ORIGINAL PAPER

Mercury contamination and health risk to crops around the zinc smelting plant in Huludao City, northeastern China

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Received: 2 June 2006 / Accepted: 19 January 2007 / Published online: 13 March 2007 Springer Science+Business Media B.V. 2007

Abstract The Huludao zinc plant in Liaoning province, northeast China was the largest in Asia, and its smelting activities had seriously contaminated soil, water and atmosphere in the surrounding area. For the first time, we investigated the total mercury (THg) content in maize, soybean, broomcorn, 22 vegetables, and the soil around their roots from eight sampling plots near the Huludao zinc plant. THg contents of the seeds of maize, soybean, and broomcorn are 0.008, 0.006, and 0.057 mg kg^{-1} , respectively, with the broomcorn being the highest, exceeding the maximum level of contaminant in food (GB2762-2005) by 4.7 times. The edible parts of vegetables are also contaminated with a range of mercury contents of $0.001-0.147$ mg kg⁻¹ (dry weight). THg contents in plant tissue decrease in the order of leaves $>$ root $>$ stalk $>$ grain. Using

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correlation analysis, we show that mercury in the roots of these plants is mainly derived from soil, and the uptake of gaseous mercury is the predominant path by which the mercury accumulated in the foliage. The average and maximum mercury daily intake (DI) of adult around the Huludao zinc plant via consuming vegetables are 0.015 and 0.051 μ g/kg/d, respectively, and those of children are 0.02 and $0.07 \mu g/kg/d$, respectively. The average and maximum weekly intakes of total mercury for adult are 2.1 and 7.1%, respectively, of the provisional tolerable weekly intake (PTWI), and 2.8 and 9.7%, respectively, of the PTWI for children.

Keywords Vegetable Crop Mercury Zinc smelting

Introduction

Human activities, including the combustion of fossil fuels and the incineration of the waste, gold mining, and intentional use of mercury, have significantly increased the emission of mercury into the atmosphere, making it a global pollutant. The global mercury assessment (UNEP, [2002](#page-8-0)) reported that the mercury content in global atmosphere had increased by three times since the industrial revolution, and much more around the industrial areas. Mercury contamination seriously threatened ecosystems and human health. Numerous recent studies have concluded that the principal human methlymercury intake was the consumption of contaminated fish, while amalgam dental filling was generally the main source of total Hg in Western countries. However, Horvat et al. [\(2003](#page-7-0)) discovered that the consumption of mercury and methylmercury from contaminated rice could also affect human health. Therefore, the knowledge of the consumption of other mercury-contaminated food was needed to assess its potential impact on human health.

Zinc smelting is regarded as one of the most important anthropogenic mercury emission sources (Nriagu & Pacyna, [1988](#page-8-0)). During the zinc smelting process, almost all mercury in the zinc ores are evaporated from the matrix, and eventually go into the atmosphere, if no pollution control technology is applied (Pacyna & Pacyna, [2002\)](#page-8-0). Mercury emissions from zinc smelting in many developing countries are not well controlled. China is one of the largest zinc producers worldwide, with the Huludao zinc plant in Liaoning, China being the largest in Asia. About 260 tons of mercury was emitted into the atmosphere from the Huludao zinc plant in the decade from 1980 to 1990 (Dong, [1988](#page-7-0)). Zinc smelting activities at the Huludao zinc plant had seriously contaminated soil, water and atmosphere in the surrounding area (Wang, Yan, & Zhang, [2005a;](#page-8-0) Wang, Sato, Xing, & Tao, [2005b](#page-8-0)). There have been previous reports on mercury contamination to the local ecosystem from other point sources such as artisanal gold mining (Egler, Rodrigues, Villas, & Beinhoff, [2006](#page-7-0)), a thermometer factory (Krishna, Karunasagar, & Arunachalam, [2003\)](#page-7-0), and mercury contamination to soil from artisanal zinc smelting using indigenous methods in Hezhang county, Guizhou, China (Feng, Li, & Qui, [2006](#page-7-0)). However, reports on mercury contaminations to vegetables and crops from zinc smelting plant have been scant or nonexistent. Here, we reported the result of such an investigation for the Huludao zinc plant in Liaoning, China. We deduced the mercury contamination in 22 vegetables and three crops in the neighboring area, discussed the mercury distribution characteristics and the possible sources of mercury in these vegetables and crops, and made an assessment of the health risk to the local residents around the Huludao zinc plant from consuming vegetables.

Methods

Study area

Huludao city $(40^{\circ}56)'$ N, $120^{\circ}28'$ E) lies on the west coastland of Liaoning Province in China. Its climate represents a typical continental monsoon with an average annual temperature of 8.7° C and an average annual rainfall of 590 mm. The primary wind direction is from northeast to southwest. Petrochemicals, nonferrous metal smelting, shipbuilding, and power generation are the primary industries of Huludao city. The Huludao zinc plant is situated at the south east of Huludao City, near the Bohai Gulf (as shown in Fig. [1\)](#page-2-0), with an area of 80,000 m^2 , and produces 330,000 t of zinc every year. In this study, vegetables were collected from areas marked as A, B, D, E, G, and H and crops from areas A, C, and F, shown in Fig. [1.](#page-2-0)

Sampling and pretreatment

In Oct 2005, 22 species of vegetables and maize, soybean, broomcorn cultivated popularly around the Huludao zinc plant were collected, along with soil samples around their roots. The total sampling number of vegetables and crops reached 187. Five to seven individual plants were collected with 5×5 m at each sampling site. The fresh vegetable samples were put in clean plastic bags and transported to the laboratory for sample treatment as soon as possible. The fresh weights (FW) of the samples were recorded. After being cleaned with Milli-Q water, each individual vegetable or crop was separated into root, stalk, leaf and fruit subsamples. All subsamples were air-dried, and the dried samples were weighed again and the dry weight (DW) recorded, and then ground to a fine powder which could pass the 0.18 mm-mesh sieves in a precleaned food blender, and stored in polythene zip-bags. The blender was extensively cleaned with Mili-Q water and ethanol to prevent any cross contamination.

Five to seven soil samples of the surface layer (a little deeper than the root concentrated layer,

 \leq 15 cm) were collected at each sampling site. Soil samples were air dried, crushed, and passed through a 0.18 mm-mesh sieve and stored at ambient temperature before analysis of the concentration of mercury.

Mercury analysis and quality control

The soil and plant samples were digested using the method of H_2SO_4 –HNO₃–V₂O₅ described by Rasmussen, Mierle, and Nriagu ([1991,](#page-8-0) p. 379). Total mercury (THg) concentration was determined using BrCl oxidation and $SnCl₂$ reduction couple with cold-vapor atomic absorption spectrometry (CVAAS). A standard reference material of soil sample [GBW 07405(GSS-5)] of 0.29 ± 0.03 mg kg⁻¹ and poplar leaf [GBW 07604(GSV-3)] of 0.026 \pm 0.003 mg kg⁻¹ provided by the China National Center for Standard Materials were used as quality control references. The recoveries of Hg in the soil and plant samples were in the ranges 94.1–102.3% and 91.7–108.31%, respectively, with the range of the certified Hg concentration. Repeated measurements of plant samples showed that the error of our method was less than 5%.

Calculation of oral intake of metals from vegetables

The daily oral intake of metals from soil through vegetables was calculated by the following:

Daily intake of mercury (DI) (μ g/kg body weight bw \langle day \rangle = concentration of mercury in vegetable (μ g g⁻¹, fresh weight) \times mean vegetable consumption (g/person/day) /reference body weight.

Weekly intake (WI) of mercury (μ g/kg bw / day) = Daily intake of mercury $(DI) \times 7$ days/ week

The World Health Organization (WHO) has established a provisional tolerable weekly intake

(PTWI) of 5 μ g/kg body weight (bw)/week for total mercury. The PTWI represents permissible human weekly exposure to those contaminants associated with the consumption of otherwise wholesome and/or nutritious foods. The term provisional expresses the tentative nature of the Joint FAO/WHO Committee on Food Additive's (JECFA) evaluation and tolerable signifies permissibility rather than acceptability for the intake of trace contaminants that have no necessary functions in food, in contrast to those of pesticides or food additives permitted for use on foods (UNEP/FAO/WHO, [1992\)](#page-8-0).

Statistical analysis

The data were statistically analyzed using the statistical packages SPSS 10.0 (SPSS Inc., USA) and Origin 7.0 (Origin Inc., USA). The measures were expressed in terms of a geometric mean when necessary. Statistical significance was computed by analysis of variance (ANOVA). The level of significance was set at $p < .05$ (two-tailed).

Results and discussion

THg in the edible part of vegetable

THg content in vegetables collected from the sampling plots are listed in Table [1.](#page-4-0) The vegetables could be classified into leafy, solanaceous fruit, leguminous, gourd, and tuberous vegetables according to their edible parts. It was accepted that the edible parts of leafy vegetable were the leaves, those of solanaceous fruit, gourd vegetables were fruits, those of leguminous vegetable were the pod and seed, and those of tuberous vegetables were the tubers. THg contents in vegetables decrease in the order of leafy, solanaceous fruit, tuberous, gourd, and leguminous vegetables. Spinach in the leafy vegetables, balsam pear in the gourd vegetables, and radish in the tuberous vegetables accumulated large amount of mercury, exceeding the levels from other vegetables. The differences in mercury contents in vegetables show that the vegetables have different absorbabilities of mercury.

THg in vegetables in other areas is quite small compared to that in the area surrounding the Huludao zinc plant (Table [2\)](#page-4-0). THg in capsicum around the Huludao zinc plant was more than in Lanmuchang, Guizhou province (Xiao, Guho, Boyle, Liu, & Chen, [2004](#page-8-0)), and the THg in leek and lettuce was more than in Thessaloniki, N. Greece (Voutsa & Samara, [1998\)](#page-8-0), although the THg in shallot was less than that in the Tapajós gold mine in Brazil (Egler et al. 2006). It is suggested that most vegetables around the Huludao zinc plant have been seriously contaminated by mercury.

THg distribution in the tissues of vegetable

Eight popular vegetables in the sampling plot were selected to investigate the mercury distribution in the tissue of vegetables. Figure [2](#page-5-0) shows that THg contents in the leaves of leek and rutabaga are higher than in the roots; THg contents in the leaves of kidney bean, carrot and potato are higher than those in the fruits; and THg content in the tissues of eggplant decreases in the order of leaf $>$ root $>$ stalk $>$ fruit, with tomato and capsicum having the similar trend. THg in the leaves of most vegetables is generally higher than that in the other tissues. The accumulation of mercury in the tissues of vegetables depends on the adsorbability of the tissues to mercury.

Mercury distribution in maize (Zea mays L.), soybean (Glycine max, L.) and broomcorn (Sorghum bicolor L)

The statistical summaries of THg concentrations inthe different tissues of maize, soybean, broomcorn collected in the studied area are shown in Fig. [3](#page-5-0). This shows that the THg content in the three crops increases in the order of $maize <$ soybean $<$ broomcorn with THg contents in the tissues of each individual crop varying asleaf $>$ root $>$ stalk $>$ grain. THg contents of the seeds of maize, soybean, and broomcorn are 0.008, 0.006, 0.057 mg kg^{-1} , respectively, with the broomcorn being the highest, exceeding the maximum level of contaminant in food (GB2762- 2005) by 4.7 times. The THg content in the root,

Kind	The vegetable	Scientific name (sample number)	The range of THg content (average) (mg kg^{-1})	Sd
Leguminous vegetable	Kidney bean	Phaseolus vulgaris L. (13)	$0.01 - 0.109$ (0.028)	0.024
	Long bean	<i>Vigna sinensis</i> (L.) Savi (3)	$0.012 - 0.032$ (0.013)	0.015
Gourd vegetable	Cushaw	Cucurbita moschata (Duch.) poiret. (5)	$0.012 - 0.059$ (0.036)	0.010
	Towel gourd	Luffa cylindrical(L.) Roem (4)	$0.005 - 0.0012$ (0.001)	0.002
	Pumpkin	Cucurbita pepo L. (5)	$0.004 - 0.031(0.017)$	0.014
	Balsam pear	Momordica charantia L. (3)	$0.01 - 0.134(0.11)$	0.021
Solanaceous fruit	Eggplant	Aolanum melongena L. (10)	$0.004 - 0.059$ (0.027)	0.013
vegetable	Tomato	Lycopersicon esculentum Mill. (11)	$0.009 - 0.074$ (0.041)	0.021
	Capsicum	Capsicum annuum L. var. grossum(L.) sendt (17)	$0.006 - 0.095(0.047)$	0.019
Leafy vegetable	Celery	Apium graveolens L. (7)	$0.025 - 0.069$ (0.039)	0.027
	Chinese cabbage	<i>Brassica pekinensis</i> (Lour.) Rupr. (11)	$0.025 - 0.13(0.079)$	0.021
	Leek	Allium tuberosum Rottl.ex Spreng. (5)	$0.032 - 0.18(0.11)$	0.038
	Shallot	Allium Fistulosum L. (6)	$0.088 - 0.18(0.13)$	0.032
	Lettuce	Lactuca sativa L. var. romana Hort (8)	$0.027 - 0.12$ (0.08)	0.015
	Spinach	Spinacia oleracea L. (4)	$0.084 - 0.19$ (0.15)	0.029
	Cole	<i>Brassica chinensis L.</i> (6)	$0.054 - 0.13(0.10)$	0.012
	Cilantro	Coriandrum sativum L. (4)	$0.005 - 0.078$ (0.050)	0.010
	Rutabaga	Brassica napobrassica(L.)Mill (3)	$0.004 - 0.091$ (0.051)	0.027
	Mustard	Brassica juncea(L.) czern.et coss.var. multiceps Tsen et Lee (3)	$0.038 - 0.051$ (0.043)	0.009
Tuberous vegetable Carrot		Daucus carota L. var. sativa Hoffm (5)	$0.009 - 0.019(0.012)$	0.004
	Radish	Rhaphanus sativus L. (7)	$0.005 - 0.112(0.081)$	0.016
	Potato	Ipomaea batatas (L.) Lam (3)	$0.004 - 0.019$ (0.011)	0.007
Leguminous vegetable			$0.01 - 0.109(0.021)$	0.01
Gourd vegetable			$0.005 - 0.13$ (0.042)	0.049
Solanaceous fruit vegetable			$0.004 - 0.095(0.038)$	0.010
Leafy vegetable			$0.004 - 0.195(0.082)$	0.038
Tuberous vegetable			$0.004 - 0.134(0.035)$	0.040
Total			$0.005 - 0.195(0.057)$	0.041

Table 1 Mercury content of vegetable in Huludao City (dry weight)

Table 2 Comparison of mercury content of vegetable in different areas (dry weight)

Site	The total mercury content in vegetable(mg·kg ⁻¹)						
	Capsicum	Chinese cabbage	Leek	Shallot	Lettuce	Carrot	Reference
Huludao Zinc plant	$0.006 - 0.10$ $0.02 - 0.13$					$0.03-0.18$ 0.09-0.18 0.03-0.12 0.009-0.02 This study	
Lanmuchang in Guizhou	$0.01 - 0.04$ $0.09 - 0.70$						Xiao et al. 2004
Thessaloniki, N. in Greece			$0.01 - 0.07$		$0.01 - 0.08$		$0.01 - 0.09$ Voutsa and Samara 1998
Tapajós gold mine in Brazil				$0.06 - 0.99$			Egler et al. 2006

stalk, leaf, and grain of broomcorn are, respectively, $3.2\times$, $4.5\times$, $2.8\times$, and $12.3\times$ as high as those of the soybean, and $4.8\times$, $10.3\times$, $4.1\times$, and $16.3\times$ as high as those of maize. Thus broomcorn is more effective in the accumulation of mercury than maize and soybean. Zhao and Yan ([1997](#page-8-0)) reported that Hg content in the maize around the Huludao zinc plant was less than that in the

Fig. 2 Total mercury distribution in tissues of vegetable

Fig. 3 Total mercury distribution in the tissues of crops

broomcorn, consistent with our current findings. THg contents in root, stalk, leaf, grain of maize in this study are $10\times$ as high as those in the Sanjiang plain (Liu, Wang, Lv, Li, & Wang, [2004\)](#page-8-0), and higher than those in the crops irrigated with mixtures of wastewater and sewage sludge in Zimbabwe (Muchuweti et al., [2006\)](#page-8-0) and those in the crops irrigated around a large-scale municipal solid-waste incineration plant in Shanghai city (Tang et al., [2005\)](#page-8-0).

THg content in the top soil

Table 3 summarizes the THg concentrations in the topsoils near the Huludao zinc plant. The

Table 3 Total mercury content in top soil of agriculture land (mg kg^{-1} , dry weight)

Sampling sites	Minimum	Maximum	Average	SD
A	0.206	0.248	0.227	0.02
B	2.462	3.797	3.129	0.67
C	0.667	0.873	0.770	0.10
D	2.272	2.792	2.532	0.26
E	0.425	0.674	0.536	0.12
$\mathbf F$	1.035	1.175	1.105	0.07
G	0.173	0.466	0.319	0.15
H	0.110	0.146	0.128	0.02
Total	0.110	3.797	1.28	1.12

topsoils of agriculture land are seriously contaminated with mercury compared to the local background mercury concentration $(0.037 \text{ mg kg}^{-1})$ in Liaoning, China), and the average mercury concentration is 1.28 mg kg⁻¹with a range of 0.110–3.797 mg kg^{-1} , significantly larger than that in Hezhang, Guizhou, China (contaminated by Hg) and other areas (Table 4).

Mercury source in vegetables and crops

The THg distribution in the tissues of vegetables and crops found in this study has the following characteristics: leaf $>$ root $>$ stalk $>$ grain (fruit). Feng et al. ([2006\)](#page-7-0) reported a similar distribution feature of mercury in tissues of maize, which was

Table 4 Comparison of mercury content in topsoil from different areas

Site	THg content in top soil of agriculture land $(mg kg^{-1}, dry)$ weight)	Reference
Huludao zinc plant Hezhang, in Guizhou, China Naboc river area, Mindanao, Philippines	0.13–3.37 $0.14 - 0.86$ $0.08 - 0.99$	This study Feng et al. 2006 Appleton, Weeks. Calvez, and Beinhoff 2006
Rwamagasa artisanal gold mining centre, Geita district, Tanzania	$0.05 - 9.2$	Taylor et al. 2005

contaminated by zinc smelting. However, Liu et al. [\(2004](#page-8-0), p. 289) found that the THg content in the tissues of maize cultivated in the Sanjiang plain had features as follows: root > leaf > stalk $>$ grain. The THg distribution in the tissues of vegetable around the Huludao zinc plant also increases as follows: leaf > root > stalk > fruit. From the perspective of the Hg source, one factor was considered important: the gaseous Hg in atmosphere could be absorbed and accumulated in the leaves of vegetables and crops, leading to a higher Hg content in leaves than in roots. Hg content in plant leaves is strongly correlated with the Hg content in the atmosphere (Wang, Mou, & Qing, [1998\)](#page-8-0). Research showed that a large amount of gaseous mercury was emitted into the atmosphere around the Huludao zinc plant with a Hg vapor concentration in the atmosphere of 0.005–0.008 mg/m³> (Zhao & Yan, [1997](#page-8-0)), significantly larger than that of Sanjiang plain (not contaminated by Hg) and other areas (Table 5). Moreover, the THg contents in the soil and in the roots of maize correlate linearly with a correlation coefficient of 0.8159 ($p < .01$, $n = 11$), but such a correlation between the Hg content in the soil and in the leaves of maize is poor, with a correlation coefficient of 0.5743 ($p > .1$, $n = 11$). This suggests that mercury in the root is mainly derived from the soil, whereas mercury in the leaves might have other sources in addition to the

Table 5 Comparison of mercury content of atmosphere in different areas

Site	THg in the atmosphere $(ng \text{ m}^{-3})$	Reference
Huludao zinc plant	5000–8000	Zhao and Yan 1997
Changchun, China	$4.7 - 79$	Li and Wang 2003
Sanjiang plain	68–97	Liu et al. 2004
Hezhang, Guizhou, China	$30 - 413$	Feng et al. 2006
Wanshan, in Guizhou, China	500-1500	Horvat et al. 2003
Lichwin, Poland	1.28–1.98	Zielonka, Hlawiczka, Fudala, Wängberg, and Munthe 2005

soil. It is believed that mercury from the atmosphere is accumulated in the above-ground part of the crops and vegetables, whereas the mercury from the soil is accumulated in the roots, in agreement with the report of Wang and Mou ([1999,](#page-8-0) p. 30). Based on this argument, we believe that mercury in the leaves of maize is mainly derived from atmosphere.

Health risk from consumption of vegetables

The daily ingestion rates of vegetables for adult and children are 301 and 231.5 g/person/day (Wang et al., [2005a](#page-8-0), [b\)](#page-8-0). The average and maximum mercury daily intakes (DI) of adults are 0.015, 0.051 μ g/kg/d and of children are 0.02, 0.07 μ g/kg/d, respectively (Table 6), exceeding the daily intake of adult and child in Tianjin (Wang et al., [2005,](#page-8-0) [b](#page-8-0)) and Korea (Lee et al., [2006\)](#page-8-0). The average and maximum mercury weekly intakes (WI) of adults are 0.105, $0.357 \mu g/kg$ week and of children are 0.14, 0.49 μ g/kg/week, respectively. Mercury could be ingested by a person through many pathways. The average and maximum weekly intake of total mercury for an adult only by consuming the vegetables around the Huludao zinc plant are 2.1 and 7.1%, respectively and 6.3 and 8.3%, respectively, for a child. The adverse effect to a child of consuming contaminated vegetables is more than

Table 6 Health risk analysis of vegetable to the local villagers

		Adult	Child
Vegetable		301.4	231.5
intake			
(g/person/			
day)			
Reference		55.9	32.7
weight (kg)			
Daily intake	Average	0.015	0.020
$(\mu$ g/kg body	Range	$0.00013 - 0.051$	$0.00018 - 0.070$
weight/day)			
Weekly intake	Average	0.11	0.14
$(\mu$ g/kg body	Range	$0.00091 - 0.357$	$0.012 - 0.49$
weight/week)			
PTWI $(\mu g/kg)$ week)		5	5
Weekly intake /	Average	2.1	2.8
$PTWI(\%)$	Maximum	7.1	9.7

that of an adult. Therefore, we strongly suggest that local residents should avoid eating the vegetables around the Huludao zinc plant in order to reduce this health risk.

Environmental assessment of mercury in crops

Maize, soybean, and broomcorn are not primary food staples ingested by the local residents. Rice, the primary food, is not cultivated around the Huludao zinc plant. Therefore, the health risk analysis of consumption of maize, soybean, and broomcorn was not investigated. However, the THg content in the roots, stalks, and leaves of maize were $3\times$, $14\times$, and $22\times$ as high as in grain, respectively. The THg contents in the roots, stalks and leaves of soybean were $10\times$, $28\times$, and $43\times$ as high as in grain, respectively. The THg contents in the roots, stalks and leaves of broomcorn were $10\times$, 28 \times , and 43 \times as high as in grain. Most of the mercury in the crops from the environment through stomata or other points of entry is accumulated in the inedible part of the crops (Wind-ham, Weis, & Weis, [2003\)](#page-8-0), whereas Hg in the edible part is less than in the inedible part of crops due to wrapping of leaves or pods. The local villagers often use the inedible part of the crops as fuel instead of coal, contributing to Hg concentration in the atmosphere. The THg contents in the leaves of maize, soybean were similar to the Hg content of coal that was produced in Liaoning province (0.2 mg kg^{-1}) (Wang, Kang, & Chen, [1996\)](#page-8-0), and that of broomcorn was $3.5\times$ as high as the THg content of coal. As a result, the burning of the inedible parts of mercury-contaminated crops causes worse pollution. So other energy source should be taken into account instead of coal and the inedible parts of crops. In addition, the local villagers usually took the root, stalk, leaves, grain of crop as fodder for livestock, so Hg might be transferred in this way through the food chain to the local villages. The contaminated crop would then affect the health of the local villagers indirectly.

Conclusion

The results indicated that maize, soybean, broomcorn and 22 vegetables from eight sampling plots around the Huludao zinc plant were seriously contaminated by Hg. The THg content in vegetables decreased in the order: leafy, solanaceous fruit, tuberous, gourd leguminous vegetables. THg in the inedible part of broomcorn is higher than that in maize and soybean. The THg content in plant tissues decrease in the order: leaves > root $>$ stalk $>$ grain. Using the correlation analysis, we show that the Hg in the root of plant is mainly derived from the soil, and the uptake of gaseous Hg is the predominant pathway by which Hg accumulates in the foliage. The average and maximum weekly intake of total mercury for an adult only by consuming the vegetable around the Huludao zinc plant are 2.1% and 7.1%, and 2.8% and 9.7% for a child. Other uses of crops could also contaminate the local environment and affect the health of local villagers indirectly.

Acknowledgements The authors would like to acknowledge the support of the Knowledge Innovation Project of the Chinese Academy of Sciences (KZCX3-SW-437). We would like to thank Prof. Fuke Yi from the Northeast Institute of Geography and Agricultural Ecology, the Chinese Academy of Sciences and Dr. Zhongzhu Liang from Jilin University for their help with this paper.

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