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# Fraction distribution and bioavailability of soil heavy metals under different planting patterns in mangrove restoration wetlands in Jinjiang, Fujian, China

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ABSTRACT

The mangrove restoration wetland project in Jinjiang Estuary, Fujian Province, was started in April 2014, and the wetland was restored using vegetation restoration technology. Prior to restoration, the area was a mix of *Spartina alterniflora* beaches, muddy beaches, and abandoned quarries, which were not environmentally robust. Six species of mangrove plants were used in the wetland, including *Kandelia obovata*, *Aegiceras corniculatum*, *Avicennia marina*, *Acanthus ilicifolius*, *Bruguiera gymnorhiza*, and *Rhizophora stylosa*. The plants were planted according to three mixed patterns and three planting densities. Finally, the whole wetland was divided into 27 plots, and three plant-free control areas were set up in the area close to the vegetation area. In this study, 30 topsoil samples were collected (April 2019), fraction concentrations of heavy metals (Cu, Cr, Zn, and Pb), and their bioavailability and spatial distributions were determined, and the relationships between their fractions and planting patterns were analyzed. The results showed that among the nine planting patterns, the pattern "*A-K-C*, 0.5×0.5 m" was the most different from the other models, and the fraction of most heavy metals obtained the lowest value of soil metal content in this model. The secondary-phase fraction (SPF) of heavy metals, including acid-soluble, reducible, and oxidizable fractions, is considered to be a direct and potentially hazardous fraction to organisms. In this study, Cu, Zn, and Pb had the greatest SPFs among all the metals (35.29, 33.45, and 51.58%, respectively). Compared with the relevant results before restoration, it was found in after five years of mangrove restoration, the SPF of Cu, Cr, Zn and Pb decreased by 41.31, 22.89, 27.06, and 22.13%, respectively, indicating that the direct and potential toxicity of these four elements to the environment decreased. The risk of heavy metal release decreased from medium and high pollution levels to low pollution levels or even no pollution levels. For most metals, the fraction distributions were controlled by clay, silt, pH, and soil organic matter. The research methods and results can provide a theoretical and scientific basis for further study of wetland vegetation, and in addition, aid in selecting feasible restoration methods for further wetland restoration.

## 1. Introduction

The Jinjiang estuary wetland in Fujian Province is located at the junction of the Jinjiang River and the East China Sea. It is not only an important natural ecological resource in Jinjiang, but also an important protected wetland in southern Fujian. However, due to the discharge of upstream industrial and domestic sewage, surrounding aquaculture farms, and battlefield wastewater, the wetland is seriously degraded, and heavy metal pollution is becoming increasingly severe. Therefore, it is of practical significance to repair the wetland (Deng, 2019; Su et al., 2019). Currently, the technology used for wetland remediation mainly includes hydrodynamic remediation, basement remediation, and phytoremediation technologies (Nilsson and Aradottir, 2013; van Proosdij et al., 2010; Weinstein et al., 2019). Among them, phytoremediation technology is applied to seriously damaged or degraded coastal wetlands and includes planting of mangrove plants, which are used as a

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suitable and effective restoration technique (Duarte et al., 2012; Romanach et al., 2018). The Jinjiang Estuary Wetland in Fujian Province was repaired by this type of phytoremediation (planting mangroves).

As major pollutants, heavy metals exist widely in water, atmosphere, and soil environments. Heavy metals have attracted a wide array of attention because of their high biological toxicity, resistance to degradation, and gradual accumulation in Yoshihiko biomass (Rahman and Singh, 2019; Shao et al., 2020; Sundaramanickam et al., 2016). Estuarine wetlands are usually located close to cities, and a large number of heavy metals from watersheds, the atmosphere, and the oceans enter and are stored wetlands, making estuaries a key accumulation area of heavy metals and other pollutants (Cheng et al., 2012). In addition, the influence of vegetation on heavy metals in mangrove wetlands is also impressive; whether through the absorption of heavy metals by plants, or the direct impact of roots and plant litter, each will have a certain impact on the precipitation of heavy metals in the soil (Kamaruzzaman et al., 2011; Li et al., 2018). The total concentration of heavy metals in soil is a useful index for evaluating soil pollution (Zhang et al., 2009). However, the total concentration of heavy metals cannot provide sufficient information about the bioavailability and toxicity of heavy metals (McLaughlin et al., 2000; Nyamangara, 1998; Rodriguez et al., 2009). The mobility of heavy metals, their bioavailability, and related ecological toxicity to plants depend, to a large extent, on their fractions (Dabek-Zlotorzynska et al., 2003; Tessier et al., 1979). In these fractions, the exchangeable forms are considered bioavailable. The bound forms of organic substances related to carbonates, iron, and manganese oxides can also be bioavailable, while most of the residues are not available to plants or microorganisms (De Groot et al., 2013; He et al., 2005; Makela et al., 2012). Therefore, the components of heavy metals must be considered in the study of mangrove restoration of soil heavy metal pollution in wetlands.

Sequential extraction procedures (SEPs) are widely applied to assess heavy metal speciation in soils (Guevara-Riba et al., 2004). These procedures use a series of selective reagents to dissolve the heavy metals of different components, thus making a more realistic estimate of their actual environmental impact (Gleyzes et al., 2002). The SEPs proposed by the European Community Bureau of Reference (BCR) are widely used to analyze different heavy metal fractions (Bhattacharyya et al., 2008; Massolo et al., 2012; Yuan et al., 2016). In mangrove restoration wetlands, the coverage and diversity of vegetation will affect the main productivity of the wetland and affect the distribution of heavy metals in the mangrove wetland (Chen et al., 2018; Ciarkowska, 2017; Usman et al., 2013). Previous studies have shown that planting vegetation affects the physical and chemical properties of soil, as well as the solubility, mobility, and toxicity of heavy metals (Perry and Berkeley, 2009; Zhou et al., 2010). Therefore, it is particularly important to further explore the effects of different planting patterns on heavy metals in mangrove wetlands. In addition, to better study the distribution of heavy metals under different vegetation planting patterns, it is necessary to study the spatial structure of heavy metals and the statistical correlation between different heavy metals (Ciarkowska, 2017).

In China, there are few studies focused on the forms of heavy metals in soil and bioavailability of artificial restoration wetlands, and studies on the relationship between wetland restoration models (including vegetation types and planting density) are even rarer. Therefore, the main objectives of this study were as follows: 1. To analyze the heavy metal content in the soil of a mangrove restoration wetland in Jinjiang, Fujian; 2. To investigate the relationship between different forms of heavy metals in the soil and vegetation restoration model; 3. To explore the spatial distribution and controlling factors of non-heavy metal SPF in the study area; 4. To evaluate the polluted water of heavy metals in the wetland using the land accumulation index and risk assessment system; and 5. To compare the results of this study with the relevant results before the restoration of the wetland. The above research clarifies the environmental significance of wetland restoration projects and provides a theoretical and scientific basis for the development of related wetland restoration projects.

## 2. Materials and methods

### *2.1. Research regional background*

The Xianshicun Estuary Wetland (east of Jinjiang Bridge to Liuyuan Sluice) in the north of Chennai Town on the south bank of the Jinjiang River, Fujian Province, is located in the experimental area of the Quanzhou Bay Estuary Wetland Nature Reserve (24°47–21"- 24 °59-50" N, 118°42–46"- 118 °44-44" E) (Fig. 1A). The area has a typical subtropical marine climate. The annual average temperature is 24.4 °C, the highest temperature at the end of the year was 32 °C, the minimum temperature was 10 °C, and the annual average precipitation is 1095.4 mm. The main rainfall occurred in May and June (Chen et al., 2019). The restoration of mangrove plants in this area is a mixed area of *Spartina alterniflora* beach, muddy beach, and abandoned sand quarry. The beach area is approximately 659.32 m long, and the widest part is approximately 523.80 m. *S. alterniflora* needed to be cleared and arranged prior to experimental procedures, and the total area cleared was 8.4192 $hm^2$. The total area of mangrove restoration was 18.3800 $hm^2$.

### *2.2. Recovery model design*

Before carrying out the wetland restoration vegetation planting, we carried out a survey to better understand the mangrove system in the area, and finally selected six kinds of mangrove plants for wetland restoration, including *Kandelia obovata*, *Aegiceras corniculatum*, *Avicennia marina*, *Acanthus ilicifolius*, *Bruguiera gymnorhiza*, and *Rhizophora stylosa*. When selecting seedlings, *K. obovata* hypocotyl seedlings are required to grow mature, robust, full plants, free from diseases and insect pests, and the terminal buds of hypocotyl should not be broken prior to use. *B. gymnorhiza*, *R. stylos*, *A. corniculatum*, *A. marin*, and *A. ilicifolius* are afforested through the use of container seedlings. Seedlings of these species are required to grow robust and disease-free, up to 30 cm in length and with a seedling age of 1 year. According to the tidal level distribution of these six types of plants, a mixed model was designed: *A. corniculatum* and *A. marina* were distributed in the middle and low tide zones, which were mainly planted in the front tidal flat, while *A. ilicifolius*, *B. gymnorhiza*, and *R. stylosa* were distributed in the middle and high tidal levels. It is mainly planted in the most energy abundant part of the shore, and *K. obovata* was planted in all plots (Fig. 1B). In this field experiment design, we used a two factors (mixed plant type and planting density) three-level random block design. The restored wetland was divided into 27 sample plots, each with an area of about 0.35 ha, and set up three non-plant control areas for a total of 30 sample plots. The labels for the different recovery models are listed in Table 1.

### *2.3. Collection and pretreatment of soil samples*

Sampling was carried out in late April 2019 (the fifth year after the mangrove wetland was restored). The topsoil (0–10 cm) of 30 plots (including three bare land areas without plants) were collected using a PVC tube. Five duplicate soil samples were collected from each sampling site and mixed. All samples were packed in polyethylene bags and immediately sent to the laboratory. Several samples were naturally dried and stored in self-sealing bags with labels, while the rest were stored in cryogenic refrigerators. Part of the dry sediment samples were ground with a mortar, sifted into 20-mesh and 100-mesh sizes, and then stored in self-sealing bags with corresponding labels for experimental analysis of soil physical and chemical properties and heavy metals. Another part of the unground soil sample was used for the particle size analysis.

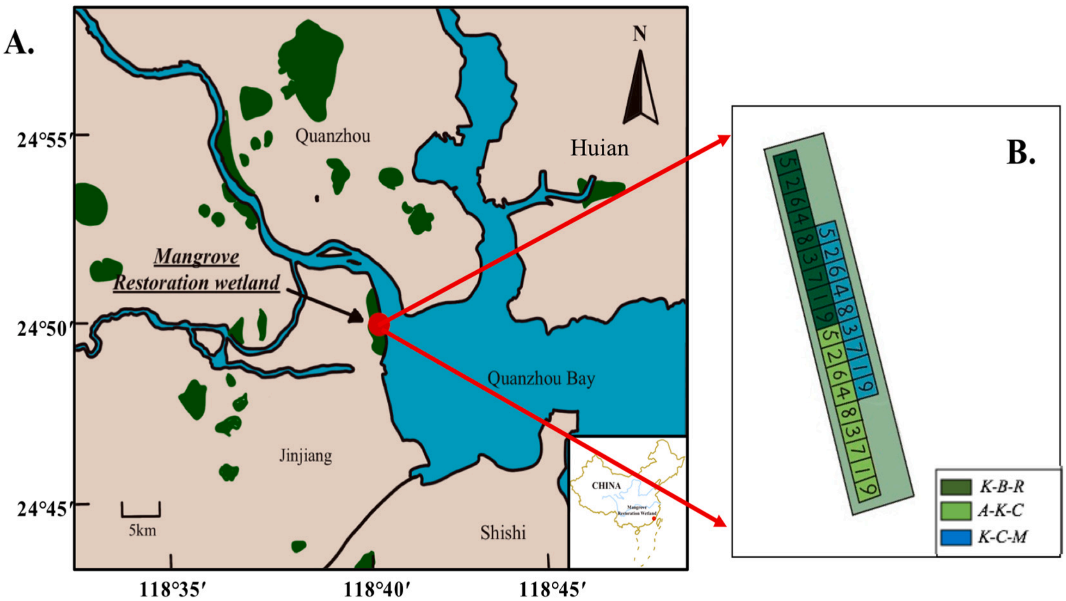


**Fig. 1.** A. Location map of mangrove restoration wetland; B. Location map of each mixed pattern. The planting density of plots 1, 2, 3 is 1.0 × 1.0 m; the planting density of plots 4, 5, 6 is 1.0 × 0.5 m; the planting density of plots 7, 8, 9 is 0.5 × 0.5 m.

**Table 1**
Labels of different recovery patterns.

| Mixed Plant Type | Planting Density | | |
|---|---|---|---|
| | 1.0 × 1.0 m | 1.0 × 0.5 m | 0.5 × 0.5 m |
| *K-B-R* (*Kandelia obovata* + *Bruguiera gymnorhiza* + *Rhizophora stylosa*) | I#1 | I#2 | I#3 |
| *A-K-C* (*Acanthus ilifolius* + *Kandelia obovata* + *Aegiceras corniculatum*) | II#1 | II#2 | II#3 |
| *K-C-M* (*Kandelia obovata* + *Aegiceras corniculatum* + *Avicennia marina*) | III#1 | III#2 | III#3 |

### 2.4. Chemical analysis of soil samples

Soil pH was measured using a pH meter (PB10) in a 1:5 soil/water suspension. The potassium dichromate oxidation method was used to analyze soil organic matter (Morona et al., 2017). The total phosphorus (TP) content of in the soil was determined by the method described by Lu (Lu et al., 2016). Total nitrogen (TN) in the soil was determined by $K_2S_2O_8$ oxidation (Lu et al., 2005). Soil particle size was analyzed using sodium hexametaphosphate as a dispersant and a laser particle size analyzer (Mastersizer 2000) to analyze soil particle size (PS) (Zobeck, 2004).

The BCR SEP was designed to separate heavy metals into four operationally defined fractions: acid-soluble/exchangeable (F1), reducible (F2), oxidizable (F3), and residual fractions (F4) (Quevauviller et al., 1997). The SPF of a specific heavy metal is the sum of the heavy metals F1, F2, and F3. The SPF is generally considered to be a direct and potentially dangerous part of the organism because the binding capacity of F1, F2, and F3 in the soil phase is much lower than that of F4 (Bruder-Hubscher et al., 2002). The BCR sequential extraction program was used for the analysis of metal fractions (Rauret et al., 1999; Wenzel et al., 2001), and the extraction procedure is described in detail in the Supporting Information. Cu, Cr, Zn, and Pb in these samples were analyzed using a 7800 ICP-MS (Agilent Co., Ltd.) at the Analysis and Testing Center of Huaqiao University. The method for determining the total amount of heavy metals in soil is the same as that used to determine the residual state. Quality assurance and quality control were evaluated using two blanks, method blanks, and standard reference materials (GBW07304). The recoveries of the five standard metal samples were between 95 and 105%. Each sample was run twice, and the standard error was always less than 5%.

### 2.5. Pollution level of heavy metals

Using Igeo (Geo-Cumulative, $I_{geo}$) and RAC (Risk Assessment Code, *RAC*), the pollution degree of heavy metals in mangrove restored wetland soil was evaluated (Jain, 2004; Singh et al., 2005). Table 2 shows the classification of various pollution indicators.

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

$$I_{geo} = log_2 \frac{C_j}{1.5C_{bi}} \tag{1}$$

Notes: $C_i$: the measured concentration of heavy metal (mg/kg); $b_i$: the geochemical background value of heavy metal (Cu = 22.4, Cr = 40.7, Zn = 83.60, Pb = 39 mg/kg) (Liu, 1995).

Risk Assessment Code (*RAC*):

$$RAC = \frac{Exchange\ fraction + Water/acid\ soluble}{Total\ metal\ content} \times 100\% \tag{2}$$

Notes: the sum of the exchangeable fraction + water/acid soluble is the weak acid extraction state ($F_1$) in the BCR continuous extraction method of this study.

### 2.6. Data analysis

The one-way analysis of variance (ANOVA) and Duncan multiple

**Table 2**
Classification of various pollution indicators.

| $I_{geo}$ | | *RAC* | |
|---|---|---|---|
| Rank | Pollution level | Rank | Pollution level |
| ≤0 | Practically no pollution | <1% | Practically no pollution |
| 0–1 | Unpolluted to moderately pollution | 1–10% | Low pollution |
| 1–2 | Moderately pollution | 11–30% | Moderately pollution |
| 2–3 | Moderately to heavily pollution | 31–50% | Heavily pollution |
| 3–4 | Heavily pollution | >50% | Extremely pollution |
| 4–5 | Heavily to extremely pollution | | |
| >5 | Extremely pollution | | |

range tests was used to determine the differences of soil characteristics, total amount and form content of heavy metals under different planting patterns of mangrove restoration wetland. Pearson correlation was used to analyze the relationship between the total amount of heavy metals and the forms of heavy metals in soil. The linear regression analysis model was used to explore the relationship between the percentage of second phase fraction (SPF) and soil characteristics. Ward cluster analysis was used to analyze the total amount of heavy metals and SPF under different planting patterns. The data processing and statistical analysis were completed by Excel 2019 and SPSS 25.0, and the charts were drawn by Origin 2018, PowerPoint 2019 and Word 2019.

## 3. Results

### 3.1. Total concentrations of soil heavy metals

Table 3 shows the descriptive statistics of soil clay, silt, sand, pH, SOM, TP, and TN contents of mangrove plants under different planting patterns. The contents of each physical and chemical index in the plant area were significantly different from those in the non-plant control area ($p < 0.05$), in which SOM, TP, and TN in the plant area were significantly higher than those in the non-plant control area. The rising ratios were 17.48, 26.67, 99.99, and 123.83%, respectively. The pH value of soil in the plant area is 6.63–7, which is neutral as a whole. According to the one-way ANOVA analysis, there were significant differences in the contents of TP and TN under different planting patterns in the plant area, and the TP contents were 716.79–1293.23 mg/kg. The TN content was 636.49 to 1056.01 mg/kg. Among all the parameters in Table 3, the changes in SOM, TP, and TN were the largest (coefficient of variation; CV > 15%), while those of silt were the smallest (CV = 3.17%).

Table 4 shows descriptive statistics (Mean, SE, Min, Max and CV) of the total concentrations of heavy metals in all soil samples. The ranges of concentrations ($mg \cdot kg^{-1}$) of the four elements in soil from the plant-covered area were Zn (166.21–231.82) > Pb (61.25–100.34) > Cr (20.88–29.75) > Cu (13.88–21.78). Among all the samples, the CV of Pb was the highest (14.16%), and the maximum/minimum ratio of Pb was 1.64. In comparison, the CV and maximum/minimum ratio of other heavy metals were < 14% and 1.37–1.57, respectively. The concentration of the four elements in the soil of the plant area was significantly higher than that in the soil of the non-plant control area ($p < 0.05$), and the concentration of the four elements in the soil of the plant area increased by 10.96 (Cu), 18.95 (Cr), 13.86 (Zn), and 14.60% (Pb), respectively. The total concentration of Cu and Cr was 11.92% and 38.98%, respectively, which was lower than the background value. In comparison, the total concentration of Zn and Pb was 163.20% and 112.72% higher than this study, respectively. Besides, it is worth noting that the concentrations of the four elements are the lowest in the botanical area II#3 (*A-K-C*, 0.5 × 0.5 m), which were $Cu_{min} = 13.88$ mg/kg, $Cr_{min} = 21.78$ mg/kg, $Zn_{min} = 166.22$ mg/kg, $Pb_{min} = 61.25$ mg/kg, respectively, and the concentrations of these four elements in this model are significantly different from those in other models ($p < 0.05$).

### 3.2. Descriptive analysis of fractions of soil heavy metals

Table 5 shows the descriptive statistical analysis of the forms of heavy metals in soil in mangrove restoration wetlands. The score estimated by BCR SEPs in this study was expressed as a percentage of the total concentration in the soil (per portion, %). The F1 of Cu and Zn were the highest (10.13% and 10.75%, respectively), while the F1 of Cr and Pb was less than 2%. The F2 in Zn and Pb were the highest (12.84% and 22.53%, respectively), while F2 in Cu and Cr were lower (5.41% and 0.65%, respectively). The F3 of all heavy metals ranged from 3.75% to 22.30%, with the highest Zn value and the lowest Cr value. The F4 were the main component of Cu, Cr, Zn, and Pb, accounting for 15.94, 23.24, 144.28, and 36.79%, respectively. It was observed, as shown by the data in Table 5, that the CV of all heavy metals in the F1 was the highest (> 23%), while the CV observed in the F3 was the lowest (< 14%). The CV of different fractions was arranged in the order F1 > F2 > F4 > F3. The CV values of Cr and Pb in the F1 were very high, reaching 54.69% and 41.19%, respectively. The CVs of the F1 in Zn, Cu, and Zn were 23.85%

**Table 3**
Physicochemical properties of surface soil in wetland after 5-years restoration.

| Vegetation Types | Planting Density | Number | Particle Size (%) | | | pH | SOM (mg/kg) | TP (mg/kg) | TN (mg/kg) |
|---|---|---|---|---|---|---|---|---|---|
| | | | Clay | Silt | Sand | | | | |
| | | | <2 μm | 2–20 μm | >20 μm | | | | |
| Control | | | 4.19 ± 0.92a | 77.19 ± 0.56ab | 18.62 ± 0.42ab | 7.23 ± 0.23a | 21.29 ± 7.9ab | 549.7 ± 8.43 a | 388.49 ± 59.14 a |
| *K-B-R* | 1.0 × 1.0 m | I#1 | 5.68 ± 0.56b | 80.31 ± 2.93b | 13.89 ± 3.57b | 6.08 ± 0.17b | 32.99 ± 5.19b | 1176.74 ± 73.45 de | 901.02 ± 82.56 bcd |
| | 1.0 × 0.5 m | I#2 | 5.48 ± 0.14ab | 75.41 ± 6.33ab | 19.1 ± 6.2ab | 6.10 ± 0.14b | 27.31 ± 8.67ab | 716.79 ± 14.43 ab | 851.42 ± 92.03 cd |
| | 0.5 × 0.5 m | I#3 | 5.24 ± 0.63ab | 73.59 ± 3.76a | 21.17 ± 4.23a | 6.34 ± 0.03c | 23.56 ± 0.55ab | 892.89 ± 53.56 bc | 785.28 ± 176.78 df |
| *A-K-C* | 1.0 × 1.0 m | II#1 | 4.67 ± 0.4ab | 76.71 ± 3.54ab | 18.48 ± 3.75ab | 6.64 ± 0.06d | 30.69 ± 2.86b | 1254.68 ± 81 e | 1056.01 ± 84.63 b |
| | 1.0 × 0.5 m | II#2 | 4.81 ± 0.64ab | 80.73 ± 1.8b | 14.46 ± 2.33ab | 6.62 ± 0.14d | 30.12 ± 1.83b | 1241.82 ± 55.76 e | 1008.48 ± 81.31 bc |
| | 0.5 × 0.5 m | II#3 | 4.66 ± 0.35ab | 80.59 ± 1.64b | 14.75 ± 1.4ab | 7.00 ± 0.12e | 28.35 ± 12.95ab | 1184.67 ± 60.53 de | 963.02 ± 94.09 bc |
| *K-C-M* | 1.0 × 1.0 m | III#1 | 4.52 ± 0.28ab | 76.84 ± 1.71ab | 18.64 ± 1.43ab | 7.00 ± 0.03e | 31.13 ± 5.68b | 1293.23 ± 40.53 e | 857.62 ± 18.94 cd |
| | 1.0 × 0.5 m | III#2 | 4.9 ± 0.53ab | 78.29 ± 4ab | 16.8 ± 4.52ab | 6.97 ± 0.05e | 21.57 ± 5.21ab | 1138.46 ± 68.58 de | 766.68 ± 92.58 ef |
| | 0.5 × 0.5 m | III#3 | 4.34 ± 1.63ab | 78.37 ± 1.14ab | 17.26 ± 2.69ab | 6.95 ± 0.06e | 16.99 ± 0.58a | 994.62 ± 97.27 cde | 636.49 ± 69.04 f |
| Mean | | | 4.92 | 77.87 | 17.17 | 6.63 | 26.97 | 1099.32 | 869.56 |
| Standard error | | | 0.45 | 2.47 | 2.44 | 0.38 | 5.24 | 192.60 | 130.51 |
| Min | | | 4.34 | 73.59 | 13.89 | 6.08 | 16.99 | 716.79 | 636.49 |
| Max | | | 5.68 | 80.73 | 21.17 | 7.00 | 32.99 | 1293.23 | 1056.01 |
| Coefficient of variation (%) | | | 9.18 | 3.17 | 14.22 | 5.74 | 19.43 | 17.52 | 15.01 |
| Rate over the control group (%) | | | 17.48 | 0.88 | −7.78 | −8.25 | 26.67 | 99.99 | 123.83 |

Notes: the data in the table is the mean ± standard deviation. Different lowercase letters indicate significant differences in the indicators of different regions at the 0.05 level.

**Table 4**
Distribution characteristics of total heavy metals under different planting patterns (mg/kg).

| Vegetation Types | Planting Density | Number | Cu | Cr | Zn | Pb |
|---|---|---|---|---|---|---|
| Control | | | 17.78 ± 1.01a | 20.88 ± 1.84a | 193.25 ± 14.09a | 72.39 ± 5.13a |
| *K-B-R* | 1.0 × 1.0 m | I#1 | 20.67 ± 0.48bc | 22.1 ± 0.7ab | 228.62 ± 19.74bc | 94.13 ± 3.38de |
| | 1.0 × 0.5 m | I#2 | 21.67 ± 0.86c | 23.42 ± 0.09abc | 245.51 ± 9.04c | 100.34 ± 8.78c |
| | 0.5 × 0.5 m | I#3 | 20.95 ± 0.53bc | 25.88 ± 2d | 229.68 ± 7.96bc | 90.77 ± 4.06 cd |
| *A-K-C* | 1.0 × 1.0 m | II#1 | 21.78 ± 1.86c | 29.75 ± 2.64c | 212.82 ± 7.39ab | 83.39 ± 5.99bc |
| | 1.0 × 0.5 m | II#2 | 21.19 ± 0.83bc | 24.33 ± 1.82bc | 231.82 ± 18.89bc | 86.2 ± 2.75bcd |
| | 0.5 × 0.5 m | II#3 | 13.88 ± 0.77d | 21.78 ± 1.83ab | 166.21 ± 6.39d | 61.25 ± 3.16f |
| *K-C-M* | 1.0 × 1.0 m | III#1 | 19.61 ± 0.97ab | 25.12 ± 0.73b | 219.59 ± 3.77b | 80.05 ± 3.42ab |
| | 1.0 × 0.5 m | III#2 | 19.39 ± 1.03ab | 25.59 ± 1.25b | 215.1 ± 10.39b | 77.04 ± 7.6ab |
| | 0.5 × 0.5 m | III#3 | 18.42 ± 0.95a | 25.56 ± 1.72b | 230.93 ± 11.52bc | 73.48 ± 4.8a |
| Mean | | | 19.73 | 24.84 | 220.03 | 82.96 |
| Standard error | | | 2.46 | 2.38 | 22.49 | 11.74 |
| Min | | | 13.88 | 21.78 | 166.21 | 61.25 |
| Max | | | 21.78 | 29.75 | 245.51 | 100.34 |
| Coefficient of variation (%) | | | 12.49% | 9.60% | 10.22% | 14.16% |
| Local background[a] | | | 22.4 | 40.7 | 83.6 | 39 |
| Rate over local background (%) | | | −11.92% | −38.98% | 163.20% | 112.72% |
| Rate over the control group (%) | | | 10.96 | 18.95 | 13.86 | 14.60 |

Notes: the data in the table is the mean ± standard deviation. Different lowercase letters indicate significant differences in the indicators of different regions at the 0.05 level.
[a] The background values of heavy metal concentration in coastal soils of Fujian Province(Liu, 1995).

**Table 5**
Statistical description of different fractions of soil heavy metals in mangrove restoration wetland.

| | Cu | Cr | Zn | Pb |
|---|---|---|---|---|
| Acid solution ($F_1$) | | | | |
| Mean (mg/kg) | 2.02 | 0.08 | 24.02 | 1.41 |
| Standard error | 0.48 | 0.04 | 6.24 | 0.58 |
| Min (mg/kg) | 1.03 | 0.00 | 9.45 | 0.51 |
| Max (mg/kg) | 2.63 | 0.12 | 32.29 | 2.24 |
| Coefficient of variation (%) | 23.85 | 54.69 | 25.98 | 41.19 |
| Per portion (%) | 10.13 | 0.30 | 10.75 | 1.68 |
| Reducible ($F_2$) | | | | |
| Mean (mg/kg) | 1.10 | 0.16 | 28.45 | 18.61 |
| Standard error | 0.52 | 0.07 | 5.59 | 2.97 |
| Min (mg/kg) | 0.16 | 0.01 | 15.13 | 11.33 |
| Max (mg/kg) | 1.66 | 0.25 | 34.14 | 21.21 |
| Coefficient of variation (%) | 47.64 | 46.29 | 19.65 | 15.95 |
| Per portion (%) | 5.41 | 0.65 | 12.84 | 22.53 |
| Oxidizable ($F_3$) | | | | |
| Mean (mg/kg) | 5.53 | 5.26 | 20.77 | 19.41 |
| Standard error | 0.53 | 0.71 | 1.77 | 2.72 |
| Min (mg/kg) | 4.47 | 3.75 | 16.50 | 15.15 |
| Max (mg/kg) | 6.31 | 6.11 | 22.30 | 22.30 |
| Coefficient of variation (%) | 9.63 | 13.45 | 8.52 | 13.99 |
| Per portion (%) | 28.18 | 21.22 | 9.47 | 23.50 |
| Residual ($F_4$) | | | | |
| Mean (mg/kg) | 15.94 | 23.24 | 144.28 | 36.79 |
| Standard error | 2.68 | 1.83 | 12.75 | 2.93 |
| Min (mg/kg) | 9.34 | 20.40 | 118.83 | 31.80 |
| Max (mg/kg) | 18.05 | 26.38 | 159.78 | 41.49 |
| Coefficient of variation (%) | 16.80 | 7.87 | 8.84 | 7.96 |
| Per portion (%) | 80.33 | 93.96 | 65.89 | 44.95 |
| Secondary-phase ($F_1 + F_2 + F_3$) | | | | |
| Mean (mg/kg) | 8.64 | 5.50 | 73.25 | 39.43 |
| Standard error | 1.25 | 0.73 | 12.38 | 5.19 |
| Min (mg/kg) | 5.66 | 3.93 | 41.07 | 27.23 |
| Max (mg/kg) | 10.14 | 6.30 | 81.90 | 43.95 |
| Coefficient of variation (%) | 14.42 | 13.30 | 16.90 | 13.18 |
| Per portion (%) | 35.29 | 19.10 | 33.45 | 51.58 |

and 25.98%, respectively. The CV of the F2 in Cr, Cu, and Cr was also very high, reaching 47.64% and 46.29%, respectively. The CVs of the F3 and F4 in all remaining elements were lower (< 20%), and the CV of the F3 in Cu, Zn, and the CV of the F4 in Cr, Zn, and Pb were the lowest (< 10%).

### 3.3. Distribution of heavy metals in soils of different planting patterns in mangrove restoration wetlands

Fig. 2 shows the partial characteristics and difference analysis results of heavy metal forms in soil under different planting patterns in mangrove restoration wetlands (different lowercase letters in the figure represent significant differences among different mixed vegetation types, $p < 0.05$). Through the difference analysis, we know that the contents of all elements are different under different planting patterns, and also different under different planting densities (some are significant, some are not significant). We observed that the maximum values of the F1 of the four elements in the soil with planting pattern I#, in which the F1 maximum values of Cu and Zn appeared in the planting density of I#2, while the F1 maximum values of Cr and Pb appeared in the planting densities of I# 3 and I#1, respectively. The minimum F1 values of Cu, Cr, and Zn were observed in II#3, and the F1 content in this region was significantly different from that of other planting patterns ($p < 0.05$), while the F1 minimum value of Pb appeared in the planting pattern of II#2. There were significant differences in the F2 concentrations of Cu, Cr, Zn, and Pb under different planting patterns and planting densities. The F2 of Cu and Zn was the largest in II#2 and the smallest in II#3, and the difference between the two densities was significant ($p < 0.05$). The smallest Cr (F2) was found in II#3 and the largest in II#2, the largest of Pb (F2) was in II#3, and the smallest in II#3. We observed that the F3 and F4 of the four elements were the smallest in II#3. The F3 of Cu, Cr, and Zn was the largest in II#3, F3 of Pb was the largest in II#2, F4 of Cu and Cr was the largest in II#2, and the F4 of Zn and Pb was the largest in II#3, and there were significant differences among different planting patterns. It can be concluded that *K-B-R*, *A-K-C*, and low-density planting patterns have the greatest influence on the distribution of heavy metals.

Overall, whether in the non-plant control area or the plant area, the direct and potential ecological toxicity scores of Pb were the highest among the four elements. The direct and potential ecological toxicity scores of Cu and Zn were the second highest, and the direct and potential ecological scores of Cr were the lowest. The proportions of the four parts of the four heavy metal elements were different. For example, the morphological ratio of Cu and Cr to Pb was F4 > F3 > F2 > F1, while that of Zn was F4 > F2 > F1 > F3.

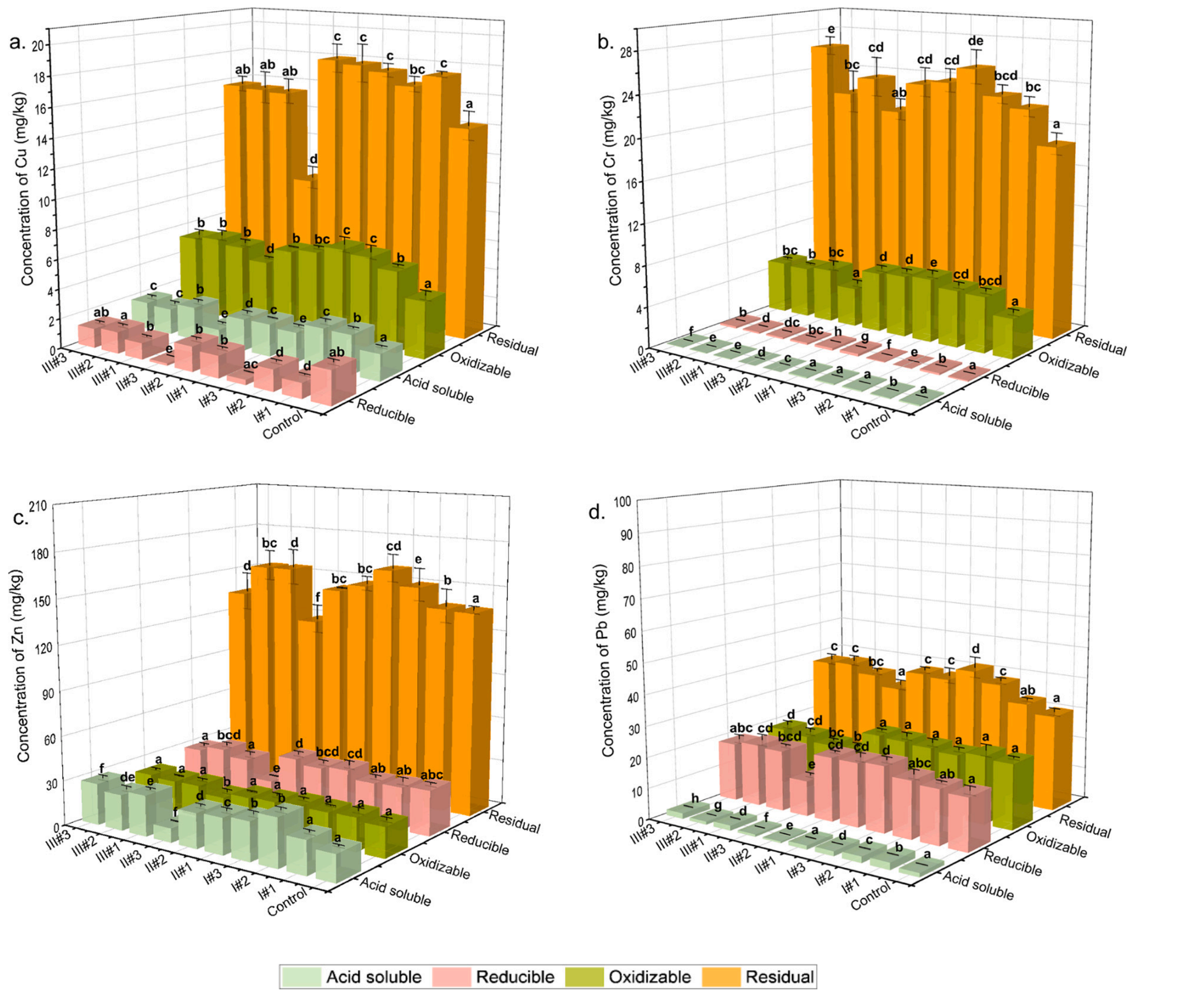


**Fig. 2.** Distribution characteristics of heavy metal fractions under different planting patterns in mangrove restoration wetlands. Different lowercase letters indicate significant differences between different mixed vegetation types ($p < 0.05$). a. Cu; b. Cr; c. Zn; d.Pb.

### 3.4. Effects of soil properties on the distribution of heavy metals under different planting patterns

The migration and immobilization of heavy metals in soils may be affected by a variety of soil characteristics. Therefore, we carried out a linear stepwise regression of the percentage of the SPF of heavy metals to evaluate the effects of soil properties on the availability of heavy metals in mangrove restoration wetlands with different planting patterns (Table 6). Among the three planting patterns, the soil properties in *K-C-M* did not affect the SPF percentage of the four elements. Still, the SPF percentage of Cu in *A-K-C* was affected by TP, while the SPF percentage of Cr was affected by clay in *K-B-R* and silt in *A-K-C*. In addition, pH in *K-B-R* and *A-K-C* had significant effects on the percentage of SPF in Zn ($p < 0.05$). However, the effects on Pb were as follows: both the SOM in *K-B-R* and the pH in *A-K-C* had significant effects on the SPF percentage of Pb, while the soil properties of *K-C-M* did not affect the SPF percentage of Pb.

**Table 6**
Linear regression models between secondary-phase fraction percentage of heavy metals and soil properties in the three planting models of mangrove restoration wetland.

| Planting models | Model | R | F | Sig. |
|---|---|---|---|---|
| Cu | | | | |
| *K-B-R* (I#) | – | – | – | – |
| *A-K-C* (II#) | $y = 5.302–0.032 \times {}_5–0.002 \times {}_6$ | 0.920 | 16.554 | 0.004 |
| *K-C-M* (II#) | – | – | – | – |
| Cr | | | | |
| *K-B-R* (I#) | $y = 35.970–0.013 \times {}_1$ | 0.799 | 12.355 | 0.010 |
| *A-K-C* (II#) | $y = 3.499 + 0.017 \times {}_2$ | 0.899 | 26.487 | 0.001 |
| *K-C-M* (II#) | – | – | – | – |
| Zn | | | | |
| *K-B-R* (I#) | $y = -34.960 + 11.120 \times {}_4$ | 0.877 | 23.289 | 0.002 |
| *A-K-C* (II#) | $y = 165.273–19.824 \times {}_4$ | 0.771 | 10.259 | 0.025 |
| *K-C-M* (II#) | – | – | – | – |
| Pb | | | | |
| *K-B-R* (I#) | $y = 32.802 + 0.419 \times {}_5$ | 0.824 | 14.769 | 0.006 |
| *A-K-C* (II#) | $y = 155.721–15.789 \times {}_4$ | 0.724 | 7.693 | 0.028 |
| *K-C-M* (II#) | – | – | – | – |

Clay content ($x_1$), Silt content ($x_2$), Sand content ($x_3$), pH ($x_4$), SOM content ($x_5$), TP content ($x_6$) and TN content ($x_7$). For some planting models, regression models were not listed since they did not pass F test at 0.05 level.

### 3.5. Risk assessment of heavy metals under different planting patterns

Fig. 3 (a.) and (b.) show the distribution of soil heavy metal accumulation index ($I_{geo}$) and risk assessment code (*RAC*) under different planting patterns in the fifth year of mangrove restoration, respectively.

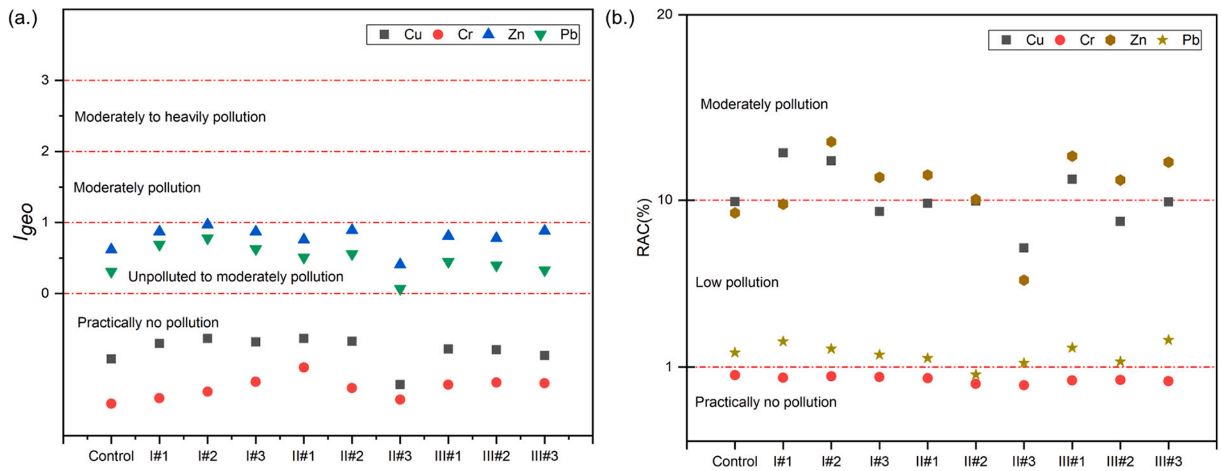


**Fig. 3.** Distribution of the RAC of heavy metals in the soil under different planting patterns in the fifth year of mangrove restoration.

The mangrove restoration wetland in this study is located in the coastal area of the Jinjiang estuary in Fujian Province, and thus, the heavy metal pollution is based on the heavy metal content from the coastal area of Fujian Province. As shown in Fig. 3a, the heavy metal elements Cu and Cr in the soil were at pollution-free levels in each planting pattern. The $I_{geo}$ of Zn was 0.41–0.97, the average value was 0.79, and it was at the level of light pollution in each planting pattern. The $I_{geo}$ of Pb was 0.07–0.78, the average value was 0.47, and it was at the level of light pollution in each planting pattern. The order of the average $I_{geo}$ of each heavy metal was Zn > Pb > Cu > Cr. It is worth noting that the $I_{geo}$ of heavy metal elements Cu, Zn, and Pb in II#3 was lower than that in the non-plant control area.

As shown in Fig. 3b, the *RAC* values of Cr and Pb were approximately 1%, and Cr was considered to cause almost no pollution, but Zn had a high *RAC*, which was generally at a medium pollution level. It is worth noting that both Cu, Cr, and Zn in II#3 reached the lowest *RAC* values

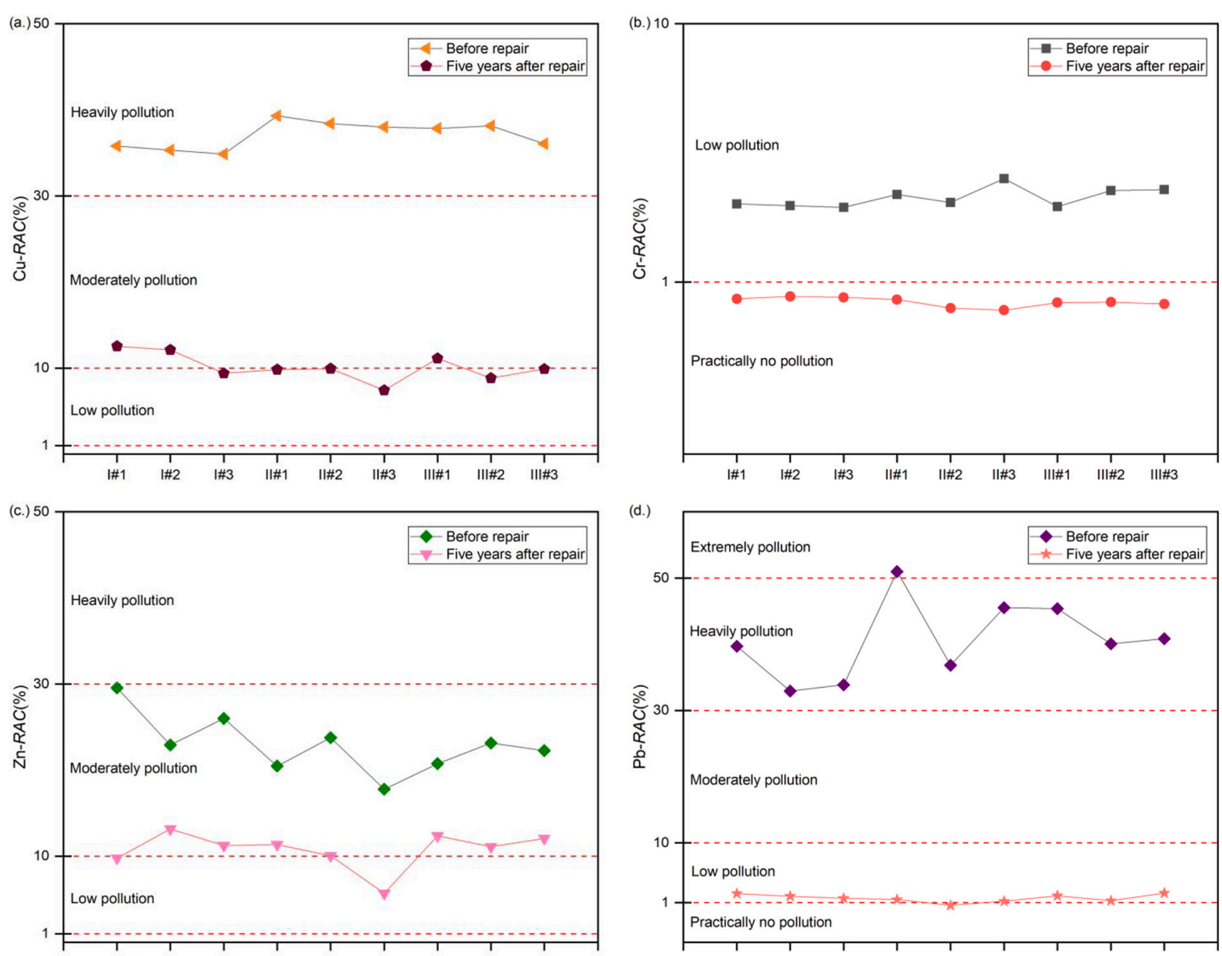


**Fig. 4.** Comparison of Cu, Cr, Zn, And Pb heavy metal elements RAC before and 5 years after mangrove restoration.

(*A-K-C*, 0.5 × 0.5 m), which were 7.42, 0.02, and 5.69%, respectively, and were lower than those in the non-plant control area, decreased by 25.14, 96.62, and 38.92%, respectively, compared with the *RAC* in the non-plant control area. The average *RAC* order of each heavy metal was Zn (*RAC* = 10.75%) > Cu (*RAC* = 10.13%) > Pb (*RAC* = 1.68%) > Cr (*RAC* = 0.30%).

Fig. 4 shows the *RAC* comparison of the four heavy metal elements (Cu, Cr, Zn, and Pb) before and 5 years after mangrove restoration. It was not difficult to find that five years after the restoration of mangrove wetlands, the pollution degree of various heavy metal elements was greatly reduced, which can, to a certain extent, explain that the restoration of mangrove wetlands was beneficial to environmental protection.

## 4. Discussion

### 4.1. Soil physicochemical properties

As shown in Table 3, the soil physicochemical properties of the mangrove restoration wetland were significantly different under different planting patterns in the fifth year. The SOM, TP, and TN in the plant area were significantly higher than those in the non-plant control area, and combined with the correlation analysis, the correlations among SOM, TP, and TN were significant ($p < 0.05$) (Table 7), which was consistent with the study conducted by Zhang et al. (Zhang et al., 2012). While there is a gradual increase in the content of organic matter during plant growth (Machado et al., 2008), studies have also pointed out that the accumulation of SOM in wetland soil is an important way to retain phosphorus (Huang et al., 2015). On the other hand, SOM can provide energy for microbial metabolism and promote the mineralization and stability of soil nitrogen (Curtis et al., 2005), which is important because the SOM, TP, and TN in the plant-covered area are higher than those in the bare area. In addition, the soil in the plant-covered area of the intertidal wetland has a stronger adsorption capacity for nitrogen and phosphorus in the water than the bare soil, which also causes the nitrogen and phosphorus content in the vegetation area to be higher than that in the control area (Hou et al., 2008; Zhang et al., 2015). Furthermore, according to the one-way ANOVA analysis, there were significant differences in the contents of TP and TN under different planting patterns in the plant-covered area. There are differences in vegetation growth, surface cover, chemical composition of root exudates, and root turnover rate under different vegetation restoration models. These factors change the content of soil nutrients, which leads to differences in soil nitrogen and phosphorus content (Li et al., 2013). Pengthamkeerat et al. showed that changes in soil physical properties can lead to changes in soil microhabitats, thus affecting the distribution, activity, and diversity of soil microorganisms, which reflects the cycle, transformation, and utilization of nitrogen in soil ecosystems under different vegetation restoration models (Pengthamkeerati et al., 2011).

### 4.2. Total heavy metals in soil

Table 4 shows the total distribution of the four heavy metals under different planting patterns. The total contents of Zn and Pb were significantly higher than the background values of heavy metals in the coastal areas of Fujian Province (p < 0.05) (Liu, 1995). The total amount of heavy metals in each model in the plant-covered area was significantly higher than that in the control area. This is because the mangrove wetlands have enhanced their ability to intercept water flow due to the growth of vegetation, coupled with the adsorption capacity of soil sediments, which in turn promoted the accumulation of heavy metals in the soil. Studies have highlighted that wetlands are often regarded as "sinks" for metals (Pavlovic et al., 2016; Stead-Dexter and Ward, 2004). In this study, the soil pH value in the plant-covered area was 8.25% lower than that in the non-plant control area (Table 3). Combined with the correlation analysis (Table 7), the pH value was significantly and negatively correlated with Cu, Zn, and Pb ($p < 0.01$); that is, in the soil area with low pH, the heavy metal content was high. This conclusion is consistent with the study by Zeng et al. (Zeng et al., 2011). The difference in the total amount of heavy metals in soil under different planting patterns is significant because there must be differences in the root system and size of different types of mangrove plants, and the enrichment ability of heavy metals is also different, as reported by Zhang et al. (Zhang et al., 2013). In addition, this previous study found that *K. obovata* and *A. corniculatum* stored heavy metals in the following order: Cu > Cr > Pb, whereas in this study, all three planting patterns contained *K. obovata* (or *Kandelia obovate*), and *A. corniculatum*. The content of these three elements in the soil under the three models is opposite to that of Zhang et al. (Zhang et al., 2013), that is, the content of heavy metals in the soil was Pb > Cr > Cu. In addition to the type of vegetation affecting the distribution of heavy metals in soil, the density of vegetation also has an effect. Different planting densities of vegetation lead to different root densities. Roots can absorb heavy metals in soil and store them in plants. The higher the planting density of vegetation, the stronger the concentration of heavy metal ions, which in turn reduces the concentration of heavy metals in soils (Liu et al., 2009; Wang et al., 2009). In this study, the contents of heavy metals from low-density vegetation plots were lower, especially the contents of four elements in II#3 (0.5 × 0.5 m) were significantly different from those in other models ($p < 0.05$), and the contents of these four elements were the lowest in II#3. Combined with the tree-like diagram, we can clearly see that the greatest difference was found between II#3 and the other models (Fig. 5a).

### 4.3. Fractions of soil heavy metals

In general, the F1 have the strongest migration ability, with F2 and F3 being called the non-residual state, mainly from human activities, and the sum of the three is called the SPF of heavy metals, whereas the F4 is difficult to migrate and transform, mainly from natural minerals (Zhang et al., 2017; Zhou et al., 2010). Table 5 shows that the main components of Cu, Cr, and Zn were F4, which was consistent with the results of Zhong et al. (Zhong et al., 2011), while the main component of Pb was F4. It is worth noting that, combined with the relevant data before wetland restoration in this study (Table 8) (Su et al., 2019), the F1 of Cu, Cr, Zn, and Pb decreased by 72.70, 92.35, 53.18, and 95.88%, respectively, after 5 years of restoration (April 2019). The SPF of Cu, Cr,

**Table 7**
Correlation analysis between soil physicochemical properties and total heavy metals.

| | Clay | Silt | Sand | pH | SOM | TP | TN | Cu | Cr | Zn | Pb |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Clay | 1 | 0.103 | −0.291 | −0.539** | −0.056 | −0.334 | −0.098 | 0.353 | −0.108 | 0.331 | 0.552** |
| Silt | | 1 | −0.981** | 0.160 | −0.493** | −0.106 | 0.012 | −0.154 | −0.126 | −0.143 | −0.177 |
| Sand | | | 1 | −0.046 | 0.480* | 0.172 | 0.003 | 0.072 | 0.133 | 0.074 | 0.064 |
| pH | | | | 1 | 0.120 | 0.517* | −0.063 | −0.561** | 0.173 | −0.564** | −0.799** |
| SOM | | | | | 1 | 0.407* | −0.147* | 0.335 | 0.608** | 0.235 | 0.118 |
| TP | | | | | | 1 | 0.018 | 0.158 | 0.446* | 0.199 | −0.179 |
| TN | | | | | | | 1 | 0.004 | 0.176 | −0.034 | −0.067 |

* Correlation is significant at the 0.05 level (2-tailed), n = 30.
** Correlation is significant at the 0.01 level (2-tailed), $n$ = 30.

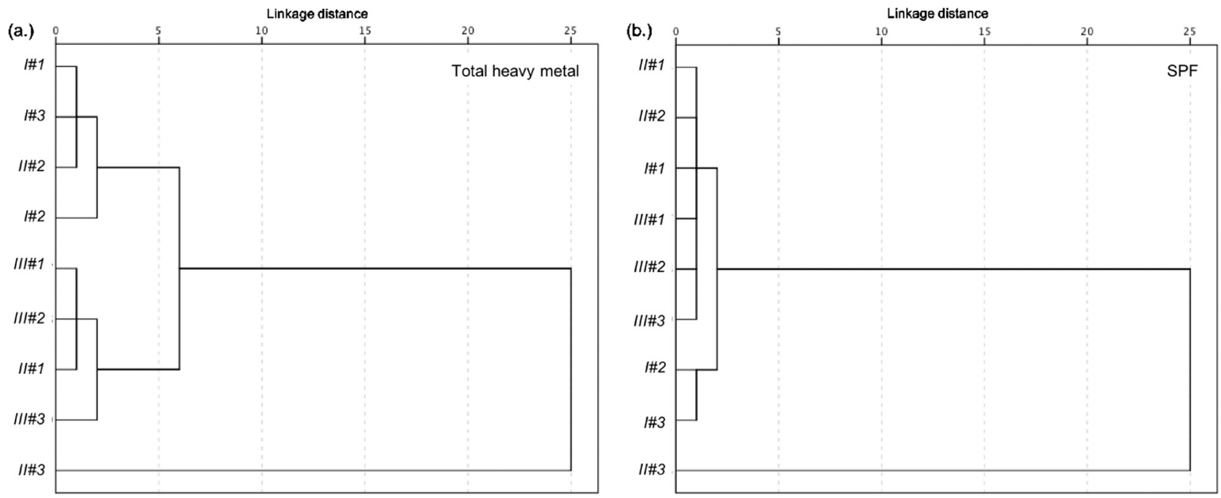


**Fig. 5.** Tree-like diagram. Agglomeration according to Ward method, the total amount of heavy metals and SPF in the soil were respectively classified and aggregated under each planting mode. (a.): Total heavy metals; (b.): SPF.

**Table 8**
Comparative analysis of the fractions of heavy metal elements before and after wetland restoration.

| | Cu | Cr | Zn | Pb |
|---|---|---|---|---|
| Acid solution ($F_1$) | | | | |
| Before wetland restoration (%) | 37.09 | 3.94 | 22.97 | 40.69 |
| 5 years after the restoration of the wetland (%) | 10.13 | 0.30 | 10.75 | 1.68 |
| Rate over the Before wetland restoration (%) | −72.70 | −92.35 | −53.18 | −95.88 |
| Reducible ($F_2$) | | | | |
| Before wetland restoration (%) | 18.53 | 10.31 | 13.61 | 19.06 |
| 5 years after the restoration of the wetland (%) | 5.41 | 0.65 | 12.84 | 22.53 |
| Rate over the Before wetland restoration (%) | −70.82 | −93.73 | −5.60 | 18.22 |
| Oxidizable ($F_3$) | | | | |
| Before wetland restoration (%) | 18.87 | 14.50 | 8.76 | 1.52 |
| 5 years after the restoration of the wetland (%) | 28.18 | 21.22 | 9.47 | 23.50 |
| Rate over the Before wetland restoration (%) | 49.38 | 46.35 | 8.11 | 1444.66 |
| Residual ($F_4$) | | | | |
| Before wetland restoration (%) | 25.51 | 71.25 | 54.67 | 38.74 |
| 5 years after the restoration of the wetland (%) | 80.33 | 93.96 | 65.89 | 44.95 |
| Rate over the Before wetland restoration (%) | 214.85 | 31.88 | 20.52 | 16.04 |
| Secondary-phase ($F_1 + F_2 + F_3$) | | | | |
| Before wetland restoration (%) | 377.54 | 115.77 | 20.60 | 73.85 |
| 5 years after the restoration of the wetland (%) | 221.58 | 89.27 | 15.03 | 57.50 |
| Rate over the Before wetland restoration (%) | −41.31 | −22.89 | −27.06 | −22.13 |

Zn, and Pb decreased by 41.31, 22.89, 27.06, and 22.13%, respectively, after 5 years of recovery (April 2019) compared with that before recovery (April 2014). The results showed that after 5 years of restoration, the bioavailability of Cr, Cu, Pb, and Zn had decreased, and the direct and potential toxicity to the environment was low (Wang et al., 2009). According to the analysis of the CV of various forms of heavy metals, we found that the CV of the F1 and F2 of all elements was between 15% and 55%, while the CV of the F3 and F4 were between 7.8% and 16.8%, indicating that the heavy metals the F1 and F2 in our study area had greater spatial variability than the F3 and F4 (Zhong et al., 2011). The non-residual states of Cu, the F2 of Cr, the F1 and F3 of Zn and the F1, F3, and F4 of Pb were not correlated with the total concentration of Cr. In addition, there was a significant correlation between the total concentration of heavy metals in most soils and different components ($p < 0.05$) (Table 9), which indicated that the total concentration of Cr had no significant effect on the forms of Cu, Cr, Zn, and Pb. Among them, it is worth noting that for Zn, the correlation coefficients of the F1, F2 and F3, which with Cu, Zn and Pb are generally higher than those of F4, indicates that the "activity" and "potential activity" of Zn in the study area are affected by the concentration of total Cu, Zn and Pb.

As with the total amount of heavy metals, the content distribution of various forms is closely related to the planting pattern of vegetation. As can be seen from Fig. 2, the contents of different forms of the four elements are significantly different under different planting patterns, and the location of each morphological extreme value of each element is also different. This is because there are some differences in the growth of each plant, the absorption rate of heavy metals, and the storage and secretion of heavy metals; therefore, there are also differences in the morphological distribution of metal elements under different vegetation planting patterns (Verbruggen et al., 2009; Xiao et al., 2015). The results showed that iron oxides and hydroxides in the rhizosphere of mangrove plants had strong adsorption capacity for heavy metals, but different planting patterns showed different adsorption capacity for heavy metals (Marchand et al., 2010). However, it is worth noting that the contents of various forms of heavy metals in soils under model II#3 were relatively low, such as the F1 of Cu, Cr, and Zn; F2 of Cu, Zn, and Pb; and the F3 and F4 of Cu, Cr, Zn, and Pb being the lowest in the soil under model II#3, which is consistent with the content distribution of total heavy metals in different models in this study. For the analysis of the maximum values of various forms, we found that most of them appeared in the low-density area in I#. The results show that the low-density area under mode I# and II# had the greatest influence on the speciation of heavy metals, and combined with the tree-map analysis, we can clearly see that II#3 is the most different from the other models (Fig. 5b). There are various reasons for this result, but it is certain that it is related to the ability of vegetation to absorb heavy metals in soil. The external environment (such as external input) also has a great influence on it (Cheng et al., 2012; Deng et al., 2019). The F1 is the most active form among the four forms, and it is easy for plants to absorb and utilize (Weis and Weis, 2004), so the content of F1 in the soil is often measured at a low level (Fig. 4). In addition, mixed cropping can affect the distribution of the F2 and F3, and the contents of F2 and F3 vary significantly among different cropping patterns, which may be due to the fact that the F1 of heavy metals cannot meet the growing needs of vegetation, while the potential forms in which F2 and F3 can be released and transformed lead to the redistribution of these two forms (Liu et al., 2018; Wang et al., 2009). Planting density affects the distribution of heavy gold forms in soil,

**Table 9**
Correlation between the total concentration of soil heavy metals and their fractions.

| Total heavy metal | Cu | | | | Cr | | | |
|---|---|---|---|---|---|---|---|---|
| | $F_1$ | $F_2$ | $F_3$ | $F_4$ | $F_1$ | $F_2$ | $F_3$ | $F_4$ |
| Cu | 0.822** | 0.565** | 0.734** | 0.897** | 0.683** | −0.147 | 0.797** | 0.284 |
| Cr | 0.080 | 0.338 | 0.374 | 0.401* | 0.415* | 0.242 | 0.462* | 0.379 |
| Zn | 0.761** | 0.517** | 0.586** | 0.777** | 0.543** | −0.326 | 0.649** | 0.500** |
| Pb | 0.831** | 0.309 | 0.633** | 0.729** | 0.672** | −0.453* | 0.679** | 0.165 |
| | Zn | | | | Pb | | | |
| Total heavy metal | $F_1$ | $F_2$ | $F_3$ | $F_4$ | $F_1$ | $F_2$ | $F_3$ | $F_4$ |
| Cu | 0.714** | 0.812** | 0.655** | 0.592** | 0.326 | 0.817** | 0.735** | 0.518** |
| Cr | 0.283 | 0.446* | 0.276 | 0.370 | −0.092 | 0.584** | 0.225 | 0.357 |
| Zn | 0.837** | 0.659** | 0.749** | 0.457* | 0.478* | 0.647** | 0.559** | 0.508** |
| Pb | 0.637** | 0.603** | 0.581** | 0.543** | 0.509** | 0.530** | 0.680** | 0.444* |

Acid solution ($F_1$); Reducible ($F_2$); Oxidizable ($F_3$); Residual ($F_4$); Secondary-phase ($F_1 + F_2 + F_3$).
* Correlation is significant at the 0.05 level (2-tailed), n = 30.
** Correlation is significant at the 0.01 level (2-tailed), n = 30.

mainly because planting density affects the aboveground and underground biomass and the number of plant roots, thereby affecting the morphological distribution of heavy metals (Ferreira et al., 2015). Therefore, the "*A-K-C*, 0.5×0.5 m" model may be applied on a larger scale in Jinjiang wetland restoration, although this system requires further exploration and experimentation.

In analyzing the effects of soil characteristics on the speciation and distribution of heavy metals (Table 6), we found that clay, silt, pH, and SOM were the controlling factors of the SPF of most heavy metals in various planting patterns. Among the fine particles separated from the surface area (sand, silt, and clay), the concentration of heavy metals is higher due to the increase in surface area, the increased content of clay minerals and SOM, and the presence of iron-manganese oxides and sulfides (Banerjee et al., 2017; Luo et al., 2011). In addition, vegetation can release oxygen, oxidize rhizosphere soil, and affect soil redox potential and pH (Weis and Weis, 2004). The reduced state of heavy metals mainly binds to iron, manganese oxides, and hydroxides, and is easily affected by pH and redox potential; the oxidizing state of heavy metals and oxygen release from roots will affect soil pH and redox potential, and oxidize organic matter and other compounds, thus affecting the distribution of reducible and oxidizable forms in soil (Tao et al., 2003). In addition, vegetation can affect the distribution of its residual state by affecting other forms of transformation. The correlation analysis of this study also found that the existing forms of heavy metals in soil can be transformed into each other (Table 10). Most of the forms showed a significant positive correlation ($p < 0.01$).

**Table 10**
Correlation coefficients between the heavy metal fractions in soils.

| | | $F_1$ | $F_2$ | $F_3$ | $F_4$ |
|---|---|---|---|---|---|
| Cu | $F_1$ | 1 | | | |
| | $F_2$ | 0.310 | 1 | | |
| | $F_3$ | 0.515** | −0.251 | 1 | |
| | $F_4$ | 0.750** | 0.335 | 0.620** | 1 |
| Cr | $F_1$ | 1 | | | |
| | $F_2$ | −0.501** | 1 | | |
| | $F_3$ | 0.346 | 0.030 | 1 | |
| | $F_4$ | 0.050 | 0.131 | 0.659** | 1 |
| Zn | $F_1$ | 1 | | | |
| | $F_2$ | 0.053** | 1 | | |
| | $F_3$ | 0.688** | 0.555** | 1 | |
| | $F_4$ | 0.584** | 0.580** | 0.328 | 1 |
| Pb | $F_1$ | 1 | | | |
| | $F_2$ | 0.133 | 1 | | |
| | $F_3$ | 0.148 | 0.463* | 1 | |
| | $F_4$ | 0.022 | 0.668** | 0.218 | 1 |

* Correlation is significant at the 0.05 level (2-tailed), n = 30.
** Correlation is significant at the 0.01 level (2-tailed), n = 30.

### 4.4. Risk assessment of heavy metals

Based on the land accumulation index and pollution risk assessment coding method, the degree of heavy metal pollution in mangrove restoration wetlands was evaluated (Fig. 4), and compared with the relevant results before wetland restoration (Fig. 5). According to the *RAC* guidelines, for any given metal, if the exchangeable carbonate and carbonate content in the soil sample is less than 10% of the total metal concentration, it is considered environmentally safe (Sundaray et al., 2011). Soils with a total metal concentration of 11% in carbonates and exchangeable parts pose a moderate risk to the environment. The release of soils with a total metal concentration of more than 50% in the same location as above must be considered very dangerous, as metals can be expected to easily enter the food chain (El-Said and Youssef, 2013). The *RAC* values of Cr and Pb in all plots in this study were less than 10%, indicating that these two elements are safe in the environment, while the *RAC* values of Cu and Zn elements above 10% in some planting patterns present a certain level of risk to the environment. To a certain extent, it can be explained that different planting patterns have a certain impact on the release risk of metal elements. However, it is worth noting that the study area has been repaired by mangrove plants for five years. Under the restoration of mangrove plants to the wetland for five years, the *RAC* of Cu, Cr, Zn, and Pb remediation decreased significantly, decreasing from medium and high pollution to low pollution or even no pollution. This is because mangrove plants have a strong adsorption capacity for metal elements (Bai et al., 2011), and coupled with the effect of long-term vegetation restoration, it will also promote the development of wetland restoration. The removal effect of heavy metals by long-term wetland reclamation is usually better than short-term (Bai et al., 2014).

## 5. Conclusions

The BCR sequential extraction procedure was used to study the speciation distribution characteristics and release risk of heavy metals in soil under different restoration models after five years of mangrove restoration in Jinjiang, Fujian Province. The conclusions are as follows: 1. Soil properties and heavy metal distributions were significantly different under different planting patterns; 2. Pattern "*A-K-C*, 0.5×0.5 m" was the most different from other models, and the fraction of most heavy metals would obtain the lowest value of soil metal content in this model. It is worthwhile to conduct more related research on this model to excavate its restoration incentives in-depth; 3. After five years of mangrove restoration, the SPF of Cu, Cr, Zn, and Pb decreased by 41.31, 22.89, 27.06, and 22.13%, respectively. The bioavailability of Pb and Zn decreased, and the direct and potential toxicity to the environment decreased; 4. Combined with the linear regression model between the percentage of the SPF of heavy metals and soil characteristics, we found that clay content, silt content, pH, and SOM in soil characteristics were the controlling factors of the SPF of most heavy metals; and 5. The mangrove restoration wetland was restored in the fifth year, and the pollution risk of Cu, Cr, Zn, and Pb decreased from medium and high pollution levels to low pollution levels or even no pollution levels.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecoleng.2021.106242.

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