



# Extremely Elevated Total Mercury and Methylmercury in Forage Plants in a Large-Scale Abandoned Hg Mining Site: A Potential Risk of Exposure to Grazing Animals

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Received: 21 December 2020 / Accepted: 21 February 2021 / Published online: 19 March 2021  
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## Abstract

Ninety-five wild forage plants (belonging to 22 species of 18 families) and their corresponding rhizosphere soil samples were collected from wastelands of a large-scale abandoned Hg mining region for total Hg (THg) and methylmercury (MeHg) analysis. The forage plant communities on the wastelands were dominated by the *Asteraceae*, *Crassulaceae*, and *Polygonaceae* families. The THg and MeHg concentrations in the forage plants varied widely and were in the range of 0.10 to 13 mg/kg and 0.19 to 23 µg/kg, respectively. Shoots of *Aster ageratoides* showed the highest average THg concentration of  $12 \pm 1.1$  mg/kg, while those of *Aster subulatus* had the highest average MeHg concentrations of  $7.4 \pm 6.1$  µg/kg. Both the THg and MeHg concentrations in the aboveground plant parts exhibited positive correlations with the THg ( $r=0.70$ ,  $P<0.01$ ) and MeHg ( $r=0.68$ ,  $P<0.01$ ) concentrations in the roots; however, these were not correlated with the THg and MeHg concentrations in their rhizosphere soils. The species *A. ageratoides*, *A. subulatus*, and *S. brachyotus* showed strong accumulation of Hg and are of concern for herbivorous/omnivorous wildlife and feeding livestock. Taking the provisional tolerable weekly intake (PTWI) values for IHg recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA in Summary and conclusions of the seventy-second meeting of the joint FAO/WHO expert committee on food additives Rome, Italy, 2010) for human dietary exposure of 4 ng/g into account, the daily intake of IHg by a 65 kg animal grazing on 1.0 kg of forage (dry weight) would be between 190 and 13,200 µg, three to five orders of magnitude higher than the permitted limit, suggesting a potential risk of exposure.

Mercury (Hg) is a global pollutant. Even at low concentrations, Hg exposure can have neurological effects and increase the risk of cardiovascular disease in humans (Driscoll et al. 2013; Peng et al. 2015). Mining and retorting of cinnabar ores are major sources of metal Hg as well as one of the major sources of anthropogenic Hg in the environment (Xu

et al. 2019a, b). Severe Hg pollution caused by abandoned mine-waste calcines (ignited residues) continues to threaten areas of historic Hg mining. These calcines are enriched with water-soluble secondary Hg compounds, such as meta cinnabar, polymorphic sulfide Hg, sulphate Hg, chloride Hg, etc. The large amounts of water-soluble Hg in effluent discharges from mine-waste calcines can readily be transformed into the more toxic methylmercury (MeHg) under suboxic conditions (Qiu et al. 2005; Lin et al. 2010). Because MeHg is considered the most harmful form of Hg due to its high lipophilicity (Agency for Toxic Substances and Disease Registry 2013), the transformation, bioaccumulation, and biomagnification of MeHg is of the greatest concern.

China has rich cinnabar deposits, and it ranks third in the world for its total reserves. Of these mines, the Wanshan Hg mine, once known as the “Mercury Capital,” was the largest elemental Hg production centre in China. Exploitation of this area in China dates back to the Qin Dynasty (220 B.C.) and has resulted in severe environmental Hg contamination and generated significant quantities of wastelands. Total

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Hg (THg) concentrations as high as 4400 mg/kg have been found in mine-waste calcines of the Wanshan Hg mine, with concentrations up to 790 mg/kg Hg in soils from paddies, and 10,000 ng/L Hg in surface water (Horvat et al. 2003; Qiu et al. 2005). The highest concentrations of Hg in the soils collected from the abandoned Hg mining region were approximately two to three orders of magnitude higher than the “probable effect concentration” of 1.06 mg/kg Hg, above which harmful effects on organisms are likely to be observed (MacDonald et al. 2000; Conko et al. 2013), posing public concerns.

Many studies have shown that mine-waste calcines sites are favourable for Hg methylation in Hg mining areas. Levels as high as 3100 µg/kg MeHg have been reported in calcines (Gray et al. 2006). Due to the extremely high levels of Hg, particularly MeHg, mine-waste calcines have become a major source of Hg in the environment surrounding mines, causing elevated concentrations of both inorganic Hg (IHg) and MeHg in soils, water, and biota (Gibb et al. 2011; Xu et al. 2017; Qian et al. 2018; Li et al. 2020). Numerous investigations have revealed elevated concentrations of THg in wild plants; for example, concentrations in goosefoot (*Chenopodium glaucum*) and ferns (*Pteris vittata* L.) from the Wanshan Hg mining region reached 100 mg/kg (Wang et al. 2011; Qian 2020), which is three to four orders of magnitude higher than the allowable limit of 0.05 mg/kg in edible plants set by the Ministry of Health of China (2017). However, few studies have focused on MeHg concentrations in plants, and most of these studies have focused on rice, in which high levels of MeHg can accumulate in the grain (Li et al. 2017). Currently, data for MeHg in wild plants from Hg-contaminated sites is considerably lacking.

Plants provide the basic food energy for animals occupying the areas of former Hg mining. The consumption of the extremely Hg-contaminated forage plants may cause Hg accumulation and biomagnification in primary consumers and introduce Hg into terrestrial food chains. Mercury, as a long-term hazard in vegetated wastes, may have critical impacts on wildlife dependent on the region (Madejón et al. 2012; Basri et al. 2020). Our recent investigations have shown that MeHg has accumulated in herbivorous wildlife in the Wanshan Hg mining region to levels that could cause health concerns (Abeyasinghe et al. 2017; Xu et al. 2019a, b). To better understand the potential risks of THg and MeHg exposure in herbivorous wildlife as well as livestock, the characterization of THg and MeHg in wild plants (forages) is an urgent necessity. It is important to know if forage plants in contaminated regions are of concern for herbivores.

In the present study, dominant wild forage plants growing on wastelands in the Wanshan Hg mining district, Southwest China were investigated. The objectives were to (1) obtain basic information on the forage plants present and their THg and MeHg levels, (2) elucidate the transfer efficiency

of THg and MeHg from the soil to the forage species and factors influencing this, and (3) clarify which species are of the greatest interest to herbivorous animals and assess the potential risk of exposure.

## Materials and Methods

### Study Area

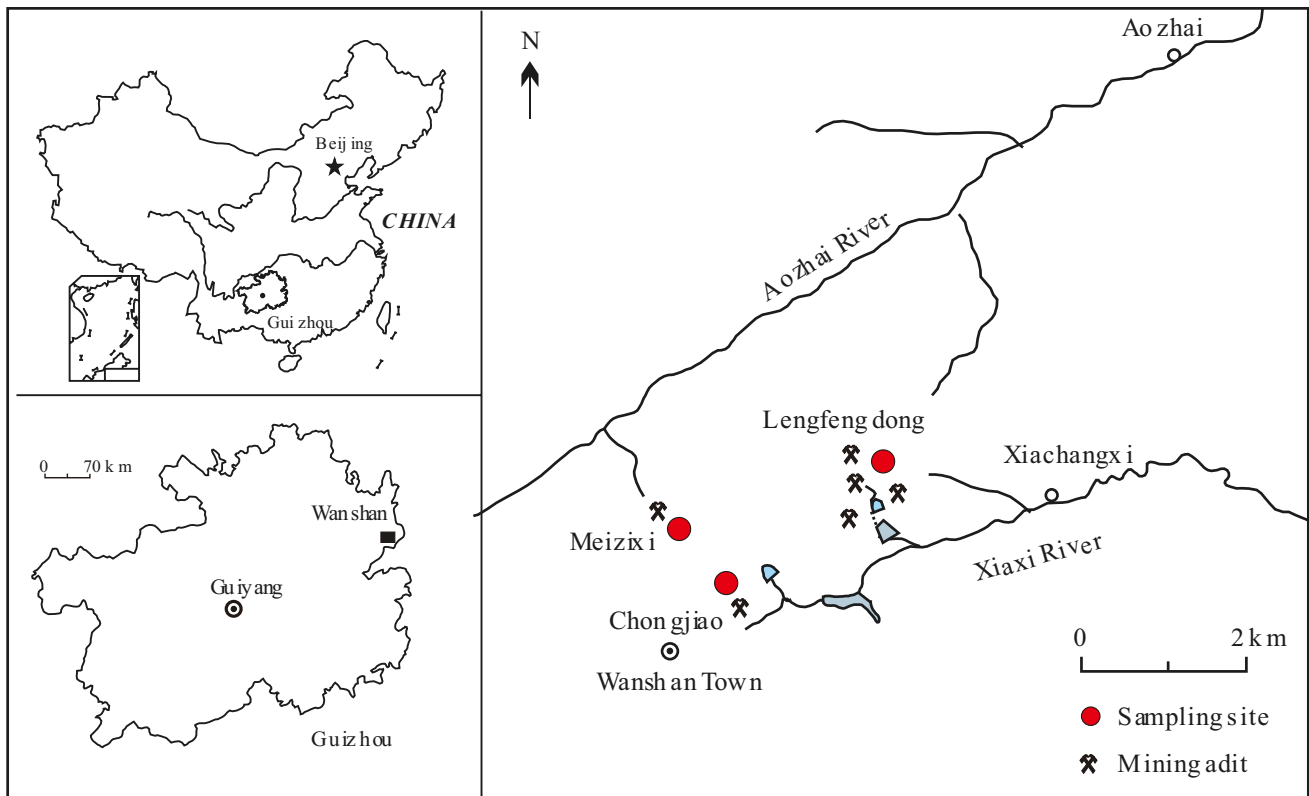
The Wanshan Hg mining district, on the eastern edge of the Yun-Gui Plateau in southwestern China (E: 109°07′–109°24′; N: 27°24′–27°38′), is the largest industrial metallic Hg production centre in China. This region has a typical karst landscape with an average elevation of 850 m. The annual average temperature is 13.4 °C, and the mean annual precipitation is 1400 mm/year. Cinnabar is the main ore mineral associated with metacinnabar, natural metallic Hg, tiemannite, sphalerite, pyrite, and stibnite. The average Hg grade of the ore deposits is higher than 0.25%.

Extensive Hg mining and retorting was performed for 630 years and ceased in 2004. Approximately 125.8 million tonnes of mine-waste calcines were introduced into the environment between the early 1950s and the late 1990s (Qiu et al. 2005). The large historic Hg mining adits of Lengfengdong and Meizixi are at the headwaters of the rivers Xiaxi and Aozhai, the major aquatic systems of the Wanshan mining district. Large mine-waste calcine piles were placed adjacent to the corresponding adits and created a significant area of Hg-contaminated wastelands. In the present study, the wastelands associated with calcine piles from Lengfengdong (LFD), Chongjiao (CJ), and Meizixi (MZX) were selected for investigation (Fig. 1; Table S1).

### Sampling and Preparation

Ninety-five samples of dominant forage plants belonging to 22 species of 18 families that are favoured by grazing animals, were collected from the wastelands. We preferentially sampled herbaceous plants rather than woody species. All of the plants were identified to the species level based on descriptions in the Flora of China ([flora.huh.harvard.edu/china/mss/welcome.htm](http://flora.huh.harvard.edu/china/mss/welcome.htm)).

During sampling, dominant forage samples were randomly taken from the wastelands, within a sampling grid of 5 × 5 m. For each sample, three or more similarly sized individual plants of the same species were collected to ensure adequate amounts of tissue for analysis. Plant samples were dug out of the ground with a shovel and separated in situ into aboveground parts (shoots) and roots. In the laboratory, the plants were washed thoroughly with tap water and then three times with deionized water (DW). Afterwards, the plants were frozen in a freezer and



**Fig. 1** Study area and sampling sites

then placed in a vacuum freeze drier ( $-50\text{ }^{\circ}\text{C}$ ) for drying. The dry plants were ground and sieved to a fine powder using an analytical mill (IKA-A11 basic, IKA, Germany) and nylon sieve (mesh size of 0.18 mm). During processing, the lab equipment was rinsed three times with ethanol cleansing to control cross-contamination among the samples. The fine-powder samples were stored in hermetic bags for analysis.

Corresponding rhizosphere soils were simultaneously collected with the plants. Approximately 0.5 kg of rhizosphere soil from the roots of individual plants was shaken onto a piece of paper, and then the total 1.5 kg of soil collected from the three individual plant roots mentioned above was mixed as the final composite sample. The soils were stored in double polyethylene plastic bags to prevent cross-contamination. After collection, all of the soil samples were air-dried in the laboratory, thoroughly mixed, and subsequently ground to a fine powder using an agate mortar and nylon sieve (mesh size of 0.075 mm). A cleansing process similar to the preparation of the plant samples was applied to control cross-contamination among the samples. The fine-powder samples were stored in double zip-lock polyethylene plastic bags for analysis.

## Sample Analysis

### Plant

For THg determination, approximately 0.1–0.2 g (accurate to 0.0001) samples were placed in plastic tubes and digested with 5 mL  $\text{HNO}_3\text{:H}_2\text{SO}_4=4\text{:}1$  (v/v) in a water bath at  $95\text{ }^{\circ}\text{C}$  for 3 h. Afterwards, 5 mL of DW and 0.5 mL of BrCl were added to the solutions and were digested for another 30 min. Finally, the digestion solution was brought to a fixed volume of 50 mL with DW. After leaving the digestion for 24 h, 400  $\mu\text{L}$  of  $\text{NH}_2\text{OH}\cdot\text{HCl}$  was added, and 5.0 mL of the liquid supernatant was transferred to a bubble bottle. Then, 400  $\mu\text{L}$  of  $\text{SnCl}_2$  was added for Hg determination by atomic absorption spectroscopy (AAS, F732-V, Shanghai Huaguang, China) (Qiu et al. 2012).

For MeHg determination, approximately 0.3–0.5 g (accurate to 0.0001) samples were weighed into Teflon tubes, and a 5-mL methanol solution with 25% KOH was added. The samples were digested for 3 h in a water bath at  $75\text{ }^{\circ}\text{C}$ , after which 1.5 mL of concentrated HCl was added to acidify the solution. Subsequently, 10 mL of  $\text{CH}_2\text{Cl}_2$  was added to the digestate, shaken for 30 min, and then separated. The

lower solvent phase  $\text{CH}_2\text{Cl}_2$  was collected in a 50-mL Teflon bottle. Approximately 30 mL of DW was added to the solvent phase in the 50-mL Teflon bottle and the MeHg back-extracted into the new water phase with a fixed volume of 50 mL. Samples of approximately 10 mL were placed into a bubbler for MeHg determination by gas chromatography-cold vapor atomic fluorescence spectroscopy (GC-CVAFS) according to the US EPA method 1630 (Liang et al. 1996; USEPA 2001) using a Brooks Rand Model III mercury detector (Seattle, WA) followed by a progressive sequence of aqueous phase ethylation, addition of 2M of acetate buffer, ethylation with 1% sodium tetraethylborate. The methyl-ethyl-mercury was purged onto Tenax traps, from which it was subsequently thermally desorbed and separated for MeHg detection.

The bioaccumulation of heavy metals from soil can be described using bioconcentration factors (BCFs) and transfer factors (TFs) (Yoon et al. 2006; Gonzaga et al. 2008). To calculate BCFs and TFs of the plants, inorganic Hg (IHg) was calculated. We defined IHg as the difference between THg and MeHg in both the plant roots and shoots according to Lin et al. (2008) and Shi et al. (2005a, b).

## Soil

For THg determination, approximately 0.1–0.2 g (accurate to 0.0001 g) soil samples were weighed and placed into plastic tubes. Then, 5 mL of DW and 5 mL of fresh aqua regia ( $\text{HCl}:\text{HNO}_3 = 3:1$ ,  $v/v$ ) were added. The samples were allowed to rest for 5 min before 1 mL BrCl was added for water bath digestion at 95 °C for 3 h. The digestate was left for 24 h. Following this, 400  $\mu\text{L}$  of  $\text{NH}_2\text{OH}\cdot\text{HCl}$  was added to remove the free halogens, and the samples were brought to a fixed volume of 50 mL with DW. Approximately 5 mL of the digestate was taken for Hg analysis, similar to the methods used for the plants.

For MeHg determination, approximately 0.3–0.4 g (accurate to 0.0001) of soil samples were weighed and placed into 50-mL plastic centrifuge tubes. Next, 1 mL of 2 mol/L of  $\text{CuSO}_4$  and 4 mL concentrated  $\text{HNO}_3:\text{H}_2\text{O} = 1:3$  ( $v/v$ ) were added. Then, 5 mL of ultrapure  $\text{CH}_2\text{Cl}_2$  was added and shaken for 30 min to extract the MeHg into the solvent. Afterwards, the  $\text{CH}_2\text{Cl}_2$  solvent phase was collected in a 50-mL Teflon bottle. Approximately 30 mL of DW was added, and then the MeHg was back-extracted into the new water phase. The extract was brought to a fixed volume of 50 mL with DW (Liang et al. 1996). Approximately 5-mL aliquots were taken for MeHg GC-CVAFS analysis, similar to the procedure used for the plants.

For the soil pH measurements, approximately 10-g soil samples were weighed and placed into plastic vials. Next, 25 mL of DW without  $\text{CO}_2$  was added, mixed for 2 min, and left to settle for 30 min (Lu 2000). The soil pH was

determined using a pH meter (PHS-3E, Shanghai Leici, China).

For soil organic matter (OM) determination, approximately 0.5–1.0 g of soil was weighed and placed into colorimetric tubes. Concentrated sulfuric acid was added and then OM measurement followed a water bath-potassium dichromate volumetric method (Lu 2000).

## Quality Assurance/Quality Control

Quality assurance/quality control (QA/QC) measures consisted of the use of a standard working curve, blanks, sample duplicates, matrix spikes, and the certified reference materials of lichen (BCR-482), lobster hepatopancreas (TORT-2), Chinese yellow–red soil (GBW07405), and estuarine sediment (ERM-CC580), as further described below and in the Supplementary Material (Table S2).

For THg, the method was validated using the reference materials BCR-482 and GBW07405. An average total Hg concentration of  $0.475 \pm 0.02$  mg/kg ( $n=5$ ) was obtained for the lichen standard BCR-482, which was within the range of the certified value of  $0.48 \pm 0.02$  mg/kg. For the soil, GBW07405 was used, and the measured concentration of  $0.32 \pm 0.02$  mg/kg ( $n=5$ ) was within an acceptable range of the certified value of  $0.29 \pm 0.04$  mg/kg.

For MeHg, the obtained value of  $75.0 \pm 3.1$   $\mu\text{g}/\text{kg}$  ( $n=5$ ) met the certified value of  $75.5 \pm 3.7$   $\mu\text{g}/\text{kg}$  for the ERM-CC580 soil standard. In addition, the obtained value of  $155 \pm 25$   $\mu\text{g}/\text{kg}$  ( $n=5$ ) met the certified value of  $152 \pm 13$   $\mu\text{g}/\text{kg}$  for the TORT-2 plant standard. The recovery of THg and MeHg in the solid samples was in the range of 95% to 109% and 87% to 108%, respectively.

## Calculations of BCFs and TFs of IHg and MeHg

In the present study, the IHg and MeHg BCFs were defined as the ratios of their concentration in the plant roots to that in the soil ( $[\text{IHg or MeHg}]_{\text{root}}/[\text{IHg or MeHg}]_{\text{soil}}$ ), reflecting the capability of the plant's roots to absorb and accumulate IHg and MeHg from the soil. The TFs were defined as the ratio of the IHg and MeHg concentration in the shoots of the plants to that in their roots ( $[\text{IHg or MeHg}]_{\text{shoot}}/[\text{IHg or MeHg}]_{\text{root}}$ ), referring to the facility of transport of IHg and MeHg from the roots to the shoots.

## Statistical Analysis

Data analyses were performed using Microsoft Excel 2010 (Microsoft Co. Ltd., USA). CorelDRAW Graphics Suits X8 (Corel Corporation, USA) was used to draw a map of the sampling sites. Other figures and a one-way ANOVA analysis were performed using GraphPad Prism version 8.0.0 for

Windows (GraphPad Software, San Diego, CA) and R v3.6.1 (R Core Team 2019).

## Results and Discussion

### Plant Species

All of the samples belonged to 22 species of 18 families. The colonizing plants on the CJ wasteland consist of 11 species *Aster ageratoides*, *Aster subulatus*, *Buddleja davidii*, *Cibotium barometz*, *Conyza canadensis*, *Corydalis edulis Maxim*, *Gynura bicolor*, *Herba artimisiae sieversianae*, *Rumex japonicus*, *Sonchus brachyotus*, and *Sonchus oleraceus*. These plants have high coverage and biomass; the *Asteraceae* family accounts for 50.0% of the plants identified. Plants, such as *C. canadensis*, are amphibious plants, which have various growth habits but have a strong reproductive capacity and can grow well in places with high Hg concentrations. On the LFD and MZX wastelands, the dominant plants include *A. ageratoides*, *B. davidii*, *H. artimisiae sieversianae*, *Houttuynia cordata*, *Oenanthe javanica*, *Primula sikkimensis*, *Portulaca oleracea*, *Primula sikkimensis*, *R. japonicas*, *A. subulatus*, *Brassica campestris*, *C. canadensis*, *H. cordata*, *Ipomoea batatas*, *Mentha canadensis*, *Plantago asiatica*, *Rumex acetosa*, *R. japonicus*, *Sedum bulbiferum*, *Sedum emarginatum*, and *S. oleraceus*. Of these plants, the

three families *Asteraceae*, *Crassulaceae*, and *Polygonaceae* account for 69.2% of the total.

The wastelands were once arable lands but currently contain a significant amount of mine-waste calcine, resulting in poor nutrient content and extremely high Hg concentrations. These conditions are conducive only to certain plant species with the capacity to grow in disturbed environments and with a high Hg tolerance. The herbaceous *Asteraceae* species accounted for 40% of the total investigated plants. These species exhibit particular characteristics, such as abundant seeds, fast bud and growth rates, high biomass, and resilience, allowing them to become the dominant plant colonies on the wastelands.

### Concentrations of THg and MeHg

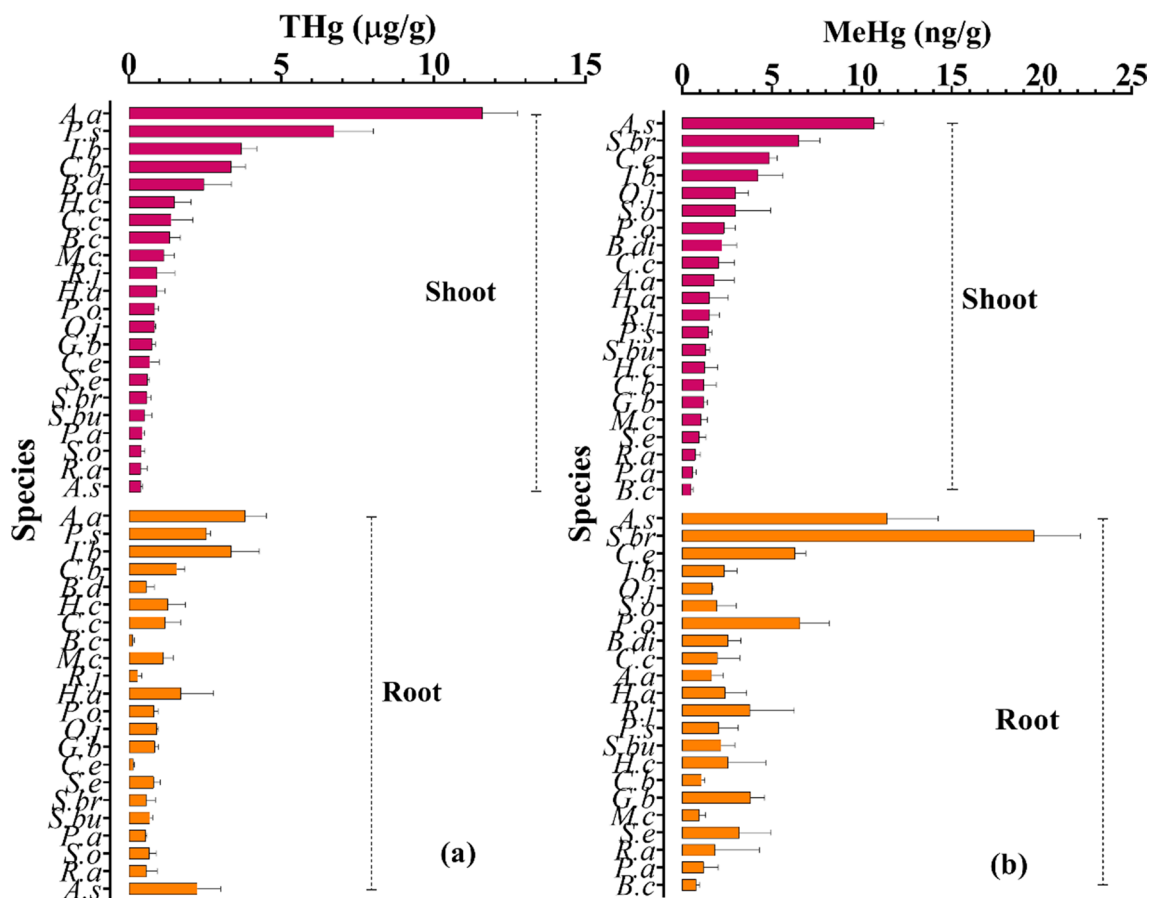
#### THg

THg exhibited a wide range of concentrations in the roots and shoots of the 22 species, ranging from 0.10 to 4.4 mg/kg and 0.19 to 13 mg/kg, respectively (Table 1; Fig. 2a). Compared with the average THg values among the different species, the shoots of *A. ageratoides* showed the highest THg level, reaching  $12 \pm 1.1$  mg/kg, followed by *P. sikkimensis* with  $6.7 \pm 1.2$  mg/kg, while the lowest THg level was in *A. subulatus* with  $0.39 \pm 0.055$  mg/kg. The *A. ageratoides* roots also showed the highest THg level of  $3.8 \pm 0.69$  mg/kg, on

**Table 1** Concentrations of THg and its BCFs and TFs in wild plants inhabiting the wastelands of the Wanshan Hg mining region, Guizhou Province, Southwest China (mg/kg)

Species (acronym)	Soil	Root	Shoot	BCFs	TFs
<i>Aster ageratoides</i> (A.a)	32 ± 5.6	3.8 ± 0.69	12 ± 1.1	0.12 ± 0.028	3.1 ± 0.37
<i>Aster subulatus</i> (A.s)	34 ± 6.5	2.3 ± 0.76	0.39 ± 0.055	0.065 ± 0.018	0.19 ± 0.09
<i>Brassica campestris</i> L. (B.c)	24 ± 7.1	0.13 ± 0.052	1.4 ± 0.34	0.006 ± 0.002	11 ± 1.9
<i>Buddleja davidii</i> (B.d)	46 ± 14	0.58 ± 0.26	2.5 ± 0.91	0.013 ± 0.004	4.5 ± 1.4
<i>Cibotium barometz</i> L. (C.b)	37 ± 3.5	1.6 ± 0.26	3.4 ± 0.46	0.043 ± 0.011	2.2 ± 0.072
<i>Conyza canadensis</i> (C.c)	31 ± 15	1.2 ± 0.50	1.4 ± 0.71	0.040 ± 0.012	1.3 ± 0.47
<i>Corydalis edulis Maxim.</i> (C.e)	24 ± 5.7	0.15 ± 0.038	0.69 ± 0.30	0.006 ± 0.003	4.5 ± 1.1
<i>Gynura bicolor</i> (G.b)	43 ± 5.5	0.86 ± 0.11	0.76 ± 0.11	0.019 ± 0.001	0.90 ± 0.22
<i>Herba artimisiae</i> (H.a)	60 ± 28	1.7 ± 1.1	0.92 ± 0.27	0.029 ± 0.014	0.72 ± 0.40
<i>Houttuynia cordata</i> (H.c)	179 ± 39	1.3 ± 0.57	1.5 ± 0.53	0.007 ± 0.003	1.4 ± 0.86
<i>Ipomoea batatas</i> (I.b)	355 ± 113	3.4 ± 0.92	3.7 ± 0.49	0.010 ± 0.003	1.22 ± 0.58
<i>Mentha canadensis</i> (M.c)	132 ± 40	1.1 ± 0.32	1.2 ± 0.34	0.009 ± 0.004	1.1 ± 0.56
<i>Oenanthe javanica</i> (O.j)	61 ± 21	0.92 ± 0.040	0.84 ± 0.041	0.02 ± 0.0008	0.92 ± 0.08
<i>Plantago asiatica</i> (P.a)	159 ± 68	0.56 ± 0.034	0.45 ± 0.070	0.004 ± 0.002	0.81 ± 0.17
<i>Portulaca oleracea</i> (P.o)	72 ± 18	0.83 ± 0.13	0.84 ± 0.13	0.012 ± 0.002	1.0 ± 0.045
<i>Primula sikkimensis</i> (P.s)	135 ± 17	2.6 ± 0.12	6.7 ± 1.2	0.019 ± 0.002	2.7 ± 0.64
<i>Rumex acetosa</i> (R.a)	85 ± 3.9	0.57 ± 0.36	0.40 ± 0.20	0.007 ± 0.003	0.73 ± 0.18
<i>Rumex japonicus</i> (R.j)	47 ± 18	0.30 ± 0.13	0.93 ± 0.57	0.007 ± 0.002	3.2 ± 1.4
<i>Sedum bulbiferum</i> (S.bu)	252 ± 23	0.69 ± 0.090	0.51 ± 0.24	0.003 ± 0.0002	0.72 ± 0.27
<i>Sedum emarginatum</i> (S.e)	53 ± 23	0.82 ± 0.21	0.61 ± 0.067	0.017 ± 0.007	0.81 ± 0.32
<i>Sonchus brachyotus</i> (S.br)	45 ± 4.9	0.58 ± 0.29	0.60 ± 0.12	0.013 ± 0.008	1.3 ± 0.77
<i>Sonchus oleraceus</i> (S.o)	121 ± 2.7	0.67 ± 0.22	0.41 ± 0.12	0.006 ± 0.003	0.69 ± 0.44





**Fig. 2** THg and MeHg in the roots and shoots of the forage plants

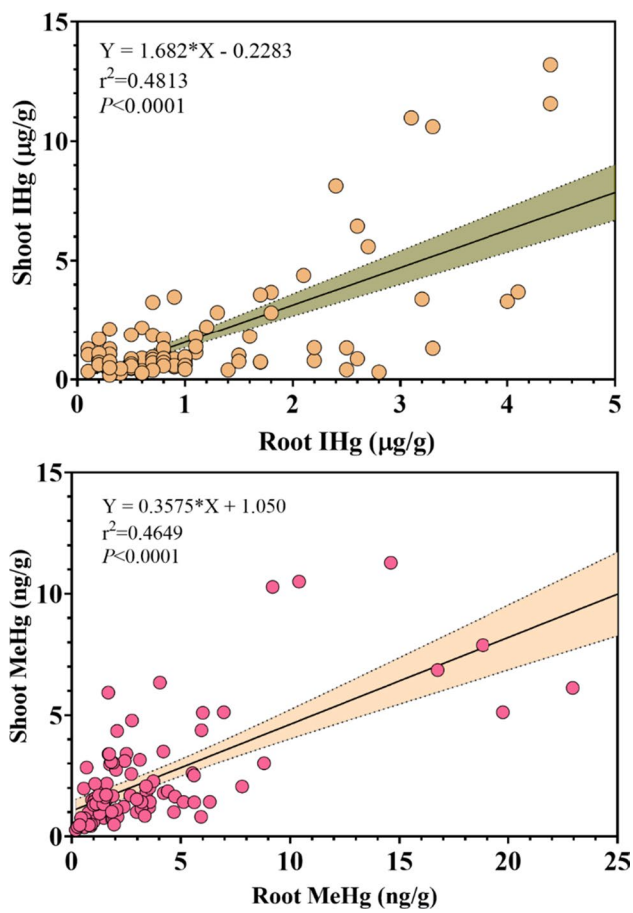
average, while the lowest THg level of  $0.13 \pm 0.052$  mg/kg was in *B. campestris*. One-way ANOVA tests showed no differences in the THg concentrations in both the roots and shoots of the plants across the different wastelands, even though significant differences in soil THg were observed between CJ and MZX ( $P=0.0007$ ) and between LFD and MZX ( $P=0.0108$ ) (Table 2; Fig. S1). A positive correlation

was detected between the shoots and roots ( $r^2=0.48$ ,  $P<0.0001$ ; Fig. 3a).

The plants in the present study exhibited a comparable level of THg (9.9 mg/kg on average) to that recently reported from artisanal and small-scale mining in the Bombana, Indonesia, although this was much higher than the THg levels observed in wild plants from the Alacrán gold mining area

**Table 2** THg and MeHg concentrations in the plants as well as their rhizosphere soils from the three sampling sites of the Wanshan Hg mining region, Guizhou Province, Southwest China

Conc		Chongjiao			Lengfengdong			Meizixi		
		Soil	Root	Shoot	Soil	Root	Shoot	Soil	Root	Shoot
THg (mg/kg)	Minimum	14	0.56	0.61	24	0.19	0.19	16	0.1	0.24
	Median	40	0.80	0.89	57	0.81	0.95	105	0.84	0.70
	Maximum	308	23	11	176	4.4	13	456	4.1	5.6
	Mean $\pm$ SD	$57 \pm 56$	$4.8 \pm 6.1$	$3.3 \pm 2.6$	$73 \pm 47$	$1.2 \pm 1.1$	$2.8 \pm 3.7$	$133 \pm 104$	$1.2 \pm 0.96$	$1.1 \pm 1.0$
MeHg ( $\mu$ g/kg)	Minimum	0.74	0.56	0.61	1.2	0.94	0.81	0.75	0.19	0.28
	Median	4.7	2.0	2.7	2.2	3.4	2.1	3.6	1.9	0.99
	Maximum	23	23	11	5.6	8.8	3.4	19	9.2	11
	Mean $\pm$ SD	$6.5 \pm 6.1$	$4.8 \pm 6.0$	$3.3 \pm 2.6$	$2.9 \pm 1.4$	$3.9 \pm 2.2$	$2.1 \pm 0.75$	$4.6 \pm 3.6$	$2.2 \pm 1.9$	$1.6 \pm 1.9$



**Fig. 3** Correlation of THg and MeHg between the roots and shoots of the forage plants

in the United States (Marrugo-Negrete et al. 2016; Basri et al. 2020), and those observed in agricultural plants of the same region. In the study area, fresh forage plants were the main feedstock for cattle and goats, hence, such elevated levels of Hg leads to high risks for potential exposure for the livestock and wildlife that consume these plants. Among the investigated forage-plant species, *A. ageratoides* recorded the highest THg concentrations both in the shoots and roots, which may be of great concern for herbivorous/omnivorous grazing animals.

### MeHg

Forage plants showed broad ranges of MeHg concentrations in the roots and shoots, ranging from 0.19 to 23 µg/kg and 0.28 to 11 µg/kg, respectively (Table 3; Fig. 2b). The highest average MeHg concentration was found in the shoots of *A. subulatus* at  $7.4 \pm 6.1$  µg/kg, followed by *S. brachyotus* with  $3.5 \pm 2.5$  µg/kg, while the lowest was in *B. camperstris* with  $0.49 \pm 0.11$  µg/kg. *S. brachyotus* exhibited the highest average MeHg concentration in its roots with  $13 \pm 10$  µg/kg,

followed by *A. subulatus* at  $9.4 \pm 8.0$  µg/kg, while the lowest was in *A. ageratoides* at  $1.1 \pm 0.76$  µg/kg. As expected, levels of MeHg in the roots exhibited a significant positive correlation to the MeHg in the shoots ( $r^2 = 0.46$ ,  $P < 0.0001$ ; Fig. 3b), suggesting a strong transport of MeHg from the roots to the aboveground parts of the plants.

Different species exhibited different capabilities for MeHg bioaccumulation. In addition to *A. subulatus*, which recorded the highest levels of MeHg concentration (greater than 10 µg/kg on average) in both the shoots and roots, species *S. brachyotus*, *C. edulis*, and *P. oleracea* also showed high MeHg concentrations, particularly in their roots, with a range of 6.3–19 µg/kg on average. Those values were comparable to those observed in rice in the same region (Zhao et al. 2016; Xu et al. 2017). Such significantly high levels of MeHg in both the shoots and roots may result in heavy body burdens of MeHg in herbivores as well as their predators because of how easily MeHg is absorbed and accumulated by organisms. This may explain the high MeHg levels in herbivores identified in the study region, as report by Abey-singhe et al. (2017).

### BCFs and TFs for IHg and MeHg

#### BCFs

Plants exhibited a wide range of BCFs for IHg and MeHg, ranging from 0.0023 to 0.16 and from 0.022 to 17, respectively. The BCFs for IHg in all species were less than 1, with the highest average value in *A. ageratoides* at  $0.12 \pm 0.028$ . For MeHg, seven species, *A. ageratoides*, *A. subulatus*, *B. davidii*, *P. oleracea*, *R. japonicus*, *S. emarginatum*, and *S. brachyotus* exhibited peak values exceeding 1.0, ranging from 1.3 to 14 on average. The *A. subulatus* showed the highest MeHg BCF of  $14 \pm 2.5$ , on average, followed by *S. brachyotus* with  $3.8 \pm 1.8$ , while the lowest value of  $0.13 \pm 0.049$  was in *I. batatas* (Tables 1 and 3; Fig. 4). The extent of bioaccumulation of MeHg in plants is likely dependent on the species. Previous studies have reported that BCFs of THg and/or IHg in plants are usually less than 0.5 (Zhang et al. 2010; Cosio et al. 2014), whereas the BCFs of MeHg were usually greater than that of THg and/or IHg (Schwesig and Krebs 2003; Tong et al. 2013), which was consistent with our data.

#### TFs

The TFs for IHg in the plants varied widely, ranging between 0.12 and 13. The lowest TF value differed from the highest by more than 100 times. Most of the species exhibited average values of TF greater than 1, and among them, *B. camperstris* exhibited the highest value of  $11 \pm 1.9$  on average. For MeHg, eight species exhibited peak values exceeding 1.0,

**Table 3** Concentrations of MeHg and its BCFs and TFs in wild plants inhabiting the wastelands of the Wanshan Hg mining region, Guizhou Province, Southwest China ( $\mu\text{g}/\text{kg}$ )

Species (acronym)	Soil	Root	Shoot	BCFs	TFs
<i>Aster ageratoides</i> (A.a)	1.4±0.15	1.6±0.65	1.8±1.1	1.2±0.58	1.1±0.45
<i>Aster subulatus</i> (A.s)	0.78±0.055	11.4±2.8	11±0.53	14±2.5	0.97±0.18
<i>Brassica campestris</i> L. (B.c)	5.8±2.7	0.79±0.18	0.49±0.11	0.16±0.088	0.63±0.006
<i>Buddleja daviddii</i> (B.d)	2.8±2.1	2.6±0.69	2.2±0.81	1.3±0.68	0.98±0.56
<i>Cibotium barometz</i> L. (C.b)	16±11	1.1±0.18	1.2±0.67	0.16±0.21	1.1±0.45
<i>Conyza canadensis</i> (C.c)	2.9±1.3	1.9±1.22	2.1±0.87	0.82±0.71	1.7±1.4
<i>Corydalis edulis</i> Maxim. (C.e)	10±1.2	6.3±0.57	4.9±0.42	0.63±0.038	0.77±0.066
<i>Gynura bicolor</i> (G.b)	11±0.99	3.8±0.77	1.2±0.21	0.34±0.10	0.32±0.10
<i>Herba artimisiae</i> (H.a)	5.0±2.4	2.4±1.2	1.5±1.0	0.76±0.73	0.62±0.20
<i>Houttuynia cordata</i> (H.c)	4.9±2.3	2.6±2.1	1.3±0.71	0.73±0.72	0.77±0.54
<i>Ipomoea batata</i> (I.b)	18±2.2	2.3±0.71	4.2±1.4	0.13±0.049	1.9±1.1
<i>Mentha canadensis</i> (M.c)	2.9±0.90	0.97±0.34	1.1±0.36	0.36±0.18	1.1±0.34
<i>Oenanthe javanica</i> (O.j)	2.9±2.3	1.7±0.057	2.9±0.70	0.80±0.42	1.8±0.37
<i>Plantago asiatica</i> (P.a)	4.4±1.6	1.2±0.79	0.59±0.19	0.38±0.36	0.67±0.43
<i>Portulaca oleracea</i> (P.o)	2.8±0.58	6.6±1.6	2.3±0.62	2.4±0.48	0.37±0.10
<i>Primula sikkimensis</i> (P.s)	3.8±0.87	2.1±1.1	1.5±0.18	0.52±0.18	0.85±0.38
<i>Rumex acetosa</i> (R.a)	4.0±1.5	1.8±2.5	0.75±0.27	0.38±0.44	1.1±0.79
<i>Rumex japonicus</i> (R.j)	2.2±1.7	3.8±2.5	1.5±0.57	2.4±2.3	0.66±0.49
<i>Sedum bulbiferum</i> (S.bu)	4.4±0.82	2.2±0.77	1.3±0.22	0.50±0.18	0.66±0.23
<i>Sedum emarginatum</i> (S.e)	2.1±0.30	3.2±1.8	0.94±0.38	1.5±0.66	0.33±0.15
<i>Sonchus brachyotus</i> (S.br)	5.9±2.2	19±2.6	6.5±1.1	3.8±1.8	0.34±0.088
<i>Sonchus oleraceus</i> (S.o)	2.5±1.5	1.9±1.1	2.9±1.9	0.79±0.26	1.5±0.45

ranging from 1.1 to 1.9, with the highest value observed in *I. batatas* (Tables 1 and 3; Fig. 4). Five species, *A. ageratoides*, *C. barometz*, *C. canadensis*, *I. batatas*, and *M. canadensis* showed high TFs for both IHg and MeHg with peak values exceeding 1.0 on average, indicating their increased ability to accumulate IHg and MeHg as compared with other species.

For all of the species, the MeHg in the roots was found at higher concentrations than that observed in the shoots, and the MeHg showed significantly higher BCFs than that of IHg, confirming that MeHg is more easily absorbed by the roots. Plant roots can absorb MeHg from the soil; the total amount of MeHg in the soil plays a critical role in controlling MeHg concentrations in rice (Meng et al. 2011). At highly Hg-contaminated sites, high amounts of MeHg can be generated due to the active methylation occurring in the water and soil, which could eventually lead to increased MeHg accumulation in plants.

Although all of the plants are capable of growing in a heavily Hg-contaminated environment, the significant variation in the Hg concentrations may be attributable to the different physiological characteristics of the plants (Marrugo-Negrete et al. 2016). Mercury is a unique heavy metal that

can exist in its elemental form in the atmosphere; mining and retorting activities resulted in an even further increase in the atmospheric Hg (i.e., one to three orders of magnitude higher than in the remote regions; Zhang et al. 2016; Xu et al. 2020). Hence, the finding that the shoots of the plants exhibited higher IHg concentrations than those of the roots in the present study may be attributed to the elevated levels of atmospheric Hg, which also can explain the elevated TFs for IHg (Fig. 5).

### Correlations Among Factors Affecting IHg and MeHg in Plants

Rhizosphere soil Hg, as well as soil pH and OM, can affect Hg levels in plants (Zhao et al. 2016; Tang et al. 2018). The average THg concentrations in the investigated plants' rhizosphere soil ranged from 18 to 261 mg/kg THg and from 1.4 to 19  $\mu\text{g}/\text{kg}$  MeHg. Soil pH values were all higher than 7.5 due to the carbonate buffering of the surrounding rock and the introduction of large amounts of MgO and CaO during the retorting of cinnabar ores.

Spearman correlation coefficients of Hg concentrations with bioaccumulation factors are shown in Fig. 6.



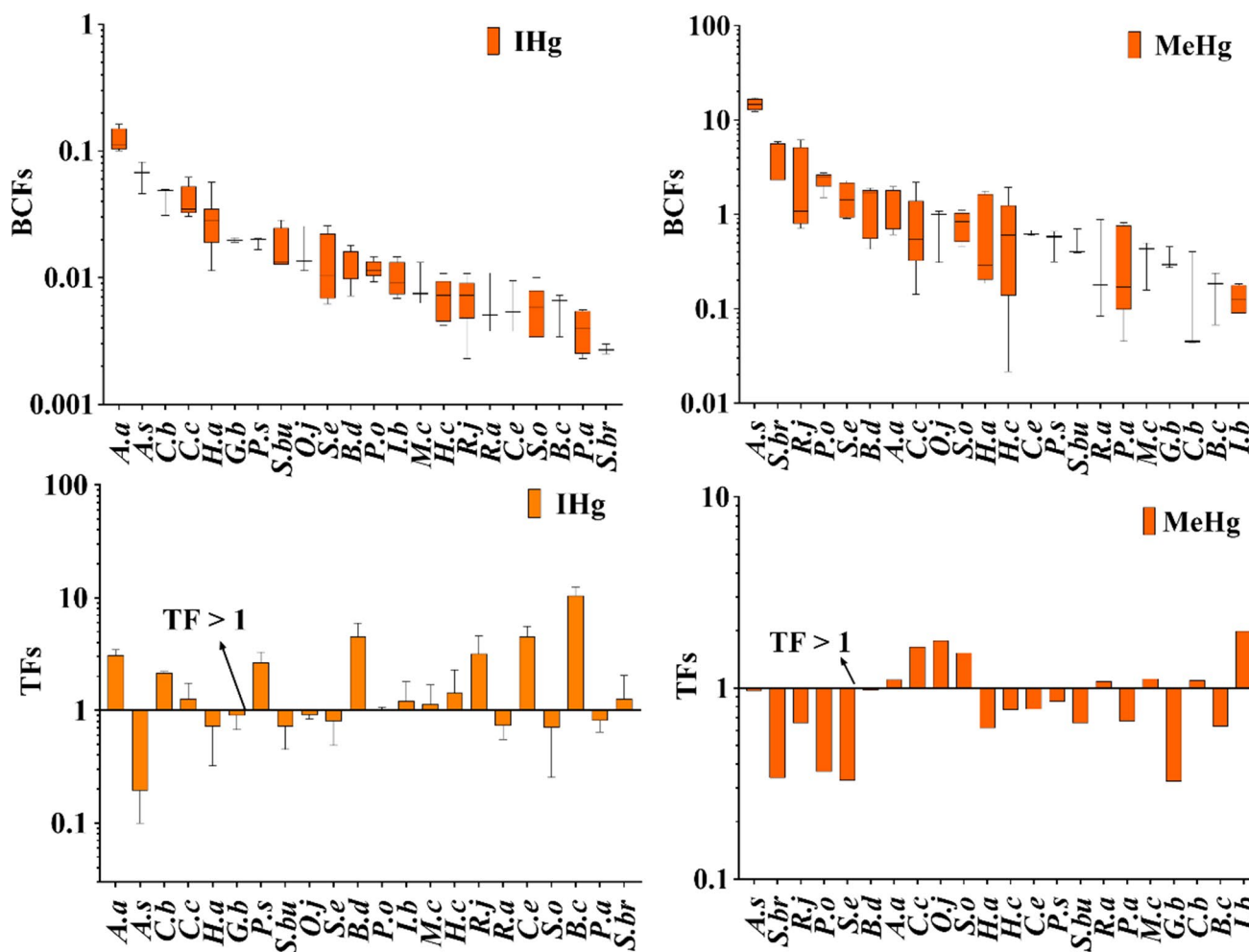
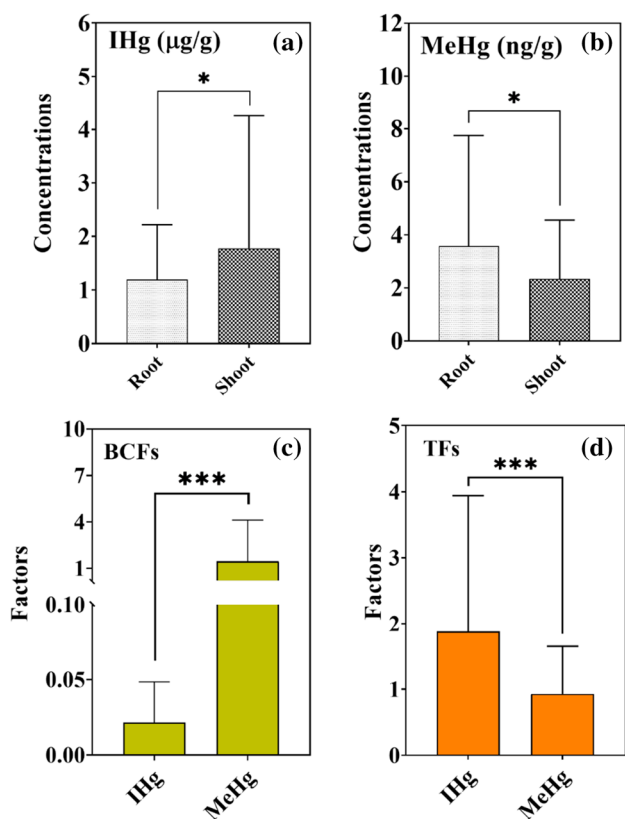


Fig. 4 Distribution of the BCFs and TFs for IHg and MeHg in the forage plants

Root IHg had the most significant correlations with shoot IHg ( $r=0.7$ ,  $P<0.01$ ) and IHg BCFs ( $r=0.66$ ,  $P<0.01$ ), as well as positive correlations with soil THg ( $r=0.31$ ,  $P<0.01$ ) and MeHg ( $r=0.24$ ,  $P<0.05$ ) but negative correlation with IHg TFs ( $r=-0.22$ ,  $P<0.05$ ), reflecting their respective contributions to the accumulation of Hg in the roots. Shoot IHg had fewer correlations with other parameters, showing positive correlation only with IHg BCFs ( $r=0.7$ ,  $P<0.01$ ) and IHg TFs ( $r=0.22$ ,  $P<0.05$ ). The concentrations of root MeHg also exhibited a strong positive correlation to MeHg BCFs as well as shoot MeHg. Interestingly, soil pH exhibited positive correlations to both root MeHg ( $r=0.25$ ,  $P<0.05$ ) and shoot MeHg ( $r=0.34$ ,  $P<0.01$ ) and likewise to MeHg TFs ( $r=0.22$ ,  $P<0.05$ ) as well as MeHg BCFs ( $r=0.17$ ,  $P<0.05$ ),

suggesting that pH was an important factor for controlling the processes of plant MeHg uptake and transport from the soil into the body. In contrast, soil OM showed negative correlations to both MeHg BCFs ( $r=-0.29$ ,  $P<0.01$ ) and MeHg TFs ( $r=-0.32$ ,  $P<0.01$ ) as well as shoot MeHg ( $r=-0.34$ ,  $P<0.01$ ).

In the present study, although soil Hg played an important role in the absorption and enrichment of Hg in the plants, it was found that the rhizosphere soil pH and OM also may play important roles in the process of Hg uptake and transfer in plants. Our results indicate that there is a complex mechanism for Hg uptake, particularly for plants growing in heavily Hg-contaminated sites.



**Fig. 5** Differences in IHg, MeHg, BCFs, and TFs between the roots and shoots of the forage plants (\* $P \leq 0.05$ ; \*\*\*\* $P \leq 0.0001$ )

### Potential Risks for Herbivores

The forage plants investigated in the present study are usually consumed by domestic animals, such as cows, goats, and poultry. Based on the ecotoxicological effects of Hg on organisms, De Vries et al. (2003) recommended a set of critical limits for Hg in plants and categorized Hg concentrations into three hazard levels: high hazard ( $> 3$  mg/kg), low-moderate hazard (0.1–3.0 mg/kg), and low hazard ( $< 0.1$  mg/kg). Considering the shoots, all of the investigated species of forage plant were at a low-moderate or greater hazard level, with approximately 15.8% of them falling into the high hazard level with greater than 3 mg/kg Hg. Moreover, compared with the nationally allowable limit of 0.1 mg/kg THg in vegetables (GB 2762-2017), the average THg concentration in the shoots of most of the forage-plant species was elevated by two to three orders of magnitude.

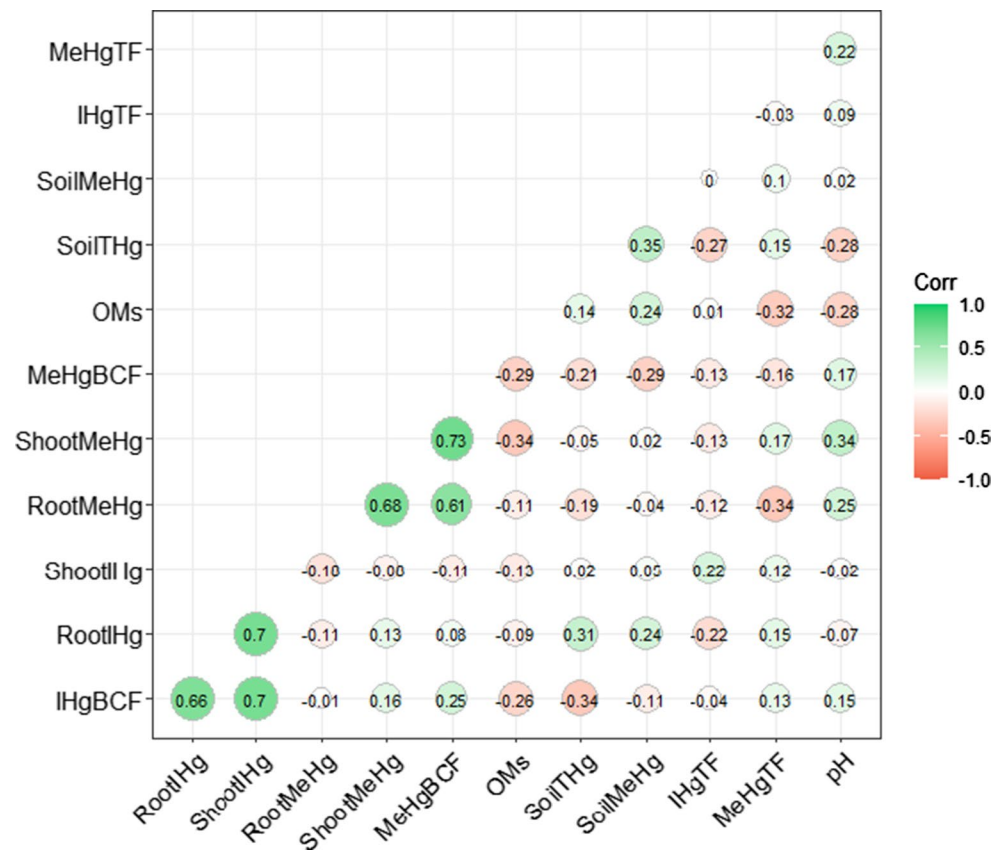
Four species, *A. ageratoides*, *P. sikkimensis*, *I. batatas*, and *C. barometz*, exhibited THg greater than 3 mg/kg in their shoots, suggesting that these plants had high Hg accumulation capabilities. These species are generally

characterized by cold and drought-tolerance, large biomass, and long growing period (Xing et al. 2010; Wang et al. 2011), resulting in their strong growth on the wastelands. Hence, the high concentrations of Hg in their shoots may cause potentially high Hg exposure risks to herbivorous/omnivorous wildlife and feeding livestock.

Currently, critical limits for Hg in plants (grass) for grazing animals are not available. Therefore, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) provisional tolerable weekly intake (PTWI) values for human intake were cited in the risk assessment for herbivores (Gramss and Voigt 2014). Taking into account the PTWI value 4 ng/g for IHg recommended by the JECFA (2010) for human dietary exposure from foods other than fish and shellfish, daily ingestion of 0.037 µg IHg by a 65 kg animal is acceptable. Grazing on 1.0 kg of the shoots of the forage-plants (dry weight) would mean that the daily intake of IHg and MeHg was between 190 and 13,200 µg, which reaches three to five orders of magnitude more than the permitted limit. Moreover, grazing animals, such as cows and sheep, are highly sensitive to contamination given the ingestion of soil along with grass intake; hence, soil heavily contaminated by both THg and MeHg is an additional point of concern.

### Conclusions

The dominant forage plants collected from the wastelands of this large-scale Hg mine exhibited high concentrations of both THg and MeHg, ranging from 0.085 to 13 mg/kg and 0.059 to 24 µg/kg, respectively. The species *A. ageratoides*, *C. barometz*, *I. batatas*, and *P. sikkimensis* exhibited THg levels greater than the high hazard level of 3 mg/kg in their shoots. Moreover, levels of MeHg comparable to that found in rice were observed in *A. subulatus*, *C. edulis*, *S. brachyotus*, and *P. oleracea*. Among those species, *A. ageratoides*, *C. barometz*, and *I. batatas*, all with TFs greater than 1.0, showed great facility in the accumulation and transfer of both IHg and MeHg to their shoots. The species *A. ageratoides*, *A. subulatus*, and *S. brachyotus* in the present study may pose the highest risks for THg and MeHg exposure to biota due to their strong Hg accumulation facility and abundant biomass. Because the investigated forage plants are widely consumed by herbivorous/omnivorous wildlife and grazing livestock, the high THg and MeHg concentrations could directly result in Hg accumulation and biomagnification in the terrestrial food chain. Thus, future studies on the plant-herbivorous-carnivorous food chain are urgently needed to clarify the risks from THg, particularly MeHg, exposure to wildlife and grazing livestock.

**Fig. 6** Pearson correlation analysis of the THg and MeHg

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s00244-021-00826-2>.

**Acknowledgements** Financial support for this work was provided by the talent introduction research project of Guizhou University (2019: 06), the National Key Research and Development Plan [2018YFC1802602], the First-Class Ecology Discipline in Guizhou Province (No. GNYL[2017]007), and the National Natural Science Foundation of China (NSFC: 41573135).

## Declarations

**Conflict of interest** The authors declare that they have no conflict of interests or personal relationships that could have influenced the work reported in the manuscript.

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