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Relative importance of different exposure routes of heavy metals for humans living near a municipal solid waste incinerator☆

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ABSTRACT

The potential health effects of toxic chemicals (e.g. heavy metals) emitted by municipal solid waste incinerators (MSWIs) are of great concern to local residents, however there have been few studies on the contributions of different exposure pathways and their subsequent effects on the body burden of residents living near MSWIs. In this study, multiple exposure routes of heavy metals including Pb, Cr, Cd and Mn were assessed by investigating the metals in foods (such as vegetables, crops, meats and fruits etc.), drinking water, ambient air and soil collected surrounding an MSWI in Shenzhen, south China. Vegetable ingestion played the most important role in the total average daily dose of Pb and Cr, and cereals were the key exposure routes for Mn and Cd. Compound-specific contaminations were observed in the investigated areas, with Pb and Cr present in the surrounding environment, having accumulated to relatively high levels in the local vegetables, and the intake of contaminated vegetable foods greatly influencing the body burden of Pb and Cr. Consistently, significantly high blood concentrations of Pb and Cr were detected in the local residents compared to a referenced population, and a lack of significant differences was found for Cd and Mn. The results possibly suggested that emission of MSWI influenced the external exposure doses of the major pathways of Pb and Cr in this study, and resulted in the different body burden of metals in humans living near a MSWI. MSWI-local food-humans is an important exposure pathway for residents living near MSWI, and thus should not be neglected in developing future strategies and policies to prevent the high risks suffered by residents living near MSWIs.

1. Introduction

The acceleration of urbanization has made waste reduction and disposal important factors restricting economic and social sustainable development (Li et al., 2015; Cheng and Hu, 2010). Although regulators consider incineration a strategic municipal solid waste treatment method (Abanades et al., 2002; Rimaitytė et al., 2007), significant concerns have been raised regarding the installation of municipal solid waste incinerators (MSWIs) due to the potential effects of the toxic chemicals they emit (Domingo, 2002). Heavy metals such as lead (Pb), cadmium (Cd), and mercury (Hg) are among the many pollutants emitted by MSWIs, as municipal wastes such as leather waste shavings, batteries and lamps contain various toxic heavy metals (Louhab and Akssas, 2006). These heavy metals are emitted and carried out of the incinerator device among the hot flue gases and fly ash to cross environmental boundaries. It is well known that heavy metals can be absorbed by plants through uptake from the soil and air deposition, and by animals and humans through food, water, air, soil/dust ingestion and skin contact (Li et al., 2011; Zhao et al., 2010) posing health risks to those living near MSWIs.

Although studies all over the world have investigated the metals present in people living near MSWIs (Chen, 2004; Cheng et al., 2007), the latter's effect on blood metals in these populations remain controversial. In one study, no statistically significant difference was found between the group living within 300 m of an MSWI in Seoul and the reference group for Pb, Cd and Hg (Lee et al., 2013). Similarly, no significant differences between exposed and unexposed subjects were observed for blood metals in a pilot study in Modena, Italy (Ranzi et al., 2013). No associations were also observed between metal exposure from the incinerators and heavy metal body burden among the population living near MSWIs in Lisbon and Madeira, Portugal (Reis et al., 2007). In comparisons, Schroijen et al. found that adolescents living near an incinerator in the Northern part of Belgium had significantly higher Pb and Cd

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concentrations in their blood samples (Schroijen et al., 2008). Significantly high concentrations of Pb and Cd were also found in people proximity to the incinerators in Germany, compared with those who were not exposed (Wrbitzky et al., 1995). Given that the heavy metals can enter the human body through different routes, such as dermal contact, inhalation and ingestion (Li et al., 2011; Zhao et al., 2010), the different results about the influences of MSWI on human blood metals obtained in various countries might be due to the different contributions of multiple exposure pathways to the local residences. The importance of site-specific multipathway exposure of heavy metals has been reported in an investigation about the exposure of children to 10 metals near the largest coking plant in China (Cao et al., 2014). However, the current investigations about health risks of residents in the vicinity of MSWIs only focused on single exposure pathways such as foods, water, or dust/soil, and no studies is available on the contributions of different exposure pathways of heavy metals and their subsequent influences on the body burden of metals in populations near the MSWI.

The present research studied an MSWI in northwestern of Shenzhen, south China. Heavy metals including Pb, Cr, Cd and Mn were measured in foods (such as vegetables, crops, meats and fruits etc.), drinking water, ambient air and soil collected in the vicinities of the MSWI. Concentrations of the metals in blood samples were also investigated in the local residents (195 adults) living about 0–3 km away from MSWI, which were compared with a group of referenced persons (230 adults) living about 5–10 km away from the MSWI. The contributions of multi-pathway exposures of different metals were assessed, and help to understand the differences of blood metals in the two groups of populations. The results of the study provided a basis for guiding further strategies and policies aimed at preventing high risk suffered by residents living in the vicinities of MSWI.

2. Materials and methods

2.1. Study sites

An MSWI in Shenzhen, Guangdong Province, China, is featured in this study. It initiated regular operation in December 2005 and had three incineration units, each of which incinerate 400 tons of waste per day. The combustion temperature of the burning treatment lines is more than 850 °C, even up to 1100 °C. The average temperature at this site is 22.3 °C. The rainy season is April to September with an annual rainfall of 1924.7 mm. As shown in the wind rose map in Fig. 1, the average annual wind speed is 2.1–3.0 m/s with the wind coming from the northeast from September to February and from the southwest from March to May. From June to August, southwest and southeast winds is dominant.

2.2. Sampling of food, total suspended particulates, soil and water samples

Fifty-three types of food samples (each comprising at least three subsamples and selected according to the local population's dietary composition) were collected over four seasons from August 2013 to July 2014 (Table S1 in supplementary material). Of all the food samples, 39 were purchased from a market about 3 km away from the MSWI (Fig. 1) and included 8 types of fruit, 22 types of vegetables, 3 types of cereals, 9 types of fish and shrimp, 4 types of meat and 3 kinds of eggs. All the commercial foodstuffs in the market were not local products, and the commercial vegetables generally came from the commodity vegetable bases in Shandong and He'nan Province. In addition to the food samples purchased from the market, a farmers' garden situated about 1 km from the MSWI was found that grew vegetables for the commercial consumption of residents living close to the MSWI and no other potential heavy metal pollution sources nearby could be found. Based on the local topographical and meteorological conditions, the garden was in the areas affected by the pollution of the MSWI. Fourteen types of vegetable samples were purchased from this garden, and 11 of the local vegetables obtained were the same as those purchased from the market: BCL = cabbage (*Brassica campestris* L.); SOL = spinach (*Spinacia oleracea* L.); GBL = garlic bolt (*Garlic bolt* L.); BCL= Chinese cabbage (*Brassica chinensis* L.); SLL = leaves of asparagus lettuce (*Strobilanthes lactucifolia Levl.*); AA = chive (*Allium ascalonicum*); LSL = leaf lettuce (*Lactuca sativa* L.); IAF = water spinach (*Ipomoea aquatica Forsk*); CAL = green pepper (*Capsicum annuum* L.); CSL = cucumber (*Cucumis sativus Linn*); VU = cowpea (*Vigna unguiculata*).

9 fly ash samples were collected from the discharge of filters from the gas cleaning system from November 2013 to July 2014. Ambient air monitoring was conducted at five stations (A1 to A5) with increases in the distance to the MSWI as shown in Fig. 1. Total suspended particulates (TSP) samples were collected on Whatman glass fiber filters with a diameter of 47 mm (0.1 mm pore-size and 99.9% collection efficiency) using a medium volume sampler (Laoying, Qingdao) operated at a constant flow rate (100 L/min) and programmed to collect 24 h samples. The medium volume samplers at all of the stations were kept at a height of 1.5 m. The TSP mass was determined by gravimetric analysis on glass fiber filters stabilized and weighed before and after sampling. Overall, 45 TSPs samples were collected over nine months from August 2013 to July 2014.

During the sampling period, 46 soil samples were collected from the areas surrounding the MSWI. Surface soil samples (0–5 cm depth; approximately 1 kg) were collected using plastic brushes, wrapped in aluminum foil and stored in sealed polyethylene bags. The soil samples were grouped into five areas, based on the distance of the sampling locations from the MSWI: 1 km north (−1.6-0 km, n = 9), within (0 km, n = 11), 1–3 km south (1–3 km, n = 20), 5–6 km south of MSWI (5–6 km, n = 4), and 9–10 km southeast (9–10 km, n = 2).

Eighteen tap water samples were collected from areas located about 0–1, 5–6 and 9–10 km away from the MSWI over a 1-year sampling period. Water samples were collected in polyethylene bottles pre-washed with concentrated nitric acid (1:1) and deionized water several times before use. To avoid possible contamination during water sampling, we turned on the water taps completely to release water for at least 5 min.

2.3. Blood sampling

A scheme devised for the collection of human blood samples was approved by the Ethical Committee of the Peking University Health Science Center. The population comprising 425 adults ranging in age from 24 to 45 years was divided into two groups: 195 from areas with potential exposure (0–3 km away from the MSWI) and 230 from the reference area (5–10 km away from the MSWI). All the participants have lived and worked in the current addresses for at least two years, and these persons spent most their daily time in the areas. In 2014, venous blood samples taken from all of the participants; that is, drawn into vacutainers containing sodium heparin anticoagulant, after thoroughly cleaning the arm with an ethanol swab. All of the samples were stored at −80 °C until analysis. Before the sample collection, we asked each participant, with the assistance of qualified public health workers, to complete a written informed consent and answer a questionnaire covering lifestyle information and social demographic characteristics including age, gender, body mass index (BMI), resident history,

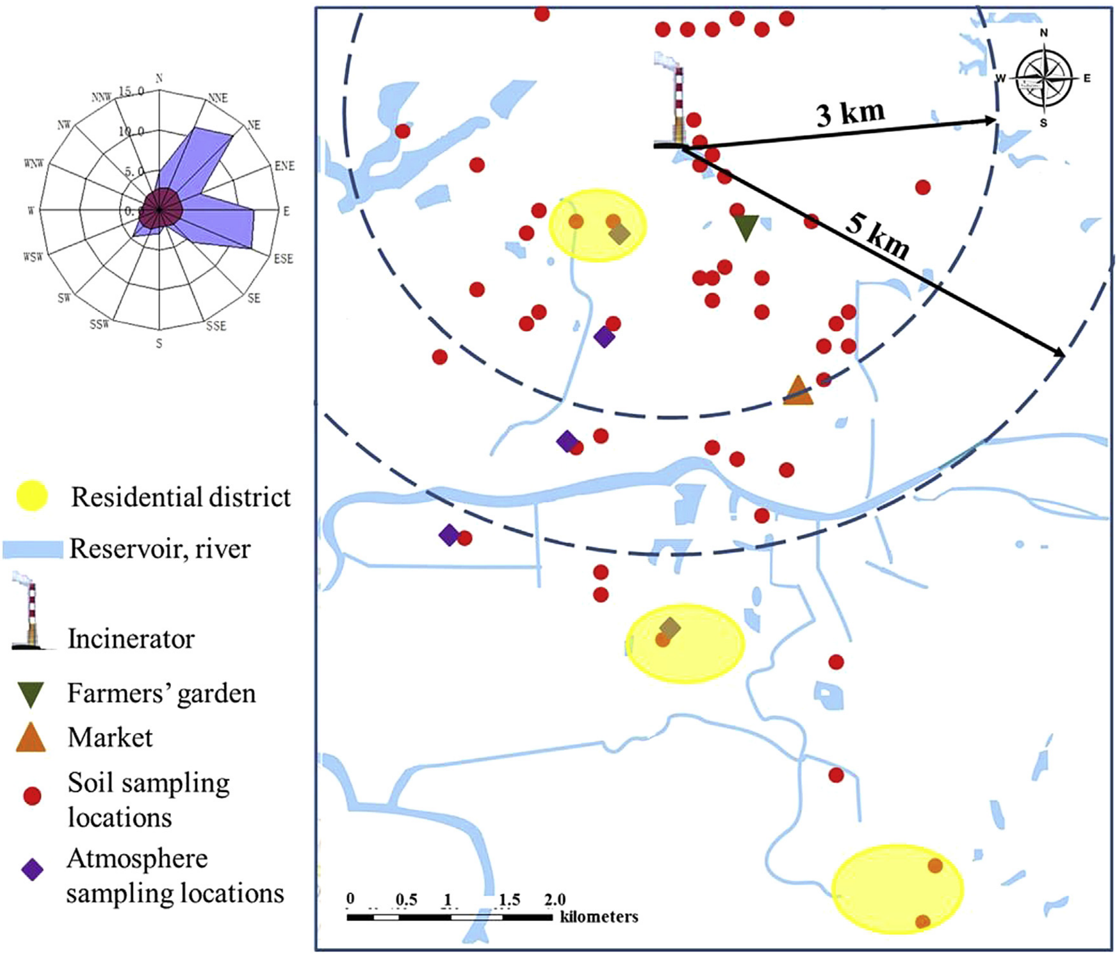

Fig. 1. Locations of the atmospheric and soil sampling sites, the market and the farmers' garden in the investigated areas surrounding the MSWI. Two population groups were collected in areas about 0–3 km and 5–10 km away from the MSWI. Wind rose map in Shenzhen from 1953 to 2008.

smoking habit and level of education (Table 1).

2.4. Metal analysis

All the collected meat, fruit and vegetable samples were rinsed by distilled water before metal analysis. Approximately 0.2-g freeze-dried food sample, 0.1-g filter membrane of each TSP sample or 0.1-g fly ash sample were transferred to a Teflon vessel, and 6 mL HNO_3 was added for digestion. The microwave digestion unit (CEM Mars-6, USA) was programmed at 1600 W with the following conditions: ramp to 120 °C in 5 min and keep for 3 min, then ramp to 160 °C in 8 min and hold for 5 min, then ramp to 180 °C in 5 min and kept for 15 min; cooling time 15 min. The digested solutions were heated at 120 °C for 4 h close to dryness and then transferred to a decontaminated tube after being cooled. The solution was diluted to 10 mL with deionized water and kept at −20 °C until ICP-MS analysis.

The soil samples were freeze-dried, ground and sieved through a 2-mm mesh screen to achieve a homogeneous grain distribution. Each approximately 0.1-g sample was combined with 6 mL HNO_3, 0.5 mL HF, and 5 mL HCl before microwave digestion. The conditions of microwave digestion unit were the same as those used for the food and air samples. The digested solution was heated and 2 mL H_2O_2 was added at 120 °C for 4 h close to dryness, and then transferred to a decontaminated tube after being cooled. The solution was diluted to 10 mL with deionized water and kept at −20 °C until ICP-MS analysis. Ten-mL tap water samples were added with nitric acid (0.1% v/v) and then kept at 4 °C until metal analysis.

Each 0.5-mL blood sample was added to 5 mL HNO_3 for

Table 1
Characteristics of the studied population.

	Distance to MSWI	
	0-3 km	5-10 km
Enrolled	195	230
Age, year	32.2 ± 4.9	33.3 ± 5.2
BMI	21.9 ± 3.3	21.9 ± 3.2
Duration of residence, year	4.4 ± 2.1	3.0 ± 1.7
Gender, n (%)		
Male	121 (62.1)	140 (60.9)
Female	74 (37.9)	90 (39.1)
Smoking, n (%)		
Current smokers	45 (23.6)	47 (20.4)
Former smokers	4 (2.1)	10 (4.3)
Non-smokers	142 (74.3)	173 (75.2)
missing	4	0
Education, n (%)		
primary school or below	0 (0)	2 (0.9)
secondary school	4 (2.1)	39 (17)
high school	90 (46.6)	102 (44.3)
bachelor degree	96 (49.7)	82 (35.7)
master degree or above	3 (1.6)	5 (2.2)
missing	2	0

microwave digestion with the temperature programed as described above. The digested solution was heated to 120 °C for 4 h close to dryness and then transferred to a decontaminated tube after being cooled. The solution was diluted to 10 mL with deionized water and kept at −20 °C until ICP-MS analysis.

2.5. ICP-MS analysis

Cr, Cd, Pb and Mn were measured by an inductively coupled plasma mass spectrometer (ICP-MS, Thermo ICP-Q, USA) with the following instrumental conditions: radio frequency power was 1500 w, RF matching was 1.38 v, sample depth was 8 mm, peristaltic pump was 0.3 r/min, nebulizer was 0.8 L/min and integration time was 0.1 s.

2.6. Quality assurance and quality control

Reagent blanks were processed simultaneously to deduct the error induced by the analytical procedure. Standard soil reference materials (GBW07403 (GSS-3)) were used to validate the analytical procedure. The recoveries for standard reference materials were 94–96%, which confirmed the reliability of the analytical protocol. We also determined recoveries for other samples by spiking them with a working standard (10 μg/L environmental sample media, 5 μg/L blood sample media), and the average recoveries of the individual species in the spiked samples varied from 82 to 102%. All of the analyses for the standard reference materials and spiking samples were performed in triplicate. The instrumental limits of detection were 0.08, 0.15, 0.003 and 0.12 μg/L for Pb, Cr, Cd, and Mn, respectively.

2.7. Average daily dose assessment

Three main heavy metal exposure pathways in local residents were considered: ingestion, inhalation and dermal contact. The risk estimates were determined based on the US Environmental Protection Agency (EPA) health risk handbook (USEPA, 2001). The risk of exposure was expressed in terms of the average daily dose (ADD) (ng kg^{-1} day^{-1}), which was calculated using Eqs. (1) (2) and (3).

The dose through the ingestion of food, water and soil was calculated using Eq (1)

$$\mathrm{ADDingest} = \frac{\mathrm{C} \times \mathrm{IngR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{1}$$

The dose through the inhalation of air and soil was calculated using Eq (2)

$$\mathrm{ADDinhale} = \frac{\mathrm{C} \times \mathrm{InhR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT} \times \mathrm{PEF}} \tag{2}$$

The dose absorbed through dermal contact with soil was calculated using Eq (3)

$$\mathrm{ADDdermal} = \frac{\mathrm{C} \times \mathrm{SA} \times \mathrm{SL} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{3}$$

where C is the concentration of metals in the matrix (ng/g or ng/m^3), IngR is the ingestion rate in mg/day, InhR is the inhalation rate in m^3/day, SA is the surface area of the skin exposed to pollutants in cm^2, SL is the skin adherence factor in mg/cm^2, EF is the exposure frequency in days/year, ED is the exposure duration in years, AT is the averaging time, BW is the residents' body weights obtained through the questionnaire-based the survey, ABS is the dermal absorption factor, and PEF is the particle emission factor representing an estimate of the relationship between soil contaminant concentrations and the concentration of these contaminants in air as a consequence of particle suspension. To calculate the ADD of each pathway, the exposure parameters were obtained from the literature (Li et al., 2015; USEPA, 2001; Liu et al., 2015a) and listed in Table S2.

2.8. Statistical analysis

In our statistical analyses, the medians, arithmetic means and range were calculated to assess the levels of heavy metal contamination in blood samples. The differences in concentrations between two groups (0–3 km and 5–10 km) and in vegetables from the farmers' garden were evaluated using the Student t-test. A one-way analysis of variance (ANOVA) was performed to look for differences in the data on concentrations in air and soil samples. A probability value of $p < 0.05$ was set as the level for statistical significance. Statistical analysis was performed using the SPSS version 22.0 statistical package.

3. Results and discussion

3.1. Heavy metals in atmospheric and soil samples

Concentrations of Pb, Cr, Cd and Mn in the fly ash were 1347 ± 358, 339 ± 127, 175 ± 44 and 556 ± 266 mg/kg, with the ranges of 760–1841, 141–559, 113–232 and 249–1029 mg/kg, respectively, which were comparable to those reported in MSWIs in Shanghai, Japan and Korea (Pb: 340–3600 mg/kg, Cr: 54–310 mg/kg, Cd: 20–410 mg/kg, Mn: 100–700 mg/kg) (Shim et al., 2005; Zhou et al., 2015). Due to the relatively high concentrations detected in fly ash, the four metals were selected as the target pollutants. Concentrations of Pb, Cr, Cd and Mn were detected in atmosphere samples collected from 5 sampling locations at 1-km-interval distances from the MSWI. As Table S3 shows, concentrations of Pb, Cr, Cd and Mn in the TSP samples from the 5 sampling sites were 47.83 ± 30.51, 19.52 ± 26.45, 2.23 ± 1.68 and 47.56 ± 42.05 ng/m^3, with the following ranges of 15.02–166.39, 5.62–202.09, 0.33–10.33 and 10.44–310.19 ng/m^3, respectively. The atmospheric concentrations of metals measured at 5 sampling sites were all below the air quality guidelines for Europe. The concentration of Cr (19.52 ng/m^3) in this study was higher than those reported in Sapporo (2.61 ng/m^3) and Tokyo (6.09 ng/m^3) (Var et al., 2000), but concentrations of Pb (47.83 ng/m^3), Cd (1.22 ng/m^3), and Mn (47.56 ng/m^3) were all lower than those reported in Ho Chi Minh (Pb: 146 ng/m^3) (Hien et al., 2001), Hong Kong (Pb: 79 ng/m^3) (Lau and Luk, 2001), Beijing (Mn: 235 ng/m^3) (Okuda et al., 2008), Tokyo (Pb: 125 ng/m^3) (Var et al., 2000), and Taiwan (Cd: 8.5 ng/m^3) (Fang et al., 2004). The highest annual average concentrations of the four metals were all found in atmospheric samples at Site A1, but no statistical differences were noted for the metal concentrations in samples collected at different sampling sites (Fig. 2). A decreased concentration trend with increasing distance from the MSWI was observed (Fig. 2), suggesting that the MSWI was a potential emission sources of the atmospheric metal contaminations measured nearby.

The metals in the soil samples could reflect the spatial distributions of the pollutants in the investigated area over time, compared with the atmospheric concentrations of metals obtained through active air sampling. The concentrations of Pb, Cr, Cd and Mn in the soil samples from the five sampling areas that were different distances from the MSWI are shown in Table S4 and Fig. 3: (Pb) 58.75 ± 33.46 mg/kg, (Cr) 37.09 ± 24.30 mg/kg, (Cd) 0.27 ± 0.18 mg/kg, and (Mn) 321.5 ± 358.8 mg/kg. The concentrations of Pb in soil the samples collected from areas around the MSWI plant (83.93 ± 42.46 mg/kg) were about two-times higher

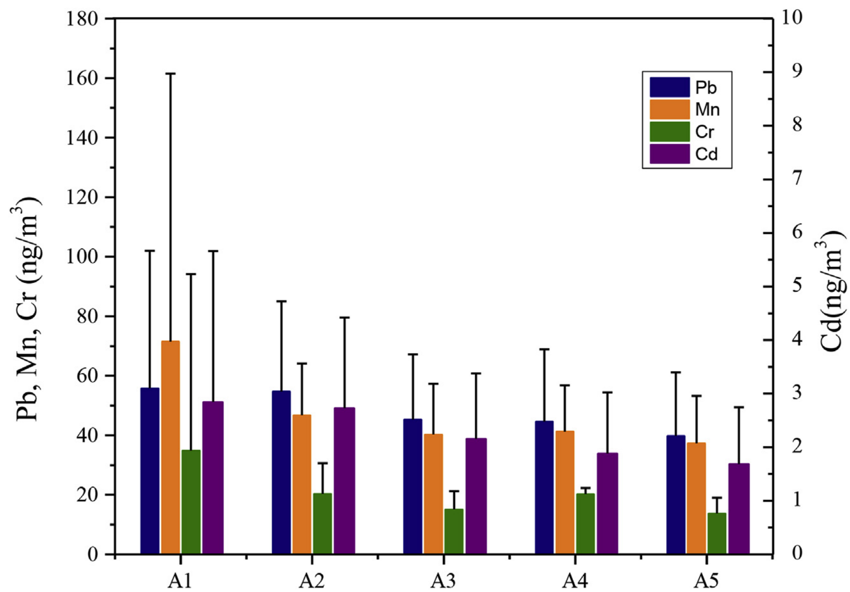

Fig. 2. Concentrations of Pb, Cr, Mn and Cd in TSPs (ng/m^3) collected from five sampling sites over a one-year sampling period.

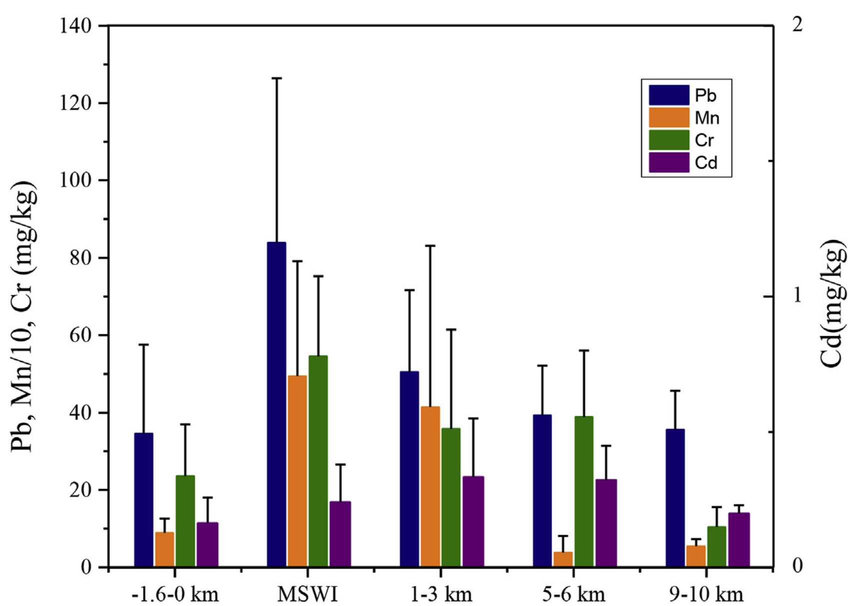

Fig. 3. Concentrations of Pb, Cr, Mn and Cd in soil samples (mg/kg) collected from five areas different distances away from the MSWI.

than those reported in soil from a MSWI in Spain (46.36 ± 29.04 mg/kg) (Nadal et al., 2005). In contrast, the concentrations of Pb in the soil around the MSWI in our study were all lower than those of urban soils in European cities such as Rome (330.8 mg/kg) (Angelone et al., 1995), La Coruma (309 mg/kg) (Calprieto et al., 2001), and Palermo (202 mg/kg) (Manta et al., 2002). Although concentrations of Mn and Cd in soils in the present study were comparable to those previously reported in soils collected near MSWIs (Mn: 320.5 ± 126.4 mg/kg; Cd: 0.22 ± 0.15 mg/kg) (Nadal et al., 2005) or in cities (Mn: 340–519 mg/kg; Cd: 0.29–0.31 mg/kg) (Manta et al., 2002; Wilcke et al., 1998), relatively high concentrations of Cr were detected in soils from the investigated areas compared with other studies (Cr: 26.4–39 mg/kg) (Calprieto et al., 2001; Wilcke et al., 1998), which is consistent with the high concentrations of Cr observed in air samples in the present study. Among the five sampling areas, the concentrations of Pb, Cr and Mn in soils collected from areas within the MSWI were 83.93 ± 42.46 mg/kg, 54.59 ± 20.69 mg/kg and 494.1 ± 297.9 mg/kg, respectively—all significantly higher than those in soils from −1.6-0 and 5–6 km from the MSWI plant ($p < 0.05$). We observed a decrease in soil concentrations of Pb, Cr and Mn with increasing distance from the MSWI, but no significant differences could be found for Cd in the soils samples collected from different areas, suggesting that the relatively low Cd emissions in the investigated areas would not result in the high accumulation in the soil surrounding MSWI. The spatial distribution of metals in the air and soils samples from the studied areas indicated that metal concentrations in the environmental matrix were affected by emissions from the MSWI.

3.2. Heavy metals in vegetables from a garden near the MSWI

Given the relatively high concentrations of Pb, Cr, Cd and Mn in the air and soil samples taken near the MSWI, the vegetables grown near MSWI were expected to accumulate high levels of heavy metals. The heavy metal concentrations in various vegetables from the farmers' garden situated about 1 km from the MSWI were further determined (Fig. 4, Table S5). The average concentrations of Pb, Cr, Mn and Cd in 14 types of vegetables in the farmers' garden were 46.36 ± 49.03 ng/g ww, 206.9 ± 389.6 ng/g ww, 3.47 ± 3.08 μg/g ww and 5.29 ± 4.27 ng/g ww, respectively. The average concentrations of Pb, Cr and Cd in leafy vegetables (53.92 ± 50.92 ng/g ww, 236.4 ± 423.4 ng/g ww and 5.92 ± 4.44 ng/g ww, respectively) were higher than those (11.13 ± 3.71 ng/g ww, 68.39 ± 11.84 ng/g ww and 2.34 ± 0.71 ng/g ww, respectively) in fruit vegetables (Fig. 4), possibly due to the fact that leafy vegetables tend to accumulate more heavy metals through the deposition. This is supported by a recent study of metal transportation in vegetables, which found that atmospheric emissions was an important source for the heavy metal accumulation in the aerial parts of vegetables (Li et al., 2015).

For comparison, we analyzed Pb, Cr, Mn and Cd in the vegetables purchased from the market, which were delivered from other cities far from the MSWI-affected areas. The average concentrations of Pb, Cr, Mn and Cd in the vegetables from the market were 13.08 ± 17.15 ng/g ww, 78.02 ± 101.11 ng/g ww, 3.47 ± 3.08 μg/g ww and 10.53 ± 21.00 ng/g ww, respectively. The metal concentrations in the market vegetables in the present study were within the range of those reported in previous studies in Bangladesh and Hong

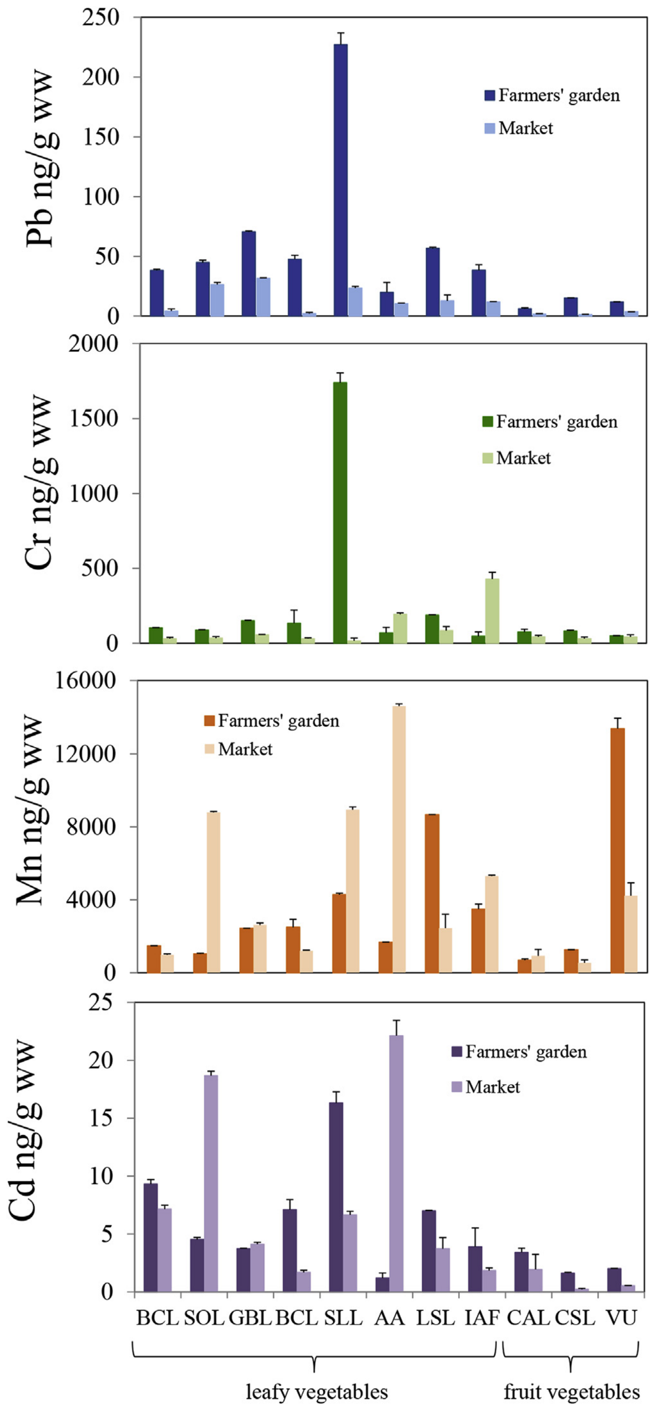

Fig. 4. Concentrations of Pb, Cr, Mn and Cd in vegetables from the farmers' garden and market.

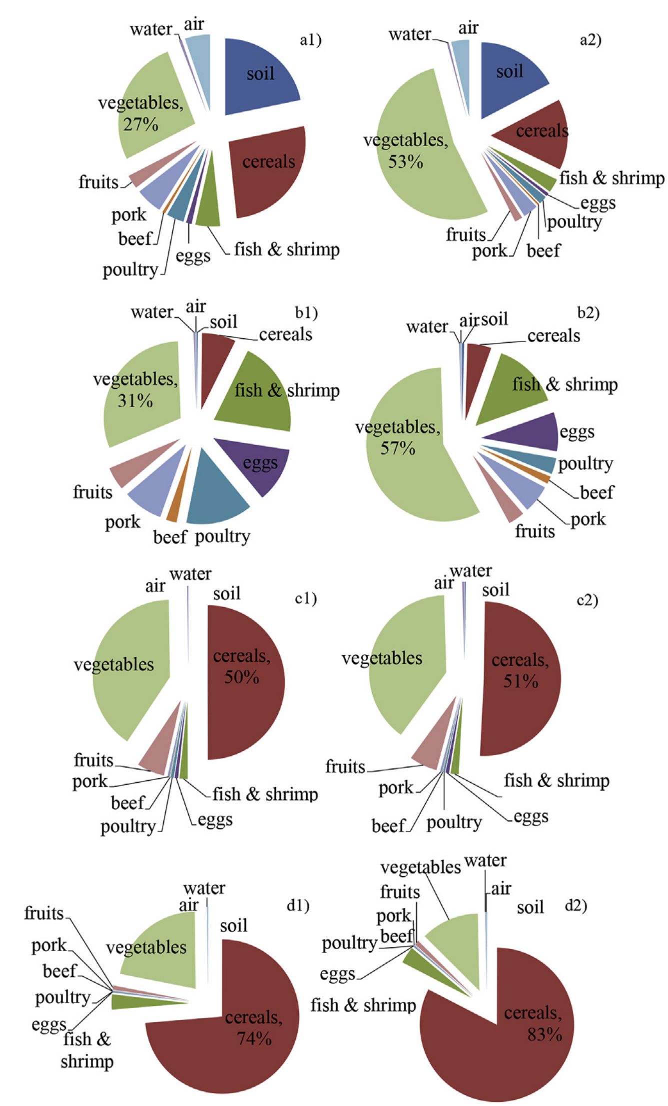

Fig. 5. Percentages of contributions to the ADD through exposure to air, water, soil and food for populations in two scenarios: those who only consumed commercial vegetables (a1: Pb; b1: Cr; c1: Mn; d1: Cd), and those who only consumed the vegetables from the garden near the MSWI (a2: Pb; b2: Cr; c2: Mn; d2: Cd).

Kong (Pb: 5–120 ng/g ww, Cr: 50–240 ng/g ww, Mn: 6.9–28.4 μg/g ww, Cd: 8–56 ng/g ww) (Shaheen et al., 2016; Hu et al., 2013). Our comparison of the heavy metal concentrations in the vegetables from the farmers' garden and those in the vegetables from the market revealed that the concentrations of Pb and Cr in the former were significantly higher than those in the latter ($p < 0.05$), but no significant difference could be found for vegetable concentrations of Cd and Mn (Fig. 4). The results were consistent with the significantly high concentrations of Pb and Cr in the air and soil samples collected near the MSWI, suggesting that Pb and Cr emitted from the investigated MSWI could be deposited in the surrounding environment to accumulate in relatively high levels in the local vegetables. The indirect intake of metals discharged from the MSWI via the edible vegetable consumption posed an excessive exposure suffered by local residents.

3.3. Daily exposure doses of heavy metals

Human exposure to heavy metals mainly occurs through the routes of ingestion, inhalation and dermal contact (Cao et al., 2014). The general population's daily exposure doses of heavy metals were evaluated based on the levels of heavy metals in environmental samples collected in areas about 5–10 km away from the MSWI in the present study (Tables S4–S7). The metal intake rates through exposure to ambient air, drinking water, food and soil were obtained from the literature (Table S2) (USEPA, 2001; Li et al., 2015; Liu et al., 2015a). The results showed that food ingestion played an important role in the total ADD, accounting for 72, 99, 99, and 99% for Pb, Cr, Cd and Mn, respectively (Fig. 5). It is interesting to note that the percentages of contribution to total exposure dose by different types of foods indicated compound-specific differences for target metals (Fig. 5). The results showed that the ingestion of vegetables played the most important role in the total ADD of Pb (27%), followed by cereals (26%), fish and shrimp (5%), poultry (4%), and pork (6%). Similarly, the ingestion of vegetables contributed the most to the total ADD of Cr (31%), followed by fish and shrimp (20%), poultry (14%), pork (8%), and cereals (7%). In comparison, the ingestion of cereals contributed predominantly to the total ADD of Mn, accounting for 50%, followed by vegetables (40%), fish and shrimp (2%), fruit (6%) and poultry (0.6%), and the ingestion of cereals accounted for the majority of the total ADD of Cd (74%), followed by vegetables (22%), fish and shrimp (3%), fruit (1%) and poultry (0.2%). The results implied that cereals were the key exposure routes for the residents' ADD of Mn and Cd, and the intake of contaminated vegetable foods greatly influencing the body burden of Pb and Cr.

The daily exposure to heavy metals experienced by residents living near the MSWI was further estimated based on the assumption that the local residents only consume vegetables from the garden near the MSWI. The contributions of different exposure pathways were calculated based on the metal concentrations in vegetables from the local farmers' garden and the environmental samples collected in areas about 0–3 km away from the MSWI. As Fig. 5 shows, contributions made by ingesting vegetables increased from 27 to 31 to 53–57% for Pb and Cr, but no obvious difference could be found regarding the percentage contributions of different exposure pathways for Mn and Cd. The results suggested that the exposure burden of Pb and Cr for residents living near the MSWI may be relatively easily affected, since person living surrounding the MSWI have relatively high consumptions of vegetables grown in the vicinities of MSWI.

3.4. Blood metals in residents

Blood samples were collected from two groups of residents: those living 0–3 km away from the MSW and those living about 5–10 km away. Table 1 showed the characteristics of the populations, and no significant differences were observed between the cases and the controls with respect to age, BMI, smoking status. Table S7 and Fig. 6 showed the concentrations of Pb, Cr, Mn and Cd in the blood samples from two areas. The concentrations of Pb, Cr, Mn and Cd in all of the blood samples were 26.86 ± 15.02 ng/mL, 3.26 ± 3.26 ng/mL, 12.61 ± 4.84 ng/mL and 1.68 ± 1.97 ng/mL, respectively. The concentrations of Pb and Mn in the blood samples in the present study were relatively higher than those reported in Canadian (Pb: 23.83 ng/mL; Mn: 10.99 ng/mL) and German (Pb: 22 ng/mL; Mn: 9 ng/mL) studies (Clark et al., 2007; Heitland and Köster, 2006), and the blood concentrations of Cr and Cd in the present study were comparable to those in Spanish and Russian (Cr: 1.31–6.65 ng/mL, Cd: 0.49–2.17 ng/mL) studies (Gil et al., 2011; Ivanenko et al., 2013). Interestingly, the concentrations of Pb and Cr in those living in near the MSWI (0–3 km) were significantly higher

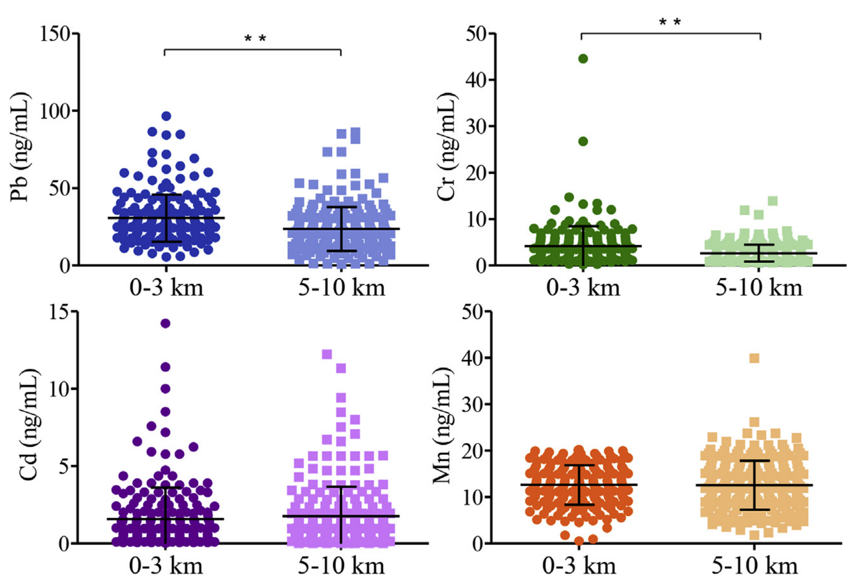

Fig. 6. Concentrations of Pb, Cr, Mn and Cd (ng/mL) in the blood samples of residents living 0–3 and 5–10 km away from the MSWI. **: $p < 0.05$.

than those in the reference area population ($p < 0.05$), and a lack of significant differences was observed in blood concentrations of Cd and Mn in those from the two areas ($p > 0.05$). Gender-based significant differences in Pb and Cr concentration were also found. The elevated blood concentrations of Pb and Cr were consistent with the significantly high contribution of vegetable ingestions to the ADD of Pb and Cr compared with those of Mn and Cd to the local residents. The results of the internal metal levels and external exposure doses indicated that the major exposure pathways were significantly affected for Pb and Cr in this study and resulted in the different body burden of metals in humans living near a MSWI. In previous studies, only the internal metal levels were investigated in persons living in the vicinities of MSWI, and different results about the health impacts of MSWI were obtained (Lee et al., 2013; Ranzi et al., 2013; Reis et al., 2007; Schroijen et al., 2008). It is possible that the different results might be due to the site-specific multi-pathway exposure of heavy metals to the local residences. In this study, we found that vegetable and cereal ingestions are important exposure pathways for Pb/Cr and Cd/Mn, respectively, and consistent high blood levels of Pb/Cr were observed in humans living in the vicinities of MSWI, who consumed lots of local polluted vegetables in a farmers' garden nearby. Thus, clarifying the relative contribution of various exposure routes of heavy metals is important to find the predominant exposure pathway influencing the internal metal levels of local residents.

Many countries and institutions have established heavy metal emission standards to protect human health against MSWIs (Liu et al., 2015b; EUR-Lex, 2000). In the present study, we showed that the external exposure doses of metals could be affected by the MSWI–local food–humans exposure pathway. As we known, MSWI was generally built far from the city due to its potential pollution to the surrounding environment, and planted vegetables can be easily found in the rural areas in the vicinity of MSWI (van Dijk et al., 2015; Li et al., 2015; Pan et al., 2016; Xiong et al., 2016). The consumption of local vegetables, however, poses a greater risk of excessive exposure to pollutants emitted from MSWIs, subsequently resulting in unexpected health risks for local residents. Therefore, we urge inhabitants to show caution in consuming vegetables grown near MSWIs, and future risk assessments should consider the MSWI–local food–humans exposure pathway.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2017.04.002.

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