

Lead in soil and agricultural products in the Huainan Coal Mining Area, Anhui, China: levels, distribution, and health implications

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Abstract Heavy metal accumulation in agricultural soil is of great concern, as heavy metals can be finally transferred to the human body through the food chain. A field survey was conducted to investigate the lead (Pb) levels and distribution in soil, agricultural products (wheat, paddy, and soybean), and fish, in the Huainan Coal Mining Area (HCMA), Anhui Province, China, to provide reference information to local inhabitants. The daily intake and target hazard quotients of Pb through food consumption were assessed. Results showed that the mean Pb concentration in soil was higher than the Huainan soil background Pb value but lower than the maximum allowance Pb concentration for agricultural soil (GB 15618-2008). The elevated Pb in soil, especially in rainy months (June to August in Huainan), might be related to Pb leaching from ambient coal gangue piles. Excessive Pb concentration was found in the grains of food crops, which would pose a potential health risk to local inhabitants. Therein, wheat showed higher Pb bioaccumulation ability than other crops. With regard to the Pb levels in muscles, fishes were

considered to be safe for consumption. The calculations on daily intake and tolerable hazard quotient of Pb suggest that the potential health hazard posed by Pb is currently insignificant for the inhabitants in the HCMA.

Keywords Lead · Soil · Agricultural products · Distribution · Health risk assessment

Introduction

Lead (Pb) is ubiquitous in the environment, with a crustal abundance of approximately 12.5 mg/kg (Wedepohl 1995). Due to its persistence and toxicity in the environment, Pb is always regarded as a predominant toxic element in environmental laws and regulations. Lead is introduced into the human body through inhalation, ingestion, and other various pathways through Pb fume or dust (Fang et al. 2014a). Lead and its compounds, catalogued as developmental neurotoxin, could cause adverse effects on the nervous, hemopoietic, cardiovascular, and endocrine systems in the human body as a result of long-term exposure (Absalon and Slesak 2010; Khan et al. 2010). The main natural Pb sources include wind-borne soil particles, volcanic emissions, sea salt spray, and wild forest fires, which only consist of 3.5 % of total Pb emissions. However, anthropogenic Pb emission is one order of magnitude higher relative to natural emission (Nriagu 1989). Coal mining and combustion are the

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predominantly anthropogenic Pb emissions into the surrounding soil, water, and atmospheric compartments.

Increasing awareness has been focused on the potentially adverse effects of heavy metals enriched in soil (Fu et al. 2008; Khan et al. 2008, 2013; Li et al. 2006a, b, 2014). Heavy metal contamination of arable soil and agricultural products in the vicinity of a mining area has been an important international environmental concern. Heavy metal uptake via roots from contaminated soil and irrigation water, or direct atmospheric deposition of contaminants onto plant surfaces, can lead to the accumulation and contamination of heavy metals in plants (Oliver 1997). A large amount of Pb could be released during coal or coal gangue mining, washing, storage as well as transportation (Zhou et al. 2014a, b). Lead may disperse into the soil around mines through weathering and leaching of tailings. Metal sulfides, especially pyrite, can be readily oxidized during natural weathering of coal gangue piles produced during intensive mining activities, from which toxic elements including Pb are released and pose potential environmental risks to the surrounding soil and water environment (Dang et al. 2002).

Cultivation of food crops in contaminated compartments can lead to the transfer of metals into the edible parts of these plants, which may result in human health risks (Ji et al. 2013). Lacatusu et al. (1996) reported that soil and vegetables polluted by Pb and Cd in Copsa Mica and Baia Mare, Romania, significantly decreased the human life expectancy by 9–10 years within the affected area. Turkdogan et al. (2003) found that the high contents of metals (Co, Cd, Pb, Mn, Ni, and Cu) in fruit and vegetables in Van region of Eastern Turkey were related to the high prevalence of upper gastrointestinal cancer rates. Similar studies have also been carried out in China (Cui et al. 2004; Fang et al. 2014b; Khan et al. 2008; Li et al. 2006a, b; Liu et al. 2005; Wang et al. 2005, 2006). It was reported that the Pb content of some vegetables was 20 times higher than the permitted standards (Maximum Levels of Contaminants in Foods of China, GB 2762-2012) in the vicinity of a Pb/Zn mine in Shaoxing, eastern China (Li et al. 2006a). Likewise, soil and food crops are contaminated by Pb in varying degrees in the vicinity of Daobaoshan mine, Guangdong Province (Zhuang et al. 2009a, b). Local inhabitants near a smelter in Nanning, Guangxi Province, were substantially exposed to Pb via consumption of vegetables (Cui et al. 2004). Recently, Zhang et al. (2012) studied the Pb distribution

in soil or sediment collected from the floodplain of Huaihe River, Anhui Province, and concluded that the subsidence land was contaminated with Pb (45.57 mg/kg on average) due to the coal mining activities in this area. However, up until now, the investigation on the spatial and temporal distribution of Pb in soil and agricultural products and its health implications is limited. Thus, in the present study, we investigate the subsidence area in the Huainan Coal Mining Area (HCMA) to (1) characterize the content and distribution of Pb in soil and agricultural products and (2) assess the potential health risks of Pb in agricultural products.

Study area

The HCMA is situated in the southeast of North China (Fig. 1). It covers an area of approximately 3200 km² (mean 180 km in length and 15–25 km in width). Coal mining activities in Huainan started one century ago. Nine coal mines are currently active, and an additional ten coal mines are planned in the near future. Raw coal production in 2011 reached 67.5 million tons (Mt) (Chen et al. 2011), with coal gangue accounting for 10–15 % of coal production (Zhou et al. 2014b). Thirty-four coal gangue piles in Huainan occupy an area of 6 km², covering 50 % of the area allocated for solid wastes.

The climate of the HCMA is characterized by warm and semi-humid monsoons, with an annual average temperature of 15.2 °C, rainfall of 922.6–926.3 mm, and wind speed of 1.3–2.9 m/s in an E or SE direction, at spring and summer, and a NE or NW direction at fall and winter.

Three sampling sites were selected in the HCMA: Xinzhuangzi Mine, Panyi Mine, and Guqiao Mine (Fig. 1). Xinzhuangzi is an old mine (built in 1947) with an annual production capacity of 4 Mt. Panyi Mine occupies an area of 58.4 km², and its production capacity is 3 Mt/year. The total area of Guqiao Mine is approximately 106 km², with an estimated coal reserve of 18.2 Mt. The construction and mining activities at HCMA have already resulted in adverse environmental effects, such as large quantities of coal gangue piles and large areas of subsidence lakes. Coal gangue production was approximately 0.5 Mt for Xinzhuangzi, 1.18 Mt for Panyi, and 1.84 Mt for Guqiao in 2010.

The HCMA spreads along the Huai River bank, and the geomorphology is composed of piedmont slopes and

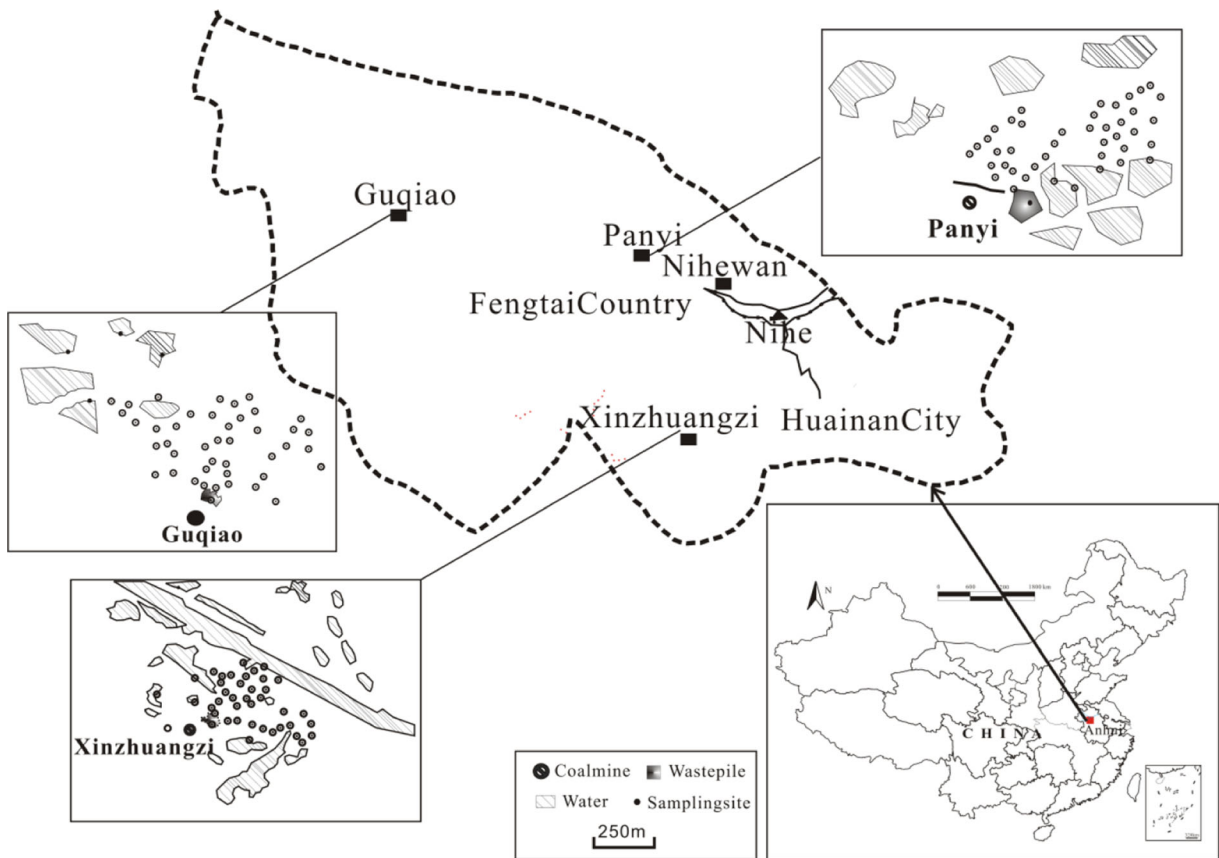


Fig. 1 Sampling locations of the Xinzhuangzi, Panyi, and Guqiao mines in the Huainan Coal Mining Area, Anhui, China

alluvial plains, suitable for crop plantation, such as paddy, wheat, soybean, etc. According to the Huainan Statistical Yearbook 2013, wheat production in Huainan rose to 594, 000 t in 2012, accounting for approximately 44 % of the total crop production. The rice production was 712,000 t, making up 53 % of the total crop production. Furthermore, approximately 19,000 t of soybean is harvested each year in the HCMA. Aquiculture in subsidence ponds accounts for 31 % of the total aquiculture. In addition, fishery harvesting was estimated to be 0.15~0.3 kg fish/m² in collapsed lakes.

Sampling and methods

Sampling

To investigate the spatial and temporal distribution of Pb, and its environmental hazard in the cultivated crops and aquatic products around the HCMA subsidence area, representative samples comprising 453 soil, 187

wheat, 40 rice, 64 soybean, and 36 fish were collected during April, June, August, and October in 2012, at the subsidence areas of Xinzhuangzi, Panyi, and Guqiao (Table 1). Surface soil samples (upper 15 cm of the soil profile) were collected using the plum-shaped distribution method. Each soil sample consisted of five sub-samples. Wheat, rice, and soybean samples were collected around the corresponding soil sampling sites, using the same plum-shaped distribution method. Individual crop samples were composed of no less than ten strains with comparable maturity. Fish samples were collected in randomly chosen collapsed lakes within the individual subsidence area. All samples were stored immediately in the sealed polyethylene bags and transported to the laboratory immediately.

Analytical methods

The soil samples were air-dried, then crushed and ground with an agate mortar to pass through a 120-mesh sieve. Approximately 0.2 g sample was placed

Table 1 Statistical description of the sampling campaign in Huainan Coal Mining Area

April	Soil	Wheat	Fish	
Xinzhuangzi	30	30	3	
Panyi	38	38	3	
Guqiao	42	42	3	
June	Soil	Wheat	Fish	
Xinzhuangzi	42	27	3	
Panyi	35	20	3	
Guqiao	45	30	3	
August	Soil	Soybean	Rice	Fish
Xinzhuangzi	39	20	3	3
Panyi	38	7	11	3
Guqiao	35	8	7	3
October	Soil	Soybean	Rice	Fish
Xinzhuangzi	40	13	5	3
Panyi	31	8	8	3
Guqiao	38	8	6	3

into a conical flask, then 8 ml concentrated nitric acid (HNO₃, analytical grade) and 2 ml perchloric acid (HClO₄, analytical grade) were added. After cold digestion within a fume hood overnight, the conical flasks were placed on an electric hot plate for thermal decomposition (100 °C for 1 h, 120 °C for 2 h, and 180 °C for 1 h) and evaporated at 210 °C. The resulting digests were cooled at room temperature and diluted to 25 ml with 5 % HNO₃ solution (GB/T 17141-1997).

Plant samples (wheat, rice, and soybean) were separated (root, stem, leaf, flesh, and husk) and oven-dried (70 °C) to constant weight and milled to small pieces. The digestion method for plant samples was modified from Gu et al. (2007) (State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, Nanjing). Approximately 2 g sample was placed in a 100-ml triangular flask in which several glass beads were put to prevent violent liquid boiling. Flasks were covered with neck bent funnels, and 10 ml mixed concentrated acid (HNO₃:H₂SO₄:KClO₄=8:1:1) was added then kept overnight in the fume hood. The cold digested samples were then fitted on an electric hot plate to undergo thermal digestion. The final solution was filtered through a 0.45-μm filter membrane and added to 50 ml with 5 % HNO₃ for the subsequent elemental determination.

Fish samples were separated into parts including scales, muscle, vertebral column, gill, viscera, fins, and swim bladder. The different parts were washed with tap

water followed by Milli-Q deionized water and oven-dried to constant weight at 80 °C. The digestion method for fish tissues was modified from a previous method (Yilmaz et al. 2007). About 0.5 g sample was weighed in a conical flask with several glass beads put in to prevent violent liquid boiling. An acid mixture including 10 ml concentrated HNO₃ and 1 ml HClO₄ was added before placing it on the electric hot plate (110 °C for 1 h). After digestion, the solution was transferred to volumetric flasks and kept in 4 °C freezer prior to the analysis.

The Pb content was determined by inductively coupled plasma mass spectrometry (ICP-MS) (X Series 2, Thermo Fisher Scientific). Each sample was determined in triplicate. Reagent blank and standard reference materials GBW07403 (soil), GBW07603 (bush leaves), and GBW10050 (prawn) were processed in parallel and subjected to quality control. The recovery of Pb was within 93–98 %.

Health risk assessment

The bioaccumulation factor (BAF), an index of the ability of the plant to accumulate heavy metals with respect to its content in the soil substrate (Ghosh and Singh 2005), is calculated as follows:

$$\text{BAF} = \frac{C_{\text{plant}}}{C_{\text{soil}}} \quad (1)$$

where C_{plant} and C_{soil} represent the heavy metal (Pb in the present study) contents of edible parts of plants and soil, respectively.

The daily intake (DI) of metals depends on both the element concentration ($[M]$) and the amount of the respective food consumed (W). The daily intake of Pb from soil through food crops is calculated as follows:

$$\text{DI} = \frac{[M] \times W}{\text{BW}} \quad (2)$$

where $[M]$ is the content of heavy metals in food crops (mg/kg); W represents the daily average consumption of food crops in this region (g/person); and BW is the body weight (kg). For adult inhabitants, the daily ingestion rates of wheat, rice, soybean, and fish are set as 140.2, 238.3, 4.2, and 29.6 g/person, respectively (Wang et al. 2009).

The THQ, which is the ratio of the exposure pollutant to the reference doses (a reference dose or RfD), is valid and useful to express the risk of non-carcinogenic effects. If the ratio is less than 1, there will not be any

obvious risk. Conversely, an exposed population of concern will experience health risks if the dose is equal to or greater than the RfD. The THQ value can be calculated as follows:

$$THQ = \frac{EFr \times ED \times FI \times MC}{RfDo \times BW \times AT} \times 10^{-3} \quad (3)$$

where THQ is the target hazard quotient; EFr is the exposure frequency (365 days/year); ED is the exposure duration (70 years); FI is food ingestion (g/person/day); MC is the metal concentration in food (mg/kg); RfDo is the oral reference dose (mg/kg/day); BW is the average body weight, which is 55.9 kg for an adult; and AT is the average time for non-carcinogens (365 days/year × number of exposure years, assuming 70 years in this study).

Results and discussion

Pb content of soil and agricultural products

The descriptive statistics on the Pb content of soil and agricultural products in the HCMA are tabulated in Table 2. The elemental concentration of Pb in soil from the HCMA is much lower than those in soil around non-ferrous metals mining and/or smelting plants in previous studies (Cui et al. 2004; Liu et al. 2005; Wang et al. 2006; Zheng et al. 2007). The mean Pb contents of soil are 26.1±11.6, 25.4±10.1, and 20.1±8.9 mg/kg for Xinzhuangzi, Panyi, and Guqiao, respectively. They are comparable to the Huainan soil Pb background value (23.52 mg/kg) (Yang et al. 1995), but lower than the threshold of Environmental Quality Standards for Soils (GB 15618-2008). According to our previous study (Fang et al. 2014b), Pb may be released from coal gangue piles through weathering as well as leaching under complex natural conditions in the HCMA.

Previous studies have demonstrated that the higher the content of heavy metals in soil, the higher the

probability of metal transfer into crops, and vice versa (Mapanda et al. 2007). The Pb contents of the wheat samples obtained in April are 23.0±1.5, 22.8±2.4, and 22.1±1.4 mg/kg for Xinzhuangzi, Panyi, and Guqiao, respectively, and decrease in June to 17.6±2.6, 15.7±2.4, and 16.0±1.2 mg/kg, respectively. The average Pb content of wheat grains is approximately 3.5 times as high as the national maximum allowance concentration (0.2 mg/kg) of Pb in foods (GB 2762-2012), which deserves further attention.

The Pb content of paddy samples decreases in the following sequence in August: Xinzhuangzi (10.0±1.4 mg/kg) > Panyi (9.2±1.8 mg/kg) > Guqiao (7.3±0.3 mg/kg). However, in October, paddy samples at Guqiao have the highest Pb content. A similar tendency is also found for Pb content in the studied soybean samples collected in October, in which the Pb contents are 6.1±0.6, 6.4±0.9, and 7.2±1.9 mg/kg for Xinzhuangzi, Panyi, and Guqiao, respectively. It is noted that Pb contents of the edible parts of paddy (0.48, 0.21, and 0.29 mg/kg in Xinzhuangzi, Panyi, and Guqiao paddy grains, respectively) have exceeded the national maximum allowance concentration (0.2 mg/kg, GB 2762-2012). In addition, the average Pb content of soybean grains at Xinzhuangzi (0.36 mg/kg) also surpasses the permissible Pb level set as 0.2 mg/kg.

The fish investigated in the present study is catalogued as a crucial species, which is one of the demersal fishes that are much closer to sediments in the ponds. Pb contents of fish samples of the mining areas are 10.8±3.6, 12.9±2.5, and 10.9±1.3 mg/kg for Xinzhuangzi, Panyi, and Guqiao, respectively.

Spatial and temporal variation of Pb in the HCMA

The temporal and spatial variation of soil Pb content among mines in the HCMA is presented in Fig. 2. Generally, Pb contents in soils collected in April and August are obviously lower than those in June and

Table 2 Temporal and spatial distribution of Pb in soil and agricultural products in HCMA (mg/kg, dry weight)

Locations	Soil	Wheat		Paddy		Soybean		Fish
		April	June	August	October	August	October	
Xinzhuangzi	26.1±11.6	23.0±1.5	17.6±2.6	10.0±1.4	9.6±0.9	6.0±0.2	6.1±0.6	10.8±3.6
Panyi	25.4±10.1	22.8±2.4	15.7±2.4	9.2±1.8	9.6±0.1	5.2±0.3	6.4±0.9	12.9±2.5
Guqiao	20.1±8.9	22.1±1.4	16.0±1.2	7.3±0.3	11.2±4.1	4.7±0.4	7.2±1.9	10.9±1.3

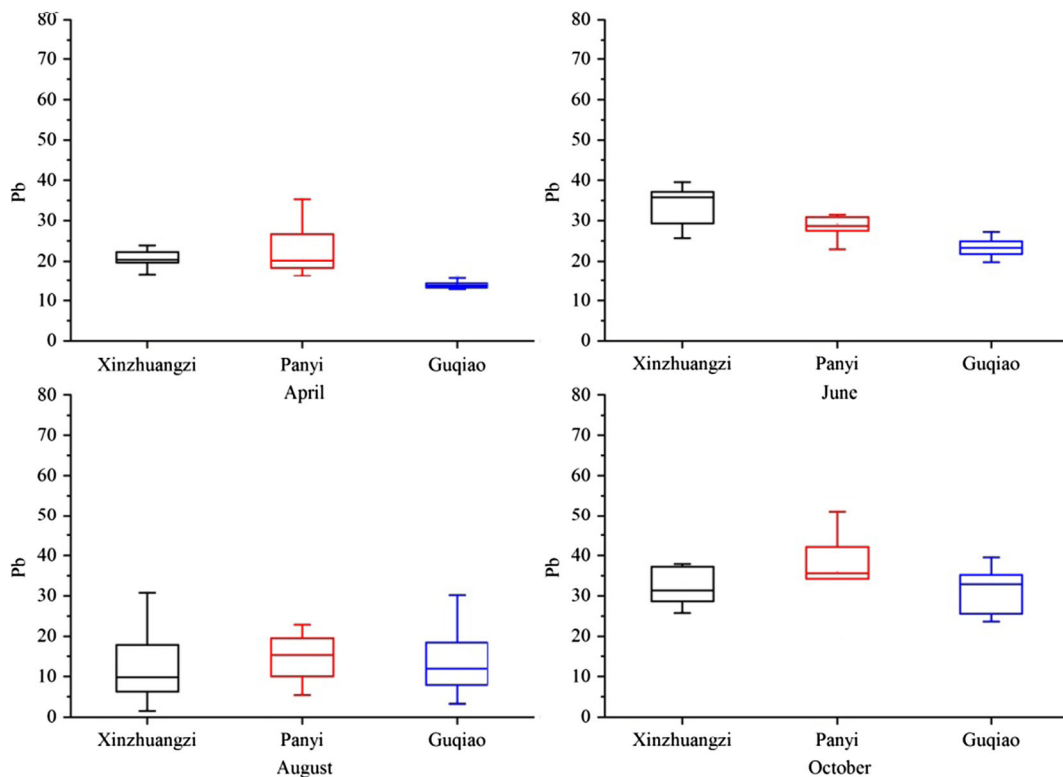


Fig. 2 Temporal and spatial variation of Pb content in soil (mg/kg) in the HCMA

October. Food crops are in the growing period in April and August, which need large amounts of water and nutrients. During this process, Pb in soil solution may be transferred into plants and thus results in the lower Pb content in soil. Furthermore, according to the Huainan Statistical Yearbook (2013), Huainan City enters into rainy season since June, and precipitation events largely concentrated in June, July, and August. The elevated Pb content in soil may relate to more Pb leaching from coal gangue piles (Fang et al. 2014b).

The internal distribution of Pb in individual parts of agricultural products is depicted in Figs. 3, 4, and 5. The Pb distribution in wheat has been discussed in our previous study (Fang et al. 2014b), where we found Pb is preferentially accumulated in the roots. Most Pb is absorbed through roots from the soil, and it is expected that the Pb content of wheat linearly correlates with the Pb content of soil (Mapanda et al. 2007). A major proportion of Pb is distributed in roots of paddy and soybean, followed by an appropriate part of Pb being enriched in their stems. In addition, some Pb is transferred and kept in the husks in soybeans. The higher Pb content in husks indicates that husks are barriers for toxic Pb entering grains that could relieve the potential

health implications for humans. However, husks are usually milled as food for livestock; thus, the Pb accumulation in husks deserves further attention when taking the food chain into consideration. As for the fish sample, Pb is predominantly concentrated in scales, vertebrae as well as viscera, which are almost discarded when consuming fish (Olmedo et al. 2013; Rashed 2001; Wang et al. 2005; Yi et al. 2011).

Bioaccumulation factors

Plants could transfer toxic elements from abiotic into biotic environments and thus represent biological barriers for the migration of toxic elements into the environment (Qi et al. 2011). The BAFs for Pb for agricultural products vary significantly between species and sampling locations (Table 3 and Fig. 6). The Pb bioaccumulation factor ranges from 0.0202 to 0.0302 for wheat, from 0.0059 to 0.0094 for paddy, and from 0.0039 to 0.0105 for soybean. The relatively lower BAF values of Pb in these crops indicate that the transfer of Pb from soil to the edible parts of agricultural products is limited. Among the three food crops, wheat shows relatively higher Pb accumulation ability whereas

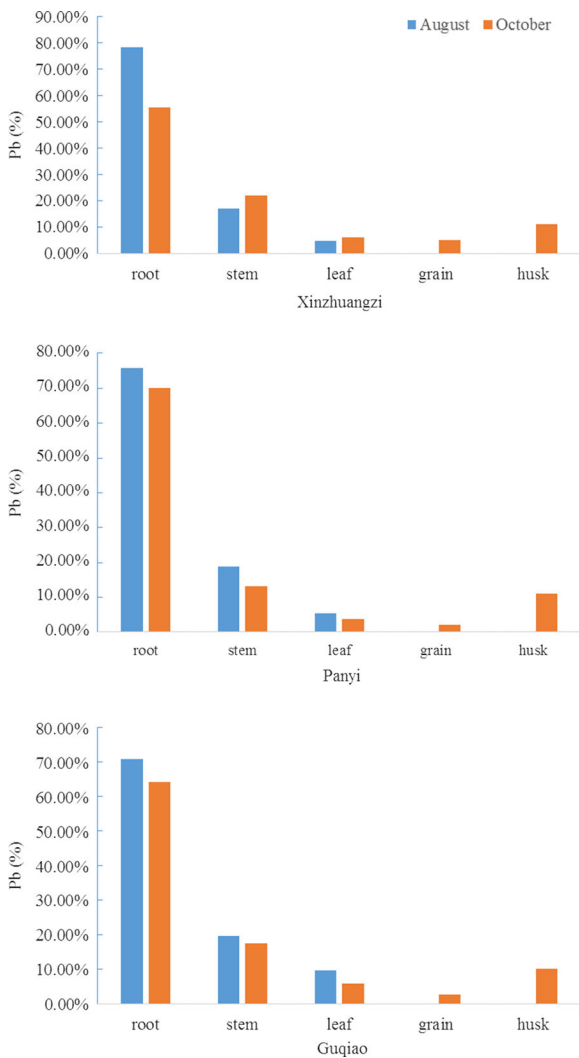


Fig. 3 Distribution of Pb in individual parts of paddy (%) in the HCMA

soybean has the lowest Pb accumulation potential. The difference in the BAFs among various locations may relate to soil characteristics (Cui et al. 2004). As shown in Fig. 6b, the BAFs decrease with increasing total Pb contents in soils, consistent with the conclusion drawn by Wang et al. (2006). It is suggested that the higher content of Pb in soil would restrain the translocation of Pb from soil to vegetables (Wang et al. 2006).

Health risk assessments

Principally, there are two major pathways for human exposure to soil contamination: soil-plant-human (food chain pathway) and soil-human (incidental soil ingestion)

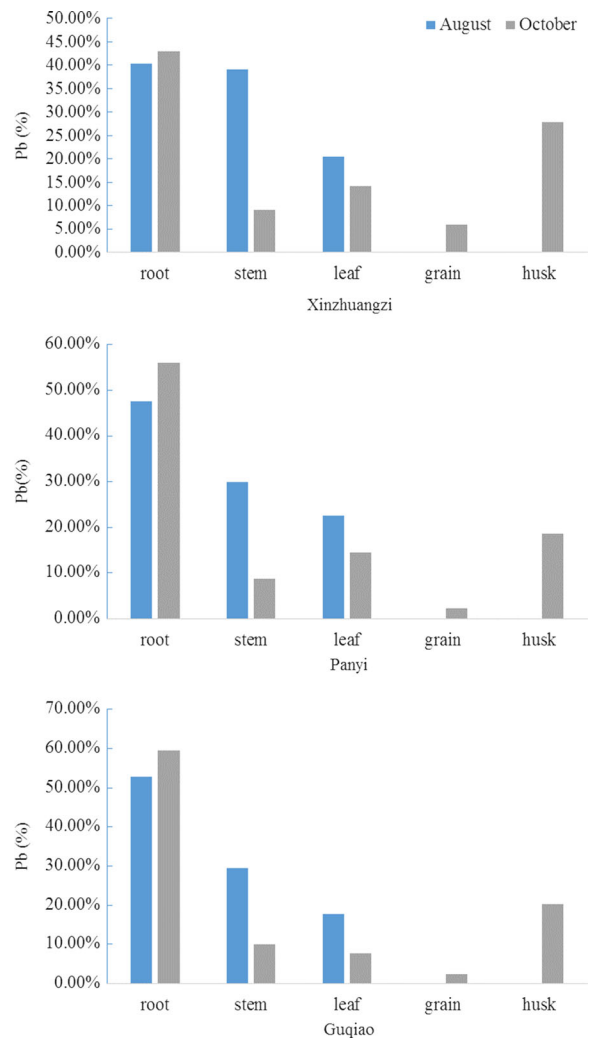


Fig. 4 Distribution of Pb in individual parts of soybean (%) in the HCMA

pathways (Cui et al. 2004). Soil-to-plant transfer of heavy metals is the major pathway of human exposure.

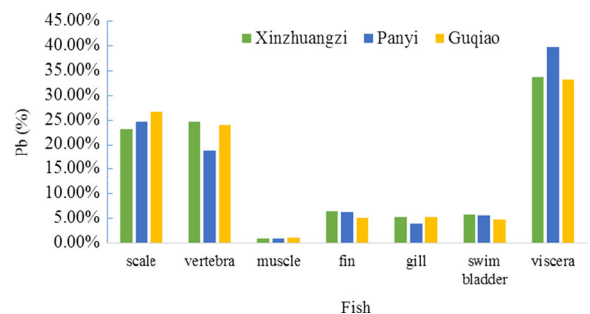


Fig. 5 Distribution of Pb in individual parts of fish (%) in the HCMA

Table 3 The mean bioaccumulation factors (BAFs) for agricultural products in HCMA

Locations	Wheat	Paddy	Soybean
Xinzhuangzi	0.0202	0.0140	0.0105
Panyi	0.0243	0.0059	0.0039
Guqiao	0.0302	0.0094	0.0058

The calculations of daily intake (DI) and target hazard quotients (THQ) are summarized in Table 4. The DI values for wheat, paddy, and soybean are highest in Xinzhuangzi Mine, followed by Guqiao Mine and then Panyi Mine. As for the daily Pb intake through fish, fishes in Guqiao Mine pose higher risks than those in other mines although their disparity is not so significant. The total Pb daily intake taking account of wheat, paddy, soybean, and fish consumption for adult inhabitants in Xinzhuangzi, Panyi, and Guqiao is 3.351, 1.484, and

Table 4 The estimated DI and THQ values for consuming agricultural products in HCMA

Locations	Wheat	Paddy	Soybean	Fish	Total
Daily intake					
Xinzhuangzi	1.212	2.060	0.027	0.052	3.351
Panyi	0.524	0.891	0.011	0.058	1.484
Guqiao	0.729	1.239	0.014	0.061	2.043
Target hazard quotients					
Xinzhuangzi	0.303	0.515	0.007	0.013	0.838
Panyi	0.131	0.223	0.003	0.014	0.371
Guqiao	0.182	0.310	0.003	0.015	0.510

2.043 $\mu\text{g}/\text{kg}/\text{day}$, respectively. The inhabitants in the HCMA are considered to be safe according to the tolerable daily intake of Pb (3.6 $\mu\text{g}/\text{kg}/\text{day}$) regulated by FAO/WHO (Ostapczuk et al. 1987). However, our conclusion might be optimistic due to the absence of vegetables in this study. Vegetables are one of the main risk contributors in addition to wheat and paddy in inhabitants' daily food consumption. Besides, although the inedible parts of crop products could not directly affect the health of inhabitants, they do have potential health risks as the local villagers would use the inedible parts of the crops as fuel instead of coal. This practice will contribute to the elevated Pb levels in the atmosphere (Zhou et al. 2014c). Pb is persistent and accumulated in the human body and is hardly to be excreted; thus, increasing concern should be paid for the Pb in food and its daily intake.

The THQ values for individual food crops are all far below unity in the HCMA, indicating that health risks associated with Pb exposure are insignificant at present. The total THQ value for Xinzhuangzi (0.838) is higher than those for Panyi (0.371) and Guqiao (0.51); these values are all below unity. This suggests that food consumption for inhabitants in the HCMA will not increase the health risks associated with Pb. Among these various foods, wheat and paddy, the staple food throughout China, are main contributors to the total THQ. Thus, heavy metal contamination in staple food needs more comprehensive and systematic investigation. Furthermore, the health risk estimated on the basis of Pb in this study may be insufficient to predict the risk associated with mixed contaminants in the HCMA.

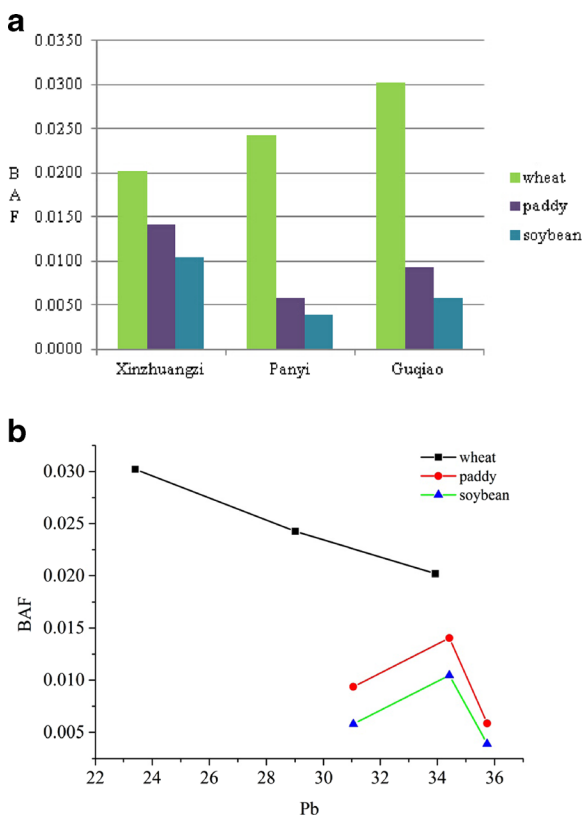


Fig. 6 The comparison of bioaccumulation factors for agricultural products in the HCMA (a) and the relationship between the BAFs and total Pb content in soil (b)

Conclusions

The spatial and temporal distribution of Pb in soil and agricultural products in the HCMA was investigated. Samples were collected from Xinzhuangzi, Panyi, and Guqiao mines, which are old-aged, medium-aged, and new-aged mines, respectively. The Pb contents in soils collected in these mines were all below the maximum allowance for Pb concentration in agricultural soil of China (GB 15618-2008), although Pb contents in the Xinzhuangzi and Panyi soils exceeded the background Pb value of the Huainan soil. Pb is preferentially concentrated in the roots of wheat. The major proportion of Pb was stocked in roots for paddy and soybean, followed by stems and husks. In spite of the barrier of husks in preventing Pb from entering grains, the Pb content in grains of wheat, paddy as well as soybean exceeded the maximum permissible Pb levels of contaminants in food (GB 2762-2012), which deserves further attention. In comparison with other crops, edible parts in wheat showed a relatively higher Pb bioaccumulation ability. Fish raised in the HCMA is confirmed to be safe for consumption considering its Pb contents in muscles. The calculation of the daily intake and target hazard quotients of Pb through daily food consumption showed that the inhabitants in the HCMA are currently safe. A large fraction of Pb was released from coal gangue piles to soils in the HCMA in the rainy season; thus, the monitoring and assessment of Pb in soil and agricultural products are of great importance.

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