

Phytoextraction of arsenic-contaminated soil with *Pteris vittata* in Henan Province, China: comprehensive evaluation of remediation efficiency correcting for atmospheric depositions

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Abstract Research on the appropriate method for evaluating phytoremediation efficiency is limited. A 2-year field experiment was conducted to investigate phytoremediation efficiency using the hyperaccumulator *Pteris vittata* on an arsenic (As)-contaminated site. The remediation efficiency was evaluated through the removal rate of As in soils and extraction rate of heavy metals in plants. After 2 years of remediation, the concentration of total As in soils decreased from 16.27 mg kg⁻¹ in 2012 to 14.58 mg kg⁻¹ in 2014. The total remediation efficiency of As was 10.39% in terms of the removal rate of heavy metals calculated for soils, whereas the remediation efficiency calculated from As uptake by *P. vittata* was 16.09%. Such a discrepancy aroused further consideration on the potential input of As. A large amount of As was brought in by atmospheric emissions, which possibly biased the calculation of remediation efficiency. In fact, considering also the atmospheric depositions of As, the corrected removal rate of As from soil was 16.57%. Therefore, the results of this work suggest that (i) when evaluating the phytoextraction efficiency, the whole input and output cycle of the element of interest in the targeted ecosystem must be considered, and (ii) *P. vittata* has the potential to be used to remediate As-contaminated soils in Henan Province, China.

Keywords Arsenic · Atmospheric depositions · Heavy metal contamination · *Pteris vittata* · Phytoextraction · Remediation efficiency

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Introduction

Soil heavy metal (HM) pollution is an environmental problem that has received increasing attention worldwide (Khan et al. 2010; Petrikova et al. 1995; Yu et al. 2012). In March 2015, the Chinese Ministry of Environmental Protection and the Ministry of Land and Resources released the first-ever results of a nationwide soil pollution survey, which revealed that one fifth of the arable lands is polluted and contaminated with HMs such as cadmium (Cd), nickel (Ni), and arsenic (As). Arsenic in soil can enter the food chain through drinking water or agricultural products, posing a risk to human health and environmental safety (Amonoo-Neizer et al. 1996; Berg et al. 2007; Phuong et al. 2008).

Given the high toxicity and large distribution of As, the remediation of As-polluted soils is an important research focus (Kim et al. 2005; Redwine 2001; Shipley et al. 2011). The phytoextraction of As-contaminated soils offers a low-cost method for soil remediation, and As may be recycled for value (Kramer 2005). A fern *Pteris vittata* L. was discovered as the first As hyperaccumulator (Ma et al. 2001). *P. vittata* shows extraordinary tolerance and accumulation to As, and it grows rapidly with large biomass, wide distribution, and easy adaptation to different environmental conditions; therefore, it has been widely applied to the phytoextraction of As from contaminated soils (Kertulis-Tartar et al. 2006; Mandal et al. 2012; Salido et al. 2003; Shelmerdine et al. 2009; Wan et al. 2016). *P. vittata* demonstrates an annual As removal efficiency ranging from 12 to 19.2%, which depends on the adopted strengthening measures (Lessl et al. 2014; Lessl and Ma 2013; Niazi et al. 2011, 2012).

The soil pollution prevention action plan of China was released in May 2015. The plan indicates that 666,666.7 ha of contaminated cultivated lands should be remediated until 2020, which means an increasing number of remediation projects in the near future. Under the present Chinese national

conditions, phytoremediation might be appropriate in terms of costs and environmental friendliness. With the increasing demand for the removal of HM from contaminated soils, the introduction of guidelines and regulations on soil remediation (especially in the evaluation of the remediation efficiency) becomes very important (Cui and Yuan 2012). At present, different methods are used to calculate remediation efficiency, and they mainly include two aspects: (i) evaluation of the reduction of As in soil, which may bring to different results when collecting soils at different depths, and (ii) assessment of the accumulation of As in plants, which may differ among plant samples collected at different times or locations. The two methods have their own advantages and disadvantages. When confronting various results using these two methods, the different input and output sources of As should be taken into account.

This paper introduces a case study of phytoextraction of As-contaminated soil in Jiyuan, Henan Province, China. The As removal efficiency was evaluated both from the aspect of the As decrease in soil and the extracted As in *P. vittata*. In addition, the input of As from atmospheric depositions when calculating the remediation efficiency was also considered.

Materials and methods

Site description

The experimental site is in Kejing Town, located in an alluvial plain, south of Taihang Mountain, Jiyuan, northwest of Henan Province (35° 07' 49.5" N, 112° 32' 50.7" E). Jiyuan has a temperate continental monsoon climate featuring four distinct seasons. The annual temperature is 14.6 °C and annual rainfall is 860 mm. The annual sunshine time is 1727.6 h.

Metallurgy is one of the main industries in Jiyuan. The lead (Pb) smelter industry started in the 1950s, with an annual Pb output reaching 8 million t in 1980. Henan Yuguang Au & Pb Co., Ltd., located in the suburb of Jiyuan City, is the biggest Pb smelter company in China. According to a survey conducted by Henan Environment Monitoring Stations, the soil around Yuguang Au & Pb Co. has been seriously contaminated, with the main contaminants being As, Pb, and Cd. The whole area of the contaminated site is approximately 10,000 m², located in the southwest of Yuguang Au & Pb Co. The area used for the remediation project is 100 m². In March 2013, *P. vittata* was transplanted at a height of 15–20 cm to the field at a density of 30 cm × 30 cm. The aboveground parts of *P. vittata* were harvested at the end of October 2013 and 2014. The roots and rhizoids were kept for growth in the next year. Regular irrigation was supplied according to the climate conditions. Fertilizers (150 kg N ha⁻¹, 90 kg K₂O ha⁻¹, and 350 kg P₂O₅ ha⁻¹) were applied in June every year. The employed fertilizers met the agriculture industry standard

for organic fertilizer NY525-2012 (2012), with As concentration lower than 15 mg kg⁻¹. The harvested *P. vittata* biomass was air-dried (water content <8%) and then incinerated. For this purpose, plant biomass was crushed to about 5 mm and then pelleted into granules of 4–5 cm long and 1 cm in diameter. After this preparation, biomass was fed from the loader (feeding port) at a constant rate of 75 kg h⁻¹ for 1 h. The combustion temperature was monitored and maintained at 850 °C ± 10 °C using a thermocouple. The flue gas was monitored, which met the national emission standard. The incinerated ash was disposed according to measures for the treatment of hazardous waste.

Chemical analyses

Soil sampling and analysis

Before phytoextraction, 11 topsoil (0–20-cm depth) samples were collected for the measurement of soil properties, including pH; organic matter; and total and available nitrogen (N), phosphorus (P), and potassium (K). A total of 72 topsoil (0–20-cm depth) samples were collected to investigate the pollution status of this site, including the concentrations of As, Zn, Cu, Ni, Pb, and Cr. To monitor the change in soil HM concentrations, the locations of these soil sampling sites were recorded. When *P. vittata* was harvested in October 2013 and 2014, soil samples were collected again at the same sampling sites. In total, 216 soil samples were collected. The detailed methods for the measurement of soil properties and HM concentrations are described in the following.

Plant material and stones were removed, and the remaining soil was oven-dried at 40 °C and sieved to 2 mm to measure soil pH; organic matter; and total N, P, and K. To measure soil HM concentrations, soil was sieved at 0.149 mm according to GB/T 22015.2-2008 (2008). Soil pH was determined in a 1:2.5 soil/water mixture (Wei and Chen 2006), and the total organic matter was calculated using the Walkley–Black method (Nelson and Sommers 1982). Total P and N in soil were determined using the titrimetric and gravimetric method with ascorbic acid (John 1970) and Kjeldahl method (Anantakrishnan and Srinivasa Pai 1952), respectively. To determine total K concentration, soil samples were digested using HNO₃-HClO₄, followed by HF, and K was detected using atomic absorption spectrophotometry according to the Chinese National Standard Method for the determination of total potassium in soil (GB 9836-88). HMs in soils were determined through HNO₃-H₂O₂ digestion in accordance with the 3050B method of USEPA (1996).

Plant sampling and analysis

Ten areas of 2 m² were randomly selected to measure the yield of *P. vittata*. The aboveground parts of *P. vittata* were

collected in 2013 and 2014. Ten plant samples (each was a mix of five individual *P. vittata* plants as a composite sample) were collected. Aboveground parts were cut at a height of 10 cm, rinsed with tap water and deionized water, dried at 60 °C until constant weight, and sieved at 2 mm for the analysis of HMs. Plant samples were digested with a mixture of HNO₃-HClO₄ according to Chen et al. (2002). The As concentration was determined using an atomic fluorescence spectrometer (Haiguang AFS-2202, Beijing Kechuang Haiguang Instrumental Co., Ltd., Beijing, China). The concentrations of Zn, Cu, Ni, Pb, and Cr were determined by inductively coupled plasma mass spectrometry (ELAN DRC-e, PerkinElmer, USA).

Atmospheric depositions sampling and analysis

The remediation site was in the downwind of Yuguang Au & Pb Co. Therefore, atmospheric depositions were monitored every month to evaluate the accumulation of As in the remediation site caused by atmospheric emissions.

The whole remediation area was separated into three sub-areas. Three dust tanks were set at the center of these three sub-areas for the sampling of atmospheric depositions. The tank was made of polymethylmethacrylate with the internal diameter being 15 cm and height being 30 cm. The sampling locations were established under the following rules: (1) no high buildings or trees nearby and avoiding local pollution sources, (2) located 5–12 m above the floor and far from the platform to avoid platform dust, (3) the dust collecting cylinder bracket should be stable and firm to avoid being blown down by the wind, and (4) the dust tank must not be easy to damage and must be easy to replace. In addition, a control dust tank was set in a rural area about 50 km from the remediation site, without pollution sources nearby. All these four dust tanks were placed on top of existing buildings at 5 m from the floor.

Before use, tanks were cleaned, immersed in 10% (v/v) HCl for 24 h, and rinsed with deionized water in the laboratory. The clean tanks were brought to the sampling points. Subsequently, 2% HNO₃ was added in the tank. When sampling, deionized water was added to a depth of 5–10 cm to avoid secondary dust emission. Samples were collected 12 times from May 2012 to April 2013. At the end of each month, the dust tanks were retrieved. First, debris was removed and the remaining dust and solution were completely transferred to a beaker. The beaker was then transported to the laboratory and freeze-dried under vacuum at -50 °C until constant weight. The weight difference between the mass of beaker plus dust and the mass of the sole beaker was considered as the mass of the collected dust (M_d). The dust was digested with nitric acid and hydrochloric acid and then measured for the As concentration. The difference in As concentration between samples collected at the contaminated area and that at the control site was considered the concentration of As in dust (C_d).

Data quality control

For quality control, certified standard reference materials for soils (GBW07401) and plants (GBW07603) from the China National Standard Materials Center were digested along with experimental samples. The results for reference materials were in the allowable error ranges.

Data processing

Data were analyzed with SPSS statistic program package (Release 11.0, SPSS Inc., now IBM SPSS, Armonk, NY, USA). One-way ANOVA was performed to test the significance of treatment effects, and multiple comparisons were analyzed using the least significant difference method. The ArcGIS software (kriging interpolation method) was used to obtain a map of As distribution (Yang et al. 2008).

The bioconcentration factors of As in *P. vittata* were calculated as follows (Fayiga et al. 2004):

$$BF = \frac{C_{\text{plant}}}{C_{\text{soil}}} \quad (1)$$

where BF is the bioconcentration factor, C_{plant} is the above-ground concentration of As in *P. vittata*, and C_{soil} is the concentration of As in soil.

The removal rate of As from soil was calculated as follows:

$$Rr = \frac{C_{\text{sb}} - C_{\text{sa}}}{C_{\text{sb}}} \quad (2)$$

where Rr is the removal rate of As from soil, C_{sb} is the As concentration in soil before remediation (mg kg^{-1}), and C_{sa} is the As concentration in soil after remediation (mg kg^{-1}).

Upon considering atmospheric deposition, the calculation formula of the As removal rate (Rr_n) was corrected as follows:

$$Rr_n = \frac{C_{\text{sb}} + C_n - C_{\text{sa}}}{C_{\text{sb}} + C_n} \quad (3)$$

where Rr_n is the As removal rate considering the impact of atmospheric depositions, C_{sb} is the As concentration in soil before remediation (mg kg^{-1}), C_n is the annual increase in soil As concentration caused by atmospheric depositions, and C_{sa} is the As concentration in soil after remediation (mg kg^{-1}). C_n was calculated as follows:

$$C_n = \frac{C_d \times M_d \times D_s \times DE_s}{A_t} \quad (4)$$

where C_d is the As concentration in dust, M_d is the mass of dust in the tank, D_s is the density of the soil, DE_s is the depth of soil (20 cm), and A_t is the area of the tank.

The phytoextraction ratio was calculated as follows:

$$Pr = \frac{C_p \times M_p}{C_{sb} \times M_r} \tag{5}$$

where Pr is the phytoextraction ratio, C_p is the As concentration in the aboveground parts of *P. vittata* after phytoextraction, M_p is the aboveground biomass of *P. vittata* per unit area, C_{sb} is the As concentration in soil before remediation (mg kg^{-1}), and M_r is the soil mass per unit area that plant roots can reach. In practice, M_r was calculated for the 0–20-cm soil layer.

Results

Soil pollution status before remediation

Before soil remediation, the pH ranged from 6.83 to 7.64. The organic matter ranged from 15.06 to 18.92 g kg^{-1} . The total N, P, and K ranged from 0.73 to 0.93 g kg^{-1} , from 0.47 to 0.56 g kg^{-1} , and from 16.5 to 19.7 g kg^{-1} , respectively. According to the fertility evaluation guidelines (Zheng et al. 2004), soil in the remediation site was of medium fertility.

The concentrations of As, Zn, Cu, Ni, Pb, and Cr in soil before phytoextraction are reported in Table 1. Compared with the values recommended by China’s Environmental Quality Standard for Soils (GB15618-1995, grade II for soil $6.5 \leq \text{pH} < 7.5$: As $\leq 30 \text{ mg kg}^{-1}$; Zn $\leq 250 \text{ mg kg}^{-1}$; Cu $\leq 100 \text{ mg kg}^{-1}$; Ni $\leq 50 \text{ mg kg}^{-1}$; Pb $\leq 300 \text{ mg kg}^{-1}$; Cr $\leq 200 \text{ mg kg}^{-1}$), soil could be considered not contaminated. However, compared with the HM background values in soils of Henan Province (Wei et al. 1990), the concentrations of As, Zn, Cu, and Pb were significantly higher than the background value ($p < 0.01$), suggesting an exogenous input of HMs. Compared with the screening levels of As recommended by US Environmental Protection Agency (EPA), the As

concentrations were much higher than the value allowed for residential areas (0.39 mg kg^{-1}) or industrial lands (1.6 mg kg^{-1}).

Even though the As concentration in the soil was lower than the value recommended by China’s Environmental Quality Standard for Soils, the As concentrations in 7% of the wheat and maize samples exceeded the national standard for food (0.5 mg kg^{-1}). This value indicated a potential threat of As on human health.

A high heterogeneity in As distribution was observed in soil. High concentrations of As were mainly distributed in the eastern part of the remediation site (Fig. 1a), being as high as 24 mg kg^{-1} .

As concentrations in soil after remediation

After 1 year from planting the As hyperaccumulator *P. vittata*, the As concentration in soil decreased to 14.99 mg kg^{-1} , within a range of $13.34\text{--}17.45 \text{ mg kg}^{-1}$. After 2 years of remediation, the As concentration in soil decreased to 14.58 mg kg^{-1} , within a range of $12.47\text{--}17.32 \text{ mg kg}^{-1}$. The distribution of As also indicated an apparent decrease in the As concentration (Fig. 1b), especially in the eastern part of the site.

As accumulation in *P. vittata*

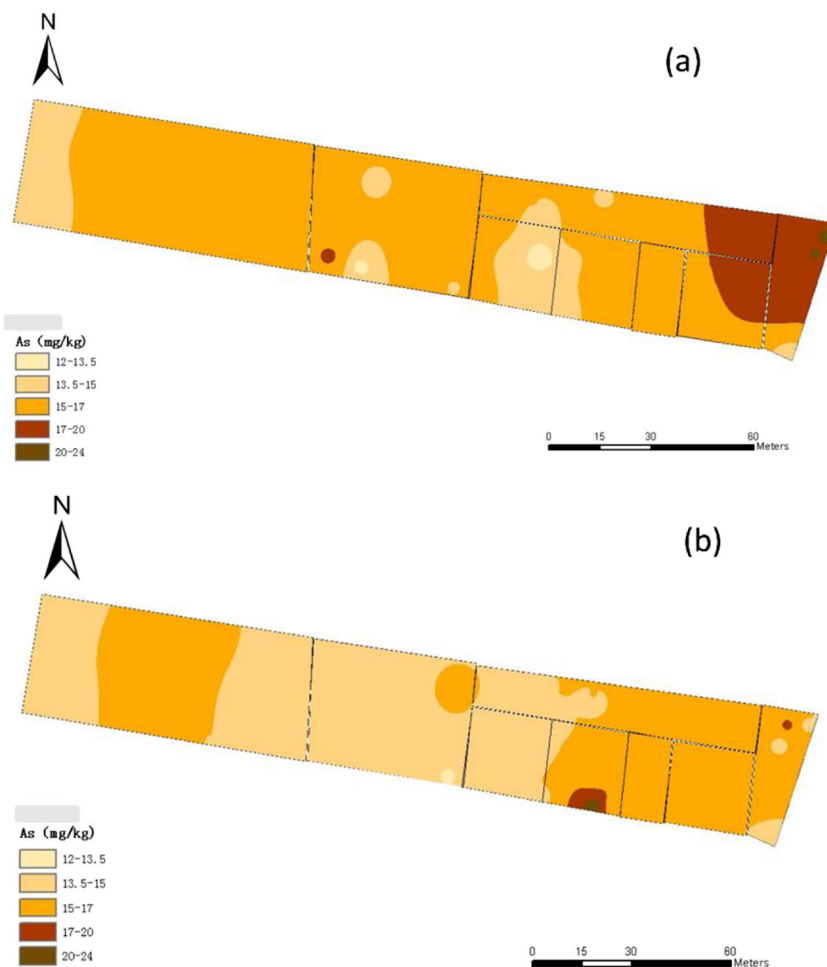
In 2013, the aboveground biomass of *P. vittata* was, on average, 7000 kg ha^{-1} (dry weight), and the As concentration was 419.6 mg kg^{-1} . In 2014, the aboveground biomass of *P. vittata* was 6909 kg ha^{-1} (dry weight), and the As concentration was 409.8 mg kg^{-1} . Therefore, no significant difference in biomass and As concentration of *P. vittata* was found between 2013 and 2014 (Table 2). The bioconcentration factor of *P. vittata* reached 28, confirming the high accumulating ability of *P. vittata*. Results indicated that *P. vittata* grew well on this remediation site, and it could continuously take up As from soil.

Table 1 Heavy metal concentration in soil before phytoextraction in 2012

Items	As	HM concentration (mg kg^{-1})				
		Zn	Cu	Ni	Pb	Cr
No. of samples	72	72	72	72	72	72
Average \pm std	16.3 ± 1.3	110.5 ± 1.5	28.9 ± 4.2	25.4 ± 6.2	259.2 ± 33.5	39.0 ± 7.5
Medium	15.96	108.9	29.26	23.64	250.5	38.63
Max.	21.22	158.05	38.43	41.41	291	72.41
Min.	12.81	71.03	5.4	10.48	218.7	7.87
Reference values ^a	30	250	100	50	300	200
Background values in Henan Province	11.4	60.1	19.7	29.9	19.6	63.8

^a China’s Environmental Quality Standard for Soils (GB15618-1995, grade II for soil $6.5 \leq \text{pH} \leq 7.5$)

Fig. 1 Distribution of As before (a) and after (b) remediation



Evaluation of remediation efficiency in terms of the removal rate of As from soil

After 1 year of remediation, the removal rate of As (R_r) in soil was 7.87%, which decreased to 2.74% in the second year. In total, the phytoremediation ratio of As after 2 years of remediation was 10.39% (Table 3). Although a decrease was observed, analysis of variance indicated no significant differences in soil As concentration among different years ($p > 0.05$).

Upon comparing the decrease in soil As concentration and the extracted As by *P. vittata*, it is evident that As was taken up from soil, but As in soil did not show an apparent decrease. By

Table 2 Average biomass and As concentration in the aboveground parts of *P. vittata*

Year	As (mg kg^{-1})	Yield per unit (kg ha^{-1})
2013	419.6	7000
2014	409.8	6909
Average	414.7	6954

multiplying biomass and the As concentration, it can be calculated that 5768 g of As was removed from 1 ha of soil after 2 years of remediation. The calculated phytoextraction ratio (Pr) was then 16.09%. The gap between 10.39% (calculated for the soil) and 16.09% (calculated for the plant) was further investigated.

Analyses on atmospheric depositions showed that the dust collected at the remediation site contained a considerable amount of As compared with the dust collected at the control site (Fig. 2). In the control site, the deposited As was lower than 1 kg km^{-2} in 30 days, whereas the deposited As in the remediation site reached up to 20 kg km^{-2} in 30 days. These results indicated significant As atmospheric depositions in the remediation area, which may have led to the insignificant variation in soil As concentration after the 2 years' remediation, despite the continuous extraction of As from soil by *P. vittata*.

Therefore, by considering the effect of atmospheric depositions (i.e., after subtracting the increase in soil As caused by atmospheric deposition), the corrected removal rate of As in soil (R_{r_n}) reached 16.57% after 2 years of remediation (Table 4). This finding was basically in accordance with the

Table 3 Removal rate of As from soil from 2012 to 2014

Year	Average As concentration in soil (mg kg ⁻¹)	Range (min-max) (mg kg ⁻¹)	Removal rate (Rr, %)	Total phytoextraction ratio (Pr, %)
2012	16.27	12.81–21.22		
2013	14.99	13.34–17.45	7.87	
2014	14.58	12.47–17.32	2.74	10.39

efficiency calculated by determining the accumulated As in *P. vittata*.

Discussion

Potential of *P. vittata* in removing As from soil

Nearly 30 years have passed since the first proposal of removing HMs from contaminated soil using plants (Chaney 1989). During this period, a considerable amount of studies has been conducted toward the screening of hyperaccumulators and the interaction between trace elements and plants; however, reports on the field success of phytoextraction are still limited (Robinson et al. 2015).

Through the current study and several previous publications, *P. vittata* showed potential to remove As from soils with As concentrations ranging from 16.3 to 1184 mg kg⁻¹ (Liao et al. 2004; Wan et al. 2016; Xie et al. 2010). In these studies, the remediation efficiency showed a large variability. One reason for the different remediation efficiencies is the variation in As accumulating ability. The observed As concentration in *P. vittata* ranged from 409 to 1700 mg kg⁻¹. In addition, the remediation efficiency depended not only on the accumulating ability of *P. vittata* but also on the environmental

conditions, such as soil As concentration, As speciation, and soil properties.

An earlier study reported an As removal amount of 15.5 kg ha⁻¹ after 2 years of remediation (Xie et al. 2010); this value was higher than the value obtained in the current study (about 5.8 kg ha⁻¹). The primary reason for this difference was that the original As concentration before remediation at that site was much higher (1184 mg kg⁻¹). It has been found that in soils with As concentration ranging from 26 to 126 mg kg⁻¹, *P. vittata* removed 46–66% total As from soils over seven harvests in 3.5 years (Lessl et al. 2014; Lessl and Ma 2013), which was apparently higher than the current study. However, this finding may mainly result from the higher soil As concentration, increased harvest frequency, and addition of extra *P.* Compared with another phytoextraction experiment on the As-contaminated site (soil As concentration before remediation being 58 mg kg⁻¹), which showed an extraction rate of 7.84% (Liao et al. 2004), the phytoextraction rate in the current study apparently increased. This increase was primarily due to the improved fertilization technology that increased the biomass of *P. vittata* in the field (from 1500 to 5000 kg ha⁻¹).

In addition, the current study indicated the importance of pollution source control during soil remediation. Despite the evidence that *P. vittata* continuously took up As from soil, As

Fig. 2 Atmospheric depositions of As in the control and remediation sites

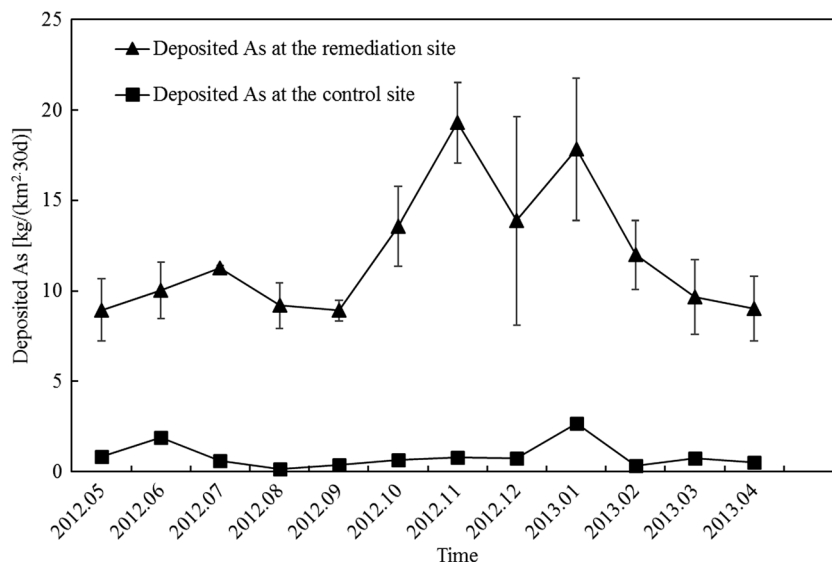


Table 4 Corrected removal rate of As from soil from 2012 to 2014, taking into account atmospheric depositions

Year	Average As concentration in soil (mg kg ⁻¹)	Increase in As caused by atmospheric depositions (mg kg ⁻¹)	Annual phytoextraction ratio (Pr, %)	Total phytoextraction ratio (Pr, %)
2012	16.27			
2013	14.99	0.60	11.24	
2014	14.58	0.60	6.41	16.57

in soil displayed a negligible decrease. This discrepancy was primarily due to the lack of pollution control, particularly atmospheric depositions from a nearby Pb smelter. The pollution source must be continuously monitored for a successful remediation project.

Evaluation of phytoextraction efficiency

With the development of phytoextraction technology, an appropriate method for evaluating the efficiency of phytoextraction becomes increasingly important. The use of the removal rate of HMs from soil is the most common method. However, some researchers have suggested that using the reduction in soil concentration as an indicator may not be sufficient because of large differences in soil HM concentration given that the plant roots can only reach the surface soil (Niazi et al. 2012). Therefore, both the decrease in soil HM concentration and the increase in hyperaccumulator HM concentration must be considered as essential indexes. However, in the current study, the amount of As extracted by *P. vittata* and the decreased amount of As in soil did not match. Therefore, sometimes, these two indexes are not sufficient for a correct remediation assessment. In fact, the complete input and output cycle of HMs in an ecosystem should be considered. The input of HMs into soil may include atmospheric depositions, irrigation and fertilization, canopy leaching, and hyperaccumulator litters (Barcelo and Poschenrieder 2011). The output of HMs from soil includes hyperaccumulator extraction, surface runoff, soil infiltration, and plant surface evaporation. In the remediation site described in the current study, atmospheric depositions were considered one of the most important inputs. Only after considering this input, the phytoextraction efficiencies calculated from the hyperaccumulator removal and soil decrease matched.

A recent study proposed that the ultimate goal of any remediation process should be to remove the contaminant(s) from the polluted site and restore the continued capacity of a soil to perform or function according to its potential (i.e., its health) (Hernandez-Allica et al. 2006). Therefore, further research on the appropriate evaluation method for soil remediation efficiency is necessary.

Conclusion

Through a 2-year phytoextraction project, As hyperaccumulator *P. vittata* was found to exhibit huge biomass and high As accumulating ability, with the aboveground biomass and As concentration being 7000 kg ha⁻¹ (dry weight) and 419 mg kg⁻¹, respectively. The remediation efficiency was calculated according to the decrease in soil As concentration and the accumulation of As in *P. vittata*. According to the decrease in soil As concentration, after 2 years of remediation, As in soil decreased from 16.27 to 14.58 mg kg⁻¹, with a calculated remediation efficiency of 10.39%. However, by considering atmospheric depositions, the corrected remediation efficiency was 16.57%, which was basically in accordance with the remediation efficiency calculated by determining the accumulation of As in *P. vittata* (16.09%). Therefore, the As input from atmospheric depositions must be considered when evaluating the efficiency of a remediation process.

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