

# Heavy metal contamination in soils and vegetables and health risk assessment of inhabitants in Daye, China

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## Abstract

**Objective:** This study was performed to evaluate the state of heavy metal contamination in soil and vegetables and assess the health risk of inhabitants in the mine-affected area and area far from the mine (reference area) in Daye, China.

**Methods:** The heavy metal concentrations in soil and vegetable samples were detected by inductively coupled plasma mass spectrometry. Residents' exposure parameters were obtained through a questionnaire survey. A health risk assessment model recommended by the United States Environmental Protection Agency was used to evaluate the residents' risk of oral exposure.

**Results:** The copper, lead, cadmium, and arsenic concentrations in soil and in vegetables were higher in the mine-affected area than in the reference area. The health risk of residents in the reference area was within the acceptable range (hazard index < 1, carcinogen risk < 10<sup>-4</sup>). In the contaminated area, however, the mean hazard index was 2.25 for children and 3.00 for adults, and the mean carcinogen risk was 4.749 × 10<sup>-4</sup> for children and 0.587 × 10<sup>-4</sup> for adults.

**Conclusions:** Potential health risks exist for inhabitants near the mine area. Cadmium and arsenic should be paid more attention as risk sources.

## Keywords

Heavy metal, health risk assessment, soil, vegetable, Daye, hazard index, carcinogen risk

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## Introduction

The smelting process in metal mines can produce large amounts of waste, resulting in high accumulation of heavy metals in the soil and in the river and underground water surrounding the mining area.<sup>1</sup> Similarly, the levels of heavy metal contamination in the soil, air, river, and crops from mining-affected areas are reportedly higher than those in non-mining areas in many countries including China,<sup>2,3</sup> the United States,<sup>4</sup> and South Korea.<sup>5</sup> In addition to causing environmental damage, heavy metals in soil, air, and other media in mining-affected areas enter plants by absorption through the vegetable roots and dust on leaves.<sup>6,7</sup> Ingestion of heavy metals can upset the body chemistry, especially because these metals do not undergo decomposition within the body and have a high affinity for many body systems.<sup>8</sup> Increasing evidence indicates that oral exposure is the most important way for heavy metals in the environment to enter the body; contaminated soil can result in heavy metal translocation into the food chain, and high consumption of contaminated vegetables can pose a serious risk to the human body.<sup>9,10</sup>

Studies of the accumulation and origin of heavy metals in soil<sup>11</sup> and the potential ecological hazards associated with this process<sup>12,13</sup> have revealed an increasing trend. This intensive research has attracted attention to the effects of heavy metals on human health, especially in infants and children.<sup>14–17</sup> Among many toxic elements, lead, arsenic, and cadmium are considered to be potential carcinogens and are associated with the development of several diseases, especially cardiovascular, kidney, nervous system, blood, and bone diseases.<sup>18</sup> Although copper is an essential element for humans, a high concentration in soil and vegetables leads to human toxicity.<sup>19</sup>

China is one of few countries with a wide range of mineral resources and a high degree of self-sufficiency in minerals. Mining activities alone have generated about 1,500,000 hectare (ha) of wasteland in China,<sup>20</sup> and this figure is increasing at a rate of 46,700 ha per year.<sup>21</sup> Daye is one of the most important metal ore-concentrating areas in China and has a 3000-year metallurgical mining history since the Shang Dynasty. Heavy metal contamination of soil, surface water, and underground water in Daye caused by long-term mining activities has attracted extensive attention;<sup>22,23</sup> however, research of the health risks to residents is still in the initial stage. Previous research of such health risks focused more on the contamination level of environmental media than on the risk of heavy metal exposure. This is because such studies more commonly used the exposure parameters from guidelines published by the United States Environmental Protection Agency (USEPA) or data from literature references for residents,<sup>24</sup> which might not have been completely in line with the actual situation of the local population.

The aim of the present study was to estimate the environmental impact of heavy metals in farmland soil and vegetables sampled both from the mine-affected area and area away from mining activities (reference area) in Daye, China. The transformation characteristics of heavy metals from soil to vegetables in the reference and contaminated areas were studied in detail. At the initial stage, a questionnaire survey was administered to the residents in Daye to explore the exposure parameters of the local population specifically associated with vegetable consumption. The overriding concern was that the health risk of local inhabitants from the reference and contaminated areas are based on the actual inhabitant exposure parameters and heavy metal concentrations. The research techniques and results

of this study can provide a reference for mining pollution control and health risk assessment in similar area around the world.

## Materials and methods

### Ethical approval

This study protocol was approved by a professional expert group (Expert Group on the Special Investigation Project of Environment and Health in Daye, Hubei Province) that was designated by the Hubei Provincial Center for Disease Control and Prevention and the Hubei Academy of Environmental Sciences.

### Study site description

Daye city lies to the southeast of Hubei province in central China. The present

study included two areas, namely the mine-affected area (i.e., contaminated area) in the northeast of Daye city and an area away from the mining activities (i.e., reference area) in the west of Daye city. (Figure 1). In total, 32 sampling sites are randomly distributed in farmland in the selected villages, with 2 sampling sites in every village. The reference area includes 14 sampling sites in 7 villages: Shangwang (sampling sites 1 and 2), Zhushan (sampling sites 3 and 4), Fandao (sampling sites 5 and 8), Wuduan (sampling sites 6 and 7), Mingshan (sampling sites 9 and 10), Shangzhuang (sampling sites 11 and 12), and Yangqiao (sampling sites 13 and 14) (Figure 1). The contaminated area includes 18 sampling sites in 9 villages: Guantang (sampling sites 15 and 16), Changle (sampling sites 17 and 18), Chunguang (sampling sites 19 and 20), Huajing (sampling sites 21 and 22), Wangjizhuang (sampling sites 23 and 24), Luojiao (sampling sites 25 and 26), Luodiao (sampling sites 27 and 28), Weiwang (sampling sites 29 and 30), and Yangqiao (sampling sites 31 and 32).

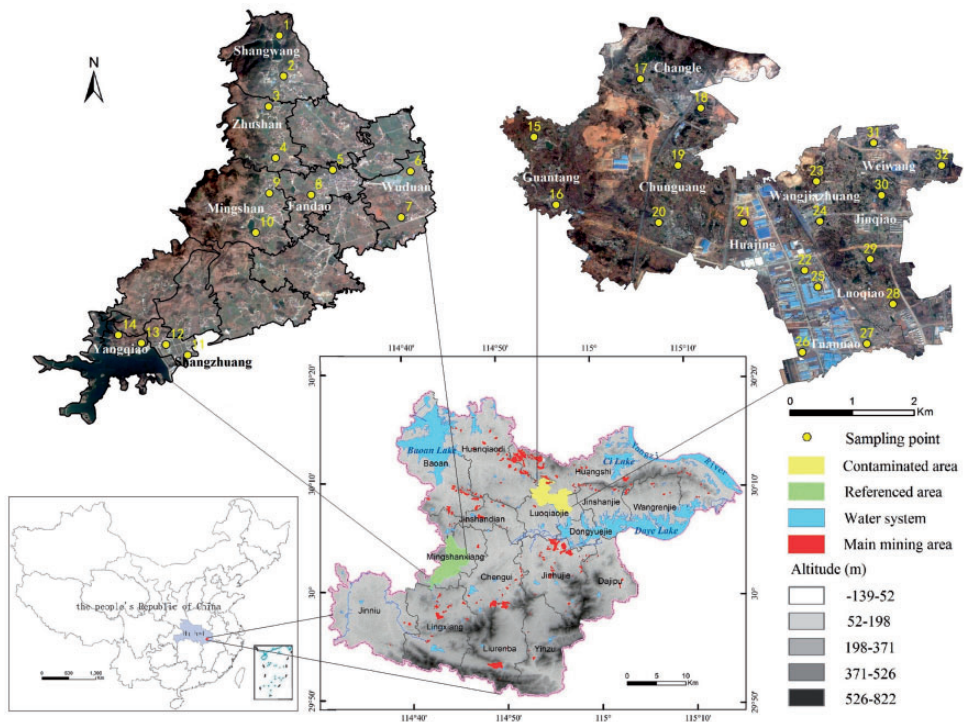


Figure 1. Distribution of sampling points.

and 22), Wangjiazhuang (sampling sites 23 and 24), Luoqiao (sampling sites 25 and 28), Tuannao (sampling sites 26 and 27), Jinqiao (sampling sites 29 and 30), and Weiwang (sampling sites 31 and 32). The villages and sampling sites in the two study areas were selected with consideration of the inhabitants' settlement and distribution. The reference area has socioeconomic characteristics similar to those of the contaminated area. The geographical locations of the sampling points are shown in Figure 1.

### Sampling and pretreatment

**Sample collection.** In total, 32 surface soil samples (1 kg each) were collected from the top 0- to 20-cm layer at the sampling sites with reference to HJ/166-2004 (China, 2004).<sup>25</sup> This is because most crops' roots are present in the top 20 cm of soil,<sup>26</sup> which is regularly turned over by local farmers. At each sampling site, one subsample was collected within a distance of 5 m surrounding the site to form a composite sample. A total of 1 kg of soil was selected from the mixed samples by the quartile method, stored in a polyethylene zip-bag, and immediately transported to the laboratory. In total, 204 vegetable samples were collected directly from the land from which the soil samples were collected, including 7 kinds of vegetables: cowpea (*Vigna unguiculata* (Linn.) Walp), water spinach (*Ipomoea aquatica* Forsk), amaranth (*Amaranthus tricolor* L), sweet potato leaves (*Ipomoea batatas* Lam), tomato (*Lycopersicon esculentum* Miller), eggplant (*Solanum melongena* Linn), and pepper (*Capsicum annum* Linn. var. *grossum* (L.) Senti). The specific sampling quantities were 21 cowpea, 21 water spinach, 6 amaranth, 24 sweet potato leaves, 18 tomato, 15 eggplant, and 18 pepper samples in the contaminated area and 15 cowpea, 12 water spinach, 6 amaranth, 12 sweet potato

leaves, 6 tomato, 15 eggplant, and 15 pepper samples in the reference area.

**Sample pretreatment and analysis.** The soil samples were air-dried at room temperature, then pulverized and sieved through a 1-mm stainless steel mesh to obtain a fine homogenous powder. The samples were stored in polyethylene bags at 4°C prior to analysis. The edible portions of the vegetables were thoroughly washed with tap water and deionized. After being primarily air-dried at room temperature and milled by a ceramic-coated grinder, the vegetable samples were frozen at 18°C until chemical analysis.

The soil pH was measured in ultrapure water (1.0:2.5 soil:solution ratio, dry w/v). The soil samples were digested according to "Acid Digestion of Sediments, Sludges and Soils" (USEPA Method 3050b).<sup>27</sup> The vegetable samples were digested according to "Determination of Lead in Food, Arsenic, Iron, Calcium, Zinc, Aluminium, Sodium, Magnesium, Boron, Manganese, Copper, Barium, Strontium, Titanium, Tin, Cadmium, Chromium and Vanadium Content by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) Method" (DB53/T 288-2009).<sup>28</sup>

The concentrations of copper (Cu), lead (Pb), arsenic (As), and cadmium (Cd) in vegetables and soil were measured by inductively coupled plasma mass spectrometry (NexION 350x; PerkinElmer, Waltham, MA, USA). Each batch of testing blank and parallel samples were analyzed simultaneously. The relative deviation between the parallel samples was controlled within 10%.

### Population survey

The exposure parameters from the Exposure Factors Handbook,<sup>29</sup> such as the inhabitants' weight and vegetable intake, are not suitable for calculation of the residents' health risk. Thus, the

questionnaire regarding the exposure parameters of the Daye area mainly covered the problems related to calculation of dietary route exposure, including sex, age, height, weight, and other individual characteristics as well as food types, consumption, frequency, time, and other dietary exposure parameters. Four hundred questionnaires were issued, and 389 valid responses were obtained. After data input, PASW Statistics, version 18.0 (SPSS Inc., Chicago, IL, USA) was used for statistical analysis.

### Data analysis

**Transfer factor.** The transfer factor (TF) of heavy metals was calculated by Equation (1), which represents the potential capability of heavy metal transmission from soil to the edible parts of a vegetable:<sup>30</sup>

$$TF = \frac{C_{veg}}{C_{soil}} \quad (1)$$

where  $C_{veg}$  is the heavy metal concentration (on a fresh weight basis) in the edible parts of the vegetable, and  $C_{soil}$  is the total heavy metal concentration (on a dry weight basis) in the corresponding soil where the vegetable was grown.

**Health risk assessment and exposure parameters.** The health risk assessment model was provided by USEPA (2007) and allowed for quantitative assessment of the human health risks of Cu, Pb, As, and Cd. The mean daily dose (ADD) was used to measure the amount of daily heavy metal intake associated with local inhabitants' vegetable consumption. The formula for calculating ADD for a single heavy metal through the diet is shown in Equation (2):<sup>31</sup>

$$ADD(\text{mg}/\text{kg} \cdot \text{d}) = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (2)$$

where  $C$  (mg/kg on a fresh weight basis) is the food contaminant concentration,  $IR$  (kg/person/d) is the daily vegetable consumption,  $EF$  (365 days/year) is the exposure frequency,  $ED$  is the exposure duration,  $BW$  (kg) is the body weight, and  $AT$  is the mean exposure time for noncarcinogens.

The hazard index (HI) for the noncarcinogen risk of a variety of heavy metals was calculated using Equation (3):<sup>32</sup>

$$HI = \sum_{i=1}^n (ADD_i \div RfD_i) \quad (3)$$

where  $i$  is the heavy metal species and  $n$  is the type of heavy metal. Cu is a noncarcinogen and As, Cd, and Pb are carcinogens. The carcinogen risk (CR) of heavy metal intake through vegetable consumption was calculated using Equation (4):

$$CR = \sum_{i=1}^n (ADD_i \times SF_i) \quad (4)$$

The  $IR$  was obtained through the questionnaire investigation and statistics presented in "China dietary and nutrition survey and tracking research."<sup>33</sup> The  $IR$  for children and adults was 153.84 and 373.98 g/d, respectively. The  $BW$  for children and adults was 18.77 and 61.00 kg, respectively. For the noncarcinogen risk,  $AT$  is the product of  $EF$  and  $ED$ ;  $ED$  for children and adults was 4.54 and 42.61 years, respectively, which are the mean ages according to a local population survey. For the  $CR$ ,  $AT$  is the whole life expectancy (74.80 years for children and 64.95 years for adults).

According to the USEPA Integrated Risk Information System statistics, the recommended reference dose values for oral intake of As, Cd, Pb, and Cu are 0.0003, 0.001, 0.004, and 0.04 mg/kg per day,



respectively. The carcinogen slope factor for As, Cd, and Pb are 1.5, 0.38, and 0.0085 mg/kg per day, respectively.<sup>34,35</sup>

The HI was used to characterize inhabitants' noncarcinogen risk of consuming vegetables. If the HI is <1, the exposed population is unlikely to experience obvious adverse effects. The acceptable risk range for CR is  $10^{-4}$  according to the USEPA.

## Results and Discussion

### Heavy metal concentrations in farmland soil

Table 1 summarizes the heavy metal concentrations in farmland soil in the study area. The soil from the contaminated area and reference area was slightly acidic, with a pH of <7. There were significant variations of the heavy metals in the soil samples from both the contaminated and reference areas in the following order: Cu > Pb > As > Cd ( $p < 0.05$  for all).

In the reference area, the maximum values of all heavy metals in soil were below the maximum allowable level (MAL) according to the Environment Quality Standard for Soils regulated by the Environmental Protecting Administration of China (EPAC).<sup>36</sup> The concentrations of Pb, As, and Cd were all below those in the natural background soil in Daye.<sup>37</sup> However, the concentration of Cu in soil from the reference area was

slightly higher than the soil background value, which may be related to the abundance of Cu ore and the increasingly intensive mining activities. Thus, the soil from the reference area has not been contaminated but may still be affected by mining activities.

All measured heavy metal concentrations in the surface soil samples from the contaminated area were significantly higher than those in the reference area ( $p < 0.05$ ) and in the background soil of Daye. The mean Cu, Pb, As, and Cd concentrations in soil from the contaminated area were 4.53, 2.57, 2.50, and 10.16 times higher than those in the reference area, respectively ( $p = 0.001, 0.001, 0.000, \text{ and } 0.000$ , respectively). In addition, the mean concentration of Cu and Cd in soil from the contaminated area reached 128.48 and 55.16 mg/kg, respectively, which was found to exceed the MAL. However, the concentrations of As and Cu were within the safe limits set by the EPAC. These results suggest that the soil in the contaminated area has been greatly affected, even contaminated, by mining and smelting activities and may not be suitable for crop production. Additionally, Cd was found to be the primary contaminant.

Moreover, the level of heavy metal contamination differed in all soil sample types, which was reflected by the maximum value, minimum value, and standard deviation of each element. This shows that there are

**Table 1.** Heavy metal concentrations in soil (mg/kg)

Heavy metal	Contaminated soils (n = 18)		Reference soils (n = 14)		MAL	Background value
	Range	Mean	Range	Mean		
Cu	32.35–321.99	128.48 ± 101.36	17.33–47.04	28.33 ± 11.31	50	25.63
As	10.16–56.42	20.09 ± 11.60	4.74–10.48	8.01 ± 2.14	30	16.58
Cd	0.20–2.45	1.38 ± 0.81	0.03–0.22	0.13 ± 0.05	0.3	0.175
Pb	19.54–87.56	55.16 ± 22.40	17.88–24.83	21.41 ± 2.26	250	43.35

Concentrations are presented in mg/kg. The mean is presented with the standard deviation. Cu, copper; As, arsenic; Cd, cadmium; Pb, lead; MAL, maximum allowable level.

differences in the degree of soil contamination in different villages even within the same region, which may be mainly related to the specific terrain, soil types, and farming activities.

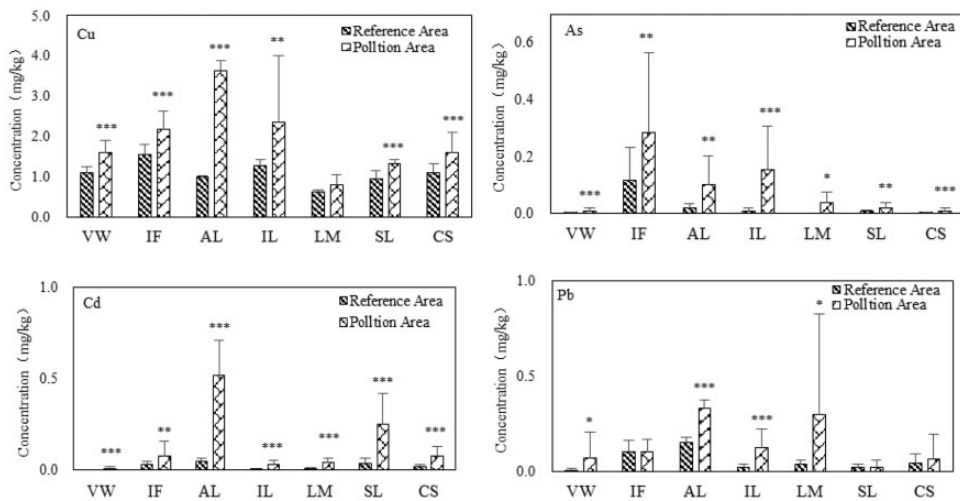
### Assessment of heavy metal contamination in vegetables

The heavy metal concentrations in the edible portions of vegetables grown in contaminated soils were compared with those grown in soil from the reference area. The seven vegetables examined were cowpea, water spinach, amaranth, sweet potato leaves, tomato, eggplant, and pepper.

The mean heavy metal concentrations in vegetables from both the contaminated and reference areas decreased in the order of  $\text{Cu} > \text{Pb} > \text{Cd} > \text{As}$ , which is not completely consistent with the above-mentioned order of heavy metal concentrations in soil (i.e.,  $\text{Cu} > \text{Pb} > \text{As} > \text{Cd}$ ). The concentrations of Cu, Pb, Cd, and As in vegetables from the contaminated area ranged from 0.463 to 6.672, 0.000 to 1.474, 0.000 to 0.711, and 0.000 to 0.942 mg/kg, respectively. The

concentrations of Cu, Pb, Cd, and As in vegetables from the reference area ranged from 0.618 to 1.548, 0.006 to 0.153, 0.000 to 0.048, and 0.000 to 0.095 mg/kg, respectively, which were significantly lower than those in vegetables from the contaminated area ( $p < 0.05$  for all) (Figure 2). This result is in agreement with the findings that plants grown in topsoil contaminated with heavy metals are more likely to be contaminated.<sup>38</sup>

As shown in Figure 2, the heavy metal concentrations in different vegetables are quite different. Overall, leafy vegetables such as spinach, amaranth, and potato leaf seemed to accumulate more heavy metals in their edible parts than other types of vegetables such as cowpeas and pepper. This is consistent with the conclusions of Yang et al.<sup>39</sup> A study by Yana et al.<sup>40</sup> showed that heavy metals in the leaves of vegetables do not only come from the soil but are also greatly affected by contamination from smelting waste gases, whereas heavy metals in the fruits and roots of vegetables mainly come from



**Figure 2.** Heavy metal concentrations in the edible parts of vegetables. The error bars indicate the standard deviation, and the asterisks indicate significant differences in heavy metal concentrations between vegetable grown in mine-affected and reference soils. \* $P < 0.1$ , \*\* $P < 0.05$ , and \*\*\* $P < 0.001$ .

soil. In addition, the heavy metal concentrations in vegetables are related to the different growth stages of vegetables in the same season because the ability of vegetables to enrich heavy metals varies with the growth stages.<sup>41</sup> This may be the reason why heavy metals in the edible portions of leaf vegetables is much higher.

To evaluate the heavy metal contamination of vegetables grown in soil, the heavy metal concentrations in this study were compared with the MAL set by China, the Food and Agriculture Organization (FAO),<sup>42</sup> and the European Communities (EC)<sup>43</sup> (Table 2). Cu is an essential element for the human body and is therefore not regulated by Chinese food standards. However, the FAO (2011) regulates the maximum Cu level at 40 mg/kg (based on fresh weight), while the EC (2006) limits the Cu level at 20 mg/kg (based on fresh weight).

The mean concentrations of Cu, Pb, Cd, and As in vegetables from contaminated soil in this study were lower than the MAL set by China, the FAO, and the EC. However, not all vegetables are safe to enter the food chain; the degree of contamination greatly differs among different kinds of vegetables. Contamination in amaranth species was the most severe among the sampled vegetables. Only the concentrations of Cd and Pb in amaranth species were 2.6 and 1.1 times higher, respectively, than the MAL

set by China,<sup>44</sup> the FAO, and the EC. Additionally, the concentrations of Cd in pepper and eggplant were 5.0 and 1.6 times higher, respectively, than the MAL. The concentrations of Cu, Pb, and As for both pepper and eggplant did not exceed the MAL. However, all four heavy metals in cowpeas, spinach, and sweet potato leaves were below the MAL.

### Transfer of heavy metals from soil to vegetables

The TF can be used to assess the potential ability of heavy metals to move from soil to plant tissues. The TF of heavy metals from soil to vegetables in the contaminated and reference areas is shown in Figure 3. The mean TF value for Cu, As, Cd, and Pb from soil to vegetables in the contaminated area reached 0.021, 0.007, 0.095, and 0.003, respectively. The mean TF value for Cu, As, Cd, and Pb from soil to vegetables in the reference area reached 0.043, 0.003, 0.139, and 0.003, respectively. The TF of Cu and Cd from soil to vegetables was obviously lower in the contaminated than reference area. The TF of the four heavy metals in the two regions both showed an order of Cd > Cu > As > Pb, which is consistent with the conclusions reported by Liu et al.<sup>45</sup> and Cai et al.<sup>46</sup> In addition, the order was not the same as in the report by Choudhury,<sup>47</sup> which is more likely related

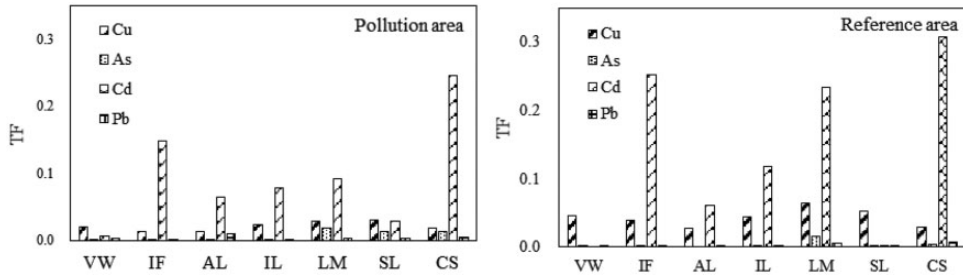
**Table 2.** Recommended maximum levels and heavy metal concentrations for vegetables

Standard	Cu	As	Cd	Pb
FAO, 2011	40	0.05 (Inorganic)	0.2 leafy; 0.05 others	0.3
EC, 2006	20	NR	0.2 leafy; 0.05 others	0.3
CMH, 2012	NR	0.5 (Total)	0.2 leafy; 0.05 others	0.3 leafy; 0.1 others
Contaminated area mean	1.930 ± 0.494	0.123 ± 0.087	0.143 ± 0.079	0.145 ± 0.148
Reference area mean	1.087 ± 0.151	0.022 ± 0.022	0.020 ± 0.011	0.055 ± 0.029

Levels are presented in (mg/kg, fresh weight). Data are presented as mean ± standard deviation.

Cu, copper; As, arsenic; Cd, cadmium; Pb, lead; FAO, Food and Agriculture Organization; EC, European Union Standards; CMH, Chinese Ministry of Health; NR, Not recommended.





**Figure 3.** Transfer factor of heavy metals from soil to vegetables in contaminated area and reference area.

to the physical and chemical properties of the soil and the type of plant sampled.

The TF of the different vegetables varied greatly, showing an order of amaranth > spinach > eggplant > pepper > tomato > sweet potato leaf > cowpea. The TF of leafy vegetables such as spinach and amaranth was higher than that of other vegetables such as eggplant, pepper, and tomato, which is in agreement with the findings of Xu et al.<sup>48</sup>

Additionally, the contamination of soil by Cd was the most severe, so the accumulation of Cd from soil to vegetables should be paid more attention (although the mean concentration of Cd in vegetables was lower than the MAL recommended by Chinese Ministry of Health).

### *Health risk of local inhabitants by contaminated vegetables*

The noncarcinogen risk values of children and adults in the contaminated and reference areas were calculated according to the consumption of vegetables and the heavy metal concentrations in vegetables (Table 3). The total HI of the four heavy metals for children and adults in the contaminated area ranged from 0.72 to 7.70 and 0.54 to 5.76, respectively. The total HI of children and adults in the reference area ranged from 0.28 to 1.66 and 0.19 to 1.20, respectively. The total HI of residents

in the contaminated area, including both adults and children, were higher than those the reference area. The total HI calculated in the reference area was <1 if the local residents consumed each vegetable at the same frequency, suggesting that the noncarcinogen risk of heavy metal exposure through vegetables was generally safe. Remarkably, residents in the contaminated area were exposed to a serious noncarcinogenic health risk.

In the contaminated area, the HI values for Cu and Pb in both children and adults were <1, indicating that the noncarcinogen risk caused by only oral Cu or Pb was not obvious. Notably, the total noncarcinogen risk had a mean of 77% for As and Cd in both children and adults in the contaminated area. Similarly, Lim et al.<sup>49</sup> reported a high risk for Cd and As.

The mean noncarcinogen risk contribution rate of Cu, As, Cd, and Pb for residents in the reference area was 30%, 27%, 22%, and 15%, respectively. The situation in the reference area was greatly different from that in the contaminated area, mainly due to the different heavy metal concentrations in vegetables between the two different regions.

The CR values of adults and children in the contaminated area ranged from  $5.49 \times 10^{-5}$  to  $1.25 \times 10^{-3}$  and from  $6.80 \times 10^{-6}$  to  $1.55 \times 10^{-4}$ , respectively, and those of adults and children in the reference area

**Table 3.** Hazard risk index of vegetable intake

Vegetable	Age	Contaminated area					Reference area				
		Cu	As	Cd	Pb	Total	Cu	As	Cd	Pb	Total
VW	Children	0.33	0.18	0.07	0.14	0.72	0.23	0.02	0.00	0.01	0.72
	Adults	0.25	0.13	0.05	0.11	0.54	0.17	0.01	0.00	0.01	0.19
IF	Children	0.45	2.96	0.63	0.21	4.26	0.32	0.88	0.26	0.21	1.66
	Adults	0.34	2.22	0.47	0.16	3.18	0.24	0.66	0.19	0.16	1.24
AL	Children	0.74	2.06	4.22	0.67	7.70	0.21	0.26	0.39	0.31	1.17
	Adults	0.56	1.54	3.16	0.50	5.76	0.15	0.19	0.30	0.24	0.88
IL	Children	0.48	2.12	0.22	0.26	3.09	0.26	0.13	0.00	0.04	0.44
	Adults	0.36	1.59	0.17	0.19	2.31	0.20	0.10	0.00	0.03	0.33
LM	Children	0.16	0.25	0.35	0.61	1.37	0.13	0.00	0.07	0.08	0.28
	Adults	0.12	0.19	0.26	0.46	1.03	0.09	0.00	0.06	0.06	0.21
SL	Children	0.27	0.27	2.07	0.05	2.66	0.20	0.10	0.29	0.04	0.63
	Adults	0.20	0.20	1.55	0.04	1.99	0.15	0.08	0.22	0.03	0.47
CS	Children	0.33	0.11	0.65	0.13	1.22	0.22	0.04	0.13	0.09	0.48
	Adults	0.25	0.09	0.49	0.10	0.91	0.17	0.03	0.10	0.07	0.36
Mean	Children	0.40	1.14	1.17	0.30	3.00	0.22	0.20	0.16	0.11	0.77
	Adults	0.30	0.85	0.88	0.22	2.25	0.17	0.15	0.12	0.08	0.53

Cu, copper; As, arsenic; Cd, cadmium; Pb, lead; VW, *Vigna unguiculata* (Linn.) Walp (cowpea); IF, *Ipomoea aquatica* Forsk (water spinach); AL, *Amaranthus tricolor* L (amaranth); IL, *Ipomoea batatas* Lam (sweet potato leaves); LM, *Lycopersicon esculentum* Miller (tomato); SL, *Solanum melongena* Linn (eggplant); CS, *Capsicum annuum* Linn. var. *grossum* (L.) Sendt (pepper).

**Table 4.** Carcinogen risk value of vegetable intake ( $10^{-4}$ )

Vegetable	Age	Contaminated area				Reference area			
		As	Cd	Pb	Total	As	Cd	Pb	Total
VW	Children	0.049	0.016	0.003	0.068	0.004	0.000	0.000	0.005
	Adults	0.396	0.129	0.024	0.549	0.035	0.000	0.002	0.037
IF	Children	0.809	0.146	0.004	0.960	0.239	0.060	0.004	0.304
	Adults	6.542	1.183	0.035	7.761	1.936	0.484	0.035	2.455
AL	Children	0.563	0.974	0.014	1.551	0.070	0.091	0.006	0.167
	Adults	4.553	7.875	0.112	12.54	0.563	0.736	0.052	1.351
IL	Children	0.580	0.051	0.005	0.636	0.035	0.000	0.001	0.037
	Adults	4.688	0.413	0.043	5.144	0.287	0.002	0.007	0.296
LM	Children	0.069	0.080	0.013	0.162	0.000	0.017	0.002	0.019
	Adults	0.561	0.644	0.102	1.307	0.000	0.139	0.014	0.152
SL	Children	0.073	0.477	0.001	0.551	0.028	0.067	0.001	0.096
	Adults	0.593	3.856	0.008	4.457	0.229	0.541	0.007	0.776
CS	Children	0.031	0.150	0.003	0.183	0.010	0.030	0.002	0.042
	Adults	0.251	1.209	0.022	1.482	0.082	0.240	0.015	0.337
Mean	Children	0.311	0.271	0.006	0.587	0.055	0.038	0.002	0.096
	Adults	2.512	2.187	0.049	4.749	0.447	0.306	0.019	0.772

Cu, copper; As, arsenic; Cd, cadmium; Pb, lead; VW, *Vigna unguiculata* (Linn.) Walp (cowpea); IF, *Ipomoea aquatica* Forsk (water spinach); AL, *Amaranthus tricolor* L (amaranth); IL, *Ipomoea batatas* Lam (sweet potato leaves); LM, *Lycopersicon esculentum* Miller (tomato); SL, *Solanum melongena* Linn (eggplant); CS, *Capsicum annuum* Linn. var. *grossum* (L.) Sendt (pepper).

ranged from  $3.70 \times 10^{-6}$  to  $2.45 \times 10^{-4}$  and from  $5.00 \times 10^{-7}$  to  $1.67 \times 10^{-6}$ , respectively. The CR for residents in the contaminated area was significantly higher than that in the reference region ( $p=0.03$ ). Additionally, the CR of heavy metals for children was significantly higher than that for adults ( $p=0.03$ ), which is consistent with previous studies.<sup>50</sup> Please click here to enter your response. However, the result that the CR was higher for adults than children differed from the findings reported by Lanhua et al.,<sup>51</sup> who may have artificially expanded the continuous exposure time of children. The exposure time was defined as the mean age of the residents, mainly taking into account the changes in the future residence and living environment of residents with different ages.

The CR contribution rate of different heavy metals for residents in both the contaminated and reference areas showed the order of  $As > Cd > Pb$  (Table 4). The CR for As was close to that of Cd, and both were the main source of the CR. As had a low contamination level in vegetables but a high CR for residents; this was mainly due to the high carcinogen slope factor of As determined by the biological toxicity and carcinogenicity.

## Conclusion

The concentrations of four heavy metals near the mine were significantly higher than those in the reference area, indicating that the soil in the contaminated area is greatly affected by mining and smelting activities. Contamination of soil with Cd was the most serious, with a mean concentration of 4.6 times higher than the maximum permitted level set by the China Ministry of Environmental Protection. The mean concentrations of Cu, Pb, Cd, and As in different parts of vegetables in contaminated soils were lower than the MAL set by China, the FAO, and the EC,

but the Cd and Pb concentrations in some vegetables exceeded the MAL. The heavy metal concentrations in vegetables are largely dependent on the ability to accumulate heavy metals. The TF of the four heavy metals showed the order of  $Cd > Cu > As > Pb$ . From the viewpoint of the soil concentration and enrichment ability, Cd should be paid more attention. Residents in the reference area seem to be safe. However, residents in the contaminated area are exposed to a serious health risk with an mean of a 77% contribution from As and Cd. The total HI of the four heavy metals in children and adults in the contaminated area ranged from 0.72 to 7.70 and 0.54 to 5.76, respectively. The CR values for adults and children in the contaminated area ranged from  $5.49 \times 10^{-5}$  to  $1.25 \times 10^{-3}$  and from  $6.80 \times 10^{-6}$  to  $1.55 \times 10^{-4}$ , respectively. Similarly, both Cd and As were the main source of CR.

From a health risk perspective, the environmental quality standards and food safety standards of some heavy metals, such as As, may need to be improved. The concentration of As meets the MAL in vegetables but appears to largely contribute to the human health risk because of the higher toxicity to cells or organisms. The sources and transformation of heavy metal contamination require further study to provide a basis for health risk management from the perspective of an entire mining area.

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## Declaration of conflicting interests

The authors declare that there is no conflict of interest.

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