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# Phytoremediation of multi-metal contaminated copper tailings with herbaceous plant and composite amendments

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# Abstract

After the closure of the large tailing pond of copper sulfide mine, ecological restoration must be carried out. The ecological restoration method of directly using tailing as a vegetation matrix to replace guest soil is a new method. The key to this method lies in improving the substrate environment and carry out phytoremediation. A field test was performed to carry out an enhanced phytoremediation technique for multi-metal contaminated copper tailings by Sudan grass (*Sorghum sudanense* (Piper) Stapf.), ryegrass (*Lolium perenne* L.), and Bermuda grass (*Cynodon dactylon*), using conditioner (TH-LZ01) and straw combination into composite amendments as soil amendments, aimed to obtain the maximum of phytoremediation effect. The 5% conditioner and 0.5% straw ($C_2S_2$) were recommended as the optimum chemical proportions for amendment materials. We could find that the chlorophyll, catalase activity, plant length and fresh weight in the 5% conditioner and 0.5% straw ($C_2S_2$) applied treatments were notably higher when compared with those of other treatments, indicating that composite amendments could alleviate the toxicity of metals to plants. In addition, adding composite amendments can obviously improve the restoration effect of Sudan grass, ryegrass and Bermuda grass on the pH, water content, electrical conductivity and organic matter of copper tailings. Compared with no treatment, the absorption of metal in leaves of plants treated with composite amendments is lower, but that in roots is enhanced, revealing that the stability of metal in roots is enhanced by composite amendments treatment to a great extent. By applying composite amendments, the metal removal effect of ryegrass appeared to be particularly effective, and by the following order: Cd (48.5%) > Zn (45.8%) > Pb (27.7%) > Cu (21.9%), followed by Sudan grass and Bermuda grass.


# 1 Introduction

Mineral mining inevitably leads to a large number of tailings, of which copper tailings emissions were as high as a billion tons (Si et al., 2022). The copper tailings have the characteristics of loose structure, severe habitat destruction, poor water retention, high heavy metal content, extreme pH, poor nutrient, etc (Soltani et al., 2017). The environmental impact is persistent, universal, serious and regional (Valentín-Vargas et al., 2018). The massive storage of tailings not only occupies a large number of land resources, causing serious environmental pollution, but also, the heavy metals and other pollutants contained in the tailings are lost through runoff, leaching and other ways, causing pollution to the soil, water and air around the mining area, and entering the human body through the food chain enrichment, threatening human health (Kasowska et al., 2018; Xie et al., 2020; Chen et al., 2015). Therefore, finding efficient methods to improve tailings and control the migration of heavy metal pollutants is very important. The common methods of tailings remediation include physical remediation, chemical remediation and phytoremediation (Cai et al., 2021a). Traditional physical and chemical remediation methods have disadvantages such as high repair cost, easy to damage soil structure, poor stability effect, and easily cause secondary pollution (Cai et al., 2021b; Safari et al., 2015). Phytoremediation has been widely concerned for its high efficiency and environmental friendliness and has a good application prospect. The key to ecological restoration of tailings reservoir is vegetation reconstruction. Due to the high content

of heavy metals in tailings and the lack of nutrient elements (such as N and P), the growth of plants in the tailings reservoir is limited. It is a new method to treat copper tailings by directly using tailings as vegetative substrates instead of guest soil. Amendments are added to tailings, heavy metal-resistant plants are planted simultaneously, and soilless reclamation vegetation restoration technology is established to stabilize the tailings reservoir, improve the landscape and control pollution (Acosta et al., 2018). The application of single amendments, which are often applied in large amounts or require repeated application, has the potential to damage the soil structure, increase the risk of surface and groundwater contamination, and cause plant nutrient deficiencies (Li et al., 2019a; Tariq et al., 2016). For example, excessive application of phosphorus-containing materials may cause eutrophication of water bodies. The research shows that the compound addition of two or more amendments can effectively improve the condition of poly heavy metal pollution (Radziemska et al., 2019). Wang et al. (2018) used pig manure, lime, dust, mushroom slag and other materials as amendments for lead and zinc tailings. They found that lime and pig manure can increase the pH value of tailings, thereby reducing the effectiveness of Pb and Zn. Chiu et al. (2006) applied manure and domestic sludge to lead and zinc tailings and at the same time selected vetiver grass (*Chrysopogon zizanioides* L.). and ryegrass (*Lolium perenne* L.), which had strong tolerance and found that the contents of N, P and K in tailings environment could be increased. It can promote plant growth and effectively reduce the availability of Pb, Zn and Cu in tailings.

Organic fertilizers, peat, medical stone, sludge, calcite, frankincense and other organic and inorganic amendments are often used to stabilize contaminated soil (Lou et al., 2017; Elyamine et al., 2018). Through adsorption, precipitation and ion exchange, these amendments changed the occurrence form of heavy metals in polluted soil, reduced the biological availability of heavy metals, and changed the physiological and growth state of repair plants (Shi et al., 2017; Zhang et al., 2019). Because of its rich nutrients and organic matter, crop straw can improve the matrix's water retention ability. Its nutrient release is slow, and the nutrient use time is long; it can chelate heavy metals, reduce toxicity, and is widely used in the matrix improvement of heavy metal tailings (Tang et al., 2020; Ogawa et al., 2020).

Combining the amendments and heavy metal-resistant plant to remediate, realizing the soilless reclamation, and the in-situ remediation method has the advantages of low cost, no damage to the site structure, no secondary pollution, relative reliability, environmental protection, and has broad application prospects (Bonilla et al., 2015; Wang et al., 2019; Samsuri et al., 2019). However, this technology is still in the laboratory research stage, with few field application experiments. In this paper, field experiments were conducted to study the effects of heavy metal-resistant herbaceous plants (Sudan grass, ryegrass, Bermuda grass) planted after adding the self-developed tailings conditioners and straw combination into the composite amendments on the physical and chemical properties and heavy metal content of the tailings, aiming to provide the theoretical basis for soilless reclamation and ecological restoration of the tailings reservoir.

# 2 Materials and methods

# 2.1 Study Area

The test area locates in Dexing Copper Mine, Sizhou Town, Dexing City, Eastern Jiangxi Province (29°0'3" N 117°43'33" E) (Fig. 1). The tailings pond covers an area of 26.961 $km^2$, the catchment area is 1.835 $km^2$, the height of the dam crest is 30.0 m, and the total storage capacity is 1640.6×104 $m^3$. The climate of the mining area is subtropical monsoon humid, dry and windy in spring, hot and rainy in summer, warm in autumn and cool at night, cold in winter and less snow. The annual average temperature reaches 17℃, whereas the extreme maximum temperature is 38.9℃, and the extreme minimum temperature is -9.7℃. In addition, the temperature difference between day and night is large, ranging from 5.4 to 15.6℃. Beyond that, the annual average rainfall maintains at 700–800 mm, and the year with the least rainfall is only 284.4 mm. Moreover, the test site is located in 4# tailing pond in Xiyuan Dagou, north of Dashan Concentrator, Asia's largest tailing pond.

# 2.2 Experimental material

The copper tailings samples in the current study were taken from the surface (5–15 cm) of 4# tailing pond (29°02'46.89" N 117°47'35.98" E) of Dexing Copper Mine in Jiangxi Province. Sudan grass, ryegrass and Bermuda grass were the plant raw materials, and the seeds were purchased from Jiangsu Qianhua Baimei Seed Industry Co., Ltd. There have been a lot of studies and elaboration on the preparation and application of conditioner for copper tailings fertilizer restoration by the previous staff of the research group (Wang et al. 2023). The conditioner preparation and determination process for fertilizer restoration and subsequent application methods are not described here. In addition, its components by weight percentage are 50 wt% peat, 5 wt% medical stone, 5 wt% calcite, 2 wt% frankincense, 35 wt% organic fertilizer, 2 wt% water retaining agent, and 1 wt% binder. We took straw from Manhu Village, Xinjian District, Nanchang City, Jiangxi Province (28°21'10" N 116°25'2" E). The plants selected were based on the previous experimental results of the research group (Ai et al. 2020).

The basic physicochemical properties of copper tailing and conditioner are shown in Table 1.

Table 1
The basic physicochemical properties of copper tailing and conditioner.

| Indicator | Copper tailing | TH-LZ01 |
|---|---|---|
| pH | 8.05 ± 0.09 | 6.42 ± 0.04 |
| EC (μS/cm) | 134.63 ± 5.97 | 3.93 ± 0.71 |
| SOC (g/kg) | 10.20 ± 0.20 | 39.86 ± 7.43 |
| WC (%) | 3.01 ± 0.06 | 15.33 ± 1.20 |
| TN (g/kg) | 0.06 ± 0.01 | 3.14 ± 0.12 |
| AP (mg/kg) | 5.15 ± 0.58 | 24.65 ± 88.44 |
| AK (mg/kg) | 8.28 ± 0.06 | 27.12 ± 268.76 |
| Total Cu (mg/kg) | 674.58 ± 12.95 | 7.22 ± 0.36 |
| Total Cd (mg/kg) | 2.71 ± 0.23 | 0.29 ± 0.08 |
| Total Pb (mg/kg) | 27.13 ± 2.57 | 3.14 ± 0.30 |
| Total Zn (mg/kg) | 409.26 ± 8.49 | 65.46 ± 3.84 |

EC (electrical conductivity), SOC (organic matter content), WC (water content), TN (total nitrogen content), AP (available phosphorus content), AK (available potassium content). TH-LZ01 (the self-developed tailings conditioners). The results are expressed as the mean ± standard error.

# 2.3 Preliminary experiment

The pre-experiment was performed on the rooftop of the 6th floor of the experimental building in the Nanchang Campus of Jiangxi University of Science and Technology for eight weeks. Ryegrass was used as plant material for thinning a week after germination. In addition, every 50 seedlings were moved to a 1500 g copper tailings pot (top diameter: 23.5 cm, bottom diameter: 12.8 cm, height: 14cm). Then, the chosen plants were grown based on six various soil treatments, respectively, CK (100% original tailings), $C_1$ (99% original tailings + 1% conditioner), $S_1$ (99.9% original tailings + 0.1% straw), $C_1S_1$ (98.9% original tailings + 1% conditioner + 0.1% straw), $C_2S_2$ (94.5% original tailings + 5% conditioner + 0.5% straw), as well as $C_3S_3$ (89% original tailings + 10% conditioner + 1% straw). In addition, three parallels were set for each group. In addition, when planting, the fixed root was watered carefully using extensive management. In this study, it needs to be pointed out that watering was carried out under the condition that there was no rain for a long period. Finally, five plants were selected by the five-point sampling method every week to measure the growth height and fresh weight of plants.

# 2.4 Field test

In July 2021, an experimental plot of 400 $m^2$ was established in the tailings pond, and the plot was divided into eight subgroups labeled as CK, CS, 1#, 2#, 3#, 4#, 5#, and 6#, respectively. CK was the control group of copper tailings without taking any measures, and CS was the control group without planting plants with the addition of composite amendments. Ryegrass, Sudan grass and Bermuda grass, planted with copper tailings without any measures were labeled as 1#, 2# and 3#, respectively. In contrast, ryegrass, Sudan grass and Bermuda grass, planted with the composite amendment were labeled as 4#, 5# and 6#, respectively. Each cell has an area of 10.0 m × 5.0 m and is arranged randomly. The spacing between the cells was 0.7 m, and a drainage ditch with a depth of 0.3–0.5 m and a width of 0.3 m was dug outside the perimeter of each cell. One week before planting, except CK, 1#, 2# and 3#, each plot was improved by a self-developed improver with an additional amount of 1 kg/$m^2$ and mixed with the surface tailing sand. After plowing, the sowing density was about 25 g/$m^2$, and the seeding method was broadcast. After seeding, add straw with a length of about 50 cm, and the covering density is 0.1 kg/$m^2$. Watering the root water when planting while adopting extensive management, only watering when there is no rain for a long time. After eight months of sowing, three quadrats were randomly selected from various fields, and three plants with similar growth were selected from each quadrat. The stem, leaves and root samples were obtained by using gardening scissors. A sample of copper tailings (10 cm deep) in the quadrate was then collected with a shovel and sifted with a 1.2×1.2 cm screen to remove larger stones. After mixing well, put it into a sealed sample bag by quartering method and place it in a low-temperature incubator. The mass of copper tailings in the sealed sample bag was controlled to about 100 g, and the water content, pH, electrical conductivity, organic matter and heavy metal of copper tailings were determined in the laboratory.

# 2.4 Determination method

Copper tailings pH was determined by the water extraction potential method (Zhou et al., 2015), organic matter content was determined by the potassium dichromate volumetric method and external heating method (Ma et al., 2021), water content was determined by oven drying method (Quan et al., 2017), electrical conductivity (EC) of copper tailings was measured by Orion162A conductivity meter (Sukweenadhi et al., 2018), and plant chlorophyll content was determined by ethanol extraction colorimetry (Akram et al., 2016). The ultraviolet absorption method (UV-1800PC) was used for catalase activity (Cha et al., 2015), and the heavy metals in the copper tailings matrix and plant body (aboveground and underground parts) were dissolved by aqua regia reflux method and nitric acid-perchloric acid method, respectively (Yao et al., 2022). The effective state of heavy metals was extracted by 0.005 mol/L DTPA. ICP-OES was used to determine heavy metals (Rudin et al., 2017).

# 2.5 Data analysis and processing

The data obtained by the experiment were processed and analyzed by adopting Excel 2010, SPSS 20 and SPSSAU online data analysis platforms. The Origin 9.1 software was employed for plotting.

The enrichment coefficient (BCF) was $BCF_{root}$ and $BCF_{shoot}$, respectively, indicating the enrichment ability of plant roots and aboveground parts to heavy metals in soil (Yanqun et al., 2005). At the same time, the

transport coefficient (TF) denotes the ability of plants to transfer heavy metals from roots to aboveground parts (Yoon et al., 2006). The calculation formula is presented as follows:

$BCF_{root} = C_{root}/C_{soil}$

$BCF_{shoot} = C_{shoot}/C_{soil}$

$TF = C_{shoot}/C_{root}$

where $C_{soil}$, $C_{shoot}$ and $C_{root}$ refer to absorptions of Cd, Cu, Pb and Zn ($mg \bullet kg^{-1}$) in soil samples, aboveground parts and roots of plants, respectively.

With the purpose of assessing the capability of the plants to absorb the metal ions from the soil, the removal rate (%) of heavy metals could be calculated using the formula (Li et al., 2019b):

Removal rate (%) = $(C_0\text{-}C)/C_0 \times 100\%$

where $C_0$ and C suggest the heavy metal concentrations ($mg \bullet kg^{-1}$) before and after the experiment separately.

# 3 Results

# 3.1 The plant growth responses

Chlorophyll content in plants is closely related to photosynthesis and nutrient absorption. As shown in Fig. 2 (a), the changing trend of chlorophyll content of plants under different treatment measures is the same. The chlorophyll content of ryegrass under the improvement measure of applying 5% conditioner and covering 0.5% straw ($C_2S_2$) in the composite treatment has the maximum value, and the chlorophyll content of ryegrass under the treatment of 100% copper tailings (CK) has increased by 129.84%. Compared with the control group, the chlorophyll content of ryegrass treated with 1% conditioner ($C_1$) and 0.1% straw ($S_1$) was not significantly different ($P > 0.05$), indicating that the single application of conditioner or straw had no significant effect on the chlorophyll content of plants. Compared with the control group, the chlorophyll content of ryegrass under the combined treatment of conditioner and straw was significantly different ($P < 0.05$). It can be seen from the figure that the effect of $C_2S_2$ treatment on the chlorophyll content of plants is higher than that of other combined treatments.

CAT is an important antioxidant enzyme and plays an important role in plant resistance to stress. The catalase activity of plants is shown in Fig. 2 (b). There was no significant difference in the catalase activity of ryegrass under the conditions of single-factor treatment with 1% conditioner ($C_1$) and single-factor treatment with 0.1% straw covered ($S_1$) ($P > 0.05$). Compared with the control group, the catalase activity of ryegrass under all complex treatments had significant differences ($P < 0.05$), and the effect of treatment was better than that of single-factor treatment. The catalase activity of ryegrass under the

combined treatment of 10% conditioner and 1% straw ($C_3S_3$) decreased compared with that under the combined treatment of 5% conditioner and 0.5% straw ($C_2S_2$), indicating that the catalase activity of ryegrass was not higher with the added amount of conditioner and straw.

In accordance with plant height (Figs. 3a and 3b) and fresh weight (Figs. 3c and 3d), the optimum chemical ratio of composite amendments as the copper tailings amendments were identified in this study. Plant height and fresh weight increased with the increase in planting time, exhibiting a good linear positive correlation. With the increase in planting time, the height and fresh weight of the copper tailings treated with conditioner ($C_1$) were 6.67–15.30 cm and 0.8–5.2 g, respectively, compared with those of the straw treatment group ($S_1$) and the control group (CK). Sudan grass's height and fresh weight increased by 59.4-481.1% and 254.5–300.0%, respectively, when treated with composite amendments ($C_1S_1$) compared with $C_1$. Compared to $C_1S_1$ and $C_3S_3$, the plant height and fresh weight of Sudan grass treated with $C_2S_2$ are 21.9% and 2.6% higher, and 30.5% and 17.9% higher. As a result, $C_2S_2$ will be used as the experimental treatment of copper tailings in the later stage to satisfy the requirements of soil environmental quality without influencing the normal growth of plants.

# 3.2 Physicochemical properties of copper tailings

As presented in Fig. 4a, compared with the CK group's pH value, tailings with three herb plants planted directly without any amendment are featured with no significant change, ranging from 7.91 to 8.18. Nevertheless, the pH value of the tailings lowered slightly using composite amendments; that is, the pH value of the tailings in the CS group decreased by 7.33% relative to the control group. The pH value of the plant rhizosphere tailings in the tailings with the addition of composite amendments and planting herbaceous plants were decreased in comparison with that in the CS group ($p < 0.05$), which were 7.06, 7.12 and 7.03, respectively, indicating that adding composite amendments and planting herbaceous plants can reduce the pH of tailings. Additionally, the electrical conductivity of copper tailings repaired by herbaceous plants showed a decreasing trend. The order of electrical conductivity in three herbaceous habitats was 3# (Bermuda grass) > 1# (ryegrass) > 2# (Sudan grass), and the average soil conductivity is 120.20 $\mu s \bullet cm^{-1}$, 117.47 $\mu s \bullet cm^{-1}$ and 86.67 $\mu s \bullet cm^{-1}$, separately. Moreover, the remediation effect of 3#, 1# and 2# groups is more obvious, which can be decreased by 34.34%, 29.61% and 31.46% relative to the control group, respectively, implying that the combined application of composite amendments can promote the electrical conductivity of copper tailings repaired by herbs.

The water and organic matter content of copper tailings in various habitats is shown in Fig. 4(b). The water content of copper tailings in the control group is too low (2.97%), which is not conducive to plant growth, whereas copper tailings with different treatments can increase to different degrees. Among them, the water content of copper tailings with Sudan grass is the highest, which is 4.59%, compared with other groups planting herbs without amendments. Compared with other treatment groups, the water content of copper tailings is shown to be significantly increased by applying amendments and planting ryegrass, which is 16.16%. The order of the organic matter content of copper tailings in various habitats is CS > 5#

(Sudan grass) > 6# (Bermuda grass) > 3# (Bermuda grass) > 2# (Sudan grass) > 4# (ryegrass) > 1# (ryegrass) > CK, with 29.72 g•kg$^{-1}$, 13.08 g•kg$^{-1}$, 12.51 g•kg$^{-1}$, 12.29 g•kg$^{-1}$, 10.67 g•kg$^{-1}$, 10.53 g•kg$^{-1}$, 10.34 g•kg$^{-1}$, and 10.20 g•kg$^{-1}$ respectively. Compared with CK, the ratios were about 2.91 times, 1.28 times, 1.23 times, 1.20 times, 1.05 times, 1.03 times, and 1.01 times of the control group, separately, showing that composite amendments have a certain repair impact on the organic matter of copper tailings.

# 3.3 The uptake of heavy metals in plant tissues

The contents of Cd, Cu, Pb, and Zn in plant tissues are displayed in Fig. 5. Cd contents in the roots and leaves of the three herbs directly planted without any amendments are as follows: ryegrass (10.23 mg•kg$^{-1}$) > Bermuda grass (8.01 mg•kg$^{-1}$) > Sudan grass (6.93 mg•kg$^{-1}$). In contrast, the Cu content is ryegrass (2033.74 mg•kg$^{-1}$) > Bermuda grass (1762.44 mg•kg$^{-1}$) > Sudan grass (1616.04 mg•kg$^{-1}$), and the Pb content is Sudan grass (27.01 mg•kg$^{-1}$) > ryegrass (20.10 mg•kg$^{-1}$) > Bermuda grass (18.71 mg•kg$^{-1}$). In addition, Zn content is ryegrass (504.93 mg•kg$^{-1}$) > Sudan grass (475.75 mg•kg$^{-1}$) > Bermuda grass (458.59 mg•kg$^{-1}$). With the use of composite amendments, the contents of Cd and Cu in tissues of ryegrass, Sudan grass and Bermuda grass decrease to a certain degree (Figs. 4a and 4b). Specifically, the contents of Cd and Cu in roots are 3.59, 4.18 and 4.17 mg•kg$^{-1}$, and 1004.55, 1065.45 and 1114.48 mg•kg$^{-1}$, respectively, while those in leaves are 1.07, 1.30 and 1.01 mg•kg$^{-1}$, and 134.98, 108.64 and 135.18 mg•kg$^{-1}$, accordingly. However, Pd and Zn contents in plant tissues increased (Figs. 3c and 3d). Moreover, compared with CR, CV and CF, Pd content in ryegrass and Bermuda grass was 46.77% and 43.19%, while Zn content in Sudan grass and Bermuda grass was 8.41% and 10.51%, respectively.

# 3.4 The translocation and bioconcentration factors of heavy metals

The enrichment coefficient (BCF) and the transport coefficient (TF) can evaluate the ability of plants to extract and transport heavy metals from copper tailings to aboveground parts. TF, $BCF_{shoot}$ and $BCF_{root}$ values of the three herbaceous plants under different treatments are shown in Fig. 6. According to TF data, Zn is the most easily transferable metal, followed by Cd, Cu, and Pb. The $BCF_{root}$ of the three herbaceous plants for Cd and Cu is all greater than 1.00, and it ranges from 1.74 to 2.61 and 1.41 to 1.95, respectively, indicating that the three plants have a strong capability of enriching Cd and Cu in the underground part of the soil. In addition, the $BCF_{shoot}$ of Cd, Cu, Pb and Zn is less than 1.00. Especially Pb and Zn are lower, and $BCF_{shoot}$ is 0.06 ~ 0.14 and 0.32 ~ 0.36, respectively. Compared with 1#, 2#, and 3# groups, TF and $BCF_{shoot}$ decreased by 7.5–70.6% and 7.1–72.9% based on the use of composite amendments, respectively, whereas the $BCF_{root}$ of Pb and Zn increased by 57.4% and 23.4% on average, accordingly.

# 3.5 Concentrations of total heavy metal, the bioavailability and removal rates of heavy metals

Based on the analysis and determination of the full amount of heavy metal in copper tailings (Fig. 7a), the total amount of heavy metal, namely Cd, Cu, Pb and Zn, in copper tailings of CK was lower than that of the three plant treatments. The contents of Cd, Cu and Pb in 3# treatment are lower than those of 1# and 2# treatments, while the content of Zn in 3# treatment is higher than that of 1# and 2# treatments. Additionally, the three treatments showed no obvious difference in Cd, Pb and Zn contents. The contents of Cd, Cu, Pb and Zn in 4# treatment increased by 12.9%, 14.4%, 9.9% and 13.1%, respectively, using composite amendments. Compared with 3#, the contents of Cd, Cu and Zn in 6# treatment were elevated by 23.9%, 3.4% and 6.1%, separately. These results demonstrate that amendments can stabilize heavy metals in copper tailings and foster plant growth, thus effectively controlling heavy metal loss with soil and water with plant roots.

Figure 7b shows that the contents of effective Cd, Cu, Pb and Zn in copper tailings of the CK group are 0.29 $mg•kg^{-1}$, 46.24 $mg•kg^{-1}$, 1.11 $mg•kg^{-1}$ and 45.42 $mg•kg^{-1}$, separately. In comparison with no amendments, the contents of available Cd, Cu, Pb and Zn in copper tailings treated by three plants have no significant reduction effect, and the reduction rates are 4.6–11.6%, 9.8–17.4%, 17.2–19.3% and 10.7–16.6%, respectively. However, using composite amendments, the available content of heavy metals is decreased dramatically, with reduced rates of 27.4–43.7%, 42.0-50.9%, 38.9–52.0% and 29.8–38.7%, respectively. These results suggest that adding composite amendments to copper tailings can effectively lower the available content of heavy metals in copper tailings, thus weakening the toxicity of heavy metals in copper tailings. In addition, the removal rates of Cd, Cu, Pb and Zn in copper tailings under different treatments are shown in Fig. 6c. Among the herbs planted directly on the original copper tailings, Sudan grass possesses the highest removal rates of Cd, Cu, Pb and Zn, which are 12.1%, 9.8%, 6.7% and 13.8% respectively. In contrast, the removal efficiency targeting Cd, Cu, Pb and Zn by herbaceous plants is observably improved, and that of ryegrass is the most obvious, featuring 48.5%, 21.9%, 27.7% and 45.8%, respectively.

# 4 Discussion

Heavy metal pollution is a severe environmental concern, claiming plant productivity in contaminated areas. Even though certain plants can adapt to some levels of these stresses, their growth rates and ultimately survivals can also be influenced. According to the plant growth status in Fig. 2 and Fig. 3, compared with other treatments, plant chlorophyll content, catalase activity, plant height and plant fresh weight in pure copper tailings were lower, which was mainly due to the high heavy metal content in copper tailings, which reduced root vitality and limited the absorption of water and nutrients by seedlings, followed by the effect of sterility. The combined application of composite amendments significantly increases the chlorophyll, catalase activity, plant length and fresh weight, which can create a favorable ecological niche for phytoremediation. The physicochemical properties of copper tailings are mainly enhanced because the conditioner provides sufficient nutrients for plants (Bi et al., 2017). In addition, due to the straw with porous structure, larger specific surface area, high cation exchange capacity and large surface functional groups, much heavy metal on the surface of ligand binding sites is provided. A large

number of complexes are formed, while it also becomes part of the heavy metal passivation that is difficult to be absorbed by the plant morphology, thereby lowering the toxicity of heavy metals to plants (Wang et al., 2021). However, it indicates that the higher the dosage of composite amendments, the better the plant growth and repair effect, which is similar to the previous study by Li et al. (2019), when 5%, 10% and 15% attapulgite and biochar are supplemented to the soil contaminated by complex heavy metals, and 10% attapulgite and 10% biochar generate the best passivation effect.

Figure 4 shows that the application of composite amendments is beneficial for the restoration effect of herbaceous plants on pH, water content, electrical conductivity and organic matter of copper tailings. As a certain number of alkaline substances, including lime and calcium hydroxide, are put into mineral processing to recover $Cu^{2+}$ in acidic mine wastewater, copper tailings are alkaline (Mustapha et al., 2019). The pH of copper tailings is elevated slightly after being repaired by different plants, maybe due to the fact that copper tailings are the products of the beneficiation process and contain a large number of heavy metals, such as Cd, Cu, Pb, and Zn (Ma et al., 2016). When combining composite amendments remedies the copper tailings, the enrichment of heavy metals in copper tailings by plant roots is promoted. Meanwhile, $Cd^{2+}$, $Cu^{2+}$, $Pb^{2+}$ and $Zn^{2+}$ in copper tailings are enriched in plant roots, and the pH value of copper tailings shows a certain downward trend (Geng et al., 2019). On the other hand, the conditioner contains a large amount of peat, the pH of peat is acidic, which can effectively neutralize the alkaline degree of copper tailing and may be related to the selective enrichment of some basic ions in the rhizosphere of these herbs to reduce soil alkalinity (Liu et al. 2018). Composite amendments, three plants backfilling materials processing of copper water content increased significantly, mainly because water retention agents can effectively inhibit the evaporation of conditioner, improve copper backfilling of saturated water content, reduce the saturated hydraulic conductivity of copper backfilling, and slow the rate of copper backfilling release water, thus reducing the backfilling materials copper water infiltration and erosion (Peng et al., 2021). In addition, plant roots can reduce soil erosion from surface runoff or wind erosion, enhance aggregate soil structure, and increase copper tailings' water retention capacity (Pang et al., 2003). The decrease in electrical conductivity of each group after phytoremediation can probably be due to the fact that the water content of copper tailings before remediation is much lower than that of the experimental group (Zhu et al., 2020). In addition, the increase in water content of each group after phytoremediation influences the salinity of copper tailings, leading to the decreasing electrical conductivity of each group after phytoremediation (Lin et al., 2019). Furthermore, the organic matter of copper tailings increases significantly using the composite amendments, mostly due to that the organic fertilizer in straw and the peat and organic fertilizer in the conditioner are rich in organic matter, thereby increasing the organic matter content of copper tailings (Shi et al. 2017). Relative to the CS group, the organic matter content of the herbaceous plants planted after adding the amendment decreases, and ryegrass consumes the most organic matter in copper tailings, which may be caused by the short growth time of herbaceous plants, and the greater consumption of organic matter than the generation, causing the reduction of organic matter content in copper tailings (Cortes-Flores et al., 2020).

The concentration of heavy metals in plants reflects the bioavailability of heavy metals and the ability of plants to absorb, transport and accumulate heavy metals. Figure 5 shows that the heavy metal content in the overground part is lower than that in the underground part, which may be related to the Cd, Cu, Pb, Zn super accumulation plants of ryegrass, Sudan grass and Bermuda grass (Ding et al., 2017; Liu et al., 2016). More Cd, Cu, Pb, and Zn can be accumulated in the stems and leaves of the overground part. Using composite amendments, the concentration of Cd and Cu in plants decreases, which may be associated with the "dilution effect" of plants; that is, the concentration of heavy metals in plants growing in soil decreases with the increase of plant biomass; in other words, it is equivalent to the concentration of heavy metals diluted by more biomass (Xie et al., 2018). The supplementation of organic amendments can reduce the absorption of heavy metals in the parts mentioned earlier of plants in an effective way, which may relate to the adsorption and fixation of heavy metals by the amendments and the formation of relatively stable complexes through ion exchange (Wang et al., 2020). The concentrations of Pb and Zn in plant roots increase under 4#, 5# and 6# treatments compared with those without amendment, conforming to the previous studies by Radziemska et al. (2019), finding that the use of soil amendments significantly increases the accumulation of heavy metals, especially Pb, Zn and Cr, in the red fescue root system. Here, it should be noticed that the increased heavy metal content in plant roots may result from releasing heavy metal chelators by plant roots, which provides more binding sites (Yan et al., 2020). In addition, soil amendments that lead to larger root-specific surface areas may also result in heavy metal accumulation in roots (Bossolani et al., 2021).

The enrichment and transport coefficients listed in Fig. 6 show that ryegrass, Sudan grass and Bermuda grass used in this study are classified as non-hyper enriched plants of Cd, Cu, Pb and Zn, with $BCF_{shoot}$ of target metals being all greater than one and BCF values being much higher than those of aboveground parts. Some studies indicate that most metals, including Pb, Zn, Mn and Hg, are not easily transported in plant tissues because of their high toxicity. Samsuri and Liu et al. (2019, 2020) reported that ryegrass and Sudan grass had a strong ability to absorb metals, while metal transfer from roots to aboveground parts was relatively low, except Pd and Zn. Lou et al. (2017) found that Bermuda grass had high Cd accumulation and transfer capacity. In addition, each metal has a different binding ability that influences its enrichment and adsorption/desorption processes in plants. According to the transport coefficient and enrichment coefficient results, the order of transport and accumulation capacity of the four heavy metals in plants is Zn > Cd > Cu > Pb. Among them, Pb has the lowest mobility, which is mainly due to the relatively low mobility of Pb itself. In addition, compared with other heavy metals, Pb is an immobilized heavy metal, which can be easily combined with the carboxyl group in the cell wall, and the root acts as a physical barrier through the Casparian zone in the endoderm (Zhang et al., 2020). TF and BCFshoot decreased significantly compared with 1#, 2#, and 3# groups. At the same time, $BCF_{root}$ increases significantly using composite amendments, probably because soil conditioners can decrease the binding of metal ions to extracellular cation exchange sites in the extracellular body and the cell wall (Lin et al. 2019). Thus, the heavy metal flux through the ectoplasm into the root of the stele is increased.

The total amount, available state and removal rate of heavy metals in contaminated soil are affected by heavy metals, amendments and plant species. Figure 7a shows that after planting ryegrass, Sudan grass and Bermuda grass, the total amount of heavy metal in copper tailings increases to a certain extent, mainly because the forms of heavy metals in soil are seriously affected by pH value (Kinuthia et al., 2020). After planting plants, the pH value of copper tailings increases, which can reduce the bioavailability of heavy metals, including Cu, Zn and Cd, since the increase of pH can increase the number of negative charges on the surface of soil particles, which plays a main role in the fixation of heavy metal ions on soil particles; in addition, the conversion of heavy metals in soil into more stable hydroxide or carbonate precipitation is promoted (Su et al., 2019; Hamid et al., 2018). Using amendments, the total number of heavy metals in copper tailings increases compared with 1#, 2# and 3#, probably due to the fact that organic amendments such as conditioner and straw include a large number of organic acids, including folic acid and amino acids (Karami et al., 2011). Groups rich in these organic acids, such as -COOH, -OH and -CHO, can react with heavy metals in a complex way, which can lower the activity of heavy metals and enhance their stability in the soil, further increasing the difficulty of absorption by animals and plants (Huang et al., 2020). Figure 7b reveals that three herbs and composite amendments repair copper tailings under different treatments. It is possible that some proteins or polypeptides in plant roots can be combined with Cd, Pb, Cu and Zn to form precipitation or binding states, reducing the content of effective heavy metal states in the soil to protect plants from heavy metal toxicity (Chen et al., 2019). Huang et al. (2021) claimed that organic amendments could notably change the distribution of Cd, Pb, Cu and Zn forms in soil from easily exchangeable to bioavailable forms. The organic fertilizer in the conditioner can chelate with heavy metals. Since straw has a large surface area and is rich in oxygen-containing functional groups, including carboxyl and phenol hydroxyl, heavy metals can be bonded to the surface of straw through chelation, which can thereby lower the bioavailability of heavy metals (Zheng et al., 2019). As shown in Fig. 7c, without adding amendments directly after planting three kinds of herbs, all target metal removal rates are lower than 20%, concerning the removal efficiency of Sudan grass on Cd, possibly because the minimum content of Cd in contaminated soil, and Sudan grass as plants as the accumulation of Cd is relatively high; thus, the highest removal rate appears (Nugroho et al., 2021). Using composite amendments, the removal rate of all heavy metals elevates obviously, while the increase of Pd removal rate is the least, which may be related to the fact that Pd is not easily transported in plant tissues due to its high toxicity. In addition, the high specific surface area, high porous microstructure, abundant surface charges and surface functional groups of soil amendment may become the factors that can exert influence on the removal of heavy metals (Mahajan et al. 2020).

# 5 Conclusions

In the current work, three plant species, ryegrass, Sudan grass, and Bermuda grass, were chosen to assess their potential for field-scale experimental phytoremediation. Besides, the findings of plant growth parameters revealed a higher efficiency of 5% conditioner and 0.5% straw to phytoremediation for heavy metals concerning chlorophyll, catalase activity, plant length, biomass production, pH, water content, electrical conductivity, and organic matter were elevated notably. The combination effects of ryegrass

and composite amendments can be proposed as one of the best options for phytoremediation of heavy metal-contaminated soil. As a result, the method can also provide a green alternative to conventionally costly and environmentally unfriendly physical-chemical technologies.

# Declarations


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### Conflicts of interest/Competing interests

We claim that we do not have any commercial or associative interest representing a conflict of interest in association with the work submitted.

### Availability of data and material

All data involved in the current work are available upon request by contact with the corresponding author.

### Code availability

Not applicable.

### Authors' contributions

Weiwei Wang carried out the data analyses and wrote the manuscript;

Jinchun Xue contributed to the conception of the study;

Jiajia You contributed significantly to analysis;

Zhengliao Zhang and Hui Qi performed the experiment;

Xiao Zhang helped perform the analysis with constructive discussions;

### Ethics approval

Not applicable' for that section

### Consent to participate

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### Consent for publication

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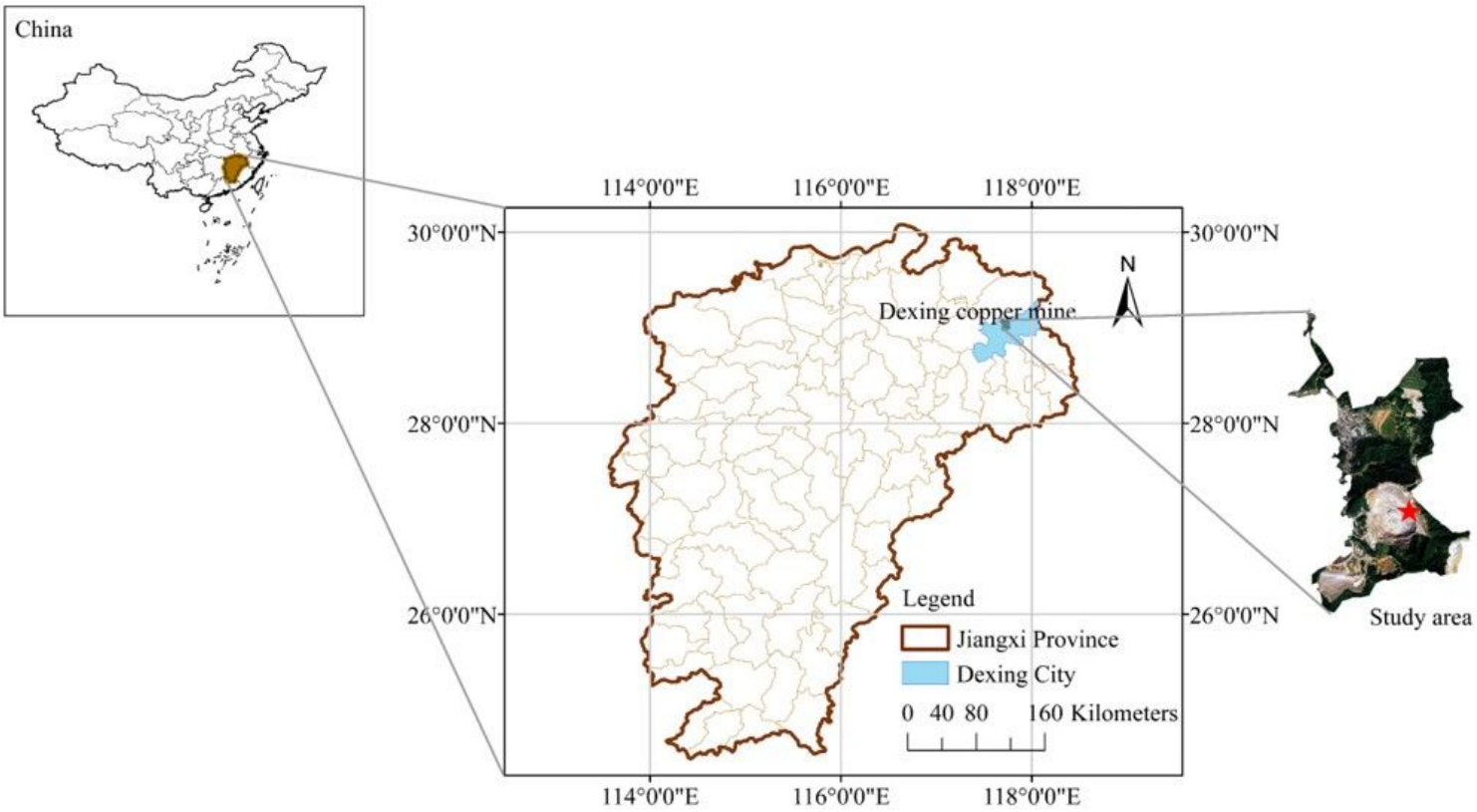
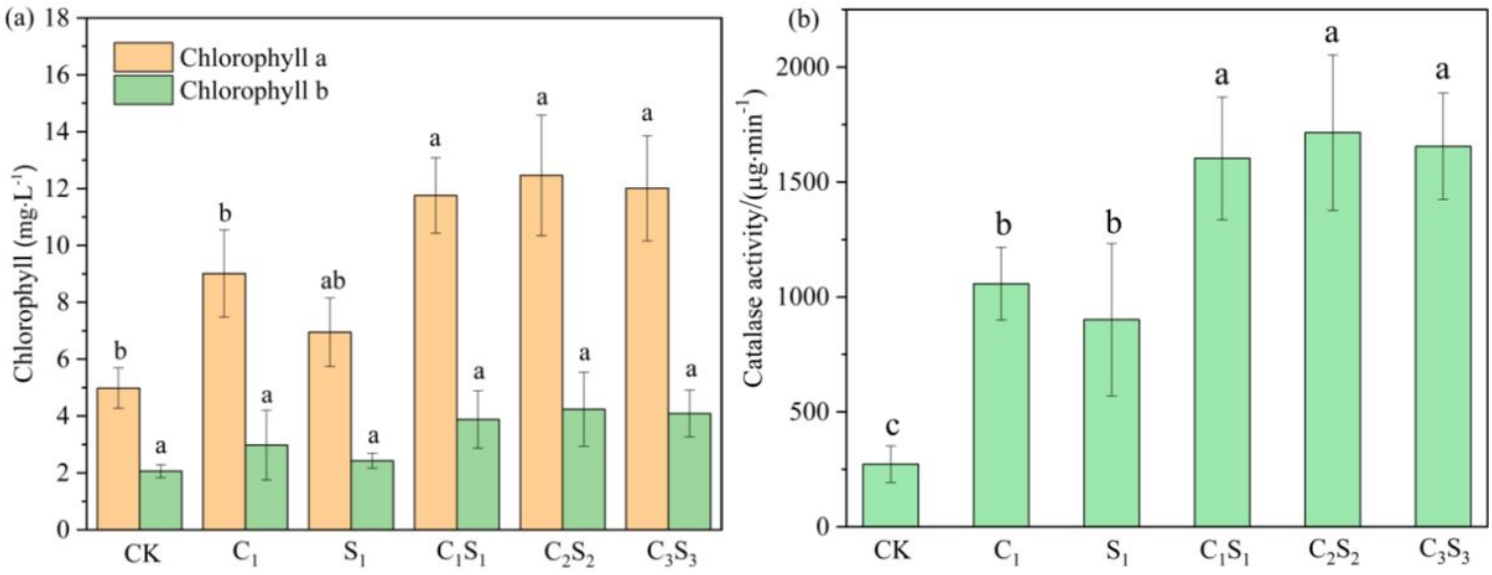
# Figures




## Figure 1

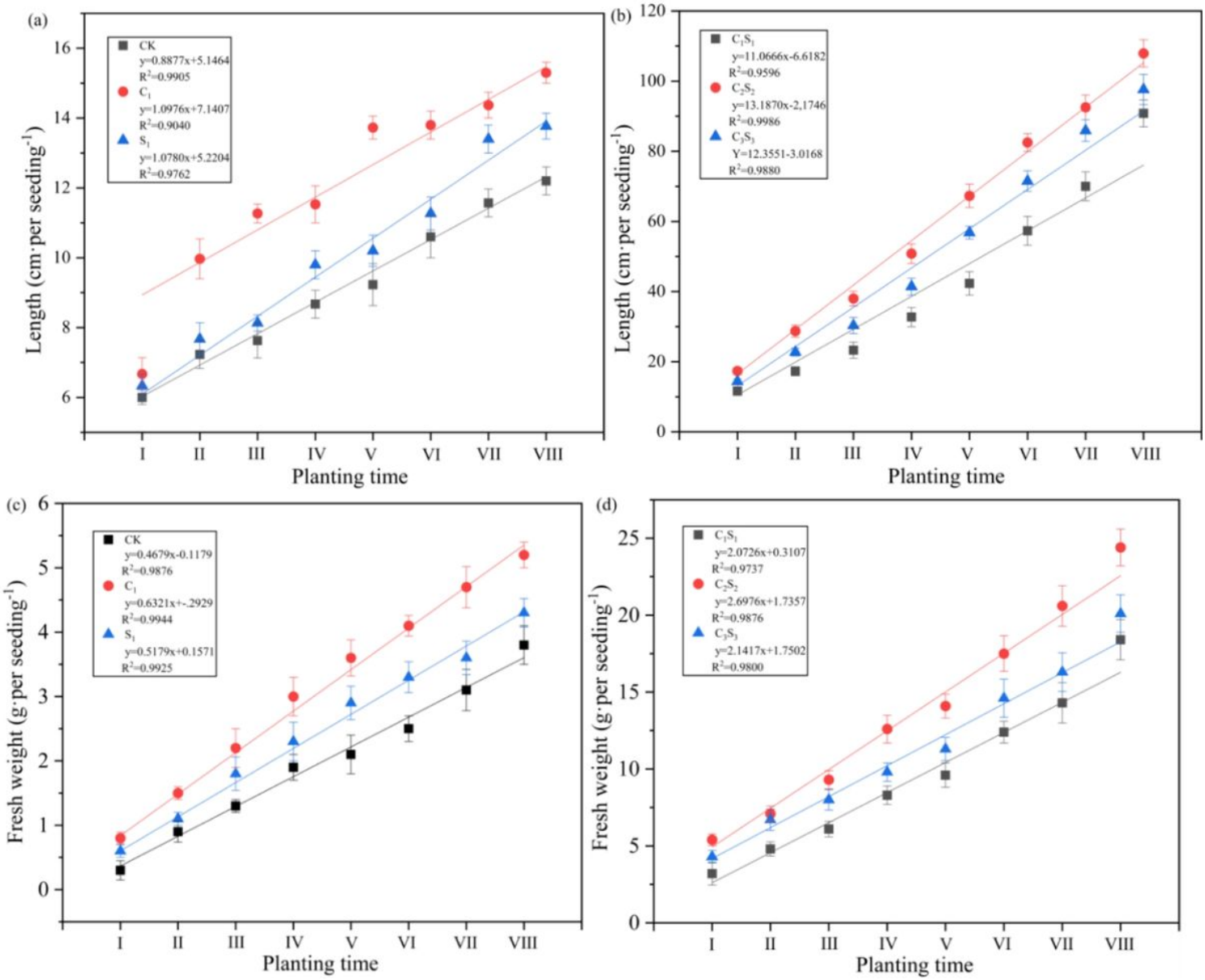
Geographical location map of the research district.




## Figure 2

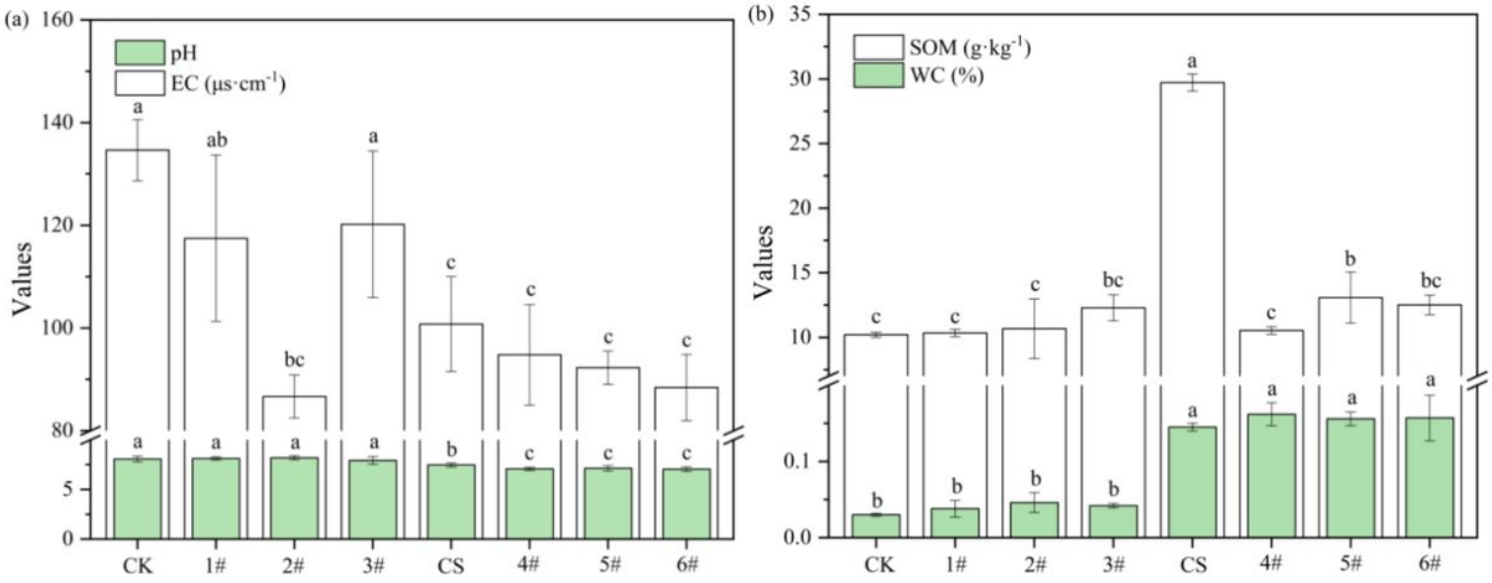
The effect of different treatments on chlorophyll and catalase activity. CK (100% original tailings), $C_1$ (99% original tailings +1% conditioner), $S_1$ (99.9% original tailings +0.1% straw), $C_1S_1$ (98.9% original tailings +1% conditioner +0.1% straw), $C_2S_2$ (94.5% original tailings +5% conditioner +0.5% straw), and

$C_3S_3$(89% original tailings +10% conditioner +1% straw). Error bars showed the standard deviation of three replicates. The same letter suggested no obvious difference between treatments ($P$>0.05).



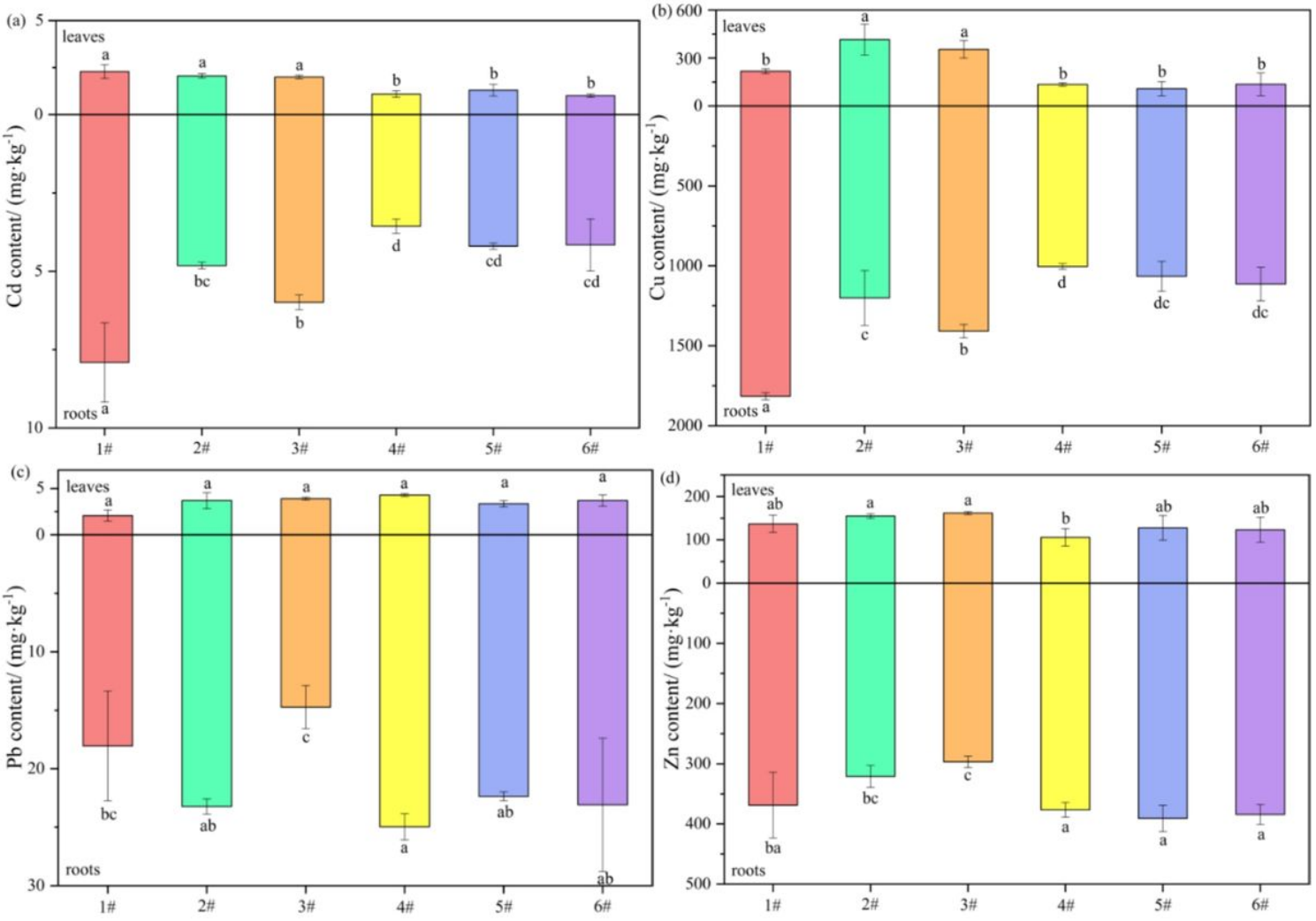

## Figure 3

Pearson's correlations between plant height, fresh weight and experimental time. CK (100% original tailings), $C_1$ (99% original tailings +1% conditioner), $S_1$ (99.9% original tailings +0.1% straw), $C_1S_1$ (98.9% original tailings +1% conditioner +0.1% straw), $C_2S_2$ (94.5% original tailings +5% conditioner +0.5% straw), and $C_3S_3$(89% original tailings +10% conditioner +1% straw). The first to the eighth weeks of planting time is denoted as , , , , , , , and respectively. Error bars showed the standard deviation of three replicates.



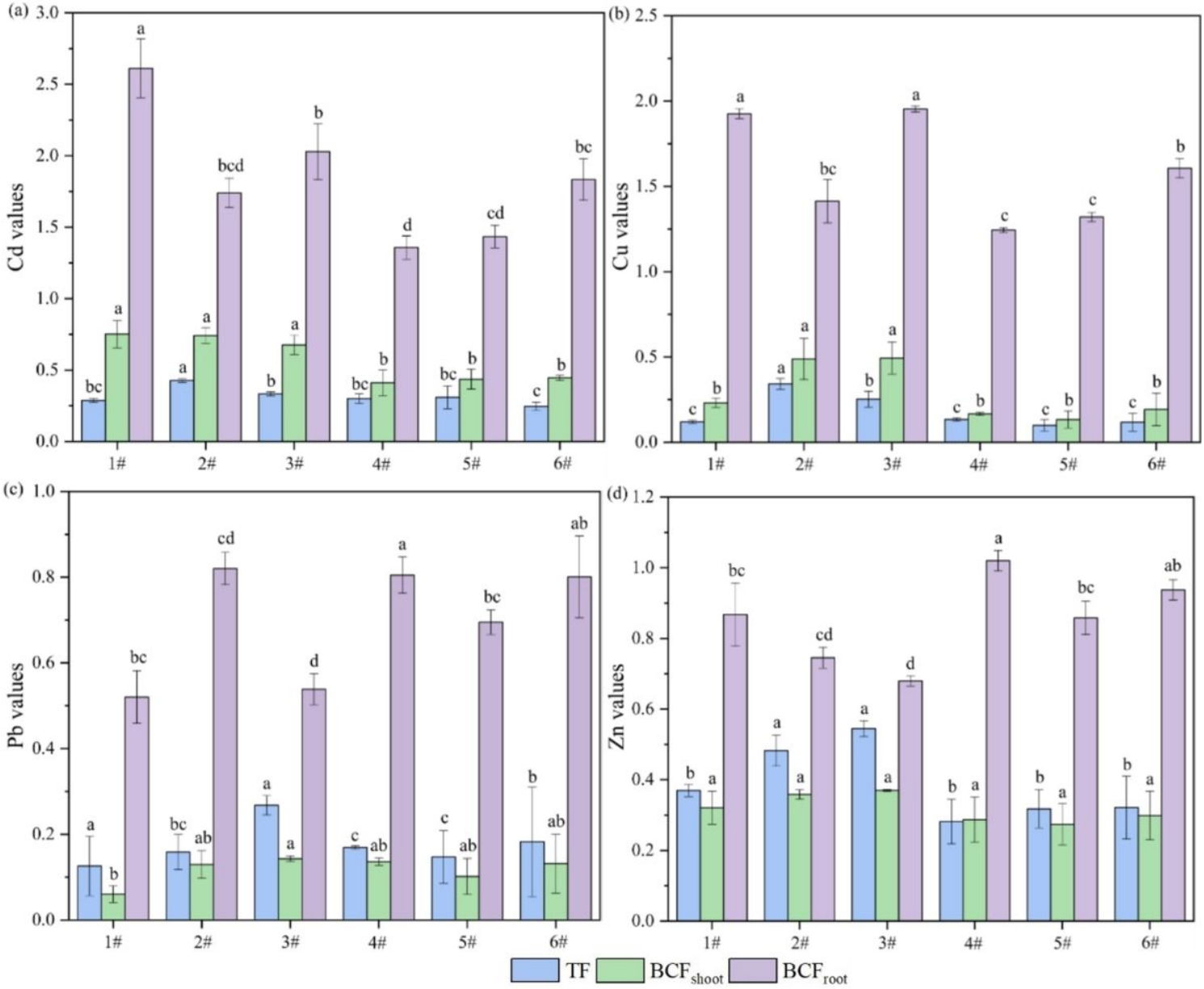

## Figure 4

The effect of different treatments on the physicochemical properties of copper tailings. The non-amended, amendment-assisted, non-amended or amendment-assisted and planted ryegrass, Sudan grass, and Bermuda grass were CK, CS, 1#, 2#, 3#, 4#, 5# and 6#, respectively. Electrical conductivity (EC), water content (WC), and soil organic matter (SOM). Error bars showed the standard deviation of three replicates. The same letter suggested no obvious difference between treatments ($P$>0.05).



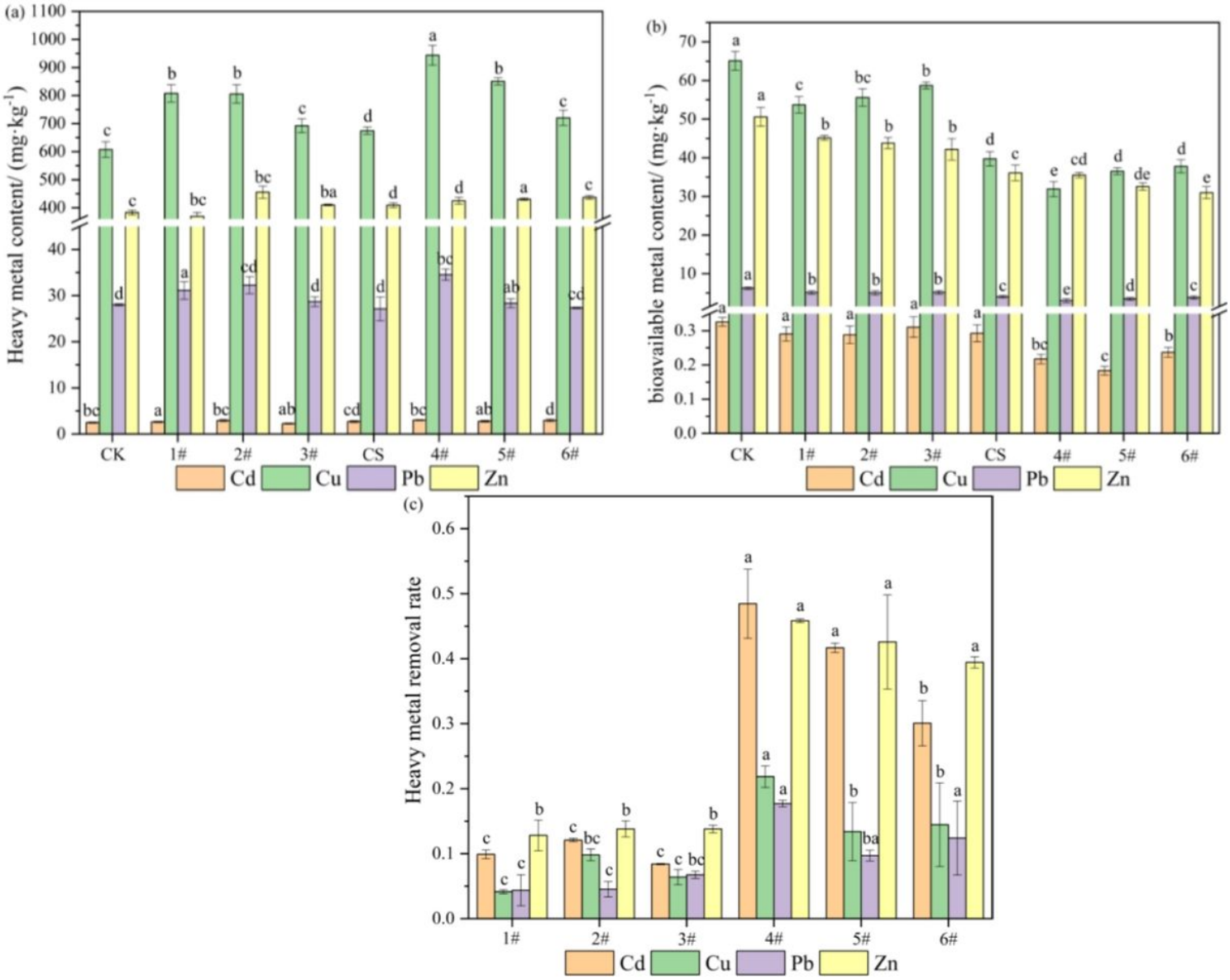

## Figure 5

Effects of different treatments on metal concentrations in plant tissues. The non-amended or amendment-assisted and planted ryegrass, Sudan grass, and Bermuda grass are referred to as CK, CS, 1#, 2#, 3#, 4#, 5# and 6#, respectively. Error bars represent the standard deviation of three replicates. Moreover, the same letter indicates no obvious difference between treatments ($P$>0.05).




## Figure 6

The translocation (TF) and bioconcentration ($BCF_{shoot}$ and $BCF_{root}$) factors of metals. The non-amended or amendment-assisted and planted ryegrass, Sudan grass, and Bermuda grass are referred to as CK, CS, 1#, 2#, 3#, 4#, 5# and 6#, respectively. Error bars represent the standard deviation of three replicates. The same letter indicates no obvious difference between the treatments ($P$>0.05).




## Figure 7

Effects of different treatments on the total amount, effective state and removal rate of metals. The non-amended or amendment-assisted and planted ryegrass, Sudan grass, and Bermuda grass are referred to as CK, CS, 1#, 2#, 3#, 4#, 5# and 6#, respectively. Error bars show the standard deviation of three replicates. In addition, the same letter indicates no obvious difference between treatments ($P$>0.05).