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Heavy Metal Contamination in Soil and Soybean near the Dabaoshan Mine, South China*1

ZHUANG Ping, LI Zhi-An*2, ZOU Bi, XIA Han-Ping and WANG Gang

South China Botanical Garden, Chinese Academy of Sciences, Guangzhou 510560 (China)

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ABSTRACT

Concentrations of Pb, Cd, Cu, Zn, Cr and Ni in soybean (*Glycine max* L.) grown near the Dabaoshan Mine were investigated, and their potential risk to the health of inhabitants was estimated. In the Fandong (FD) and Zhongxin (ZX) villages, which are near the Dabaoshan mineral deposit, concentrations of Pb (0.34 mg kg^{-1} for FD), Cd (0.23 mg kg^{-1} for ZX) and Cr (1.14 and 1.75 mg kg^{-1} for FD and ZX, respectively) in the seeds of soybean exceeded the tolerance limit set by Chinese standards. The estimated daily intakes (EDIs) from consumption of soybean seeds for FD inhabitants were 0.570, 0.170, 38.550, 142.400, 1.910 and 14.530 μg d^{-1} kg^{-1} boby weight for Pb, Cd, Cu, Zn, Cr and Ni, respectively. Our results indicate that soybeans grown in the vicinity of the Dabaoshan Mine accumulate some metals, and the seeds pose a potential health risk to the local inhabitants.

Key Words: accumulation, cadmium, estimated daily intake, health risks, target hazard quotient

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INTRODUCTION

Agricultural soil contamination with heavy metals is of increasing worldwide concern because of food safety issues and potential health risks (McLaughlin *et al.*, 1999). Excessive dietary ingestion of heavy metals may lead to a number of human and animal health disorders, which can induce systemic health problems, such as decreased immunological defenses, impaired psycho-social behavior, disabilities associated with malnutrition and a high prevalence of upper gastrointestinal cancer (Türkdoğan *et al.*, 2002; Järup, 2003).

Heavy metals in soils are derived from both natural (geogenic) sources and anthropogenic contamination. The contribution of metals from anthropogenic sources in soils can be much higher than the contribution from natural sources (Nriagu and Pacyna, 1988). With rapid economic development in recent decades, numerous human activities including mining, smelting, agriculture, waste disposal, and traffic and transportation have resulted in the release of significant quantities of heavy metals to the environment (Navarro *et al.*, 2008). Heavy metal uptake from contaminated soil *via* plant roots and direct deposition of contaminants from the atmosphere or soil particles onto plant surfaces are two major contributors to heavy metal accumulation in the edible part of food crops (Pruvot *et al.*, 2006). Hence, dietary intake of heavy metals has been regarded as a main pathway of human exposure to heavy metals (Kabata-Pendias and Mukherjee, 2007).

The potential health risks through food chain contamination by heavy metals have been evaluated by the target hazard quotient (THQ), a ratio of determined dose of a pollutant to the oral reference dose (RfDo) (USEPA, 1989). In general, numerous investigations have focused on estimating the potential human health risks of vegetables (Kachenko and Singh, 2006; Hao *et al.*, 2009), rice and wheat grain (Huang *et al.*, 2008) in contaminated sites. Little attention has been paid to the assessment of health risks *via* consumption of soybean (Angelova *et al.*, 2003; Shute and Macfie, 2006), although soybean is a staple food in diets of many parts of the world. Bojinova *et al.* (1994) reported that soybean and other beans belong to a group of crops that strongly accumulate heavy metals. Therefore, the objectives of this study were to quantify the concentrations of heavy metals (Pb, Cd, Cu, Zn, Cr and Ni) in contaminated soils and different parts of the soybean plant and evaluate the potential health risks of soybean consumption for the local inhabitants around the Dabaoshan Mine.

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*2Corresponding author. E-mail: lizan@scbg.ac.cn.

MATERIALS AND METHODS

Study area

The Dabaoshan Mine (24° 31′ 37″ N, 113° 42′ 49″ E) is located in the northern region of Guangdong Province, southern China (Fig. 1). This area has a humid subtropical climate with an average annual temperature of 20.3 °C and rainfall of 1 762 mm. The minerals in the ore mainly consist of pyrite, pyrrhotite, and chalcopyrite, with minor components of sphalerite, chalcocite, galena, limonite, calaverite, and native bismuth. Since mining began in the 1970s, large quantities of mine wastes and acid mine drainage (AMD) have been drained downslope into the Chuandu and Hengshi rivers. Water from these rivers has been used to irrigate agricultural lands for food crops, which has led to severe deterioration and contamination of the surrounding environment. Certain regions and villages around the Dabaoshan Mine have been termed endemic cancer regions/villages in China, because esophageal cancers, liver cancer, and other cancers were reported frequently in humans and poultry, with a mortality rate approaching 56% in humans. Fandong and Zhongxin villages are near the Dabaoshan mineral deposit. Fandong (FD) village (24° 33′ 78″ N, 113° 43′ 94″ E) is located on the mountaintop mine area (altitude at 500 m), which is directly affected by mining activities due to a mine tailing dam 0.5 km away from this village. Zhongxin (ZX) village (24° 35′ 72″ N, 113° 39′ 94″ E), located in the northern downstream side of the Dabaoshan Mine, approximately 6–8 km away from the mine area, and is contaminated by the wastewater (the Chuandu River) of mining operation (Zhuang *et al.*, 2009).

Sampling and pre-treatment

Soybean (*Glycine max* L.) samples grown in garden soils of FD and ZX villages and corresponding soils were collected. Soil and soybean samples were composited from at least five discrete samples taken in a 10 m × 10 m block for every soybean field. Soil samples were taken with a 5-cm inner diameter stainless steel auger from the 0–15 cm soil layer, and were placed in plastic bags. Each soybean sample was separated into root, stem, leaf, pod and seed. Grain samples were washed with tap water to remove any attached particles, rinsed three times with distilled water, and oven-dried at 70 °C to constant weight. Dried samples were ground using a stainless steel grinder appropriate for heavy metals analysis. The collected soil samples were air-dried and sieved through a 2-mm polyethylene sieve to remove stones and plant roots.

Sample analysis

Soil pH was measured in suspension (soil:water ra-

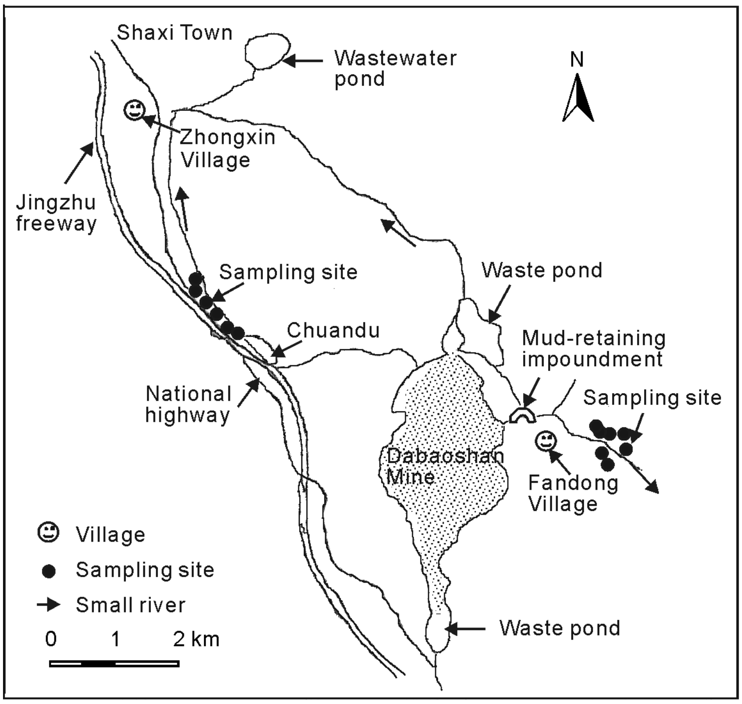

Fig. 1 Map of the Dabaoshan Mine showing the location of sampling sites at the Fandong and Zhongxin villages. Additional mining-affected areas are drained by several small rivers in the vicinity of the Dabaoshan Mine.

tio of 1:2.5). Soil samples were digested in preparation for total metal analysis using a concentrated acid mixture (HNO_3:$HClO_4$:HF ratio of 3:1:1). For soybean samples, 0.5 g of dried sample was digested with HNO_3 and $HClO_4$ in a 5:1 ratio until a transparent solution was obtained. The total concentrations of heavy metals were determined by inductively coupled plasma-atomic emission spectrometry (Perkin Elmer Optima 3300DV, USA). Lead and Cd were analyzed using graphite furnace atomic absorption spectrophotometry (GBC932AA), due to the low concentrations in soybean seed.

Quality control

Double distilled water was used throughout the study. Glassware was properly cleaned, and the reagents were of analytical grade. Certified reference soils (GBW 08303) and two reference plant materials (rice: GBW 10010 and tea: GBW 10016) were digested in a similar manner. Blank and drift standards were run after twenty determinations to maintain instrument calibration. The coefficient of variation of replicate analyses was determined for the measurements to calculate analytical precision.

Soil metal contamination assessment

The degree of metal contamination in the soils was assessed by the geoaccumulation index (I_{geo}) proposed by Muller (1969):

$$I_{\mathrm{geo}} = \log_2(C_{\mathrm{n}}/1.5B_{\mathrm{n}}) \tag{1}$$

where C_{n} represents the measured concentration of the examined metal n in the soil sample, B_{n} is the natural background concentration of the metal n, and the factor 1.5 is used because of possible variations in background values due to lithological variability. In this study, B_{n} is the background content of element n in Guangdong Province soil (CNEMC, 1990). Based on the geoaccumulation index I_{geo}, the degree of metal contamination of the samples was classified as: Grade 0 ($I_{\mathrm{geo}} \leq 0$), uncontaminated; Grade 1 ($0 \leq I_{\mathrm{geo}} \leq 1$), uncontaminated to moderately contaminated; Grade 2 ($1 \leq I_{\mathrm{geo}} \leq 2$), moderately contaminated; Grade 3 ($2 \leq I_{\mathrm{geo}} \leq 3$), moderately to heavily contaminated; Grade 4 ($3 \leq I_{\mathrm{geo}} \leq 4$), heavily contaminated; Grade 5 ($4 \leq I_{\mathrm{geo}} \leq 5$), heavily to extremely contaminated; and Grade 6 ($5 \leq I_{\mathrm{geo}} \leq 6$), extremely contaminated. Grade 6 is an open class and comprises all values of the index higher than Grade 5. The elemental concentrations in Grade 6 may be at least a hundredfold greater than the geochemical background value.

Metal bioaccumulation factor (BAF)

Heavy metal concentrations of soils and soybean were calculated on the basis of dry weight. The metal bioaccumulation factor, a ratio of crop to soil metal concentration, was calculated as follows:

$$\mathrm{BAF} = C_{\mathrm{plant}}/C_{\mathrm{soil}} \tag{2}$$

where C_{plant} and C_{soil} represent the heavy metal concentrations in the seeds of soybean and soils, respectively, on a dry weight basis.

Estimated daily intakes (EDIs) of heavy metals

The estimated daily intakes of heavy metals (Pb, Cd, Cu, Zn, Cr and Ni) depended on both the metal concentration in the seeds of soybean and the amount of soybean consumption. The EDI of metal was determined by the following equation:

$$\mathrm{EDI} = (C\times \mathrm{CR})/\mathrm{BW} \tag{3}$$

where C is the concentration of heavy metals, CR represents the average daily consumption rate of food in this region, and BW is the body weight. During the harvesting season, the local inhabitants had an average soybean consumption of 100 g d^{-1} person^{-1}. The metal intakes were compared with the tolerable daily intakes for metals recommended by the World Heath Organization (WHO, 1993).

Target hazard quotient

The health risks from consumption of soybean by the local inhabitants were assessed based on the THQ, which is a ratio of determined dose of a pollutant to a reference dose level. If the ratio is less than 1, the exposed population is unlikely to experience obvious adverse effects. The method of estimating risk using THQ was provided in the USEPA (United States Environmental Protection Agency) risk-based concentration table (USEPA, 1997, 2007), and is based on the equation:

$$\mathrm{THQ} = (C \times \mathrm{CR} \times \mathrm{EF} \times \mathrm{ED})/(\mathrm{BW} \times \mathrm{AT} \times \mathrm{RfDo}) \tag{4}$$

where exposure frequency (EF) is assumed to be 365 d year^{-1}; exposure duration (ED) is assumed to be 70 years; body weight (BW) is assumed to be 60 kg; averaging time (AT; *i.e.*, EF × ED) is assumed to be 25 550 d; and RfDo is the oral reference dose (mg kg^{-1} d^{-1}). Oral reference doses for Pb, Cd, Cu, Zn, Cr and Ni set by USEPA Integrated Risk Information System database were 0.04, 0.3, 0.004, 0.001, 1.5 and 0.02 mg

kg^{-1} d^{-1}, respectively (USEPA, 1997, 2007).

RESULTS AND DISCUSSION

Concentrations of metals in soil

The properties and total concentrations of heavy metals (Pb, Cd, Cu, Zn, Cr and Ni) in soil samples at FD and ZX villages are listed in Table I. Soils at the two villages were moderately acidic. The total concentrations of Pb, Cd, Cu, Zn, and Cr in the soils at FD were 5, 2, 5.5, 4.8 and 1.5 times, respectively, higher than those of ZX soils. The mean I_{geo} values suggested that the soils at FD village were moderately to heavily contaminated (Grade 3) by Pb, heavily contaminated (Grade 4) by Cd and Zn, and extremely contaminated (Grade 6) by Cu; and the soils at ZX village were moderately to heavily contaminated (Grade 3) by Cd and Cu.

According to Grade II of the Environmental Quality Standard for Soils of China (CERSPC, 1995), the soils at FD were primarily contaminated by Cd, Cu and Zn, with the average values over three times as much as the maximum permissible concentrations of metals in agricultural soils (Table I). A few soil samples at ZX village presented higher concentrations of the 6 heavy metals than the permissible limits of Chinese standards. Although soil Cr and Ni at the two villages did not show significantly elevated concentrations, they were still slightly higher than the background values in the soils of Guangdong Province. The concentrations of heavy metals, except for Ni, decreased with distance from the mine area.

Metal concentrations in soybean

Table II shows the average concentrations of Pb, Cd, Cu, Zn, Cr and Ni in different tissues of soybean. The leaves of soybean accumulated a higher level of Cd than other tissues of soybean, being 1.33 and 2.84 mg kg^{-1} for FD and ZX, respectively. Similar results were reported by Liu *et al.* (2005) for maize plants grown on heavily contaminated soils, which accumulated substantial amounts of Cd in their leaf tissues. The Cd concentration in the leaves of ZX soybean was two times higher than FD soybean, though the Cd contamination in the ZX soil was not severe. Exceptionally high concentrations of Zn (310.00 mg kg^{-1} for FD and 275.00 mg kg^{-1} for ZX) were observed in the leaves of soybean, followed by roots, stems, seeds, and pods. The above results were similar to the findings of Angelova *et al.* (2003) that Zn and Cd were accumulated largely in the leaves of peas and beans grown in soils near the nonferrous metal works in Bulgaria. Previous study found that the translocation of foliar absorbed Pb to fruits or seeds of plants was insignificant (Chamberlain, 1983), while other literature reported that cereal grains could accumulate substantial amounts of Pb *via* foliar absorption (Bi *et al.*, 2009). The relatively high concentration of Pb and Cd in the leaves, pods of soybean at ZX might be due to atmospheric deposition from downwind of mine area. This observation was consistent with the fact that aerial emissions can be a major contributor to contamination of leguminous crops (Pilc *et al.*, 1999; Zhang *et al.*, 2010). The results were similar to the data of Liu *et al.* (2005), who reported metal concentrations in pulses grown around the Chenzhou Pb/Zn mine in China. The Cu concentrations in seed of soybean grown at FD and ZX villages were greater than and equal to the maximum level (MHPRC, 2005), which were likely due to the severe Cu contamination in soil (Table I). The Ni concentrations in the portions of soybean at FD village

TABLE I

Selected properties of soils at Fandong (FD) and Zhongxin (ZX) villages

Soil property	FD soils ($n = 20$)		ZX soils ($n = 20$)		Background value[b]	Standard value[c]
	Mean	I_{geo}[a]	Mean	I_{geo}		
pH (H_2O)	5.80±0.45[d]	-	5.30±0.36	-	-	< 6.5
Organic matter (%)	3.30±0.50	-	3.10±0.13	-	-	
Total Pb (mg kg^{-1})	262.40±37.20	2.27±0.22	52.50±5.30	−0.05±0.12	36	250
Total Cd (mg kg^{-1})	1.04±0.24	3.50±0.32	0.51±0.14	2.45±0.34	0.06	0.3
Total Cu (mg kg^{-1})	997.00±235.00	5.24±0.39	181.20±19.80	2.82±0.14	17	50
Total Zn (mg kg^{-1})	645.00±113.00	3.16±0.27	133.70±12.50	0.91±0.11	47.3	200
Total Cr (mg kg^{-1})	84.60±38.10	0.05±0.54	55.10±5.50	−0.46±0.12	50.5	150
Total Ni (mg kg^{-1})	20.30±5.60	−0.16±0.47	24.10±0.92	0.16±0.05	14.4	40

[a] Geoaccumulation index.

[b] Metal background value in the soils of Guangdong Province.

[c] Grade II of Environmental Quality Standard for Soils (GB15618-1995) (CERSPC, 1995).

[d] Mean±standard deviation.

TABLE II

Concentrations of heavy metals in the different tissues of soybean grown at Fandong (FD) and Zhongxin (ZX) villages

Heavy metal	Maximum level		Site	Root	Stem	Leaf	Pod	Seed
	Food[a)]	Fodder[b)]						
	mg kg^{-1}			mg kg^{-1} DW[c)]				
Pb	0.2	5	FD	22.70±10.30[d)]	5.71±3.60	3.77±2.23	3.96±0.15	0.34±0.01
			ZX	14.20±2.17	13.70±3.42	12.10±1.88	6.85±0.52	0.15±0.0
Cd	0.2	0.5	FD	1.59±0.57	1.22±0.47	1.33±0.26	0.45±0.25	0.16±0.02
			ZX	1.11±0.75	0.94±0.42	2.84±1.14	0.72±0.37	0.23±0.06
Cu	20	-	FD	129.00±65.00	41.70±21.50	20.00±6.84	21.70±10.10	23.10±4.61
			ZX	47.90±28.50	19.90±2.55	16.40±2.38	14.10±1.81	16.20±0.93
Zn	100	-	FD	153.00±35.10	137.00±64.50	310.00±92.80	79.70±23.90	85.40±17.00
			ZX	46.50±32.90	38.70±10.20	275.00±85.70	55.80±11.90	57.40±7.14
Cr	1.0	10	FD	10.40±6.12	5.91±3.28	1.36±0.45	4.11±2.52	1.14±0.30
			ZX	6.37±1.42	4.78±1.09	3.31±0.27	3.78±0.32	1.75±0.16
Ni	-	-	FD	12.80±6.50	7.16±4.16	5.39±2.86	12.10±8.13	8.72±1.49
			ZX	33.50±15.40	15.80±1.28	6.67±2.09	12.90±1.93	11.90±4.84

[a)]The maximum levels of contaminants in foods (GB 2762-2005) for Pb, Cd, and Cr; the tolerance limit of copper in foods (GB 15199-1994) for Cu; and the tolerance limit of zinc in foods (GB 13106-1991) for Zn.
[b)]The hygienical standard for feeds (GB 13078-2001) for Pb, Cd and Cr.
[c)]DW = dry weight.
[d)]Mean±standard deviation ($n = 20$).

were generally low, being comparable with those recorded in soybean grown at ZX village.

In China, the tolerance limit is 0.2 mg kg^{-1} for both Pb and Cd in food and 1.0 mg kg^{-1} for Cr (MHPRC, 2005). The concentrations of Pb (for FD), Cd (for ZX) and Cr (both FD and ZX) in the seeds of soybean exceeded the tolerance limits (Table II). Therefore, soybeans grown around the mine area could pose a potential health risk to local inhabitants. According to the Tolerance Limit of Heavy Metals for Feed of China (CERSPC, 2009), the concentrations of Cd in the leaves of soybean grown at two villages exceeded the standard limit (0.5 mg kg^{-1}) for animal feeds. The Pb concentration in the leaves of soybean at ZX village exceeded the fodder maximum permissible level (5 mg kg^{-1}), with an average level 2.4 times the maximum permissible level (Table II). This indicated that leaves of soybean were unsafe as animal feeds. Standard limits for Zn and Cu have not been established because animal feeds need rather high contents of these trace elements (MHPRC, 1991, 1994).

Bioaccumulation factor of metals from soils to seeds and leaves of soybean

Soil-to-plant transfer is one of the important pathways of human exposure to metals through the food chain. Bioaccumulation factor (BAF) is a common parameter often used in the study of environmental contamination. Table III displays the BAFs calculated for heavy metal transfer from soils to seeds and leaves of soybean. The trends in the BAFs of heavy metals in leaves were in a descending order as Cd > Zn > Ni > Cu > Cr > Pb for FD and Cd > Zn > Ni > Pb > Cu > Cr for ZX. The high BAFs of Cd and Zn for leaves of soybean were similar to the results reported by Khan *et al.* (2008) for Brassica species. Gu *et al.* (2005) suggested that Cd can bind with enzymes instead of Zn when the two metals simultaneously enter plant cells, because Zn and Cd affect nucleic acid metabolism in the same manner.

The BAF values of Cd from soils to leaves of soybean were 5.610 for ZX and 1.279 for FD, which were

TABLE III

Bioaccumulation factor (BAF) values of heavy metals from soil to seeds and leaves of soybean at Fandong (FD) and Zhongxin (ZX) villages

Site	Transfer	Pb	Cd	Cu	Zn	Cr	Ni
FD	From soil to leaves	0.001	0.096	0.023	0.148	0.014	0.431
	From soil to seeds	0.014	1.279	0.020	0.537	0.016	0.266
ZX	From soil to leaves	0.003	0.455	0.090	0.429	0.032	0.493
	From soil to seeds	0.230	5.610	0.091	2.054	0.060	0.277

higher than those of any other metal examined. In this study, Pb and Cr are less mobile in soybean plants, which strongly agreed with the observation of Gigliotti *et al.* (1996). BAFs of heavy metals from soil to both seeds and leaves of soybean for ZX were remarkably higher than the corresponding BAFs of FD, except for Ni. The significant difference in BAFs between the two villages may be related to soil nutrient management and soil properties (Cui *et al.*, 2004). In this work, soybeans accumulated relatively high Cd in their leaves and seeds despite fairly low concentrations of Cd in the soils. These results are consistent with reports that Cd is a readily mobile metal and the presence of Zn in the soil may enhance the accumulation of Cd (Shute and Macfie, 2006).

Health risk of consuming soybean seeds

The estimated dietary intakes and target hazard quotients of heavy metals for adults around Dabaoshan Mine through consumption of soybean are listed in Table IV. The trends of EDIs for heavy metals through consumption of soybean were in the order of Zn > Cu > Ni > Cr > Pb > Cd for the inhabitants of FD. The ZX inhabitants *via* soybean consumption ingested higher amounts of Cd, Cr and Ni than FD inhabitants. The estimated daily intake values for both FD and ZX were less than the RfDo limits. Though the EDIs of metals from soybean consumption were lower than those of rice and vegetables from our previous studies (Zhuang *et al.*, 2009), attention should be paid to the soybean consumption since the EDIs of both Cu and Ni were greater than 50% of the RfDo values.

The THQs of individual heavy metals for both villages were all below 1, but THQ values of Cu and Ni approached 1. The THQs of Cr for both villages were minimal (0.001), in agreement with previous studies (Huang *et al.*, 2008). It has been reported that exposure to two or more contaminants may result in additive and/or interactive effects (Hallenbeck, 1993). The total metal THQs for soybean were 2.475 and 2.437 for FD and ZX villages, respectively (Table IV). The present results might indicate that great attention should be paid to the potential health risk to the inhabitants in the vicinity of the Dabaoshan Mine.

CONCLUSIONS

The high I_{geo} values of soils indicated severe soil contamination by Pb, Cd, Cu and Zn at FD village near the mine area. Cadmium and Pb concentrations in soybean seeds were above the national maximum permissible levels for food. Among the selected heavy metals, potential health hazards of Cu and Ni were greatest and Cr was the least. The analysis of the estimated dietary intakes and target hazard quotients for six selected heavy metals showed that there was a potential health risk to the local inhabitants consuming contaminated soybean.

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TABLE IV

Estimated daily intakes (EDIs)[a)] and target hazard quotients (THQs) of metals for adults around the Dabaoshan Mine through consumption of seeds of soybean at Fandong (FD) and Zhongxin (ZX) villages

Site	Parameter	Pb	Cd	Cu	Zn	Cr	Ni	Sum
FD	EDI (μg d^{-1} kg^{-1} BW[b)])	0.570	0.170	38.550	142.400	1.910	14.530	-
	THQ	0.142	0.167	0.964	0.475	0.001	0.727	2.475
ZX	EDI (μg d^{-1} kg^{-1} BW)	0.267	0.383	27.069	95.653	2.917	19.792	-
	THQ	0.067	0.383	0.677	0.319	0.002	0.990	2.437

[a)]Recommended dietary intakes = 100 g d^{-1} $person^{-1}$.

[b)]BW = body weight.

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