



# Concentrations and health risks of heavy metals in soils and crops around the Pingle manganese (Mn) mine area in Guangxi Province, China

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## Abstract

Heavy metal concentrations in agricultural fields and their ecological risks are a source of extensive concern worldwide. A field survey was conducted to investigate the present situation and health risks of heavy metals (Mn, Pb, Zn, Cu, and Cd) and crops (corn, green soybean, sweet potato, persimmon, cassava, and Chinese chestnut) around the Pingle manganese (Mn) mine site in Guangxi Province, China, which was abandoned 20 years ago. The results showed that the ranges of Mn, Pb, Zn, Cu, and Cd were 2706.54–6760.16, 229.37–275.84, 160.39–215.48, 58.11–75.30, and 5.78–6.98 mg kg<sup>-1</sup>, respectively, which were approximately 15.38–38.41, 11.76–14.15, 2.12–2.85, 2.09–2.71, and 21.64–28.75 times greater than their respective background values. Most of these concentrations exceeded the national standard for soil quality (grade 2) developed by the Ministry of Environmental Protection of China (1995). Contamination assessments based on the single contamination index ( $P_i$ ), Nemerow multi-factor index ( $P_{com}$ ), and potential ecological risk index (RI) showed that the sampled soils were severely polluted with these heavy metals, especially Pb and Cd. Similarly, the crops were enriched with Pb and Cd, with concentrations of 10.22–41.78 and 2.33–5.37 mg kg<sup>-1</sup>, respectively, which were much higher than the threshold values of the national food standards. Contamination assessments with  $P_i$  and  $P_{com}$  also showed that the crops were severely polluted with Pb and Cd. The bioaccumulation factor values for Cd were highest among the heavy metals, with an average mean of 0.66 in these six crops, demonstrating that Cd readily accumulates in these crops. An assessment showed that the health risk for adults living in the mining-impacted areas was significant. Our study strongly recommends that heavy metal contamination in agricultural soils and crops grown around the Pingle Mn mining-affected areas should be treated to mitigate the health risks.

**Keywords** Heavy metal · Soil pollution · Foodstuffs · Health risk · Mining-affected

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## Introduction

Anthropogenic activities (mining, smelting, and industries) are important factors contributing to increased heavy metal concentrations in agriculture fields. Heavy metals in soil can create serious problems, such as the accumulation of high heavy metal concentrations in plants/animals that are then consumed by humans. Increasing evidence shows that heavy metal pollution in mining-affected areas has caused damage to the health of the local residents (Lei et al. 2015; Roba et al. 2016; Nawab et al. 2016; Fan et al. 2018). Therefore, it is important and urgent to investigate the heavy metal concentrations in agricultural soils and crops around mine sites and to assess the resulting health risks.

Guangxi Province is famous for being “the hometown of non-ferrous metals” with over 6800 kinds of mines and for

having both the highest reserve quantity and exploitation quantity of manganese (Mn) ore in China (Li 2006). Furthermore, Guangxi is one of the most ecologically fragile regions in China, as it is located in one of the three world's largest karst geomorphic areas. The karst areas in China are the largest in the world, and the karst areas in Guangxi Province account for approximately 60% of this province. These areas are intrinsically more vulnerable than others because of a variety of factors such as geology, geomorphology, hydrogeology, biodiversity, and climate.

At the same time, Guangxi Province has some of the most acidic precipitation in China as it is located in a region prone to acidic precipitation in East Asia. Combined with natural conditions and environmental issues caused by humans, the ecological environment in Guangxi Province is very vulnerable.

Due to dense populations and scarce land resources, residents who live around mine sites must cultivate agriculture crops in the mining-affected soils. However, to date, little is known about how severely these agriculture fields have been contaminated with heavy metals or what the dominant pollutants are. A human health risk assessment has also not been performed to investigate the contamination levels of heavy metals in crops and the influences on people who consume agricultural products from this area. In previous studies, investigations of heavy metals in crops and vegetables have been conducted in various areas (Ávila et al. 2016; Roba et al. 2016; Nawab et al. 2016; Bi et al. 2018; Chaparro et al. 2018). However, few works have been undertaken in karst formation areas that experience acid deposition.

The Pingle Mn mine lies south of the Guilin region of Guangxi Province, China, which is characterized by karst formations and acid deposition. The Pingle Mn mine is classified as one of the large- and medium-sized Mn mines in Guangxi. The mining activities began in 1958 and continued until 1998. In this paper, we consider whether the agricultural soil is polluted by heavy metals, what the main pollutant is, what the contamination degree is, and how great the health risk associated with crops grown around the mine is. The main part of this study was to quantify the amounts of Cu, Zn, Pb, Cd, and Mn in the agricultural soils and the edible parts of representative crops, i.e., corn, green soybean, sweet potato, persimmon, cassava, and Chinese chestnut. The degree of heavy metal contamination in the agricultural soils and crops as well as the associated health risks was the main focus of this paper.

## Materials and methods

### Study site

The Pingle Mn mine (24° 37' N, 111° 40' E) lies south of the Guilin region of Guangxi Province, China, which is well

known for its spectacular karst landforms mainly developed in Devonian and Carboniferous limestone (Deng et al. 1988). Acidic precipitation is very severe in this area. The annual average pH values of precipitation are < 5.6, with the lowest pH values even reaching 2.32–3.15, and the frequency of acid rain is approximately 36.3–88.4% (Cheng et al. 2010). The climate in this area is subtropical monsoon with an annual average temperature of 19.9 °C, an average rainfall of 1636 mm, and an average humidity of 78%. This area is hilly, and crops are mainly cultured around the foothills of the mountains. The shortest horizontal distance between the crops and the mine tailing area was approximately 1.0 km. The crops were irrigated by irrigation canals and ditches.

### Sampling collection

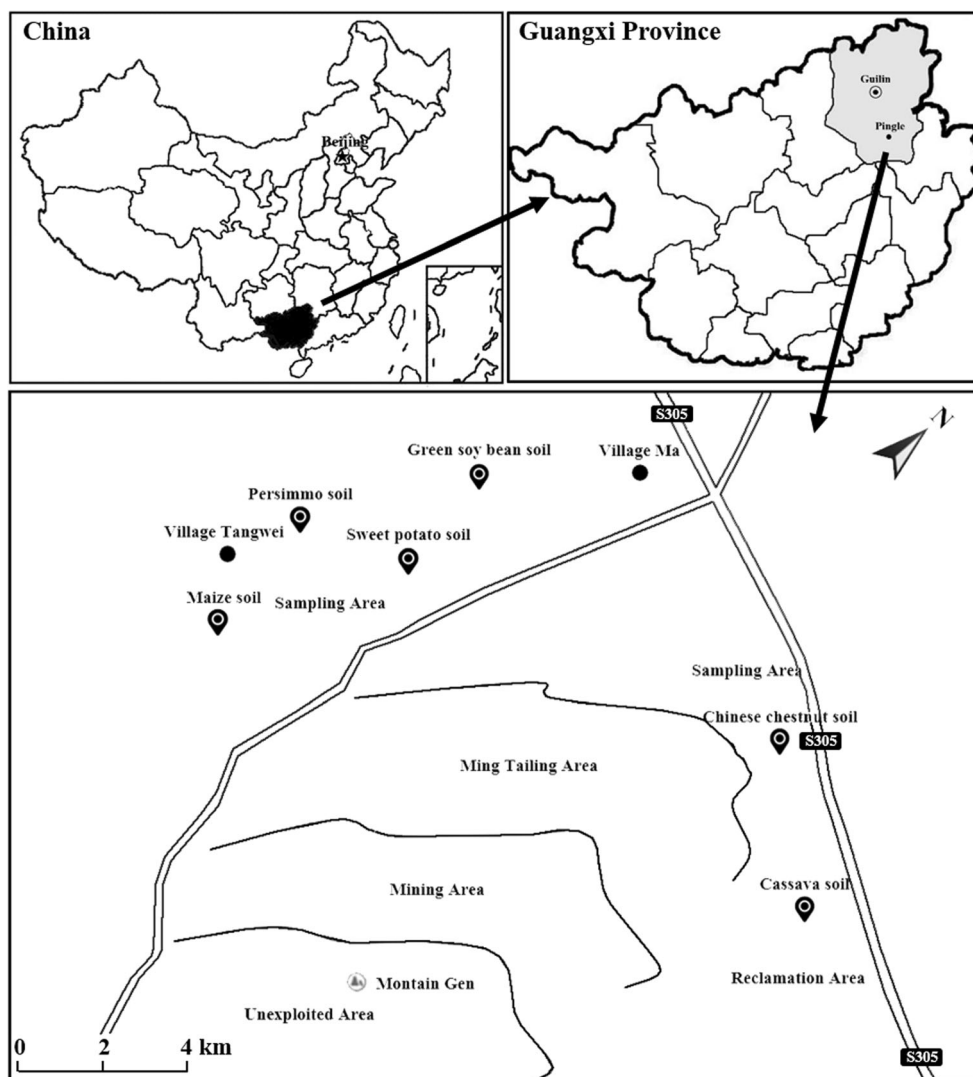
Six crop species were selected for this study: maize (representing cereals), green soybean (representing pulses), cassava and sweet potato (representing tuber crops), Chinese chestnut (representing nuts), and persimmon (representing fruits), which represented the major crop types growing in this area in the sampling season of October 2014. Six sampling sites were selected for each crop based on the “S”-curve sampling method, Torx points sampling method, or random sampling method according to the actual situations (Fig. 1). A total of 108 soil and plant samples were collected. The plant samples consisted of the edible parts of the six studied crops. Each plant sample contained 3–5 individual plants, and the total mass was approximately 1 kg. Samples of the corresponding rhizospheric soils (0–20-cm depth) were taken concurrently. Each soil sample was mixed by pooling six subsamples and then dividing these into three parallel samples. All soil samples were sealed in clean polyethylene plastic bags and brought to the laboratory for measurements.

### Sample pretreatments

Soil sample pretreatments included the following steps: soil samples were air-dried, ground, and passed through a 2-mm sieve and sealed in clean polyethylene plastic bags for analysis of heavy metals.

Plant sample pretreatments included the following steps: all fresh crop samples were thoroughly cleaned with tap water by hand and then washed three times with deionized water. After absorbing the remaining water, the samples were put into an oven to dry for 30 min at 105 °C, and then, they continued to dry at 70 °C until reaching a constant weight. After peeling the inedible parts of the crops, i.e., the shells of the Chinese chestnut and green soybean; the skins of the cassava, sweet potato, maize, and persimmon; and the stigmas of the corn, all samples were ground, homogenized, and then stored in clean polyethylene plastic bags for analysis of heavy metals.

**Fig. 1** The map of the study area and the sampling plots



## Sample analyses

Soil sample measurements included the following: 0.25-g air-dried soil was weighed and put into a 50-ml cleaned digestion tube, and then 8 ml of mixed digestants ( $\text{HCl}:\text{HNO}_3$ , v:v = 1:1) was added for soaking overnight. The mixture was digested for 8–10 h at temperatures of 140–160 °C according to the reaction process. When white precipitate was generated at the bottom of the tube, digestion was terminated, and the digested liquid was cooled to room temperature. The residuum was dissolved by 1%  $\text{HNO}_3$  then transferred into a volumetric flask, diluted to 50 ml, and mixed. The concentrations of Mn, Pb, Zn, Cu, and Cd were measured using an atomic absorption spectrometer (AAAnalyst800, USA). All data was corrected by blank sample values, i.e., mixed digestants, which were prepared in the same way. The soil pH value was measured using a pH meter (Leici pHS-25, China) at a ratio of 1:2.5 (soil:water), after shaking for 0.5 h and then equilibrating for 0.5 h.

Plant sample measurements included the following: the dried plant materials (0.25 g) were digested with 5 ml  $\text{HNO}_3$  and 1 ml  $\text{HClO}_4$  for 4 h. The detailed digestion and measurement procedures were similar to those for the soil.

## Contamination assessments

The single contamination index ( $P_i$ ) (Lee et al. 2008; Lei et al. 2015) and the Nemerow multi-factor index ( $P_{com}$ ) (Ogunkunle and Fatoba 2013) are usually used to assess the contamination level of soil or food. The  $P_i$  and  $P_{com}$  are defined as in Eqs. 1 and 2, respectively.

$$P_i = C_i/S_i \quad (1)$$

where  $P_i$  indicates the single contamination index;  $C_i$ , the measured concentration of the examined metal  $i$  in the soil; and  $S_i$ , the background concentration of metal  $i$ . The evaluation results are divided into five grades, i.e.,  $P_i \leq 0.7$ , safe; 0.7

$< P_i \leq 1.0$ , warning;  $1 < P_i \leq 2$ , light pollution;  $2 < P_i \leq 3$ , moderate pollution; and  $P_i > 3$ , heavy pollution.

$$P_{com} = \left[ \left( \bar{P}^2 + P_{max}^2 \right) / 2 \right]^{1/2} \tag{2}$$

where  $P_{com}$  indicates the composite contamination index;  $\bar{P}$ , the average value of the single-factor index; and  $P_{max}$ , the maximum value of the single-factor index. The evaluation results are divided into five grades, i.e.,  $P_{com} \leq 0.7$ , safe;  $0.7 < P_{com} \leq 1.0$ , warning;  $1 < P_{com} \leq 2$ , light pollution;  $2 < P_{com} \leq 3$ , moderate pollution; and  $P_{com} > 3$ , heavy pollution.

### Risks assessments

Soil ecological and human health risk assessments serve as direct evaluations of food for cultivation and consumption safety. In this paper, monomial potential ecological risk factor ( $E_i$ ), potential ecological risk index (RI), bioaccumulation factor (BAF), adult weekly intake (average weekly dose [AWD]), and health risk assessment (HRA) were applied for the heavy metal risk assessment. The RI, proposed by Hakanson (1980), integrates the concentrations of heavy metals with respect to their ecological effect, environmental effect, and toxicity and has been used to assess the heavy metal pollution and ecological hazards in soils. The detailed calculation processes are calculated according to Eqs. 1, 3, and 4 (Hakanson 1980; Bian et al. 2015; Cai et al. 2015; Nuapia et al. 2018).

$$E_i = T \times P_i \tag{3}$$

$$RI = \sum E_i \tag{4}$$

where  $E_i$  indicates monomial potential ecological risk factor. The evaluation results are divided into five grades, i.e.,  $E_r < 40$ , low potential ecological risk;  $40 \leq E_r < 80$ , moderate potential ecological risk;  $80 \leq E_r < 160$ , considerable potential ecological risk;  $160 \leq E_r < 320$ , high potential ecological risk; and  $E_r \geq 320$ , very high potential ecological risk.  $T_i$  indicates the metal toxicity factor of metal  $i$ ; the standardized values of Cd, Pb, Cu, Zn, and Mn are 30, 5, 5, 1, and 1  $\text{mg kg}^{-1}$ , respectively.  $P_i$  indicates the single contamination index and RI indicates the potential ecological risk index. The evaluation results are divided into five grades, i.e.,  $RI < 150$ , low ecological risk;  $150 \leq RI < 300$ , moderate ecological risk;  $300 \leq RI < 600$ , considerable ecological risk;  $600 \leq RI < 1200$ , very high ecological risk; and  $RI \geq 1200$ , extremely high ecological risk.

The BAF is used in the risk assessment to estimate the trophic transfer of contaminants, such as metals, from soils and can be helpful in the prediction of risks associated with this transfer (Coelho et al. 2018). In this paper, the BAF is the ratio of the metal concentration in the edible parts of crops to

the concentration of the metal in the agricultural soil (Eq. 5) according to Noor-ul-Amin et al. (2013).

$$BAF = \text{metal}_{crops} / \text{metal}_{soils} \tag{5}$$

AWD and HRA were used to assess the human health risks as defined in Eqs. 6 and 7.

$$AWD = (C_m \times W \times 7) / BW \tag{6}$$

$$HRA = AWD / Rfd \times 100 \tag{7}$$

where AWD indicates the average weekly dose ( $\text{mg kg}^{-1} \text{ week}^{-1}$ ) of metal by body weight (BW, the average rural adult body weight is 60 kg);  $C_m$ , heavy metal concentration in the crops;  $W$ , dietary reference intake weight (the values of cassava, persimmon, sweet potato, Chinese chestnut, maize, and green soybean were 10.5, 42.5, 9.3, 33.1 and 8.3  $\text{g BW}^{-1} \text{ day}^{-1}$ , respectively, Cheng et al. 2005); BW, body weight;  $Rfd$ , the permitted weekly tolerable upper intake levels of heavy metals (the values of Cu, Zn, Pb, Pb, and Mn were 3.5, 7.0, 0.025, 0.007, and 0.42  $\text{mg kg}^{-1} \text{ week}^{-1}$ , respectively; China’s Environmental Protection Agency 1990; the Environmental Research Institution of Guangxi 1992); and HRA, health risk assessment ( $HRA < 100$  means the exposed population is assumed to be safe).

### Data and statistical analysis

The data were processed with Microsoft Excel 2010 and SPSS13.0 software. The graphic work was performed using Excel 2010. All the values were expressed as the mean  $\pm$  standard deviation (SD) of three replicates.

## Results and discussions

### Soil heavy metal concentrations and contamination assessments in soils

#### Soil pH value and heavy metal concentrations

Soil pH value is considered a master variable in soils, as it controls many chemical processes that take place. It specifically affects the availability of plant nutrients and toxic substances by controlling the chemical forms of these materials (Ávila et al. 2016). The pH values ranged from 4.83 to 5.94 (Table 1), which were close to those of the background soil (5.3), indicating that the mining activities had no substantial effects on the soil pH value. Naturally occurring soils in Guangxi Province, which are developed from the karst landscape, warm and humid climate, and slight aeolian erosion, are generally acidic (Cheng et al. 2010). All these conditions are favorable for heavy metal

**Table 1** Soil pH value and heavy metal concentrations in the agricultural soil around Pingle Mn mine in Guangxi Province, China (mg kg<sup>-1</sup>)

Samples	pH	Cu	Zn	Pb	Cd	Mn
Cassava soil	4.83 ± 0.19	58.10 ± 0.93	160.39 ± 5.62	229.37 ± 5.71	5.78 ± 0.25	2706.54 ± 46.86
Persimmon and sweet potato soil	5.55 ± 0.40	69.05 ± 1.07	203.89 ± 6.52	254.01 ± 3.31	6.85 ± 0.29	6760.16 ± 182.52
Chinese chestnut soil	4.88 ± 0.94	58.13 ± 0.89	167.57 ± 4.83	241.01 ± 0.69	6.50 ± 0.26	4654.94 ± 177.51
Maize soil	5.20 ± 0.81	75.30 ± 2.26	215.48 ± 15.43	272.13 ± 3.34	6.98 ± 0.18	6329.74 ± 111.17
Green soybean soil	5.94 ± 0.28	69.76 ± 0.69	188.79 ± 4.68	275.84 ± 4.28	5.78 ± 0.30	6440.73 ± 101.72
Background value of soil in Guangxi province*	5.3*	27.8*	75.6*	19.5**	0.267*	176**
GB15618-1995 (MEP 1995)	< 6.5	≤ 150	≤ 200	≤ 250	≤ 0.3	–

\*Data cited from China's Environmental Protection Agency (1990); \*\*Data cited from the environmental research institution of Guangxi (1992). Values are means ± SD ( $n = 3$ )

activation, erosion, and leaching from the soil, which can result in ecological risks (McCauley et al. 2009).

The heavy metal with the highest mean concentration in the agricultural fields was Mn, followed by Pb, Zn, Cu, and Cd. The concentrations of Mn, Pb, Zn, Cu, and Cd were 2706.54–6760.16, 229.37–275.84, 160.39–215.48, 58.11–75.30, and 5.78–6.98 mg kg<sup>-1</sup>, respectively, which were approximately 15.38–38.41, 11.76–14.15, 2.12–2.85, 2.09–2.71, and 21.64–28.75 times their respective background values. Most of these concentrations were over the standards of the national soil quality standard (grade 2, GB15618-1995, MEP 1995). Among all the investigated heavy metals, Mn had the highest absolute concentration, whereas the concentration of Cd exceeded the level of its standards by the greatest degree. According to exceedance of the corresponding standards, Mn and Cd may be the primary heavy metal pollution factors in this mining-affected area, which is consistent with other reports from Guangxi Province (Lai et al. 2007; Jiang and Li 2013). Soils planted with persimmon, sweet potato, and green soybean showed the highest concentrations of Mn and Pb; soils planted with maize showed the highest concentrations of Zn, Cu, and Cd; and soils planted with cassava had the lowest concentration of Mn, Pb, Zn, Cu, and Cd. Further studies are needed to understand whether these differences were due to the differences in crop species or in the soil background or both.

Pearson's correlation was used to determine the pH value and inter-metal relationships among the heavy metals in the soils. This correlation provides the most valuable information about heavy metal concentrations and their sources (Muhammad et al. 2011). Further details are given in supplemental materials (S Table 1).

### Contamination degrees and potential ecological risks

The  $P_i$  evaluations of the heavy metals (Mn, Cd, Cu, Pb, and Zn) in the rhizospheres were carried out in this study based on the environmental quality standard for soils (MEP 1995). The values of  $P_i$  (Table 2) demonstrate that the  $P_{Mn}$  values in the sampled sites were highest, followed by the  $P_i$  values for Cd,

Pb, Cu, and Zn. The  $P_{Cu}$  and  $P_{Zn}$  values were lower than 3, and the other values were all greater than 3 and reached the heavy pollution grade, indicating that the agricultural soils have been severely contaminated with the heavy metals Mn, Cd, and Pb.  $P_{com}$  allows the assessment of soil quality at a much more comprehensive level than evaluations based on a single element (Brady et al. 2015). The assessment results of  $P_{com}$  further demonstrated that the sampled soils were severely polluted as the values were far greater than 5.

$E_r$  represents the ecological risk of each element. The  $E_r$  values for Cu, Zn, and Mn were all lower than 30, while the  $E_r$  values for Cd were much higher than 320 (Table 3), indicating Cd played a major role in soil pollution. This result is similar to results from the other five Mn mine sites of Guangxi Province as reported by Lai et al. (2007). Therefore, special attention should be paid to Cd in this study area since this nonessential, toxic element can be incorporated in organisms (Kirkham 2006). RI represents the sensitivity of the biological community to a toxic substance and illustrates the potential ecological risk posed by the overall contamination. To quantify the overall potential ecological risk posed by the heavy metals in the agricultural soils, the RI values were computed (Table 3). The RI values (723.77–955.71) for the sampled soils were all above 600, demonstrating that the agriculture soils around the Pingle Mn mine had very high potential ecological risks.

### Heavy metal concentrations and health risk assessments of the edible parts of crops

#### Heavy metal concentrations

Uptake from the rhizosphere and accumulation by crop plants represent the main entry pathways for potentially health-threatening toxic metals into human and animal food (Clemens 2006). The heavy metal concentrations in the edible parts of crops collected from agricultural fields around the Pingle Mn mine areas are shown in Table 4. The sequential concentrations in crops were Mn > Pb > Zn > Cu > Cd, while the sequential magnitudes of exceedance of the standards were

**Table 2** Concentration assessments of heavy metals in the agricultural soil around the Pingle Mn mine in Guangxi Province, China

Sampling sites	Single contamination index ( $P_i$ )					Nemerow multi-factor index ( $P_{com}$ )
	$P_{Cu}$	$P_{Zn}$	$P_{Pb}$	$P_{Cd}$	$P_{Mn}$	
Cassava soil	2.09 <sup>M</sup>	2.12 <sup>M</sup>	11.76 <sup>H</sup>	21.64 <sup>H</sup>	15.38 <sup>H</sup>	17.04 <sup>H</sup>
Persimmon/sweet potato soil	2.48 <sup>M</sup>	2.70 <sup>M</sup>	13.03 <sup>H</sup>	25.67 <sup>H</sup>	38.41 <sup>H</sup>	29.55 <sup>H</sup>
Chinese chestnut soil	2.09 <sup>M</sup>	2.22 <sup>M</sup>	12.36 <sup>H</sup>	24.36 <sup>H</sup>	26.45 <sup>H</sup>	21.00 <sup>H</sup>
Maize soil	2.71 <sup>M</sup>	2.85 <sup>M</sup>	13.96 <sup>H</sup>	26.14 <sup>H</sup>	35.96 <sup>H</sup>	27.92 <sup>H</sup>
Green soybean soil	2.51 <sup>M</sup>	2.50 <sup>M</sup>	14.15 <sup>H</sup>	28.75 <sup>H</sup>	36.60 <sup>H</sup>	28.51 <sup>H</sup>

$P_i$ , single contamination index; the evaluation results are divided into 5 grades, i.e.,  $P_i \leq 0.7$ , safe;  $0.7 < P_i \leq 1.0$ , warning;  $1 < P_i \leq 2$ , light pollution;  $2 < P_i \leq 3$ , moderate pollution;  $P_i > 3$ , heavy pollution.  $P_{com}$ , Nemerow multi-factor index; the evaluation results are divided into 5 grades, i.e.,  $P_{com} \leq 0.7$ , safe;  $0.7 < P_{com} \leq 1.0$ , warning;  $1 < P_{com} \leq 2$ , light pollution;  $2 < P_{com} \leq 3$ , moderate pollution;  $P_{com} > 3$ , heavy pollution. *C* clean, *L*, light pollution, *M* medium pollution, *H* heavy pollution

Pb > Cd > Zn > Cu. The concentration ranges of Pb and Cd were 10.22–41.78 and 2.33–5.37 mg kg<sup>-1</sup>, respectively, thus exceeding the national food standards by 147–496 times and 9–128 times, respectively. The concentrations of Zn and Cu exceeded the threshold values prescribed in the national food standards by 166% and 150%, respectively. The corresponding value of Mn was calculated due to the lack of a comparable standard. In terms of the crop species, the highest concentrations of Mn, Pb, Zn, Cu, and Cd were present in Chinese chestnut, sweet potato, maize, Chinese chestnut, and sweet potato, respectively; the lowest concentrations existed in maize, green soybean, persimmon, maize, and persimmon, respectively.

**Heavy metal pollution indices and contamination degrees**

In terms of the environmental risk evaluation, the risk estimation can be conducted based on a description of bioindicator organisms and biological parameter evaluations, which provide a more realistic idea of the true impacts of the contaminants in an ecosystem than the classical approach of direct metal quantification in a studied matrix (Coelho et al. 2018). In this study, the BAF values in different edible parts of plants

collected from the agricultural fields around the Pingle Mn mine were calculated (Fig. 2). The overall BAF values were in the order of Cd > Zn ≈ Cu > Pb > Mn. The BAF values for Cd were in range of 0.41–0.95, with an average mean of 0.66 in these six crops, and these values were much higher than the ranges and means of the other four heavy metals, i.e., 0.02–0.27 and 0.13 for Cu, 0.083–0.32 and 0.17 for Zn, 0.11–0.15 and 0.13 for Pb, and 0.0029–0.12 and 0.28 for Mn, respectively. These results demonstrated that Cd accumulated much more easily in crops than the other heavy metals, and more attention should be paid to this element, as it is highly dangerous to human health (Ghosha and Indra 2018). In terms of crop species, the BAF values for Cu, Pb, and Mn were highest in Chinese chestnut, and the BAF values for Zn and Cd were highest in green soybean. The lowest BAF values for Cu, Pb, and Mn were found in maize, while the lowest BAF values of Zn and Cd were found in persimmon. The overall BAF values were in the order of Cd > Zn ≈ Cu > Pb > Mn. The values were all less than 1, similar to the study results in Nawab et al. (2016) but lower than the results in Khan et al. (2010), who noted that the BAF values for Cd, Ni, and Pb were greater than 1 for most of their studied vegetables. Similarly, our results

**Table 3** Potential ecological risk assessment of the agricultural soil around Pingle Mn mine in Guangxi Province, China

Sampling sites	Individual potential ecological risk factor ( $E_i$ )					Ecological risk (RI)
	$E_{Cu}$	$E_{Zn}$	$E_{Pb}$	$E_{Cd}$	$E_{Mn}$	
Cassava soil	10.45 <sup>L</sup>	2.12 <sup>L</sup>	58.81 <sup>M</sup>	649.31 <sup>VH</sup>	3.08 <sup>L</sup>	723.77 <sup>VH</sup>
Persimmon/sweet potato soil	12.42 <sup>L</sup>	2.70 <sup>L</sup>	65.13 <sup>M</sup>	769.97 <sup>VH</sup>	7.68 <sup>L</sup>	857.90 <sup>VH</sup>
Chinese chestnut soil	10.46 <sup>L</sup>	2.22 <sup>L</sup>	61.80 <sup>M</sup>	730.81 <sup>VH</sup>	5.29 <sup>L</sup>	810.57 <sup>VH</sup>
Maize soil	13.54 <sup>L</sup>	2.85 <sup>L</sup>	69.78 <sup>M</sup>	784.09 <sup>VH</sup>	7.19 <sup>L</sup>	877.46 <sup>VH</sup>
Green soybean soil	12.55 <sup>L</sup>	2.50 <sup>L</sup>	70.73 <sup>M</sup>	862.62 <sup>VH</sup>	7.32 <sup>L</sup>	955.71 <sup>VH</sup>

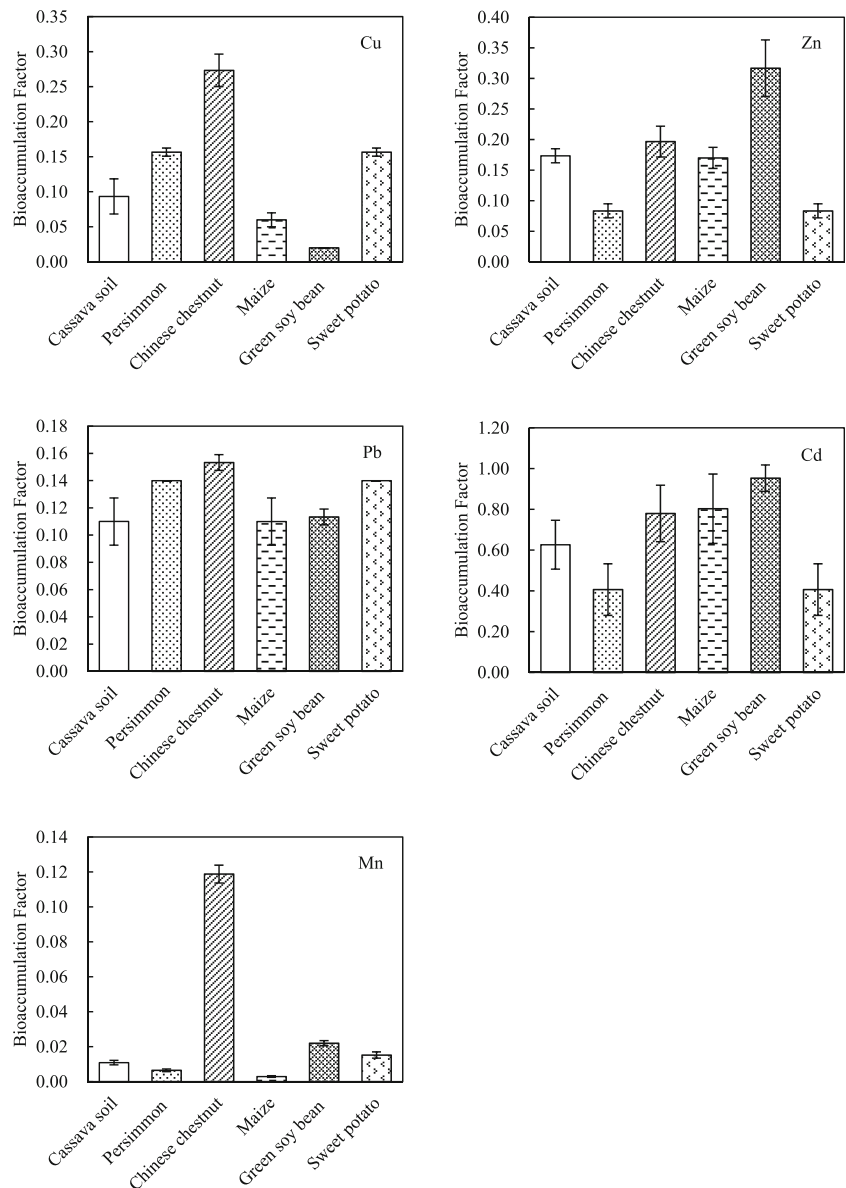
$E_i$ , monomial potential ecological risk factor; standards,  $E_r < 40$ , low potential ecological risk (L);  $40 \leq E_r < 80$ , moderate potential ecological risk (M);  $80 \leq E_r < 160$ , considerable potential ecological risk (C);  $160 \leq E_r < 320$ , high potential ecological risk (H),  $E_r \geq 320$ , very high potential ecological risk (VH).  $T_i$ , the metal toxic factor; the standardized values of Cd, Pb, Cu, Zn, and Mn are 30, 5, 5, 1, and 1 respectively.  $P_i$ , single contamination index. RI, the potential ecological risk index; the evaluation results are divided into 5 grades, i.e.,  $RI < 150$ , low ecological risk (L);  $150 \leq RI < 300$ , moderate ecological risk (M);  $300 \leq RI < 600$ , considered ecological risk (C);  $600 \leq RI < 1200$ , very high ecological risk (VH);  $RI \geq 1200$ , extremely high ecological risk (EH)

**Table 4** Heavy metal concentrations in the edible parts of crops (fresh weight) around Pingle Mn mine in Guangxi Province, China ( $\text{mg kg}^{-1}$ )

Crop species	Cu	Zn	Pb	Cd	Mn
Cassava	$4.68 \pm 0.80$	$22.86 \pm 0.27$	$21.24 \pm 2.71$	$3.01 \pm 0.23$	$24.7 \pm 2.34$
Persimmon	$9.16 \pm 0.14$	$14.74 \pm 0.63$	$30.89 \pm 0.36$	$2.33 \pm 0.41$	$37.31 \pm 1.92$
Sweet potato	$10.64 \pm 0.53$	$21.05 \pm 1.29$	$41.78 \pm 0.02$	$5.37 \pm 0.24$	$80.07 \pm 5.92$
Chinese chestnut	$12.05 \pm 0.94$	$24.81 \pm 1.75$	$27.48 \pm 0.67$	$3.83 \pm 0.40$	$98.33 \pm 0.46$
Maize	$4.1 \pm 0.37$	$31.29 \pm 1.76$	$25.90 \pm 2.77$	$4.91 \pm 0.83$	$16.02 \pm 1.49$
Green soybean	$4.92 \pm 1.16$	$19.80 \pm 3.90$	$10.22 \pm 0.60$	$2.44 \pm 0.58$	$57.52 \pm 2.65$
Average values in crops	7.59	22.43	26.25	3.65	52.28
Threshold values in food of the national food standard	10–20	5–100	0.1–0.3	0.05–0.2	–
	GB 15199-1994	GB 13106-1994	GB 2762-2005	GB 2762-2005	–

– means there is no standard

**Fig. 2** Heavy metal bioaccumulation factor (BAF) values for the selected heavy metals in crops around the Pingle Mn mine in Guangxi Province, China



**Table 5** Heavy metal pollution indices and the corresponding pollution degrees of the crops around Pingle Mn mine in Guangxi Province, China

Crop species	Single contamination index ( $P_i$ )					Nemerow multi-factor index ( $P_{com}$ )
	$P_{Cu}$	$P_{Zn}$	$P_{Pb}$	$P_{Cd}$	$P_{Mn}$	
Cassava	0.23 <sup>S</sup>	0.23 <sup>S</sup>	70.80 <sup>H</sup>	15.05 <sup>H</sup>	–	52.34 <sup>H</sup>
Persimmon	0.46 <sup>S</sup>	0.15 <sup>S</sup>	102.97 <sup>H</sup>	11.65 <sup>H</sup>	–	75.60 <sup>H</sup>
Sweet potato	0.53 <sup>S</sup>	0.21 <sup>S</sup>	139.27 <sup>H</sup>	26.85 <sup>H</sup>	–	102.80 <sup>H</sup>
Chinese chestnut	0.60 <sup>S</sup>	0.25 <sup>S</sup>	91.60 <sup>H</sup>	19.15 <sup>H</sup>	–	67.71 <sup>H</sup>
Maize	0.21 <sup>S</sup>	0.31 <sup>S</sup>	86.33 <sup>H</sup>	24.55 <sup>H</sup>	–	64.14 <sup>H</sup>
Green soybean	0.25 <sup>S</sup>	0.20 <sup>S</sup>	34.07 <sup>H</sup>	12.20 <sup>H</sup>	–	25.46 <sup>H</sup>

The data calculated according to the ceiling values of the threshold values (Table 4);  $P_i$ , single contamination index; the evaluation results are divided into 5 grades, i.e.,  $P_i \leq 0.7$ , safe;  $0.7 < P_i \leq 1.0$ , warning;  $1 < P_i \leq 2$ , light pollution;  $2 < P_i \leq 3$ , moderate pollution;  $P_i > 3$ , heavy pollution.  $P_{com}$ , Nemerow multi-factor index; the evaluation results are divided into 5 grades, i.e.,  $P_{com} \leq 0.7$ , safe;  $0.7 < P_{com} \leq 1.0$ , warning;  $1 < P_{com} \leq 2$ , light pollution;  $2 < P_{com} \leq 3$ , moderate pollution;  $P_{com} > 3$ , heavy pollution. C clean, L, light pollution, M medium pollution, H heavy pollution; –, not assess here for shortage of National Food Hygiene Standards of Mn

were well below the findings of Noor-ul-Amin et al. (2013). The differences in metal uptakes may be attributed to the different capabilities of metal absorption and translocation within the plants (Vousta et al. 1996).

The evaluations of  $P_i$  of heavy metals (Mn, Cd, Cu, Pb, and Zn) in the edible parts of crops were carried out for the five heavy metals in this study based on threshold values from the national food standard. Table 5 presents the  $P_i$  values of the heavy metals in the edible parts of the crops. The  $P_i$  values of both Cu and Zn were below 0.7, suggesting that the crops were not polluted by Cu and Zn. In contrast, the corresponding values of both Pb and Cd were far greater than 3, with the highest  $P_{Pb}$  value reaching 139.27, suggesting that the crops were heavily contaminated by these two heavy metals. The results were consistent with those of previous studies in some other areas in Guangxi Province by Lai et al. (2007), Yuan

et al. (2010), and Huang et al. (2011). The analysis of  $P_{com}$  (25.46–102.80) further showed that all crops were severely contaminated by heavy metals. The sequential  $P_{com}$  values in the crops were sweet potato > persimmon > cassava > Chinese chestnut > green soybean > maize.

### Human health risk assessments

Soil pollution by heavy metals ultimately affects human health via air absorption, skin contact, food chains, etc. Among these processes, food chains are the most common pathway (Khan et al. 2015). The AWD and HRA (%) values calculated for the health risk assessments for adults are presented in Table 6. The highest values for AWD varied among the crop species and heavy metals. Pb represented the highest health risk in all the crops due to its lowest  $Rfd$  value ( $0.007 \text{ mg kg}^{-1} \text{ week}^{-1}$ ),

**Table 6** Human health risk assessments for adults around Pingle Mn mine in Guangxi Province, China

Crop species	Contents in adult ( $\text{mg kg}^{-1} \text{ week}^{-1}$ )	$HRA_{Cu}$ ( $\text{mg kg}^{-1} \text{ week}^{-1}$ )	$HRA_{Zn}$	$HRA_{Pb}$	$HRA_{Cd}$	$HRA_{Mn}$
Cassava	AWD	0.006	0.028	0.028	0.026	0.030
	HRA (%)	0.16	0.40	112.01	371.70	7.20
Persimmon	AWD	0.045	0.073	0.073	0.153	0.045
	HRA (%)	1.30	1.04	292.34	2188.04	44.05
Sweet potato	AWD	0.013	0.026	0.026	0.051	0.098
	HRA (%)	0.37	0.37	103.15	731.15	23.35
Chinese chestnut	AWD	0.013	0.027	0.027	0.030	0.128
	HRA (%)	0.37	0.38	107.68	425.94	30.57
Maize	AWD	0.016	0.121	0.121	0.100	0.062
	HRA (%)	0.45	1.73	483.33	1428.82	14.73
Green soybean	AWD	0.005	0.019	0.019	0.010	0.055
	HRA (%)	0.14	0.27	76.69	141.38	13.20
$Rfd$	3.50*	7.00*	0.025**	0.007*	0.42*	3.50*

AWD, the average weekly dose of metal by adult ( $\text{mg kg}^{-1} \text{ week}^{-1}$ ). HRA, health risk index,  $HRA < 100$  meant the exposed population is assumed to be safe.  $Rfd$ , the permitted weekly tolerable upper intake levels ( $\text{mg kg}^{-1} \text{ week}^{-1}$ ). \*Data from Deng et al. 2013; \*\*Data from Lei et al. 2015



while the corresponding HRA values for Cu, Zn, and Mn were minor compared to those for Cd and Pb. The HRA values for all investigated heavy metals in this study were greater than those reported by Khan et al. (2013) in Swat, by Lei et al. (2015) in Hunan Province of China, by Nawab et al. (2016) in northern Pakistan, and Singh et al. (2010) in the dry tropical areas of India. High background values in the soil may be another factor contributing to the high concentrations of heavy metals in foodstuffs.

The concentration ranges of Cd were 2.33–5.37 mg kg<sup>-1</sup> in the sampled crops, which were much higher than the threshold values of the national food standard (0.02–0.5 mg kg<sup>-1</sup>). Excess Cd in the body is harmful to the kidney, liver, lungs, brain, prostate, testes, muscles, heart, immune system, and central nervous system (Ghosha and Indra 2018). The residence time of Cd in the body is as long as 20–30 a, therefore, the health risk from Cd is a long-term problem. Furthermore, Pb exposure has been listed as one of the 67 important risk factors contributing to the global burden of disease (Lim et al. 2012). High exposure to Pb can seriously damage the kidney, liver, central nervous system, and hematologic systems (Cao et al. 2014; McClure et al. 2016). Based on the diets of urban and rural residents in Guangxi Province, the intake of Pb per one BW was as high as 370 µg kg<sup>-1</sup> in 1 week via consumption of these crops. Although the corresponding data on children are currently unavailable, we have reasons to think children may suffer much greater health risks based on previous studies in other areas (Lim et al. 2012; Cao et al. 2014; McClure et al. 2016) due to their vulnerability to heavy metal contamination, their frequent hand-to-mouth behavior, their higher intake rates, and their undeveloped neural system (Cao et al. 2014).

The high concentrations of Cd and Pb in foodstuffs may be due to mafic and ultramafic rocks and open dumping of mining wastes (Nawab et al. 2015). In this area, the soil background values for Cd and Pb were as high as 0.267 and 19.5 mg kg<sup>-1</sup>, respectively (China's Environmental Protection Agency 1990; Environmental Research Institution of Guangxi 1992), which have much higher values than those in other areas in China. Additionally, acidic precipitation is a vital factor for the activation, erosion, and transformation of heavy metals (McCauley et al. 2009; Liu et al. 2010). Acid rain is a serious environmental issue in Guangxi Province. Data from ten monitoring stations showed that the annual average pH values of precipitation were < 5.6, with the lowest values even reaching 2.32–3.15, and the frequency of acid rain was approximately 36.3–88.4% (Cheng et al. 2010). Thus, acid rain could undoubtedly accelerate the activation, erosion, and leaching of heavy metals, which could ultimately induce greater ecological security issues. Therefore, it would be interesting to study the migration and transformation of these heavy metals under acidic precipitation conditions in the future. Furthermore, this study area is part of a karstic landscape, which is characterized by distinctive landforms

related to dissolution and dominantly subsurface drainage (Liang and Xu 2014; Gutiérrez et al. 2014). The direct connection between the surface and the underlying high-permeability aquifers makes karst aquifers extremely vulnerable to pollution (Gutiérrez et al. 2014). This characteristic means heavy metals within this system are more prone to migration and diffusion and more likely to cause extensive pollution. Severe pollution with heavy metals in agricultural soils substantially increases the threat to sustainable agricultural and regional development. Therefore, soil amelioration, selection of crop species selection that resists the bioaccumulation of heavy metals, and field management technology, as well as other measures, should be implemented immediately to mitigate the health risks.

## Conclusion

The concentrations of heavy metals (Cu, Zn, Pb, Cd, and Mn) in agricultural soils and six representative crops, as well as their contamination degrees and their ecological and human health risks, were studied in the Pingle Mn mine in China. The heavy metal concentrations in the soils generally exhibited the following order: Mn > Pb > Zn > Cu > Cd; the Mn and Cd concentrations in the agricultural soils exceeded those of the national standard (MEP 1995) by 15.38–38.41 and 21.64–28.75 times, respectively. Assessments with the indices  $P_i$  and  $P_{com}$  showed that the sampled soils were severely polluted by these heavy metals, especially Cd and Pb. The BAF values for Cd were much higher than those for the other four heavy metals. The analysis with the potential ecological risk index further demonstrated that the soils were all at extremely high risk of heavy metal pollution. The concentrations of these heavy metals in the crops had the same order as in the soils. Ecological risk assessments with  $P_i$  and  $P_{com}$  also showed that the crops were severely polluted by Pb and Cd. The HRA values for the adults living in mining-impacted areas were high. Our study strongly recommends that heavy metal contamination in agricultural soils and crops grown around mining-affected areas should be managed to mitigate the health risks.

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