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A combined process coupling phytoremediation and *in situ* flushing for removal of arsenic in contaminated soil

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ABSTRACT

Phytoremediation and soil washing are both potentially useful for remediating arsenic (As)-contaminated soils. We evaluated the effectiveness of a combined process coupling phytoremediation and *in situ* soil flushing for removal of As in contaminated soil through a pilot study. The results showed that growing *Pteris vittata* L. (*P.v.*) accompanied by soil flushing of phosphate (*P.v.*/Flushing treatment) could significantly decrease the total As concentration of soil over a 37 day flushing period compared with the single flushing (Flushing treatment). The *P.v.*/Flushing treatment removed 54.04% of soil As from contaminated soil compared to 47.16% in Flushing treatment, suggesting that the growth of *P. vittata* was beneficial for promoting the removal efficiency. We analyzed the As fractionation in soil and As concentration in soil solution to reveal the mechanism behind this combined process. Results showed that comparing with the control treatment, the percent of labile arsenate fraction significantly increased by 17% under *P.v.*/Flushing treatment. As concentration in soil solution remained a high lever during the middle and later periods (51.26–56.22 mg/L), which was significantly higher than the Flushing treatment. Although soil flushing of phosphate for more than a month, *P. vittata* still had good accumulation and transfer capacity of As of the soil. The results of the research revealed that combination of phytoremediation and *in situ* soil flushing is available to remediate As-contaminated soils.

Introduction

Arsenic (As) is a toxic metalloid, which pose a high risk to large human populations. The contamination of As in Bangladesh groundwater was first reported in the mid-1990s (Nickson et al., 1998), and since then, a lot of investigations and studies involving As contamination had been done for the recent decades. The utilization of As-contaminated water for irrigation has resulted in elevated As levels in agricultural soils and therefore agricultural products (Ramirez-Andreotta et al., 2013; Tong et al., 2014). Since the extensive mining activities and irrigation of wastewater with high As, China has become one of the most severely As-contaminated countries (Yu et al., 2015; Zhang et al., in press).

Phytoremediation is a promising technology for the remediation of contaminated soil. An As-hyperaccumulator, *Pteris vittata* L. (*P.v.*) has extraordinary tolerance and accumulation capacity of As (Ma et al., 2001; Chen et al., 2002). A series

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of trials had shown positive results of this emerging technology. *P. vittata* removed 46%–66% total As from soils over 7 harvests in 3.5 years (Lessl et al., 2014), 8% of the total As was removed in 7 months through growing this fern (Liao et al., 2004). However, the remediation efficiency of this technology is relatively low and a long remediation period is required (Shelmerdine et al., 2009; Luu et al., 2014).

Increasing availability of soil As, which can be achieved by applying appropriate amendments, is of significance for promoting phytoremediation efficiency. Studies have found that addition of phosphate fertilizers can significantly increase the biomass and As accumulation of *P. vittata* grown in As-contaminated soils, these phosphate fertilizers were solid-state and thoroughly mixed with contaminated soils (Tu and Ma, 2003; Liao et al., 2008). Some phosphate fertilizers which are more soluble would provide more readily available phosphate to the plant (Fayiga and Ma, 2006; Yan et al., 2012). Based on existing results, we cannot help to think that maybe adding liquid phosphate is more effective for phytoremediation. Actually, the remediation process involves adding liquid solution into the contaminated soils called "*in situ* soil flushing" (Wasay et al., 2001). *In situ* soil flushing could permanently remove the contaminants within a relatively short period of time (Yun et al., 2015). However, this technology is not effective in low permeability or heterogeneous soils (Abumaizar and Smith, 1999). Obviously, plant root growth could loosen the soil and increase soil permeability to a certain degree. Moreover, the root exudate of *P. vittata* altered rhizospheric chemical composition and enhanced As bioavailability in the soil (Mandal et al., 2014; Xu et al., 2014). Hence, we hypothesized that growing *P. vittata* may in turn accelerate the efficiency of *in situ* flushing.

The overall objective of this research was to determine the availability of a combined process coupling phytoremediation and *in situ* flushing for remediation of As-contaminated soils. The specific objectives were to evaluate (1) the impacts of soil flushing (using phosphate) on As accumulation of *P. vittata*, and (2) whether growing *P. vittata* is beneficial for promoting the efficiency of flushing.

1. Materials and methods

1.1. Pilot study

The experiment included three treatments: (1) flushing (*in situ* flushing of 0.8 mol/L KH_2PO_4 without growing *P. vittata*); (2) *P. vittata* (growing *P. vittata* without *in situ* flushing); and (3) *P.v.*/Flushing (growing *P. vittata* combined with *in situ* flushing of 0.8 mol/L KH_2PO_4).

The experiment was conducted in a closed brick-powder flushing tank (1.5 m × 1.5 m × 1.5 m) which contained (from top to bottom) seven layers. The schematic diagram of the remediation mode was shown in Fig. 1. Our early study has shown that the barrier adsorption material (mixture of activated carbon and iron oxide (volume ratio of 1:1), and its initial As concentration was 2.87 mg/kg) had a high adsorption capacity to As. In addition, soil solution collectors were placed in Layer B (at 17 cm) and Layer F (at 62 cm), and there was a wastewater collection port at the bottom of the flushing tank. The three treatments were conducted in three flushing tanks simultaneously.

1.2. Soil preparation and plant transplantation

The contaminated soils were collected at a depth of 0–30 cm from an As-contaminated farmland in Chenzhou, Hunan Province (25°48′N and 113°02′E). After removing stones, plant, animal residues and other impurities, the sample was air-dried, and passed through a 2 mm sieve. The soil properties were as follows: pH 8.38, organic matter content of 31.90 g/kg, total P of 0.6 g/kg, cation exchange capacity of 34.52 cmol/kg, total As concentration of 209.30 mg/kg, and a soil particle size distribution of 60.93% sand, 23.68% silt, and 15.39% clay.

P. vittata seedlings were collected from a local greenhouse (Chenzhou, Hunan) and were selected for the following characteristics: a height of 5–10 cm, 4–5 pairs of pinnae, and equally sized rhizomes. The seedlings were then cultivated in a greenhouse (at 22–25°C, and with a relative humidity of 75%–80%) at the Institute of Agricultural Sciences, Fangshan District, Beijing. After 60 days of cultivation, nine seedlings with identical growth status were transplanted to the *P.v.*/Flushing and *P. vittata* treatments tanks, respectively.

1.3. Sample collection

Prior to the start of the experiment, each tank was watered with 300 L of groundwater (As concentration is 0.014 mg/L). During the experiment, the Flushing and *P.v.*/Flushing tanks was each flushed with 1000 L of KH_2PO_4 solution per day, at a rate of 150 L/hr for 37 days, and soil solutions were collected at 8:00, 14:00, and 18:00 daily. The collected solutions were filtered through a 0.45 μm filter before testing.

Soil samples were collected from Layer B (0–20 cm) on days 7, 12, 17, and 37, and at the end of the experiment, collecting soils from Layer D and F. After cultivation for 37 days, the plants were harvested from the *P.v.*/Flushing and *P. vittata* tanks.

1.4. Chemical analysis

The As concentration of soil and plant was analyzed by the method reported by Yan et al. (2012). The soil samples were air-dried and passed through a 0.85 mm sieve, and the As fractions were extracted from soil samples using the five-step. Soil samples were treated sequentially with 1 mol/L NH_4Cl to separate labile arsenate (L-As), 0.5 mol/L NH_4F to separate aluminum-bound As (Al–As), 0.1 mol/L NaOH to separate iron-bound As (Fe–As), and 0.5 mol/L H_2SO_4 to separate Ca-bound As (Ca–As). Finally, the samples were digested with HNO_3 and H_2O_2 to determine residual As (O–As). The fresh plant samples were rinsed with deionized water, heated at 105°C for 30 min, dried at 60°C to achieve constant weight. 0.5 g of dry tissue samples was weighted, mixed with HNO_3 (10 mL) and $HClO_4$ (2 mL, 70%–72%), and digested over a heating plate until it became a clear liquid.

The digested plant and soil samples were analyzed for total As using AFS (AFS-9130, Titan Instruments, Beijing, China). All