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RESEARCH ARTICLE

# Distribution fractions and potential ecological risk assessment of heavy metals in mangrove sediments of the Greater Bay Area

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## Abstract

The restoration of mangrove in coastal wetlands of China has been started since the 1990s. However, various pollutants, especially for heavy metals (HMs), contained in wastewater might present a significant risk to mangrove forests during the restoration. In this study, sediments of five typical mangrove wetlands with varying restoration years and management measures in the Greater Bay Area were collected to evaluate the distribution fractions and potential ecological risk of HMs. Cd (0.2–1.6 mg/kg) was found in high concentrations in the exchangeable fraction (37.8–71.5%), whereas Cu (54.2–94.8 mg/kg), Zn (157.6–332.6 mg/kg), Cr (57.7–113.6 mg/kg), Pb (36.5–89.9 mg/kg), and Ni (29.7–69.5 mg/kg) primarily presented in residual fraction (30.8–91.9%). According to the geo-accumulation index ($I_{geo}$) analysis, sediment Cd presented a high level of pollution ($3 \le I_{geo} \le 4$), while Zn and Cu were associated with moderately pollution ($1 \le I_{geo} \le 2$). Besides, high ecological risk of Cd was found in sediments of five mangroves, with risk assessment code (RAC) ranging from 45.9 to 84.2. Redundancy analysis revealed that the content of $NO_3^-$-N was closely related to that of HMs in sediments and, pH value and $NO_3^-$-N concentration affected the distribution of HMs geochemical fractions. High concentration of HMs in QA and NS sampling sites was caused by the formerly pollutants discharge, resulting in these sediments still with a higher HM pollution level after the plant of mangrove for a long period. Fortunately, strict drainage standards for industrial activities in Shenzhen significantly availed for decreasing HMs contents in mangrove sediments. Therefore, future works on mangrove conversion and restoration should be linked to the water purification in the GBA.

**Keywords** Mangrove restoration · Heavy metals · Ecological risk assessment · Coastal Sediment · Redundancy Analysis

## Introduction

Despite occupying no more than 0.06% of the earth's surface, mangrove ecosystem actually played an important role in water purification, wind/waves protection, and carbon restoration (De Lacerda and Linneweber 2002; FIELD et al. 1998; Hamilton and Casey 2016). Due to the negative effects of anthropogenic activities, e.g., urbanization and aquaculture, coastal areas for mangrove forests have been significantly decreased by ~ 40% worldwide over the past half-century (Herbeck et al. 2020). Fortunately, with improved environmental protection awareness and the development of social economy in developing countries, especially in China, the conservation and restoration of mangrove forests in coastal wetlands have been rapidly established since the 1990s (Herbeck et al. 2020). However, the continuous discharge of pollutants from anthropogenic activities accomplished by industrial development might still be a seriously threat to mangrove forests, especially for those in restoration processes (Lewis et al. 2011; Maiti and Chowdhury 2013). Nowadays, scarce information is presented concerning the potential ecological threats of anthropogenic pollutants on mangrove forests with varying restoration years.

As representative pollutants, heavy metals (HMs), mainly present in industrial wastewater discharge into estuary areas

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(Algül and Beyhan 2020; Sun et al. 2022), resulting in a significant risk to the mangrove ecosystem (Feng et al. 2017; Tam and Wong 2000b; Zhang et al. 2014). The particularity of mangrove ecosystem was benefit for fine sediments, organics, and minerals deposition, indirectly promoting the accumulation of HMs in the relevant sediments (Prasad and Ramanathan 2008; Shao et al. 2009). In addition, the geochemical fractions of HMs in sediments might be transformed with acid dissolution and complexation effects (Zhang et al. 2017). It is generally agreed that bioavailable HMs in sediments could be easily accumulated by mangrove forests, resulting in a serious risk to plant health. This was mainly attributed to two reasons: (i) anions, e.g., $Cl^-$, $SO_4^{2-}$, migrated with tidal process could complex with HMs (Hirsch et al. 1989; Weggler et al. 2004); (ii) protons, organic acids and amino acids secreted by microorganisms and mangroves availed for soil acidification and lead to the increase amount in bioavailable HMs fractions (Wang et al. 2021). Furthermore, mangrove forests also played a significant part in the uptake of HMs from sediments, but this effect is significantly different due to the differences of mangrove growth stages (Feng et al. 2017). However, few research has considered the relationships between HMs distribution characteristics and external pollutants or physicochemical properties of mangrove sediments in restoration stages.

The Greater Bay Area (GBA), located in (sub)tropical regions, includes Hong Kong, Macao, and nine cities in Guangdong. There are obvious differences in industrial distribution and economic development levels due to varying development emphasis among these cities. Comparatively, cities on the east bank of the GBA have more electronic manufacturing factories, resulting in certain differences in pollutant discharge amount during the industrial activities. In addition, there are certain differences in the treatment standards of industrial wastes among these cities with varying urban environmental policies (Yang et al. 2012), resulting in the different fractions and concentrations of HMs discharged into surrounding mangrove ecosystem. Furthermore, due to the variations in starting time of mangrove protection, mangrove planting years varied significantly between different regions, which might affect the tolerance of mangroves to HMs pollutants (Bai et al. 2011). Therefore, it is of great significance to understand the impacts of human activities and urban development on the HMs distribution features of mangrove wetlands in the GBA.

Based on the above mentioned, we hypothesized that the concentration distribution and geochemical fraction of HMs in the sediments of mangrove with different restoration years would be significantly influenced by the industrial distribution, economic development level, and management requirements in different cities of GBA. Therefore, five representative mangroves sites with certain differences in planting years and industrial activities were selected for sediments sampling in the GBA. The study's objectives were (i) to determine the relationship between sediment HMs concentration and urban effluent nutrients (e.g., AP, $NH_4^+$, $NO_3^-$), (ii) to estimate the influences of varying physiochemical properties on the geochemical fractions of HMs in sediments, and (iii) to evaluate the effects of industrial development and environment protection on HMs accumulation in mangrove sediments.

## Materials and methods

### Study area

The GBA is located in southern China, with an annual mean temperature of 21.4–22.4 °C and an annual average precipitation of 1662.7 mm (2020). As one of the most economically developed regions in China, extensive industrial and anthropogenic activities in this area have caused a relatively serious HM contamination. It has been confirmed that surface water in this area contained with trace metals from mining, industrial, and agricultural activities (Zhao et al. 2017). According to the annual detection data of National Estuary in 2021, the main pollutants in aquatic environment were nitrogen and HMs. For instance, total nitrogen concentrations in river sections of Guangzhou (GZ), Shenzhen (SZ), Zhuhai (ZH), and Jiangmen (JM) were 2.2, 0.8, 0.6, and 0.5 mg/L, respectively.

Coastal ecosystems, especially mangrove wetland system, with outstanding HM and nutrient retention capacity, play an important role in the environmental protection in terrestrial-marine transitions (Yan et al. 2017; Yim and Tam 1999). Therefore, to reveal the influence factors, e.g., human activity, regional protection policies, and restoration age, on the ecological function of mangrove wetlands, five mangrove wetland sites were selected within the GBA. Two sampling sites with three replicates of each mangrove wetland in Beidou (BD), Qi'ao Island (QA), Nansha (NS), Hai'ou Island (HO), and Futian (FT) (Fig. 1), which are located in JM, ZH, GZ, GZ, SZ, respectively. Among these sites, QA, NS, HO, and FT were located on the western and eastern side of the Pearl River, respectively, while for BD, it was located on the western side of Tan River. All these sampling sites were belonged to Pearl River basin. Notably, NS site belongs to natural mangrove forest (at least 40 years), while the mangrove restoration ages of mangrove forest at BD, QA, HO, and FT sites are approximately 10, 20, 5, and 3 years, respectively. Given that population density and protective measure of the local situation, mangroves in BD, QA, and FT sites as affected by human activity are relatively weaker than that in NS and HO sites. The invasion of alien species, e.g., *Spartina alterniflora, Mikania micrantha Kunth*, and *Leucaena*

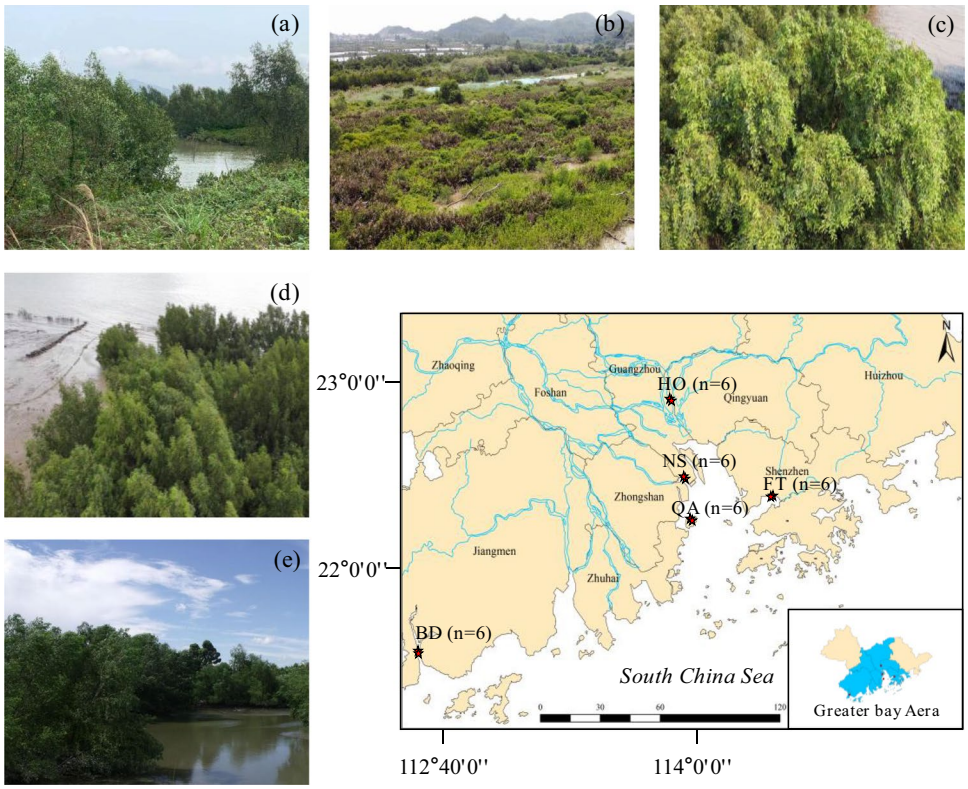


**Fig. 1** Location map of study area and sampling sites in the GBA. Note: panels (**a**), (**b**), (**c**), (**d**), and (**e**) represent mangrove wetlands in BD, QA, NS, HO, and FT sites, respectively

*leucocephala*, in QA site occurred 18–24 years ago. While FT site was introduced with *Aegiceras corniculatum*, *Kandelia obovataa sheue*, and *Avicennia marina* 3 years ago, after the removal of the alien species *Sonneratia apetala*, which has been planted for 17 years. The environmental department of SZ has issued strict industrial effluent discharge standard since 2016, which is a stricter exhaustion standard when compared with the relevantly national discharge standards performed in other cities, such as GZ and JM.

## Sediment sampling and chemical analyses

Mangrove surface sediments at two depths of 0–20 and 20–40 cm were collected from BD, QA, NS, HO, and FT sites in the GBA (Fig. 1) in April 2021, and the entire sampling process lasted for 2 weeks. After collection, they were immediately stored in car refrigerators and then prepared for subsequent analyses in the laboratory. After 2 weeks of air drying at room temperature, all samples were screened through a 2-mm sieve to remove small gravel and coarse debris. Each dried sample was ground until every particle passed through a 0.149-mm nylon sieve for determination of physicochemical properties and HMs fractions.

Sediment pH was determined using a calibrated pH meter (HQ40D, Hach, USA) with the supernatant collected from soil–water mixture liquid at a soil–water weight ratio of 1:5. Sediment organic carbon (SOC) was analyzed with an elemental analyzer (Vario TOC Cube, Elementar, Germany). The concentrations of nitrogen ($NH_4^+$-N and $NO_3^-$-N forms) and available phosphorus in sediments were detected according to the standards of HJ 634–2012 and NY/T 1121.7–2014, respectively. The total HMs, e.g., Cu, Zn, Cd, Cr, Pb, Ni, were determined by ICP-MS (NexION 350, Shimadzu, Japan) after digestion with hydrochloric acid and nitric acid in the ratio of 1:3. According to a five-step process suggested by Tessier et al. (1979), sediment HMs were extracted and classified into five fractions, including exchangeable fraction (F1), carbonate fraction (F2), Fe–Mn oxide fraction (F3), organically bound fraction (F4), and residual fraction (F5), respectively. For HMs concentration tests, blank and standard reference materials (GBW07401) were injected into ICP-MS for evaluating the determination process's accuracy and precision. The recovery rate of HMs samples spiked with standards was maintained at the range of 95–105%.

## Geo-accumulation index ($I_{geo}$)

The geographic accumulation index ($I_{geo}$) was used to evaluate the contamination situation of HMs in mangrove sediments, which is defined by Eq. (1) (Muller 1969) as follows:

$$I_{geo} = \log_2 \frac{C_n}{1.5 \times B_n} \quad (1)$$

where $C_n$ and $B_n$ represent the measured concentration of HMn in the sediment and the geological background value of HMn, respectively. The constant term 1.5 takes the consideration of the fluctuation of HMs background value in sediments caused by possible lithological variations (Ma et al. 2016). Zn, Cr, Cu, Ni, Cd, and Pb in the GBA have background concentrations of 47.3, 50.5, 17, 18.2, 0.056, and 36 mg/kg, respectively (Chen et al. 1991). According to $I_{geo}$ values, the pollution situation of each HM is divided into seven levels: $I_{geo} \leq 0$: unpolluted, $0 < I_{geo} \leq 1$: weakly polluted, $1 < I_{geo} \leq 2$: moderately polluted, $2 < I_{geo} \leq 3$: moderately to heavily polluted, $3 < I_{geo} \leq 4$: heavily polluted, $4 < I_{geo} \leq 5$: heavily to and extremely polluted; $I_{geo} > 5$: extremely polluted.

### Risk assessment code (RAC)

The mobility and bioavailability of HMn are assessed by the RAC according to its proportion of exchangeable and carbonate fractions to the total fractions in sediments, which is defined by Eq. (2) as follows:

$$RAC_i = \frac{Metal^i_{ef} + Metal^i_{cf}}{Metal^i_t} \times 100 \quad (2)$$

where $Metel_{ef}$, $Metel_{cf}$, and $Metel_t$ represent the concentration of HM in exchangeable fraction, carbonate fraction, and the sum of all fractions, respectively. According to assessment results, the RAC indices are divided into five levels: $RAC \leq 1$: no risk (NR), $1 < RAC \leq 10$: low risk, $10 < RAC \leq 30$: medium risk, $30 < RAC \leq 50$: high risk, and $50 < RAC \leq 100$: very high risk (Marrugo-Negrete et al. 2017).

### Statistical analysis

The differences in physicochemical properties and HM concentration among different mangrove sediments (SPSS 20.0, IBM, USA) were identified by the One-way analysis of variance (ANOVA) and $p < 0.05$ was considered as the significant level. Pearson's correlation test was used to assess the correlation analysis between HMs content and sediment physicochemical properties. Furthermore, redundancy analysis (RDA) was used with canoco 5 for windows to evaluate the relationships between HMs and sediment physicochemical properties.

## Results

### Physicochemical properties of mangrove sediments

The physicochemical properties of mangrove sediments for surface (0–20 cm) and bottom (20–40 cm) layers in the GBA are presented with Table 1. Five sites of mangrove sediments showed almost no statistically significant difference in pH between two layers ($p > 0.05$); higher pH values were found at BD and NS sites. The SOC concentration increased with planting years, especially for mangroves that planted over 20 years. With the increase of planting years, the SOC content significantly increased with depth in the QA site ($p < 0.05$). The content of $NH_4^+$-N in sediments of two layers for five sites were substantially higher than that of $NO_3^-$-N. In addition, $NH_4^+$-N contents in sediments of two layers for five sites showed no obvious

**Table 1** Physicochemical properties of mangrove sediments (two layers: 0–20 and 20–40 cm) collected from five sampling sites in the GBA (Data: average value ± standard deviation)

| Site types | Mangrove age (years) | Sediment layer (cm) | pH | SOC (g/kg) | $NH_4^+$-N (mg/kg) | $NO_3^-$-N (mg/kg) | Available P (mg/kg) |
|---|---|---|---|---|---|---|---|
| Site BD ($n=6$) | 10 | 0–20 | $7.3 \pm 0.2^{A1}$ | $23.5 \pm 3.6^{A1}$ | $20.1 \pm 2.7^{A1}$ | $2.6 \pm 0.3^{A1}$ | $6.5 \pm 0.5^{A1}$ |
| | | 20–40 | $7.3 \pm 0.2^{A1}$ | $22.6 \pm 1.6^{A1}$ | $13.8 \pm 1.9^{B1}$ | $1.9 \pm 0.2^{B1}$ | $6.6 \pm 1.2^{A1}$ |
| Site QA ($n=6$) | 20 | 0–20 | $6.7 \pm 0.2^{A2}$ | $33.3 \pm 2.2^{A2}$ | $14.6 \pm 2.0^{A2}$ | $3.0 \pm 0.3^{A12}$ | $10.3 \pm 1.2^{A2}$ |
| | | 20–40 | $7.0 \pm 0.5^{A1}$ | $30.4 \pm 1.3^{A2}$ | $12.0 \pm 1.3^{B1}$ | $0.7 \pm 0.3^{B2}$ | $9.0 \pm 3.4^{A2}$ |
| Site NS ($n=6$) | > 40 | 0–20 | $7.1 \pm 0.5^{A12}$ | $24.0 \pm 1.7^{A1}$ | $9.1 \pm 1.2^{A3}$ | $3.3 \pm 0.6^{A2}$ | $8.6 \pm 0.4^{A12}$ |
| | | 20–40 | $7.7 \pm 0.4^{B2}$ | $18.1 \pm 2.2^{B3}$ | $14.7 \pm 3.1^{B12}$ | $2.8 \pm 0.6^{A3}$ | $5.7 \pm 0.5^{B1}$ |
| Site HO ($n=6$) | 5 | 0–20 | $6.6 \pm 0.4^{A2}$ | $21.0 \pm 3.8^{A1}$ | $11.9 \pm 1.3^{A23}$ | $4.3 \pm 1.0^{A3}$ | $13.2 \pm 3.0^{A3}$ |
| | | 20–40 | $6.8 \pm 0.2^{A1}$ | $18.2 \pm 2.2^{B3}$ | $16.7 \pm 2.4^{B2}$ | $2.8 \pm 0.1^{B3}$ | $11.2 \pm 1.9^{A3}$ |
| Site FT ($n=6$) | 3[c] | 0–20 | $6.9 \pm 0.4^{A12}$ | $20.5 \pm 1.1^{A1}$ | $7.5 \pm 0.3^{A3}$ | $1.1 \pm 0.5^{A4}$ | $22.9 \pm 3.6^{A4}$ |
| | | 20–40 | $7.1 \pm 0.9^{A1}$ | $21.1 \pm 1.3^{A13}$ | $10.6 \pm 4.6^{B3}$ | $1.2 \pm 0.3^{A4}$ | $28.2 \pm 1.6^{B4}$ |

a. [AB] Values in each column with the same letters are not significantly different (LSD) among different sediment layers in same sediment type ($p < 0.05$). b. [123] Values in each column with different numbers represent significant differences (LSD) among different mangrove sediment types in same sediment layer ($p < 0.05$). c. Mangrove forests have been planted again for about three years due to the elimination of alien invasion species

**Table 2** Total concentrations of HMs, including Cu, Zn, Cd, Cr, Pb, and Ni, in mangrove sediments (two layers: 0–20 and 20–40 cm) collected from five sampling sites in the GBA

| Site types | Mangrove age (years) | Sediment layer (cm) | Cu (mg/kg) | Zn (mg/kg) | Cd (mg/kg) | Cr (mg/kg) | Pb (mg/kg) | Ni (mg/kg) |
|---|---|---|---|---|---|---|---|---|
| Site BD ($n=6$) | 10 | 0–20 | $54.2\pm2.5^{A1}$ | $241.7\pm15.3^{A1}$ | $0.2\pm0.02^{A1}$ | $72.4\pm14.9^{A1}$ | $74.2\pm2.7^{A1}$ | $40.4\pm9.3^{A1}$ |
| | | 20–40 | $64.3\pm4.4^{B1}$ | $260.3\pm8.9^{B1}$ | $0.3\pm0.04^{A1}$ | $75.3\pm8.4^{A1}$ | $48.9\pm2.4^{B1}$ | $45.5\pm7.5^{A1}$ |
| Site QA ($n=6$) | 20 | 0–20 | $77.8\pm4.8^{A2}$ | $332.6\pm14.2^{A2}$ | $0.6\pm0.04^{A2}$ | $83.8\pm4.2^{A2}$ | $50.4\pm3.2^{A2}$ | $45.4\pm1.4^{A1}$ |
| | | 20–40 | $84.4\pm3.0^{A2}$ | $317.2\pm11.0^{A2}$ | $0.6\pm0.04^{A2}$ | $93.2\pm4.1^{B2}$ | $51.4\pm3.7^{A1}$ | $50.2\pm3.4^{A1}$ |
| Site NS ($n=6$) | >40 | 0–20 | $55.7\pm5.3^{A1}$ | $328.4\pm13.0^{A2}$ | $0.7\pm0.06^{A2}$ | $70.8\pm2.8^{A1}$ | $52.9\pm1.4^{A2}$ | $38.7\pm2.3^{A13}$ |
| | | 20–40 | $89.0\pm1.3^{B23}$ | $286.8\pm14.0^{B12}$ | $1.1\pm0.07^{B3}$ | $64.4\pm4.1^{B3}$ | $36.5\pm1.9^{B2}$ | $53.6\pm15.0^{B1}$ |
| Site HO ($n=6$) | 5 | 0–20 | $92.1\pm4.4^{A3}$ | $306.2\pm19.2^{A2}$ | $1.6\pm0.02^{A3}$ | $113.1\pm25.4^{A3}$ | $89.9\pm5.9^{A3}$ | $69.5\pm4.2^{A2}$ |
| | | 20–40 | $94.8\pm6.3^{A3}$ | $283.2\pm17.1^{B12}$ | $1.0\pm0.18^{B3}$ | $113.6\pm17.8^{A4}$ | $72.8\pm5.3^{B3}$ | $59.5\pm13.4^{B2}$ |
| Site FT ($n=6$) | 3[c] | 0–20 | $79.7\pm10.0^{A23}$ | $157.6\pm9.3^{A3}$ | $0.4\pm0.03^{A1}$ | $57.7\pm4.1^{A4}$ | $51.5\pm6.6^{A2}$ | $29.7\pm5.8^{A3}$ |
| | | 20–40 | $71.1\pm4.8^{B12}$ | $273.1\pm18.0^{B1}$ | $0.3\pm0.05^{A1}$ | $75.3\pm8.3^{B1}$ | $61.2\pm1.2^{B4}$ | $32.3\pm2.2^{A3}$ |

a. $^{AB}$ Values in each column with the same letters are not significantly different (LSD) among different sediment layers in same sediment type ($p<0.05$). b. $^{123}$ Values in each column with different numbers represent significant differences (LSD) among different mangrove sediment types in same sediment layer ($p<0.05$). c. Mangrove forests have been planted again for about three years due to the elimination of alien invasion species

pattern, however, $NO_3^-$-N concentrations in the surface layer (0–20 cm) of five sites were higher than that of the bottom layer (20–40 cm). The content of available P in mangrove sediments was significantly higher at the FT site than at other sampling sites ($p<0.05$), but there was no statistically significant difference in available P between two layers among five mangrove sediment sites ($p>0.05$).

The average concentrations of HMs in sediments of two layers for five sites ranged from 0.2–369.7 mg/kg and demonstrated the following pattern: Zn > Cr > Cu ≈ Pb > Ni > Cd (Table 2). Generally, the mean values of HMs in two layers of sediments collected from BD and FT sites were lower than that of QA, NS, and HO sites in most cases. In the BD site, the contents of Cu, Cd, Cr, and Ni in the surface layer (0–20 cm) of sediments were similar to that of the bottom layer (20–40 cm) ($p>0.05$), and the concentration of Zn and Pb in the surface layer (0–20 cm) of sediments was significantly lower and higher than that

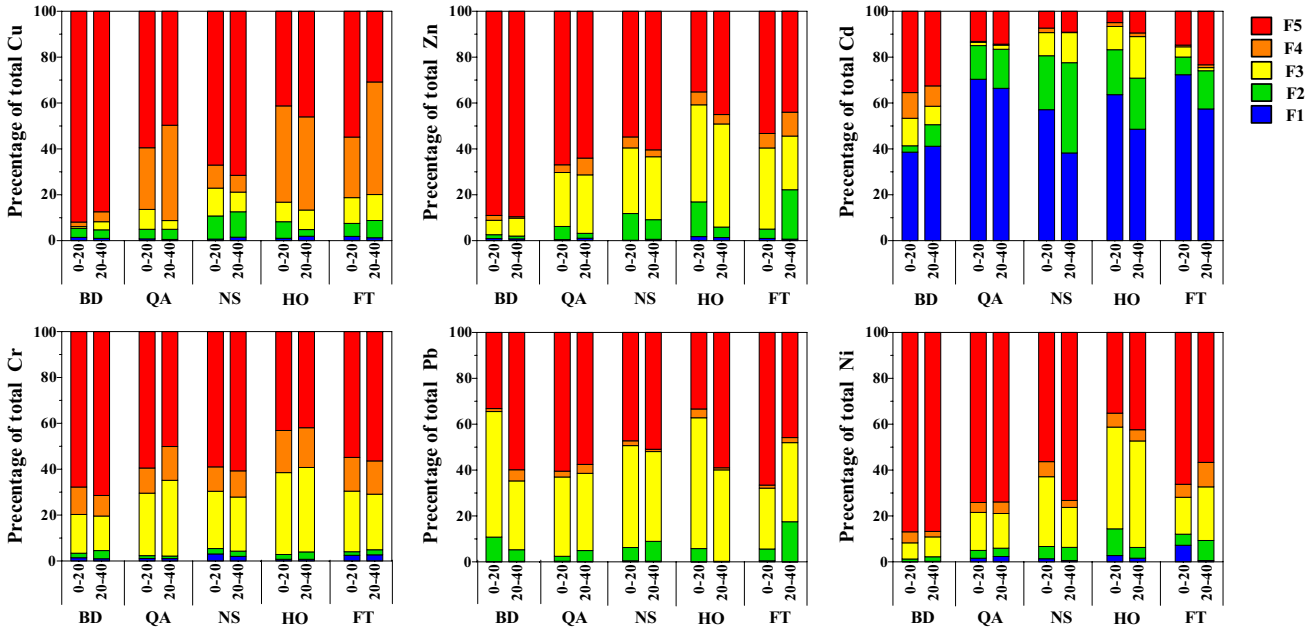


**Fig. 2** Percentages of five geochemical fractions for Cu, Zn, Cd, Cr, Pb, and Ni in two layers (0–20 cm, 20–40 cm) of sediments collected from BD, QA, NS, HO, and FT sites, respectively. HMs was classified into exchangeable fraction (F1), carbonate fraction (F2), Fe–Mn oxide fraction (F3), organically bound fraction (F4), and residual fraction (F5), respectively

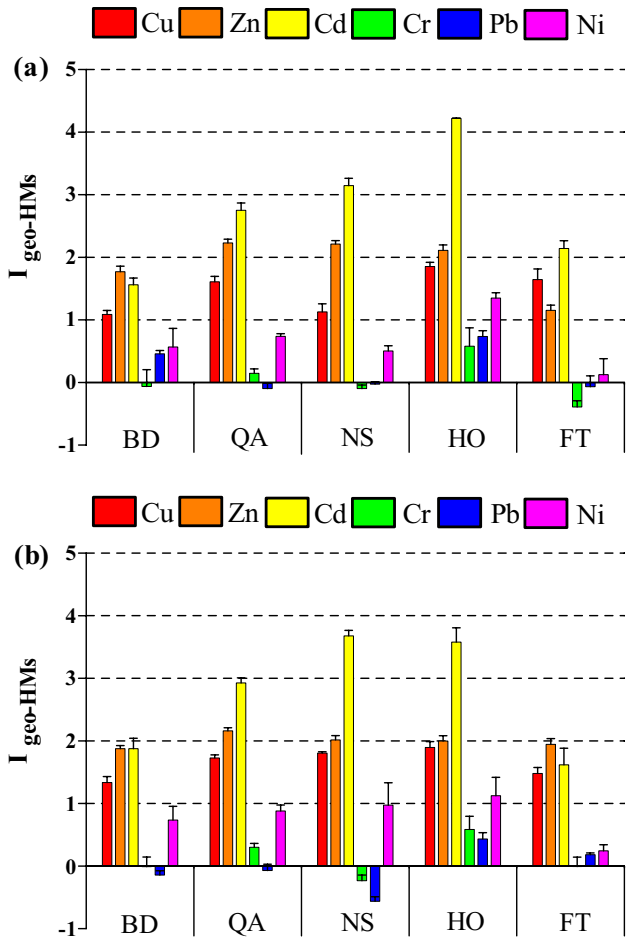


**Fig. 3** The geo-accumulation index ($I_{geo}$) of HMs in mangrove sediments (two layers: 0–20 and 20–40 cm) collected from BD, QA, NS, HO, and FT sites, respectively. Dotted lines represent different pollution levels

of the bottom layer (20–40 cm), respectively ($p < 0.05$). There was no significant difference in HMs content in the two layers of sediments from the QA site except Cr. In addition, much more Zn, Cr, and Pb in the surface layer (0–20 cm) of NS site and Cd, Pb, Ni in the surface layer (0–20 cm) of HO site were found compared with that of the corresponding bottom layer (20–40 cm) ($p < 0.05$). The concentrations of Zn, Cr, and Pb in the surface layer (0–20 cm) of the FT site were significantly lower than those in the bottom layer (20–40 cm) ($p < 0.05$).

## Heavy metals fractions in mangrove sediments

The mean percentages of five geochemical fractions for six HMs in two layers of sediments collected from five mangrove sites in the GBA are illustrated in Fig. 2. Although some minor differences in five extraction fractions were found among these sediment samples, the distribution of HMs fractions exhibited with similar patterns at two sediment layers.

As shown in Fig. 2, the exchangeable fraction of Cd presented with the largest percentage (mean value: 38.6% in BD, 70.4% in QA, 57.1% in NS, 63.6% in HO, and 72.3% in FT) among five sequentially extraction phases. Furthermore, the percentage of residual fraction of Cd in BD and FT sites, and the carbonate fraction of Cd in QA, NS, and HO sites also had a relatively high proportion in the surface sediments. The ratio of exchangeable Cd showed a decreasing trend, whereas an opposite tendency was observed for the percentage of carbonate and residual Cd with the increasing depth of sediments. Generally, much higher percentage of exchangeable Cd was observed in mangrove sediments when compared with that of other HMs ($p < 0.05$).

Cr, Pb, Zn, and Ni fractions in two sediment layers followed the order: residual > Fe–Mn oxide > organic-bound > carbonate-bound > exchangeable. The ratios of residual (organic-bound) Cr, Pb, Zn, and Ni ranged from 46.9–62.1% and 33.2–66.5%, respectively. For Fe–Mn oxide fraction, they varied in ranges of 24.2–37.2%, 26.5–54.7%, 6.3–44.9%, and 7.0–46.4%, respectively. For sediment Cu, it distributed mostly in residual form in BD and NS, accounting for 89.6 and 69.2% of total amount, respectively. However, Cu in QA, HO and FT was primarily in the organic-bound and residual state, the former and latter fractions accounted for 34.2–41.3% and 42.8–54.6%, respectively.

## Heavy metals pollution and ecological risk assessment

The $I_{geo}$ evaluation for six HMs in two sediment layers of five mangrove sites in the GBA is illustrated in Fig. 3. Except for Ni, there was no discernible difference in $I_{geo}$ values between the surface layer (0–20 cm) and the bottom layer (20–40 cm) ($p > 0.05$). The pollution levels of these metals demonstrated the following pattern: Cd > Zn > Cu > Ni > Pb > Cr, $I_{geo}$ values among five sites were as followed: HO > NS > QA > FT > BD. In general, Cd in most sediments at five sites was classified as "heavily polluted" ($3 < I_{geo} \leq 4$). Comparatively, Zn and Cu in these sites showed "moderately polluted" ($1 < I_{geo} \leq 2$) according to $I_{geo}$ analysis. The other metals, including Cr, Pb, and Ni, were found with negative $I_{geo}$ values among five sediment sites, indicating an "unpolluted" or "weakly polluted" level.

According to RAC analysis, Cd in mangrove sediments presented with a high ecological risk, ranging from 45.9 to 84.2 (Fig. 4). There was no significant difference in the RAC value of Cd between surface (0–20 cm) and bottom (20–40 cm) sediments ($p > 0.05$). In general, the risk level tendency of these metals among different sampling sites are similar to that of $I_{geo}$. According to $I_{geo}$ evaluation, Cu and Zn were moderately polluted in mangrove sediment, while their RAC values were

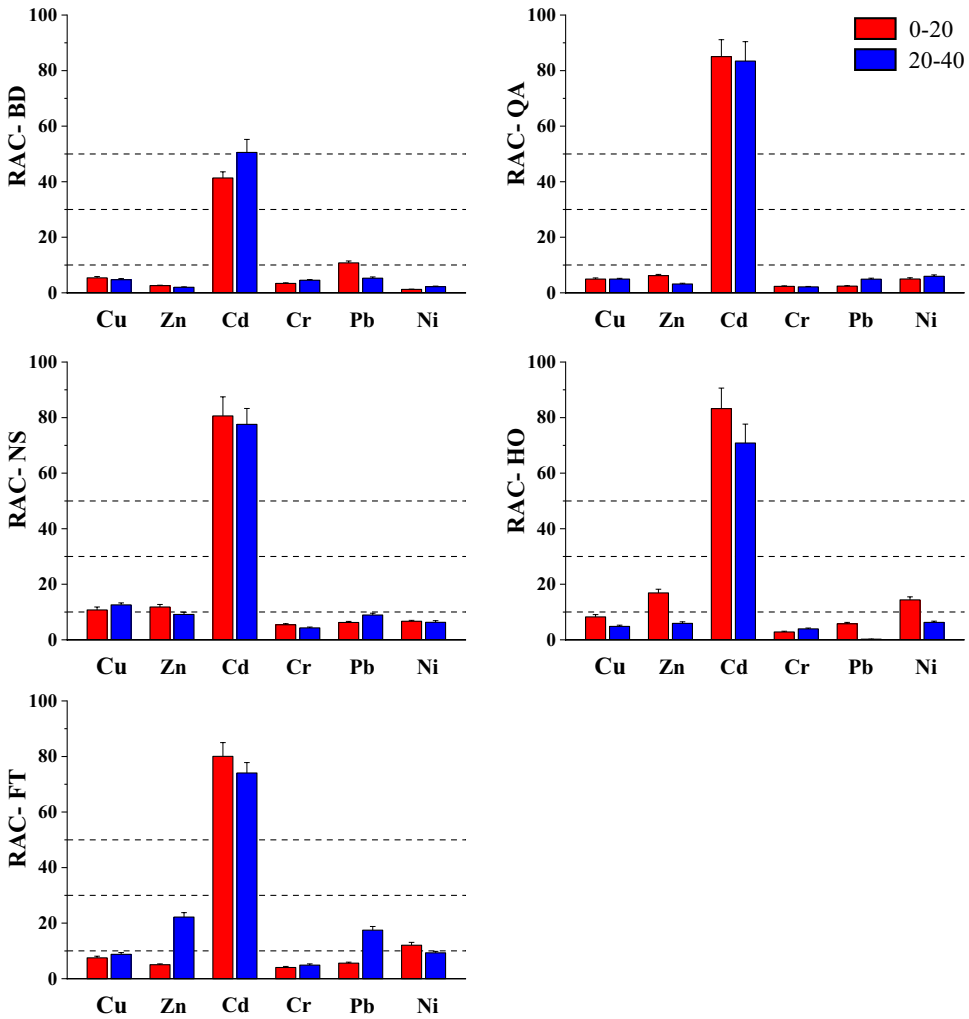


**Fig. 4** The risk assessment code (RAC) values of Cu, Zn, Cd, Cr, Pb, and Ni in mangrove sediments (two layers: 0–20 and 20–40 cm) collected from BD, QA, NS, HO, and FT sites, respectively. Dotted lines represent different risk levels

low ecological risk with the ranges of 4.7–12.6 and 1.9–22.2, respectively. Furthermore, the RAC value of Cr in five mangrove sediments showed a low risk (2.3–5.6), while for other metals, including Pb and Ni, ranged from 0.3 to 17.5, suggesting a "medium risk" or "low risk" level. Higher RAC value of Cu, Zn, Pb, and Ni in the surface layer (0–20 cm) of HO site was found compared with that of the corresponding bottom layer (20–40 cm) ($p < 0.05$), while the RAC value of Zn and Pb in the surface layer (0–20 cm) of FT site was lower than these in the bottom layer (20–40 cm) ($p < 0.05$).

## Relationships between heavy metals and sediment physicochemical properties

The RDA of HMs (Zn, Cr, Cu, Ni, Cd, and Pb) and physicochemical properties (pH, SOC, $NH_4^+$-N, $NO_3^-$-N, and available P) among five mangrove sediments were illustrated in Fig. 5. In general, over 75% of changes in the total content of six HMs can be explained by sediment physicochemical properties. The total concentrations of HMs distributed in five mangrove sediments were strongly affected by $NO_3^-$-N and $NH_4^+$-N. According to the RDA ordination diagram, sediments sampled from five mangroves sites exhibited a certain level of similarity, especially for BD and FT. The Pearson correlation analysis also revealed a significant correlation between the concentrations of HMs and sediment nitrogen, especially for $NO_3^-$-N ($p < 0.05$) (Table 3). In addition, the total concentrations of most HMs in these sediments showed a significant correlation with each other.

The physicochemical properties of mangrove sediments could also be used to assess the variations of geochemical fractions of HMs (Fig. 6). In general, $NO_3^-$-N was closely related to Fe–Mn bound fraction of six metals except for Cu, and the linear regression correlation of $NO_3^-$-N and Fe–Mn bound state in Table 4 re-confirmed the former result. Notably, the linear relationship was more obvious at QA, NS, and HO sites with relatively serious HM pollution levels. Furthermore, RDA results

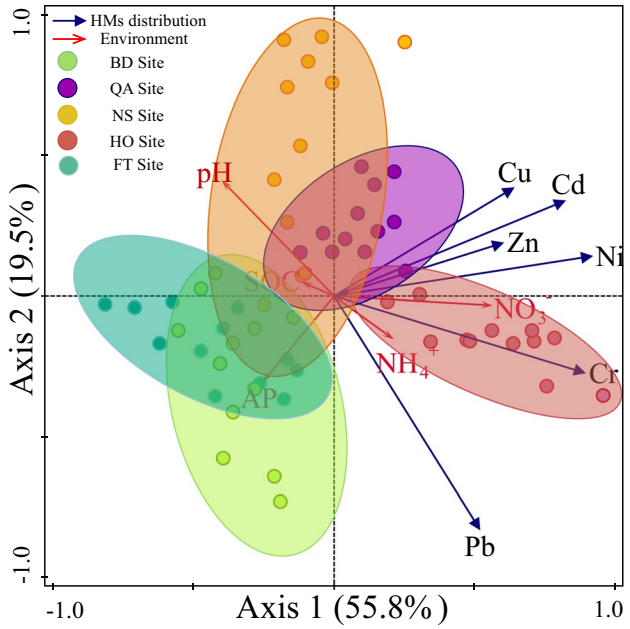


**Fig. 5** The results of redundancy analysis (RDA) between physicochemical properties and total HMs in mangrove sediments collected from BD, QA, NS, HO, and FT sites in the GBA

indicated that the distribution of exchangeable Cd in mangrove sediments was similar to that in carbonate fraction, resulting in higher RAC values in the GBA.

## Discussions

### Heavy metals fractions in different mangrove sediments

Comparatively, Cd in sediments exhibited relatively higher percentages of exchangeable and carbonate-bound fractions, resulting in a more serious ecological risk of Cd in mangrove wetlands due to their easy migration, transformation and bio-absorption. It is generally agreed that soil chloride ($Cl^-$) availed for Cd leaching from solid soil and promoted its mobility (Weggler et al. 2004) due to the complexation effect between two ions (Hirsch et al. 1989). In this study, the salinity values of surface water of these sites were 2.3%, 1.3%, 0.2%, 0.6%, and 2.0% in BD, QA, NS, HO, and FT, respectively, in which the exchangeable Cd proportion at QA, NS, HO, and FT sites was significantly positive correlated with salinity. In addition, Cd had a specific affinity for carbonates and might participate with carbonate minerals under alkaline condition, resulting in a high proportion of carbonated Cd (Ahdy and Youssef 2011). This study also implied that sediments pH in the GBA presented neutrally to weakly alkaline conditions (Table 1), which rationalized a high amount of carbonate-bound Cd.

The distribution of Cr, Ni, and Zn fractions in GBA mangrove sediments was mainly concentrated in residual fraction, derived from natural weathering and their compounds with silicate, leading to a low ecological risk (Ngiam and Lim 2001; Xiao et al. 2015). Sediments Cu exhibited a higher proportion in organic-bound fraction, which is consistent with previous study (Esmaeilzadeh et al. 2016). Organic matter in sediments serves as the main ligand for Cu complexation and avails for the formation of organic-bound Cu (Krupadam et al. 2007). However, the change of redox potential in sediments could drive its release, causing an adverse effect to mangrove wetland (Wu et al. 2016). The highest proportion of organic-bound Cu was found in HO and FT sites, resulting from the frequently withered leaves with Cu replaced Mg in chlorophyll during the first several years after mangrove planting (Wang et al. 2018). Specifically, Pb showed a higher percentage in Fe–Mn oxide, which can be explained by its coprecipitation with Fe and Mn (Li et al. 2014). What is more, the higher concentration of Fe

**Table 3** Spearman correlation analysis among total contents of HMs and physicochemical properties of mangrove sediments in the GBA ($n=60$)

| | pH | $NH_4^+$-N | $NO_3^-$-N | SOC | Available P | Cu | Zn | Cd | Cr | Pb | Ni |
|---|---|---|---|---|---|---|---|---|---|---|---|
| pH | 1.000 | | | | | | | | | | |
| $NH_4^+$-N | 0.135 | 1.000 | | | | | | | | | |
| $NO_3^-$-N | −0.227 | 0.222 | 1.000 | | | | | | | | |
| SOC | −0.257 | 0.010 | −0.155 | 1.000 | | | | | | | |
| Available P | −0.302 | −0.478** | −0.377 | −0.218 | 1.000 | | | | | | |
| Cu | −0.266 | −0.051 | 0.116 | −0.225 | 0.110 | 1.000 | | | | | |
| Zn | −0.104 | 0.150* | **0.441*** | −0.040 | −0.394 | 0.151 | 1.000 | | | | |
| Cd | −0.241 | 0.017 | **0.678**** | −0.267 | −0.252 | **0.655**** | **0.437*** | 1.000 | | | |
| Cr | −0.502** | 0.207 | **0.371*** | −0.350 | −0.105 | **0.551**** | **0.506**** | **0.594**** | 1.000 | | |
| Pb | −0.533** | 0.206 | **0.439*** | −0.237 | 0.143 | 0.163 | 0.076 | **0.372*** | **0.677**** | 1.000 | |
| Ni | −0.230 | 0.155 | **0.367*** | −0.222 | −0.277 | **0.466**** | 0.299 | **0.625**** | **0.547**** | 0.325 | 1.000 |

a. * Significant correlation at $p<0.05$; ** Significant correlation at $p<0.01$. b. The marks in bond represent a significant correlation among $NO_3^-$-N and heavy metals in mangrove sediments ($p<0.05$)

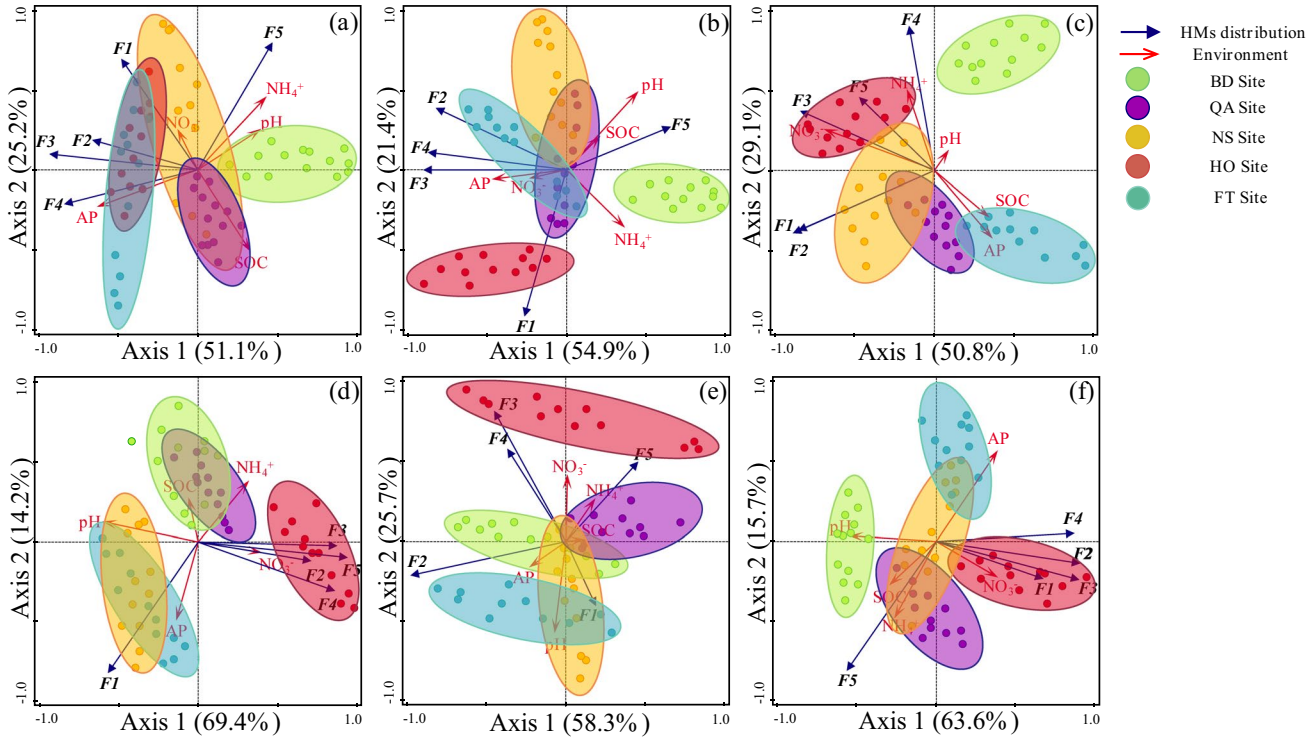


**Fig. 6** The results of redundancy analysis (RDA) between HMs concentrations in different five geochemical fractions and physicochemical properties in mangrove sediments collected from BD, QA, NS, HO, and FT sites in the PRE. Panels (**a**), (**b**), (**c**), (**d**), (**e**), and (**f**) represent Cu, Zn, Cd, Cr, Pb, and Ni, respectively

and Mn (Fe: 2.4–4.9%, Mn: 278.9–1101.2 mg/kg) in the GBA was beneficial for its formation in Fe–Mn oxide.

## Effects of sediment physicochemical properties on heavy metals distribution

It is agreed that environmental conditions should be responsible for the changes in bioavailability and mobility of HMs in sediments, whereas the interactive effects between mangroves and their surrounding environment were more intensive due to the frequent transitions of land-salt water and long immersion period of mangroves in the GBA (Bartoli et al. 2012; Tam and Wong 1995), which contributes to a more complex sediment environment. For instance, the distribution of Cr may be directly impacted by the increase in $Fe_2O_3$ and clay content in sediment (Liu et al. 2015). The increased nutrient levels in the water above will result in increased concentrations of HMs in the sediment (Liu et al. 2022), and the content of HMs in sediments may also significantly varied with different species of mangrove plant (He et al. 2014). In this study, HMs content in sediments was negatively correlated with soil pH value (Fig. 5), indicating that lower pH value availed for the mobilization of HMs (de Souza Machado et al. 2016). RDA results also confirmed that pH was negatively correlated with exchangeable fractions of HMs, except for Cu and Cr (Fig. 6), while the residual and organic-bound states of Cu, Zn, and Cd in sediments presented a positive correlation with sediment pH. This demonstrated that HMs adsorption capacity of sediment gradually weakened with increasing pH value, thus affecting the distribution of HMs geochemical fractions in sediments (Bang and Hesterberg 2004).

Spearman correlation analysis indicated that sediment nitrogen content was significantly related to HMs, particularly for $NO_3^-$-N concentration (Table 3; $p < 0.05$), which could be explained by the anthropogenic sources of HMs and $NO_3^-$-N. Relevant research has demonstrated that HMs are typically derived from domestic and industrial sewage through the source tracing analysis in mangrove sediments (Liu et al. 2014). Nowadays, the study focused on the sewage treatment plant further found that $NO_3^-$-N showed a significant correlation with HMs in effluent (Qiao et al. 2022). Considering the developed industrial level of the GBA, it seems reasonable that $NO_3^-$-N showed a significant correlation with HMs in these five mangrove sediments. In addition, HMs content in Fe–Mn oxide fractions was strongly correlated with $NO_3^-$-N, especially in the QA, NS, and HO sites, which supports the former

**Table 4** The linear regression of HMs in Fe–Mn oxide fraction against to $NO_3^-$-N in mangrove sediments of BD, QA, NS, HO, and FT in the GBA

| Site types | Metal-types | Regression equation | Adj $R^2$ | $F$ value |
|---|---|---|---|---|
| BD | Cu-F3 | $Y=-2.7497X+7.4618$ | 0.0469 | 1.6895 |
| | Zn-F3 | $Y=-7.5754X+34.6767$ | 0.1220 | 2.9457 |
| | Cd-F3 | $Y=0.0073X+0.0108$ | −0.4864 | 0.3508 |
| | Cr-F3 | $Y=1.3131X+15.8519$ | −0.1168 | 0.0583 |
| | Pb-F3 | $Y=38.1183X-57.3740$ | 0.0492 | 2.2116 |
| | Ni-F3 | $Y=-1.6035X+6.9666$ | −0.0293 | 0.5705 |
| QA | Cu-F3 | $\boldsymbol{Y=-1.5257X+7.8364}$ | **0.6433** | **17.2379**$^{**}$ |
| | Zn-F3 | $\boldsymbol{Y=-5.6569X+98.0280}$ | **0.4730** | **9.0797**$^{*}$ |
| | Cd-F3 | $\boldsymbol{Y=-0.0272X+0.0941}$ | **0.4231** | **7.6027**$^{*}$ |
| | Cr-F3 | $Y=4.1053X+18.3021$ | −0.0587 | 0.2226 |
| | Pb-F3 | $Y=-2.6740X+25.4184$ | −0.0531 | 0.2944 |
| | Ni-F3 | $\boldsymbol{Y=-1.7393X+13.0415}$ | **0.2397** | **5.4153**$^{*}$ |
| NS | Cu-F3 | $\boldsymbol{Y=-1.8076X+12.7328}$ | **0.2603** | **6.3862**$^{*}$ |
| | Zn-F3 | $\boldsymbol{Y=31.5373X-10.3138}$ | **0.4296** | **11.5448**$^{**}$ |
| | Cd-F3 | $Y=-0.1368X+0.5276$ | −0.0231 | 0.6830 |
| | Cr-F3 | $Y=5.7720X+2.1230$ | −0.0749 | 0.0244 |
| | Pb-F3 | $Y=19.0438X-39.4772$ | −0.0455 | 0.3897 |
| | Ni-F3 | $\boldsymbol{Y=5.0341X-4.8843}$ | **0.2640** | **6.7468**$^{*}$ |
| HO | Cu-F3 | $\boldsymbol{Y=-0.1584X+8.5162}$ | **0.3265** | **7.7887**$^{*}$ |
| | Zn-F3 | $\boldsymbol{Y=1.5640X+122.9617}$ | **0.3818** | **9.6496**$^{**}$ |
| | Cd-F3 | $Y=-0.0157X+0.2253$ | 0.0482 | 2.0116 |
| | Cr-F3 | $Y=-0.9562X+38.7867$ | −0.0393 | 0.4705 |
| | Pb-F3 | $\boldsymbol{Y=14.9639X-12.7005}$ | **0.2617** | **6.4669**$^{*}$ |
| | Ni-F3 | $Y=2.1598X+21.6094$ | −0.0093 | 0.8699 |
| FT | Cu-F3 | $\boldsymbol{Y=-8.8846X+18.9624}$ | **0.7456** | **42.0430**$^{***}$ |
| | Zn-F3 | $Y=78.9006X-33.0277$ | 0.0448 | 1.6566 |
| | Cd-F3 | $Y=-0.1273X+0.1598$ | −0.0581 | 0.2306 |
| | Cr-F3 | $Y=29.1721X-17.5863$ | −0.0440 | 0.4099 |
| | Pb-F3 | $Y=72.6319X-68.0773$ | −0.0217 | 0.7026 |
| | Ni-F3 | $Y=26.9721X-25.5607$ | −0.0462 | 0.3806 |

a. $X$ and $Y$ mean the concentration of $NO_3^-$-N and HMs in Fe–Mn oxide fraction, respectively. b. $^{*}$, $^{**}$ and $^{***}$ present the significant correlation at $p<0.05$, $p<0.01$ and $p<0.001$, respectively. c. The marks in bond represent a significant correlation between the concentrations of $NO_3^-$-N and HMs in Fe–Mn oxide fraction in mangrove sediments ($p<0.05$)

hypothesis of this study. Furthermore, relevant investigation found the overlying water with higher N and P levels could contribute to more HMs release into the sediment (Fu et al. 2014). Hence, $NO_3^-$-N content presented a significant positive correlation with the majority of exchangeable HMs, except for Cr and Pb, suggesting that $NO_3^-$-N enrichment could increase the ecological risks of majority HMs to mangrove systems. The reasons may be concluded into (i) sulfur autotrophic denitrification process availed for the production of $SO_4^{2-}$, resulting in the transformation of HMs from stable oxidizable state to their exchangeable fraction (Shao et al. 2009); (ii) the changes of rhizosphere metabolism after nitrogen absorption, inducing the secretion of organic acids and the mobilization of HMs (Wang et al. 2021).

## Human activities and mangrove forest planting years impacted on heavy metals distribution

It is generally agreed that human activities, i.e., sewage discharging and mineral smelting, have a significant influence on the pollution concentrations and geochemical fractions of HMs (Cai et al. 2007; Ogundiran and Osibanjo 2009). Due to the huge differences in social economy and industrial activities in the GBA areas, mangroves in GZ and SZ are greatly exposed to external HMs drain pressure than those in ZH and JM (Feng et al. 2017). Comparatively, HMs in mangrove sediments of BD site were highly concentrated in residual fraction and remained at moderate pollution levels, indicating that those HMs might be more impacted by geological

sources than by industrial activities (Cai et al. 2007; Liang et al. 2022). Notably, a relatively low pollution level was also found in FT, located in the SZ region, which might be attributable to the implementation of water pollution control since 2016, resulting in the surrounding water purification (Huang et al. 2021; Zhou et al. 2021). This confirmed that strict social management strategies could provide the improvement of aquatic environment and decrease the HMs pollution levels in mangrove sediments (Tam and Wong 2000b).

A longer mangrove forest planting years mean that those plants could accumulated more HMs from sediments, resulting in the decrease of HMs in coastal sediments (Feng et al. 2017). However, results of this study were inconsistent with the former reports, which might be caused by (i) the QA, NS, and HO sites were frequently accompanied by a continuous input of HMs with industrial wastewater discharge under national discharge standards. Despite there were no industrial factories distributed in HO site (Wang et al. 2022), the HO is an island in the GBA, which means that the water flow of Pearl River can change the hydraulic condition and cause more HMs from upper reaches to enrich on the surface sediment in this area (Gonçalves et al. 1992); (ii) the re-cultivation of mangrove forests in the FT site has been lasted for over 15 years, whereas the plants in sampling site of FT was reclaimed in recent 3 years; (iii) aquatic salinity varied with sampling sites, resulting in a significant impact on the absorption and distribution of sediment HMs (Hirsch et al. 1989). Despite there was no clear tendency for the contribution of HMs decrease in the sediments with different planting years of mangrove. Cd in all sediments exhibited a high ecological risk value, indicating that the inflow of exogenous Cd exceeded the absorption capacity of mangroves at length (Feng et al. 2017; Li et al. 2007). Compared to the previous studies (Feng et al. 2017; Li et al. 2007; Tam and Wong 2000a), the highly ecological risk of Cd and Zn in the sediments of mangrove was still found in the GBA. However, the present study also obtained that the ecological risk level of Cu and Pb has been increased and decreased in recent years in the sediments of mangrove of GBA.

## Conclusions

Despite ecological protection project has been performed for mangrove wetland, sediments in the five sampling sites were still seriously contaminated by Cd and moderately contaminated by Cu and Zn in the GBA. Affected by the sediment and overlying water properties, Cd in sediments was highly concentrated in exchangeable and carbonate-bound fractions, resulting in a high RAC value due to their easy migration, transformation, and bio-absorption. A strong correlation between the concentrations of HMs and $NO_3^-$-N was found in mangrove sediments due to the continuous input of HMs and $NO_3^-$-N in the Pearl River. Further analysis demonstrated that pH value, available P, and nitrogen concentration affected the distribution of HMs geochemical fractions in sediments. High concentration of HMs in QA and NS sampling sites was caused by the formerly pollutants discharge, resulting in these sediments still exhibited with a higher HM pollution level after the plant of mangrove for a long period. Therefore, future development of mangrove conversion and restoration should be linked to the water purification management in the GBA.

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**Author contribution** Huan-Zhan Zhou, Hui-Min Jiang, and Ze-Xiang Cai: methodology, investigation. Jun-Feng Wang: writing—review and editing, funding acquisition. Zhi-Min Xu: review and editing, project administration, supervision, funding acquisition. Guan-Hui Tang, Song Ding, Sheng-Teng Liu: review and editing.

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**Data availability** The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

## Declarations

**Competing interests** The authors declare that they have no competing interests.

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