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Source apportionment of heavy metal and their health risks in soil-dustfall-plant system nearby a typical non-ferrous metal mining area of Tongling, Eastern China☆

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ABSTRACT

The agricultural land-atmospheric dustfall-plant system around the mining area is at high risks of heavy metal pollution caused by mining-smelting activities. In this study, 118 samples (including rhizospheric soils, background soils, soil-forming parent rocks, crops, vegetables, medicinal plants and atmospheric dustfall) were collected nearby Tongling Cu-Fe-Au mining area, Eastern China. We studied the concentrations, migration, sources, and health risks through consumption of two main crops (corn and rice), six kinds of vegetables, and medicinal plants (Fengdan, *Paeonia ostii*) for six metal elements (Cu, Zn, Cr, Cd, Pb and Hg). Results revealed Cr and Cd in soils, and Cd, Cr, Pb, Cu and Zn in dustfall showed a relatively high contamination degree. The mean contents of Cr and Pb in corn kernels, as well as Cd, Cr and Pb in rice grains and all vegetables, and Cr in Fengdan cortex moutan exceeded the corresponding food safety limits in China. The transfer capability of Cr in corn kernels and rice grains, Pb in edible vegetables, and Cd in cortex moutan were the strongest, respectively. Health risk assessment results showed Cr had the greatest non-carcinogenic risk, followed by Pb and Cd. The results of pearson's correlation analysis (CA), hierarchical cluster analysis (HCA), and principal component analysis (PCA) indicated Zn-Cr, Pb and Cd-Cu-Hg in the plants might derive from different geochemical end-members. Source apportionment based on lead isotope showed that mining-smelting activities were the major source of Pb in atmospheric dustfall and agricultural soils, with the average contribution rates of 66% and 50%, respectively. Vehicle emissions from diesel fuels (50%–68%) and mining-smelting activities (16%–25%) contributed mainly to Pb accumulation in plants. Hence, our study suggested the accumulations of Pb in plants might be mainly from the direct foliar uptake of atmospheric Pb related to vehicle emissions and mining-smelting activities.

1. Introduction

With metal mineral resources exploited and utilized by human beings, the environment was seriously contaminated by heavy metals (HMs) (Li et al., 2014). Mining and metallurgical activities, including digging, transporting, various kinds of industry beneficiation process and smelting, have caused a large amount of HMs to be released into the surrounding environment (Wang et al., 2004; Sun et al., 2013, 2014; Li et al., 2014; Oyebamiji et al., 2018). Long-time unconscious exposure to metal pollutants can have potential harm to the local inhabitants (Bi et al., 2018). Acting as dispensable elements, Pb, Cd, and Hg have shown carcinogenic effect at lower concentrations (Oliver, 1997; Żukowska and Biziuk, 2008; Sun et al., 2016). Cu, Zn, and Cr are considered to be essential and may be

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ingested by plants and humans at a safe range (FAO, 1995; USEPA, 1995; Tchounwou et al., 2012). However, they can be toxic to human beings and other animals when they exceed the safe threshold levels. These metals can cause hazards such as headache, neurologic involvement and liver disease (USEPA, 2000; Kabata-Pendias and Mukherjee, 2007).

Soil-to-plant transfer of HMs through root uptake and their subsequent consumption, together with inhalation of particles polluted by HMs are the major exposure pathways for humans in contaminated areas. Once these metal elements are migrated into soils, they will be enriched in local crops (i.e., wheat and rice, Zeng et al., 2008), vegetables (i.e., tomato, chili, eggplant, green bean, cucurbit, cucumber, etc., Liu et al., 2009; Xu et al., 2013; He et al., 2015), and also other plants (e.g., medicinal plant) that can influence the health of the local residents (Zheng et al., 2007; Shen et al., 2017).

However, soils are not the only heavy metal pollution sources for plants (Bi et al., 2018). Dustfall with metal elements released through metallurgical industries and mining activities may be directly/indirectly absorbed on the above-ground tissues of plants via wet-dry atmospheric deposition. In addition, metal elements in atmospheric dustfall are very easy to get into the top surface of soils in crop and vegetable areas through wind-driven transport and surface runoff (Chen et al., 2010; Bi et al., 2018). Hence, soil-to-plant and dustfall-to-plant transportation are two major pathways of metal elements entering into the plants. Although some results for heavy metal pollution in soil-vegetable systems have been reported in recent years (Liu et al., 2005; Zhuang et al., 2009; Pan et al., 2016; Hu et al., 2017), there have been few detailed and systematic studies of the sources and risks for HMs in the more complex soil-dustfall-plant system for the mine environment.

Tongling, located in the middle-south of Anhui Province, eastern China, is a very important industrial city famous for the mining and smelting of nonferrous metals. Previous studies have shown that soils, vegetables, medicinal plants (Fengdan, *Paeonia ostii*), surface and groundwater in the Tongling mining area have suffered from varying degrees of heavy metal pollution (e.g., Xu et al., 2013; Shen et al., 2017). Information on the migration enrichment behaviors and the potential health risks of HMs in the soil-dustfall-plant system near the mining area are still insufficient. Moreover, there have no studies that focused on analyzing the sources of potentially toxic metals in plants. Therefore, it will be necessary to determine the levels and sources of metal elements, and to evaluate potential health risks in soil-dustfall-plant system.

The objectives of this work are to: (1) investigate the levels and migration of six metal elements in soil-dustfall-plant system downstream along the Fenghuangshan- Xinqiao mine in the Tongling area; (2) identify the dominant sources of HMs by statistical analysis methods; (3) estimate the relative contribution of different sources in plants using lead isotopic ratios; and (4) discuss the potential risks of HMs via consuming edible plants grown in Tongling mining area.

2. Materials and methods

2.1. Study area

Tongling between 117°42′00″-118°10′60″ E and 30°45′12″-31°07′56″ N (Fig. 1), belongs to the north subtropical humid monsoon climate. Annual mean temperature, rainfall and wind speed are ~16.4 °C, ~1400 mm and 3.1 m/s (the maximum is 24 m/s), respectively. The dominant wind direction in Tongling area is obviously regular throughout the year, with NE wind in winter (20%) and SW wind in summer (18%). Soil types in Tongling area are chiefly red soils. Geologically, Tongling polymetallic deposits, located at the middle section of the famous middle-lower Yangtze River Cu-Fe-Au belt (Wang et al., 2018) in central-eastern China with more than 3000 years of mining and smelting history, whose metal minerals mainly contain magnetite, hematite, chalcopyrite, pyrite, siderite, as well as small amounts of metal sulfide-bearing minerals (e.g., chalcocite, galena, and sphalerite, etc., Xu et al., 2008). Weathering and oxidation process of these metal sulfides will release a large amount of HMs (Cu, Cd, Pb, Cr, etc.) and acid anions (carbonates and sulfates) into the surrounding environment of the mining area under extreme acidic conditions, which would post a health risk to local residents.

2.2. Sample collection and pre-treatment

Based on the consideration of local main crops, vegetables and medicinal plants and their spatial distribution in Tongling mining area, three categories of representative plants grown in the study area were collected in summer during June and August 2017, including 15 samples of two main crops (corn and rice), 46 samples of six kinds of vegetables (e.g., chili, eggplant, tomato, green bean, cucumber and cucurbit) and 7 samples of medicinal plants (Fengdan, *Paeonia ostii*). At every plant sampling site we randomly collected 3–5 replicate plants to make up a composite sample. Rhizospheric soils were obtained by gently shaking the root for a while. Atmospheric dustfall samples were gathered in dust barrels near the mining areas and suspected pollution enterprises according to the prevailing wind direction during the sampling season. Soils and dustfall were combined into a composite sample using 5 subsamples, respectively. Samples collected were all placed into polythene sealing bags and sent to the Sample Pretreatment Lab within 4 h. Total 29 rhizospheric soil samples corresponding to plants investigated in the mining area, 1 background surface soil sample located in the hillside of Fenghuangshan mountain, 2 soil-forming parent rocks (granodiorite and granite), 68 plant samples involving 9 species (*Zea mays* L., *Oryza sativa*, *Capsicum annuum*, *Solanum melongena*, *Lycopersicon esculentum*, *Vigna unguiculata*, *Cucumis sativus* L., *Lagenaria siceraria (Molina) Standl.*, and *Paeonia ostii*), along with 18 atmospheric dustfall samples collected using a dust cylinder fixed 2 m above the roadside pole were acquired in this study (Fig. 1 and Table S1).

Every fresh plant sample was cleaned in tap water, rinsed 3–5 times with Milli-Q ultrapure water and then dissected into root, stem, leaf, skin and/or pulp, and recorded the fresh weight. Each separated plant sample was then desiccated for 0.5 h at 105 °C and heated to constant weight at 70 °C for 72 h, then ground using a super-centrifugal grinder (ZM 200, Retsch, Germany). The soil and dustfall samples were dried using a freezedryer under vacuum and −45 °C, then triturated and sieved to 200 mesh. All samples were stored according to specifications before chemical analysis.

2.3. Chemical analysis and instrument measurement

2.3.1. Determination of total metal elements

Determination of soil pH was carried out in a soil-water mixture system by a digital pH meter (HACH HQ40d). Hg was quantified using a direct mercury analyzer (DMA-80, Milestone). The contents of other five HMs (Cu, Zn, Cd, Cr and Pb) were measured using an inductively coupled plasma mass spectrometer (ICP-MS, Agilent Technologies 7500 Series) with various digestion methods. Soils and dustfall were digested by HNO_3-HF-$HClO_4$ (sub-boiling grade), volume ratio 5:5:3, respectively. Plant samples were digested with HNO_3-$HClO_4$ (sub-boiling grade), volume ratio 10:1. Finally, each dissolved sample was transferred to a cleaned PET bottle with 1 g In added as an internal standard, then diluted to 80 g before ICP-MS analysis.

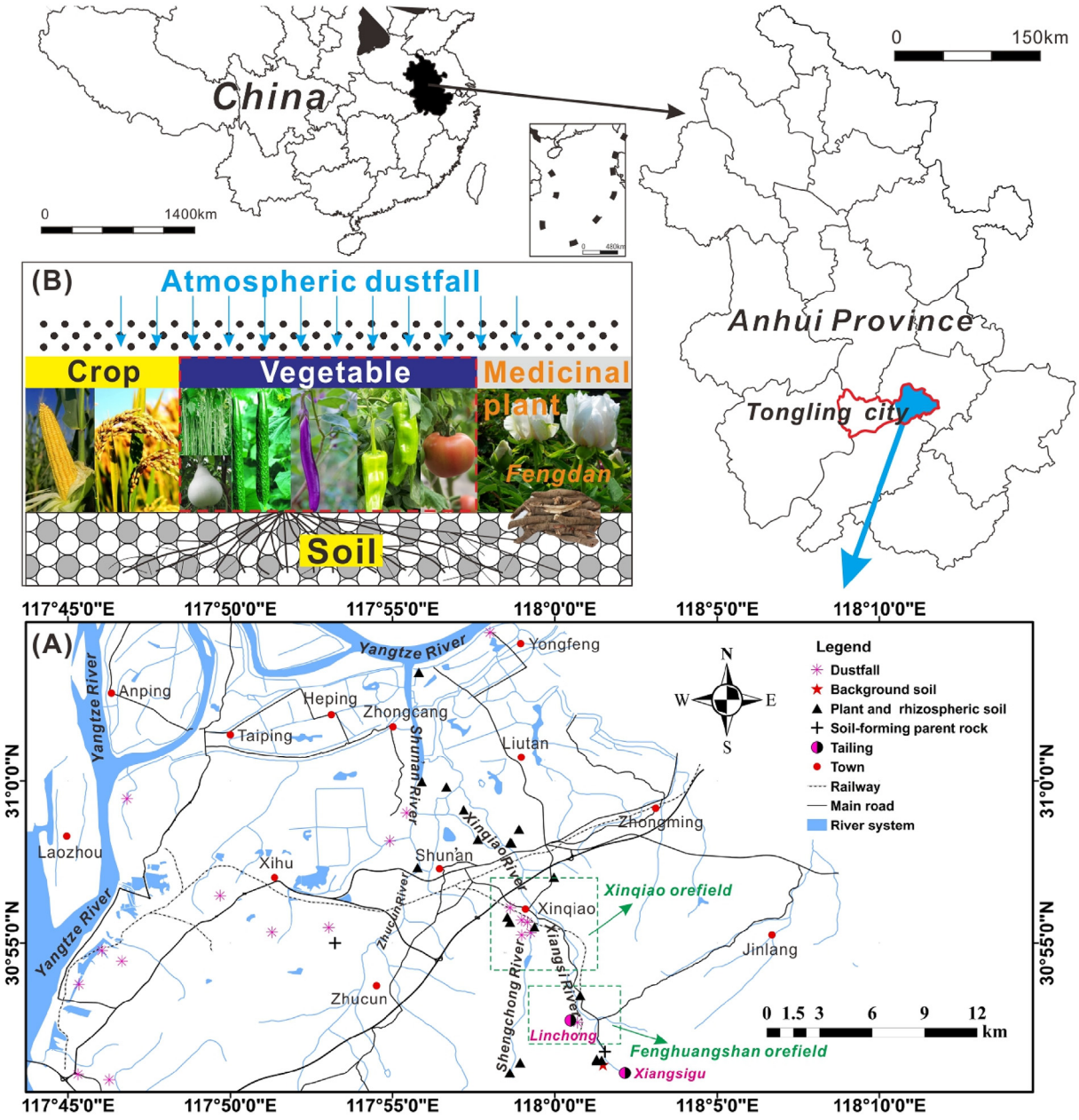

Fig. 1. Sampling sites (**A**) and soil-dustfall-plant system (**B**) in the Tongling mining area, southeastern China.

During the analysis of metal elements, Chinese standardized reference materials including GBW07405 (GSS-5) for soils and dustfall, GBW (E) 100349 for plants were used for monitoring the whole analysis procedure, respectively. The precision represented by the relative deviations of the data were <5% for Cu, Pb, Zn and Hg, and <10% for Cd and Cr after three repeated experiments of standardized reference materials, respectively. Quality assurance (QA) and Quality control (QC), which showed good precision throughout, were conducted using simultaneous, reagent blanks and duplicates (accounting for 10–20% of the total number of samples). The recovery rates of the reference materials were about 90–110% for all the metals.

2.3.2. Measurement of Pb isotope

To analyse Pb isotopic composition, soil/dustfall sample (~100 mg) was digested in Teflon® pots on a hotplate using a mixture of distilled acids including 8 mL HNO_3 and 2 mL HF. For plant samples, to ensure sufficient semaphores for lead isotopic measurement, the mass of samples had to be greater than for the other analyses. The dried residue was re-dissolved in HBr + HNO_3 solution for all samples and then loaded into a column with 50 mm of AG1-X8 anion exchange resin to separate Pb, and the extracted Pb was purified in a second column. The lead isotopic measurement was carried on a solid source thermal ionization mass spectrometer (TIMS, Finnigan MAT 262, Germany) at the University of Science and Technology of China (USTC). Analysis results of the lead isotope for standard solutions of NBS 981 were consistent with the recommended values.

2.4. Data analysis

2.4.1. Assessment of pollution levels

Index of geoaccumulation (I_{geo}) was used to estimate the accumulation level of the targeted metal element in soils or dustfall (Wei and Yang, 2010). The formula was originally defined as (Muller, 1969):

$$I_{geo} = \log_2\left[\frac{SC_i}{1.5BV_i}\right] \quad (1)$$

where SC_i was the concentration of metal element i in the investigated samples (soils and dustfall), BV_i was the background value of the metal element i in Tongling soils (AEMC, 1992). 1.5 was the modified proportional coefficient and was used to correct inevitable variations resulted from diagenesis. Muller (1969) proposed 7 classes of soil quality from the calculated I_{geo} value for each metal, and the detailed classifications were presented in Table S2.

A pollution index (PI) of single metal element and a mean pollution index (MPI) of the six metal elements were calculated in this study to further assess the pollution levels, as in the following equation (Wei and Yang, 2010):

$$PI_i = \frac{SC_i}{BV_i}$$

$$MPI = \frac{1}{n}\sum_{i=1}^{n} PI_i \quad (2)$$

where $n = 6$, SC_i was the concentration of metal element i in the investigated samples (soils or dustfall), BV_i was the background value of the targeted metal element i. The values of BV_i for Cr, Zn, Pb, Cu, Cd and Hg were 88.56, 85.58, 47.79, 32.15, 0.09 and 0.05 mg/kg in Tongling area, respectively (AEMC, 1992). According to calculated MPI values, four classifications of environment quality were listed in Table S2 (Wei and Yang, 2010).

2.4.2. Migration of metal elements

The transfer factor (TF) can reflect the potential risk of human exposure to heavy metal elements from soils to edible plant tissues through food chain transmission. The values were obtained on the base of the following equation (Khan et al., 2010):

$$TF = \frac{C_p}{C_s} \quad (3)$$

where C_p was the concentration of metal element in the tissue of plants based on dry weight; C_s was the concentration of metal element in related rhizospheric soil based on dry weight.

2.4.3. Calculation of contributions for separate Pb sources

A binary mixing model could be used to calculate the contribution rate of each end-member to lead in samples (Shotyk et al., 1997):

$$X_A\% = \frac{R_S - R_B}{R_A - R_B} \times 100 \quad (4)$$

where X_A represented the percentage contributions (%) of source A. R_S, R_A and R_B represented the $^{206}Pb/^{207}Pb$ ratio of sample, source A, and source B, respectively.

If the three major sources of pollution and the isotopic characteristics of samples are known, a ternary mixing model can be established using the following equations (Chiaradia et al., 1997):

$$R_S = f_1R_1 + f_2R_2 + f_3R_3$$

$$N_S = f_1N_1 + f_2N_2 + f_3N_3$$

$$f_1 + f_2 + f_3 = 1 \quad (5)$$

where R_S represents the $^{206}Pb/^{207}Pb$ ratio of sample; N_S represents the $^{208}Pb/^{206}Pb$ ratio of sample; R_1, R_2, R_3 represent the $^{206}Pb/^{207}Pb$ ratio of source 1, 2, 3, respectively; N_1, N_2, N_3 represent the $^{208}Pb/^{206}Pb$ ratio of source 1, 2, 3, respectively; f_1, f_2, f_3 represent the percentage contributions (%) of source 1, 2, 3, respectively.

2.4.4. Calculation of health risk

The mean values of estimated daily intake (EDI) are used to quantify human dietary exposure to HMs. The EDI were acquired using the following equation (Bi et al., 2018):

$$EDI(\mu g/kg/day) = \frac{C_m \times W_p}{B_w} \quad (6)$$

where C_m represented the concentration of each metal element in the edible part of plants (mg/kg, all plants based on wet weight except for medicinal Fengdan based on dry weight); W_p was the daily intake of the edible part of plants (16 g/day for maize, Fan et al., 2018; 345 g/day for vegetables, Khan et al., 2010; 12 g/day for cortex moutan, Shen et al., 2017; and 372 g/day for rice, Zhuang et al., 2009); B_w was the average body weight of Tongling adult residents in the present investigation (60 kg).

A target hazard quotient (THQ) of each metal element to estimate the potential hazard to local residents was calculated in this study using the following equation:

$$THQ = \frac{F_e \times D_e \times EDI}{RfD_o \times A_t} \quad (7)$$

where F_e and D_e were the frequency (365 days/year) and duration (70 years) of the average exposure, respectively; RfD_o was the reference dose of daily oral exposure, of which the values for Cu, Zn, Cr and Cd are 40, 300, 3 and 1 μg/kg/day, respectively (USEPA, 2018). The values of RfD_o for Pb and Hg were 1.5 μg/kg/day and 0.14 μg/kg/day obtained from EFSA (2010) and JECFA (2010), respectively; A_t is the average time for non-carcinogenic effects (365 days/year × number of exposure years, assuming 70 years in the study). A THQ value ≥ 1 was considered to be potential harms for the exposed population; while $THQ < 1$ was relatively safe.

2.4.5. Statistical analysis

Statistical analysis was processed with SPSS version 22 (SPSS Inc., Chicago, USA) and Microsoft Office Excel 2007. The plotting and typesetting of all the graphs were done with Origin 8.5 (OriginLab Corp., Massachusetts, USA) and CorelDRAW X5. Hierarchical cluster analysis (HCA) was employed to systematically classify the metal elements in all samples. Pearson's correlation analysis (CA) and Principal component analysis (PCA) were used to explore the sources and exposure pathways of metal elements in three types of environmental media (rhizospheric soils, atmospheric dustfall and plants), with the levels of significance ($p < 0.05$ and $p < 0.01$).

3. Results and discussion

3.1. HMs in soil, dustfall and plant

The six targeted metal concentrations in investigated rhizospheric soils, atmospheric dustfall and plants were listed in Table 1

Table 1
Heavy metal concentrations in rhizospheric soils and atmospheric dustfall in the study area (mg/kg).

Location		pH	Cu	Zn	Cd	Cr	Pb	Hg
Rhizospheric soil (n = 29)	Minimum value	2.65	17.10	42.95	0.05	31.48	ND	0.03
	Maximum value	7.77	340.43	859.25	10.20	2647.50	390.25	0.17
	Mean	6.20	95.58	192.12	1.34	202.22	72.61	0.06
	SD	1.27	80.61	154.91	2.11	486.53	83.67	0.03
Atmospheric dustfall (n = 18)	Minimum value		333.20	832.90	1.85	58.94	108.60	0.04
	Maximum value		3579	76310	12.34	526.20	394.56	0.23
	Mean		1422.71	6228.23	5.47	285.65	244.64	0.12
	SD		1002.52	17116.66	5.47	110.31	72.15	0.05
Background values of Tongling[a]			32.15	85.58	0.09	88.56	47.79	0.05
Chinese standard for agriculture soil[b]			≤50.00	≤200.00	≤0.30	≤150.00	≤90.00	≤1.8

SD = Standard deviation, ND = Below detection limit.
[a] Research report on soil environmental background in Anhui province (AEMC, 1992).
[b] GB 15618-2018 (MEEPRC, 2018).

and Table S3. The rhizospheric soils had a limited range of pH values, with pH varying from acidic to nearly neutral (mostly between 6.0 and 7.5), consistent with the pH values (6.5–7.5) previously reported by Xu et al. (2013) for the vegetable soils from the Tongling mining area. In comparison with the values of the five HMs (Cd, Cu, Zn, Cr, Pb and Hg) in the Tongling background soil (AEMC, 1992), the average content of HMs in soils was 14.89, 2.97, 2.24, 2.28, 1.52 and 1.2 times of the background value, respectively; the average contents in atmospheric dustfall exceeded the corresponding soil background values by 3.23, 44.25, 72.78, 60.78, 5.12 and 2.4 times, respectively. In contrast to the Chinese agricultural soil environmental quality control Criteria (GB15618-2018; MEEPRC, 2018), the average content of only three (Cd, Cu and Cr) HMs in soils was 4.47-, 1.91- and 1.35-fold of the suggested corresponding maximum allowable levels, respectively. However, the mean content of above five HMs (Cd, Cu, Zn, Cr and Pb) in dustfall was all significantly higher than the threshold values issued by MEEPRC (2018), respectively, suggesting comparatively higher degrees of contamination for these five HMs in dustfall in this region. The Hg levels in soils and dustfall were slightly higher than the background values (AEMC, 1992) but obviously lower than threshold values (GB15618-2018; MEEPRC, 2018), respectively, indicating there was very weak Hg pollution in this region.

The pollution degrees in soils and dustfall commonly showed the Cd > Cu > Zn > Pb > Hg > Cr sequence in this study (Table 2). The I_{geo} and *PI* values of Cd for soils (I_{geo} = 2.31, *PI* = 14.89) and dustfall (I_{geo} = 5.2, *PI* = 60.75) were the highest among the investigated HMs, indicating the soils and dustfall in this region were heavily contaminated by Cd. Values of *MPI* in the soils and dustfall were 4.19 and 31.41 (Table 2), placing them in the high and extremely high pollution classes, respectively, and together indicating that soils and dustfall in the study were severely polluted by these HMs.

The concentrations of six targeted HMs accumulated in the edible parts of plants (i.e., leaves, grains, pulps, etc.), would have an indirect/direct influence on local people's health. Differences between metal accumulation capabilities and soil characteristics, might result in different levels of enrichment of HMs in different plant tissues. (Fig. 2). In corn tissues, corn cob and bran had the highest levels of Cd, Cr and Pb, corn stigma had the greatest values of Cu, Zn and Hg, and corn kernels had the lowest levels of HMs apart from Cr. (Table S3 and Fig. 2). In rice tissues, rice roots showed almost the highest levels of other five metal elements except Cr, which might be due to the direct contribution of the soil to the root. Rice leaves had the highest levels of Cr element, suggesting atmospheric deposition may be a source. The average contents of HMs in the different types of vegetables investigated in this study varied greatly, which might be explained by their different propensities for absorbing and accumulating metals (Xu et al., 2013). As a whole, we found the Zn accumulations in the six investigated vegetables were the most, followed by Pb, Cu, Cr, and Cd, with Hg the least,

Table 2
Geoaccumulation index (I_{geo}), pollution index (*PI*) and mean pollution index (*MPI*) of heavy metals in agricultural soils and atmospheric dustfall in the study area.

Location			Cu	Zn	Cd	Cr	Pb	Hg
Rhizospheric soil (n = 29)	I_{geo}	Minimum value	−1.5	−1.58	−1.43	−2.08	−2.84	−1.32
		Maximum value	2.82	2.74	6.24	4.32	2.44	1.18
		Mean	0.53	0.3	2.31	−0.53	−0.28	−0.48
		SD	1.15	0.85	1.62	1.33	1.16	0.66
	PI	Minimum value	0.53	0.5	0.56	0.36	ND	0.60
		Maximum value	10.59	10.04	113.33	29.89	8.17	3.40
		Mean	2.97	2.24	14.89	2.28	1.52	1.21
		SD	2.51	1.81	23.42	5.49	1.75	0.71
	MPI	4.19						
	Class	Highly						
Atmospheric dustfall (n = 18)	I_{geo}	Minimum value	2.79	2.7	3.78	−1.17	0.6	−0.84
		Maximum value	6.21	9.22	6.51	1.99	2.46	1.60
		Mean	4.49	3.98	5.2	0.97	1.71	0.51
		SD	1.1	1.53	0.64	0.68	0.44	0.58
	PI	Minimum value	10.36	9.73	20.59	0.67	2.27	0.84
		Maximum value	111.32	891.68	137.13	5.94	8.26	4.53
		Mean	44.25	72.78	60.75	3.23	5.12	2.30
		SD	31.18	200.01	28.08	1.25	1.51	0.90
	MPI	31.41						
	Class	Severely						

SD = Standard deviation, ND = Below detection limit.

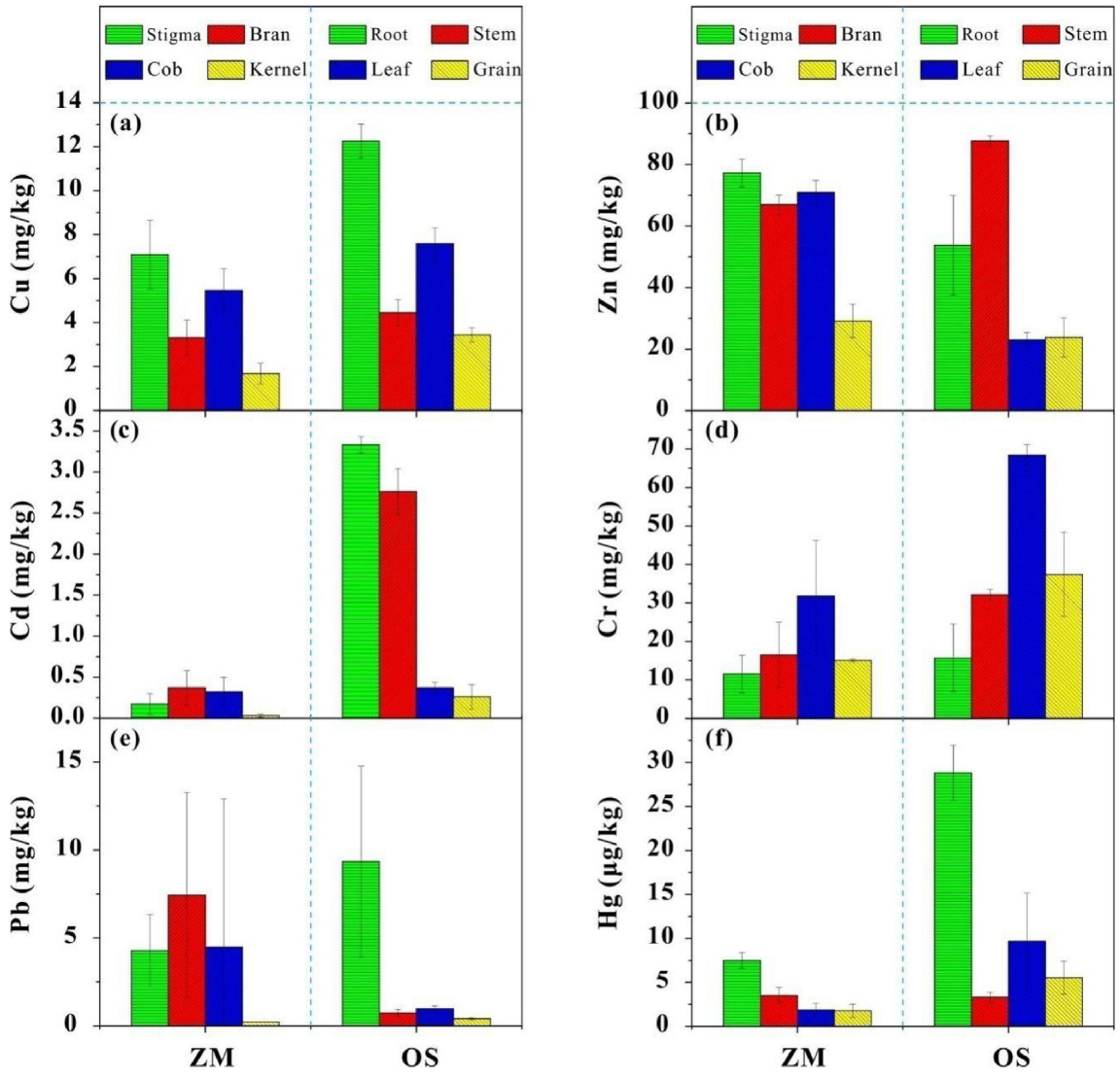

Fig. 2. Concentrations (based on wet weight) of HMs in the different tissues for the main crops including ZM (corn) and OS (rice) in the Tongling mining area.

basically consistent with previous results reported by Xu et al. (2013). Besides, we also discovered most HMs accumulated in the pulps and seeds were more than in the skins (Table S3 and Fig. 3). In Fengdan tissues, cortex moutan accumulated the most concentration of Cu and Zn, leaves gathered the most levels of Pb and Hg, and stems had the highest Cr concentration (Table S3 and Fig. S1). Compared with the threshold values issued by NHFPCPRC and CFDA (2017) (GB2762-2017), the average contents for Cr and Pb in the edible corn kernels, Cd, Cr, and Pb in the rice grains and various vegetable tissues (especially the edible portions), and Cr in Fengdan all exceeded the recommended food threshold values (Table S3), respectively, indicating latent health risks to the residents in the study area.

3.2. Transfer of HMs in soil-to-plant system

The *TF*s for six targeted HMs for different edible tissues of plants were calculated (Table S4 and Fig. S2) in this study, and results suggested the transfer capabilities of HMs from soil to plant were not the same. The mean *TF* trends for HMs were in the ascending order of Pb < Cd < Cu < Hg < Zn < Cr in corn kernels, Pb < Hg < Cu < Zn < Cd < Cr in rice grains, Cr < Hg < Cu < Pb < Zn < Cd in vegetables, Pb < Hg < Cr < Cu < Zn < Cd in Fengdan cortex moutan, and Cr < Pb < Cu < Cd < Zn in Fengdan seeds (Shen et al., 2017), respectively. Besides, we also found the *TF* value of a given metal element in different species of plants also had a great change (Table S4 and Fig. S2). The values of *TF* for six HMs in rice grains were all higher than that in corn kernels, indicating that enrichment abilities of most HMs in the rice grains were stronger than that in corn kernels (Fig. S2).

Although the *TF* order of Cu < Zn < Cd in vegetables was very similar to earlier reports (Zheng et al., 2007; Xu et al., 2013), the mean *TF* values of Pb in cucurbits and cucumbers were higher than in leafy vegetables planted in study area (Xu et al., 2013). Existing documents showed the direct absorption from atmosphere of vegetable leaves might be the main route for the Pb enrichment in the aboveground parts of plants (Uzu et al., 2010). Hereby, atmospheric Pb seemed to be the main cause of pollution of the investigated cucurbits and cucumbers. Also, we found the mean *TF*s of Cd varied significantly from 0.066 to 13.494 among different vegetable species, with the average of 4.244, which was the highest among different vegetable species, and tomatoes (*TF* = 13.494) and eggplants (*TF* = 12.513) had stronger transfer capacities for Cd than

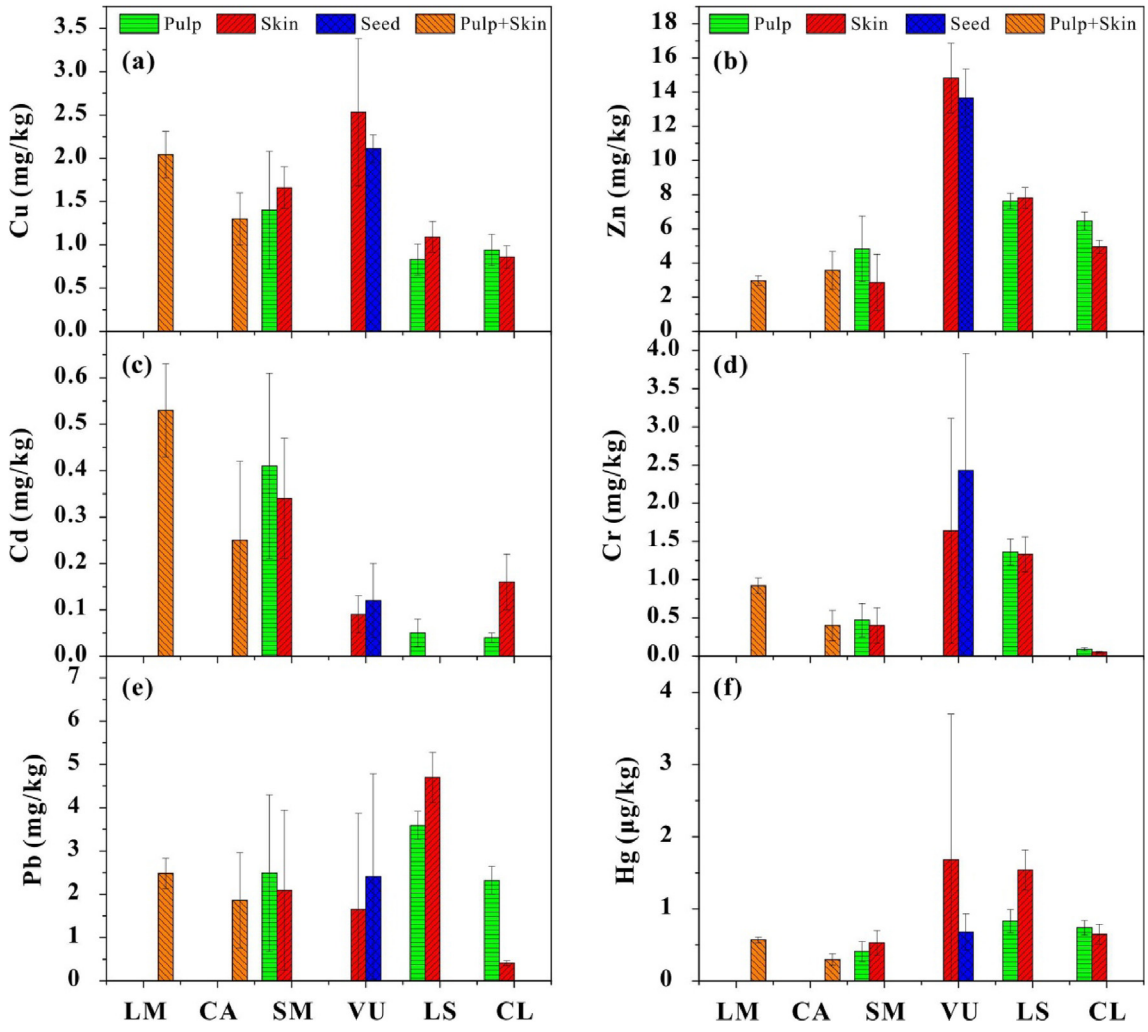

Fig. 3. Heavy metal concentrations (based on wet weight) in the different tissues for the typical vegetables in the Tongling mining area (Abbr. such as LM, CA, SM, VU, LS and CL were presented in Table S1).

other vegetables (Table S4 and Fig. S2). The higher accumulation and exchange for Cd element from soils to vegetables might be related to the Cd geochemical properties. The *TF*s for Cu element in tomatoes and the *TF*s for Zn element in cucumbers were the highest compared with other vegetables, which indicated both of them had stronger transfer capacities for Cu and Zn than other investigated vegetables, respectively (Table S4 and Fig. S2). This may be because Cu and Zn are indispensable nutrient elements for plants, and they are readily transported from the soils to the various tissues of the vegetables. The *TF*s for Cr and Hg were much lower and exhibited no obvious variation among different vegetables, indicating that their transfer abilities were very weak. Comparing the *TF* values of six targeted metals in all investigated vegetables, we found cucumber had a relatively much lower *TF* values for Cu, Cr and Cd element. Hence, we suggest that metal-excluding vegetable cultivars similar to cucumber can be grown in the Tongling area.

As shown in Fig. S2, Fengdan cortex moutans had higher *TF* values than Fengdan seeds (Fig. S2), indicating that the ability to enrich HMs in Fengdan root was stronger than in seed, which was in accord with Shen et al. (2017).

3.3. Source identification of HMs in plants

To further explore the sources of these target pollutants, three statistical analysis methods (CA, HCA and PCA) were used to determine the migration paths and hidden associated relationships among metal elements.

The results of the CA are presented in Table S5 and Table S6. For the targeted HMs in soils, the correlation coefficients for the element pairs Pb-Zn, Pb-Cu, Pb-Cd, Pb-Hg, Cu-Zn, Cu-Cd and Cu-Hg were 0.807, 0.745, 0.561, 0.786, 0.586, 0.672 and 0.634 ($p < 0.01$), respectively. Hg had a positive correlations with Zn ($r = 0.553$, $p < 0.01$) and Cd ($r = 0.375$, $p < 0.05$), respectively. For the targeted HMs in atmospheric dustfall, the element pairs Pb-Cd ($r = 0.659$, $p < 0.01$), Cu-Hg ($r = 0.677$, $p < 0.01$), Cu-Cd ($r = 0.550$, $p < 0.05$) and Hg-Cd ($r = 0.495$, $p < 0.05$) also showed very good correlations, respectively. From those results, we found Pb, Cd, Hg and Cu could have similar risk sources and geochemical migration. Moreover, the negative correlation among Cr and the other five targeted metal elements in the soils and weak correlation in atmospheric dustfall, further indicated that Cr likely originated from different sources compared with other five metals. The obvious positive correlations among Zn and other four targeted HMs (Pb, Cd, Hg and Cu) in the soils, as well as the consistent negative correlations among Zn and these four targeted HMs in dustfall, which jointly suggested that Zn in the soils and atmospheric dustfall had different sources.

HCA was performed for the soils, atmospheric dustfall, and plants in the study area to deeply discuss the possible sources of the

hazardous targeted HMs in this study. Three major clusters for the soils were identified in Fig. S3a: cluster 1 (Zn-Pb-Hg), cluster 2 (Cu-Cd) and cluster 3 (Cr). For atmospheric dustfall, four major clusters were distinguished in Fig. S3b: cluster 1 (Cu-Hg), cluster 2 (Cd-Pb), cluster 3 (Zn) and cluster 4 (Cr). For plants, three main clusters were distinguished in Fig. S3c: cluster 1 (Cd-Hg-Cu), cluster 2 (Pb) and cluster 3 (Zn-Cr). To sum up, results of the HCA showed most of HMs (Cu, Cd, Pb, and Hg) might have a common source. Cr in soils, along with Zn and Cr in atmospheric dustfall were clearly distinct from other metals in HCA, indicating that they could be derived from separate sources. These results of HCA in soils and atmospheric dustfall were in good agreement with the findings of the CA.

In order to further identify the sources of HMs for the plants, we conducted a detailed PCA of the six targeted HMs in the investigated plants. Two factors (PC1 and PC2) were identified in this study (Fig. S3d), which accounted for approximately 53.94% of the total variation (Table S7). The first factor (PC1) could explain 30.63% of total variance, and it possessed not only a positive loadings of Zn and Cr, but also a negative loadings of Pb. The second factor (PC2) could account for 23.31% of total variance, and it had a positive loadings of Cu, Cd and Hg. We found that PCA results for plants are very similar to above HCA results. The finding would jointly suggest that Zn-Cr, Pb, and Cd-Cu-Hg in the plants might derive from different geochemical end-members.

The previous studies had shown that Zn and Cr in plants might chiefly originate from such sources as local industrial emissions (e.g., non-ferrous metal smelting, Pan et al., 2017) and traffic pollution (e.g., automobile tire wear, Sprynskyy et al., 2011). Pb might mainly come from intensive vehicular emissions as well as mining activities by way of atmospheric deposition (Chen et al., 2005). In contrast, Cu-Cd-Hg appeared to be from other sources such as human activities and natural parent materials (e.g., Sawut et al., 2018; Bi et al., 2018).

3.4. Source apportionment based on lead isotope

In this work, 145 lead isotopic samples including agricultural soils, plants (crops, vegetables and medicinal Fengdan), atmospheric dustfall, coals, ores and minerals, background soils and soil-forming parent rocks in Tongling area, as well as various automobile fuels from the province adjacent to Anhui province, were collected and presented in Table S8 and Fig. 4.

As illustrated in Fig. 4, the Pb isotopes of atmospheric dustfall fell between diesel and ores and minerals, which fitted a binary mixing model. Based on the data of Table S8, the average $^{206}Pb/^{207}Pb$ ratios of diesel and ores and minerals were 1.159 and 1.179, respectively. After taking the $^{206}Pb/^{207}Pb$ isotope ratios of atmospheric dustfall into Eq. (4), the calculation result indicated that the contribution of diesel and ores and minerals to the concentration in the dustfall were 33.75% ranged from 5% to 70% and 66.25% ranged from 30% to 95%, respectively (Fig. 5a), clearly indicating local mining-smelting activities were the major source of Pb in atmospheric dustfall in Tongling area.

In Fig. 4, the $^{206}Pb/^{207}Pb$ (1.177–1.193) and $^{208}Pb/^{206}Pb$ (2.079–2.104) for the agricultural soils near the Tongling mining area basically distributed between anthropogenic emissions (e.g., coal combustion and mining-metallurgy) and natural reservoirs (the soil-forming parent rock and background soil). However, it was worth noting that the Pb isotopic ratios for agricultural soil varied greatly, indicating it had a complex Pb sources and the contribution ratio of different end-members might be different. Hence, we used a ternary mixing model to quantify the relative importance of Pb inputs from three end-members according to Fig. 4: natural source (the soil-forming parent rocks and background soils, $^{206}Pb/^{207}Pb = 1.201$ and $^{208}Pb/^{206}Pb = 2.066$), ores and minerals ($^{206}Pb/^{207}Pb = 1.179$ and $^{208}Pb/^{206}Pb = 2.092$), and coal combustion ($^{206}Pb/^{207}Pb = 1.178$ and $^{208}Pb/^{206}Pb = 2.101$). After taking the Pb isotope ratios of agricultural soils into Eq. (5), the result showed

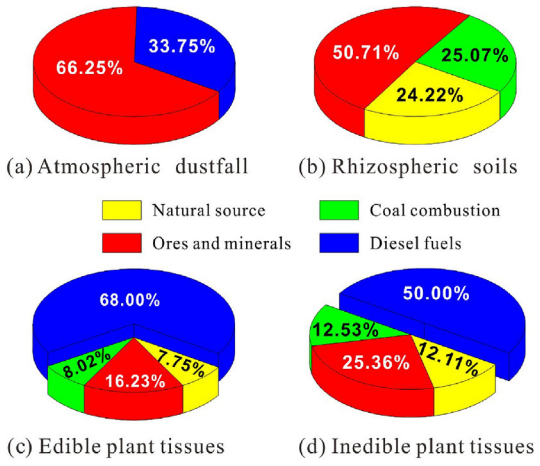

Fig. 5. Relative contribution of each end-member to Pb in atmospheric dustfall (a), rhizospheric soils (b), edible plant tissues (c) and inedible plant tissues (d). (Natural source is the same as Fig. 4).

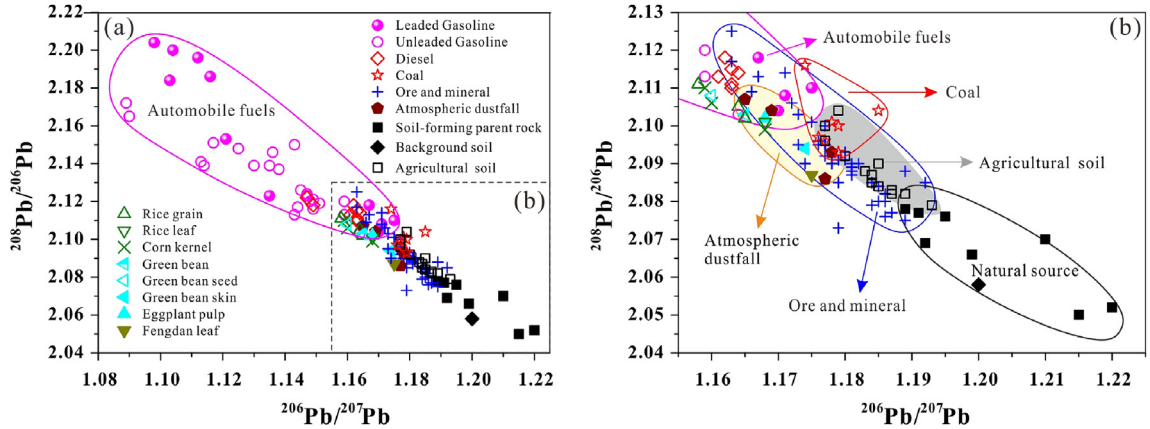

Fig. 4. Plot of $^{206}Pb/^{207}Pb$ vs. $^{208}Pb/^{206}Pb$ for the investigated samples in Tongling mining area and automobile fuels from the province adjacent to Anhui Province, China (Data from Table S8, natural source including soil-forming parent rocks and background soils).

that the contributions of the natural source, ores and minerals, and coal combustion to the Pb concentration in the rhizospheric soils were 24.22% (0–63.64%), 50.71% (0–100%) and 25.07% (0–100%), respectively (Fig. 5b), showing Pb in agricultural soils also mainly derived from the local mining-smelting operations, but the contribution of lead emissions from coal combustion could not be ignored.

It was difficult to distinguish the Pb origins in different plant tissues based solely on the total metal contents. In this study, we found Pb isotopic compositions of the plants investigated near Tongling mining area mostly plotted between diesel fuels ($^{206}Pb/^{207}Pb = 1.159$) and agricultural soil ($^{206}Pb/^{207}Pb = 1.184$) fields (Fig. 4). After taking the $^{206}Pb/^{207}Pb$ isotope ratios of plants into Eq. (4), the calculation result using a two-mixed model showed that the contribution rates of the diesel fuels and agricultural soils to the Pb concentration in the edible tissues were 68.00% and 32.00%, respectively. Combined with the aforementioned sources of lead in agricultural soils in the study area, it was known ores and minerals, coal combustion and natural processes would also be the three major sources in the edible tissues of plants, accounting for 16.23%, 8.02% and 7.75%, respectively (Fig. 5c). Lead isotopic tracing results indicated the main Pb sources in the in the edible tissues of plants were vehicle exhaust (diesel) and mining-smelting activities. Using a similar apportionment method, the results indicated that the Pb source in the inedible parts of the plants also originated mainly from diesel, ores and minerals, coal combustion and natural processes, with the average contribution rates of 50.00%, 25.36%, 12.53%, and 12.11%, respectively (Fig. 5d). In summary, the main Pb sources in the plants were the local anthropogenic emissions (diesel fuels and mining-metallurgy activities) rather than the background soils. Automobile emissions from diesel contributed significantly more to the accumulation of lead in the plant edible tissues (68.00%) than that of the inedible plant tissues (50.00%), which further suggested the atmospheric deposition was an important way for Pb to migrate into local plants. Our understanding was very consistent with the existing reports that atmospheric deposition was the main way for plants to accumulate lead via direct foliar uptake (Klaminder et al., 2005; Bi et al., 2009).

It was worth noting that Pb isotope compositions of Fengdan cortex moutans collected from Xiangsigu valley were close to the agricultural soil and ores and minerals, far from Pb isotope ranges of the automobile fuels and natural sources (Fig. 4). This meant that Pb sources in Fengdan were different from other plants investigated, mainly from local mining-metallurgy activities, rather than automobile exhaust.

3.5. Health risk assessment of HMs

Table S9 and Fig. S4 showed the exposure levels of metal elements in crop, vegetable and medicinal Fengdan to the inhabitants. The *THQ* for each metal element obtained increased in the order of $Hg < Cd < Cu < Zn < Pb < Cr$, $Hg < Zn < Cu < Cd < Pb < Cr$, $Hg < Zn < Cu < Cd < Cr < Pb$ and $Hg < Zn < Cu < Cd < Pb < Cr$ via consuming corn kernels, rice grains, vegetables and Fengdan, respectively (Fig. S4). Among the six targeted HMs, the *THQ* of Cr element in corn kernels and Fengdan, and three elements (Cr, Pb and Cd) in rice grains and vegetables were greater than 1, which suggested the uptake of Cr via consuming corn kernel and Fengdan, and Cr, Pb, Cd via consuming rice grains and vegetables would pose a noteworthy health danger.

To sum up, the potential health risk of Cr element was maximal to local residents in comparison to other metal elements investigated in the Tongling mining area, followed sequentially by Pb and Cd. In combination with daily diet habits, if we considered the overall intake of six HMs via consuming all investigated plants, although the *THQ* of some metal elements such as Cu, Zn, and Hg in the rice grains and Fengdan was less than 1, respectively, a underlying low health risk to the local population still could not to be underestimated.

4. Conclusion

The levels of HMs investigated in the soil-dustfall-plant system in Tongling mining area varied with the metal types and environmental media. The concentrations of six HMs in atmospheric dustfall were clearly higher than in soils. Cr and Cd in soils, and Cd, Cr, Pb, Cu and Zn in dustfall showed a relatively high contamination degree. The mean concentrations of Cr and Pb in corn kernels, Cd, Cr, Pb in rice grains and vegetables, and Cr in Fengdan cortex moutan all exceeded the suggested food safety limits in China, respectively. The ability of rice grains to enrich HMs was higher than corn kernels. Cadmium had the strongest enrichment ability in vegetables. The ability of Fengdan roots to enrich HMs was stronger than Fengdan seeds. The results of statistical analysis indicated that Zn and Cr in plants might chiefly originate from anthropogenic activities and traffic pollution, Pb mainly from anthropogenic sources, whereas Cu, Cd, and Hg derived from human activities and natural processes. Results of lead isotope tracing further revealed that mining-smelting activities were the major Pb sources in atmospheric dustfall and agricultural soils, whereas diesel vehicle emissions was the dominant Pb sources in plants and the atmospheric deposition was an important way for plants to absorb Pb. Risk assessments showed Cr had the greatest health risks to local inhabitants, followed sequentially by Pb and Cd.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.113089.

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