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ORIGINAL PAPER

Residents health risk of Pb, Cd and Cu exposure to street dust based on different particle sizes around zinc smelting plant, Northeast of China

Qiuhong Zhou · Na Zheng · Jingshuang Liu · Yang Wang · Chongyu Sun · Qiang Liu · Heng Wang · Jingjing Zhang

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Abstract The residents health risk of Pb, Cd and Cu exposure to street dust with different particle sizes (<100 and <63 μm) near Huludao Zinc Plant (HZP) was investigated in this study. The average concentrations of Pb, Cd and Cu in the <100-μm and <63-μm dust were 1,559, 178.5, 917.9 and 2,099, 198.4, 1,038 mg kg^{-1}, respectively. It showed that smaller particles tended to contain higher element concentrations. Metals in dust around HZP decreased gradually from the zinc smelter to west and east directions. There was significantly positive correlation among Pb, Cd and Cu in street dust with different particle sizes. The contents of Pb, Cd and Cu in dust increased with decreasing pH or increasing organic matter. Non-carcinogenic health risk assessment showed that the health index (HI) for children and adult exposed to <63-μm particles were higher than exposed to <100-μm particles, which indicated that smaller particles tend to have higher non-carcinogenic health risk. Non-carcinogenic risk of Pb was the highest in both particle sizes, followed by Cd and Cu. HI for Pb and Cd in both particle sizes for children had exceeded the acceptable value, indicated that children living around HZP were experiencing the non-carcinogenic health risk from Pb and Cd exposure to street dust.

Keywords Street dust · Health risk assessment · Heavy metal · Zinc smelting · Particle size

Q. Zhou · N. Zheng (✉) · J. Liu · Y. Wang · C. Sun · Q. Liu · H. Wang · J. Zhang
Northeast Institute of Geography and Agricultural Ecology, Chinese Academy of Sciences, Shengbei Street 4888#, Changchun City 130102, Jilin, China
e-mail: zhengnalzz@neigae.ac.cn

Q. Zhou · C. Sun · Q. Liu · H. Wang · J. Zhang
University of Chinese Academy of Science, Beijing 100049, China

Introduction

Street dust containing large amount of heavy metals affects both the urban environmental quality and the human health (Charlesworth et al. 2010; Hu et al. 2011; Shi et al. 2013). Street dust has adverse impacts on human health through heavy metal intake from the liable floating dust by ingestion, inhalation and dermal contacts (Shi et al. 2011; Faiz et al. 2012; Reis et al. 2014). Specifically, child is more liable to suffer the harmful effects from dust pollutants for their lower immunity and more hand-mouth activities (Zheng et al. 2010b; Lu et al. 2014; Chen et al. 2014a). Street dust comprises various heavy metals including Pb, Cd and Cu. Infant exposure to Pb may have highly toxic influences on nervous system and thus reduce children's intelligence (Han et al. 2011; Cao et al. 2014; El-Desoky et al. 2014), while the effects on adults exhibit the same symptoms at a higher blood lead level (Gracia and Snodgrass 2007). The Itai-itai disease is caused by excess cadmium intake. Human liver is a primary target organ of copper-induced toxicity, and excessive copper intake also induces toxicity indirectly by interacting with other nutrients, for example, excess copper intake produces anemia by interfering with iron transport and/or metabolism (Stern et al. 2007). Furthermore, foods, air and drinking water may be contaminated by heavy metals in street dust and thus affect indirectly urban environment quality and human health. Consequently, research on heavy metals in street dust is becoming a hot spot around the world, which mainly studies the concentration, source identification and spatial distribution (Gunawardana et al. 2011; Atiemo et al. 2012; Liu et al. 2014; Huang et al. 2014, Qiao et al. 2014; Yoshinaga et al. 2014; Chen et al. 2014b; Zibret et al. 2013).

Particle size was found to have a significant influence on risk assessment of human exposure to toxic chemicals in dust (Cao et al. 2012). Previous studies reported that the concentration of toxic metal in street dust generally increases as particle size decreases, for increasing specific surface area (Tanner et al. 2008; Mercier et al. 2011; Zhao et al. 2014). Fine particles of particle size <100 μm is liable to suspend when winds up or vehicular traffic moves and thus enter into human (mouth or nose) through breath. Thoracic dust (particle size <10 μm) could enter into lungs via nose and throat (Shi et al. 2011). Some scholars proposed that when sampling dust, more attention should be paid to particle sizes less than 100 μm (Cao et al. 2012). Particles in the <63 μm fraction is dominant of the total metal loading (Zhu et al. 2008). Xu et al. (2012) turned out that <63 μm fraction has higher As concentrating than other fractions. Fine particles adhere better to human's hands or skin than larger particles (Fang et al. 2013). Choate et al. (2006) concluded that only the soil particles with diameters less than about 63 μm adhere to human's skin. So except for <100 μm dust, particle with diameter <63 μm is also chosen as our research object.

The objectives of the present study were as follows: (a) distribution of Pb, Cd and Cu in different particle sizes (<100 and < 63 μm), (b) effects of pH and organic matter to metals accumulation and (c) assessment of population health risk due to Pb, Cd and Cu exposure from <100-μm street dust and <63-μm dust.

Materials and methods

Sampling and preparation

Study area

As the largest zinc plant in Asia, Huludao Zinc Plant (HZP) is a state enterprise of non-ferrous metal smelter. It is near the Liaodong Gulf which situated at the southeast of Huludao (40°56′N, 120°28′E) and lies to the west coastland of Liaoning Province in China. This area belongs to the temperate continental monsoon climate with the annual average temperature 8.7° Cand the annual average rainfall of 590 mm. As atmosphere circulation, northeaster winds prevailed in the autumn and the winter, while the southwester prevailing in the summer of the study area. The soil type is mainly brown soil. The annual production capacity of non-ferrous metals was 430,000 tons in 2006, including 330,000 tons of zinc, 600 tons of cadmium. It has 13 smoke stacks higher than 60 m and six smoke stacks higher than 100 m (Lu et al. 2010). Smoke and dust discharged from HZP contains organic and inorganic pollutants, which will eventually deposited on the land surface around the HZP.

Sampling

A total of 72 samples of street dust from 28 sites were collected along the Jinhu Road and the Jinhubei Road near the south of HZP under stable weather conditions in September 2011. During the sampling days, it is sunny since 1 week before. The distance between HZP and sample sites was within 2.5 km. Each path between the two streets set up one sample site (Fig. 1). About 500–1,000 g sample was collected by sweeping using polyethylene brush on street surface within a 5 m^2 radius circle around each sampling site. The dust samples were transferred to a sealed polyethylene bag before being transported to the laboratory. Simultaneously, the detail information of sampling point coordinates and the surrounding land use characteristics was recorded. When samples were carried back to laboratory, they were left to dry at room temperature for several days and subsequently divided into four parts by quartering method, one part was conserved, and then the others were divided into two parts, finally the two parts were sieved, in all-

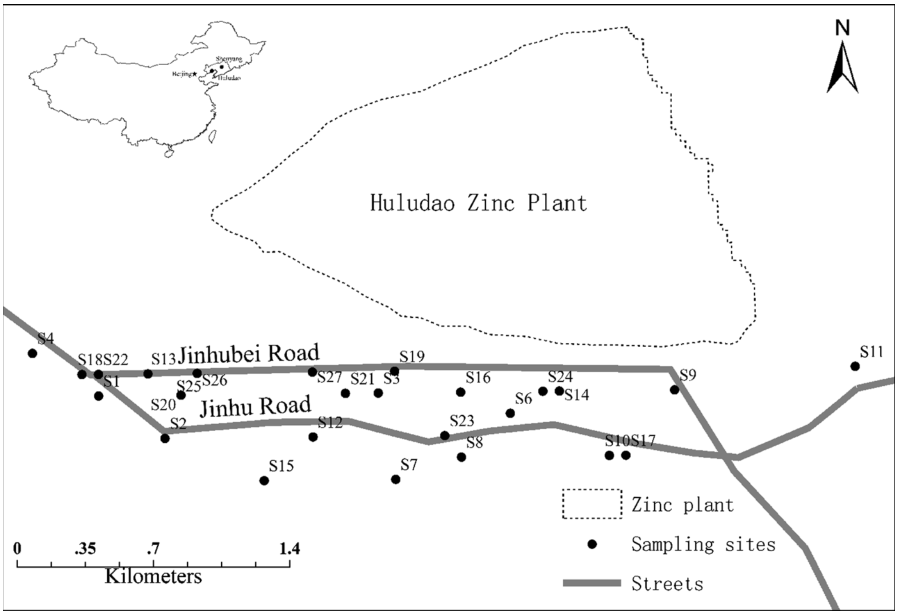

Fig. 1 Sampling sites of street dust around the HZP

Table 1 Analytical results obtained on certified reference materials (mg kg^{-1})

Elements	Observations	Observed ± SD	Certified ± SD	Recovery %
Pb	8	539.8 ± 37	552 ± 29	97.8
Cd	8	0.49 ± 0.05	0.45 ± 0.06	109.6
Cu	8	158.6 ± 3.6	144 ± 6	109.7

plastic sieving sets, through 100 and 63 μm, preserved in sealed valve bag.

Heavy metal determination and related analysis methods

About 0.15–0.20 g samples were digested with 5 ml HNO_3, 2 ml $HClO_4$ and 5 ml HF (GB/T 17138-1997). Pb and Cu concentrations were measured by atomic absorption spectrophotometer (AA-6300C, SHIMADZU, Japan). Cd was determined by ICP-MS (Thermo Fisher, X series II). Laboratory blanks and a soil standard reference material [GBW 07405(GSS-5)] were used to guarantee the accuracy and precision of the analytical method. Accuracy of the analytical method was given as the percent recovery for each element (Table 1). The dust pH and organic matter were determined using standard methods recommended by the Chinese Society of Soil Science. pH was measured in a 1:2.5 (sample, g/deionized water, ml) mixture with a pH meter according to the soil pH determination standard method (Lu 2000).

In order to test the accuracy of the experimental process, reagents used are top grade pure and reagent water was deionized water. In experimental process, all vessels used were soaked with nitric acid of 15 % solution soak after 24 h, with tap water, garnished with deionized water flushing three times.

Data analysis

Correlation analysis of the data was conducted by SPSS 16.0, and related statistical charts were finished by Origin 8.7. The spatial distribution of Pb, Cd and Cu contents in street dust were mapped by ArcGIS software.

Exposure model and health risk assessment method

Exposure model and parameter

Health risk assessment mainly consists of estimating the quantity of the heavy metal exposed to human body and evaluating the relationship between the exposed dose and the negative health effects, which cover pollutants identification, exposed pathway determination, exposure calculation and exposed risk assessment, in details. In this study, model developed by the US Environment Protection Agency (USEPA 1996) was used to calculate the exposure dose of people to Pb, Cd and Cu in the street dust near the HZP. The model applied in this study is based on the following assumptions underlie: (1) exposure to street dust particles happens via three main pathways including ingestion, inhalation and dermal contact, (2) heavy metal in the street dust exposed to the human body could be absorbed completely, (3) intake rate, particle emission and volatilization for street dust could be approximated by those developed for soil. Exposure is expressed in terms of a daily dose (mg kg^{-1} day^{-1}), and the doses received through each of the three pathways were calculated with Eqs. (1)–(3) adopted from the USEPA (1996, 2011a).

All of the three heavy metals in this study display non-carcinogenic risk, meanwhile Cd shows carcinogenic risk (IRAC 2011). For carcinogens, the lifetime average daily dose (LADD) used in the assessment of cancer risk has been calculated as a weighted average through inhalation exposure as shown in Eq. (4). In this study, only carcinogenic risk of Cd through inhalation is quantified, because USEPA only gave the inhalation reference dose of Cd.

$$\mathrm{ADD_{ing}} = C \times \frac{\mathrm{IngR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times \mathrm{CF} \quad (1)$$

$$\mathrm{ADD_{inh}} = \mathrm{C} \times \frac{\mathrm{InhR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{PEF} \times \mathrm{BW} \times \mathrm{AT}} \quad (2)$$

$$\mathrm{ADD_{dermal}} = C \times \frac{\mathrm{SL} \times \mathrm{SA} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times \mathrm{CF} \quad (3)$$

$$\mathrm{LADD} = \frac{\mathrm{C} \times \mathrm{EF}}{\mathrm{AT} \times \mathrm{PEF}} \times \left(\frac{\mathrm{InhR_{child}} \times \mathrm{ED_{child}}}{\mathrm{BW_{child}}} + \frac{\mathrm{InhR_{adult}} \times \mathrm{ED_{adult}}}{\mathrm{BW_{adult}}} \right) \quad (4)$$

where $\mathrm{ADD_{ing}}$ (mg kg^{-1} day^{-1}) is dose contacted through ingestion, $\mathrm{ADD_{inh}}$ (mg kg^{-1} day^{-1}) is dose contacted through inhalation, $\mathrm{ADD_{dermal}}$ (mg kg^{-1} day^{-1}) is dose absorbed through dermal contact with street dust and LADD (mg kg^{-1} day^{-1}) is the lifetime average daily dose for carcinogens;

Exposure parameters are listed as follows:

IngR is the ingestion rate; in this study, 200 mg day^{-1} for children and 50 mg day^{-1} for adult (USEPA 2001, Winifre et al. 2014); EF is the exposure frequency; in this study, 180 day $year^{-1}$ for street dust exposure (USEPA 2001); ED is the exposure duration; in this study, 6 years for children and 24 years for adult (USEPA 2001); BW is the average body weight; in this study, 18 kg for children and 60 kg for Chinese adult (Lu et al. 2014; Fang et al. 2013); AT is the averaging time: for non-carcinogens ED $\times$ 365 days; for carcinogens 78 $\times$ 365 days (USEPA 2011a); CF is conversion factor (10^{-6} kg mg^{-1}); InhR is inhalation rate; in this study, 7.6 m^3 day^{-1} for children and 20 m^3 day^{-1} for adult (Ferreira-Baptista and De Miguel 2005); PEF is particle emission factor; in this study, 1.36 $\times$ 10^9 m^3 kg^{-1} (USEPA 2001); SL is skin adherence factor; in this study, 0.2 mg cm^{-2} day^{-1} for children and 0.07 mg cm^{-2} day^{-1} for adult (USEPA 2001); SA is the exposed skin area; in this study, 2,800 cm^2 day^{-1} for children and 5,700 cm^2 day^{-1} for adult (USEPA 2001); ABS is dermal absorption factor (unitless); in this study, 0.001 for both children and adult (Zheng et al. 2010a).

The *C* (exposure-point concentration) values in Eqs. (1)–(4), combined with the values for the exposure factors shown above, are considered to yield an estimate of the "reasonable maximum exposure" (USEPA 1989), and is the upper limit of the 95 % confidence interval for the mean (95 % UCL). In this study, the concentrations of Pb, Cd and Cu in street dust approximate log-normal distributions and the 95 % UCL has been calculated by Eq. (5).

$$C_{95\%\mathrm{UCL}} = \exp\left\{ \bar{X} + 0.5 \times s^2 + \frac{s \times H}{\sqrt{n-1}} \right\} \quad (5)$$

where $\bar{X}$ is the arithmetic mean of the log-transformed data, *s* is the standard deviation of the log-transformed data, *H* is the *H*-statistics (Zheng et al. 2010a) and *n* is the number of samples.

Hazard quotient (HQ) and hazard index (HI)

The doses calculated with Eqs. (1)–(3) for each exposure pathway are subsequently divided by the corresponding reference dose (RfD) to yield a hazard quotient (HQ) (Eq. 6). The HQ assumes that there is a level of exposure (RfD), below which it is unlikely for sensitive populations to experience adverse health effects. If the exposure level exceeds this threshold (HQ > 1), potential adverse health effects may occur. As a rule of thumb, the greater the value of HQ is above unity, the greater the level of concern (USEPA 1989).

Hazard index (HI) is equal to the sum of the HQ (Eq. 7). The approach assumes that simultaneous sub-threshold exposures to several chemicals could result in adverse health effects. It also assumes that the magnitude of the adverse effect will be proportional to the sum of the ratios of the sub-threshold exposures to acceptable exposures (USEPA 1989). In this study, Hazard index methods and cancer risk methods were used to assess population health risk of metals exposure to street dust near the HZP.

For carcinogens, the dose was multiplied by the corresponding slope factor (SF) to produce a level of cancer risk (Eqs. 8–9) (USEPA 1996, 2001). The threshold of the risk value is 10^{-4}–10^{-6} above which environmental and regulatory agencies consider the risk unacceptable (USEPA 2002).

$$\mathrm{HQ} = \frac{D}{\mathrm{RfD}} \quad (6)$$

$$\mathrm{HI} = \sum \mathrm{HQ}_i \quad (7)$$

$$\mathrm{Risk}_i = \mathrm{LADD} \times \mathrm{SF}_i \quad (8)$$

$$\mathrm{Risk} = \sum \mathrm{Risk}_i \quad (9)$$

where HQ is the hazard quotient, indicating the non-carcinogenic risk of single contamination; D is the exposure dose; RfD (mg kg^{-1} day^{-1}) is the corresponding reference dose; HI is the hazard index, indicating the total non-carcinogenic risk from the three exposure pathways; SF is the corresponding slope factor, indicating the maximum probability carcinogenic risk of human expose of a certain dose of some pollutants.

Result and discussion

Characteristics of metals Pb, Cd and Cu in street dust based on different particle size

The statistical characteristics of Pb, Cd and Cu in street dust around HZP are presented in Table 2. As shown in Table 2, in the <100-μm-particle-size dust, Pb, Cd and Cu were 1,559, 178.5 and 917.9 mg kg^{-1}, respectively, which were 72, 1,653 and 46 times as high as the background value in soil of Liaoning province. While in the <63-μm particles, Pb, Cd and Cu were 2,099, 198.4 and 1,038 mg kg^{-1}, respectively, which were 97, 1,837 and 52 times as high as the background value. Metals in the <63 μm particle size were higher than those in <100 μm particle size. This observation was agreed with some studies which have suggested that smaller particles tended to contain higher element concentrations (Zhao et al. 2010). Based on the data, metals in street dust are possible from anthropogenic sources, which is consistent with the previous study (Zheng et al. 2010a). Cd in <63 μm particles approximated normal distribution, the other metals in different particle size dust all showed skewed positively distribution. According to CV value, heavy metals of street dust deviancy could grade into three levels: Weak variation (<10 %), moderate variation (10–30 %) and strong variability (>30 %). As is shown in Table 2, heavy metals in dust near HZP all belonged to strong variability, Pb, Cd and Cu in street dust varied very much. There was significant difference between metal concentrations in <100-μm dust and metal concentration in <63-μm dust.

Inter-element relationships provide information on the sources and pathways of metals (Al-Khashman 2007). Significance relationships were found among Pb, Cd and Cu contents in the different particle size. It indicated that Pb, Cd and Cu were similar and had a similar source. These results account for the same source or similar geochemical nature of Pb, Cd and Cu. Zn, Pb and Cu usually associated in minerals, for their analogical chemical behavior (Li and Thornton 2001), which also indicated that Pb, Cd and Cu in street dust are probably from waste gas emission of HZP.

Pb, Cd and Cu in street dust around HZP in this study were compared to other major cities around the world in Table 3. Metals in street dust around HZP were higher than other cities polluted by vehicle traffic and fossil fuel combustion, such as Greater Toronto, Jharia and Beijing (Tofan et al. 2013; Nazzal et al. 2014; Tanner et al. 2008). They were also higher than Hong Kong, Baoji, Guangzhou, Xi'an and Hermosillo (Li et al. 2013; Wang et al. 2014; Huang et al. 2014; Winifre et al. 2014; Volgyesi et al. 2014) in which heavy metals mainly come from industry pollution. Compared to the heavy metals in the street dust from

Table 2 Pb, Cd and Cu concentration distribution feature of street dust around HZP (mg kg^{-1})

Element	Mean (mg kg^{-1})	Median (mg kg^{-1})	SD (mg kg^{-1})	Max (mg kg^{-1})	Min (mg kg^{-1})	CV %	*n*	Soil background value (mg kg^{-1})
<100 µm								
Pb	1,559	1,433	1,078	4,634	117.3	69.14	28	21.6
Cd	178.5	147.6	124.9	510.2	5.68	69.98	28	0.11
Cu	917.9	785.4	656.9	2,831	135.6	71.56	28	19.8
<63 µm								
Pb	2,099	1,803	1,527	7,274	173.2	72.79	28	21.6
Cd	198.4	183.6	122.8	478.2	9.28	61.93	28	0.11
Cu	1,038	966.2	697.6	3,198	193.7	67.22	28	19.8

Table 3 A comparison of the heavy metal concentration in street dusts around the HZP and other selected cities (mg kg^{-1})

Cities	Pb	Cd	Cu	References
Dust around HZP (<100 µm)	1,559	178.5	917.9	This study
Dust around HZP (<63 µm)	2,099	198.4	1,038	This study
Xianyang	77.3	0.13	132.7	Shi et al. (2013)
Guangzhou	387.53	2.14	192.36	Huang et al. (2014)
Shanghai	155.6	3.32	160.2	Yin et al. (2013)
Shanghai urban area	236.6	0.97	257.4	Shi et al. (2010)
Baoji	1,586.2	5.5	178.2	Lu et al. (2009), Wang et al. (2014)
Tianjin	61.11	55.47	0.045	Yu et al. (2014)
Zhuzhou	956	41.4	139	Li et al. (2013)
Ma'an	105	3.21	26.42	Al-Khashman (2013)
Ajka	139	1.77	46.1	Volgyesi et al. (2014)
Nanjing	82.65	4.73	102.83	Li et al. (2013)
Jharia Towm	67.8	0.78	56.8	Tofan et al. (2013)
Greater Toronto Area	205.1	0.053	186	Nazzal et al. (2014)
Guiyu	20,600–22,900	9.34–66.8		Leung et al. (2008)
China B.[a]	26	0.097	22.6	CNEMC (1990)

[a] China B. = background values in soil of China

other sources such as weathering and corrosion of building materials, atmospheric deposition and commercial activities, it is still higher in this study (Shi et al. 2010; Yu et al. 2014). But except for Cd the contents of Pb and Cu were lower than the printed circuit board recycling workshop in Guiyu, Guangdong province, China (Leung et al. 2008).

Spatial distribution of Pb, Cd and Cu in street dust

Spatial distribution of Pb, Cd and Cu contents in street dust near HZP was conducted by Kriging interpolation of Arc GIS (Fig. 2). Pb, Cd and Cu in the street dust decreased gradually from the middle to the border when taking HZP as core. At the same longitude, Pb, Cd and Cu in dust from Jinhubei Road (near HZP) apparently were higher than that in Jinhu Road. Pb, Cd and Cu in the two roads decreased gradually from the nearest site from the zinc smelter to west and east direction, which indicated Pb, Cd and Cu had the same pollution source, the emission gas discharged by HZP. Pb, Cd and Cu contents ranked as residual area around HZP < Jinhu Road < Jinhu north Road (Table 4).

Effects of pH and organic matter to metals

pH is one of the main parameters that influences the mobility, bioavailability and distribution of heavy

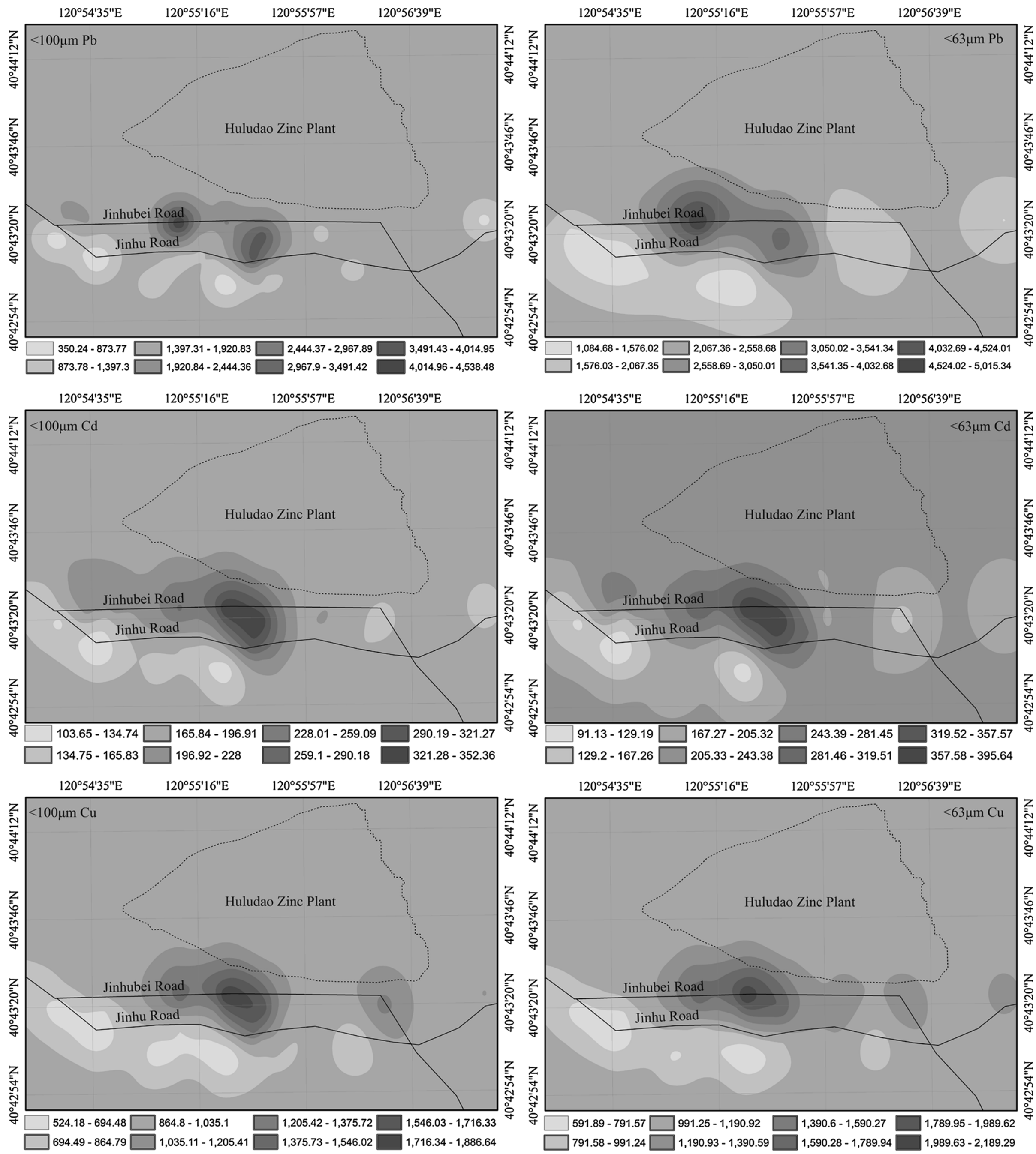

Fig. 2 Distribution of heavy metals in different particle size

metal (Yang et al. 2011). The available heavy metals would increase with pH of the surrounding environmental decreasing. pH value of street dust around HZP ranged 6.97–9.28 with the mean value of 7.71, higher than the background value (6.6), displayed alkalescency. pH and Cd (including <63 and <100 μm) have strong negative relations ($p = 0.01$), while pH and Pb (<63 μm) have significant negative correlations ($p = 0.05$). Pb, Cd and Cu contents in dust decreased with pH increasing. The excessive inputs of

Table 4 Correlation coefficients of the heavy metal contents, pH and organic matter ($n = 28$)

Element	<100 μm			<63 μm		
	Pb	Cd	Cu	Pb	Cd	Cu
<100 μm						
Pb	1					
Cd	0.857**	1				
Cu	0.778**	0.863**	1			
<63 μm						
Pb	0.966**	0.825**	0.743**	1		
Cd	0.838**	0.973**	0.794**	0.838**	1	
Cu	0.731**	0.856**	0.933**	0.746**	0.838**	1
PH	−0.364	−0.49**	−0.281	−0.406*	−0.515**	−0.355
Organic matter	0.410*	0.437*	0.474*	0.420*	0.412*	0.523**

** $p < 0.01$; * $p < 0.05$

sub-alkaline building materials (e.g., cement and calcareousness) into the street dusts may have resulted in dust alkalization (Yang et al. 2011). Heavy metal's adsorption is generally small at low soil Ph value, at high pH value, metals would be removed, thus the heavy metals concentrations decrease, which is in accordance with the negative correlations of pH and Cd (both in <100 μm particle and <63 μm particles)as well as Pb in <63-μm particles in this study (Bradl 2004).

Organic matter in street dusts around HZP ranged between 2.18 % and 11.34 %, with a mean value of 6.87 %, higher than the background value of 2.81 (CNEMC 1990). Pb, Cd and Cu had a positive correlation ($p = 0.05$) with organic in street dust except that Cu (<63 μm) and organic matter have a significant relationship at $p = 0.01$. Significant correlation was found between the content of Cu and percent of organic matter in street dust, which indicated that strong specific (covalent) interaction of Cu with organic matter (Banerjee 2003). Pb, Cd and Cu concentrations increase with the organic matter.

Organic matter contents depend on the organic matter accumulation of sources. Nevertheless, these sources of organic matter are impacted not only by nature deposit dust organic matter, but also by the accumulated organic matter resulted from human activities, including house refuse, vehicle exhaust. For these reasons, street dust organic matter showed a high level. Organic matter has many active groups, such as hydroxyl and carboxyl, which are liable to interact with heavy metals via diverse pathways, for example, complexation, chelation and redox (Bradl 2004). Consequently, organic matter in street dust has a positive correlation with the contents of heavy metals, which shows that the interactions between organic matter and dust cause more easily adsorption of heavy metal.

Health risk assessment

Pb, Cd and Cu intake due to street dust

The exposure doses for both children and adult were calculated with Eqs. (1)–(5). As for the three exposure pathways, ingestion was the dominant way for both subpopulations, followed by dermal contact, inhalation contributed least to non-carcinogens risk exposure dose, which are similar to the previous reports (Zheng et al. 2010a, b; Xu et al. 2012). Exposure dose of Pb, Cd and Cu contributed by ingestion was two orders of magnitude higher than the corresponding dermal dose and was three to four orders of magnitude higher than the inhalation value. The non-carcinogenic exposure doses among different heavy metal decreased in the order of Pb > Cu > Cd. The accumulative dose resulting from ingestion, inhalation and dermal contact for children were 13.3, 1.3 and 4.7 times higher than adult, respectively. Non-carcinogenic dose of heavy metal for <63-μm particles was higher than <100-μm particles. As for carcinogenic risk, the lifetime average daily dose of Cd in <100- and <63-μm particles was 1.11×10^{-8} and 1.21×10^{-8} mg kg^{-1} day^{-1}, respectively.

Except dust pathway, diet intake is also an important pathway for human metals exposure. Zheng et al. (2007a, b) reported that total diet target hazard quotients (TDHQ) of Pb, Cd and Cu intake for children via diet pathway were 0.331, 0.618 and 1.099, respectively, while for adults, TDHQ were 0.364, 0.749 and 1.220, respectively. Compared with this study, non-carcinogenic risk of Pb and Cd from dust exposure pathway is higher than that from diet intake, which indicates that reducing exposure dose of heavy metals in street dust is an important channel for protecting children from environmental heavy metals. Furthermore, comprehensive evaluation of heavy metal human exposure risk in diverse dwelling environment is necessary to prevent human health from heavy metal pollutants hazards.

The potential health risk of trace elements in the street dust

Values of reference dose except for Pb were taken from the US Department of Energy's RAIS compilation (USDE 2004), and reference doses for Pb have been derived from the WHO (1993) Guidelines for Drinking Water. Inhalation-specific toxicity data were available only for Cd, as for Pb and Cu toxicity values considered for the inhalation route are the corresponding oral reference doses, on the assumption that, after inhalation, the absorption of the particle bound toxicants will result in similar health effects as if the particles had been ingested (USEPA 2011b; Zheng et al. 2010a, b). The slope factor of Cd was taken from Zheng et al. (2010a, b).

Values of hazard quotient (HQ) and hazard index (HI) for non-carcinogenic elements in street dust around the HZP are listed in Table 5. From which we can see, the total hazard indexes (HIs) for children and adult exposed to <63-μm particles is higher than <100-μm particles. It indicated that the smaller particles tend to have higher non-carcinogenic health risk. Among the three exposed pathways, ingestion appeared to have the highest contribution to the overall figure of non-carcinogenic risk followed by dermal contact and inhalation turned out to be the least contributor to health risk. Inhalation of re-suspended particles through the mouth and nose is almost negligible when compared to the other two exposure routes. Similar results were obtained in previous studies (De Miguel et al. 2007; Zheng et al. 2010a, b; Han et al. 2011; Xu et al. 2012). The HQ of Pb in <63 μm particles was the largest contributor through ingestion and inhalation; however, for dermal contact the largest contributor was Cd in <63 μm particles. The total value of HI exposed to <63 μm particles for children is 12.29, which is far higher than 0.96 for adults. The percentage of HQ values for ingestion and dermal are 98.11 and 1.88 % in childhood, the values for adult are 94.76 and 5.18 %. The value of HI in <100-μm particles for children being 9.34 is higher than adult being 0.73. The percentage of HQ values for ingestion and dermal are 97.87 and 2.12 %in childhood, the values for adult are 94.10 and 5.82 %. Hazard indexes (HI) of all the three trace metals for adult were close to 1, indicating that it has potential non-carcinogenic health risk for adult when expose to street dust around HZP. Among the three non-carcinogenic elements, values of non-carcinogenic risk decrease in the order of Pb > Cd > Cu. The present study shown that HIs of Pb and Cd in both particle sizes for children had exceeded the "safe" value of HI = 1, indicated that children living around HZP were experiencing non-carcinogenic health risk from Pb and Cd exposure to street dust. We proposed that reasonable city function division should take into consideration in urban planning; the residential areas as well as the cultural and educational areas, especially the kindergarten and elementary school should be moved to the clean area far away from the zinc smelter plant. The content of Pb and Cd in street dust around HZP should be reduced to protect children from the hazard of Pb.

For carcinogenic risk, only Cd was assessed through the ingestion exposure modes of street dusts. The risk value for Cd in street dust with the particle sizes <100 and <63 μm was 7.00×10^{-8} and 7.59×10^{-8} (Table 5), respectively, both of which were within the range of threshold values (10^{-6}–10^{-4}) above which environmental and regulatory agencies consider the risk unacceptable. The carcinogenic risk levels of Cd in this study indicated that carcinogenic risk of Cd due to street dust exposure can be acceptable around HZP. However, except for Cd there are some other carcinogenic substances in street dust, e.g., As; therefore, the carcinogenic risk for people exposure to street dust around HZP is more than the value in this study.

As the largest zinc plant, HZP is gradually surrounded by residential area with the development of Huludao since the plant had been built in 1973.

Table 5 Hazard quotient and risk for arsenic in street dust around HZP

Items	<100 µm				<63 µm			
	Pb	Cd	Cu	Total	Pb	Cd	Cu	Total
C (95 % UCL) (mg kg^{-1})	1,978	226.9	1,173		2,691	246.1	1,308	
RfD_{ing} (mg kg^{-1} day^{-1})	1.40E−03	1.00E−03	4.00E−02		1.40E−03	1.00E−03	4.00E−02	
RfD_{inh} (mg kg^{-1} day^{-1})	3.50E−03	5.71E−05	4.00E−02		3.50E−03	5.71E−05	4.00E−02	
RfD_{dermal} (mg kg^{-1} day^{-1})	5.25E−04	2.50E−05	1.20E−02		5.25E−04	2.50E−05	1.20E−02	
SF_{inh} (mg kg^{-1} day^{-1})		6.30E+00				6.30E+00		
Child								
HQ_{ing}	7.74E+00	1.24E+00	1.61E−01	9.15E+00	1.05E+01	1.35E+00	1.79E−01	1.21E+01
HQ_{inh}	8.65E−05	6.08E−04	4.49E−06	6.99E−04	1.18E−04	6.60E−04	5.01E−06	7.83E−04
HQ_{dermal}	5.78E−02	1.39E−01	1.50E−03	1.99E−01	7.87E−02	1.51E−01	1.67E−03	2.31E−01
$HI = \sum HQ_i$	7.80E+00	1.38E+00	1.62E−01	9.34E+00	1.06E+01	1.50E+00	1.81E−01	1.23E+01
Risk		7.00E−08				7.59E−08		
Adult								
HQ_{ing}	5.81E−01	9.33E−02	1.20E−02	6.86E−01	7.90E−01	1.01E−01	1.34E−02	9.05E−01
HQ_{inh}	6.83E−05	4.80E−04	3.54E−06	5.52E−04	9.29E−05	5.21E−04	3.95E−06	6.18E−04
HQ_{dermal}	1.24E−02	2.98E−02	3.20E−04	4.24E−02	1.68E−02	3.23E−02	3.58E−04	4.95E−02
$HI = \sum HQ_i$	5.93E−01	1.24E−01	1.24E−02	7.29E−01	8.07E−01	1.34E−01	1.38E−02	9.55E−01
Risk		7.00E−08				7.59E−08		

Heavy metals were emitted into the environment in large quantities through atmospheric deposition during zinc smelting process, which will undoubtedly do harm to the residents around HZP. In this study, HIs of Pb, Cd and Cu in <100 μm particles for children were 7.8, 1.38 and 0.16 higher than results obtained by Zheng et al. (2010a), which covered the whole city. For adult, the HIs of Pb and Cu in this study were also higher than values studied by Zheng et al. (2010a), which indicated that the health risk for residents around HZP was higher than other residents in Huludao. Human health risk from metals of dust is also higher than that from stairway and sidewalk in Huludao and other cities (De Miguel et al. 2007; Zheng et al. 2010a; Xu et al. 2012).

Selection of dust fraction is significant and will have a high impact on exposure risk calculation results (Cao et al. 2012). Finer particles are more easily re-suspended into the air especially for particles with diameters less than 10 μm (PM10). Metals in this fraction can be easily adsorbed via inhalation and accumulated into human upper respiratory tract during breathing (Feng et al. 2014; Cao et al. 2012; Kong et al. 2011). While for dust with diameters less than 63 μm, exposure is more presumed to occur via ingestion (usually accidental, but for some, particularly toddlers, deliberate) and dermal contact besides inhalation (Cao et al. 2012). So dust with particle size less than 63 μm is more appropriate for health risk assessment.

Health risk assessment of heavy metals involves many parameters, while the parameters change with environment, age, sex and race. Besides, during health risk assessment we always suppose that heavy metal in the street dust exposed to the human body can be absorbed completely. In fact, when heavy metals enter our body, only a part of metal can be absorbed and the other part will be expelled out directly. There is a close relationship between heavy metal speciation and bioaccessibility of metals. Therefore, further research is needed to accurately access health risk of heavy metal exposed to street dust.

Conclusions

The average concentrations of Pb, Cd and Cu in the <100 μm dust were 1,559, 178.5 and 917.9 mg kg^{-1}, respectively. While in the <63-μm street dust, the values were 2099, 198.4 and 1,038 mg kg^{-1}, suggesting smaller particles tended to contain higher element concentrations. The concentrations of Pb, Cd and Cu in street dust around HZP are higher than other cities polluted by vehicle traffic, fossil fuel combustion, industry pollution, weathering and corrosion of building materials.

The contents of heavy metals in Jinhu Road and Jinhubei Road decrease gradually from the nearest site from the zinc smelter to west and east direction, which indicates the three metals have the same pollution source, and the waste gas discharged by HZP is the main source of Pb, Cd and Cu in the street dust. Correlation between Pb, Cd and Cu contents in the different particle size is significance strong. It indicated that Pb, Cd and Cu were similar and had a similar source. The contents of Pb, Cd and Cu in dust increased with decreasing pH or the increasing organic matter.

Non-carcinogenic health risk assessment results showed that the total hazard indexes (HIs) for children and adult exposed to <63-μm particles are 12.29 and 0.96, respectively, which are higher than 9.34 and 0.73 when exposed to <100 μm particles, indicated that the smaller particles tend to have higher non-carcinogenic health risk. Among the three exposed pathways, non-carcinogenic risk value decreased in the order of ingestion > dermal contact > inhalation. For the three non-carcinogenic elements, risk of Pb was the highest in both particle sizes, followed by Cd, while Cu contributed the least risk to human health. The hazard index (HI) of Pb and Cd in both particle sizes for children had exceeded the “safe” level of HI = 1, indicated that children living around the HZP were experiencing the non-carcinogenic health risk from Pb and Cd exposure to street dust. The carcinogenic risk levels of Cd in this study were within the threshold values (10^{-6}–10^{-4}).

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