1996; Machado et al., 2002; Kehrig et al., 2003; Wu et al., 2014). In the Qinzhou Bay, the average deposition rate of mangrove wetlands has been 0.93 cm/a since the 1920s (Gan et al., 2013). Therefore, core sediment deposition in this study occurred over the past 60–100 years, and the concentrations of heavy metals in the sediment layer of 0–20 cm can basically indicate the trends of heavy metals in the study area over the past 20–30 years. In the past 20 to 30 years, China experienced rapid development of the era of reform and opening-up, during which the mangrove landscape in the Qinzhou Bay was significantly affected by human activities and was converted into aquaculture ponds, salt pans, or constructed areas (Li and Dai, 2015). Meng et al. (2016) and Xia et al. (2016) found a significant degradation in the mangrove ecosystem from 1968 according to ^{210}Pb dating, which corresponded to an increase in human activities, especially shrimp-pond reclamation and artificial seawalls.

In contrast, the concentrations of Cu and Zn in this study were similar to those in the 0–45 cm core sediment in the Hainan Island, with a low industrialization level (Qiu et al., 2011), while in the Zhujiang (Pearl) River Estuary with higher levels of urbanization and industrialization, the concentrations of Cr, Cd, Cu, Zn and Pb in the mangrove sediments within a 0–70 cm depth in Nansha and 0–45 cm depth on Qi'ao Island were higher than those in this study (Liu et al., 2011; Wu et al., 2014). These differences further proved that the levels and processes of urbanization and industrialization deeply affect the distribution and pollution of heavy metals in mangrove wetlands (Zuo et al., 2009; Percival and Outridge, 2013; Senthilkumar et al., 2013).

4.2 *Association between heavy metals in the tissues of A. corniculatum and sediments*

The distributions of Cd, Cu and Pb in different parts of *A. corniculatum* in this study were similar to those in Shantou (Zhou et al., 2011). The concentrations of Zn in roots and leaves of *A. corniculatum* in Ting Kok, Hong Kong, reported by Chen et al. (2003), were 26–44 μg/g and 12–22 μg/g, respectively, and were close to the concentrations of Zn in this study, while the concentrations of Cu and Pb were higher than those in this study. The concentrations of Zn, Pb, Cu and Cd in this study were lower than those in the roots of *A. corniculatum* in Mai Po Tung, Hong Kong, while Cr was higher (Che, 1999). In the leaves of *A. corniculatum* of Newington (Saenger et al., 1991), the concentrations of Cu and Zn were 9.6 μg/g and 30 μg/g, respectively, which were higher than those in this study, but the concentration of Pb (1.6 μg/g) was lower than that in the Qinzhou Bay in this study. Overall, the widespread spatial variability of heavy metals in different parts of *A. corniculatum* in different areas was probably due to the difference in the pollution level of heavy metals in the local environmental media, especially sediment (Chen et al., 2003).

Higher concentrations of Cr, Cd, Cu, and Pb in the roots of *A. corniculatum* than in other parts in this study may indicate that the roots act as a barrier for heavy metal translocation. Moreover, higher concentrations of Zn in the stems of *A. corniculatum* might indicate that the stems can take up and store Zn (Analuddin et al., 2017). Mangrove plants accumulate different amounts of trace elements in their aerial tissues because some metals are required as nutrients for growth and survival. The uptake of metals is primarily influenced by plant metabolic requirements, e.g., Cu and Zn, which are essential micro-nutrients and are more mobile than non-essential elements, such as Pb, resulting in variable metal accumulation in plant tissue (MacFarlane et al., 2007). In this study, the data did not show a significant positive linear trend in trace heavy metal accumulations in mangrove roots with increasing sediment concentrations except for Cu (r=0.536, p<0.05). This observation is also true for the correlation between Cr, Cd, Zn and Pb elements in leaves, stems and fruits. The differences in heavy metals among different parts of *A. corniculatum* may suggest that at low metal concentrations, plants translocate metals from the root to the stem to meet metabolic requirements, while at higher concentrations, metal translocation is restricted to prevent toxicity; thus, leaves and fruits only show a slight response to toxic levels of metals present in the underlying sediments, e.g., Pb and Cd (Macfarlane et al., 2003; Wu et al., 2014).

4.3 *Role of A. corniculatum in restoration of sites polluted with heavy metals*

The BCFs of Cu in roots and leaves of *A. corniculatum* in Ting Kok were 1.62 and 1.66, respectively (Chen et al., 2003). These values are much higher than those measured in the Qinzhou Bay in this study (0.17 and 0.08, respectively, Table 3) and those in Futian, Shenzhen (Tam et al., 1995), in addition to the values reported by Saenger et al. (1991) and Zhou et al. (2011). The BCFs of Zn in the roots and leaves of *A. corniculatum* in the Qinzhou Bay were 0.31 and 0.2, respectively, which are similar to the results reported by Zhou et al. (2011) and Che (1999) but much lower than those reported by Chen et al. (2003). High BCFs of Pb in the roots of *A. corniculatum* were reported by Zhou et al. (2011) and Chen et al. (2003), i.e., 1.30 and 0.94, respectively, and these values are higher than the value in this study (0.3). In general, the roots of *A. corniculatum* can absorb and accumulate Cr, Cu and Cd relatively easily.

The BCFs of Pb and Cu in the roots of *K. obovata* and of Zn in the roots of *Avicennia marina* were 0.95, 1.34 and 1.21, respectively (Chen et al., 2003; MacFarlane et al., 2003), which are much higher than those in the roots of *A. corniculatum* in this study. On the Hainan Island (Qiu et al., 2011), the BCFs of Cr, Cd, Cu, Zn and Pb in the various parts of *R. stylosa*, *A. corniculatum*, *Bruguiera sexangula*, and *Sonneratia caseolaris* ranged from 0.02 to 0.61, and the highest BCFs of Zn and Cu were found in fruits, of Pb and Cr were found in stems, and of Cd were found in roots. The BCFs of heavy metals in *Sonneratia apetala* and *Cyperus malaccensis* in Nansha were as low as ND–0.098 and 0.298–0.07 (Wu et al., 2014). Previous studies reported that the mobility of Zn, Cu and other essential elements were higher than that of Pb, Cr and other non-essential elements in mangrove plants, such as *A. marina* and *A. corniculatum* (Chen et al., 2003; MacFarlane et al., 2003, 2007; Zhou et al., 2011). In *S. apetala,* the BCFs of heavy metals were low, but the TFs of Pb, Cr and Zn were no less than 1, indicating that valid translocation of heavy metals from roots to the aboveground organs occurred (Wu et al., 2014). In general, *A. corniculatum* shows relatively high bioconcentration capability for Cd compared with other mangrove plant species and might be used as a restoration species together with *S. apetala* in intertidal zones contaminated by Cd (MacFarlane et al., 2007; Zhou et al., 2011).

The TF of Zn in this paper was higher than that reported by Chen et al. (2003) but lower than that in the leaves of *A. marina* reported by Alongi et al. (2003) and Chen et al. (2003). The TFs of Cr, Cu, Zn and Pb in the leaves of *A. corniculatum* in this study were lower than those in previous reports, while that of Cd was higher (Chen et al., 2003; Zhou et al., 2011). Weis and Weis (2004) reported that Zn could be rapidly accumulated in the leaves of some mangrove plants, but the TF value of Zn in the leaves of *A. corniculatum* was 0.65 in this study, which was lower than that in the stems.

These differences between BCFs and TFs of heavy metals in different parts indicated that the bioaccumulation of heavy metals in mangroves was not necessarily correlated with the translocation of the heavy metals. In this study, although the BCFs of heavy metals in different parts of *A. corniculatum* in Qinzhou Bay were low, the TFs were relatively high. Moreover, *A. corniculatum* has a high biomass; thus, this species can absorb and accumulate a large amount of heavy metals such as Zn and Cu (with high TFs) and Cd (with a high BCF). *A. corniculatum* may be a suitable mangrove plant for the restoration of intertidal zones contaminated by Zn, Cu and Cd (Tam and Wong, 1995; Thomas and Fernandez, 1997; Usman et al., 2013; Wu et al., 2014; Wang et al., 2015). However, it is worth noting that maintaining a high diversity in mangrove ecosystems is very important for the restoration of heavy metals in coastal zones due to the poor pollution mitigation ability of single-species communities. Therefore, it is necessary to investigate additional mangrove species to address the complex situation of polluted coasts.

5 Conclusions

The present study revealed that the concentrations of Cd, Cu, Zn, Cr and Pb in surface sediments in the Qinzhou Bay were at the low to moderate pollution level compared with other reports worldwide and posed a slight potential ecological risk. The pollution of heavy metals in the surface sediments of the *A. corniculatum* community in QZG was relatively high compared with the three other sampling sites due to the influence of the harbour. Core sediments at depths of 0–20 cm showed significantly higher levels of heavy metals compared with those in the deeper layers. In the *A. corniculatum* plants, the accumulation of Cr was mainly restricted to the roots, while Cd, Cu, Zn and Pb showed considerable translocation potential to the aboveground part (stem), indicated by relatively high TF values. According to the results of the present study, we conclude that *A. corniculatum* can be chosen as a potential bioremediation species in wetlands polluted by Zn, Cu and Cd because of its strong bioconcentration ability for Cd and considerable transport capacity for Zn and Cu.

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