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Heavy Metal Concentrations and Accumulation Characteristics of Dominant Woody Plants in Iron and Lead−Zinc Tailing Areas in Jiangxi, Southeast China

Yanglong Li [1,2,3], Chaoqun Wang [4], Chaowu Yan [5], Shaowen Liu [6], Xiangteng Chen [1,2,3], Mansheng Zeng [7], Yuhong Dong [1,2,3] and Ruzhen Jiao [1,2,3,*]

[1] Research Institute of Forestry, Chinese Academy of Forestry, Beijing 100091, China
[2] State Key Laboratory of Tree Genetics and Breeding, Chinese Academy of Forestry, Beijing 100091, China
[3] Key Laboratory of Tree Breeding and Cultivation of State Forestry Administration, Chinese Academy of Forestry, Beijing 100091, China
[4] Biogeochemistry of Agroecosystems, University of Goettingen, 37077 Goettingen, Germany
[5] Forestry Bureau of Xinyu City, Xinyu 338000, China
[6] Standard Technology Engineering (Qingdao) Company Limited, Qingdao 266109, China
[7] Experimental Center of Subtropical Forestry, Chinese Academy of Forestry, Fenyi 336600, China
* Correspondence: jiaorzh@caf.ac.cn

Abstract: Phytoremediation using woody plants can effectively reduce heavy metal (HM) concentrations in soils. However, the remediation capacity of woody plants depends greatly on plant species and soil environmental conditions. In order to evaluate the HM remediation potential of woody plants from different tailing areas, the HM accumulation characteristics of roots, shoots, and leaves of 12 dominant native woody plants growing in iron and lead-zinc tailing areas were analyzed. The results showed that the concentrations of Cd, As, Ni, Mn, and Cr in most plants in the two tailing areas exceeded the level of normal plants. The distribution of different elements in plants was generally as follows: root > leaf > shoot for Pb and As; root > shoot > leaf for Cr; and leaf > shoot > root for Zn, Ni, and Mn. The distribution of Cu and Cd in plants varied with the type of HM pollution in the two tailing areas. There were significant ($p < 0.05$) negative correlations between available phosphorus in the soil and Pb, Cd, and Zn in the plant roots when the soil was heavily polluted with Pb, Cd, and As; similarly, there were significant ($p < 0.01$) negative correlations between readily available potassium in the soil and Pb, Zn, and Ni in plant roots. Based on the higher than average concentration of HMs in plants, and higher bioconcentration factors and translocation factors, some plants were considered woody plant species with phytoremediation. Slash pine (*Pinus elliottii*) and indian azalea (*Rhododendron simsii*) had strong enrichment and translocation abilities for Cd, oriental white oak (*Quercus glauca*) and beautiful sweetgum (*Liquidambar formosana*) for Mn and paulownia (*Paulownia fortunei*) for Zn. The plants listed above can be used as potential species for phytoremediation in iron and lead-zinc tailing areas.

Keywords: woody plants; phytoremediation; bioconcentration; enrichment coefficients; heavy metal pollution; abandoned mines; metal accumulation

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1. Introduction

China is rich in mineral resources, which plays an important role in the development of the national economy. However, mineral mining is considered to be the main source of toxic contaminants in soil [1–5]. According to the latest national soil pollution survey bulletin, 33.4% of the survey sites in mining areas exceed the national standard, and the soil around non-ferrous metal mining areas is more seriously polluted by cadmium, arsenic, and lead [6]. Toxic heavy metals (HMs) in waste rocks, tailings, and smelleding slags generated by the mining process are transferred to the soil and groundwater in mining areas through oxidation and the impact of the natural environment [1,7]. The accumulation of toxic HMs in soils reduces soil quality and contaminates crops and groundwater, which affects ecosystems and human health [8,9]. Soil HM pollution has raised global concern due to its crypticity, persistence, and high toxicity [10,11].

The remediation methods for soil HM pollution mainly include physical remediation, chemical remediation, and biological remediation. Phytoremediation is defined as a low-cost biological remediation technique that uses plants and associated soil microorganisms to reduce the concentration of HMs in the soil [12]. The plants transfer HMs from roots to aboveground parts through transpiration and achieve the purpose of removing HMs from the soil by harvesting the plants [13]. Some exudates produced by plant roots can cause precipitation of metals and reduce the mobility and biotoxicity of HMs in the soil [14]. It has been estimated that phytoremediation is about 60%–80% less expensive than conventional physicochemical remediation [15]. In addition, the disposal of plants using technologies, such as incineration, pyrolysis, and hydrothermal upgrading, can create biomass energy while reducing the risk of secondary pollution of HMs from phytoremediation [16]. It has been widely used as a sustainable remediation method to improve soil quality and mitigate metal toxicity due to its advantages of high efficiency, economy, and ecological sustainability [13,17]. Phytoremediation technology includes phytostabilization, phytovolatilization, and phytoextraction [18], the core of which is the screening of hyperaccumulators, which is defined as plant species that accumulate higher concentrations of specific HMs in the aboveground parts. To date, more than 500 plant species have been identified as hyperaccumulators, most of which belong to the herbaceous families of Asteraceae, Brassicaceae, Caryophyllaceae, Poaceae, Violaceae, and Fabaceae [12,19]. However, herbaceous phytoremediation is limited by the low biomass of herbaceous plants, and its remediation efficiency may remain unsatisfactory when applied directly to phytoremediation of polluted sites [20]. In contrast, woody plants can accumulate more HMs than herbaceous plants due to their well-developed root systems, higher above-ground biomass, and the planting of woody plants with phytoextraction and/or phytostabilization capabilities can not only reduce the mobility of HMs but also effectively prevent the expansion of soil HM pollution caused by soil erosion [21,22]. Phytoremediation using fast-growing and high-biomass tree species, such as poplar, willow, eucalyptus, birch, and paulownia, is an effective technology to reduce soil HM pollution [23]. It is also an effective plant option to use nitrogen-fixing woody plants, such as false indigo (*Amorpha fruticosa*), to fix nitrogen in the air, improve soil nutrients, enhance plant adaptability to HM stress in tailings, and revegetate without using chemical fertilizers [24]. However, woody hyperaccumulators are regionally dependent, and phytoremediation capacity depends on soil conditions [25,26]. Therefore, screening indigenous dominant plants that adapt to local soil conditions may have greater remediation potential in future phytoremediation applications [27,28].

Jiangxi province is a typical distribution area of acidic soil in the southeast of China. Due to the rich mineral resources, a large number of mines have been exploited, which has aggravated soil acidification, soil infertility, and HM pollution. In order to find plants with HM remediation potential, we selected the Jiulong iron mine and Yangtiangang lead-zinc mine in Xinyu City, Jiangxi Province as the study areas, which both have similar climatic and soil conditions but belong to different tailing types. In this study, a total of 12 different species of native dominant woody plants were selected to analyze the concentration and accumulation characteristics of HMs in roots, shoots, and leaves. The aim of this study is to analyze the HM pollution types in different tailing areas, analyze the potential soil factors affecting the uptake of HMs by plants, screen woody plants that can enrich or transport HMs, and evaluate their potential for future application as woody phytoremediation species.

2. Materials and Methods

2.1. Study Site Description

The study sites are located in two tailing areas where phytoremediation experiments were conducted: Jiulong iron tailing area and Yangtiangang lead-zinc tailing area, Yushui District, Xinyu City, Jiangxi Province, China (27°32′–27°51′ N, 114°43′–114°52′ E) (Figure 1). The climate of the study areas is a mid-subtropical monsoon with an annual average temperature of 18.6 °C, an annual average precipitation of 1264.5 mm, and an annual sunshine hours of 1753.1 h. The soil type in the two study sites is red soil developed from granite, metamorphic rocks, gravels, and carbonate rocks. In the 1950s, large-scale mining activities were carried out in the study areas, and the random accumulation of slag waste slags during the mining process had a great impact on the local ecological environment. The Yangtiangang mine was again mined on a large scale in 2008, causing serious pollution of the surrounding water, which drew the attention of the local government and was shut down after government intervention. The Jiulong mine is still being mined in a relatively less polluted cave mine, but the problems of vegetation destruction and water pollution cannot be ignored. Based on this situation, local phytoremediation was started in 2013, and more than 30 species of trees, shrubs, and herbs were planted, and the water quality of nearby rivers has improved. However, the growth of the tree species used for phytoremediation is uneven, and the effect of phytoremediation is still unclear.

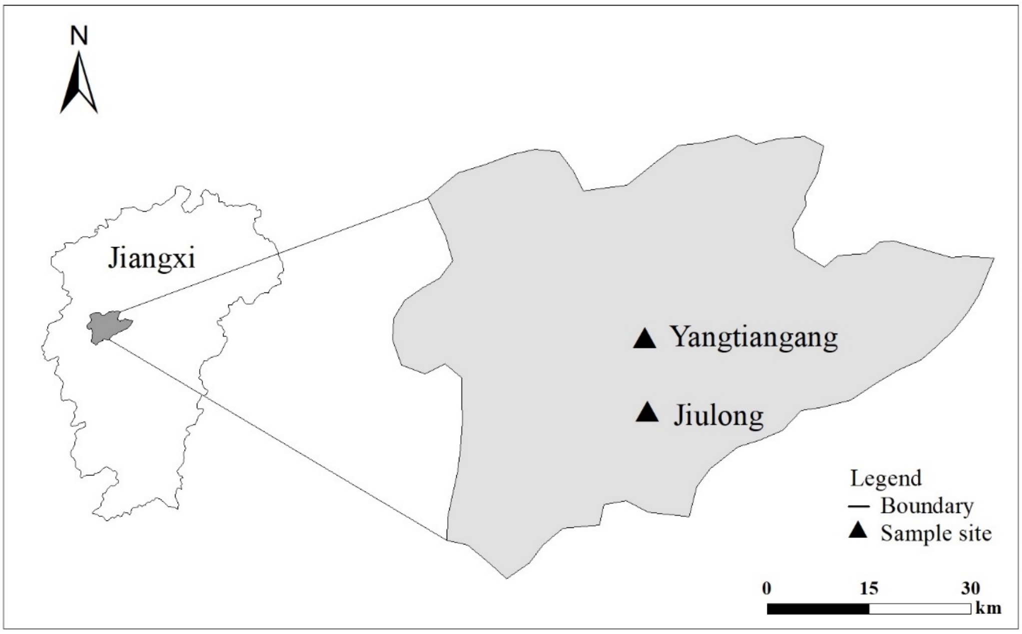

Figure 1. Location map and different sampling sites of investigated tailing areas.

2.2. Sample Collections

Plant samples were collected in May 2021. Well-grown woody plants growing on the southern slope of tailing areas were selected as the dominant plants, as shown in Table 1. Three plants with the same tree height, diameter at breast height, and growth vigor were randomly selected for one mixed sampling. The plant samples of annual shoots and new leaves of the current year were collected from the upper, middle, and lower layers of the crown and mixed well. Root samples were fine roots less than 2 mm in diameter, which were collected from soil at a depth of 0–20 cm. Meanwhile, the soil attached to the roots within 2 cm were also collected as root zone soil, and stones, weeds, and twigs were removed. Three duplicated samples of each plant species were obtained. The information of the sampling points was recorded by a global positioning system (GPS). All samples were immediately placed in clean polyethylene self-sealing bags and transported back to the laboratory for subsequent analysis.

Table 1. List of dominant plant species growing in the study area.

Sample Sites		Family	Species	Life Form
Jiulong	1	Anacardiaceae	*Rhus chinensis* Mill.	Deciduous tree
	2	Taxodiaceae	*Cunninghamia lanceolata* (Lamb.) Hook.	Evergreen tree
	3	Pinaceae	*Pinus massoniana* Lamb.	Evergreen tree
	4	Betulaceae	*Alnus cremastogyne* Burk.	Deciduous tree
	5	Pinaceae	*Pinus elliottii* Engelmann	Evergreen tree
	6	Leguminosae	*Spatholobus suberectus* Dunn	Evergreen vine
	7	Fagaceae	*Quercus glauca* Thunb.	Evergreen tree
Yangtiangang	8	Pinaceae	*Pinus massoniana* Lamb.	Evergreen tree
	9	Fagaceae	*Castanopsis sclerophylla* (Lindl. et Paxton) Schottky	Evergreen tree
	10	Anacardiaceae	*Rhus chinensis* Mill.	Deciduous tree
	11	Ericaceae	*Rhododendron simsii* Planch.	Deciduous shrub
	12	Hamamelidaceae	*Liquidambar formosana* Hance	Deciduous tree
	13	Rosaceae	*Photinia* × *fraseri* Dress.	Evergreen shrub
	14	Scrophulariaceae	*Paulownia fortunei* (Seem.) Hemsl.	Deciduous tree

2.3. Sample Analysis

Soil samples were air-dried, ground, and sieved through 2 mm for soil pH, available phosphorus (AP), and readily available potassium (AK) assays and 0.149 mm for HMs assays. Each soil sample (0.1 g) was digested with 8 mL aqua regia (HNO_3:HCl = 1:3) [29]. For the purpose of removing surface dirt of plant samples, root, shoot, and leaf samples were rinsed with running water for 24–36 h and then repeatedly washed several times with deionized water. The plant samples were dried at 85 °C for 30 min, then dried at 65 °C to a constant weight. The dried plant samples were crushed to a size <2 mm using a grinder. Each plant sample (0.2 g) was digested with 8 mL of 65% HNO_3 [30]. Each digested sample was filtered through 0.45 µm aqueous filter membrane and diluted to 25 mL volume with deionized water. Soil AP and AK were extracted from soil (5 g) using 50 mL solution of a mixture of 0.03 mol L^{-1} NH_4F and 0.025 mol L^{-1} HCL and 50 mL solution of 1 mol L^{-1} NH_4Ac (pH = 7.0), respectively. The concentrations of Pb, Cd, Cu, Zn, Cr, Ni, Mn, AP, and AK were measured by inductively coupled plasma mass spectrometry (ICP-MS, Thermo ICAP-RQ, USA). The concentrations of As were measured by atomic fluorescence spectrometry (AFS, Persee PF32, China). The recoveries of each element by standard addition were within 95.45%–104.55% for Pb, 94.29%–105.71% for Cd, 95.76%–104.24% for Cu, 93.94%–106.06% for Cr, 96.97%–103.03% for Zn, 96.68%–103.32% for Mn, 96.67%–103.33% for Ni, and 92.98%–107.02% for As. Soil pH (soil:water = 1:2.5) was measured with a conventional pH meter (Mettler Toledo FE20, China).

2.4. Assessment of Plant HM Enrichment and Transport Capacity

To assess phytoextraction or phytostabilization and to screen plant species with phytoremediation capabilities [31], the bioconcentration factor (BCF) reflecting the HM enrichment capacity and translocation factor (TF) indicating the ability of plants to transfer HMs from the belowground (root) to the aboveground (shoot and leaf) were calculated as [13]:

$$BCF_{leaf/shoot/root} = \frac{C_{leaf/shoot/root}}{C_{soil}}$$

$$TF_{leaf/shoot/root} = \frac{C_{leaf/shoot}}{C_{root}}$$

where *C* is the concentration of each heavy metal in each plant organ or soil.

2.5. Risk Assessment of Soil HM Pollution

The risks of soil HM pollution in tailing areas were assessed using the single-factor pollution index (P_i) and the geoaccumulation index (I_{geo}). The P_i is the most widely used method for assessing soil HMs and is calculated as follows:

$$P_i = \frac{C_i}{S_i}$$

where C_i is the concentration of each HM element in the soil; S_i is the risk threshold values; Pb, Cd, Cu, Zn, As, Cr, and Ni refer to the risk screening value of pH $\leq$ 5.5 in the Chinese standard for soil pollution risk control of agricultural land [32]; and Mn refers to the standard in the reference of Li et al. [33]. Based on the P_i values, the risk level of soil HM pollution was classified as: $P_i \leq 0.7$, clean; $0.7 < P_i \leq 1.0$, low pollution; $1.0 < P_i \leq 2.0$, mild pollution; $2.0 < P_i \leq 3.0$, moderate pollution; $P_i > 3.0$, heavy pollution [34].

I_{geo} takes into account the influence of anthropogenic pollution, environmental geochemistry, and natural rock formations on the background value and is a more scientific and objective method for evaluating HM pollution in soil, which is calculated as:

$$I_{geo} = \log_2\left[\frac{C_n}{1.5B_n}\right]$$

where C_n is the concentration of each HM element in the soil; B_n is the local background value of the HMs element; and 1.5 was the correction index. Based on the I_{geo} values, the risk level of soil HMs pollution was classified as: $I_{geo} \leq 0$, clean; $0 < I_{geo} \leq 1.0$, mild to moderate pollution; $1.0 < I_{geo} \leq 2.0$, moderate pollution; $2.0 < I_{geo} \leq 3.0$, moderate to heavy pollution, $3.0 < I_{geo} \leq 4.0$, heavy pollution; $4.0 < I_{geo} \leq 5.0$, heavy to extreme pollution; $I_{geo} > 5$, extreme pollution [35].

2.6. Statistical Analysis

The experimental data were processed using Microsoft Excel (Version 2019) and IBM SPSS (Version 26.0). Figures were drawn with ArcGIS (Version 10.2) and GraphPad Prism (Version 9.0). One-way analysis of variance (ANOVA) was conducted to determine the differences in HM concentrations in plants between different tree species, with $p < 0.05$ considered significant for Duncan's test. Pearson correlation analysis and principal components analysis were used to identify the potential relationship between heavy metal concentrations in plants and soil properties.

3. Results

3.1. Soil Characteristics and Pollution Assessment

The soil characteristics in the study area are shown in Table 2. The soil was acidic (pH 2.95–5.92), and the average concentrations of AP and AK in the soil was < 10 mg kg^{-1} and 100 mg kg^{-1}, respectively, indicating that the soil was nutrient-poor and not conducive to plant growth. The concentrations of all HMs in the Jiulong and Yangtiangang tailing areas were above the background values. The concentrations of Cd, Cu, and Mn in the Jiulong tailing area exceeded the threshold values by 1.03, 1.07, and 1.89 times. The concentrations of Pb, Cd, Cu, Zn, and As in the Yangtiangang tailing area exceeded the threshold values by 18.32, 3.20, 1.20, 1.70, and 55.16 times. The soils of the two tailing sites were complexly polluted with multiple HM elements. The I_{geo} and P_i values of the eight HM elements (Figure 2) showed that there was mild Cd and Cu pollution and moderate Mn pollution in the Jiulong tailing area and mild Cu pollution, moderate Zn pollution and heavy Pb, Cd, and As pollution in the Yangtiangang tailing area. Furthermore, the coefficients of variation of Cd, Cu, and Mn in Jiulong and Pb, Cd, Zn, and As in Yangtiangang were all greater than 0.4, indicating that there was a large spatial variability of HM pollution in the tailing waste sites.

Table 2. Soil characteristics and reference values in the Jiulong iron tailing area and Yangtiangang lead–zinc tailing area.

Sample Sites	Jiulong			Yangtiangang			Reference Value	
	Range [a]	Mean	CV	Range	Mean	CV	Background [b]	Threshold [c]
pH	4.49–5.92	4.83	0.10	2.95–4.41	3.70	0.11	–	–
AP	1.24–3.02	2.15	0.25	1.13–15.87	8.72	0.51	–	–
AK	38.89–158.01	86.16	0.52	61.7–140.75	95.17	0.27	–	–
Pb	27.08–47.66	35.24	0.17	67.86–5944.23	1282.57	1.58	32.1	70
Cd	0.12–0.72	0.31	0.57	0.10–2.95	0.96	1.09	0.1	0.3
Cu	17.93–102.94	53.53	0.65	33.41–84.78	59.87	0.27	20.8	50
Zn	61.19–96.42	81.86	0.15	117.25–820.47	339.08	0.75	69.0	200
As	3.36–30.61	15.53	0.64	150.45–4454.35	2206.37	0.65	10.4	40
Cr	31.33–82.73	64.57	0.24	51.75–77.48	64.89	0.15	48.0	150
Ni	29.40–53.51	40.41	0.19	16.06–55.39	34.82	0.30	19.0	60
Mn	470.83–3612.76	2267.58	0.46	678.92–1045.57	856.82	0.15	258.0	1200

[a], Element concentration unit: mg kg^{-1}. [b], Background values of the soil environment in Jiangxi Province in 2006. [c], The threshold values of Pb, Cd, Cu, Zn, As, Cr, and Ni refer to the risk screening value of pH $\leq$ 5.5 in the Chinese standard for soil pollution risk control of agricultural land [32], and Mn refer to the standard in the reference of Li et al. [33].

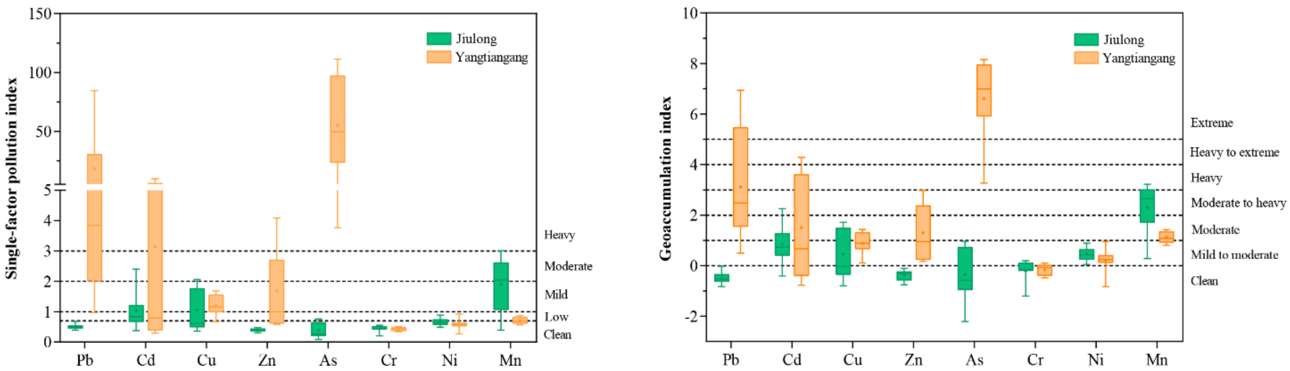

Figure 2. The single–factor pollution index and the geoaccumulation index of eight HM elements in two tailing areas.

3.2. HM Concentrations in Dominant Plants

The HM concentrations determined in woody plants in the study area are depicted in Figure 3. The range of various HMs in plants was: 0.404–476 mg kg^{-1} for Pb, 0.025–1.47 mg kg^{-1} for Cd, 2.76–48.8 mg kg^{-1} for Cu, 8.19–200 mg kg^{-1} for Zn, 0–713 mg kg^{-1} for As, 2.11–24.4 mg kg^{-1} for Cr, 1.52–36.1 mg kg^{-1} for Ni, and 29.1–2898 mg kg^{-1} for Mn. In the Yangtiangang tailing area, where Pb, Cd, Zn, and As pollution is more severe, the corresponding element concentrations in plants were higher. The concentrations of HMs in different organs of the plant varied, which were generally as follows: root > leaf > shoot for Pb and As; root > shoot > leaf for Cr; and leaf > shoot > root for Zn, Ni, and Mn. The distribution of Cu and Cd in plants in the two tailing areas was different, with shoot > leaf > root for Cu and shoot > root > leaf for Cd in the Jiulong tailing area; and root > shoot > leaf for Cu and shoot > leaf > root for Cd in the Yangtiangang tailing area. The concentrations of HMs also varied greatly among the sampled plant species. The highest Cd and Cu concentrations were found in *R. simsii*; the highest Cr and Pb concentrations were found in *R. chinensis*; the highest Mn concentrations were found in *Q. glauca*; the highest Ni concentrations were found in *P. massoniana*; the highest Cu and Zn concentrations in the shoots and leaves were found in *P. fortunei*, while the highest Cu and Zn concentrations in the roots were found in *R. simsii* and *R. chinensis*, respectively.

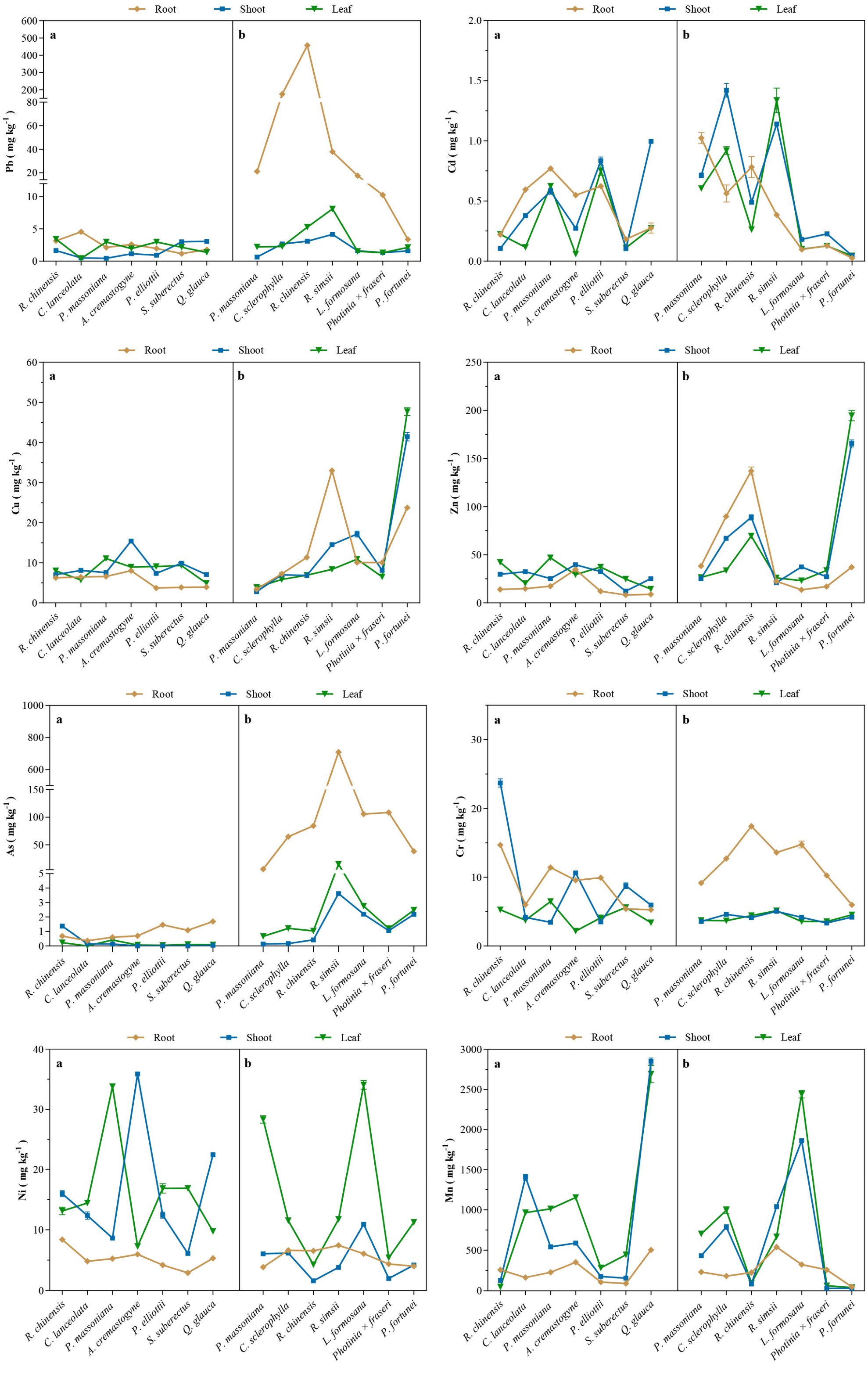

Figure 3. Heavy metals in the roots, shoots, and leaves of each woody plant in the Jiulong iron tailing area (**a**) and Yangtiangang lead−zinc tailing area (**b**).

3.3. Correlation Analysis and Principal Components Analysis between the Concentrations of HMs in Plant Roots and the Concentrations of HMs, AP, and AK in Soil

The correlation analysis between HM concentrations in plant roots and AP, AK, and HM concentrations in soil are shown in Figure 4. The Pb and As concentrations in roots positively correlated with Pb and As concentrations in soils ($p < 0.01$), respectively, while the Zn concentrations in roots negatively correlated with Cr concentrations in soils ($p < 0.01$), in both study areas. It is noteworthy that there was a significant ($p < 0.05$) correlation between AP and AK concentrations in soils and some HM concentrations in plant roots in both study areas. Significant positive correlations were found between AP concentrations and plant root Pb and Cd concentrations, and between AK and plant root Pb, Cr, and Ni concentrations in the Jiulong iron tailing area. However, such correlations were completely reversed in the Yangtiangang lead-zinc tailing area.

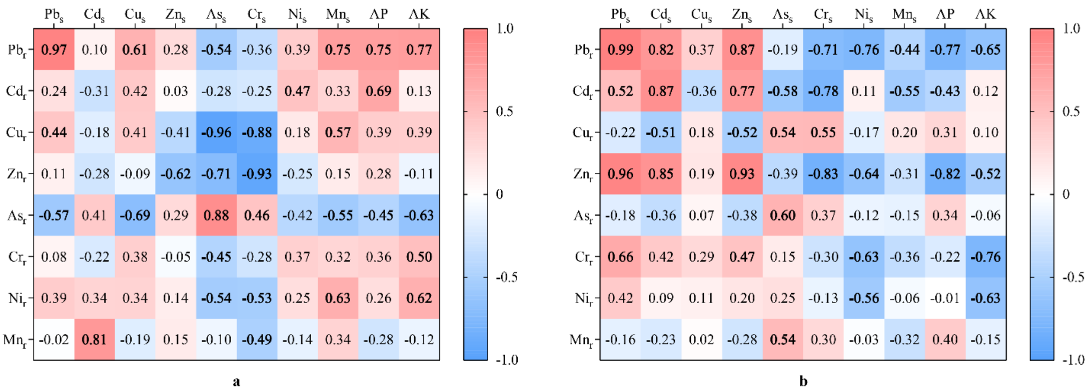

Figure 4. The Pearson correlation coefficients between plant root HM concentrations and soil HM, AP, and AK concentrations in the Jiulong iron tailing area (**a**) and Yangtiangang lead-zinc tailing area (**b**). The 's' and 'r' in the subscript denote soil and root, respectively. Bold numbers indicate the statistically significant ones, and correlation is significant at the 0.05 level (2−tailed).

Two principal components (PC 1 and PC 2) were extracted through principal components analysis of each sample analyzed. As shown in Figure 5, the determination of the relevance among HMs in plant roots and HMs, AP, and AK in soil were grouped into two classes of components. The concentration of As in soils was close to that of As in plant roots in both study areas. In addition, the closest relevance was shown between AK, AP, Pb, Cu, Mn, and Ni in soil and Pb, Ni, Cd, and Cr in plant roots in Jiulong. In Yangtiangang, Pb, Cd, and Zn in plant roots and Pb, Cd, and Zn in soil were close to each other. AP and AK in the two tailing areas had higher loadings in component 1. In Jiulong, soil AP had significant positive correlations with Pb and Cd in plant roots, and soil AK had significant positive correlations with Pb and Ni in plant roots, while in Yangtiangang, all of these relationships showed negative correlations.

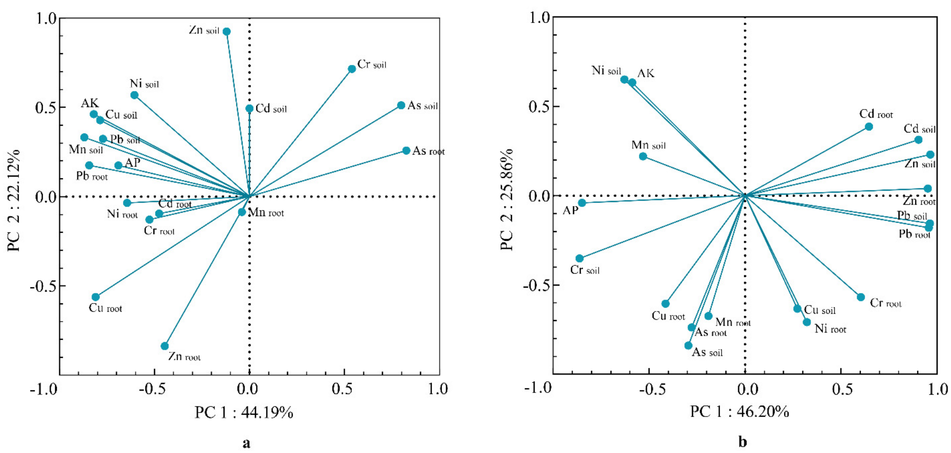

Figure 5. The loading plot from principal component analysis in selected samples from the Jiulong iron tailing area (**a**) and Yangtiangang lead−zinc tailing area (**b**).

3.4. Evaluation of HM Accumulation and Translocation Potential

The BCFs and TFs of woody plants in the study areas are depicted in Figures 6 and 7. There were differences in the capacity of plants to accumulate and transport various HMs. The BCFs of plant roots, shoots, and leaves to the six HMs with pollution risk in soil all showed Cd > Mn > Zn > Cu > Pb > As. The TFs of plant shoots and leaves to the six HMs all showed Mn > Zn > Cd ≈ Cu > Pb > As. The BCFs of most plants for Pb, Cu, Zn, and As were <1, and the TFs of all plants for As were <1, which indicated that these elements were not easily accumulated and translocated to these organs. The enrichment and transport capacities of most woody plants were more pronounced for Cd, and most of the BCFs and TFs > 1. Among the plant samples collected, *R. simsii* showed the best enrichment and transport capacity for Cd. BCF_{leaf}, BCF_{shoot}, and BCF_{root} of *R. simsii* were 8.41, 7.48, and 2.61, respectively, and the TF_{shoot} and TF_{leaf} were 2.87 and 3.23, respectively, which far exceeded 1. The BCF_{shoot}, BCF_{leaf}, Tf_{shoot}, and TF_{leaf} for Mn of *Q. glauca* and *L. formosana* were greater than 1, and the BCF_{shoot}, BCF_{leaf}, Tf_{shoot}, and TF_{leaf} for Zn of *P. fortunei* were greater than 1.

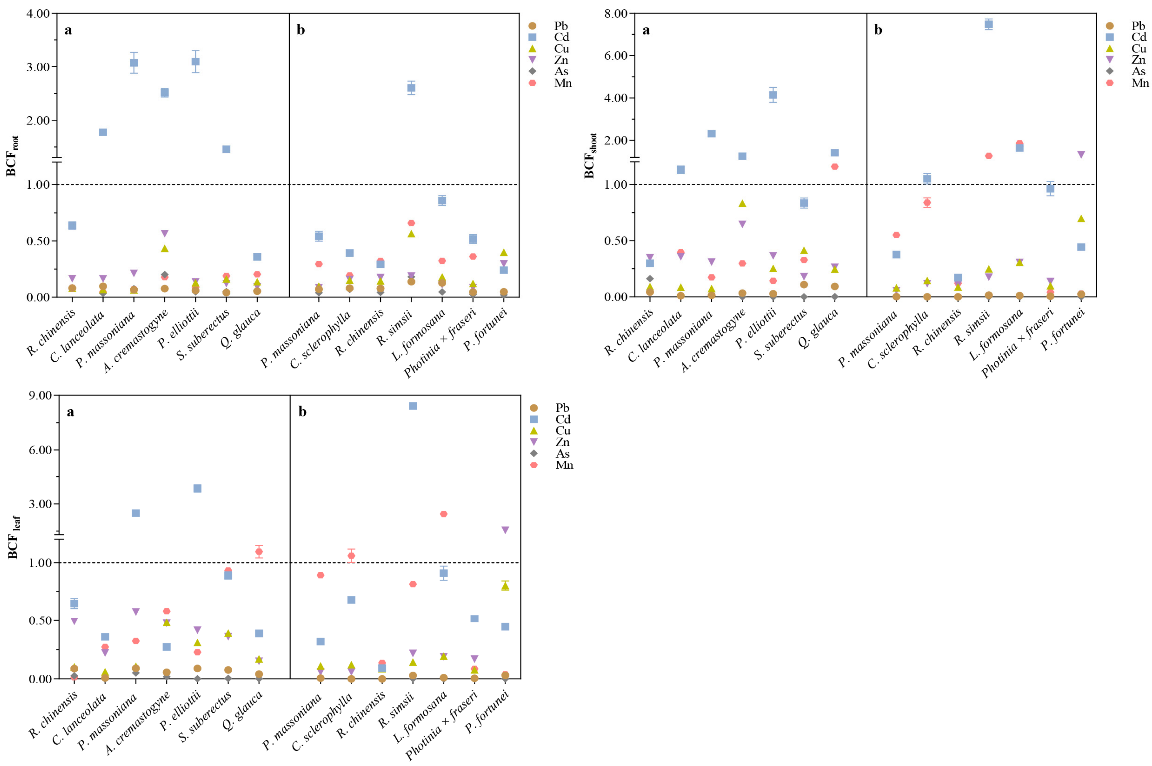

Figure 6. The BCFs of heavy metals in the roots, shoots, and leaves of each woody plant in the Jiulong iron tailing area (**a**) and Yangtiangang lead−zinc tailing area (**b**).

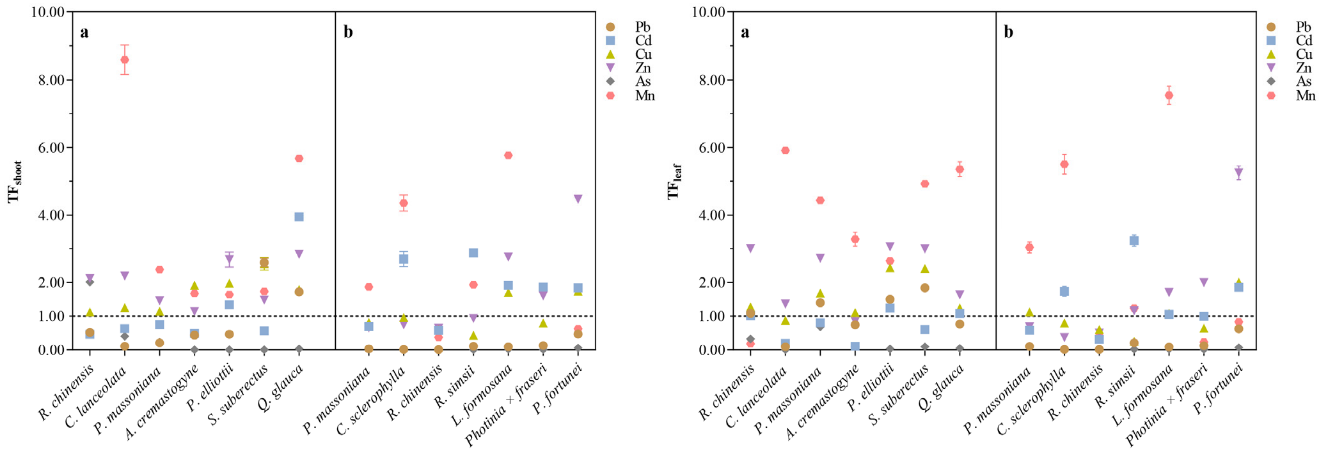

Figure 7. The TFs of heavy metals in the shoots and leaves of each woody plant in the Jiulong iron tailing area (**a**) and Yangtiangang lead-zinc tailing area (**b**).

4. Discussion

4.1. Concentrations of HMs in Woody Plants and Potential Influencing Factors

In general, the normal range of HM concentrations in plants is 0.1–41.7 mg kg^{-1} for Pb, 0.2–0.8 mg kg^{-1} for Cd, 0.4–45.8 mg kg^{-1} for Cu, 1–160 mg kg^{-1} for Zn, 0–1 mg kg^{-1} for As, 0.2–8.4 mg kg^{-1} for Cr, 0.1–10 mg kg^{-1} for Ni, and 1–700 mg kg^{-1} for Mn [36–39]. The HM concentrations of plants in the study area far exceeded general plants. Some of the plant roots have good adsorption capacity for Pb, As, and Cr in the soil, and the concentrations of these elements accumulated in roots are higher than those in aboveground shoots and leaves. It has been demonstrated that the concentrations of HMs in plants increase with increasing concentrations of HMs in the environment, but the rate of HM transport from plant roots to shoots decreases, which indicates high plant availability of the metals, as well as its limited mobility once inside the plant [40,41]. As a result, the uptake of Pb, As, and Cr by woody plants in this study is mainly confined to the roots [42,43].

The concentrations of As, Zn, Mn, and Ni in the leaves were higher than those in the shoots, which could be attributed to the role of the shoots as a transferring tissue for these elements [44]. In this study, the concentrations of Mn, Zn, Cu, and Ni in most plants were higher than those of plant Pb, Cd, Cr, and As, which was related to the specific roles played by these elements in plants. Mn, Zn, Cu, and Ni are essential trace elements for plant growth and development and are involved in processes such as enzyme and protein synthesis and photosynthesis in plants [45]. The content of these elements in plants exceeding the normal value might cause cell damage and leaf yellowing [46–48]. Cd, Pb, Cr, and As are not essential for plant growth and are strongly biotoxic to plants [49]. The presence of As and Cr in plants will induce free radical production and cause oxidative stress, which in turn leads to reduced plant biomass, leaf yellowing, and necrosis [50,51], while Pb and Cd inhibit seed germination, nutrient synthesis, and uptake and damage [52–54].

A general positive correlation between HM concentrations in plants and soil has been reported [55–58]. Similar results were observed in the present study. Both *R. chinensis* and *P. massoniana* showed stronger enrichment ability for Pb, Cd, As, and Zn in Yangtiangang, where the pollution of these four HMs was more serious. In Jiulong, where Mn pollution was more pronounced, both *R. chinensis* and *P. massoniana* showed a stronger enrichment ability for Mn. The correlation analysis also showed that the uptake of Pb and As by plant roots mainly depends on the concentrations of the corresponding two elements in the soil, with a significant positive correlation, indicating that the bioavailable concentrations of Pb and As in the soil may be related to the total amount of soil Pb and As. It is worth noting that the average concentrations of Pb (1282.57 mg kg^{-1}) and As (2206.37 mg kg^{-1}) in soil collected from Yangtiangang exceeded the pollution risk screening value by 18.32 times and 55.16 times; however, some plants, such as *R. chinensis* and *C. sclerophylla*, could still grow well under such extreme environments. These plants may develop defense strategies against high concentrations of HMs under long-term natural selection, such as the sequestration of HMs by the root system and the activation of antioxidant enzyme systems to reduce the oxidative stress caused by HMs in plants [59,60].

Previous research has explained that soil pH plays the most important role in determining metal morphology, mineral surface solubility, migration, and final bioavailability [61–63]. The soil pH in this study area exhibited strong acidity. Since a lower pH generally decreases the sorption of metal ions on soil particles, the competition between free metal ions and other cations increases, thus increasing the metal concentrations in the soil solution [28,64]. In other words, the migration and bioavailability of HMs will increase as soil pH decreases, which can exacerbate HM toxicity to plants and is likely to cause extensive pollution. The exudates produced by plant roots, such as amino acids and polysaccharides, can chelate with HM ions such as Pb^{2+}, Cu^{2+}, and Cd^{2+} to form stable metal complexes and fix them in the area outside plant roots [53]; thus, selection of appropriate plants for phytoremediation might be important not only for reducing HM levels, but also for reducing HM migration and diffusion [65].

Phosphorus and potassium are essential nutrient elements for plant growth, and the reasonable addition of fertilizers containing phosphorus and potassium can effectively promote plant growth and produce a biological dilution effect on HMs in plants by increasing biomass. In the remediation of soil HMs, phosphate compounds are often used to increase the immobilization of metals, thereby reducing their mobility and bioeffectiveness in the soil [66–68]. Similarly, potassium can immobilize HMs in the soil, reducing the uptake of HMs by plants [69]. It can also suppress the production of reactive oxygen species and help plants to antioxidize against HM stress, reducing the toxic effects of HMs, such as Pb, Cd, Zn, and As, in plants [70,71]. Chen et al. [72] found that the abilities of wheat to accumulate Zn, Cu, Pb, and Ni from soil were suppressed by phosphorus addition, while its accumulation of As was enhanced. Similar results were obtained in the Yangtiangang lead-zinc tailing area in this study. The negative correlations between AP concentrations in soils and the concentrations of Pb, Cd, and Zn in plant roots indicate that AP may be involved in complexation with Pb, Cd, and Zn, reducing the concentrations of available HM elements in soils and reducing the uptake of these elements by plants. Xing et al. [73] found that the application of higher rates of phosphate in soils with higher Cd and Pb concentrations resulted in lower available Pb and Cd in the soil, and grain Pb, Cd, and Zn concentrations were negatively affected by the phosphorus application. In addition, the negative correlations between AK concentrations in soils and the concentrations of Pb, Zn, Cr, and Ni in plant roots indicate that AK may reduce the uptake of these elements in plants and reduce the toxic effects of such elements on plants. It seems that there were potential effects between soil AP and AK on the uptake of HMs by woody plants, and a reasonable addition of phosphorus and potassium to soil may be helpful for plants to resist the toxic effects of HMs. Interestingly, similar findings to those of previous studies were only found in Yangtiangang, while completely opposite conclusions were obtained in Jiulong. In the two study areas, the cumulative contribution rate of components 1 and 2 in the principal component analysis was not high, being around 70%, indicating that the HMs absorbed by plants and soil physicochemical properties may still be affected by many factors, such as difference of plant species, degree of soil pollution, and content of soil organic matter and nitrogen [38,74,75]. The mechanism of soil properties on the uptake of HMs by plants remains to be further investigated.

4.2. Accumulation and Transport Characteristics of HMs in Woody Plants

The elements causing HM pollution in the two study areas are complex. There may be competition among multiple metals for soil adsorption sites, and one metal tends to be mobilized with increasing concentrations of another metal, thereby enhancing its bioavailability and toxicity [76]. The additive effect of multi-metal complex pollution may exacerbate the toxicity of HMs on soil and plants and affect the ability of plants to take up

HMs [77]. However, some woody plants in this study had a good enrichment capacity for a variety of HMs. *R. simsii* could enrich both Cd and As, and *L. formosana* could enrich both As and Mn, which reflected the potential of these plants in remediating complex HM-polluted soils. Differences in the uptake and translocation of HMs by different woody plants may be related to differences in the type of soil pollution [78,79]. The phytoextractive capacity of plants may be altered by soil complex contaminants, which is mainly influenced by a high concentration of a particular metal in the soil. For example, a mixed treatment of Cd and Zn significantly reduced the accumulation of Cd and Zn in poplar and willow, possibly due to ionic competition between the metals [80,81]. There were large differences in Pb and As pollution between the two tailing areas, which may explain the differences in Cu and Cd uptake by plant roots and leaves [82].

In general, a BCF > 1 indicates that the tree species has a high enrichment capacity for HM and can effectively reduce the concentration of HM in the soil [83]. TF > 1 indicates that the tree species can effectively transfer HM from the belowground to the aboveground, reduce the toxic effects caused by high concentrations of HM in the plant roots, and have the potential for phytoextraction [84,85]. Wang et al. [86] concluded that woody plants with a BCF greater than 0.4 are hyperaccumulators due to their high biomass, but there is a lack of experimental corroboration. No matter which of the above criteria is used for evaluation, similar to previous studies, plants growing in the two tailing areas have a stronger enrichment and transport capacity for Cd, which reflects the higher mobility of Cd in plants compared with other HM elements [87,88].

According to Baker and Brooks [89], hyperaccumulators refer to plants whose above-ground HM concentration is more than 10 times higher than that of typical plants. Through experiments and model validation, van der Ent et al. [20] concluded that the criteria for hyperaccumulators of certain metal elements should be set to Cd > 100 mg kg^{-1}, Cu, Cr > 300 mg kg^{-1}, Pb, Ni, As >1000 mg kg^{-1}, Zn > 3000 mg kg^{-1}, and Mn > 10,000 mg kg^{-1}. Although none of the plants were found to be hyperaccumulators in this study, some plants with high BCF, high TF or high BCF, and low TF are still found, and such plants have the potential to be applied for phytoextraction or phytostabilization in HM-polluted areas [90]. In addition, some plants can avoid excessive intake of HM ions and have strong stress avoidance and tolerance to environments with extremely high concentrations of soil HMs through the tolerance of the protoplasm, despite their low BCF and TF for HMs [91]. These plants can be used to re-green the tailing areas, which can help to conserve water and soil and reduce the migration of HMs. Based on the following criteria, tree species with HM remediation potential were classified. Some plants had HM concentrations in organs above the normal range, with BCFs and TFs for HMs of shoots and leaves > 1. Such plants are considered to have the potential for phytoextraction. Some plants are considered to have an ultra-high tolerance to HMs when they are grown in soils exceeding 20 times the threshold value for HMs, despite the fact that plant HMs levels are in the normal range and BCFs are < 1. The native woody species with potential for remediation of polluted soil in tailing areas were screened in this study (Table 3). *R. simsii* and *P. elliottii* have high BCFs and high TFs for Cd, *Q. glauca* and *L. formosana* have high BCFs and high TFs for Mn, *P. fortunei* has high BCFs and TFs for Zn, and these plants are considered to have the potential of phytoextraction for Cd, Mn, and Zn, respectively, and could be used as woody phytoremediation species for Cd, Mn, and Zn pollution in acidic red soil regions. Caparros et al. [92] also found that *P. fortunei* had a strong ability to accumulate Zn, and its annual increment of biomass is very high, up to 150 t ha^{-1}, and can be used for phytoremediation to reduce soil HM levels. As shown in Table 3, *C. sclerophylla* and *R. chinensis* are able to tolerate extreme high concentrations of Pb and As in the soil, and *L. formosana*, *P. fortunei*, *Photinia* × *fraseri* and *R. simsii* are able to tolerate extreme high concentrations of As in the soil. Pb generally exists as precipitates of carbonate, hydroxide, and phosphate in soil [93], and As generally exists in sulfide minerals or as precipitates of insoluble arsenate in soil [94]. The low availability of Pb and As limits their uptake by plants, so there are some limitations to the phytoremediation of Pb and As. Although the BCFs and TFs of these plants for Pb and As are far < 1, these plants are still valuable resources for the remediation of Pb and As pollution in tailing areas, and the specific mechanisms of their tolerance to HMs need further study.

Table 3. Woody plants with potential for remediation of heavy metal pollution in study areas.

Function	Heavy Metal	Dominant Plant
Phytoextraction	Cd	*P. elliottii*, *R. simsii*
	Mn	*Q. glauca*, *L. formosana*
	Zn	*P. fortunei*
Ultra-high tolerance	Pb, As	*C. sclerophylla*, *R. chinensis*
	As	*L. formosana*, *P. fortunei*, *Photinia* × *fraseri*, *R. simsii*

5. Conclusions

A total of 12 dominant woody plant species in the study areas showed good adaptability to HMs. The woody plants tend to have a stronger enrichment and transport capacity for Cd, with BCFs > 1 and TFs > 1 in most plants. Although no hyperaccumulators were found in this study, some woody plant species with HM remediation ability, such as *P. elliottii* and *R. simsii* for Cd, *Q. glauca* and *L. formosana* for Mn, and *P. fortunei* for Zn, were screened as phytoextraction plants in HM-polluted soils, which provide a reference for future species selection for HM soil remediation. The specific tolerance mechanisms of *C. sclerophylla*, *R. chinensi*, *L. formosana*, *P. fortunei*, *Photinia* × *fraseri*, and *R. simsii* to HMs need further study, and they may be potential woody plant species for restoration after Pb and As pollution in the future. There may be potential effects between soil AP and AK on the uptake of HMs by woody plants, and a reasonable addition of phosphorus and potassium to the soil may be helpful for plants to resist the toxic effects of HMs.

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