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Accumulation and Distribution of Rare Earth Elements (REEs) and Non-REEs With Vetiver Grass in The Abandoned Ion-Absorbed Rare Earth Mine

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Research Article

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Abstract

Overexploitation of rare earth elements (REEs) has caused serious desertification and environmental pollution, and ecological restoration of mines has attracted increasing national attention. In this paper, experiments involved land plowing, organic fertilizer broadcasting and vetiver cultivation were carried out to repair abandoned ion-absorbed rare earth mines (REM). Toxic metals content and pH in mining soil, distribution and transportation of toxic metals in the soil – vetiver grass system were investigated in detail.

Results revealed that the abandoned REM soil was weakly acidic (pH=4.09) and rich in various toxic metals composed of REEs (La, Ce, Nd, Y, Gd, Dy) of 657.57mg/kg and Non-REEs (Pb, Cu, Se, As, Cd) of 109.98mg/kg. The distribution pattern in vetiver grass illustrated that toxic metals accumulation was mainly concentrated in the roots instead of shoot, and then the cumulative concentration of REEs in roots were much greater than that of Non-REEs. Furthermore, vetiver grass exhibited preferential accumulation of Cd, Se and REEs during the absorption process (from soil to root) and preferential accumulation of Pb, Cu and As during the translocation process (from root to leaf). The adsorption behavior of toxic metals by vetiver was confirmed due to these observed irregular particles in the scanning electron microscopy.

Introduction

As a strategic resource, rare earth elements were widely used in various industries, such as clean energy (Judge et al 2017), agricultural (Abdelnour et al 2019), military and other fields (Massari et al 2013), especially, the middle and heavy REEs were closely related to cutting-edge technology products (Dushyantha et al 2020). Ion-absorbed rare earth ores were the main source of medium and heavy REEs, which was widely distributed at seven provinces in southern China, such as Ganzhou area, Jiangxi (Tang et al 2018). Numerous scholars (Bao et al 2008) have discovered that REEs attached to ion absorbed REM were easily desorbed through ion exchange when encountering with positive ions (such as Na+, NH_4^+ and H^+). However, the leaching process of REM using ammonium sulfate in forms of the pool leaching, dump leaching and in-situ leaching (Liu et al 2014) might not only induced the release of associated toxic metals (Liu et al 2020), but also resulted in a series of environmental issues, such as co-pollution of associated toxic metals and REEs, soil fertility degradation, land desertification, headwater pollution and downstream farmland damage (Xie et al 2020, Feng et al 2012, Zhou et al 2015). Furthermore, these pollutants (ammonium nitrogen, associated toxic metals and REEs) remained in REM potentially polluted the surface water and groundwater through surface runoff and leakage, which posed serious health risks to humans (Huang et al 2009, Rao et al 2017). Thus, abandoned rare earth mine disposal and restoration were especially urgent.

Recently, compared with traditional engineering techniques for remediating contaminated soil, phytoremediation was a convenient, cost-effective, non-destructive and promising solution for extracting pollutants from contaminated soil, especially the sites with contamination spread over a large area (Wang et al 2017, Cioica et al 2019, Punia 2019). It was well known that toxic metals and other pollutants in the contaminated soil were extracted by plant accumulation in harvestable shoot parts (phytoextraction) or root uptake and translocation into shoots (phytostabilization) (Pedron et al 2011, Banerjee et al 2016). After phytoextraction, these harvestable plants could be disposed of incinerated to provide thermal energy or recover valuable metals (Sas-Nowosielska et al 2004). However, restoration of ion-absorbed REM was extremely slow and difficult owing to the hostile growing conditions, such as lack of organic matter and toxic metals contamination (Pang et al 2003). Thus, the appropriate plant species selection to resist these unfavorable conditions was essential for rehabilitating abandoned rare earth mines.

Vetiver grass, as a fast-growing perennial C4 grass, possessed tall stem (1-2m) and extensive strong root system (up to 3-4m deep) (Vargas et al 2015). Moreover, vetiver was very popular in India due to its special economic value including its roots could be processed into volatile essential oil and leaves could be used as feeds for cattle, goats and horses (Chen et al 2020). Importantly, vetiver grass tolerated various adverse conditions, such as prolonged drought, extreme temperatures, acidity, alkalinity and high concentrations of toxic metals (Danh et al 2009, Roongtanakiat et al 2008). Thus, among various types of plants, vetiver grass was considered to be one of the most promising plants for mine restoration due to its unique morphological and physiological characteristics (Shu et al 2002). Nevertheless, recent studies of vetiver grass have solely reported on the soil erosion control (Mondal et al 2020), landfill rehabilitation and mine site stabilization, such as iron ore mine, gold mine tailings, coal mines and lead mine tailings (Banerjee et al 2019, Wari et al 2019). There were currently few researches that have been tested to evaluate the extraction capacity of REEs and Non-REEs using Vetiver grass from abandoned ion-absorbed rare earth mines.

Therefore, the objectives of the present study were (1) to determine physical-chemical parameters and toxic metals content of contaminated sites, (2) to explore the feasibility analysis for vegetation restoration of rare earth mine, (3) to investigate toxic metals uptake ability of the roots and shoots of vetiver and (4) to assess the transportation of these toxic metals in the soil-vetiver grass system.

Materials And Methods

Study area

The experiments were conducted in the abandoned ion-adsorption rare earths mines, had a high background level of Non-REEs and REEs in the mines, located at Dingnan County, Ganzhou City, Jiangxi Province (Fig. 1). The climate of the study site was warm and moist, with a mean annual temperature of 18.5–19.5°C and a mean annual precipitation of 1440-1556nm (Wei et al 2001).

Two abandoned heap leaching rare earth mines were selected for research in Dingnan County: REM-1 and REM-2, the site area of REM-1 and REM-2 were approximately 664.75m2 and 709.66m2, respectively. Two selected test sites had become exposed hillside and sandy soil landfill, and almost no plants can survive. The main reason was that ammonium sulfate was used to leach rare earth mines, resulting in a large amount of ammonia nitrogen and toxic metals remaining in the mines, which inhibited plant growth (Fig. 1a, c).

Planting experiment and sample preparation

The experiments land was evenly broadcasted with 75 g/m2 organic fertilizer at the soil surface, and then plowed to a depth of 80cm with excavator. Vetiver grass seedlings were purchased and each fresh plant sapling was pruned (the shoots were originally 15–25 cm high and the roots 5–15 cm long). The pretreated vetiver seedlings were planted at 50-60cm row spacing, 40-60cm cluster spacing and 6–10 tillers for each slip. All plants were watered with running water every other day to ensure high survival rate during the first two months.

In each sampling site, the plants (root and shoot) were sampled at harvest and their corresponding rhizospheric soils was taken at different growth time (0, 36, 90, 164, 246d), and then REEs (La, Ce, Nd, Y, Gd, Dy) and Non-REEs (Pb, Cu, Se, As, Cd) content in samples were detected after drying and acid digestion. Before analysis, all the samples were stored at 4°C to keep fresh.

Soil and Plant Sample Analyses

All soil samples were sieved and dried to constant weight, and then underwent acid digestion with nitric acid (HNO_3), sulfuric acid (H_2SO_4) and phosphoric acid (H_3PO_4). The toxic metal content of soil sample was analyzed using atomic absorption spectrophotometry (AAS, AA6880, Shimadzu, Japan).

Freshly roots and shoots of Vetiver were washed with deionized water to remove any adhering soil particles, and then were oven-dried for 72 h at 70°C until it achieved a constant weight. The plant samples were dissolved with nitric acid (HNO_3), sulfuric acid (H_2SO_4) and phosphoric acid (H_3PO_4) after it was homogenized in a mortar and pestle for the elemental analysis using inductively coupled plasma mass spectrometry (ICP-MS, Aglient 7700X, America) (Ng et al 2016).

The crystal morphology of plant samples (roots and shoots) was analyzed by scanning electron microscope (SEM, JSM-7500F, JEOL Ltd, Tokyo, Japan).

Data Analyses

The biological concentration factor (BCF) and biological accumulation coefficient (BAC) were defined as the ratio of toxic metals content in the roots and shoots to those in the soil, respectively. The translocation factor (TF) represented the ratio of toxic metals content in the above-ground part to those in the plant roots. Similarly, the percentage of metals uptake efficacy (MUE) were defined as the ratio of toxic metals content in the above-ground part to those in plants (Kafil et al 2019). These indicators were used to assess the ability for toxic metals accumulation and translocation upwards in the soil – vetiver grass system.

Results And Discussion

REM soil analysis and vegetation restoration

The physico-chemical parameters and toxic metals content in REM-1 and REM-2 prior to vetiver planting were presented in Table 1. The results indicated that the unrepaired soil pH of REM-1 and REM-2 were 4.45 and 4.09, respectively. Moreover, the total concentrations of Non-REEs in two experiment soils were 109.98mg/kg and 107.51mg/kg, respectively. Noticeably, Pb content was higher than other Non-REEs.

The total soil REEs (542.78 and 657.57mg/kg) in abandoned rare earth mine, which might become the main source of REEs in the future, were much higher than the average levels (177 mg/kg) for the soils in China (Liang et al 2005). In addition, within sampling sites, the Ce content (431.56 and 463.64 mg/kg) in the rare earth mine was higher than that of any other rare earth element, probably because the parent material of rare earth mine for experiment in Dingnan County was primarily granite, rich in high-content Ce. Compared with other REEs, Ce was more difficult to leaching from the leaching of rare earth mines with ammonium sulfate, which caused its higher abundance in abandoned REM. Furthermore, the concentration of Y and La were 89.90 and 55.58 mg/kg, which were higher than that of Dy and Gd.

The selection of appropriate plant species was thought to be a key step to successful revegetation. In this study, it was very successful that vetiver grass was selected as a plant to restore abandoned REM (Fig. 1b, d). It was obvious that vetiver grass exhibited a strong tolerance to acidic soil and various toxic metals, and good growth ability, which indicated that it was feasible to achieve vegetation restoration and ecological restoration with vetiver grass in mining areas.

Table 1
The physico-chemical parameters and toxic metal content in REM-1 and REM-2

Element	pH	Cu	Pb	As	Se	Cd	La	Ce	Nd	Y	Gd	Dy
REM-1	4.45	29.17	77.60	2.38	0.66	0.17	37.65	431.56	19.00	41.80	5.36	7.41
REM-2	4.09	34.60	69.30	2.69	0.74	0.18	55.58	463.64	27.90	89.90	7.71	12.84

Metal concentrations and distribution pattern in REM soil

Remediation and degradation experiments of Non-REEs in REMs were estimated, and the results were shown in Fig. 2. Compared with the other elements (Cu, As, Se and Cd) content, the residual pb content in REM-1 and REM-2 was the highest, the concentrations of other metals in REM-1 and REM-2 were in the order Cu > As > Se > Cd. Besides, a significant reduction in the content of all Non-REEs with the growth time of vetiver grass in rare earth mines was observed during the experimental period. The main reason was that the vetiver grass roots continuously absorbed and extracted these toxic metals from the soil during the growth process.

Toxic metals concentrations and distribution pattern in vetiver grass

Extraction experiments of Non-REEs (Pb, Cu, As, Se, and Cd) and REEs (La, Ce, Nd, Y, Gd and Dy) from REM with vetiver grass were analyzed, and the results were shown in Fig. 3. The concentrations of Non-REEs and REEs in roots were consistent with those in the initial soil, but those in the shoot were slightly different at harvest. This indicated that the adsorption capacity of vetiver was closely related to the metal content in the contaminated soils, which was similar to other research conclusions (Liang et al 2005). In addition, toxic metals concentration in the tissue of vetiver grass decreased in the order of root □ shoot, which showed that vetiver grass accumulated large amounts of metals in the roots and restricted their translocation to the shoots. These findings boldly speculated that when vetiver grass was used for the rehabilitation of sites contaminated with these metals, their roots can be centralized processing through harvested, while shoots can be safely harvested to feed animals due to these toxic metals were almost concentrated in roots, not in shoots.

As can be seen from Fig. 3, Among the Non-REEs (Pb, Cu, As, Se, and Cd) studied in the experiment, the concentration of Pb in root was far more than that of other Non-REEs, nevertheless, Cu content in the stems was the largest. Similarly, Unlike the stems, the REEs contents in the roots were extremely large, especially Ce (100.1mg/kg in REM-1) and Y (101.1mg/kg in REM-2), which revealed that vetiver grass was a hyperaccumulator plants for REEs.

The distribution and transportation of toxic metal in the soil-vetiver grass system

In order to study the accumulation and translocation of Non-REEs and REEs in the soil – vetiver grass system, the biological concentration factor (BCF), biological accumulation coefficient (BAC), translocation factor (TF) and percentage of metal uptake efficacy (MUE) were used to evaluate the absorption capacity of vetiver for Non-REEs and REEs (Fig. 4).

Obviously, the BCF value of Non-REEs and REEs were significantly higher than BAC value, indicating that these toxic metals absorbed by vetiver were mostly retained in the roots. Similarly, according to the lower TF value (Fig. 4b), it was found that Cd, Se and REEs performed extremely poor translocation capability from root to shoot,which indicated that the accumulation of these metals by vetiver mainly depend on the roots instead of shoot. In addition, different BCF and TF value in the Fig. 4 indicated that vetiver grass had selective absorption and translocation of Non-REEs and REEs in the soil – vetiver system. Figure 4a showed that vetiver grass roots had a strong accumulation capacity for Cd, Se and REEs (except Ce) due to their BCF value exceeding 1. On the contrary, the Pb, Cu and As accumulation by vetiver relies on the joint action of the root and stem of vetiver, due to their high TF value, the MUE value of these metals just confirmed this view.

SEM analysis

To observe clearly the surface topography of vetiver roots and shoot, the SEM micrograph of vetiver roots and shoot obtained at initial and harvest were shown in Fig. 5. As can be seen from the SEM micrograph of vetiver roots at planting (A) and harvest (B), the initial root had a smoother surface with larger gaps, while the harvested root surface was rough and rich in a lot of irregular particles. These observed irregular particles might be composed of Non-REEs and REEs extracted by vetiver grass from REM. In addition, the SEM micrograph of the vetiver shoots at planting (C) and harvest (D) showed noticeable differences in morphology. Figure 5C showed that the vetiver shoots at planting have sufficient moisture and abundant stomata, on the contrary, those at harvest were shriveled and had fewer stomata (Fig. 5D). FA Melato (Melato et al 2016) reported that excessive concentration of toxic metals in contaminated soils disturb the physiological system, including transpiration and respiration, which resulting in the reduction of water content and stomata in the plant. These changes in the plant stems in this study were in agreement with the results of previous studies (Singh et al 2004).

Conclusions

In this paper, the experimental soil was weakly acidic (4.09) and rich in various toxic metals composed of REEs (657.57mg/kg) and Non-REEs (109.98mg/kg) with the highest content of Ce. After 246 days of remediation with vetiver grass, Non-REEs concentrations in REM soil have decreased, at the same time, all toxic metals accumulation in vetiver grass was mainly concentrated in the roots instead of shoot, which indicated that the ecological restoration technology with vetiver was effective in repairing abandoned REM. In addition, vetiver grass was considered to be an effective REEs phyto-stabilizer, owing to the high considerably REEs accumulation in its roots. Furthermore, vetiver grass exhibited preferential accumulation of Cd, Se and REEs during the absorption process (from soil to root) and preferential accumulation of Pb, Cu and As during the translocation process (from root to leaf).

SEM micrograph results revealed that Non-REEs and REEs in the soil were absorbed and extracted mainly depended on vetiver roots. Besides, excessive toxic metals reduced the water content and the number of stomata in shoot, which might disturb the physiological system.

Declarations

Authors declarations

Ethics approval and consent to participate

Not applicable

Consent for publication

Not applicable

Availability of data and materials

All data generated or analysed during this study are included in this published article.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

CZF contributed to the conception of the study. CZF, KLH, LTT and LP performed the experiment. KLH performed the data analyses and wrote the manuscript. CZF and WLY helped perform the analysis with constructive discussions. All authors read and approved the final manuscript.

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Figures

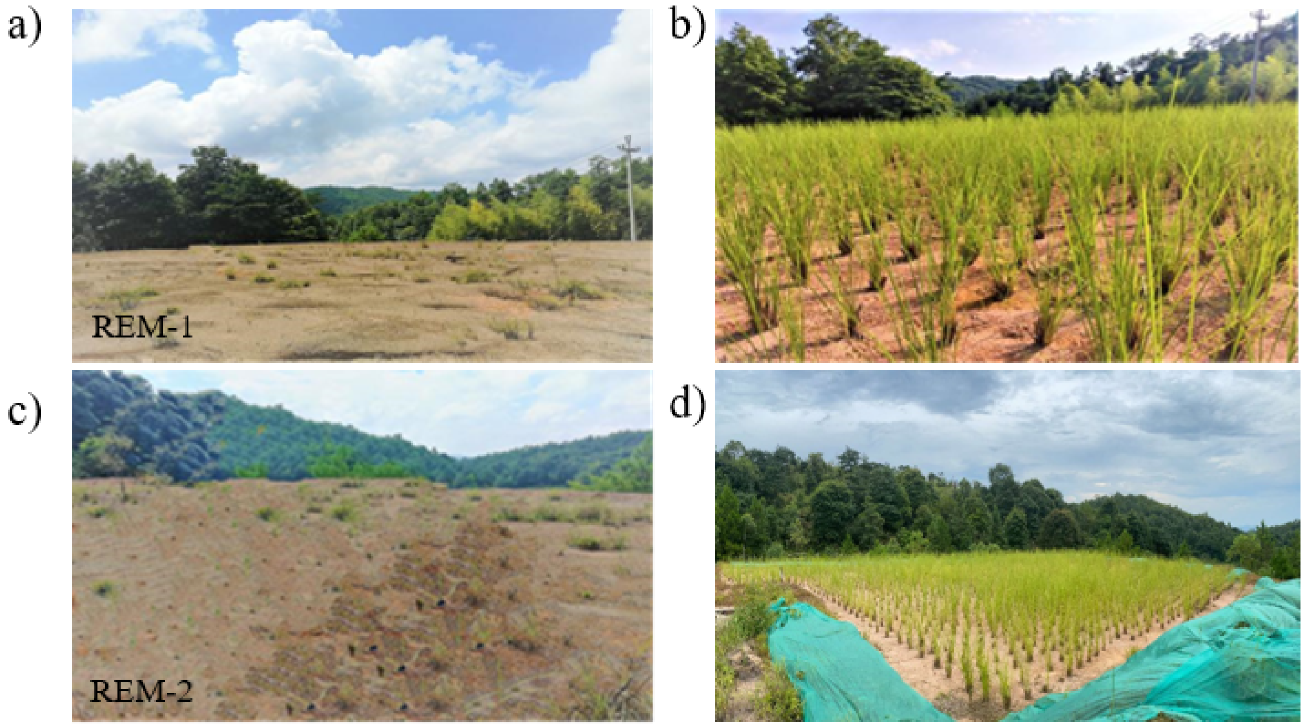

Figure 1

The original appearance and ecological restoration condition of abandoned rare earth mine. a and c were Original mined land of REM-1 and REM-2, respectively; b and d were the land of REM-1 and REM-2 with ecological restoration for 8 months, respectively.

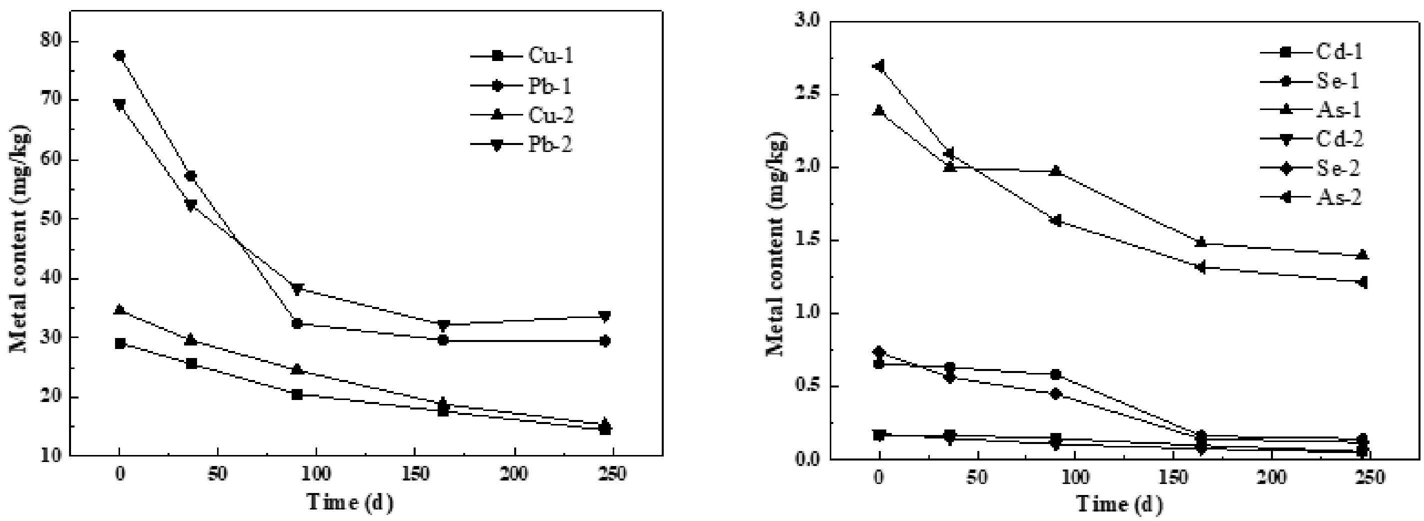

Figure 2

Metal concentrations in sampling site soils of REM-1 and REM-2

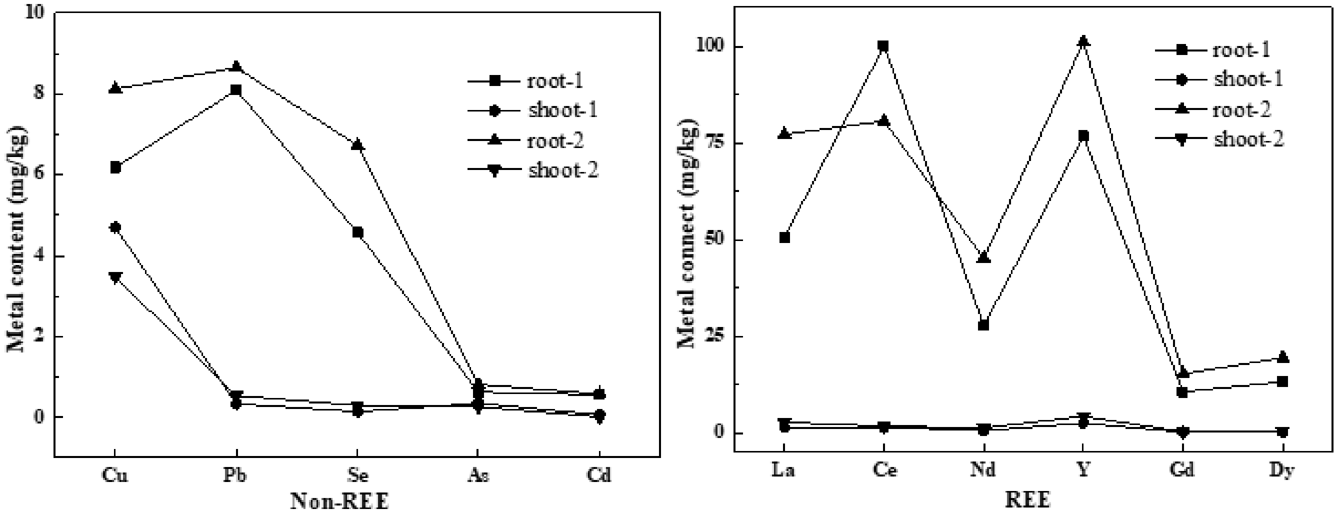

Figure 3

Toxic metals concentrations in sampling site soils of REM-1 and REM-2

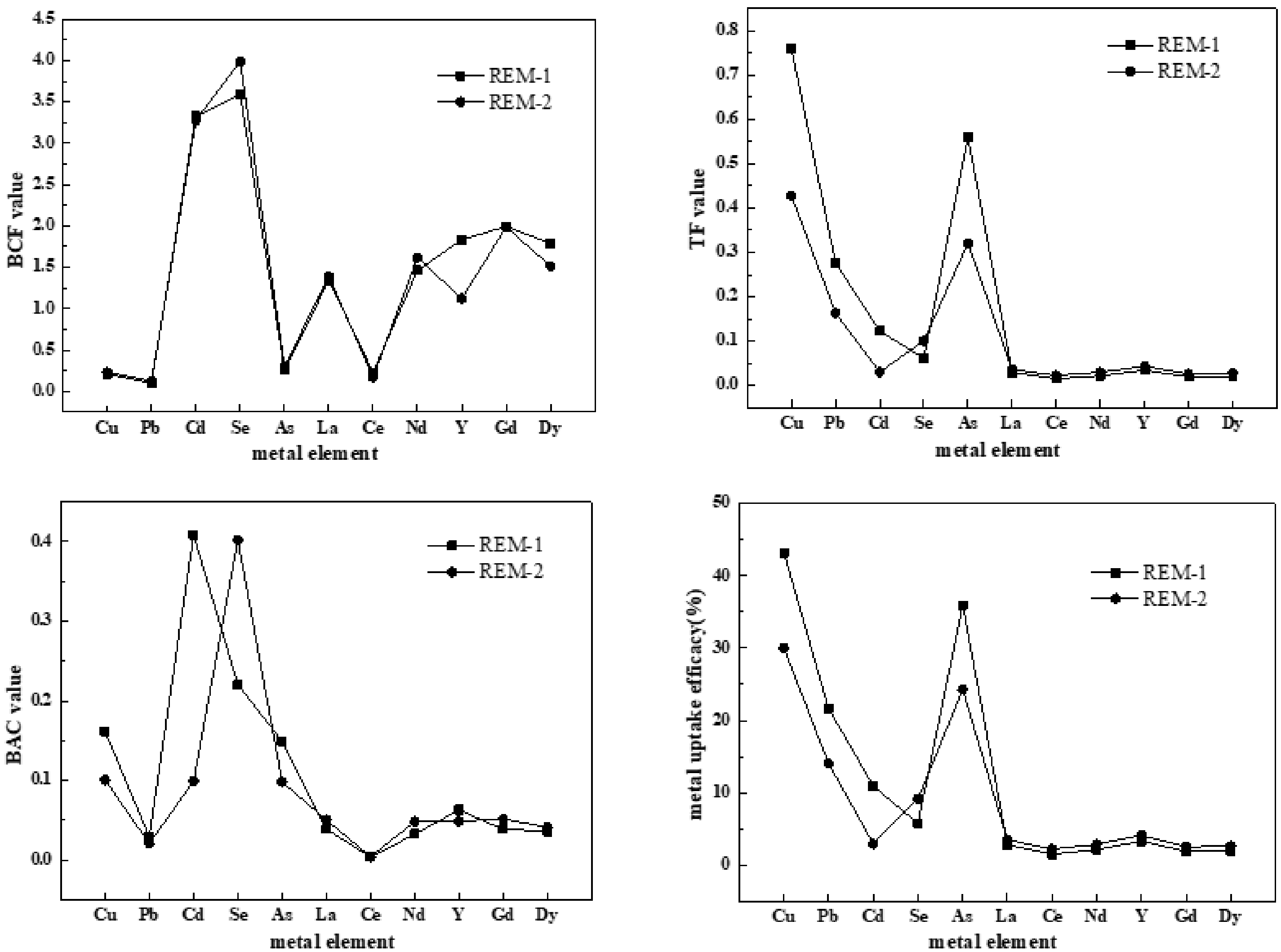

Figure 4

a soil-root BCF for toxic metals of REM-1 and REM-2 b root-shoot TF for toxic metals of REM-1 and REM-2 c soil-shoot BAC for toxic metals of REM-1 and REM-2 d metal uptake efficacy for toxic metals of REM-1 and REM-2

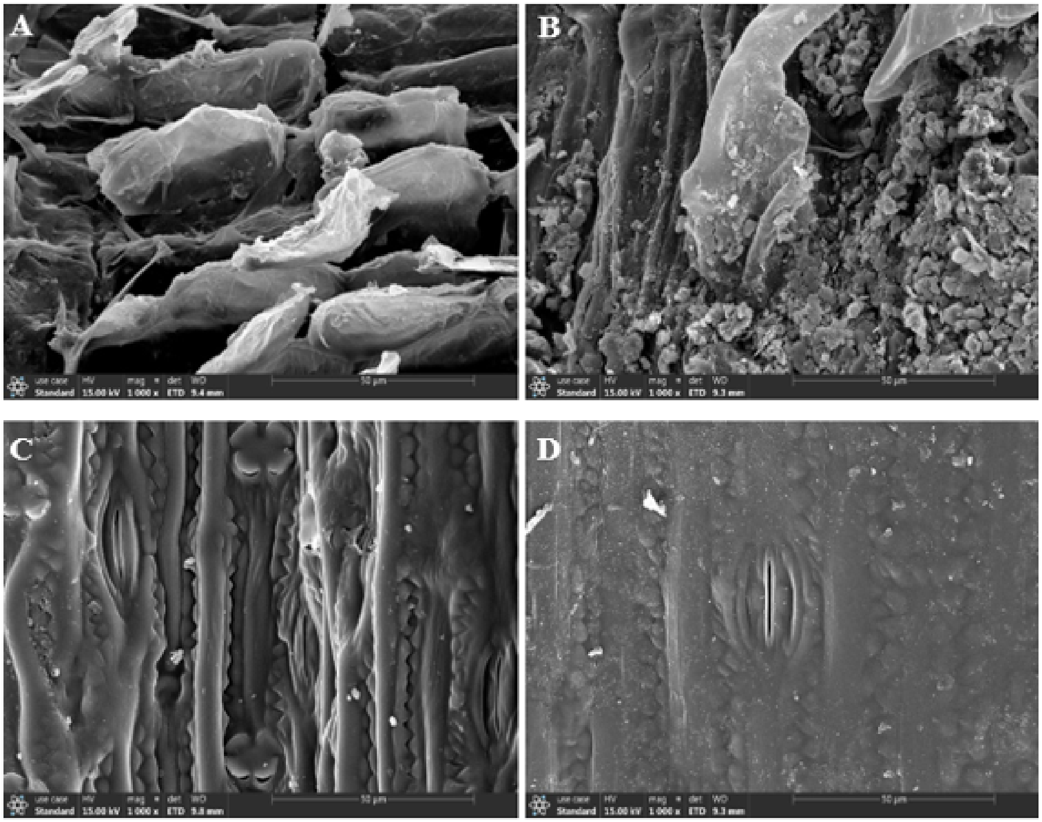

Figure 5

SEM micrographs of vetiver roots at planting (A) and harvest (B); the vetiver shoots at planting (C) and harvest (D)