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Two potential multi-metal hyperaccumulators found in four mining sites in Hunan Province, China

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ABSTRACT

Multi-metal pollution in mines and their surrounding areas causes major environmental and health problems. An investigation on the soil quality in four mining sites revealed serious heavy metal (HM) contamination. The contents of As, Pb, Zn, Cd, Cu, and Sb showed high HM pollution. Compared with the national second soil environmental quality standard, the over-standard rates of As, Pb, Zn, Cd, Cu, and Sb were 97.8%, 80.0%, 82.2%, 68.9%, 88.9%, and 73.3%, respectively. Phytoextraction is an emerging remediation technology for HM-contaminated soil. However, hyperaccumulators that can simultaneously extract multi-metals have been rarely reported. A field survey was conducted in the four mining sites; results showed that *Pteris vittata* L. and *Viola principis* H. de Boiss were two potential multi-metal extractors. Two plant species could thrive on soils severely contaminated with multiple metals. *P. vittata* accumulated 4106 mg As kg^{-1}, 499.5 mg Pb kg^{-1}, and 321.5 mg Sb kg^{-1} in the aboveground parts; by comparison, *V. principis* accumulated 1032 mg As kg^{-1}, 2350 mg Pb kg^{-1}, and 1201 mg Cd kg^{-1} in the aboveground parts. The bioaccumulation factor of *P. vittata* for As, Pb, Zn, and Sb was >1. The bioaccumulation factor of *V. principis* for As, Pb, Zn, and Cd was also >1. Therefore, *P. vittata* is an As hyperaccumulator and a Pb and Sb accumulator. By contrast, *V. principis* is a Cd, Pb, and As hyperaccumulator.

1. Introduction

Multi-metal pollution in mines and their surrounding areas has been considered as one of the most serious environmental problems in China (Qiu et al., 2009). This status is caused primarily by the characteristics of mineral sources in China; for instance, a considerable percentage of ore deposits in China accounts for lean ore, small and medium-sized deposits, or paragenetic minerals (Fu and Zhong, 2008). The mined ore is only a small fraction of the total volume of mined materials. Additional metals occurring naturally in raw ore become inevitable by-products. Zinc (Zn) and lead (Pb) smelters release large amounts of arsenic (As) and cadmium (Cd) into the environment (Adriano, 2001); as a consequence, As–Pb–Zn becomes one of the most common multi-metal contamination types. According to a recent review, lead–zinc and antimony mines are of priority control mine categories; southern provinces are selected as priority control provinces (Li et al., 2014).

The soils in mining and smelting sites are often contaminated by multi-metals as a result of particulate matter deposition, solid waste disposal, and wastewater discharge (Esmaeili et al., 2014; Rey et al., 2013). Heavy metals (HMs) accumulate in the upper layer of soil and remain for long periods because of their non-degradable nature; as such, HMs pose risks to food production and human health (Wang et al., 2015). Therefore, efficient techniques should be developed to remediate such multi-metal-contaminated lands.

Phytoextraction is an emerging remediation technology for HM-contaminated soil (Luo et al., 2014). Hyperaccumulators with a unique capacity of enriching HMs in the aboveground parts are keys to the success of this technology (Reeves and Baker, 2002). However, the lack of appropriate hyperaccumulators is one of the most challenging limitations to the promotion of phytoextraction technology, particularly in multi-metal-contaminated soil.

Over 400 hyperaccumulator species have been found; of these hyperaccumulator species, 320 are nickel (Ni) hyperaccumulators. The hyperaccumulators that can simultaneously extract multi-metals are rare. Soils in mining areas are often contaminated by multi-metals, whereas the presently identified hyperaccumulators only tolerate and accumulate one single toxic element. This contradiction has affected multi-metal extraction efficiency. To solve this problem, researchers investigated multi-metal hyperaccumulators (Abu Bakar et al., 2013; Keller, 2006; Marchiol et al., 2004; Shahid et al., 2012). For instance, *Thlaspi* and *Sedum* species and *Potentilla griffithii* are Zn and Cd co-accumulators (Papoyan et al., 2007; Qiu et al., 2011; Zhang et al., 2011). Mining sites in China with a long history of multi-metal mining were selected to increase the number of potential multi-metal hyperaccumulators in China. We hypothesized that plants have gradually gained the ability to tolerate and accumulate HMs after these plants underwent a long-term acclimation in these mining areas. On the basis of previous studies (Lei et al., 2008; Wan et al., 2014), we

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considered *Pteris vittata* L. and *Viola principis* H. de Boiss as candidates with great hyperaccumulator potential. As, Pb, Cd and Sb were the main HM targets because of their high toxicity, wide distribution, and high possibility of co-existence in the same sites.

Considering the long-term development target of remediating typical multi-metal-contaminated mining sites through phytoextraction technology, we preliminarily investigated soil HM pollution, and the HM contents in the potential multi-metal hyperaccumulators to screen appropriate plant materials for multi-metal phytoextraction.

2. Materials and methods

Soils and plants were collected from four mining sites located in Hunan in south China. Hunan is rich in mineral deposits and is known as "the hometown of nonferrous metals." More than 80 kinds of mineral deposits have been detected. Among these mineral resources, antimony (Sb) has the highest reserves in the world, and lead (Pb), zinc (Zn), and realgar have the highest reserves in China. The four mining sites included the Baoshan polymetallic mining site, Bofang Cu mining site, Shuikoushan polymetallic mining site, and Xikuangshan Sb mining site (Fig. 1).

Baoshan Cu–Mo–Pb–Zn–Ag polymetallic ore deposit located in Chenzhou City is one of the most well-known deposits in Nanling non-ferrous metal belt; this area is characterized by multiple element mineralization and large ore reserve (Bao et al., 2014). The Pb and Zn content in the Baoshan ore is as high as 6.29%. Bofang Cu ore deposit located in Hengyang City contains high Cu, Pb, Zn, Sb, and Ag contents. The Cu, As, and Pb contents in the ore are >5%, >1.5%, and >8.5%, respectively. Shuikoushan Pb–Zn–Cu–Ag–Au polymetallic ore deposit is located in Hengyang City, one of the largest Pb–Zn mining area in China (Hu and Wu, 2005). The reserve of Pb–Zn concentrate is higher than two million tons. Xikuangshan Sb ore deposit, located in Lengshuijiang City, is the largest Sb ore in the world; this ore deposit contains large amounts of Sb, As, Hg, Zn, and Cu (Mo et al., 2013). The reserve of Sb is higher than two million tons. High As and Pb contents are also found in this ore deposit because of the co-existence of As, Pb, and Sb oxides and sulfides. These four mining sites have a long history of mining. According to official records, Baoshan ore deposit has been mined for more than 40 years, and Shuikoushan and Xikuangshan have been mined for more than 110 years. According to unofficial accounts, these sites have a mining history of more than 1000 years.

We investigated 32 areas (Table 1) in these four mining sites; each area covers a size of 3 m × 3 m with *P. vittata* or *V. principis* growing as the main species. Soil samples were collected from the four sampling points on the diagonal part (depth of 0 cm to 15 cm) of each area, mixed together on site once, dried, and sieved; plant roots and stones were excluded. The remaining components were mixed again and transported to our laboratory. Three plants were collected randomly as replicates and analyzed separately.

HMs in soils was determined through HNO_3–H_2O_2 digestion in accordance with the 3050B method of USEPA (1996). Plant samples were dried, ground, and digested with a mixture of HNO_3–$HClO_4$ (Chen et al., 2002). To perform quality control, we simultaneously digested the samples of certified standard reference materials for soils (GSS-1) and plants (GSV-2) from the China National Standard Materials Center with the experimental samples. The As concentrations were determined using an atomic fluorescence spectrometer (Haiguang AFS-2202, Beijing Kechuang Haiguang Instrumental Co., Ltd., Beijing, China). The concentrations of other HMs were determined using an inductively coupled plasma mass spectrometer (ICP-MS ELAN DRC-e, PerkinElmer, the USA).

Over-standard rates were calculated to evaluate the contamination status of the soil in the mining sites. An over standard rate indicates the percentage of samples with HM concentrations higher than that recommended by China's Environmental Quality Standard for Soils (GB15618-1995, Grade II for soil 6.5 ≤ pH < 7.5: As ≤30 mg kg^{-1}; Pb ≤ 300 mg kg^{-1}; Zn ≤ 250 mg kg^{-1}; Cd ≤ 0.60 mg kg^{-1}; Cu ≤ 100 mg kg^{-1}; Cr ≤ 200 mg kg^{-1}) in all of the collected samples.

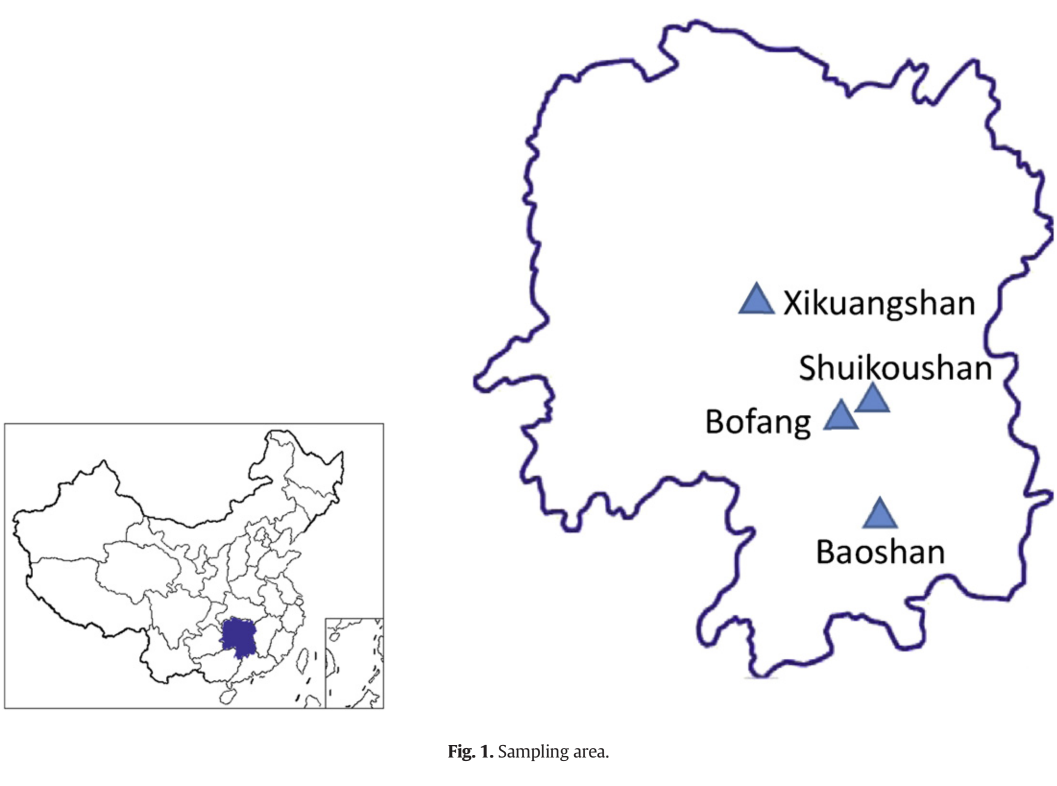

Fig. 1. Sampling area.