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Health risks of heavy metal exposure through vegetable consumption near a large-scale Pb/Zn smelter in central China

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ABSTRACT

Smelting of nonferrous metals is an important source of heavy metals in surface soil. The crops/vegetables grown on contaminated soil potentially impose adverse effects on human health. In this study, the contamination level of five heavy metals (Hg, Pb, Zn, Cd and Cu) in ten types of vegetables grown nearby a large scale Pb/Zn smelter in Hunan Province, China and the health risk associated with their consumption are assessed. Based on the data obtained from 52 samples, we find that Pb and Cd contributed to the greatest health risk and leafy vegetables tend to be more contaminated than non-leafy vegetables. Within 4 km radius of the smelter, over 75% of vegetable samples exceeded the national food standard for Pb; over 47% exceeded the Cd standard; and 7% exceeded the Hg standard. Heavy metal concentrations in vegetables measured within the 4 km radius are on average three times more elevated compared to those found at the control area 15 km away. Heavy metals in vegetables have dual sources of root absorption from soil and leaf adsorption from atmosphere. Health risk in terms of the hazard index (HI) at contaminated areas are 3.66 and 3.14 for adults and children, respectively, suggesting adverse health effects would occur. HI for both groups are mainly contributed by Pb (48%) and Cd (40%). Fortunately, vegetable samples collected at the control area are considered safe to consume.

1. Introduction

The rapid economic development in China in the past few decades resulted in widespread environmental pollution including heavy metal contamination in soil (Chen et al., 2015). Previous studies indicated that 1/6 of the agricultural land in China faces different degrees (14.49% mild, 1.45% moderate and 0.72% serious) of heavy metal pollution (Bao et al., 2008; Zhou and Feng, 2014). Due to the accumulation and slow removal process, soil contaminated with heavy metals can pose a threat to human health through polluted crops and vegetables (Khan et al., 2015; Chen et al., 2013). The sources of heavy metal pollution can be derived from direct industrial releases from vehicle and point sources followed by atmospheric deposition (Gibson and Farmer, 1986; Thornton, 1991). The environmental concerns and health effects of Hg, Pb, Zn, Cd and Cu contamination associated with nonferrous metal mining and smelting activities have been documented extensively (e.g. Zheng et al., 2007a; Bi et al., 2009; Li et al., 2015; Feng et al., 2013; Árvay et al., 2017). Particularly, Hg, Pb and Cd are potent toxic elements and excessive intake of these elements can cause bioactive destruction (Korkmaz et al., 2018; Sevindik et al., 2017) and superimposed hazards. Lead (Pb) and zinc (Zn) smelters are regarded as one of the most important heavy metal emission sources (Nriagu and Pacyna, 1988; Li et al., 2015) because of the simultaneous release of more than twelve metals and metalloids into the environment (Sterckeman et al., 2002; Li et al., 2013). Vegetables grown on the farmland near the emission source can be contaminated by the deposited heavy metals, which leads to subsequent human exposure (Alam et al., 2003; Li et al., 2014a). Heavy metal contamination of vegetables, the human exposure and the associated health risk have gained much attention and required more studies (Storelli, 2008; Wei and Yang, 2010; Chen et al., 2016). Wai et al. (2017) and Zheng et al. (2007a) assessed the risk of heavy metal exposure via consumption of vegetables grown near non-ferrous metals smelters in East and North China. Both concluded that smelting activities adversely impacted the health of residents, especially among children. Adverse effects of heavy metal exposure have also been reported near non-ferrous metal smelters

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in France (Douay et al., 2013; Dahmani-Muller et al., 2000), Australia (Kachenko and Singh, 2006), Russia (Pollard et al., 2015).

Hunan province is a major center for non-ferrous metal production in China, potentially putting > 28,000 km^2 (13% of the total area of the province) of agricultural land at risk of contamination (Guo and Zhu, 2004). In 2015, Hunan province produced 1.04×10^6 t Pb and 1.19×10^6 t Zn, account for 23.56% and 19.48% of the national production, respectively (China Nonferrous Metals Industry Association, 2016). As the largest industrial city in Hunan, Zhuzhou has > 5500 industrial enterprises, of which Zhuzhou smelter is the largest Pb/Zn smelter in China. For decades, this smelter is the dominant heavy metal source in the study region, with 34 t of heavy metals emitted into the environment each year, such as Pb, Hg, Cd, Cu, Ni, Zn, etc. The emissions quantity accounts for more than 90% of the total heavy metal emissions in the Zhuzhou (Qin et al., 2010). From 1960 to 2011, the smelter emitted approximately 105 t of Hg into the atmosphere, of which 15 t deposited locally and resulted in an increase of Hg concentration in soil from 0.12 to 1.77 mg/kg (based on the dry weight, DW) (Wu et al., 2014). Li et al., (2011, 2013, 2014b) showed that > 10 heavy and trace metals were released into the air and agricultural soils by the Zhuzhou smelter. Located in the rural-urban fringe zone, the emissions from the Zhuzhou smelter potentially contaminate the vegetables grown and sold locally. For example, Qin et al. (2010) reported elevated Cd and arsenic (As) levels in cabbage grown nearby this smelter.

The objective of this study is to gain a comprehensive understanding of heavy metal contamination in vegetables grown near the smelter as well as the health risk associated with the consumption of the vegetables. Ten (10) types of vegetables are systematically sampled in the agricultural fields at various proximities to the Zhuzhou smelter for chemical analysis of Hg, Pb, Cd, Zn and Cu. The analytical data are utilized in calculating the hazard index (HI) for estimating the health risks caused by heavy metal exposure through vegetable consumption. The results of this study would provide guidelines for vegetable consumption in highly industrialized areas in China.

2. Methods

2.1. Study area and the smelter

Zhuzhou is situated in the east side of Hunan Province, downstream of Xiang River, and about 45 km southeast of the province capital, Changsha (Fig. 1). Located at the Xiang alluvial plain (26° 03' 05" ~ 28° 01' 07" N and 112° 57' 30" ~ 114° 07' 15" E), and 961.7 thousand population in 2015. The climate is influenced by the subtropical monsoon, with wind direction mainly from the northwestern, expect the direction of wind is southeastern in summer (June to August). The annual average temperature is 18.6 °C and annual precipitation is 1587.7 mm, which is suitable for planting a variety of crops. Thus, the region is considered a high yield area in Hunan Province (Statistical Bureau of Hunan Province, 2016). The city is also a new industrial city in China, characterized by rich mineral resources and prosperous manufacturing industry.

Founded in 1956, Zhuzhou Smelter produces 650,000 t Pb and Zn each year in recent years. The production processes of lead and zinc are pyrometallurgical and hydrometallurgical methods, respectively. The technique for particulate removal in the flue gas is a combination of cyclone and electrostatic precipitator. The SO_2 in the flue gas is recovered for H_2SO_4 production. This factory also recovered sulfuric acid and metals including copper, gold, silver, cadmium, indium and bismuth. However, during the operation process, it still emitted large amount of heavy metals into the surrounding environment as aforementioned.

2.2. Sampling and analytical methods

2.2.1. Sampling methods and preparation

In order to ensure that the sampling is representative, we took local vegetables randomly from 8 sampling areas (denoted as A to H). For each vegetable sample at each sampling site, we collected multiple plants and made up samples together. Fifty-two vegetable samples representing 10 common types of commercial vegetables, including Chinese white cabbage (*Brassica napus* L.), White Radish (*Raphanus sativus* L.), Cauliflower (*Brassica oleracea* L. var. *botrytis* L.), Chinese cabbage (*Brassica pekinensis* Rupr.), Carrot (*Daucus carota* L.), Leek (*A. tuberosum* Rottl. ex Spreng.), Cabbage (*Brassica oleracea* L. var. *capitata* L.), Celery (*Apium graveolens* L.), Garlic sprout (*Allium sativum* L.) and Lettuce (*Lactuca sativa* L.), were collected near the smelter (Fig. 1) in January 2010. All vegetables were planted in ambient environment and no waste water was irrigated. To make a thorough investigation of the spatial relationship between the pollution and the smelter, the majority of vegetable samples and the associated soil samples were taken from the contaminated areas (A, B, C, D, E, F, G) within 4 km to the smelter and the baseline reference samples were collected at a control area (H) of 15.5 km in the northeast of the smelter. Table 1 shows the sample types and location of the eight sampling areas. Each vegetable sample represent a composite made up by at least three vegetable plants, with 0.5–1.0 kg of total fresh weight. The surface soils (0–20 cm) at each area were also collected and the results of heavy metals in soils had been reported by Li et al., (2011, 2014b).

In the laboratory, all vegetable samples were thoroughly washed with tap and deionized water sequentially, separated by edible part and non-edible part, and then dried in the oven at 40 °C absolutely. During the cleaning and drying procedure, the water content of each sample was also measured. Finally, all dried samples were grinded and bagged in high pressure polyethylene (HPPE) bags for elemental analysis.

2.2.2. Analytical methods

The ground and dried samples were digested using microwave according to Chen (2007). Around 0.5 g dry vegetable sample was placed in the Teflon digestion tank, added with 5 ml double-distilled HNO_3 and heated for 30 min at 100 °C. After cooling down, 1 ml 30% H_2O_2 was added into the digestion, then put the samples in a microwave for additional 21 min. Finally, the digestion liquid was filtered with a 0.45 µm Polytetrafluoroethylene (PTFE) filter and then subject to analysis. To ensure and control the quality of test, reagent blank, duplicates, and certified reference material (CRM) standards (GBW07604, poplar leave and GBW10010, rice) were also prepared as the same procedures. Elements of Pb, Zn, Cd, Cu in digest are determined by an Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Platform ICP, UK) and Hg is determined by Cold Vapour Atomic Fluorescence Spectroscopy (CVAFS, Tekran 2500, Canada) after $SnCl_2$ reduction. The recovery of all measured elements in CRM standards remained between 88% and 107%, acceptable for elemental analysis.

2.3. Methodology of contamination and health risk assessment

2.3.1. Contamination assessment of vegetables

The level of vegetable contamination is assessed by the single-factor (P_i) and the comprehensive pollution index (P). These indices are deemed to comprehensive and practical evaluation methods (Yang et al., 2008), as shown in the following Eqs. (1) and (2):

$$P_i = \frac{C_i}{C_0} \qquad (1)$$

where P_i is the single-factor index; C_i is the concentration of the heavy metals in the vegetables (mg/kg, FW) on fresh weigh basis and C_o is the assessment criterion of each element (mg/kg, FW). C_o is the maximum allowable concentrations (MAC) in food. The regulated contamination level of China is applied for C_o for the criteria, with GB 2762-2017 for

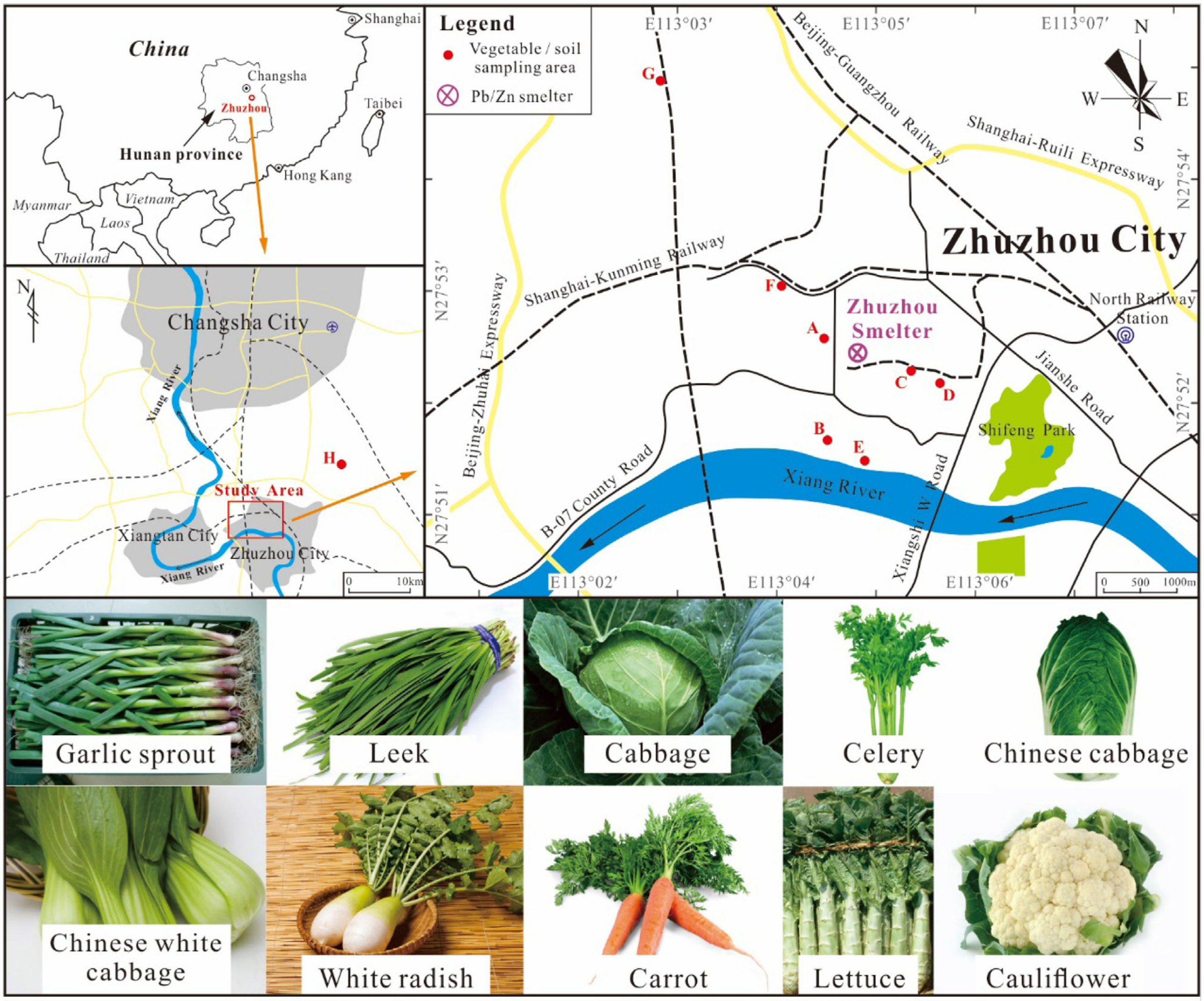

Fig. 1. Location of the sampling sites near the Pb/Zn smelter and ten kinds of vegetables sampled.

Table 1
Description of the sampling areas and the samples collected.

Sampling areas	Location	Vegetable samples (N = 52)
A	150–300 m west of the smelter (N 27°52′50′, E 113°04′27′)	(N = 8, Chinese white cabbage, Cauliflower, Cabbage, White Radish, Chinese cabbage, Lettuce, Celery, Carrot)
B	1.3 km south of the smelter (N 27°51′39′, E 113°04′27′)	(N = 7, Cauliflower, Cabbage, Carrot, White Radish, Chinese cabbage, Lettuce, Celery, Leek)
C	1.1 km east of the smelter (N 27°52′27′, E 113°05′15′)	(N = 5, Cabbage, White Radish, Chinese cabbage, Lettuce, Celery)
D	1.6 km east of the smelter (N 27°52′21′, E 113°05′45′)	(N = 6, Cauliflower, Cabbage, White Radish, Garlic sprout, Chinese cabbage, Lettuce, Celery)
E	1.7 km southeast of the smelter (N 27°51′29′, E 113°04′51′)	(N = 7, Cabbage, Garlic sprout, White Radish, Chinese cabbage, Lettuce, Celery, Leek)
F	1.6 km northwest of the smelter (N 27°53′02′, E 113°04′02′)	(N = 8, Cauliflower, Cabbage, Garlic sprout, White Radish, Chinese cabbage, Lettuce, Celery, Leek)
G	3.9 km northwest of the smelter (N 27°54′58′, E 113°02′53′)	(N = 5, Cabbage, Garlic sprout, Chinese cabbage, Lettuce, Celery)
H	15.5 km northeast of the smelter (N 27°56′07′, E 113°13′01′)	(N = 6, Cabbage, White Radish, Garlic sprout, Chinese cabbage, Lettuce, Celery)

Hg, Pb and Cd (more details in Section 3.1), GB 15199-94 for Zn (20 mg/kg) and GB 13106-91 for Cu (10 mg/kg) on a fresh weight basis.

$$P = \sqrt{\frac{\overline{P_i}^2 + P_{\max}^2}{2}} \qquad (2)$$

where P is the comprehensive pollution index; $\overline{P_i}$ is the average of single-factor index (P_i); $P_{\max}$ is the maximum of single-factor index (P_i). Both the single factor index (P_i) and comprehensive pollution index (P) are classified into five levels: $P \le 0.7$ as safe, Class I; $0.7 < P \le 1.0$ as clean, Class II; $1.0 < P \le 2.0$ as slightly polluted, Class III;

Table 2
Concentration of heavy metal in the edible parts of different kind of vegetables and the national standard for food.

Sampling areas	Hg(mg/kg, FW)	Pb(mg/kg, FW)	Zn(mg/kg,FW)	Cd(mg/kg,FW)	Cu(mg/kg,FW)
Contaminated areas (A-G)					
Leafy vegetable	0.005	1.079	11.327	0.212	0.800
(N = 28)	(0.001–0.011)[#]	(0.185–3.714)	(4.242–19.208)	(0.060–0.514)	(0.272–7.197)
Non-leafy vegetable	0.002	0.562	9.435	0.207	1.260
(N = 18)	(0.000–0.008)	(0.042–2.483)	(3.654–19.141)	(0.026–0.607)	(0.286–3.079)
Flower vegetable	0.001	0.326	14.592	0.089	0.905
(N = 4)	(0.000–0.002)	(0.139–0.696)	(8.729–19.141)	(0.071–0.124)	(0.704–1.036)
Totally average	0.004	0.877	10.587	0.210	0.980
(N = 46)	(0.000–0.011)	(0.042–3.714)	(3.654–19.208)	(0.026–0.607)	(0.272–7.197)
Control area (H)					
Leafy vegetable	0.002	0.106	3.679	0.049	0.605
(N = 4)	(0.001–0.003)	(0.060–0.150)	(2.598–5.777)	(0.034–0.063)	(0.428–0.938)
Non-leafy vegetable	0.000	0.057	2.757	0.039	0.680
(N = 2)	(0.000–0.000)	(0.052–0.062)	(2.525–2.989)	(0.026–0.052)	(0.449–0.911)
Totally average	0.001	0.090	3.371	0.045	0.630
(N = 6)	(0.000–0.003)	(0.052–0.150)	(2.525–5.777)	(0.026–0.063)	(0.428–938)
The maximum allowable concentrations (MAC) in food (mg/kg, FW)	0.01[*/**]	0.3[*] 0.1[**]	–	0.2[*] 0.1[**] 0.05[***]	–

[#] Average(Min-Max).
[*] Leafy vegetable.
[**] Non-leafy vegetable.
[***] Flower vegetable (MAC quotations from GB 2762-2017).

$2.0 < P \leq 3.0$ as moderately polluted, Class IV; and $P > 3.0$ as heavily polluted, Class V.

2.3.2. Health risk assessment

Excess exposure to heavy metals can produce either acute or chronic health effects (Yang et al., 2014). To study the health risk of heavy metals through vegetable consumption among adults and children, health risk assessment model of estimated daily intake (EDI) and target hazard quotation (THQ) are applied (Clow, 1998; Paustenbach, 2002).

$$EDI = C \times Con \tag{3}$$

$$THQ = \frac{EDI \times EF \times ED}{BW \times AT \times RfD} \tag{4}$$

where C (mg/kg) is the concentration of heavy metals in vegetable; Con (g/person/day, FW) is the daily average consumption of vegetable in the region; EF is exposure frequency (365 day/year); ED is exposure duration (76.3 years, equivalent to the average life span); BW (kg/person) represents body weight; AT is average time (365 day/year of exposure years and assuming 76.3 years in this study) (National Bureau of Statistics of China, 2015); RfD refers to a daily reference dose. The average daily vegetable intake of adults is 389 g/day (FW) and average adults body weight is considered 55.9 kg (Wang, 2005; Statistical Bureau of Hunan Province, 2016). For the children, the average daily vegetable intake is 195 g/day (FW) and body weight is 32.7 kg, respectively (Wang, 2005; Statistical Bureau of Hunan Province, 2016). RfD for Hg, Pb, Zn, Cd, Zn are 0.7, 3.5, 300, 1.0, 40 µg/kg/day, respectively (USEPA, 2009). The individual health risks of the analyzed heavy metals in the same vegetable are accumulative, that expressed as hazard index (HI). The HI of multiple heavy metals (Hg, Pb, Zn, Cd, Cu) are assessed as:

$$HI = \sum_{n=1}^{i} THQ_n;\ i = 1,\ 2,\ 3\ldots\ n \tag{5}$$

Where HI is the sum of various pollutant hazards. An HI value < 1.0 indicates minimal health impact; An HI value > 1.0 indicates potential health impacts; An HI value > 10.0 suggests serious chronic risk.

3. Results and discussion

3.1. Heavy metal in vegetables

Heavy metal concentration in edible parts of different vegetables are shown in Table 2 and Fig. 2. The maximum allowable concentration (MAC) standards for food in China (GB 2762-2017) is also shown in Table 2. The food standards for Pb and Cd vary from different kind of vegetables. For Pb, the standard is 0.3 mg/kg (FW) in leafy and 0.1 mg/kg (FW) in non-leafy vegetables. For Cd, the standard sets 0.2 mg/kg (FW) in leafy vegetables, 0.05 mg/kg (FW) in flower vegetables (in this study is for cauliflower) and 0.1 mg/kg (FW) in root or tuber vegetable (in this study suitable for lettuce, carrot and white radish). The standards for Zn and Cu in vegetables were phased out in 2011 and the new standard has not yet been set. According to the standards GB 15199-94 and GB 13106-91, the content of Zn and Cu in vegetables of this study is substantially lower than these standards (20 mg/kg for Zn and 10 mg/kg for Cu, FW).

The average concentrations of Hg, Pb, Zn, Cd and Cu in the edible part of different vegetables are 0.004, 0.88, 10.59, 0.21 and 0.98 mg/kg (FW) at the contaminated sites (A-G), respectively, and 0.001, 0.09, 3.37, 0.05 and 0.63 mg/kg (FW) at the control site (H), respectively (Table 2). The concentration of Hg, Pb, Zn and Cd at the contaminated sites were 3–10 times higher than that of the control site. The Cu content at contaminated sites was only 1.5 times higher (0.98 vs 0.63 mg/kg, FW) than that at the control site. Cu in soil (31–118 mg/kg, DW) in the vicinity to the smelter is the lightest impacted elements among the five investigated heavy metals (Li et al., 2011, Fig. 3) and the plants have a strong absorb ability for this essential element (Chen et al., 2013). Besides, leafy vegetables tend to contain more of Hg, Pb, Zn and Cd than non-leafy vegetables (Table 2 and Fig. 2), especially Hg and Pb. The average concentrations of Hg, Pb, Zn and Cd in leafy vegetables were 0.005, 1.079, 11.327 and 0.212 mg/kg (FW), respectively, and in non-leafy vegetables were 0.002, 0.562, 9.435 and 0.207 mg/kg (FW), respectively, (Table 2), leafy vegetable contains nearly 1 time more Hg and Pb than that of non-leafy vegetables. Leafy vegetables are exposed to the atmosphere and therefore can assimilate atmospheric Hg° and Pb through the foliage (Ao et al., 2017; Obrist et al., 2017; Bi et al., 2009). Since the majority of commercial vegetables are leafy, the health risks caused by the heavy metal contamination cannot be overlooked. Other

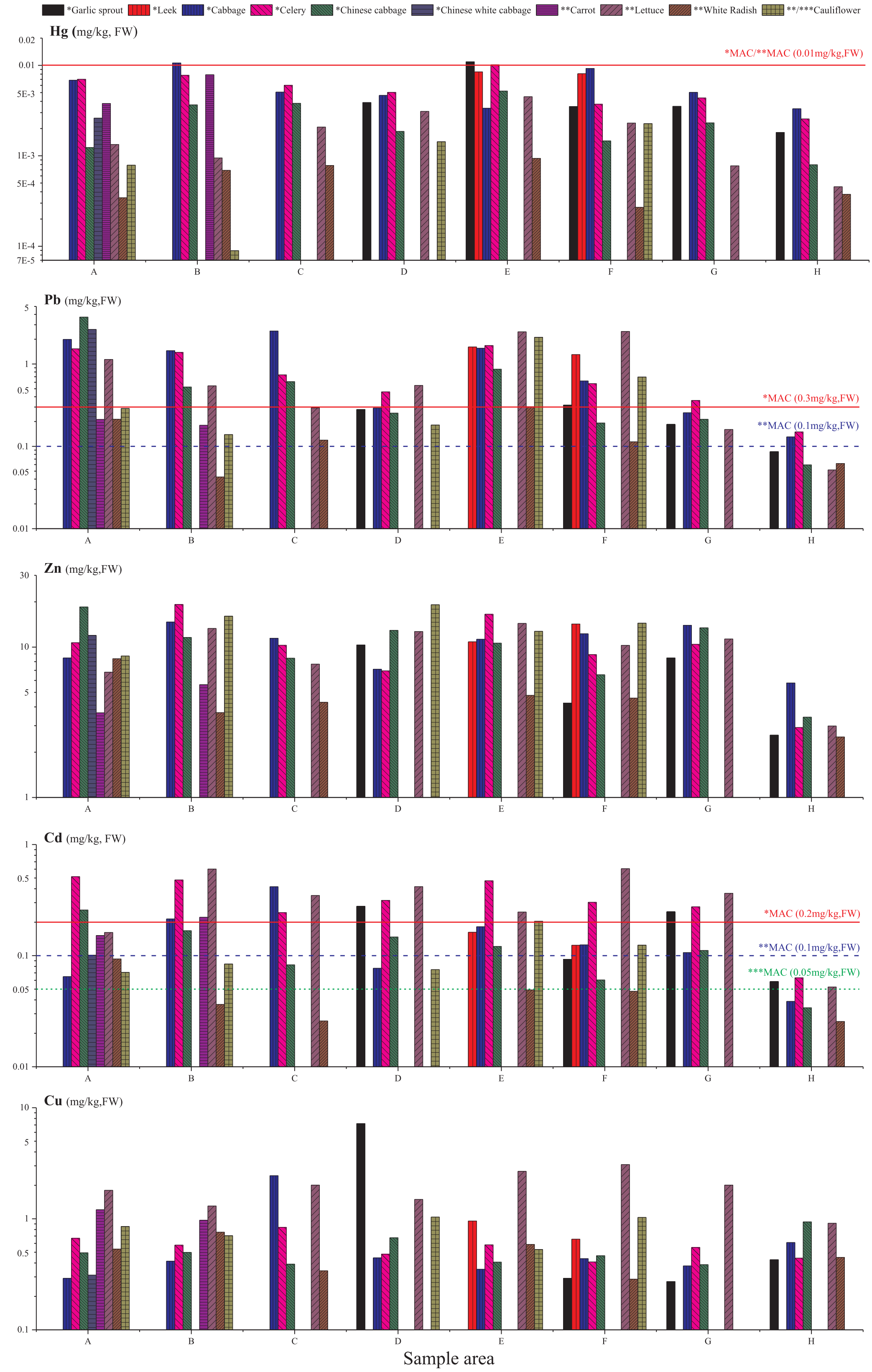

Fig. 2. Heavy metals in the edible part of different vegetables at different sampling areas around the smelter. (* Leafy vegetable; **Non-leafy vegetable, ***Flower vegetable).

parts such as stems and flowers did not show elevated heavy metal concentration, because of a range of protection mechanisms in the soil-root system, such as intracellular/extracellular sequestration, precipitation by sulphur (S) and phosphate (P), conversion of bioavailable heavy metals to into inert species through the oxidation-reduction reactions, biosorption/binding of heavy metals on the bacterial

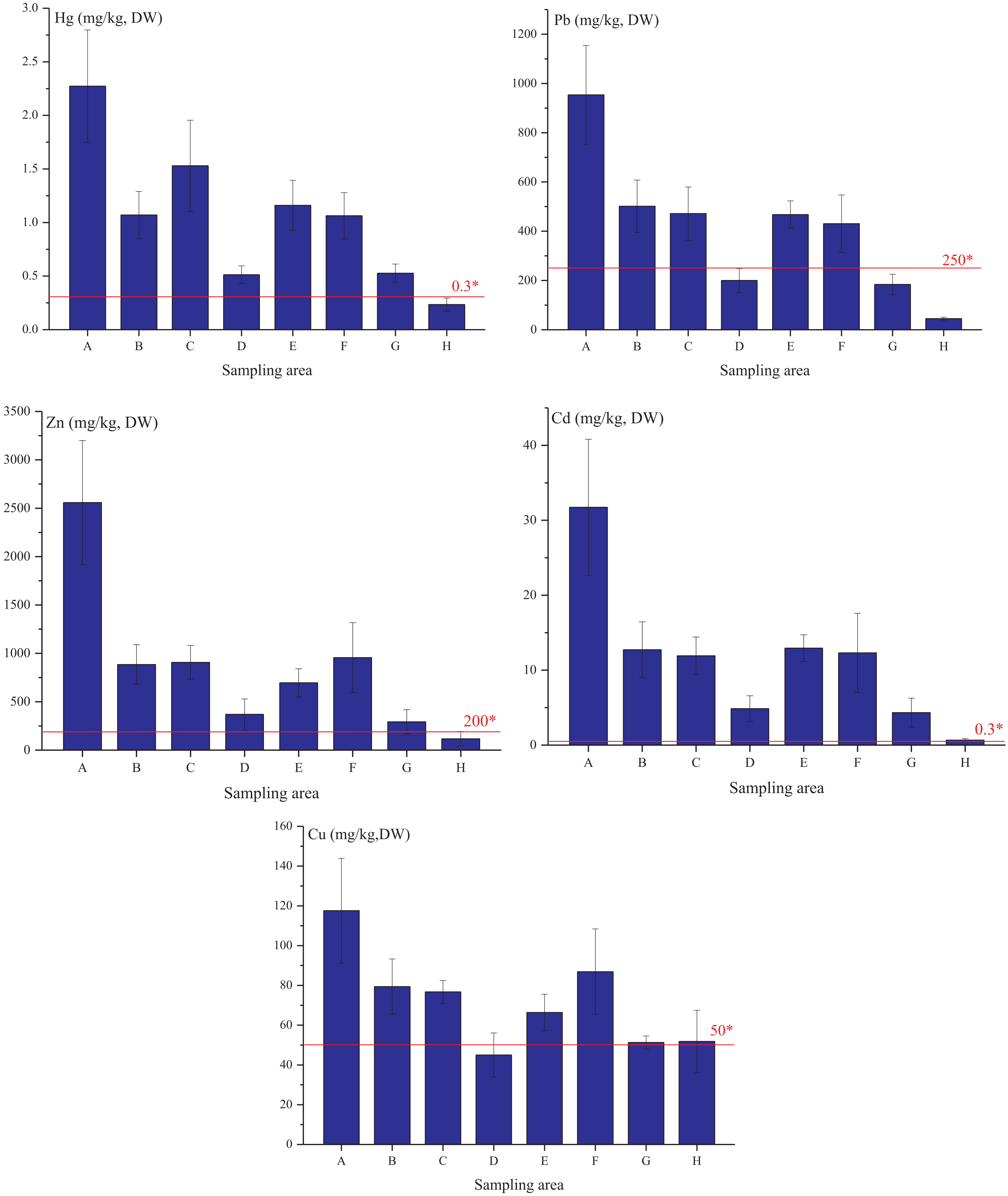

Fig. 3. The average content of heavy metals in surface agricultural soils (0–20 cm) at different areas around the smelter (based on Li et al., 2011). Error bars indicate the standard deviation; * The Grade II of Environmental Quality Standard for Soils of China (GB15618-1995).

cell wall (Liu, 2005; Etesami, 2018), can alleviate the burden of heavy metals in the stems and flowers.

Based on the MAC (Table 2), Pb and Cd in most vegetables at the contaminated areas were beyond the MAC. The percentage of samples that exceed the standards at contaminated areas (A-G) is 75% in leafy and 94% in non-leafy vegetables for Pb; 47% in leafy, 100% in flower

Table 3
Comparison of average content of heavy metals in vegetables growing in various environmental conditions (mg/kg, FW).

Region	Hg	Pb	Zn	Cd	Cu	Reference resources
Zhuzhou smelter	0.004	0.88	10.59	0.21	0.98	This study
Shanghai Industrial Zone	0.004	0.11	4.44	0.02	0.83	Zhou et al. (2017)
Zhejiang province	0.003	0.02	–	0.02	–	Huang et al. (2014)
Dongguan City, Guangdong	0.002	0.13	3.50	0.03	0.39	Cai et al. (2008)
E-iste processing site in Guangdong	–	0.52	12.12	0.24	1.84	Luo et al. (2011)
Multi-metal mining area in Yunnan	–	2.34	8.15	0.31	2.09	Li et al. (2014a)
Zn smelter in Liaoning	0.004	1.91	17.46	1.00	0.97	Zheng et al. (2007b)
Pb smelter in France	–	0.11	4.74	0.15	–	Douay et al. (2013)

vegetables and 64% in non-leafy vegetables for Cd. For Hg, only 7% samples exceed the MAC. At the control area (H), the measured concentration of heavy metals in all vegetable samples fall below the MAC. Comparing similar vegetables samples collected at other industrial areas (Table 3), such as e-waste site, multiple-metal mining/smelting area and urban industrial area, the concentration of heavy metals in this study is significantly higher, especially Pb and Cd, hinting a more serious contaminated status than other circumstances.

The soil in the farmland near the smelter is mainly a large area of acid krasnozem (red earth), with less sand and more mud Li et al. (2011). The soil pH range from 5.0 to 6.2. The heavy metal concentration in agricultural soils increases significantly from far to nearby the smelter, and most exceeded the soil contamination standard (Fig. 3). For example, Pb concentration increased from 184 mg/kg (DW) at the control area (H) to 953 mg/kg (DW) at area A with 0.2 km to the smelter, exceeded nearly 3.8 times of the Grade II (250 mg/kg, DW, soil pH < 6.5) of Environmental Quality Standard for Soils of China (GB15618-1995). Soil contamination is closely related to the total amount of heavy metal in soils, which was mainly caused by the atmospheric deposition (Li et al., 2011) and the heavy metal availability, that influenced by soil properties (such as pH, soil organic matter, Al and Si content). For the same soil, the contamination of vegetables is highly dependent on the vegetable types (Chen et al., 2016; Zhang, 1998).

Different from the obvious spatial distribution of heavy metal concentration in soils within a distance of 4 km to the smelter (Li et al., 2011), the concentration of Hg, Zn and Cd in vegetables at different contaminated areas (A-G) were not distinguished distinctly (Fig. 2), with average of 0.003–0.006, 8.44–12.02 and 0.18–0.26 mg/kg (FW) for Hg, Zn and Cd, respectively. Factors, such as soil property (water, air and pH), roots structure that influence the capacity of activating and absorption of metals, etc. may cause the content of heavy metals in plants (Zhang, 1998; Chen et al., 2016; Shahid et al., 2017). Cu and Zn are essential elements for plants, hence vegetable absorbs these elements from soil dominantly. Hg, Pb and Cd are non-essential elements for plants, so these elements are rarely absorbed by the vegetable from the soil. However, Cd can also be absorbed by the roots and then migrated to stems, leaves and other organs because the high concentration of exchangeable Cd in vegetable soils (Luo et al., 2011).

The edible portions of commercial vegetables are divided into the aboveground part (stem, leave and flower) and the underground part (root or tuber). The average concentrations of heavy metal in different organs of studied vegetables at the contaminated sites (A-G) are compared in Fig. 4. In general, Hg, Pb, Cd and Zn in leaves of carrot, radish and cabbage are comparatively higher than that of roots. This is similar to previous reports by other researchers (e.g. Qian et al., 2009; Lindqvist et al., 1991; Lou et al., 1990). However, Hg, Pb, Cd and Zn in leaves of Chinese cabbage, Chinese white cabbage, celery and lettuce are lower than that of roots. Similar results were reported regarding the distribution of Hg in Chinese white cabbage near a Pb/Zn mine in Guangxi province (Mo et al., 2016). For lettuce, the lowest Hg, Pb, Cd and Zn was found in the stem (edible part); while, for cauliflower, the stem (inedible part) always contains more Hg, Pb, Cd and Zn than other parts. As for garlic sprout, the concentration in root and leaf is almost the same. We didn't compare different parts of leeks in this study because only the leaves of leeks were collected. While the concentration of Cu in roots is higher than other parts for most vegetables except for garlic sprout. The pollution pathways of heavy metals to plants include root absorption (from soil or surface water) and leaf absorption (from atmospheric deposition) (Shahid et al., 2017). In the atmosphere, heavy metal enters the leaf through the adsorption and internalization via the cuticle and the penetration of metals via stomatal pores (Shahid et al., 2017). The size and density of stomata, the size and texture (pubescence and roughness) of leaf and cuticle of plant leaves are the most important factors for heavy metals uptake from ambient air (Abbruzzese et al., 2009; Schreiber and Schönherr, 1992). Besides, plants with massive roots and strong active transport capacity can absorb more heavy metals from the soil and the age of plant organs also has an effect on heavy metal concentration in plants (Shahid et al., 2017; Bondada et al., 2004). Thus, the leaf of white radish, carrot and cabbage can absorb more heavy metal from the atmosphere due to the larger leaf surface; and the large root/tuber of carrots and white radishes may dilute the concentration of heavy metal in the underground parts. The luxuriant root and curled-up leaves (such as Chinese white cabbage) or small proportion of leaves to the aboveground parts (such as celery and lettuce) can lead to higher heavy metal concentration in underground parts compared to the aboveground parts. Of course, this may also be related to the internal structure of plant leaves, such as stomata and cuticle (Järvinen et al., 2010; Johnson et al., 2007). The different concentrations of heavy metal between lettuce and cauliflower may be related to the difference in their active transport capability and moisture content. Isotopic evidence indicates that the amount of heavy metal absorbed via foliar transported to the root tissues was $< 1\%$ (Shahid et al., 2017; Dollard, 1986; Tso and Fisenne, 1986). Additionally, Hg and Pb in the aboveground parts, especially leaves, mainly come from the atmosphere (Ao et al., 2017; Bi et al., 2009; Yin et al., 2013). Therefore, the source of heavy metals in vegetables may be related to both soil absorption and atmosphere deposition. However, the absorption and translocations process are complex and dependent on specific vegetables and elements.

3.2. Correlation analysis of heavy metal in vegetables and soils

The correlations among the measured concentrations of heavy metals in vegetable and soil samples are shown in Table 4. The concentrations of all elements in soils are significantly correlated with each other ($p < 0.001$), suggesting the heavy metals are derived from the same source, i.e., the Pb/Zn smelter. Heavy metal contents in soil represent the intensity of atmospheric deposition in the surrounding environment and therefore have a stronger correlation with the smelter. The correlations between of the concentration of Hg, Pb, Zn and Cd in vegetables are only in the range of 0.29–0.50 and Cu only significantly correlated with Cd, which are comparatively weaker than that in soils (0.81–0.98, $p < 0.01$). This indicates that heavy metal accumulation is

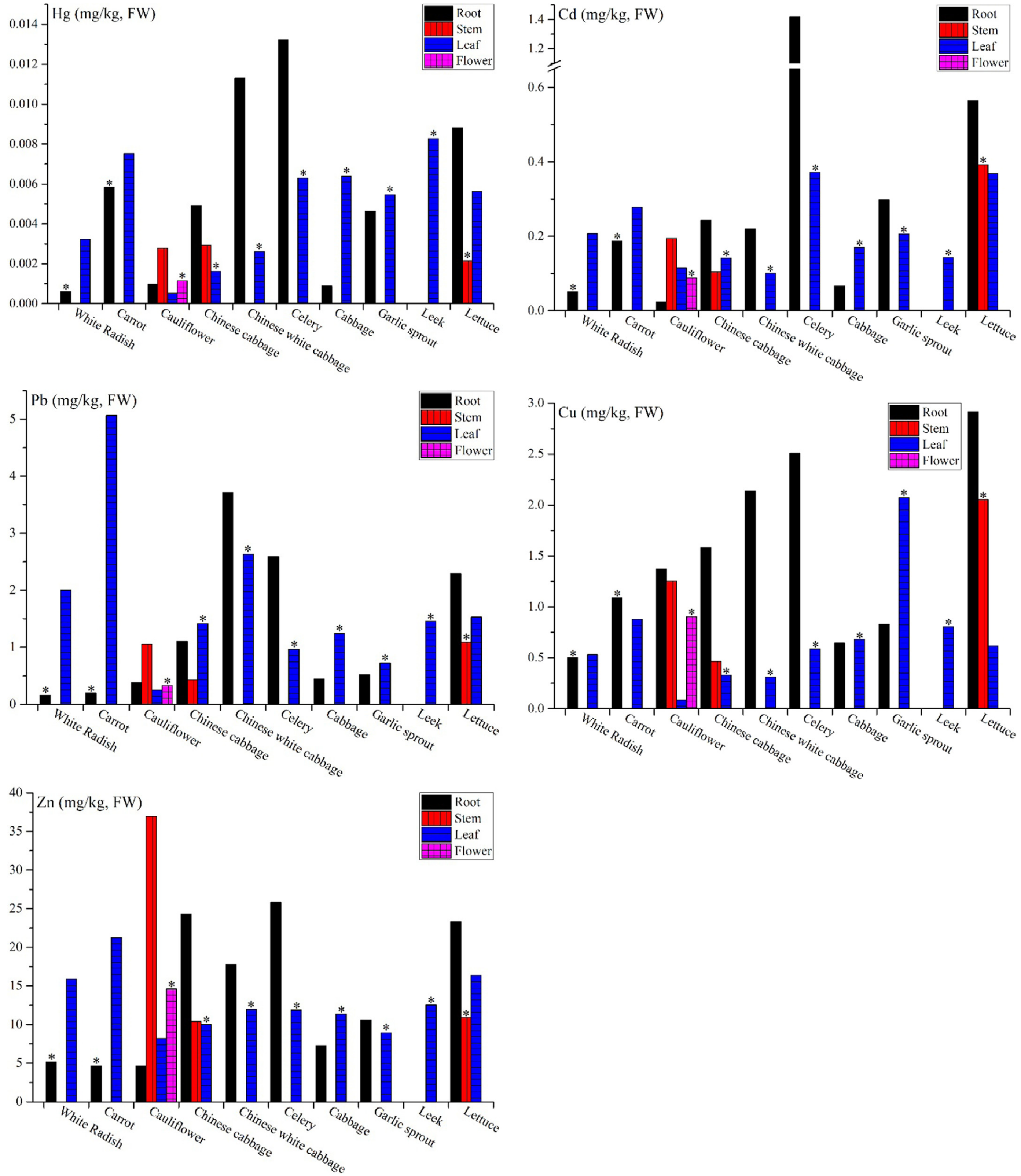

Fig. 4. Comparison of heavy metal concentrations in different parts of vegetables at the contaminated areas (A-G). * Edible parts of the vegetable.

influenced by more complicated factors including the physiology of each vegetable species, soil characteristics, availability of heavy metals in soils, as well as the exposure pathways (e.g., soil vs. atmosphere) (Douay et al., 2013; Chen et al., 2016).

3.3. Assessment of vegetable contamination and associated health risk

3.3.1. Vegetable contamination status

The level of contamination of the edible part of vegetables is assessed by the single-factor and comprehensive pollution index. The

Table 4
Correlation coefficients between heavy metal concentrations in soils (mg/kg, DW) and edible part of vegetables (mg/kg, FW).

Soil	Hg	Pb	Zn	Cd	Cu	Veget.	Hg	Pb	Zn	Cd	Cu
Hg	1					Hg	1				
Pb	0.94^{**}	1				Pb	0.40^{**}	1			
Zn	0.89^{**}	0.95^{**}	1			Zn	0.36^{**}	0.50^{**}	1		
Cd	0.91^{**}	0.98^{**}	0.98^{**}	1		Cd	0.29^{*}	0.41^{**}	0.44^{**}	1	
Cu	0.81^{**}	0.89^{**}	0.91^{**}	0.92^{**}	1	Cu	ns	ns	ns	0.36^{**}	1

ns, not significant.

* $p < 0.05$.

** $p < 0.01$.

Table 5
The result of single-factor and comprehensive pollution index assessment for the edible parts of vegetables at different sampling areas.

Sampling area	Hg		Pb		Zn		Cd		Cu		*P*	Class
	Pi	Class	*Pi*	Class	*Pi*	Class	*Pi*	Class	*Pi*	Class		
A	0.30	I	6.42	V	0.48	I	1.27	III	0.08	I	4.70	V
B	0.45	I	2.90	IV	0.60	I	2.09	IV	0.07	I	2.35	IV
C	0.35	I	3.40	V	0.42	I	1.49	III	0.12	I	2.62	IV
D	0.33	I	1.93	III	0.58	I	1.63	III	0.19	I	1.57	III
E	0.62	I	7.68	V	0.58	I	1.24	III	0.09	I	5.62	V
F	0.39	I	5.37	V	0.47	I	1.57	III	0.08	I	3.96	V
G	0.32	I	1.00	II	0.58	I	1.47	III	0.07	I	1.21	III
H	0.17	I	0.37	I	0.17	I	0.29	I	0.06	I	0.31	I

Class I, safe; Class II, clean; Class III, slightly polluted; Class IV, moderately polluted; Class V, heavily polluted.

Table 6
The result of single-factor and comprehensive pollution index assessment for the edible part of different vegetables at the contaminated areas (A-G).

Vegetable	Hg		Pb		Zn		Cd		Cu		*P*	Class
	Pi	Class	*Pi*	Class	*Pi*	Class	*Pi*	Class	*Pi*	Class		
Lettuce(N = 7)	0.21	I	10.90	V	0.55	I	3.93	V	0.21	I	8.35	V
Chinese white cabbage (N = 1)	0.26	I	8.76	V	0.60	I	0.50	I	0.03	I	6.36	V
Leek(N = 2)	0.83	II	4.85	V	0.63	I	0.72	II	0.08	I	3.57	V
Cabbage(N = 7)	0.64	I	4.13	V	0.57	I	0.85	II	0.07	I	3.06	V
Cauliflower(N = 4)	0.11	I	3.26	V	0.73	II	1.77	III	0.09	I	2.51	IV
Celery (N = 7)	0.63	I	3.21	V	0.59	I	1.86	III	0.06	I	2.46	IV
Chinese cabbage(N = 7)	0.28	I	3.03	V	0.59	I	0.68	I	0.05	I	2.25	IV
Garlic sprout(N = 45)	0.55	I	2.41	IV	0.45	I	1.03	III	0.21	I	2.02	IV
Carrot (N = 2)	0.58	I	1.96	III	0.23	I	1.87	III	0.11	I	1.67	III
White Radish(N = 5)	0.06	I	1.58	III	0.26	I	0.51	II	0.05	I	1.17	III

Class I, safe; Class II, clean; Class III, slightly polluted; Class IV, moderately polluted; Class V, heavily polluted.

results are shown in Tables 5 and 6. The contamination level of Hg, Zn and Cu at areas A-H is classified to Class I (safe), while the level of Pb and Cd at areas A-G are reached to Class III-V (slightly to heavily polluted). Based on the comprehensive pollution index (*P*), the rank of pollution at the sampling areas is E > A > F > C > B > D > G > H. Although areas B, C, D and E are not directly downwind, they are reached to class III–V (slightly to heavily polluted) because of the proximity to the smelter. Besides, other source such as train transportation of Pb/Zn ores resulted in area C to Class IV (moderately polluted). The control area shows no evidence of heavy metal contaminations. Based on the comprehensive pollution index, Fig. 5a shows that vegetables near the smelter were polluted to varying degrees, and the polluted areas were mainly located in the NW (main wind direction) and SE (secondary wind direction) side of the smelter. The pollution of Pb is the most serious (Fig. 5b), followed by Cd. The spatial distribution of Cd pollution is weakly correlated with the wind direction (Fig. 5c).

In Table 6, the comprehensive pollution index (*P*) of studied metals for vegetables is tabulated. The level of Hg, Zn and Cu contamination is classified as Class I (safe) for lettuce, Chinese white cabbage, cabbage, celery, Chinese cabbage, garlic sprout, carrot and white radish or Class II (clean) for leek and cauliflower. The contamination level of Pb and Cd appeared to be more serious: mostly in Class V (heavily polluted) for Pb. Based on the comprehensive pollution index, the pollution level rank is lettuce > Chinese white cabbage > leek > cabbage > cauliflower > celery > Chinese cabbage > garlic sprout > carrot > white radish. In this sequence, lettuce, Chinese white cabbage, leek and cabbage are in Class V (heavily polluted) and the pollution of lettuce and Chinese white cabbage (*P*:6.4–8.4) are much higher than that of the others (*P*:1.1–3.6). Cauliflower, celery, Chinese cabbage and garlic sprout are in Class IV (moderately polluted); carrot and white radish are in Class III (slightly polluted).

3.3.2. *Human health risk*

The calculated EDI, THQ and HI are shown in Table 7. EDI of Hg, Pb, Zn, Cd and Cu for samples collected at contaminated areas (A-G) are significantly greater than that of the control area (H) for both adults and children. HI of the contaminated areas is for both adults (3.66) and for children (3.14) exceeds 1, suggesting high health risks by consuming the contaminated vegetables. As a comparison, HI of the control area is regarded as safe both for adults (0.70) and children (0.60).

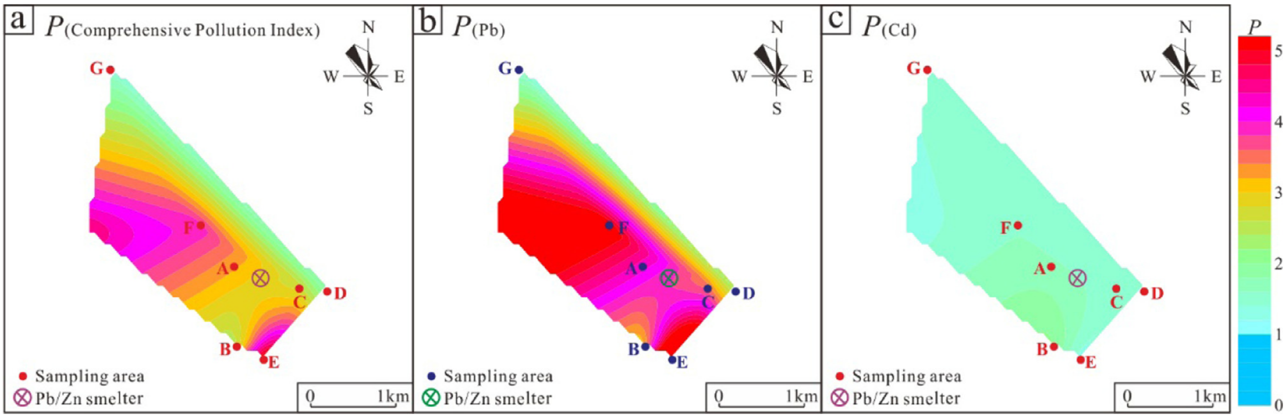

Fig. 5. Spatial distribution of vegetable pollution based on Pollution Index. a, Comprehensive Pollution Index; b, Pollution Index of Pb; c, Pollution Index of Cd.

Table 7
EDI, THQ, and HI values of heavy metal for local residents via vegetable consumption near the smelter.

Index	Population	Region	Hg	Pb	Zn	Cd	Cu
EDI (μg/kg/day)	Adults	Contaminated areas (A-G)	0.028	6.103	73.673	1.461	6.820
		Control area (H)	0.010	0.626	23.458	0.313	4.384
	Children	Contaminated areas (A-G)	0.024	5.230	63.133	1.252	5.844
		Control area (H)	0.009	0.537	20.102	0.268	3.757
RfD (μg/kg/day)	All people	All areas (A-H)	0.7	3.5	300	1	40
THQ	Adults	Contaminated areas (A-G)	0.040	1.744	0.246	1.461	0.170
		Control area (H)	0.015	0.179	0.078	0.313	0.110
	Children	Contaminated areas (A-G)	0.034	1.494	0.210	1.252	0.146
		Control area (H)	0.013	0.153	0.067	0.268	0.094
HI	Adults	Contaminated areas (A-G)	3.66		Control area (H)	0.70	
	Children	Contaminated areas (A-G)	3.14		Control area (H)	0.60	

RfD quotations from USEPA (2009).

The contribution of analyzed heavy metals to THQ is shown in Fig. 6 and Table 7. The contribution of Pb and Cd is high at 47.6% and 39.9%, respectively. This is followed by Zn (6.7%) and Cu (4.7%), and then Hg at 1.1%. It is clear that Pb and Cd are the most significant heavy metal pollutant caused by the smelter. Children are typically more vulnerable than the adults in case of heavy metal exposure through diet (Wang et al., 2015). At the control area, given the relatively safe in terms of contamination level, Pb and Cd still account for 25.8% and 45.1% of HI, respectively.

Our research indicated the local residents can be excessively exposed to Pb and Cd through vegetable consumption alone. Other exposure pathways via consumption of grain, meat, fruit, milk, drinking water, and contact with air further enhance the exposure to heavy metals (Zheng et al., 2007b; Järup, 2003; Simsek, 2000). For example, unhealthy exposure to Cd, Pb and As through rice consumption has been documented (Williams et al., 2009; Lei et al., 2015). Therefore, more studies from other exposure ways in the study area is needed in the future to obtain a panorama for this Pb/Zn smelter.

4. Conclusions

The heavy metal contamination of 10 vegetables at 8 sampling sites

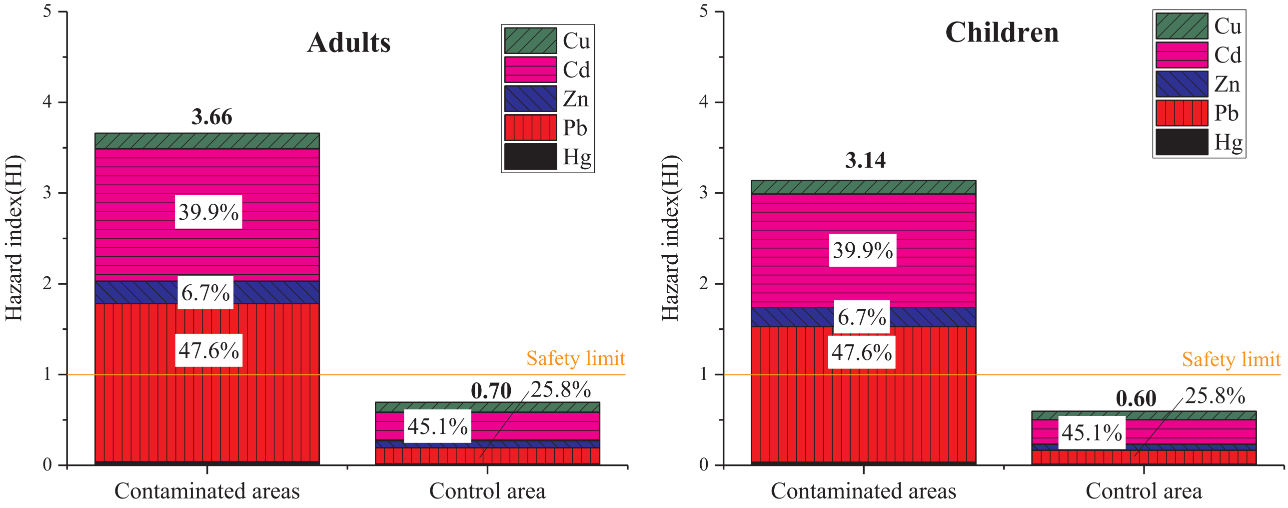

Fig. 6. Hazard index of consumption the vegetables at the contaminated and control areas near the smelter.

within 15.5 km around the Pb/Zn smelter in central China was investigated in this study. We compared the level of heavy metal contamination in vegetables near a Pb/Zn smelter, analyzed the relationship between soil pollution and distance from smelter, and assessed the health risk associated with consuming the vegetables.

The primary heavy metal contaminants in vegetables grown nearby the smelter are Pb and Cd. The contamination level caused by Hg is relatively moderate. The edible part of leafy vegetables is contaminated more seriously compared to non-leafy vegetables. It was found elevated heavy metal concentrations in the roots of Chinese cabbage, Chinese white cabbage, celery and lettuce. Particularly, leaves of white radish, carrot and cabbage and stem of cauliflower contain the highest concentrations of heavy metals. The contamination level in the edible part of vegetables decreased in the following order: lettuce, Chinese white cabbage, leek, cabbage, cauliflower, celery, Chinese cabbage, garlic sprout, carrot and white radish. The spatial distribution of heavy metal pollution is closely related to the wind direction. The hazard index of adults and children for consumption of vegetables planted at contaminated areas near the smelter is 3.66 and 3.14, respectively. The contamination of heavy metals in these vegetables, either through the uptake from the foliage or the contaminated soil, poses a health concern to local residents. Reducing the heavy metal emission and changing the agriculture practice near a known point source are needed to protect the environment and human health.

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