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Human health risk assessment of heavy metals in soil–vegetable system: A multi-medium analysis

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HIGHLIGHTS

- Flourishing private economy caused increasing heavy metal damages.
- Leafy and rootstock vegetables posed higher hazards.
- Cr has the biggest non-carcinogenesis effect while Cd generates the greatest cancer risk.
- Negative impacts on humans and the environment may cause additional costs not included in sales expenditures.

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ABSTRACT

Vegetable fields near villages in China are suffering increasing heavy metal damages from various pollution sources including agriculture, traffic, mining and Chinese typical local private family-sized industry. 268 vegetable samples which included rape, celery, cabbages, carrots, asparagus lettuces, cowpeas, tomatoes and cayenne pepper and their corresponding soils in three economically developed areas of Zhejiang Province, China were collected, and the concentrations of five heavy metals (Pb, Cd, Cr, Hg and As) in all the samples were determined. The health risk assessment methods developed by the United States Environmental Protection Agency (US EPA) were employed to explore the potential health hazards of heavy metals in soils growing vegetables. Results showed that heavy metal contaminations in investigated vegetables and corresponding soils were significant. Pollution levels varied with metals and vegetable types. The highest mean soil concentrations of heavy metals were 70.36 mg kg^{-1} Pb, 47.49 mg kg^{-1} Cr, 13.51 mg kg^{-1} As, 0.73 mg kg^{-1} for Cd and 0.67 mg kg^{-1} Hg, respectively, while the metal concentrations in vegetables and corresponding soils were poorly correlated. The health risk assessment results indicated that diet dominated the exposure pathways, so heavy metals in soil samples might cause potential harm through food-chain transfer. The total non-cancer and cancer risk results indicated that the investigated arable fields near industrial and waste mining sites were unsuitable for growing leaf and root vegetables in view of the risk of elevated intakes of heavy metals adversely affecting food safety for local residents. Chromium and Pb were the primary heavy metals posing non-cancer risks while Cd caused the greatest cancer risk. It was concluded that more effective controls should be focused on Cd and Cr to reduce pollution in this study area.

1. Introduction

There has long been concern about the issue of pollution by heavy metal because of their toxicity for plant, animal and human beings and their lack of biodegradability (Li et al., 2006; Jang et al., 2006; Zhuang et al., 2009). Soil is the primary reservoir of heavy metals in the atmosphere, hydrosphere and biota, and thus plays a fundamental role in the overall metal cycle in nature (Cao et al., 2010). Heavy metals in soil pose potential threats to the environment and can damage human health through various absorption pathways such as direct ingestion, dermal contact, diet through the soil–food chain, inhalation, and oral intake (Lu et al., 2011; Komárek et al., 2008; Park et al., 2004; Al-Saleh et al., 2004).

Vegetables play important roles in our daily diet as economic crops. However, various human activities such as mining, industrial processing like smelting, pesticides, automobile exhausts and fertilization, especially the huge annual applications of organic livestock manure, which is the traditional agricultural fertilizer, are causing elevated heavy metal concentrations in the environment in China (Zhuang et al., 2009; Cao et al., 2010; Zheng et al., 2007, 2010; Shi et al., 2011). Vegetables take up heavy metals by absorbing them from contaminated soils, as well as from deposits on parts of the vegetables exposed to the air from polluted environments (Wang et al., 2005). Chronic intakes of heavy metals have damaging effects on human beings and other animals (Zheng et al., 2007;

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Lai et al., 2010; John and Andrew, 2011). For example, Cr, Cu and Zn can cause non-carcinogenic hazardous such as neurologic involvement, headache and liver disease, when they exceed their safe threshold values (US EPA, 2000). There is also evidence that chronic exposure to low doses of cancer-causing heavy metals may cause many types of cancer. For example, Park et al. (2004) found increased lifetime risk of lung cancer death resulted from occupational exposure to dusts and mists containing hexavalent chromium. Dietary cadmium intake due to the consumption of environmentally contaminated rice and other foods was associated with an increased risk of postmenopausal breast cancer (Hiroaki et al., 2013). Acute and chronic arsenic exposure could also cause numerous human health problems. These included dermal, respiratory, cardiovascular, gastrointestinal, hematological, hepatic, renal, neurological, developmental, reproductive, immunological, genotoxic, mutagenetic, and carcinogenic effects (such as liver cancer) (Kapaj et al., 2006; Lin et al., 2013). Despite the economic benefits of industry, improved income and high crop yields due to fertilization, negative impacts on humans and the environment may cause additional costs not included in sales expenditures (Peter et al., 2012). Especially for the ubiquitous and non-biodegradable heavy metals, the negative effects persist for several decades and even longer.

The Household Responsibility System (HRS) was initiated during the late 1970s in China. It has brought a profound change to the rural economy. Farmland was allocated to each farmer household on the basis of family size. The farmers were then given the authority to manage their contracted land, including all decisions regarding production (Liu et al., 2009). In particular, vegetable fields were located very near to the villages and conveniently close to the farmers. Unfortunately, this means that the growing vegetables and soils are at high risk of contamination by local industrial pollution, since many small family-sized factories such as metal smelting and battery making businesses are located in villages due to the booming private economy.

Zhejiang province, one of the most important economic development provinces in China, has been leading the national private economy and the flourishing private enterprises bring about severe and numerous negative environmental effects. As is typical of regions in Southeast China, Zhejiang Province is densely covered with drainage ditches that form a network waterway and consequently the arable fields nearby are readily polluted by domestic and industry wastes. Many investigations have been conducted in which heavy metal pollution was evaluated using traditional methods (Granero and Domingo, 2002; Jarup, 2003; Chary et al., 2008).

In our research, particular emphasis is placed on the use of descriptive statistics in determining the effects of heavy metals pollution. This is the first study that has assessed the potential health risks of heavy metal exposure to multiple medium in such critical vegetated areas. From this data we can use various options to reduce human health hazards.

Primary objectives were: (a) to explore the current extent of local heavy metal pollution in vegetated soils and plants, (b) to determine the potential health risks of heavy metals as cumulative carcinogenic and non-carcinogenic risks via the multiple routes of ingestion, inhalation, dermal exposure and diet from the soil–vegetable system, and (c) to provide a reference for policy decision making on the prevention and treatment of heavy metal pollution.

2. Materials and methods

2.1. Study area

Zhejiang province is located in the Yangtze River delta region of Southeast China, covering a total area of 104,141 km^2 and having a total population of 5442.69 million inhabitants. With a high population density and developed industries and agriculture, Zhejiang province has 3000–4000 years of a history of food production. In this study, we selected three counties of Hangzhou, Changxing, and Shangyu in Zhejiang province as the study area. The first area is a typical suburban belt located in the northeast of Hangzhou county, the famous provincial capital, which was planted with 24.1 km^2 vegetables (30°16′16″–30°20′6″ N, 120°11′25″–120°14′58″ E). In the suburban areas in China, the heavy metals in soil are commonly affected by multiple factors including traffic, agriculture, and industry. As one of the "Top 100 counties" in China, Changxing is famous for its battery industry which could cause potential heavy metal pollution in the local environment. We sampled from the vegetated zone covering a latitude of 30°58′27″–31°02′3″ N and a longitude of 119°50′56″–119°57′16″ E. In Shangyu, another developed "Top 100 counties" in China, we sampled in a vegetated area covering from 29°59′42″–30°04′25″ N to 120°45′25″–120°49′38″ E. This sampling area is near a lead and zinc mine tailing which has been abandoned for almost a century. Due to long-term exposure to weathering, the pollutants are distributed around the mine within a heavy metal polluted area of about 800 hectares.

2.2. Sample collection

There were 268 vegetable samples (1 kg edible part of each) including 127 leafy vegetables (57 rapes, 43 celeries, 27 cabbages), 68 rootstock vegetables (26 carrots and 42 asparagus lettuces (*Lactuca sativa*)), 25 legume vegetables (25 cowpeas), and 48 solanaceous vegetables (26 tomatoes and 22 cayenne peppers) collected in 2011 from the study area. Simultaneously, 268 soil samples were collected at the vegetable sampling sites. When sampling, the study area was split into many sampling units, selected by planting mode and pollution background. Within the same sampling unit, five soil samples were collected using an "S" sampling procedure and then bulked to provide an individual composite sample. The vegetable samples were collected along the same gradients. Soil samples were taken in the immediate vicinity of the roots of the crop samples from 0 to 15 cm depth. Only the edible part of each vegetable was collected for analyses. All the soil or vegetable samples were quartered separately to provide sub-samples.

2.3. Pre-treatment and analysis of soil and vegetable samples

Soil samples were air-dried in the laboratory and sieved <2 mm. A part of the soils were ground in a porcelain mortar <100 mesh. They were stored in polyethylene bags at 4 °C prior to analysis. The edible portions of the vegetables were rinsed in distilled water and subsequently rinsed again with high-purity deionized water. After being milled by a ceramic-coated grinder, the vegetable samples were frozen at 18 °C until chemical analysis.

Soil pH (H_2O) and electrical conductivity (EC) were determined in distilled water (1:2.5 w/v). Soil mechanical composition (sand, silt, clay) was determined by a hydrometer method (Agricultural Chemistry Committee of China, 1983). Organic carbon (OC) contents were measured using the Walkey–Black wet oxidation method (Agricultural Chemistry Committee of China, 1983). Total Pb, Cd, Cr in the soils and vegetables were digested by HF–HNO_3–$HClO_4$ and analyzed by an inductively coupled plasma-mass spectrometer (ICP-MS, Agilent 7500a, USA). Total Hg and As were digested by HNO_3–HCl in a water bath and determined by a double channel Atomic Fluorescence Spectrometer with a hollow cathode lamp of Hg and As and high purity argon gas as a carrier (AFS-9100). The amounts of soil and vegetable samples for analysis were 0.5 g and 5 g, respectively. The limits of detection (LOD) for Pb, Cd, Cr, Hg and As were 11.9, 2.3, and 113.7, 2.0 and 20.0 ng L^{-1} respectively, the limits of quantity (LOQ) for these five metals were 35.7, 6.9, 341.1, 6.0 and 60.0 ng L^{-1} respectively.

2.4. Risk assessment methods

The human health risk models including carcinogenic and non-carcinogenic ones raised by US EPA, have proved successful and adopted worldwide. Currently, there is no agreed limit for acceptable

Table 1
Defining equations of daily intake via various exposure pathways[a].

Medium	Exposure pathway	Calculation formula
Soil	Ingestion	$CDI_{ingest\text{-}soil} = \frac{CS \times IRS \times EF \times ED}{BW \times AT} \times CF$
	Dermal contact	$CDI_{dermal\text{-}soil} = \frac{CS \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF$
	Diet	$CDI_{vegetable} = \frac{C_{vegetable} \times IR_{vegetable} \times EF \times ED}{BW \times AT}$
Water	Oral intake	$CDI_{oral\text{-}water} = \frac{C_{water} \times IR_{oral\text{-}water} \times EF \times ED}{BW \times AT}$
	Dermal intake	$CDI_{dermal\text{-}water} = \frac{C_{water} \times EV_{shower} \times EF \times ED \times SA \times AF \times ABS \times CF}{BW \times AT}$
Air	Inhalation	$CDI_{inhale\text{-}soil} = \frac{CS \times ET \times EF \times ED}{PEF \times 24 \times AT}$

[a] CDI = chronic daily intake; CS — exposure-point concentration: mg/kg; C_{water} — concentration in groundwater: mg/L; EF — exposure frequency: 350 d/a (USEPA, 2011); ED — exposure duration: 30 a (USEPA, 2011); ET — exposure frequency: 24 h/d (UDOE, 2011); AT — averaging time for non-carcinogens: 365 × EDd (USEPA, 2011); AT — averaging time for carcinogens: 365 × 70 d (USEPA, 2002); BW — body weight: 70 kg(USEPA, 1991);SA — exposured skin area: 5700 cm^2(USEPA, 2011); AF — adherence factor: 0.07 $mg \cdot cm^{-2}$(USEPA, 2011); ABS — dermal absorption fraction: 0.03 (As) 0.001 (other metals) (USEPA, 2011); PEF — particle emission factor: 1.36 × 10^9 m^3 kg^{-1} (USEPA, 2002); CF — units conversion factor: 10^{-6} kg mg^{-1} (USEPA, 2002); IRS — ingestion rate:100 $mg \cdot d^{-1}$ (USEPA, 2011); $IR_{vegetable}$ — ingestion rate:0.345 $kg \cdot d^{-1}$ (Wang et al., 2005).

maximum carcinogenic and non-carcinogenic risk levels in China. We therefore employed the US EPA model and their threshold values to assess the potential human health risks posed by heavy metal pollution in this study. The health risk assessment was divided into four steps: (1) hazard identification, (2) dose–response assessment, (3) exposure assessment, and (4) risk characterization (US EPA, 1989, 1992). The multiphase and multicomponent risk assessment model developed by US EPA was used to evaluate the heavy metal pollution hazard in urban residential areas (US EPA, 2004). Three sources, i.e. soil, groundwater, and air were used to calculate the intake doses of heavy metals. The metals in the soil may leach to the groundwater and cause consequent risks. Human beings could be exposed to heavy metals from vegetable soils via the following six main pathways: (1) direct ingestion of soil particles, (2) dermal contact with soil particles, (3) diet through the food chain, (4) inhalation of soil particles from the air, (5) oral intake from groundwater, and (6) dermal intake from groundwater.

The calculations for the daily exposure dose of contaminants via various exposure pathways and the detailed explanation for all the parameters are listed in Table 1 (US EPA, 2000, 1991, 2002, 2004; Wang et al., 2005).

The risk effects consist of carcinogenic and non-carcinogenic risk assessments for all the metals through ingestion, inhalation, dermal and diet exposure pathways in the study area. Cancer risk can be evaluated from:

$$\text{Cancer risk} = \text{CDI} \times \text{SF}$$

where cancer risk represents the probability of an individual lifetime health risks from carcinogens; CDI is the chronic daily intake of carcinogens (mg kg^{-1} d^{-1}); SF is the slope factor of hazardous substances (mg kg^{-1} d^{-1}).

The cumulative cancer risk can be calculated from:

$$\text{Total cancer risk} = \sum_{k=1}^{n} \text{CDI}_k \text{SF}_k$$

where CDI_k is the chronic daily intake (mg kg^{-1} d^{-1}) of substance k, SF_k is the slope factor for substance k (kg d^{-1} mg^{-1}). The acceptable or tolerable risk for regulatory purposes is within the range of 10^{-6}–10^{-4} (US EPA, 2001).

The non-carcinogenic risk from individual heavy metals can be expressed as the hazard quotient:

$$\text{HQ} = \text{CDI}/\text{RFD}$$

where the non-cancer hazard quotient (HQ) is the ratio of exposure to hazardous substances, and RFD is the chronic reference dose of the toxicant (mg kg^{-1} d^{-1}).

$$\text{Chronic hazard index} = \sum_{k=1}^{n} \text{CDI}_k/\text{RFD}_k$$

where the chronic hazard index (HI) is the sum of more than one hazard quotient for multiple substances or multiple exposure pathways, CDI_k is the daily intake of the heavy metal (k) and RFD_k is the chronic reference dose for the heavy metal k. HI values > 1 shows that there is a chance that non-carcinogenic risk may occur, and when HI < 1 the reverse applies.

2.5. Quality control and statistical analysis

In order to guarantee the accuracy of data, standard reference materials (GBW07429, from the National Institute of Metrology of China) were included in every batch of sample digestion and analysis as a part of the quality control protocol. Each sample was analyzed in triplicate and two standards were tested after every 10 samples. The calibration curves were linear within the concentration range, with the regression coefficients (R^2) > 0.999. Relative standard deviations

Table 2
Descriptive statistical analysis for the heavy metal concentrations in vegetables and soils.

	Vegetables					Corresponding soils				
	Pb	Cd	Cr	As	Hg	Pb	Cd	Cr	Hg	As
Mean	0.426	0.0472	0.4038	0.0326	0.0024	68.6444	0.7206	47.7356	0.7568	15.5107
SD	0.0386	0.0031	0.0249	0.0035	0.0005	4.1364	0.0411	0.7729	0.0932	1.5167
5	0.0118	0.0063	0.0517	0.0036	0.0004	11.5156	0.1646	26.775	0.1163	5.2159
10	0.0186	0.0088	0.0667	0.0049	0.0006	14.8971	0.1886	30.0954	0.119	5.5365
25	0.0428	0.0185	0.1032	0.0076	0.001	23.8657	0.2526	37.2641	0.3016	6.5282
Median	0.2202	0.0328	0.2616	0.0181	0.0016	39.4711	0.378	51.0079	0.5412	8.8692
75	0.5693	0.0541	0.5643	0.0356	0.0024	93.312	1.0731	56.2481	0.9509	12.1808
90	1.1274	0.0969	1.0373	0.0725	0.0031	160.2	1.7872	62.1155	1.1913	39.0555
95	1.7834	0.1501	1.2225	0.1211	0.0038	191.27	2.1367	68.9863	1.4502	52.1447
p (K–W)	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.240	0.408

P (K–W): p value of Kruskal–Wallis test among vegetable species or vegetable-corresponding soils species; 5, 10, 25, 75, 90, 95 are various percentiles of the heavy metals concentration in vegetables or soils.

(RSDs) of repeated measurements were <10%. These results showed that the elemental analysis method was both reliable and precise.

Descriptive statistics were calculated by using the SPSS software ver. 18. Origin 8 was employed for all the graph plotting. The data were displayed using the parameters of the minimum value, maximum value, mean value, the median, standard deviation and 95% UCL.

3. Results and discussion

3.1. Characterization of soil and vegetable samples

The physicochemical properties of 267 soil samples from the study area had a wide range of pH values (4.49–8.38; median: 6.4), EC

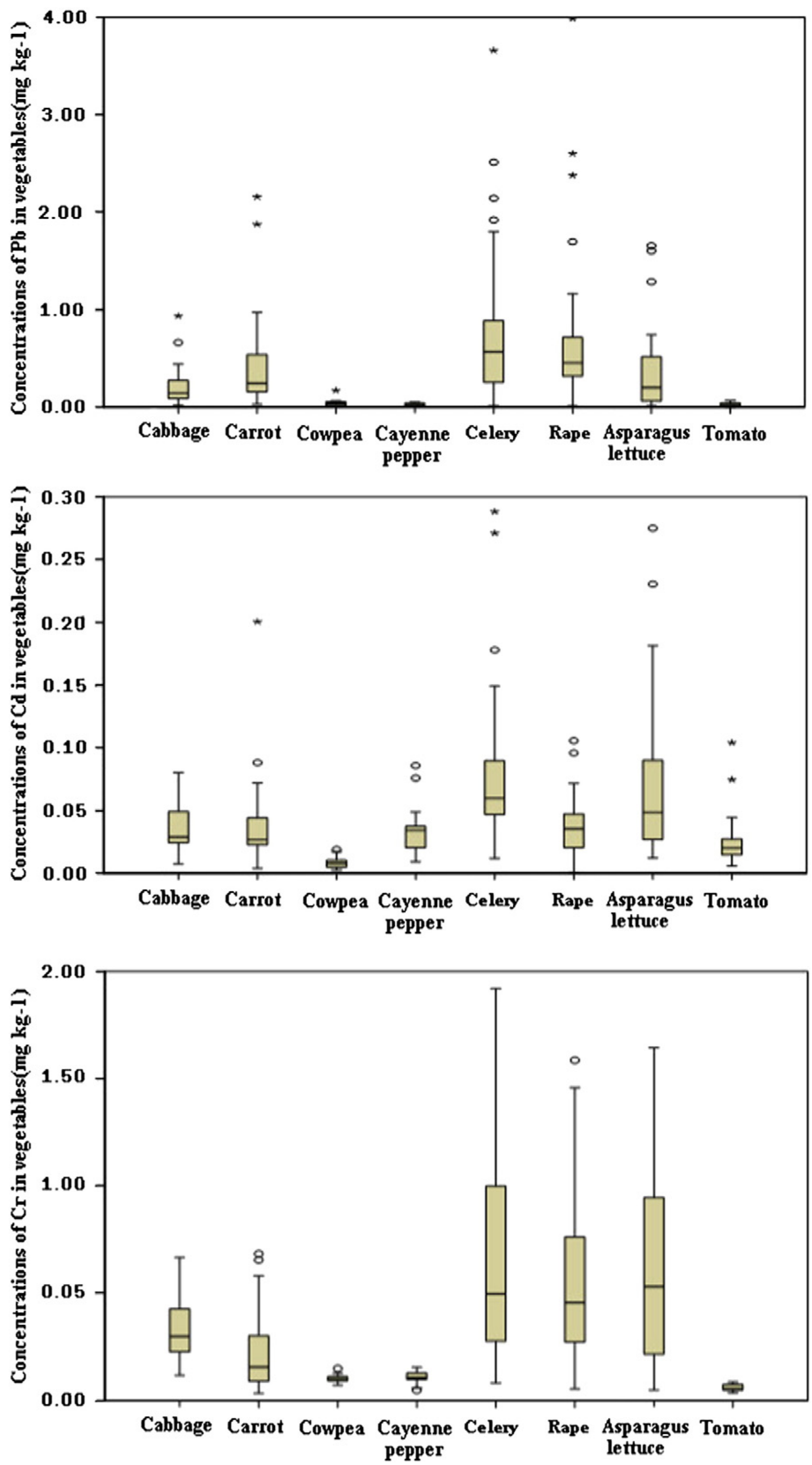

Fig. 1. Total concentrations of heavy metals in vegetables of the study area.

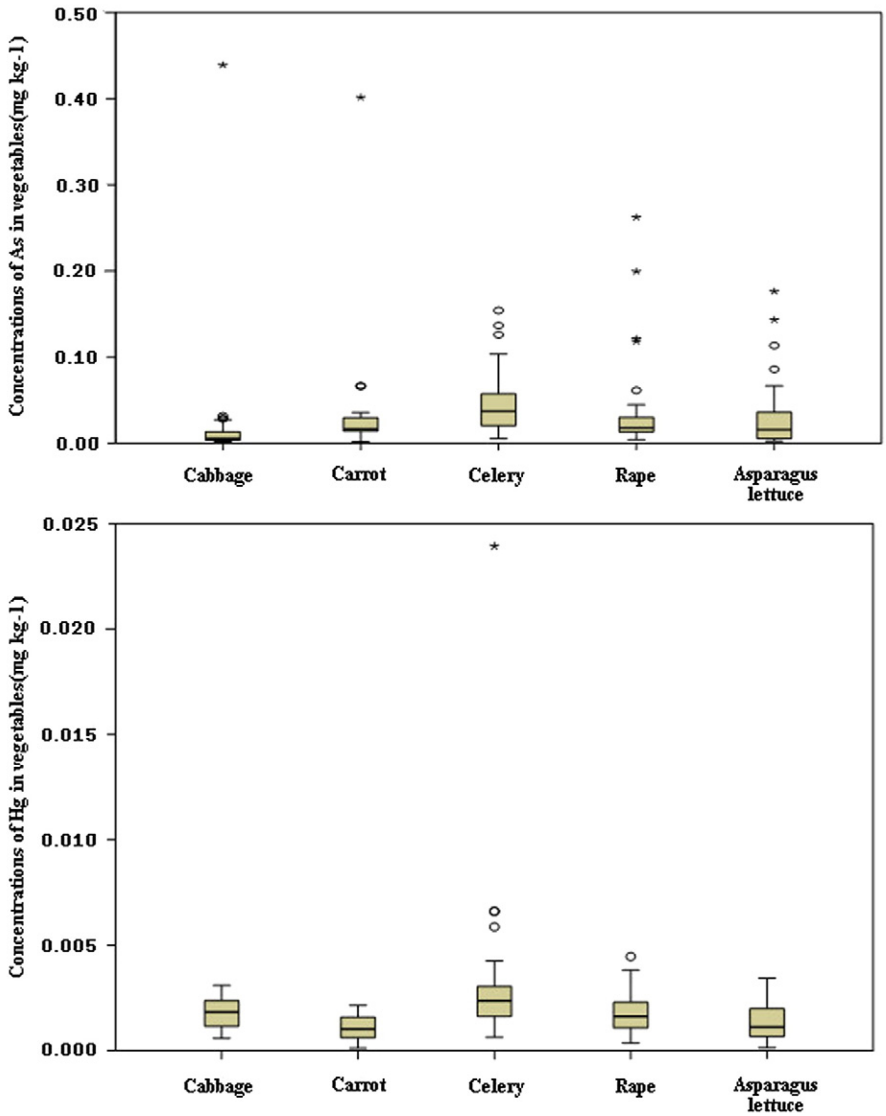

Fig. 1 (*continued*).

(60–1241; median: 315), CEC (11–29; median: 17.73 cmol kg^{-1}), OC (4.02–34.62; median: 13.88 g kg^{-1}) and contents of clay (8–70; median: 24 %), silt (4–56; median: 36%), and sand (4–58; median: 20 %). The soil basic properties varied among different vegetable planted fields. The descriptive statistical analysis for the data of all the heavy metal concentrations in the vegetables and their corresponding soils are shown in Table 2.

The average metal concentrations in the investigated vegetables and soils are shown in Figs. 1 and 2. The heavy metal concentrations varied among different vegetables (Fig. 1), due to their different accumulation abilities and various soil properties. Heavy metal concentrations, especially of Pb and Cd, in cowpea, tomato and cayenne pepper were lower than in other vegetables. Asparagus lettuce plants accumulated lower Hg concentrations but higher Cd and Cr concentrations compared to rape. Celery accumulated the highest concentration of Cr and tomato the lowest. In general, Pb concentrations in leafy vegetables and rootstock vegetables were significantly higher than in legume and solanaceous vegetables. Compared with the threshold values issued by the Chinese Ministry of Health, the Pb concentrations in leafy and rootstock vegetables were above the food safety limits, which suggests a potential risk in view of product quality and human health. In contrast, Hg and As concentrations in all the investigated vegetables, with a range of 0.013–0.044 mg kg^{-1} and 0.001–0.005 mg kg^{-1}, respectively, were below their food safety limits. Hg and As were not detected in cowpea, tomato and cayenne pepper, so these two metals in their corresponding soils were not analyzed.

The majority of vegetable soils contained high concentrations of heavy metals (Fig. 2). The highest Cd concentration was in carrot-planted soil, followed by rape-planted soil and cabbage-planted soil. There was no significant difference in mean concentrations of soil Cr between different vegetable groups. The average As concentration in leafy and rootstock soils was approximately 0.65 mg kg^{-1}, which was much higher than the background value.

There were no statistically significant correlations between soil and vegetable metal concentrations. This concurs with the non-statistically significant correlation coefficients between the heavy metal concentrations in different vegetables and their corresponding soils (0.10–0.31, $P > 0.05$) Therefore, additional environmental factors should be monitored to explore the sources of heavy metal pollution to guarantee both arable soil quality and food security.

3.2. Non-carcinogenic risk assessment in the soil–vegetable system

The non-carcinogenic risks (HQ) of all the heavy metals through different exposure routes for local adults residents were determined (Fig. 3). In this study area, the main portion of non-carcinogenic risks

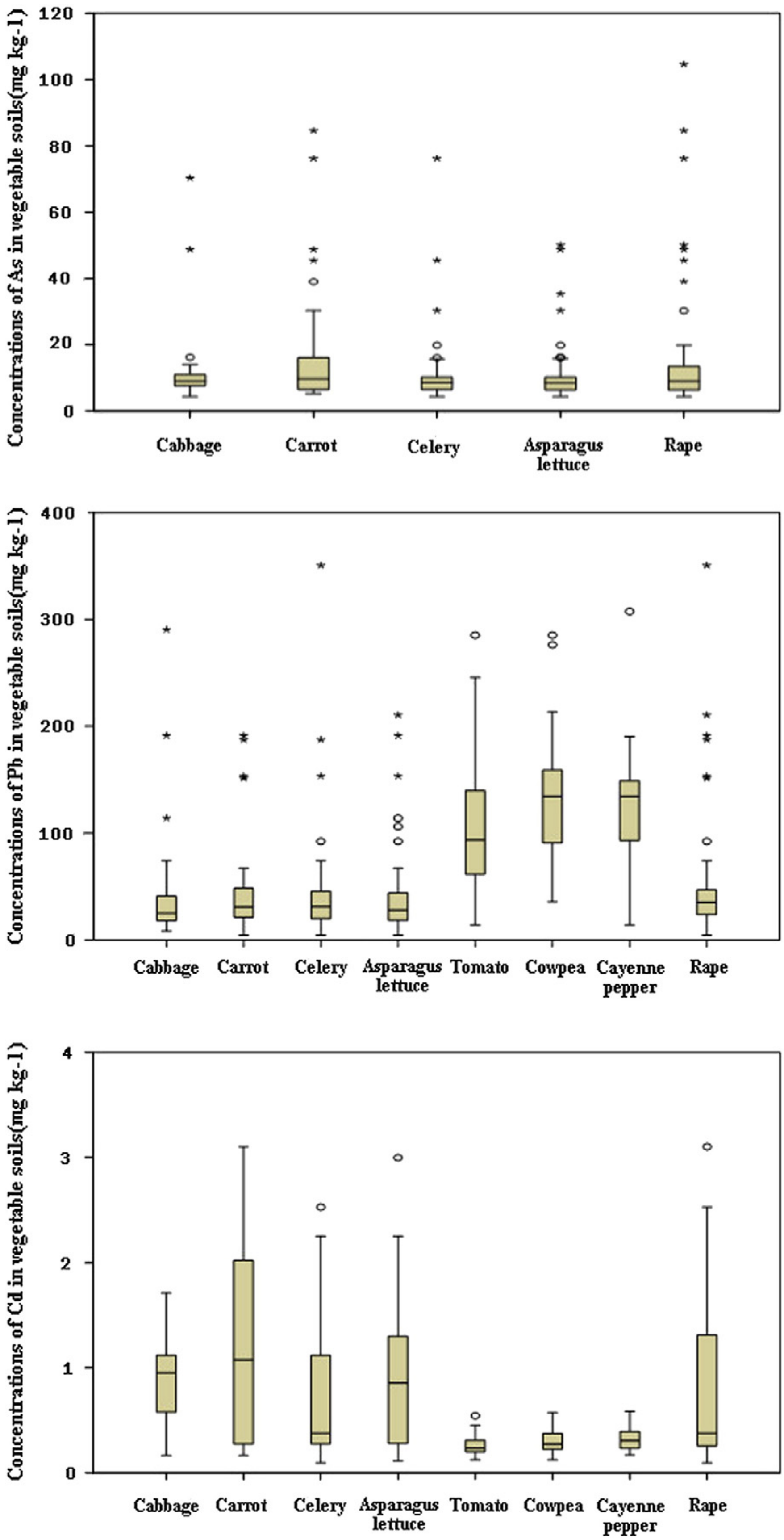

Fig. 2. Total concentrations of heavy metals in soils of the study area.

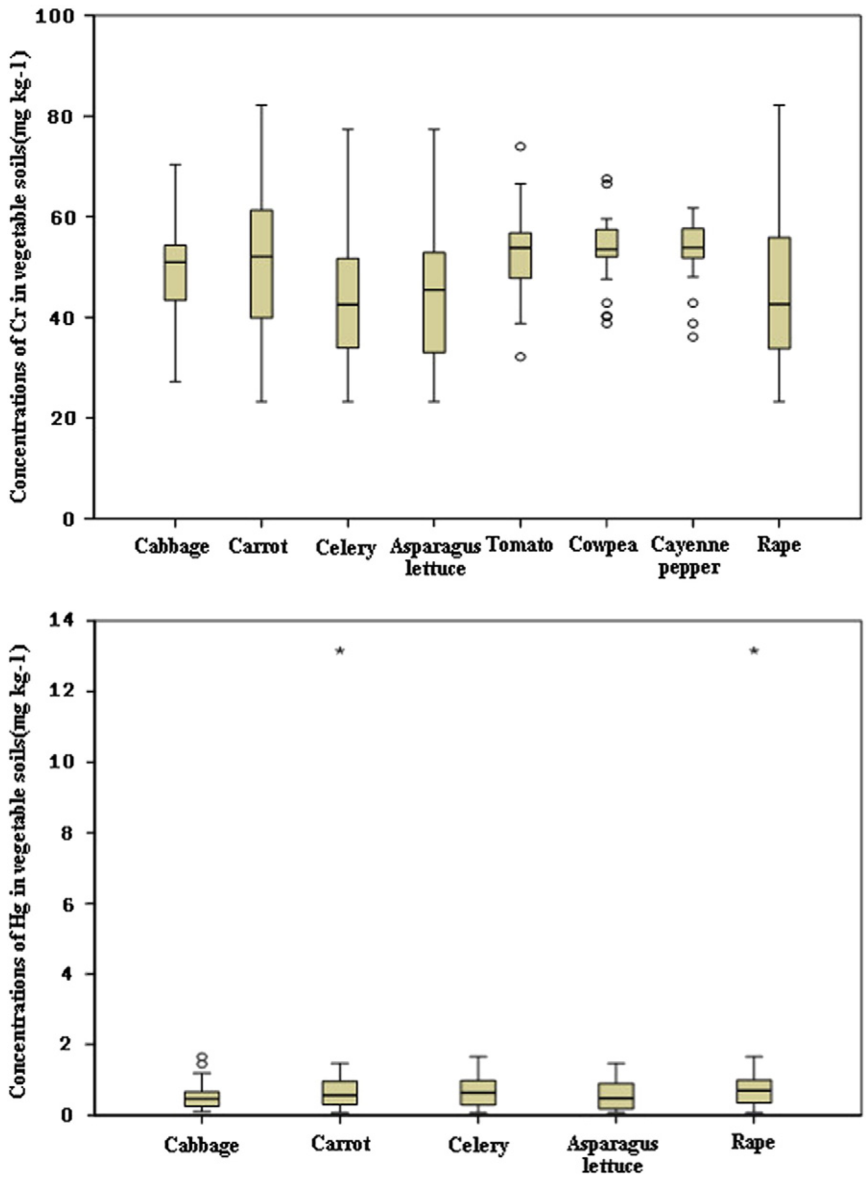

Fig. 2 (*continued*).

resulted from diet. The HQ of rape, celery, cabbage, asparagus lettuce and carrot derived from the diet are much higher than the USEPA guideline values, showing that the above-mentioned vegetables are unsafe for human consumption. The authors consider that a large daily intake of these vegetables is likely to cause a significant health hazard to the residents and consumers. Another prime non-carcinogenic risk stems from ingestion of soil, while inhalation of soil and dermal absorption soil create relatively low hazards. The contribution of drinking water and other foods except for vegetable was not included in these estimates because of their low risks which could be ignored.

Fig. 4 summarizes the separate non-carcinogenic risk of five metal pollutants. Among the metals, Pb and Cr presented relatively higher potential health risks, followed by As and Cd. In addition, the non-carcinogenic risks of Pb and Cr in soils growing leafy vegetables such as rape, celery, asparagus lettuce were significantly higher than those evaluated in other soils growing similar vegetables. It is notable that Cd presented higher non-carcinogenic risks in soils growing leafy vegetables than in the soils growing rootstock vegetables. Mercury concentrations can be considered to be safe for people living in residential areas due to their low non-carcinogenic risks. Thus we can even ignore the effect of Hg on human health. However, heavy metal risks from cabbage, carrot, cowpea, tomato and cayenne pepper were far less than the toxic thresholds set by USEPA. The risk of Hg and As in cowpea, tomato and cayenne pepper as well as their corresponding soils were not included because of a lack of data.

The results of non-carcinogenic risks of heavy metals through four exposure routes are shown in Table 3. The diet pathway, which accounted for 85.57% to 99.40%, was the dominant exposure route of all the metals to local residents. For each metal, the average risk values of all the samples did not exceed their permissible levels even though the four exposure pathways were all considered. The HQ of the pollutants decreased in the following order: Cr > Pb > As > Hg > Cd, and their risk values were 0.74, 0.69, 0.55, 0.42 and 0.24, respectively. This result was different than the order reported for indoor or street dust, where Pb ranked first (Zheng et al., 2010; Perihan, 2012). This could be due to their different primary exposure pathways for arable soil and dust. Therefore, for the non-carcinogenic risks, we could firstly reduce their hazard from diet. Then, more attention should be paid to Cr and Pb pollution.

The total non-carcinogenic hazard indexes (HI) for various heavy metals and multiple exposure pathways are summarized in Table 4. The risks from rape and celery were three times higher than the

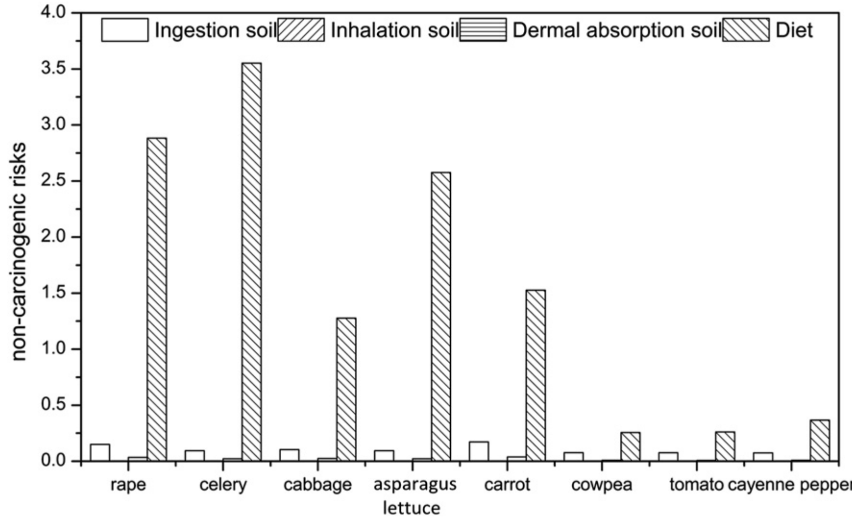

Fig. 3. Non-carcinogenic risks through four exposure pathways.

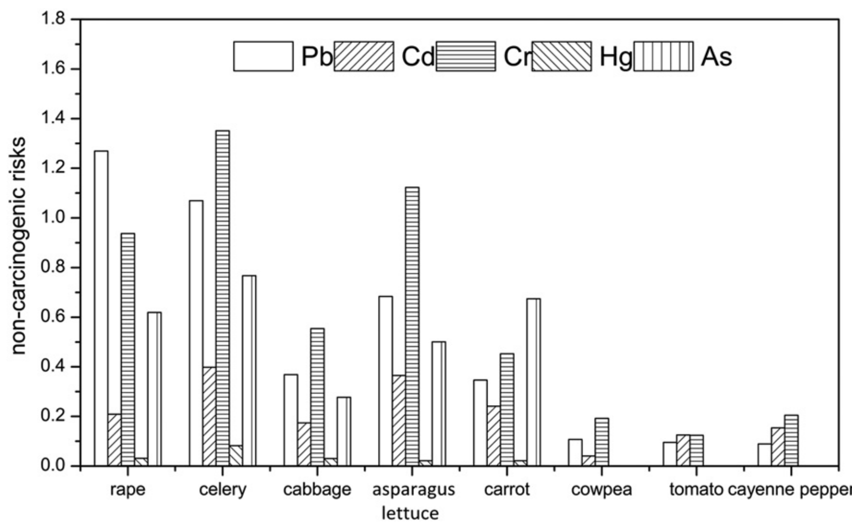

Fig. 4. Non-carcinogenic risks of five heavy metals.

threshold value of 1. The risk from asparagus lettuce is 2.69 times higher than the threshold value and the risks for cabbage and carrot were slightly above the limit. The heavy metals in cowpea, tomato and cayenne pepper may not pose a problem as a result of low HI values. Therefore, local residents could eat more solanaceous and legume vegetables instead of other vegetables in order to reduce toxicity.

Table 3
Non-carcinogenic risks (HQ) through four exposure pathways in heavy metals.

Pathway / Metal		Ingestion soil	Inhalation soil	Dermal absorption soil	Diet
Pb	Risk	0.027539	–	–	0.6657
	Contribution	3.97%	–	–	96.03%
Cd	Risk	0.000996	1.51351E-05	0.00039736	0.2363
	Contribution	0.42%	6.37E-03%	0.17%	99.40%
Cr	Risk	0.021684	0.000345669	0.006655185	0.7073
	Contribution	2.95%	4.70E-02%	0.90%	96.10%
As	Risk	0.061685	–	0.018	0.4726
	Contribution	11.17%		3.26%	85.57%
Hg	Risk	0.003059	1.62759E-06	0.000174384	0.4253
	Contribution	0.71%	3.80E-04%	4.07E-02	99.20%

3.3. Carcinogenic risk assessment in the soil–vegetable system

At the 95% confidence level, all the confidence interval values for the mean risks were calculated. The mean risk values fell within the 95% confidence intervals, thus we used the mean values in all the risk calculation formulae. The proportions of different exposure routes, as indicated from the potential cancer risk assessments (Fig. 5) were asymmetrical. Of the five investigated elements, only Cd, Cr and As were carcinogenic. However, these cancer-causing pollutants generated no significant carcinogenic effects from inhalation or dermal absorption from soil. The risk levels through diet exposure pathways in all the vegetables were from 10^{-4} to 10^{-3}, about 10^2 to 10^3 times higher than those due to inhalation and dermal absorption soil pathways, while the mean cancer risk via ingestion of soil was 10 times higher than inhalation and dermal pathways. Hence, as with the non–carcinogenic risk, diet was the dominant exposure pathway causing cancer risk when compared to the other routes. Therefore, assessing dietary exposure to heavy metal residues, as with pesticides, in food crops should be a key step in authorization procedures (Peter et al., 2012).

The results of the cancer risks for individual elements are explained in Fig. 6. Compared to two other metals of Cr and As, Cd seemed to be the predominant contaminant that created a relatively

Table 4
Non-carcinogenic hazard indexes (HI) for five heavy metals and four exposure pathways.

	Rape	Celery	Cabbage	Asparagus lettuce	Carrot	Cowpea	Tomato	Cayenne pepper
Total non-carcinogenic risks	3.07	3.67	1.41	2.69	1.74	0.34	0.34	0.45

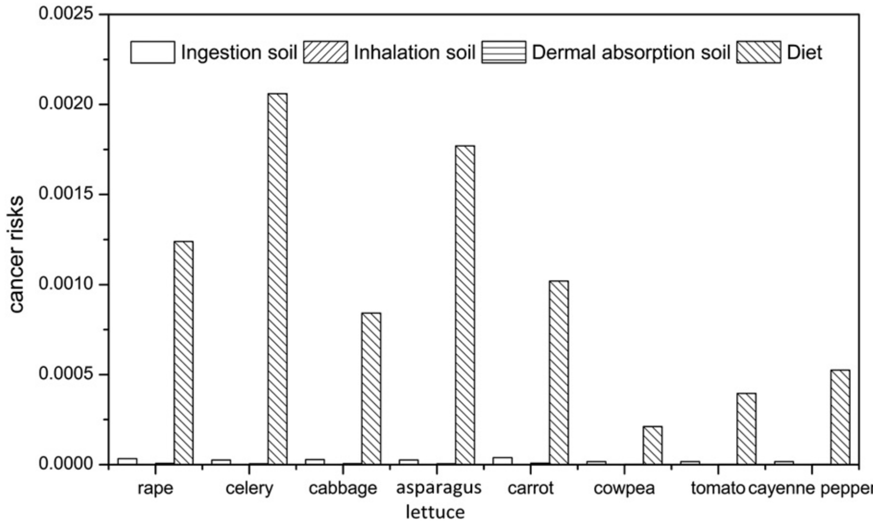

Fig. 5. Cancer risks through four main exposure pathways.

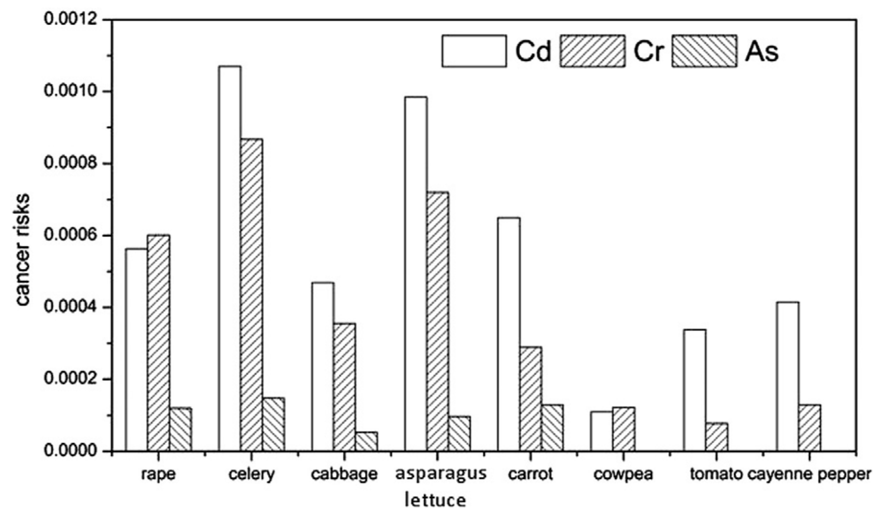

Fig. 6. Cancer risks of main heavy metals.

higher risk, followed by Cr and As. The cancer risks existed in all vegetable soils, which varied from the minimum value of 5.93×10^{-5} for Cr in tomato-planted soil to the maximum value of 1.07×10^{-3} for Cd in rape-planted soil through the diet pathway. Cowpea-planted soil posed the smallest carcinogenic hazard, consistent with the results of non-carcinogenic risks.

The cancer risks from heavy metals for the four main exposure pathways, and the total values for all routes (Table 5) ranged from 1.07E-04 to 6.41E-04. Diet pathway was the dominating exposure route for all the heavy metals to local residents which accounted for 85.55% to 99.58% of the total cancer risk. The cancer risks decreased in the order of Cd > Cr > As. Therefore, Cd appears to be the main pollutant source to produce cancer among these heavy metals.

The comprehensive assessment results of cancer risks in different kinds of vegetable soils are listed in Table 6. A wide range of cancer risks was calculated, from 2.32E-04 to2.10E-03 in the vegetable soils by the US Environmental Protection Agency. Our results showed slightly higher risks than an acceptable range of 1E-06 to 1E-04. The total combined risk for residents showed the following decreasing order: rape (2.10E-03) > cabbage (1.80E-03) > asparagus lettuce (1.27E-03) > carrot (1.07E-03) > celery (8.74E-04) > cowpea

Table 5
Cancer risks for four exposure pathways in heavy metals.

Pathway / Metal		Ingestion soil	Inhalation soil	Dermal absorption soil	Diet
Cd	Risk	2.6889E-06	3.95675E-10	–	6.38014E-04
	Contribution	0.42%	6.18E-05%	–	99.58%
Cr	Risk	1.39394E-05	1.7795E-07	2.22473E-06	4.54712E-04
	Contribution	2.96%	0.038%	0.47%	96.53%
As	Risk	1.18964E-05	1.80796E-08	3.47455E-06	9.11449E-05
	Contribution	11.17%	0.017%	3.26%	85.55%

Table 6
Cancer risks for five heavy metals and four exposure pathways.

	Rape	Celery	Cabbage	Asparagus lettuce	Carrot	Cowpea	Tomato	Cayenne pepper
Total cancer risks	2.10E-03	8.74E-04	1.80E-03	1.27E-03	1.07E-03	5.45E-04	2.32E-04	4.16E-04

(5.45E-04) cayenne pepper (4.16E-04) > tomato (2.32E-04). Likewise, leafy and rootstock vegetable showed higher cancer risks than legumes and solanaceous vegetables. These results should be considered in agricultural policy making and urban planning procedures such as land-use structure adjustment. For example, farmers could plant vegetables with low accumulation abilities such as cayenne pepper, tomato and cowpea instead of leafy and rootstock vegetables. Alternatively, polluted arable soils could be used for production of horticultural crops or taken out of agriculture and used for roads or other construction purposes. Soil remediation approaches could also be applied. However, the most important first step is to find and control the pollution sources to prevent the further pollution of agricultural soils. This would provide a powerful balance between environmental protection, food safety improvement, health risks reducing and massive savings of cost (Howard and Sammy, 2012; Peter et al., 2012).

For the risk assessment model, since the pollutants exposure procedure has the characteristics of spatio-temporal variation, the input parameters for the exposure model could be varied with time and space. Therefore, the spatial and temporal simulation of multi-pollutants, multi-sources and multi-pathways demand further studies. Although the model may provide over-estimations or underestimations, the results still can provide some valuable information and forecast to a certain extent for local government to cope with these current severe environmental problems.

4. Conclusions

The extent of heavy metal contaminations varied with metal species and vegetable types. The scenario of heavy metal contents is Pb > Cr > As > Cd > Hg, and leafy vegetables > root vegetables > solanaceous vegetables > legume vegetables.

Cr has the biggest non-carcinogenic effects while Cd generates the greatest cancer risk. Therefore, effective measures should be adopted to control Cd and Cr pollution in the study area. Certainly, the inhabitants are currently experiencing a significant potential health risk solely due to the consumption of leafy and root vegetables grown locally.

Whether for non-carcinogenic or cancer risks, leafy and rootstock vegetables posed higher hazards than solanaceous and legume vegetables. Accordingly, we recommend that local people should not consume large quantities of these vegetables, and thereby avoid large accumulations of heavy metals in the body. Furthermore, chemical risk assessments including both pesticides and heavy metals should be conducted as part of the vegetable authorization procedure.

Polluted soils can endanger human and animal health by various exposure routes. Thus, an urgent and systematic study of the heavy metals in soils growing vegetables and an assessment of pollution source apportionment is recommended since it could significantly decrease the intake of these toxic elements, and thus contribute to the improved human health of the local residents. Further studies are needed in order to assess more accurately the heavy metal intakes from vegetables by local residents and animals.

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