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Health risk from heavy metals via consumption of food crops in the vicinity of Dabaoshan mine, South China

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ABSTRACT

Heavy metal contamination of soils resulting from mining and smelting is causing major concern due to the potential risk involved. This study was designed to investigate the heavy metal (Cu, Zn, Pb and Cd) concentrations in soils and food crops and estimate the potential health risks of metals to humans via consumption of polluted food crops grown at four villages around the Dabaoshan mine, South China. The heavy metal concentrations in paddy and garden soils exceeded the maximum allowable concentrations for Chinese agricultural soil. The paddy soil at Fandong village was heavily contaminated with Cu (703 mg kg^{-1}), Zn (1100 mg kg^{-1}), Pb (386 mg kg^{-1}) and Cd (5.5 mg kg^{-1}). Rice tended to accumulated higher Cd and Pb concentration in grain parts. The concentrations of Cd, Pb and Zn in vegetables exceeded the maximum permissible concentration in China. Taro grown at the four sampled villages accumulated high concentrations of Zn, Pb and Cd. Bio-accumulation factors for heavy metals in different vegetables showed a trend in the order: Cd>Zn>Cu>Pb. Bio-accumulation factors of heavy metals were significantly higher for leafy than for non-leafy vegetable. The target hazard quotient (THQ) of rice at four sites varied from 0.66–0.89 for Cu, 0.48–0.60 for Zn, 1.43–1.99 for Pb, and 2.61–6.25 for Cd. Estimated daily intake (EDI) and THQs for Cd and Pb of rice and vegetables exceeded the FAO/WHO permissible limit. Heavy metal contamination of food crops grown around the mine posed a great health risk to the local population through consumption of rice and vegetables.

1. Introduction

Mining and processing metal ore can be a significant source of heavy metal contamination of the environment (Dudka and Adriano, 1997; Navarro et al., 2008; Singh et al., 2005). The environmental concern in mining areas is primarily related to physical disturbance of the surrounding landscape, spilled mine tailings, emitted dust and acid mine drainage (AMD) transported into rivers. Excessive accumulation of heavy metals in agricultural soils around mining areas, resulting in elevated heavy metal uptake by food crops, is of great concern because of potential health risk to the local inhabitants (Adriano, 2001; McLaughlin et al., 1999; Pruvot et al., 2006). The consumption of plants produced in contaminated areas, as well as ingestion or inhalation of contaminated particles is two principal factors contributing to human exposure to metals. Potential health risks to humans and animals from consumption of crops can be due to heavy metal uptake from contaminated soils via plant roots as well as direct deposition of contaminants from the atmosphere onto plant surfaces (McBride, 2003).

Cultivation of crops for human or livestock consumption on contaminated soil can potentially lead to the uptake and accumulation of trace metals in the edible plant parts with a resulting risk to human and animal health (Gupta and Gupta, 1998; McBride, 2007; Monika and Katarzyna, 2004). Increasing

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evidence shows that heavy metal pollution of mined areas caused health damage to the local inhabitants (Kachenko and Singh, 2006; Liu et al., 2005). It is known that serious systemic health problems can develop as a result of excessive dietary accumulation of heavy metals such as Cd, and Pb in the human body (Oliver, 1997). Although Zn and Cu are essential elements, their excessive concentration in food and feed plants are of great concern because of their toxicity to humans and animals (Kabata-Pendias and Mukherjee, 2007). Lead and Cd are considered potential carcinogens and are associated with etiology of a number of diseases, especially cardiovascular, kidney, nervous system, blood as well as bone diseases (Jarup, 2003). Lăcătuşu et al. (1996) reported that soil and vegetables polluted with Pb and Cd in Copsa Mica and Baia Mare, Romania, significantly contributed to decreased human life expectancy within the affected areas, reducing average age at death by 9–10 years. It was reported that children living around a former smelter had high blood Pb levels in France (Pruvot et al., 2006) and Brazil (Bosso and Enzweiler, 2008). Türkdoğan et al. (2002) suggested that the high prevalence of upper gastrointestinal cancer rates in the Van region of Turkey was related to the high concentration of heavy metals in the soil, fruit and vegetables. Dietary intake is the main route of exposure for most people, although inhalation can play an important role in very contaminated sites (Tripathi et al., 1997). Thus information about heavy metal concentrations in food products and their dietary intake is very important for assessing their risk to human health.

There are several thousand abandoned or operating metal based ore mines in China. Mining activities alone have generated about 1,500,000 ha of wasteland in China (MEPPRC, 2006), and this figure is increasing at a rate of 46,700 ha per year. Mining activities of Dabaoshan mine have generated large quantities of mine waste without any proper treatment since mining began in the 1970s. Previous investigations have shown that the mining activities polluted approximately 83 villages, 585×10^4 m^2 paddy fields, and 21×10^4 m^2 ponds around the mine. This has not only led to severe contamination of soil, sediments, and plants in the vicinity of the mine but has also led to health problems in people living downstream of the mine. Certain regions or villages around Dabaoshan mine have been termed endemic cancer regions/villages, because esophageal cancers, liver cancer, and other cancers were reported frequently in humans and poultry, with a mortality rate approaching 56% in humans. It is therefore important to control and limit the accumulation of heavy metals in food crops. The objective of this paper is to (1) quantify the concentration of heavy metals in soil, rice and vegetables; (2) estimate dietary intake and target hazard quotient (THQ) of heavy metals through consumption of vegetables and rice.

2. Material and methods

2.1. Description of the sampling sites

Dabaoshan mine (24°31′37″N; 113°42′49″E) is located in eastern Shaoguan city, Guangdong province, southern China (Fig. 1). This area has a humid subtropical climate with an annual average temperature of 20.3 °C and rainfall of 1762 mm. The Dabaoshan mineral deposit is a well-known polymetallic sulfide meso-hypothermal deposit, located at the boundary between Qujiang county and Wengyuan county of Guangdong Province in China. The minerals in the ore mainly consist of pyrite, pyrrhotite, and chalcopyrite with minor components of sphalerite, chalcocite, galena, limonite, calaverite, and native bismuth. Oxidation of sulfide-bearing minerals exposed to weathering has resulted in acid mine drainage (AMD), which is characterized by extreme acidity and a high level of dissolved metals (*e.g.*, Cu, Zn, Cd, Pb and As) and anions (*e.g.*, sulfates and carbonates). Since the mining operation began in the 1970s, large quantities of mine wastes and AMD have been dispersed downslope into

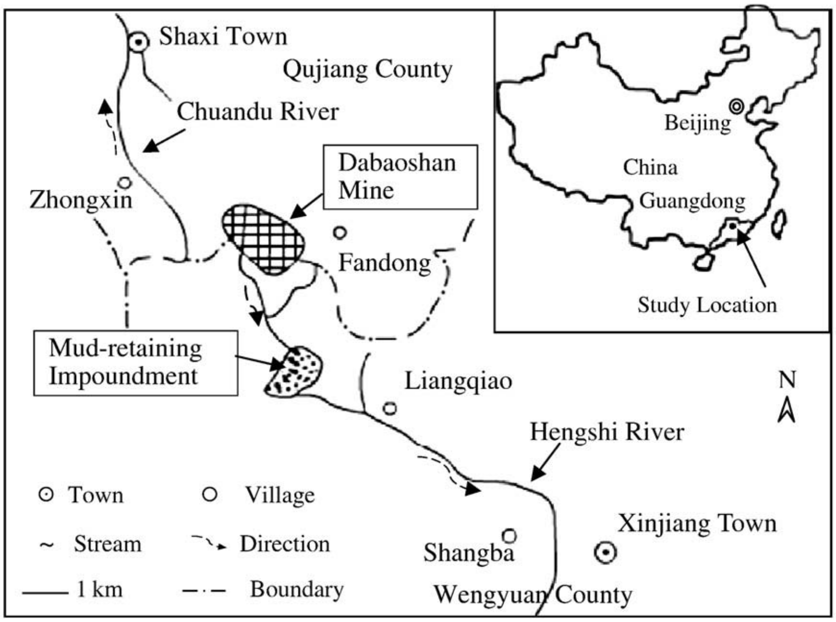

Fig. 1 – Schematic map of sampling sites in the vicinity of Dabaoshan mine area in Guangdong province (southern China). The four sampled villages included: Zhongxin (ZX), Fandong (FD), Liangqiao (LQ), and Shangba (SB).

the Hengshi River, which is mainly used to irrigate agricultural land for food crops and has led to severe deterioration and contamination of the surrounding environment.

Four sampling sites were selected in the vicinity of the Dabaoshan mine (Fig. 1). Zhongxin (ZX) village (24°35′72″N; 113°39′94″E) is located in the northern Dabaoshan mine, approximately 6–8 km away from the mine area. This area was contaminated by the wastewater (Chuandu River) of mining operation. Fandong (FD) village (24°33′78″N; 113°43′94″E), located on the mountaintop mine area (altitude at 500 m), was mainly affected by mining activities and atmospheric deposition. There was a mine tailing dam 0.5 km away from the village. Liangqiao (LQ) village (24°35′79″N; 113°42′34″E) is located approximately 3–4 km downslope from the mine on the Hengshi River. Shangba (SB) village (24°27′83″N; 113°48′16″E) is located south of the Dabaoshan mine, approximately 16–18 km away from the mine area. The agricultural soil in this region was repeatedly irrigated with polluted water from the Hengshi River.

2.2. *Sampling and pre-treatment*

Soils and plants grown at the above four villages in the vicinity of Dabaoshan mine were collected in November 2007. Taking into account regional consumption practices, the food crops included rice as the staple crop and lettuce, spinach, carrot, taro, and tomato etc. for home-grown vegetables. There are four replications for each vegetable. Vegetables were washed thoroughly with Milli-Q water, and the fresh weights (FW) of the samples were recorded. Each individual vegetable was separated into root, stalk, leaf and fruit sub-samples. All sub-samples were dried, and the dried samples were weighed again, then pulverized and stored in polythene zip-bags. At each sampling site, soil samples (0–20 com depth) were collected by a random sampling method, with paired soil and plant samples taken. The soil samples were air-dried at room temperature, then pulverized and sieved through a 1 mm stainless-steel mesh.

2.3. *Sample analysis*

The soil pH was measured in H_2O (1:2.5, soil:solution ratio, dry w/v). Diethylenetriamine pentaacetic acid (DTPA) extraction of the metals involved shaking 5.0 g of soil for 2 h with 25 ml of a solution containing 0.005 mol L^{-1} DTPA 0.01 mol L^{-1} $CaCl_2$, and 0.1 mol L^{-1} TEA (triethanolamine) buffered at pH 7.3 (Lindsay and Norvell, 1978). The suspension was then centrifuged for 30 min at 5100 *g* and filtered. Organic matter content was determined by the Walkley–Black's procedure (Nelson and Sommers, 1982). Soil samples were digested in preparation for total metal analysis using a concentrated acid mixture (HNO_3, $HClO_4$, and HF). For vegetable samples, 0.5 g of dried samples was digested with HNO_3 and $HClO_4$ in a 5:1 ratio until a transparent solution was obtained (Allen et al., 1986). The soil and plant digests were filtered and diluted to 50 and 25 ml, respectively, with distilled water. The metals in DTPA extracts and acid digests of soils and plants were analyzed by a flame atomic adsorption spectrophotometer (AAS, GBC932AA). However, Pb and Cd concentrations in vegetables were determined using graphite furnace atomic absorption spectrophotometer (GFAAs, GBC932AA).

2.4. *Quality assurance and quality control*

Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. Double distilled deionised water was used throughout the study. Glassware was properly cleaned, and the reagents were of analytical grade. Reagents blank determinations were used to correct the instrument readings. For validation of the analytical procedure, a recovery study was carried out by spiking and homogenizing several already analyzed samples with varied amounts of standard solutions of the metals. Standard reference materials (SRM) obtained from the National Research Center for CRMs (Beijing China), including soil (GBW08303), tea leaves (GBW07605), and rice powder (GBW08502), were used for validation of the analytical procedure. The results of measurements of CRMs are summarized in Table 1. 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.

2.5. *Data analysis*

2.5.1. *Bio-accumulation factor (BAF)*

Heavy metal concentrations of soils and crops were calculated on the basis of dry weight. The bio-accumulation factor (BAF), an index of the ability of the vegetable to accumulate a particular metal with respect to its concentration in the soil substrate (Ghosh and Singh, 2005), was calculated as follows:

$$\mathrm{BAF} = \frac{C_{\mathrm{plant}}}{C_{\mathrm{soil}}}$$

where C_{plant} and C_{soil} represent the heavy metal concentration in edible part of vegetables and soils, respectively.

Table 1 – Summary of measures of certified reference element concentrations (mg kg^{-1}, mean ± SD, $n=3$) in CRMs

	GBW08303			GBW07605			GBW08502		
	Certified value	Measured value	Recovery[a] (%)	Certified value	Measured value	Recovery (%)	Certified value	Measured value	Recovery (%)
Cu	120 ± 6	117 ± 0.6	98	17.3 ± 1.8	16.9 ± 1.3	98	2.6 ± 0.2	2.5 ± 0.04	96
Zn	260 ± 11	262 ± 8.9	101	26.3 ± 2.0	27.6 ± 0.7	105	14.1 ± 0.5	13.8 ± 0.3	98
Pb	73 ± 2	72 ± 0.5	99	4.4 ± 0.3	4.23 ± 0.2	96	0.75 ± 0.06	0.79 ± 0.01	105
Cd	1.2 ± 0.07	1.1 ± 0.04	98	0.057 ± 0.01	0.055 ± 0.012	96	0.02 ± 0.002	0.019 ± 0.001	95

Notes: [a]Values quoted on dry weight basis; Recovery (%) = (Mean $_{\text{measured value}}$/Mean $_{\text{certified value}}$) × 100%.

2.5.2. *Estimated daily intake (EDI) of heavy metals*

The estimated daily intake (EDI) of heavy metals (Cu, Zn, Pb, and Cd) depended on both the metal concentration in crops and the amount of consumption of the respective food crop. The EDI of metals for adults was determined by the following equation:

$$\mathrm{EDI} = \frac{C_{\mathrm{metal}} \times W_{\mathrm{food}}}{Bw}$$

where C_{metal} (μg g^{-1}, on fresh weight basis) is the concentration of heavy metals in contaminated crops; W_{food} represents the daily average consumption of crops in this region; *Bw* is the body weight. Based on the dietary intake survey by Ma et al. (2005), the local inhabitants had an average consumption per person (60 kg in body weight) of 274 and 372 g day^{-1} for vegetables and rice, respectively. The C_{metal} of rice was converted with a factor of 0.86, because home-stored rice commonly contains 14% water content. The metal intakes were compared with the tolerable daily intakes for metals recommended by the WHO (1993) and by the Food and Nutrition Board (2004).

2.5.3. *Target hazard quotient (THQ)*

The health risks from consumption of vegetables and rice by the local inhabitants were assessed based on the target hazard quotient (THQ). The THQ 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 U.S. EPA Region III risk-based concentration table (USEPA, 2007) and in Chien et al. (2002), and is based on the equation below:

$$\mathrm{THQ} = \frac{\mathrm{EFr} \times \mathrm{ED} \times \mathrm{FI} \times \mathrm{MC}}{\mathrm{RfDo} \times \mathrm{BW} \times \mathrm{AT}} \times 10^{-3}$$

where THQ is target hazard quotient; EFr is exposure frequency (365 days year^{-1}); ED is exposure duration (70 years); FI is food ingestion (g person^{-1} d^{-1}); MC is metal concentration in food (μg g^{-1}, on fresh weight basis); RfDo is the oral reference dose (mg kg^{-1} d^{-1}); BW is the average body weight, adult (65 kg); AT is averaging time for noncarcinogens (365 days year^{-1}× number of exposure years, assuming 70 years in this study). Oral reference doses were based on 4E-02, 3E-01, 4E-03, 1E-03 mg kg^{-1} d^{-1} for Cu, Zn, Pb and Cd, respectively (USEPA, IRIS, 1997, 2007).

3. Results

3.1. *Heavy metal concentration in the soil*

The characteristics and total concentration of heavy metals (Cu, Zn, Pb and Cd) in soils sampled at the four sites around Dabaoshan mine are presented in Table 2. The pH value of the soils ranged from acidic to near neutral (4.76–6.89). The paddy soils at LQ and SB are acidic (4.76). The ZX and FD soils and SB garden soil are slightly acidic, while the mean pH value of LQ garden soil is nearly neutral (6.89). The organic matter ranged between 2.38 and 4.27%, with the highest and lowest in SB paddy soil and garden soil, respectively. The percentage of DTPA-extractable fractions of heavy metals ranged between 3.7–17.4% for Cu, 6.3–13.9% for Zn, 2.8–25% for Pb, and 11.7–22.8% for Cd. The highest concentrations of Cu, Zn, Pb and Cd in soil were found at FD village, indicating remarkably severe contamination on the mountaintop of the Dabaoshan mine. Metal concentrations in paddy and garden soils declined exponentially with increased distance from the inflow of wastewater along the Hengshi River or Chuandu River. Thus, Cu and Pb concentrations in paddy soils decreased in the order of distance from the mine: FD (<1 km)>LQ (3–4 km)>ZX (6–8 km)>SB (16–18 km). The Cu, Zn and Pb concentrations in garden soils decreased in the order: LQ>FD>SB>ZX. The highest mean Cd concentration (5.5 mg kg^{-1}) was found in paddy soils of FD village, whereas Cd in paddy (4.74 mg kg^{-1}) and garden soils (4.39 mg kg^{-1}) from ZX were significantly higher compared with the other three sites.

Table 2 – Characteristics of soils collected from the study area

Sites	Soil type		pH (H_2O)	OM (%)	Total concentration (mg kg^{-1})				DTPA-extractable (mg kg^{-1})			
					Cu	Zn	Pb	Cd	Cu	Zn	Pb	Cd
ZX 6–8 km	Paddy soil	Mean	5.1	3.2	431	249	160	4.7	59(13.7%) [a]	28(11.2%)	40(25%)	0.55(11.7%)
	(n=10)	Range	4.4–6.7	1.6–4.3	109–1018	170–359	109–208	3.6–6.2	10–134	9.7–48	26–60	0.32–1.1
	Garden soil	Mean	5.4	3.4	243	234	130	4.4	23(9.5%)	23(9.8%)	16(12.3%)	0.56(12.7%)
	(n=15)	Range	4.8–6.4	2.3–4.2	88–466	311.8	105–153	2.9–5.8	11–54	10–42	12–27	0.28–0.75
FD <1 km	Paddy soil	Mean	5.2	3.5	703	1100	386	5.5	109(15.5%)	107(9.7%)	75(19.4%)	1.2(21.8%)
	(n=6)	Range	5.1–5.3	2.7–4.0	292–1313	600–1663	195–621	2.9–7.6	60–188	57–155	36–127	0.65–1.4
	Garden soil	Mean	5.9	3.3	341	481	248	4.0	54(15.8%)	67(13.9%)	34(13.7%)	0.91(22.8%)
	(n=15)	Range	5.0–6.4	2.5–3.9	171–594	346–921	177–384	3.5–5.5	35–75	49–97	22–43	0.53–1.2
LQ 3–4 km	Paddy soil	Mean	4.8	3.1	550	444	345	3.0	72(13.1%)	47(10.6%)	57(16.5%)	0.63(21%)
	(n=8)	Range	4.5–5.4	2.5–3.6	337–634	261–705	247–425	1.8–3.9	51–102	26–92	46–71	0.23–0.99
	Garden soil	Mean	6.9	3.8	348	696	297	2.4	13(3.7%)	44(6.3%)	8.4(2.8%)	0.47(19.6%)
	(n=9)	Range	6.5–7.4	3.2–4.8	184–512	634–759	281–313	2.0–2.4	6.6–20	40–46	6.8–9.8	0.27–0.61
SB 16–18 km	Paddy soil	Mean	4.8	4.3	449	501	282	3.0	78(17.4%)	60(12%)	52(18.4%)	0.61(20.3%)
	(n=8)	Range	4.7–4.9	3.9–5.1	331–580	450–537	194–408	2.5–4	60–95	53–66	35–77	0.58–0.67
	Garden soil	Mean	5.9	2.4	213	279	151	1.6	34(16%)	21(7.5%)	17(11.3%)	0.29(18.1%)
	(n=15)	Range	5.5–6.4	1.9–3.2	182–280	271–290	127–184	1.3–2.1	32–38	19–22	16–18	0.25–0.32
GB 15618–1995 [b]			<6.5		50	200	250	0.3				

[a] DTPA extracts, a ratio of DTPA-extractable to total metals in the bracket.

[b] Grade II of Environmental Quality Standard for soils (GB 15618-1995) in China.

3.2. Heavy metal concentrations in rice

Fig. 2 shows the concentration of heavy metals in the different parts of rice (*Oryza sativa*) grown in the four selected sites around Dabaoshan mine. In terms of metals distribution in rice plants, the average concentration of Cu, Zn, Pb and Cd decrease in the order: stalk>husk>grain, with the exception that concentrations of Cu and Cd in the grain were higher than those in the husk by 1.4 and 1.3 times at FD village. The metal concentrations in rice grain did not differ significantly among the four villages.

3.3. Heavy metal concentrations in vegetables

The average concentrations and range of heavy metals (mg kg^{-1}, on fresh weight basis) in the selected vegetables grown in the vicinity of Dabaoshan mine are listed in Table 3. The concentrations of heavy metals were highest for Zn, followed by Cu, with Pb and Cd being the lowest. Water spinach, celery, lettuce, Brassica, aubergine and taro showed high Cd levels in the edible part. Among the vegetables, the average concentration of Pb was highest in root vegetables (Ipomoea, taro), with the lowest (0.22 mg kg^{-1} for Ipomoea) at LQ and the highest (0.39 mg kg^{-1}) at ZX village. Taro grown at the four sampling sites simultaneously accumulated high concentrations of Zn, Pb and Cd, except for Cd at LQ. Moreover, the metal concentrations in the edible portions of tomato and bean were generally low, being comparable with those recorded in leafy vegetables.

3.4. Bio-accumulation factors from soils to food crops

Fig. 3 displays the bio-accumulation factors (BAF) calculated for heavy metal transfer from soils to vegetables and rice. The trends in the BAF for heavy metals in different vegetables were in the descending order of Cd>Zn>Cu>Pb. There was significant difference in BAF values among the four sampling sites. Relatively low BAF values were found for Cu and Zn in rice crops compared to vegetables. The BAF values of Cd varied from 0.63 (at FD village) to 2.1 (at SB village) for leaf vegetables. The average BAF values of leaf vegetables were significantly higher than those of non-leafy vegetables at all four sites. The BAFs of Pb and Cd in rice grain exceeded or equaled those in non-leafy vegetables at LQ or ZX village.

3.5. Dietary intake of metals and target hazard quotients

The estimated dietary intakes (EDI) of heavy metals for adults in the vicinity of Dabaoshan mine via consumption of vegetable and rice are presented in Table 4. The trends of EDIs for heavy metals in rice and vegetables were in the order of Zn>Cu>Pb>Cd, with intake from rice being greater than from vegetables for all metals. The highest EDIs of Cu (2322 μg d^{-1}), Zn (11693 μg d^{-1}), Pb (516 μg d^{-1}) and Cd (406 μg d^{-1}) through

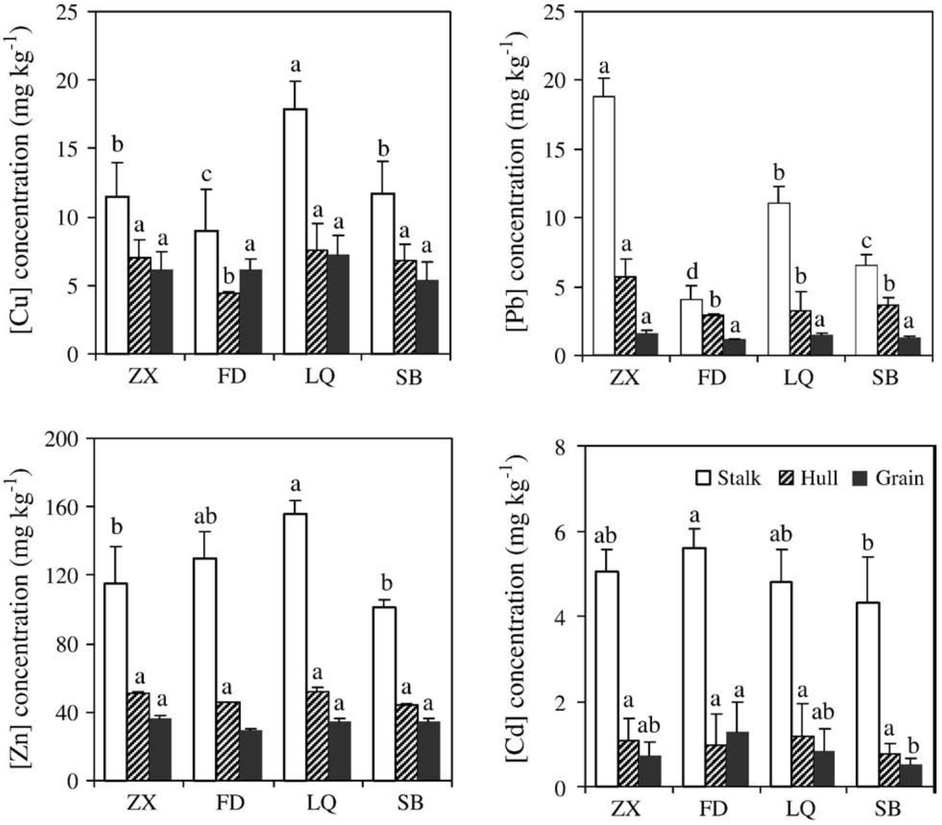

Fig. 2 – Heavy metal concentrations (mean±SD, on dry weight basis) in the different parts of rice grown in four mine-contaminated sites. The error bars indicate the standard deviation (SD). Lower case letters indicate multiple comparisons among different locations, bars with different letters are significantly different ($P<0.05$).

Table 3 – Heavy metal concentrations (mean ± SD, mg kg^{-1}, n = 4, on fresh weight basis) in most frequently consumed vegetables grown in the vicinity of Dabaoshan mine

Plant species	Cu	Zn	Pb	Cd
(a) ZX				
Brassica juncea (mustard)	1.11 ± 0.26	8.68 ± 0.81	0.21 ± 0.04	0.19 ± 0.02
Brassica oleracea (kale)	0.60 ± 0.05	8.96 ± 0.69	0.23 ± 0.03	0.32 ± 0.02
Brassica chinensis (cabbage)	1.11 ± 0.19	11.00 ± 1.03	0.12 ± 0.01	0.21 ± 0.02
Brassica rapa (Chinese cabbage)	0.53 ± 0.03	5.08 ± 0.61	0.09 ± 0.00	0.10 ± 0.01
Brassica napus (cole)	1.27 ± 0.11	8.38 ± 0.79	0.20 ± 0.03	0.28 ± 0.04
Lactuca sativa var.romana Gars (lettuce)	0.74 ± 0.05	6.90 ± 0.96	0.10 ± 0.01	0.32 ± 0.03
Lactuca sativa var.longifolia Lam (lettuce)	0.72 ± 0.03	4.57 ± 0.62	0.13 ± 0.01	0.33 ± 0.01
Lycopersicon esculentum (tomato)	0.76 ± 0.08	2.85 ± 0.31	0.06 ± 0.00	0.05 ± 0.00
Capsicum annuum (capsicum)	0.85 ± 0.04	3.47 ± 0.29	0.05 ± 0.00	0.07 ± 0.00
Solanum melongena (aubergine)	1.58 ± 0.19	4.46 ± 0.57	0.18 ± 0.01	0.28 ± 0.02
Nelumbo nucifera (lotus)	2.16 ± 0.51	5.48 ± 0.39	0.24 ± 0.02	0.05 ± 0.00
Raphanus sativus (radish)	0.45 ± 0.03	5.75 ± 0.52	0.12 ± 0.01	0.13 ± 0.01
Daucus carota (carrot)	1.33 ± 0.26	6.92 ± 0.71	0.09 ± 0.00	0.38 ± 0.05
Colocasia esculenta (taro)	3.13 ± 0.49	37.8 ± 2.39	0.36 ± 0.41	0.31 ± 0.04
Ipomoea batatas (Ipomoea)	1.90 ± 0.37	9.68 ± 0.79	0.39 ± 0.26	0.15 ± 0.01
(b) FD				
Brassica juncea (mustard)	0.85 ± 0.03	6.28 ± 0.83	0.13 ± 0.01	0.06 ± 0.00
Brassica oleracea (kale)	0.45 ± 0.02	13.8 ± 1.34	0.10 ± 0.01	0.15 ± 0.01
Brassica chinensis (cabbage)	0.67 ± 0.05	6.27 ± 0.65	0.12 ± 0.01	0.08 ± 0.00
Brassica rapa (Chinese cabbage)	0.39 ± 0.01	15.9 ± 1.61	0.09 ± 0.00	0.28 ± 0.06
Lactuca sativa var.romana Gars (lettuce)	0.95 ± 0.06	8.19 ± 0.76	0.19 ± 0.02	0.24 ± 0.03
Brassica napus (cole)	0.78 ± 0.04	21.1 ± 1.63	0.12 ± 0.01	0.28 ± 0.01
Lactuca sativa var.longifolia Lam (lettuce)	1.15 ± 0.19	13.6 ± 1.39	0.09 ± 0.01	0.02 ± 0.00
Spinacia oleracea (spinach)	1.23 ± 0.27	19.1 ± 1.08	0.14 ± 0.01	0.50 ± 0.03
Raphanus sativus (radish)	0.28 ± 0.01	3.28 ± 0.43	0.06 ± 0.00	0.04 ± 0.00
Dolichos lablab (Indian bean)	0.95 ± 0.10	7.18 ± 0.76	0.12 ± 0.01	0.03 ± 0.00
Lycopersicon esculentum (tomato)	0.79 ± 0.05	5.14 ± 0.46	0.07 ± 0.00	0.05 ± 0.00
Capsicum annuum (capsicum)	0.61 ± 0.02	2.34 ± 0.31	0.08 ± 0.00	0.05 ± 0.00
Colocasia esculenta (taro)	2.48 ± 0.31	40.2 ± 3.01	0.36 ± 0.03	0.71 ± 0.03
Dioscorea alata (yam)	2.83 ± 0.25	13.2 ± 0.19	0.38 ± 0.01	0.33 ± 0.02
Ipomoea batatas (Ipomoea)	1.60 ± 0.19	8.30 ± 0.92	0.35 ± 0.02	0.11 ± 0.01
(c) LQ				
Brassica juncea (mustard)	0.74 ± 0.03	5.07 ± 0.21	0.15 ± 0.01	0.09 ± 0.01
Brassica oleracea (kale)	0.31 ± 0.01	7.38 ± 0.35	0.01 ± 0.00	0.01 ± 0.00
Brassica rapa (Chinese cabbage)	0.84 ± 0.04	8.15 ± 0.73	0.12 ± 0.01	0.17 ± 0.01
Brassica napus (cole)	0.82 ± 0.02	7.45 ± 0.69	0.21 ± 0.03	0.13 ± 0.01
Lactuca sativa var.longifolia Lam (lettuce)	0.60 ± 0.01	5.57 ± 0.35	0.18 ± 0.02	0.19 ± 0.02
Solanum melongena (aubergine)	2.18 ± 0.19	4.25 ± 0.29	0.11 ± 0.01	0.23 ± 0.01
Capsicum annuum (capsicum)	0.78 ± 0.02	3.20 ± 0.18	0.09 ± 0.00	0.07 ± 0.00
Colocasia esculenta (taro)	2.60 ± 0.20	19.2 ± 1.46	0.18 ± 0.01	0.001 ± 0.00
Ipomoea batatas (Ipomoea)	1.69 ± 0.11	7.90 ± 0.67	0.22 ± 0.02	0.03 ± 0.01
(d) SB				
Brassica juncea (mustard)	0.75 ± 0.06	4.51 ± 0.38	0.37 ± 0.14	0.08 ± 0.00
Brassica oleracea (kale)	0.53 ± 0.04	4.99 ± 0.43	0.16 ± 0.09	0.12 ± 0.02
Brassica napus (cole)	0.94 ± 0.18	9.96 ± 0.86	0.12 ± 0.04	0.25 ± 0.01
Lactuca sativa var.romana Gars (lettuce)	0.98 ± 0.10	9.70 ± 0.69	0.13 ± 0.01	0.27 ± 0.02
Lactuca sativa var.longifolia Lam (lettuce)	0.51 ± 0.01	3.25 ± 0.28	0.07 ± 0.00	0.02 ± 0.00
Apium graveolens (celery)	1.01 ± 0.13	11.2 ± 0.49	0.11 ± 0.02	0.58 ± 0.35
Ipomoea aquatic (water spinach)	3.61 ± 0.62	9.52 ± .91	0.25 ± 0.03	0.65 ± 0.41
Daucus carota (carrot)	0.85 ± 0.16	5.70 ± 0.28	0.18 ± 0.01	0.14 ± 0.02
Phaseolus vulgaris (kidney bean)	0.98 ± 0.24	7.71 ± 0.62	0.12 ± 0.01	0.02 ± 0.00
Pisum sativum (garden pea)	0.94 ± 0.19	8.75 ± 0.64	0.10 ± 0.01	0.01 ± 0.00
Solanum melongena (aubergine)	0.96 ± 0.31	3.43 ± 0.21	0.10 ± 0.00	0.32 ± 0.31
Nelumbo nucifera (lotus)	1.60 ± 0.96	4.23 ± 0.19	0.16 ± 0.02	0.08 ± 0.01
Momordica charantia (bitter melon)	0.48 ± 0.08	3.16 ± 0.18	0.09 ± 0.00	0.12 ± 0.01
Colocasia esculenta (taro)	3.08 ± 0.58	48.1 ± 2.89	0.33 ± 0.16	0.30 ± 0.02
Ipomoea batatas (Ipomoea)	2.16 ± 0.49	7.62 ± 0.47	0.38 ± 0.31	0.05 ± 0.01

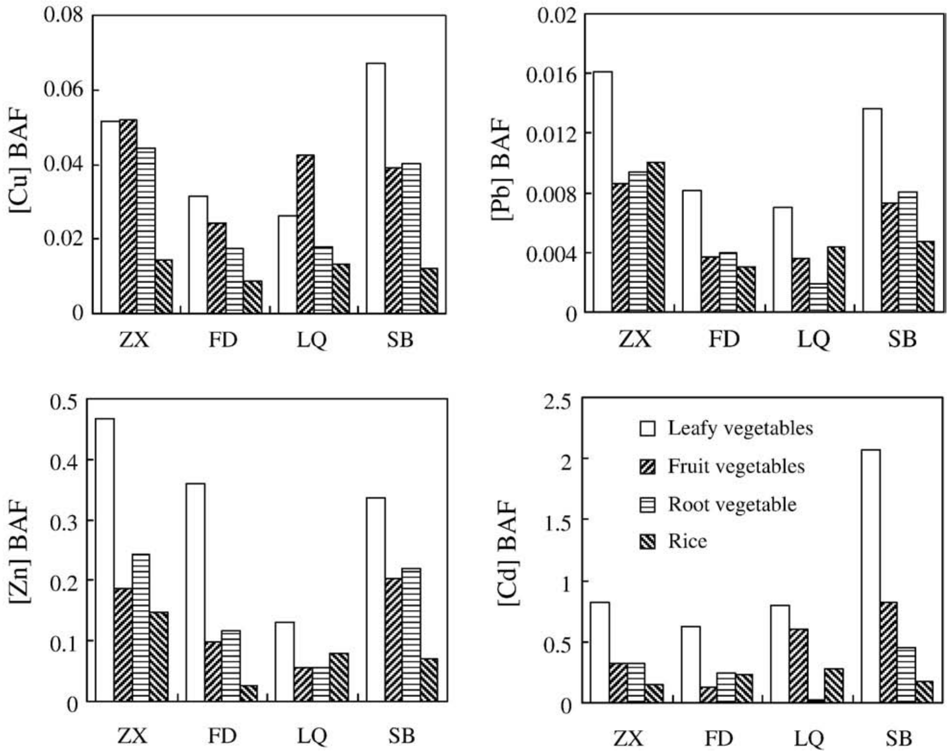

Fig. 3 – Bio-accumulation factor (BAF), a ratio of heavy metals concentration in the edible part of leafy, fruit, root vegetables and rice to that in the corresponding soil at the four contaminated villages.

consumption of rice were from LQ, ZX, ZX, and FD village, respectively. For the inhabitants of FD village, an adult was estimated to have an intake of 459 μg Cd per day via consumption of 372 g rice and 274 g vegetables only.

Table 4 – Estimated dietary intake of heavy metals (μg d^{-1}) via consumption of rice and vegetables at the four sampled villages

Type of food	Daily intake (g d^{-1})[a]	Cu	Zn	Pb	Cd
(a) ZX					
Rice	372	1977	11,693	516	229
Vegetables	274	327	2357	47	59
Total		2304	14,050	563	287
(b) FD					
Rice	372	1961	9447	371	406
Vegetables	274	290	3344	45	53
Total		2251	12,791	416	459
(c) LQ					
Rice	372	2322	11,172	476	269
Vegetables	274	339	2104	39	27
Total		2661	13,276	514	296
(d) SB					
Rice	372	1721	11,127	425	170
Vegetables	274	352	2559	49	57
Total		2073	13,685	474	226

[a] Based on study of dietary intake and nutritional status of residents in Guangdong by Ma et al. (2005).

The target hazard quotient (THQ) of each metal through consumption of both rice and vegetables in the vicinity of Dabaoshan mine decreased in the order of Cd>Pb>Cu>Zn. The metals THQ through consumption of rice and vegetables grown at four villages in the vicinity of Dabaoshan mine are given in Fig. 4. The THQ value of rice at four sites varied from 0.66–0.89 for Cu, 0.48–0.60 for Zn, 1.43–1.99 for Pb, and 2.61–6.25 for Cd. Cadmium exhibited relatively higher THQ for vegetables compared to all other metals, with ZX, SB and FD reaching values as high as 0.90, 0.87 and 0.81. The maximum value of Cd THQ for rice consumption from FD village was 6.25 followed by LQ (4.14), ZX (3.52), and SB (2.61) village. The Pb THQ values for rice consumption were more than 1 and in the order: ZX (1.33)>LQ (1.83)>SB (1.64)>FD (1.43).

4. Discussion

4.1. Soil contamination

Heavy metals present an environmental hazard in the vicinity of mining and smelting activities. Heavy metal uptake by food crops depends upon soil physicochemical characteristics and plant species. The low pH values (4.76) in the paddy soil samples from LQ and SB villages may be due to irrigation with wastewater from the Hengshi River downstream of mining activity for four decades. The pH of garden soils from LQ village is higher (pH>6.0), which may be the result of food crops being irrigated with well water. According to the China Environmental Quality Standard for Soils (NEPAC, 1995, Grade II for soil pH<6.5: Cu≤50 mg kg^{-1}; Zn≤200 mg kg^{-1}; Pb≤250 mg kg^{-1};

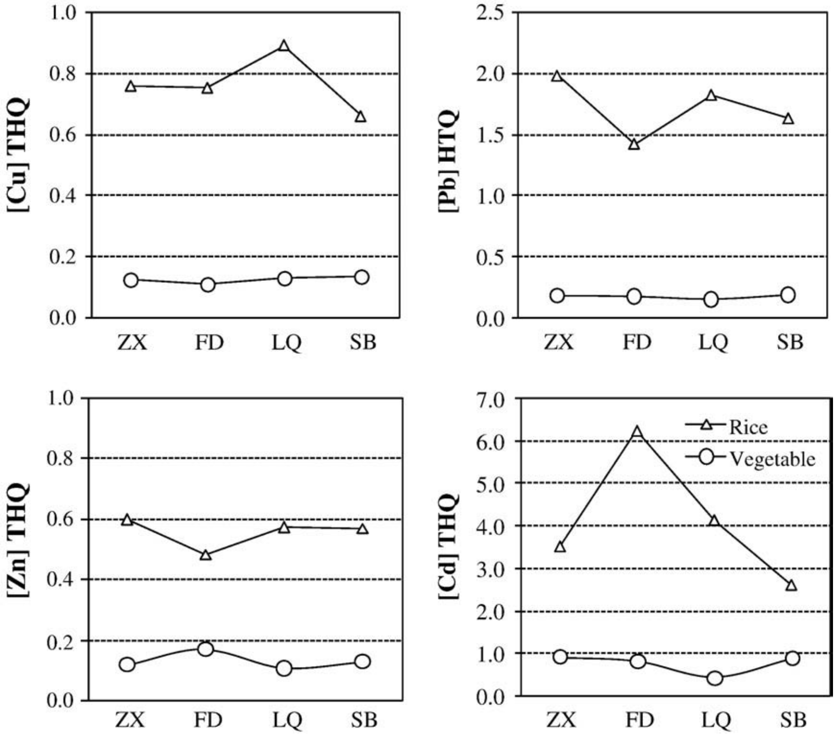

Fig. 4 – Metals THQ values through consumption of rice and vegetables grown at the four sampled villages in the vicinity of Dabaoshan mine.

Cd≤0.3 mg kg^{-1}, indicating a pollution warning threshold), heavy metal concentrations in most soil samples (paddy and garden) warranted a pollution warning, especially for FD village which is nearest to the Dabaoshan mine (Table 2). In the soil of FD village, Cd concentrations in all samples of four sampling sites revealed a moderate to serious contamination (>1.0 mg kg^{-1}, Chinese standard). The extent of soil metal contamination in the polluted location was consistent with previous study reports on the Dabaoshan mine area by Zhou et al. (2007).

However, some studies of mine-contaminated soils have observed that the speciation of metals rather than the total concentration of heavy metals is needed to evaluate the phytotoxic risk (Remon et al., 2005; Vega et al., 2004). DTPA extracts labile forms of metals that may become plant-available over time. In the present study, the proportion of total metal extracted by DTPA was greater for Cd (11–23%) than for Pb (2.8–25%), Zn (6.3–14%) or Cu (3.7–17%). A study by Singh et al. (1998) found that the percentage of total metals extracted by DTPA followed the order: Cd (38%)>Cu (28%)≥Zn (26%)>Pb (13%), indicating that Cd and Cu were the most extractable. DTPA extracts can distinguish between adsorbed metals (which are extracted) and metals contained in the structure of primary minerals from the metal ore itself (which are not extracted by DTPA) for mine contamination sites. Although the amounts of DTPA-extractable metals do not absolutely represent the actual quantities of soil metals that can be taken up by plants, they do appear to be good indicators of the potentially bioavailable quantity (Wang et al., 2006). Consequently, a substantial fraction of the metals in the soils are potentially plant-available in the vicinity of Dabaoshan mine.

4.2. *Metal contamination in food crops*

Results from present and previous studies (Lim et al., 2008; Liu et al., 2005; Pruvot et al., 2006) demonstrated that the food crops grown on contaminated soil in the vicinity of mine and smelter threatened health for the local inhabitants. Rice is Asia's staple food. The Pb and Cd concentration in rice grain grown in this area were 8 and 6.5-fold higher than the maximum permissible level (0.2 mg kg^{-1}), respectively, according to the national safety standard for milled rice (NPSF, 2002). Rice accumulated 5.3–7.5 mg kg^{-1} Cu, 18–43 mg kg^{-1} Zn, 0.24–0.80 mg kg^{-1} Pb, and 0.38–7.0 mg kg^{-1} Cd in grain grown in the Dabaoshan mine area. The Pb concentration of rice was higher than that in the Chenzhou Pb/Zn mine area, southern China (Liu et al., 2005), but other metal concentrations were similar. These results indicate that rice has some ability to transfer soil Cd and Pb into grain. Reeves and Chaney (2001) suggested that Cd is of primary concern in soil and food contamination, particularly in rice cropping systems. In corn grain grown on a Pb/Zn minesoil of Liaoning Province, the Cd and Pb concentrations exceeded the standards by 1.5 and 2.0 times (Gu et al., 2005).

In China, the maximum permissible concentrations for Cu and Zn in vegetables are 10 and 20 mg kg^{-1} (on fresh weight basis), respectively (MHPRC, 2005). It is a positive result that Cu and Zn concentrations in all the tested vegetables grown in the vicinity of Dabaoshan mine were below the maximum permissible levels (Table 3). On the other hand, the frequently high soil total Cu and Zn concentrations combined with the relatively low mean soil pH values (Table 2) suggests a strong likelihood of phytotoxicity in some crops. Chinese cabbage grown at FD village could accumulate high concentrations of Zn and Cd in the leaf. Ipomoea grown at SB village had the highest Cu and Cd concentrations in leaves. Leafy vegetables like lettuce are generally considered to accumulate Pb and Cd to a higher extent than roots/tuberous vegetables (Stalikas et al., 1997; Li et al., 2006), which is due to the fact that leafy vegetables have high translocation, high transpiration and also fast growth rates (Itanna, 2002; Muchuweti et al., 2006). In addition, they are susceptible to physical contamination by soil dust and splash because of their high foliar surface areas. Spinach grown at FD village accumulated 0.5 mg kg^{-1} Cd in the leaf, which was 2.5 times higher than the MPL (0.2 mg kg^{-1}), probably because Cd was in a labile form in soils (Hernandez et al., 2003). At ZX village (Table 3a), carrot roots showed the highest Cd concentration (0.38 mg kg^{-1}) of all vegetable crops grown there, over 3.8 times the permissible Chinese standard (0.1 mg kg^{-1}). The levels of Pb in the edible part of vegetables (except for root vegetables) were within the maximum permissible limits for vegetables in China. Ipomoea could accumulate high concentrations of Cd in the root, and Liu et al. (2005) also reported high accumulation of Cd and Pb in the leaves and stems. Taro grown in the mine-contaminated area had high concentrations of Zn, Pb and Cd, consistent with a report by Liu et al. (2005). Previous studies have found that leafy vegetables like lettuce and spinach can accumulate higher Pb and Cd concentration in the edible parts than non-leafy vegetables like carrots, suggesting that the mobility of Pb and Cd in soils depends on the crop's physiological properties (Lăcătuşu et al., 1996; Wang et al., 2006).

4.3. *Metal transfer from soil to crops*

Principally, the food chain (soil–plant–human) pathway is recognized as one of major pathways for human exposure to soil contamination. Soil-to-plant transfer is one of the key components of human exposure to metals through the food chain. Our data showed that BAF values differed significantly among locations or plant species. The difference in BAFs between locations may be related to soil nutrient management and soil properties. The results from Fig. 3 show that BAFs of food crops for these considered metals were in the order: Cd>Zn>Cu>Pb, which were strongly consistent with the reports by McBride and Liu (McBride, 2003; Liu et al., 2005). For a given metal, the transfer factor varies greatly with plant species (Alloway and Jackson, 1991; Cui et al., 2004). The high BAFs of Cd and Zn for leafy vegetables were similar to the results reported by Khan et al. (2008) for Brassica species. It is suggested that Cd can bind with enzymes instead of Zn when the two metals simultaneously enter plant cells (Gu et al., 2005), as Zn and Cd affect nucleic acid metabolism in the same manner. Consequently, Cd is easier than Zn to transfer from soil to the edible part of crops. In non-leafy vegetables, the BCF values at FD and LQ village were lower than those at ZX and SB, which might be due to the observed differences in soil properties. These findings strongly suggest that the transfer of heavy metals from soil to food crops explains the relatively high concentration of Pb and Cd in rice and vegetables, consistent with the conclusions of Cui et al. (2004) and Zheng et al. (2007).

4.4. *Health risk of local inhabitant*

To appraise the health risk associated with heavy metal contamination of rice and vegetables grown in the vicinity of Dabaoshan mine area, estimated dietary intake (EDI) and target hazard quotients (THQ) were calculated. The EDI of heavy metals was evaluated according to the average concentration of each heavy metal in each food crops and the respective consumption rate (Santos et al., 2004). The EDIs of heavy metal (Cu, Zn, Pb and Cd) through consumption of both cereal and vegetables for local inhabitants in the Dabaoshan mine area were lower than those in the Chenzhou Pb/Zn mine area located in southwest China (Liu et al., 2005). Nevertheless, the highest EDIs of Pb (563 μg d^{-1} at ZX) and Cd (459 μg d^{-1}at FD) through consumption of food crops were found to be significantly higher than the PTDI by 2.3 and 6.8 times, respectively (WHO, 1989). The biggest contribution to the intake of heavy metals came from rice, with 3–11 times as much as intake through vegetables, as shown in Table 4. Thus, perennial intake of these contaminated food crops is likely to induce adverse health effects arising largely from Pb and Cd exposure.

The THQ has been recognized as a useful parameter for evaluation of risk associated with the consumption of metal contaminated food crops (Rupert et al., 2004; Sridhara Chary et al., 2008). The THQs of Pb and Cd via rice consumption were higher than 1, and the THQ of Cd for vegetables approached 1 at three study sites, which showed that the inhabitants around Dabaoshan mine are experiencing relatively high health risk. However, Horiguchi et al. (2004) suggested that the ingested dose of heavy metals is not equal to the absorbed pollutant dose in reality, as a fraction of the ingested heavy metals may be excreted, with the remainder accumulated in body tissues where they affect human health. The THQ of heavy metals through consumption of rice in the sampled villages decreased in the order of distance from the Dabaoshan mine as follows: FD (<1 km)>LQ (3–4 km)>ZX (6–8 km)>SB (16–18 km), indicating that health risks diminish with distance from the mine (Fig. 4). The differences in the total metal THQ for various villages is largely attributable to the significantly different contributions of Cd. The THQs of Cu and Zn for rice and vegetables were generally less than 1, suggesting that the local inhabitants will not be exposed to a potential health risk from dietary Cu and Zn. The present results indicate that Cd was the major component contributing to the potential health risk, with Pb being of secondary importance, in agreement with a separate assessment for an area near a smelter in Nanning, China (Cui et al., 2004) and in the vicinity of smelters in Boolaroo, Australian (Kachenko and Singh, 2006). From the data produced in the present study, it is clear that Pb and Cd pose a potential risk to the local inhabitants through consumption of contaminated rice and vegetables. Consequently, some effective measures may be necessary to cure heavy metal contamination in soil and to reduce metal translocation from soil to edible crops in this region, and their implications for human health should be identified urgently by in-depth studies.

5. Conclusion

From the results of the rice and vegetable samples collected from this study, we obtained a better knowledge regarding the impact of the mining and smelting operation on the environment and the potential risk to human health. The extent of contamination was in the order: FD>LQ>ZX>SB, which indicated that health risks diminish with distance from the mine. By comparing the estimated dietary intake and target hazard quotients for Pb and Cd, this study determined that there was a potential health risk for the local inhabitants through consumption of contaminated food crops. Therefore, this region around Dabaoshan mine needs effective measures to cure the toxic metal contamination.

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