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ORIGINAL PAPER

Heavy metal contamination in soils and food crops around Dabaoshan mine in Guangdong, China: implication for human health

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Abstract This study was designed to investigate heavy metal (Cu, Zn, Pb, and Cd) contamination levels of soils, vegetables, and rice grown in the vicinity of the Dabaoshan mine, south China. The concentration of Cu, Zn, Pb, and Cd in paddy soil exceeded the maximum allowable concentrations for Chinese agricultural soil. The heavy metal concentrations (mg kg^{-1}, dry weight basis) in vegetables ranged from 5.0 to 14.3 for Cu, 34.7 to 170 for Zn, 0.90 to 2.23 for Pb, and 0.45 to 4.1 for Cd. The concentrations of Pb and Cd in rice grain exceeded the maximum permissible limits in China. Dietary intake of Pb and Cd through the consumption of rice and certain vegetable exceeded the recommended dietary allowance levels. The status of heavy metal concentrations of food crops grown in the vicinity of Dabaoshan mine and their implications for human health should be further investigated.

Keywords Heavy metals · Human health · Vegetables · Rice · Dabaoshan mine

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Introduction

Mining and smelting operations are important causes of heavy metal contamination in the environment due to activities such as mineral excavation, ore transportation, smelting and refining, and disposal of the tailings and waste waters around mines (Dudka and Adriano 1997; Navarro et al. 2008). Adverse environmental impacts from excessive heavy metals dispersed from mine and smelter sites include contamination of water and soil, phytotoxicity, soil erosion, and potential risks to human health (McLaughlin et al. 1999; Adriano 2001; Pruvot et al. 2006). Heavy metal contamination of agricultural soils and crops in the vicinity of mining areas has been regarded as a great environmental concern (Wcisło et al. 2002; Liu et al. 2005a; Kachenko and Singh 2006).

Several studies in China, South Korea, and the USA have shown that water (Lin et al. 2007), vegetables (Chang et al. 2005; Zheng et al. 2007), rice (Yang et al. 2006), and even fish (Schmitt et al. 2007) are often contaminated by heavy metals dispersed from mining and smelting operations. Li et al. (2006b) found that Chinese cabbage growing in the vicinity of nonferrous metals mining and smelting sites in Baiyin, China, contain high concentration of Cd exceeded the maximum permitted levels (0.05 mg kg^{-1}) by 4.5 times. In the vicinity of a Pb/Zn mine in Shaoxing, eastern China, it was reported that the respective Pb and Cd concentrations of some vegetables were 20 and 30 times higher than the permitted standards

(Li et al. 2006a). Clearly, not only the ingestion or inhalation of contaminated particles, but also the ingestion of plants produced in the contaminated area is another principal factor contributing to heavy metal of exposure for population. It has been recognized that food crops can be an important source of heavy metals for humans and animals (Dudka and Miller 1999).

Both heavy metal uptake via roots from contaminated soils and surface water, and direct deposition of contaminants from the atmosphere onto plant surfaces can lead to plant contamination by heavy metals. Lead and Cd are considered potential carcinogens and are associated with etiology of a number of diseases, especially cardiovascular, kidney, blood, nervous, and bone diseases (Jarup 2003). 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). Cultivation of crops for human or livestock consumption can potentially lead to the uptake and accumulation of these metals in edible plant parts with a resulting risk to human and animal health (Gupta and Gupta 1998; Lim et al. 2008). 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). 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. In France (Pruvot et al. 2006) and Brazil (Bosso and Enzweiler 2008), it was reported that children living around a former smelter had high blood Pb levels. 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 highly contaminated sites (Tripathi et al. 1997). Thus information about heavy metal concentrations in food products and their dietary intake is very important for assessing the risk to human health.

In China, there are over 9,000 state-owned and 30,000 private mining companies, and large amounts of hazardous wastes are released from base-metal mining and smelting operations annually. Cumulative use of land by mining was approximately 1,500,000 ha by 2006, with 60% of this area impacted by mine tailings (MEPPRC 2006). Metal ore processing usually leads to multimetal contamination of the environment, and topsoil in the vicinity of mines and smelters contains elevated concentrations of heavy metal (Dudka and Adriano 1997). Dabaoshan mine area (Guangdong, southern China) has been confirmed to have soils and waters severely pollution by heavy metals (Zhou et al. 2007; Lin et al. 2007). Mining activities during the past four decades have generated large quantities of mine waste materials without any proper treatment. It has been reported that mining activities polluted approximately 83 villages, 585 ha of paddy fields, and 21 ha of ponds around this mine. In the vicinity of Dabaoshan mine area, the number of cancer cases (esophageal cancer, liver cancer, etc.) is about nine times above the normal incidence of cancer, and the mortality rate approaches 56% (Liu et al. 2005b). Environmental surveys conducted by the Ministry of Health have shown that children living around the mine area had higher blood lead levels than those living in noncontaminated sites. This exposure has been probably attributed to the consumption of drinking water and crops contaminated by mining activities. It is therefore the objective of this present investigation to (1) quantify the concentrations of heavy metals in soils and crops grown around Dabaoshan mine, and (2) evaluate the potential health risk of humans via consumption of rice and vegetables.

Materials and methods

Description of the sampling sites

Dabaoshan mine (24°31′37″N; 113°42′49″E) is located at Shaoguan city, Guangdong, southern China. The Dabaoshan mineral deposit is located at the boundary between Qujiang county and Wengyuan county of Guangdong province in China. This area has a humid subtropical climate with an annual average 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, and calaverite. Since the mining operation began in the 1970s, large quantities of mine wastes and acid mine drainages (AMD) have been dispersed downslope into the Hengshi and

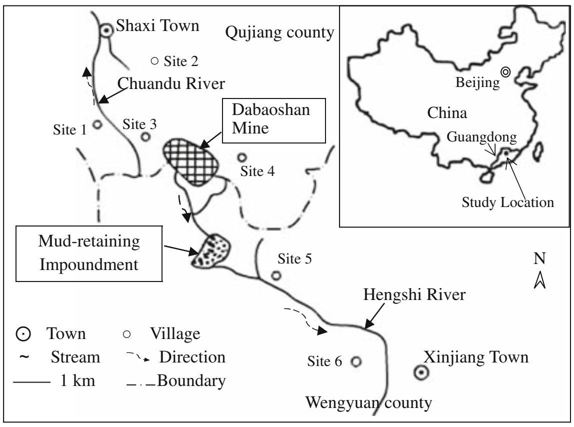

Fig. 1 The sampling location map of the Dabaoshan mine area. The symbol (○) represents the six sampling sites around the Dabaoshan mine

Chuandu Rivers, which are mainly used to irrigate agricultural land for field crops and vegetables, which has led to severe deterioration and contamination of the surrounding environment. Six sampling sites were selected to collect plant and soil samples in the vicinity of the Dabaoshan mine (Fig. 1).

Sample collection and preparation

Plant samples (rice and vegetables) and corresponding soil samples (paddy and garden soil) were collected from six sampling sites around Dabaoshan mine in August 2007. Sampling errors associated with the heterogeneous character of heavy metal concentrations in soils were minimized by collecting five soil cores combined as a single composite sample. According to the regional consumption practices, the following crops were sampled: rice (*Oryza sativa*), leafy vegetables [including five varieties of Brassica, lettuce (*Lactuca sativa* var. *romana* Gars), celery (*Apium graveolens*), spinach (*Spinacia oleracea*)], and non-leafy vegetables [taro (*Colocasia esculenta*), lotus (*Nelumbo nucifera* Gaertn), radish (*Raphanus sativus*), carrot (*Daucus carota*), aubergine (*Solanum melongena*), tomato (*Lycopersicon esculentum*), and capsicum (*Capsicum annuum*)]. All samples were collected at the normal stage of consumption, taking a representative sample of the crop. The water contents of vegetables are listed in Table 1. Soil and plant samples were collected and stored in polyethylene bags in the field, and transferred to the laboratory as soon as possible for analysis. Vegetables were thoroughly washed with tap water to remove dust and other particles, followed by Mili-Q water. The cleaned plant samples were weighed and separated into edible and nonedible parts, dried in an oven at 80°C to constant weight, then reweighed to determine water content, and finally ground to powder for analysis.

Table 1 The edible part and water content of vegetables

Species	Plant tissue	Water content (%)
Brassica juncea (mustard)	Leaf	91
Brassica oleracea (kale)	Leaf	90
Brassica chinensis (cabbage)	Leaf	94
Brassica rapa (Chinese cabbage)	Leaf	95
Brassica napus (cole)	Leaf	92
Lactuca sativa var. *romana* Gars (lettuce)	Leaf	94
Apium graveolens (celery)	Leaf + straw	90
Spinacia oleracea (spinach)	Leaf	91
Colocasia esculenta (taro)	Root	66
Raphanus sativus (radish)	Root	93
Nelumbo nucifera Gaertn (lotus)	Tuber	86
Daucus carota (carrot)	Root	93
Solanum melongena (aubergine)	Fruit	89
Lycopersicon esculentum (tomato)	Fruit	94
Capsicum annuum (capsicum)	Fruit	92

Chemical analysis

The soil samples were air-dried at room temperature, then pulverized and sieved through a 150-mesh stainless-steel screen. Samples were wet-digested with a concentrated acid mixture (HNO_3, $HClO_4$, and HF). Crop samples were digested with HNO_3 and $HClO_4$ in 5:1 ratio until a transparent solution was obtained (Allen et al. 1986; Markert et al. 1996). The soil and plant digested solutions were cooled to room temperature, filtered, transferred quantitatively to 50 and 25 ml volumetric flasks, respectively, made up to volume with distilled water, and kept in clean plastic vials before metal analysis. The total metal concentrations were determined by flame atomic absorption spectrophotometer (AAS, GBC932AA). Lead and Cd concentrations in leaf tissues were, however, determined using graphite furnace atomic absorption spectrophotometry (GFAAS, GBC932AA).

Quality control

Glassware was properly cleaned, and the reagents were of analytical grade. Double-distilled water was used throughout the study. The accuracy of instrumental methods and analytical procedures were validated using the following certified reference materials (CRMs) obtained from the National Research Center for CRMs (Beijing, China): trace elements in soil (GBW08303), and poplar leaves (GBW07604). Furthermore, each analytical batch contained at least a method blank, and standard solutions were analyzed after every 20 sample solutions as a check on instrument performance. Accuracy of the analytical method was given as percentage recovery for each of the elements. The results are presented in Table 2.

Data analysis

The dietary intakes (EDI) of heavy metals were estimated from the average concentrations of heavy metals in rice (dry weight, DW) and vegetables (fresh weight, FW), and consumption rate of 0.372 kg DW and 0.274 kg FW per day by an adult (60 kg in body weight) reported from a total diet study of Guangdong's rural residents, China (Ma et al. 2005). All statistical analyses were performed using the statistical package SPSS 11.0 for Windows (SPSS Inc., USA). Difference in heavy metal concentrations among different plants was detected using one-way analysis of variance (ANOVA), followed by multiple comparisons using the least significant difference (LSD) test.

Results and discussion

Heavy metals concentration in soils

The selected properties and total metal concentrations of the contaminated soils are presented in Table 3. The paddy soil was acidic (pH 4.97) and the mean pH value of garden soil was slightly acidic (6.07), which indicated all the sampled topsoils to be somewhat to strongly acidic, possibly attributed to continuous

Table 2 Summary of measures of certified reference element concentrations (mg kg^{-1}, mean ± standard deviation (SD), $n = 3$) in CRMs

	GBW08303 ($n = 3$)			GBW07604 ($n = 3$)		
	Certified value	Measured value	Recovery (%)	Certified value	Measured value	Recovery (%)
Cu	120 ± 6	117 ± 0.6	98	9.4 ± 0.5	9.3 ± 1.0	101
Zn	260 ± 11	262 ± 8.9	101	36 ± 6.1	37 ± 3.0	97
Pb	73 ± 2	72 ± 0.5	99	1.47 ± 0.03	1.5 ± 0.3	98
Cd	1.2 ± 0.07	1.1 ± 0.04	98	0.33 ± 0.01	0.32 ± 0.07	103

Table 3 Selected properties and total metal concentrations in soils of Dabaoshan mine area (Guangdong, southern China)

		pH (H_2O)	OM (%)	Total concentration (mg kg^{-1})			
				Cu	Zn	Pb	Cd
Paddy soil ($n = 32$)	Mean	4.97	3.38	502	498	278	3.92
	Range	4.54–5.71	2.73–4.27	276–703	181–1100	153–386	3.0–5.5
	SD	0.45	0.54	142	325	101	1.08
Garden soil ($n = 28$)	Mean	6.07	3.37	271	349	190	3.13
	Range	5.21–7.34	2.38–3.64	102–349	176–696	110–296	1.6–4.9
	SD	0.75	0.55	132	201	79	1.22
Standards values[a]		<6.5		50	200	250	0.3
Background values				35	100	35	0.2

OM organic matter

[a] Grade II of environmental quality standards values for soils of China, GB15618-1995

irrigation of paddy soils with contaminated river water. Organic matter contents of paddy soils were equal to those of garden soils. Average total concentrations of Cu (502 mg kg^{-1}), Zn (498 mg kg^{-1}), Pb (278 mg kg^{-1}), and Cd (3.92 mg kg^{-1}) in paddy soils were found to be always higher than those of garden soils. The Cu, Zn, and Cd concentrations in paddy soil were 10, 2.5, and 13 times, respectively, above grade II of environmental quality standards values for soils of China (MEPPRC 1995). For garden soils, the heavy metal concentrations were significantly higher than the environmental quality standard value and the threshold of natural background in China, except for Pb. The high heavy metal concentrations in the studied area resulted from continuous dispersal downstream from the tailings and waste waters of the large-scale mine and smelter (Zhou et al. 2007). These results corroborate several findings that elevated heavy metals levels in soils were ubiquitous in the vicinity of mines and smelters in South Korea (Chang et al. 2005) and Australia (Kachenko and Singh 2006).

Heavy metal concentrations in rice

The concentrations of Cu, Zn, Pb, and Cd (mg kg^{-1}, dry weight basis) in rice are presented in Fig. 2. The trends of heavy metal concentrations in different parts of the rice plant were in the order: straw > hull > grain. This result was in accord with a report by Fu et al. (2008). According to the maximum recommended levels of contaminants in foods of China (MHPRC 2005), the average concentrations of Pb

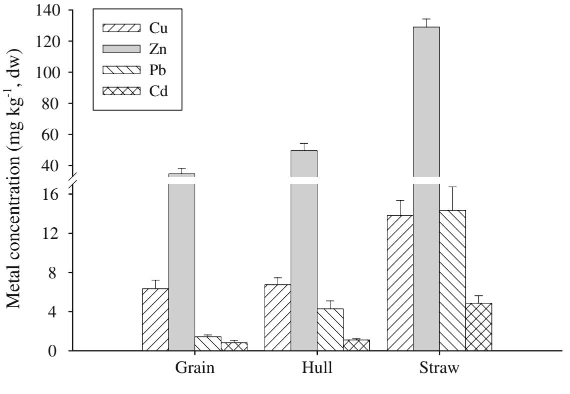

Fig. 2 Metal concentrations (mean ± SD, $n = 18$) in different parts of rice crops grown in the vicinity of Dabaoshan mine

(1.44 mg kg^{-1}) and Cd (0.82 mg kg^{-1}) in rice grains were 7.2 and 4 times, respectively, higher than the permissible values (both 0.2 mg kg^{-1}, dry weight basis) for cereals. Comparing these data with the concentrations of Pb (0.69 mg kg^{-1} DW) and Cd (0.23 mg kg^{-1} DW) in rice contaminated by E-waste (Fu et al. 2008), the local rice grown around the Dabaoshan mine was confirmed to be severely contaminated by the mining operations. The concentrations of Cu (6.34 mg kg^{-1}) and Zn (34.8 mg kg^{-1}) in rice grain were lower than in rice from Chenzhou Pb/Zn mine, China (7.5 mg kg^{-1} and 43 mg kg^{-1}, respectively) (Liu et al. 2005b). In China, the maximum permissible concentrations for Cu and Zn in rice grains are 10 mg kg^{-1} and 50 mg kg^{-1}, respectively (MHPRC 1991, 1994). Cu and Zn concentrations in rice were therefore below the permissible levels.

Heavy metal concentrations in vegetables

The concentrations of Cu, Zn, Pb, and Cd (mg kg^{-1}, dry weight basis) in the edible part of leafy vegetables around Dabaoshan mine are presented in Fig. 3. The average concentrations of heavy metals in leafy vegetable samples were in the descending order: Zn > Cu > Cd > Pb. The concentrations of Cu, Zn, Pb, and Cd were compared with the maximum permissible level (MPL) of contaminants recommended for fresh leaf vegetables in China (MHPRC 1991, 1994, 2005): 10 mg kg^{-1} for Cu, 20 mg kg^{-1} for Zn, 0.3 mg kg^{-1} for Pb, and 0.2 mg kg^{-1} for Cd (fresh weight basis). In general, the concentrations of Cu, Zn, and Pb were within these recommended values. Pb and Cd levels in vegetables of the present study were similar to those reported in vegetables (radish, carrot, leek, potato, etc.) from the abandoned mining area in France (Pruvot et al. 2006). Half of vegetable samples exceeded the MPL for Cd, with the highest (4.1 mg kg^{-1}) in lettuce leaves followed by spinach (3.6 mg kg^{-1}). Similar levels were reported by Kachenko and Singh (2006) in lettuce (0.21 mg kg^{-1} FW) and spinach (0.36 mg kg^{-1} FW) grown on contaminated soils at Boolaroo, Australia. In general, several studies identified that both spinach and lettuce appear to be relatively high accumulators of Cd (Alexander et al. 2006).

Figure 4 shows the concentrations of heavy metals (mg kg^{-1}, dry weight basis) in edible parts of nonleafy vegetables grown on contaminated site.

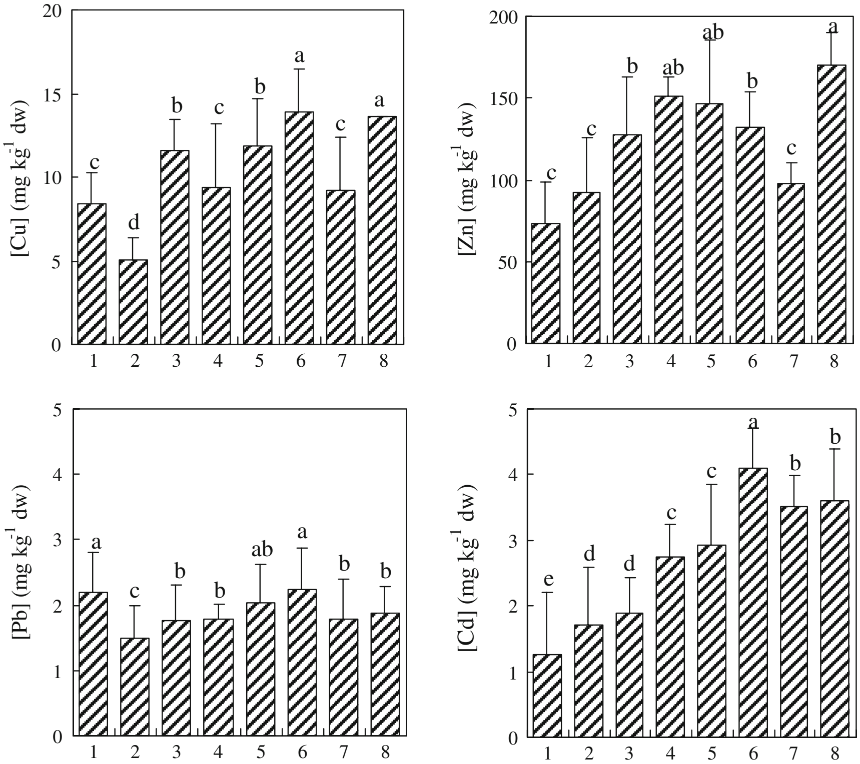

Fig. 3 Heavy metal concentrations in nine subspecies of leafy vegetables (mean ± SD) grown around the Dabaoshan mine. *Lower-case letters* indicate multiple comparisons among different plants, *bars with different letters* are significantly different ($P < 0.05$). *1 Brassica juncea*, *2 Brassica oleracea*, *3 Brassica chinensis*, *4 Brassica rapa*, *5 Brassica parachinensis*, *6 Lactuca sativa* var. *romana* Gars, *7 Apium graveolens*, *8 Spinacia oleracea*

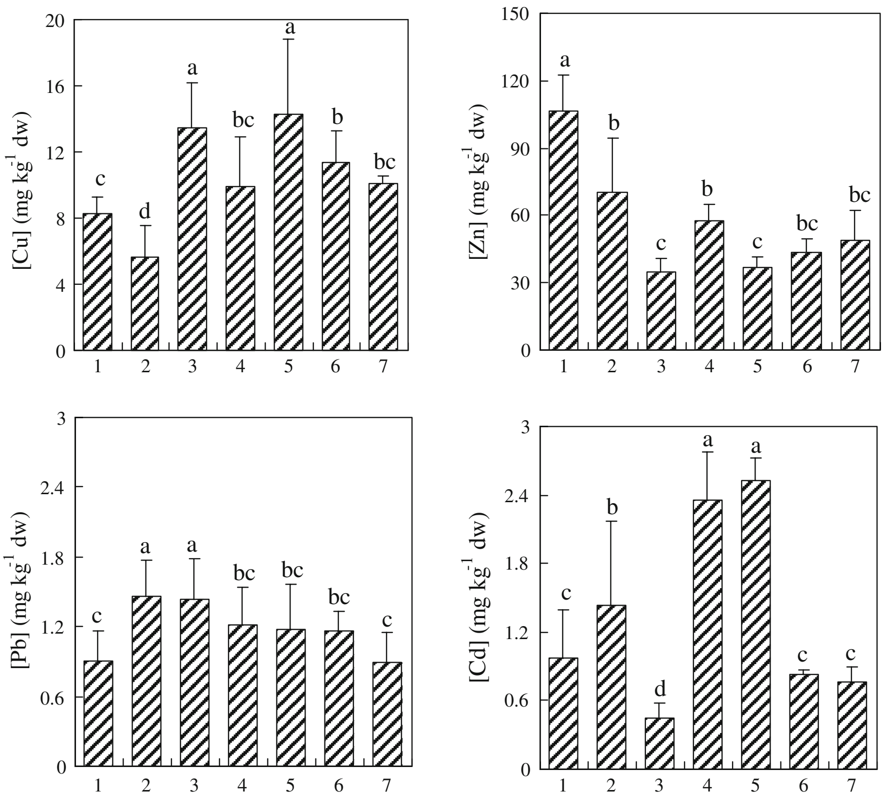

Fig. 4 Heavy metal concentrations in seven subspecies of nonleafy vegetables (mean ± SD) grown around the Dabaoshan mine. *Lower-case letters* indicate multiple comparisons among different plants, *bars with different letters* are significantly different ($P < 0.05$). *1 Colocasia esculenta*, *2 Raphanus sativus*, *3 Nelumbo nucifera* Gaertn, *4 Daucus carota*, *5 Solanum melongena*, *6 Lycopersicon esculentum*, *7 Capsicum annuum*

When compared with the MPL for vegetables regulated in China, the levels of Pb and Cd in most vegetables were higher than the MPL (0.1 mg kg^{-1} FW for both Pb and Cd). Among the tested nonleafy vegetables, taro had significantly high concentrations of Zn and Cd (107 and 0.97 mg kg^{-1}, respectively) in the edible part. The present results were consistent with previous findings by Liu et al. (2005b), who found that taro could accumulate high levels of Zn and Cd in the edible part. Surprisingly, aubergine was strongly associated with high concentrations of Cd (2.52 mg kg^{-1}). The Cd concentrations in vegetables of the present study ranged from 0.45 mg kg^{-1} (in lotus) to 2.52 mg kg^{-1} (in aubergine). The present results are in line with survey reports from Japan that approximately 7% of aubergine and 10% taro contained Cd concentrations above the permissible limit (Arao et al. 2008). No evidence of phytotoxicity of Cu and Zn in the nonleafy vegetables analyzed could be found from the concentrations of Cu and Zn in the food crops grown in the vicinity of Dabaoshan mine.

Dietary intake of heavy metal

Local inhabitants who consume rice and vegetables grown in the Dabaoshan mine are exposed to heavy metal contamination. Comparison of estimated daily intake (EDI) with tolerable daily intake (TDI) for Cu, Zn, Pb, and Cd is presented in Table 4. Exposure to contaminants and related health risks were expressed in terms of TDI, a reference value established by the Food and Agriculture Organization (FAO)/World Health Organization (WHO) expert committee, which jointly proposed that the recommended TDI limits of Pb and Cd be 3.6 μg kg^{-1} day^{-1} and 1 μg kg^{-1} day^{-1}, respectively (WHO 1993). Based on the status of metal levels in rice grain, the EDI of Pb (8.9 μg kg^{-1} day^{-1}) and Cd (5.1 μg kg^{-1} day^{-1}) were remarkably above the TDI limits, indicating that exposure to Pb and Cd was remarkably high. The present study showed that the average EDIs in rice grain from Dabaoshan mine were similar to the values observed in Chenzhou Pb/Zn mine, China (Liu et al. 2005b). From consumption of vegetables, the average values of EDI were 4.72 μg kg^{-1} day^{-1} for Cu, 43 μg kg^{-1} day^{-1} for Zn, 0.66 μg kg^{-1} day^{-1} for Pb, and 0.85 μg kg^{-1} day^{-1} for Cd (Table 4). Although the average metal EDIs of vegetables were less than the TDI limits, the contribution of vegetables to the intake of Cd of local inhabitants may pose potential health risk due to high Cd level in more than half of crop species. Moreover, no adverse effects were thus expected from Cu and Zn ingestion via consumption of rice and vegetables grown in the vicinity of the Dabaoshan mine. Furthermore, as rice is a common food consumed daily in China, heavy metals in rice may contribute substantially to the total daily intake estimated, taking precedence in vegetable consumption.

Cadmium and Pb are nonessential elements and their presence even at very low concentration causes adverse health effects to human health (Mahaffey 1990). Dietary cadmium accumulates principally in the kidneys and liver (McLaughlin et al. 1999; Muchuweti et al. 2006). The present results indicate that there is a serious potential health risk associated with Cd and Pb in rice and vegetables that threatens inhabitants in the vicinity of Dabaoshan mine. Similar results were reported by Cui et al. (2004),

Table 4 Average concentrations of heavy metals in rice and vegetables (mg kg^{-1} DW) and the estimated dietary intake (EDI) by an adult (60 kg body wt) from Dabaoshan mine relative to the Tolerable dietary intake (TDI)

Metals	Average concentrations		EDI (μg kg^{-1} day^{-1})		TDI (μg kg^{-1} day^{-1})
	Rice	Vegetables	Rice	Vegetables	
Cu	6.34	10.40	39	4.72	167[a]
Zn	34.8	89.00	216	43	667[a]
Pb	1.44	1.53	8.9	0.66	3.6[b]
Cd	0.82	1.99	5.1	0.85	1.0[b]

[a] Food and Nutrition Board, Institute of Medicine. (2004). *Dietary reference intakes (DRI): tolerable upper intake levels, elements*. National Academy of Sciences

[b] WHO (1993)

who suggested that local inhabitants near a smelter in Nanning, China, were exposed to Pb and Cd via consumption of vegetables, although no risk was found for Cu and Zn. Therefore, the status of heavy metal contamination of food crops around mine regions and the implications for human health should be identified urgently by more detailed study.

Conclusions

Soils and food crops were contaminated with Pb and Cd in the vicinity of Dabaoshan mine (Guangdong, southern China). The concentrations of heavy metals in the soils exceed the corresponding maximum allowable concentration levels for agricultural soils in China. Rice grain could accumulate unacceptably high concentrations of Pb and Cd. The estimated dietary intakes of Pb and Cd from food crops grown in the contaminated soils exceeded the tolerable daily intake limits. Rice and vegetables consumed by the local inhabitants in the vicinity of Dabaoshan mine could thus expose them to dangerous levels of Pb and Cd. Therefore, the crops grown in the vicinity of the Dabaoshan mine present a potentially serious health risk to the local inhabitants.

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