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Assessment of heavy metal pollution in vegetables and relationships with soil heavy metal distribution in Zhejiang province, China

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Abstract There are increasing concerns on heavy metal contaminant in soils and vegetables. In this study, we investigated heavy metal pollution in vegetables and the corresponding soils in the main vegetable production regions of Zhejiang province, China. A total of 97 vegetable samples and 202 agricultural soil samples were analyzed for the concentrations of Cd, Pb, As, Hg, and Cr. The average levels of Cd, Pb, and Cr in vegetable samples [Chinese cabbage (*Brassica campestris* spp. Pekinensis), pakchoi (*Brassica chinensis* L.), celery (*Apium graveolens*), tomato (*Lycopersicon esculentum*), cucumber (*Colletotrichum lagenarium*), cowpea (*Vigna unguiculata*), pumpkin (*Cucurbita pepo* L.), and eggplant (*Solanum melongena*)] were 0.020, 0.048, and 0.043 mg kg^{-1}, respectively. The Pb and Cr concentrations in all vegetable samples were below the threshold levels of the Food Quality Standard (0.3 and 0.5 mg kg^{-1}, respectively), except that two eggplant samples exceeded the threshold levels for Cd concentrations (0.05 mg kg^{-1}). As and Hg contents in vegetables were below the detection level (0.005 and 0.002 mg kg^{-1}, respectively). Soil pollution conditions were assessed in accordance with the Chinese Soil Quality Criterion (GB15618-1995, Grade II); 50 and 68 soil samples from the investigated area exceeded the maximum allowable contents for Cd and Hg, respectively. Simple correlation analysis revealed that there were significantly positive correlations between the metal concentrations in vegetables and the corresponding soils, especially for the leafy and stem vegetables such as pakchoi, cabbage, and celery. Bio-concentration factor values for Cd are higher than those for Pb and Cr, which indicates that Cd is more readily absorbed by vegetables than Pb and Cr. Therefore, more attention should be paid to the possible pollution of heavy metals in vegetables, especially Cd.

Keywords Heavy metal · Pollution · Vegetable · Soil · Health risk

Introduction

Agricultural soil has both direct and indirect influences on public health via food consumption; it is therefore of great importance to protect this resource and ensure its sustainability. Anthropogenic activities, such as mining and industrial processing, have led to increased release of heavy metals into agricultural soils (Wong et al. 2002). Heavy metal contamination has become an increasingly severe problem in China (Wang et al. 2005; Huang et al. 2007; Khan et al. 2008) and leads to growing public concern over the potential accumulation of heavy metals in agricultural soils, which poses a great threat to human health through the food chain (Wang et al. 2008).

In the food consumption structure, the proportion of vegetables has increased with the improvement of living

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standards, and vegetables are one of the most important pathways through which heavy metals enter the food chain and affect human health (Wang et al. 2004). There is a raised concern over heavy metal pollution in vegetables and its potential risks to human health (Cui et al. 2004; Wang et al. 2008; Huang et al. 2014).

Industrial emissions and irrigation water might be the primary pollution pathways for heavy metals into agricultural soils (Jiang et al. 2006). Once the heavy metals are released into agricultural soils, they could be accumulated by vegetables (Hao et al. 2009). For example, Luo et al. (2011) noted the high Cd level in broccoli and high Pb level in lettuce growing in contaminated soil at an electronic waste processing site. Gupta et al. (2008) observed 17.79 mg kg^{-1} for Cd and 57.63 mg kg^{-1} for Pb in radish collected from a wastewater-irrigated area in Titagarh, Indian. Human exposure to heavy metal contaminations of soils and vegetables is a matter of health concern. The intake of vegetables containing heavy metals has a potential health risk to consumers. It is therefore imperative to investigate heavy metal pollution in vegetables and the relationships between the metal concentrations in vegetables and the corresponding soils.

Zhejiang province, China, is one of the most rapidly developing regions with a high population density and has experienced accelerated industrialization and urbanization in recent years. Intensive production has gradually become the mainstream of agricultural development. Although some studies have reported heavy metal contamination in Zhejiang province (Fu et al. 2008; Tang et al. 2010), to our knowledge, there are few studies concerning heavy metal contamination in vegetables and health risk assessment. Hence, we conducted a study with the following objectives: (1) to investigate the contamination of heavy metals in vegetables and soils in Zhejiang province, (2) to examine the relationships between heavy metal concentrations in vegetables and the corresponding agricultural soils, and (3) to explore spatial variations and potential sources of heavy metals. Results obtained from this study are expected to provide some insights for reducing the risk of heavy metals to human health.

Materials and methods

Sample description

All of the sampling sites were chosen in the main vegetable production regions in Zhejiang province, China, including Hangzhou City (120°19′ E, 30°26′ N), Jiaxing City (120°76′ E, 30°77′ N), Ningbo City (121°56′ E, 29°86′ N), Shaoxing City (120°58′ E, 30°01′ N), and Taizhou City (121°27′ E, 28°41′ N). A total of 97 vegetable samples were collected from vegetable yards, and 202 agricultural soil samples (0–20 cm in depth) were also obtained from the corresponding sites. All of the samples were collected from June to October, 2010, put in plastic bags, and transported to the laboratory on the day of sampling.

The vegetable samples were Chinese cabbage (*Brassica campestris* spp. Pekinensis), pakchoi (*Brassica chinensis* L.), celery (*Apium graveolens*), tomato (*Lycopersicon esculentum*), cucumber (*Colletotrichum lagenarium*), cowpea (*Vigna unguiculata*), pumpkin (*Cucurbita pepo* L.), and eggplant (*Solanum melongena*).

Soil classification is Ustic Cambosols from Hangzhou and Ningbo cities, Udic Ferrisols from Shaoxing and Taizhou cities, and Stagnic Anthrosols from Jiaxing City. These soils were affected by different anthropogenic sources of pollution: steel plant, mining, and highway emission.

Sample analysis

Vegetable samples were washed carefully with tap water and deionized water. The concentrations of Cd, Pb, and Cr in vegetables were determined as described by Fu et al. (2011). Briefly, the samples were digested as follows: a 5–10-g vegetable sample was added to a 100-mL round-bottom flask. Then, 10 mL concentrated nitric acid was mixed with the sample and heated for 120 °C. Hydrogen peroxide at 1 mL was added periodically until the digestion was complete, i.e., a clear solution was reached. Finally, the digest was transferred to a 50-ml volumetric flask, made up to volume, and filtered. The concentrations of heavy metal in the filtrate were determined using inductively coupled plasma-mass spectrometry (Thermo Electron Corporation, Xserie2) following a standard procedure.

Soil samples were air-dried and sieved through a 2- and 0.149-mm nylon mesh, respectively, for pH, organic matter measurement, and total metal analysis. Soil organic matter was determined by the modified Walkley–Black titrimetric procedure (Storer 1984). Soil pH was measured using 1:2.5 (*w*/*v*) soil/de-ionized water ratio. For determination of total Cd, Pb, Cr, As, and Hg in soil, portions of each 0.20 g of soil samples were digested with HNO_3–$HClO_4$–HF (5:1:1) (Shentu et al. 2008). These soil properties are listed in Table 1.

In the analysis of metals, two certified reference materials (soil GSBZ 50013-88 and plant NCSZC73008) approved by the General Administration of Quality Supervision, Inspection, and Quarantine of the People's Republic of China were used in the digestion and analysis as part of the QA/QC protocol. Recoveries were ranged between 94.2 and 103.4 % for all of the metals in the soil (GSBZ 50013-88) and plant reference materials (NCSZC73008). Detection limits were defined as three times the standard deviation of ten runs of blank measurements. Detection limits of Cd, Pb, As, Hg, and Cr were 0.001, 0.005, 0.005, 0.002, and 0.01 mg kg^{-1}, respectively.

Statistical analyses

Linear regression analysis was performed using the statistical package SPSS 18.0 for Windows (CoHort Software, Berkeley, CA, USA). The data for soils and vegetables showed normal distributions, and the degree of confidence in the procedure was $a<0.05$.

Results and discussion

Heavy metal contents in vegetables

The contents of heavy metals (Pb, Cd, and Cr) in 97 vegetable samples (Chinese cabbage, pakchoi, celery, tomato, cucumber, cowpea, pumpkin, and eggplant) are shown in Figs. 1, 2, and 3. As and Hg contents in these vegetables were below the detection level (0.005 and 0.002 mg kg^{-1}, respectively). The average levels of Cd, Pb, and Cr were 0.020, 0.048, and 0.043 mg kg^{-1}, respectively. Compared with the maximum levels of contaminants in foods cited from GB2762-2012 (National Health Agency of China 2012), the Pb and Cr concentrations in all of the vegetable samples were below the threshold levels (0.3 and 0.5 mg kg^{-1}, respectively), except that two eggplant samples exceeded the threshold level for Cd concentration (0.05 mg kg^{-1}).

The ability of heavy metal absorption varied in different vegetables. The Pb contents in celery and pakchoi, 0.072–0.137 and 0.081–0.109 mg kg^{-1}, respectively, were much higher than those in cabbage, cowpea, cucumber, eggplant, pumpkin, and tomato (0.021–0.068, 0.039–0.067, 0.013–0.067, 0.034–0.059, 0.025–0.085, and 0.011–0.041 mg kg^{-1}, respectively) (Fig. 1), which might be attributed to the strong ability of Pb absorption by celery and pakchoi (Hashmi et al. 2007). The Cd contents in cabbage, celery, and pakchoi, respectively, of 0.017–0.068, 0.032–0.057, and 0.022–0.053 mg kg^{-1} were slightly higher than those in cucumber, eggplant, tomato, cowpea, and pumpkin (0.004–0.022, 0.014–0.051, 0.012–0.039, 0.002–0.012, and 0.001–0.011 mg kg^{-1}, respectively) (Fig. 2). In general, Cd contents in leafy and stem vegetables are higher than those in fruit vegetables. The results are in agreement with previous reports (Voutsa et al. 1996; Wang et al. 2004; Alexander et al. 2006) that leafy vegetables had the highest concentration of trace elements, followed by stem vegetable, root vegetable, and fruit vegetable. The Cr contents in cabbage, celery, and pakchoi, respectively, of 0.029–0.099, 0.098–0.167, and 0.038–

Table 1 Physio-chemical properties of the soils

Soil site		Total Cd (mg kg^{-1})	Total Cr (mg kg^{-1})	Total Pb (mg kg^{-1})	Total As (mg kg^{-1})	Total Hg (mg kg^{-1})	pH	OM (g kg^{-1})
Hangzhou	Mean	0.39	81.4	30.3	7.57	0.42	6.1	31.5
	Range	0.07–1.50	41.2–168.1	20.7–67.3	4.51–18.05	0.15–0.81	4.09–8.15	17.2–49.3
Jiaxing	Mean	0.26	82.7	29.5	7.89	0.42	6.1	31.3
	Range	0.13–3.25	83.0–136.4	11.9–41.2	5.00–12.20	0.11–0.97	4.14–7.89	14.1–45.1
Ningbo	Mean	0.34	82.9	30.1	7.6	0.42	6.04	29.6
	Range	0.11–0.35	73.1–108.9	13.4–55.0	2.31–24.53	0.09–1.96	2.86–8.52	7.65–43.2
Shaoxing	Mean	0.34	82.6	33.6	7.36	0.38	6.03	30.9
	Range	0.12–0.50	36.0–120.8	19.3–65.8	3.31–12.50	0.19–2.27	3.86–8.12	8.14–53.3
Taizhou	Mean	0.21	82.8	27.4	6.39	0.3	6.13	28.6
	Range	0.06–0.59	16.4–195.0	17.0–45.5	2.19–10.70	0.06–0.49	3.95–8.47	13.2–57.8

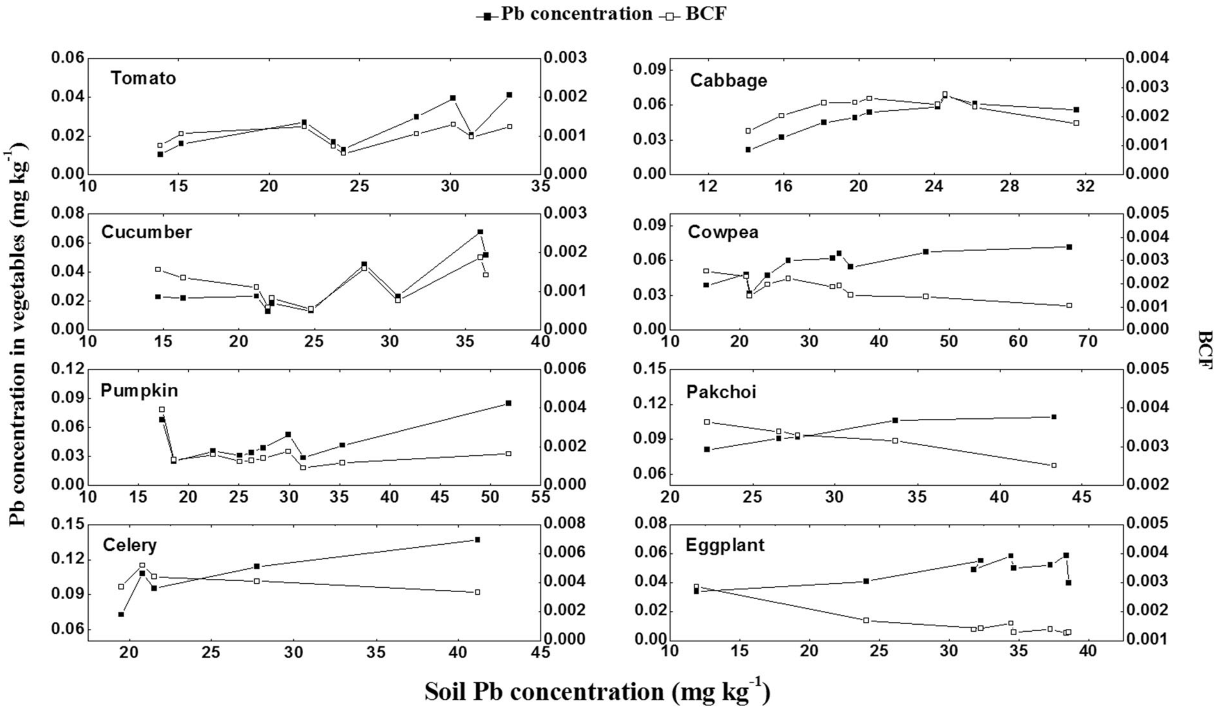

Fig. 1 Lead concentrations in edible parts of vegetables and the corresponding BCF values

0.093 mg kg^{-1} were higher than those in cowpea, cucumber, eggplant, pumpkin, and tomato (0.020–0.043, 0.013–0.024, 0.012–0.035, 0.021–0.062, and 0.017–0.040 mg kg^{-1}, respectively) (Fig. 3).

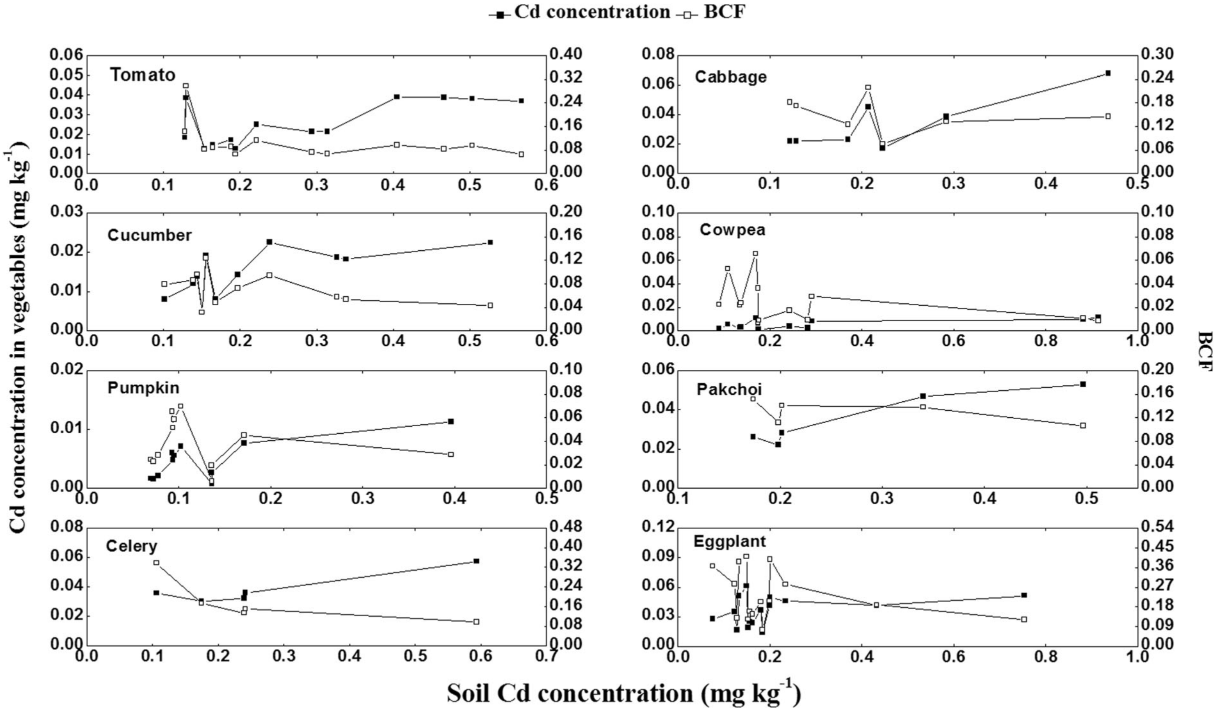

Fig. 2 Cadmium concentrations in edible parts of vegetables and the corresponding BCF values

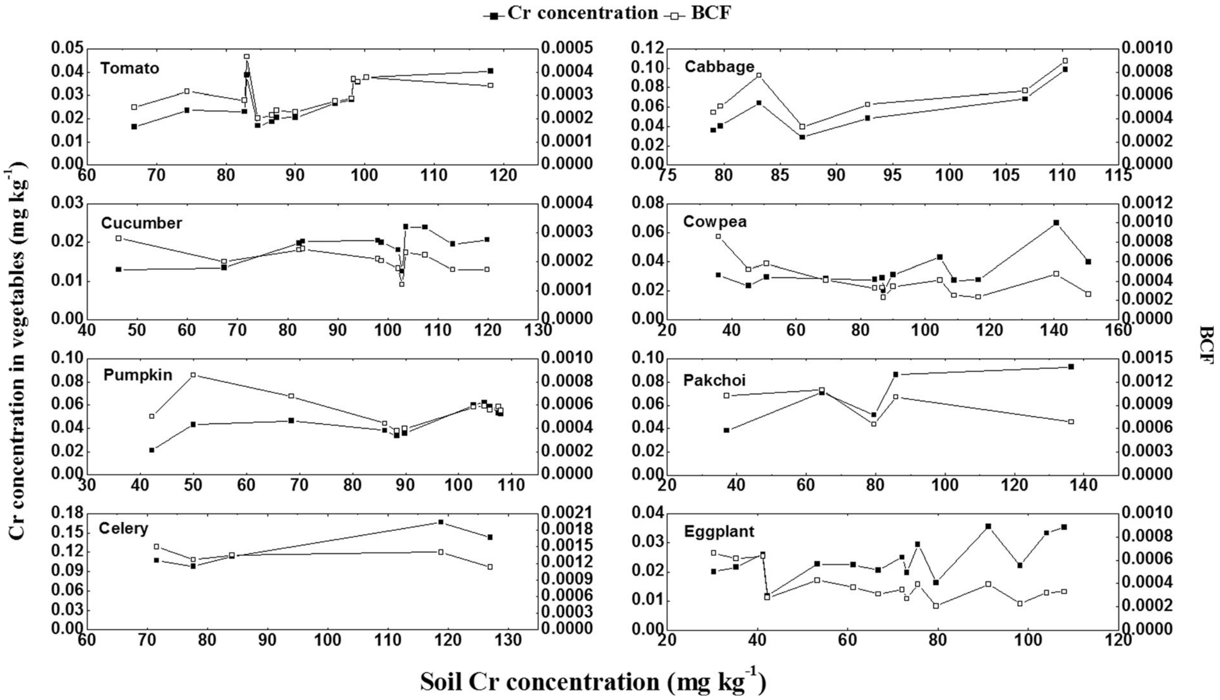

Fig. 3 Chromium concentrations in edible parts of vegetables and the corresponding BCF values

Soil contamination of heavy metals

Heavy metal concentrations in 202 soil samples within the corresponding vegetable yards are presented in Table 1. Table 2 also lists the critical limits of soil metal concentrations of China as cited from GB 15618-1995 (National Environmental Protection Agency of China 1995). The critical limits were defined as the values above which toxicity was considered to occur. Soil pollution conditions were assessed in accordance with the second level of GB 15618-1995; 50 and 68 soil samples from the investigated sites exceeded the maximum allowable level for Cd and Hg, respectively. The concentrations of Pb, As, and Cr were below the soil quality criteria. For soil Cd, the polluted areas are mainly concentrated in Hangzhou, Ningbo, and Shaoxing. Industrial pollution, sewage irrigation, and fertilizer application could have contributed to Cd contamination in these sites (Liu et al. 2006). For soil Hg, the polluted areas are mainly concentrated in Hangzhou, Ningbo, Jiaxing, and Shaoxing. Long-term use of fertilizers and pesticides has led to significant accumulation of Hg in the topsoils (Huang et al. 2007).

Table 2 Environmental quality standard for soils in China cited from GB 15618-1995 (mg kg^{-1})

Levels	First level	Second level			Third level
pH	nature	<6.5	6.5~7.5	>7.5	>6.5
Cd ≤	0.20	0.30	0.60	1.0	1.0
Hg ≤	0.15	0.30	0.50	1.0	1.5
As paddy field ≤	15	30	25	20	30
Dry land ≤	15	40	30	25	40
Pb ≤	35	250	300	350	500
Cr paddy field ≤	90	250	300	350	400
Dry land ≤	90	150	200	250	300

To understand the effect of soil property on heavy metal accumulation, the correlativity between the five heavy metals and soil properties (pH and OM) was analyzed (Table 3). The correlation coefficients showed a significant ($p<0.01$) positive correlation between OM contents and soil Pb, Cd, and Hg concentrations, which was probably attributed to fertilizer and sludge application. Meanwhile, the correlation coefficients showed a significant ($p<0.01$) positive correlation between pH and soil Pb and Hg concentrations, which was probably due to sewage sludge application (Liu et al. 2006).

Table 3 Pearson partial correlations matrix for the heavy metals, pH, and OM in agricultural soils

	As	Pb	Cd	Hg	Cr	pH
Pb	0.229**					
Cd	0.219**	0.329**				
Hg	0.147*	0.452**	0.165*			
Cr	0.139	−0.145	0.089	0.039		
pH	0.007	−0.376**	−0.07	−0.34**	0.11	
OM	0.104	0.492**	0.200**	0.206**	0.102	−0.265**

*Significant level at 0.05

**Significant level at 0.01

Spatial distributions of soil heavy metals

A multivariate statistical treatment was used to identify geochemical associations between soil heavy metals (Fig. 4). The analyzed metals are clustered into three groups as discussed in the following paragraphs.

Group I: Hg–Cd

The main source of heavy metal pollutants is the excessive use of various fertilizers and pesticides that contained heavy metals such as Cd and Hg. For example, Cd is found in phosphatic fertilizers because Cd is commonly present as an impurity in phosphatic rocks. Yang et al. (2005) measured Cd and Hg contents in phosphatic fertilizers of 0.009–2.58 and 0.005–0.69 mg kg^{-1}, respectively. Excessive doses of phosphate fertilizers have been applied to vegetable yards for over 30 years in China, resulting in accumulation of heavy metals within agricultural soils (Huang et al. 2007). It is known that Cd and Hg pollute agricultural soils in China because of the use of agrochemicals. Our results are consistent with previous surveys that report significant associations between Cd and Hg in fertilized soils (Wong et al. 2002; Huang et al. 2007).

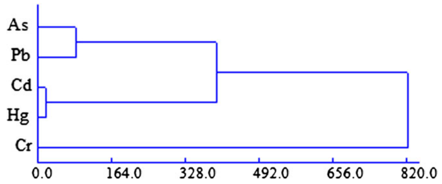

Fig. 4 Dendrogram of heavy metal elements in Zhejiang agricultural soils

Group II: As–Pb

Agricultural soils in China are usually contaminated by heavy metals derived from atmospheric deposition (Chen et al. 1999). Previous studies have confirmed that atmospheric inputs can be a significant contributor to As, Pb, Zn, and Cu loading in agricultural soils (Berthelsen et al. 1995; Alloway et al. 1999; Gray et al. 2003). The main sources of heavy metals in the atmosphere are traffic, energy production, mining, metal smelting and refining, waste incineration, and manufacturing processes. The contamination of agricultural soils by atmospheric heavy metals mainly occurs by wind-blown fine granule material that originates from industrial areas (Alloway et al. 1999). Metals of As and Pb deposited on the soil surface are then gradually incorporated into the soil, thereby contributing to overall soil As and Pb contamination.

Group III: Cr

This element group comprises primary element within rocks and soils. In terms of Cr, it is the seventh most abundant element on earth (McGrath and Smith 1995). Chromium abundance on the earth's crust ranges from 100 to 300 μg g^{-1} (Shewry and Peterson 1976). Where present in natural soils, Cr was derived from the weathering of parent material and subsequent pedogenesis. Chromium occurs in soils at a concentration from 10 to 150 mg kg^{-1} (Adriano 2001). Minor variations in Cr concentrations among the areas are probably related to variations in the contents of soil clay/sand or organic matter, which appear to influence the concentrations of Cr (Vaselli et al. 1997).

Relationship between metal concentrations in vegetables and the corresponding soils

Simple correlation analysis was performed to obtain the relationship between metal concentrations in vegetables and the corresponding soils, and the correlation coefficients are summarized in Table 4. For some metals, such as Cd, they exist primarily in soluble or exchangeable, readily bioavailable forms (Lasat 2002). These bioavailable forms can be easily taken up by plants, which explained the reason for positive correlation for all vegetables surveyed. Among all the heavy metals studied, vegetables contaminated by Cd have relatively higher potential health risks, especially for pakchoi

Table 4 Correlation coefficients between soil metal concentrations and metal concentrations in edible parts of vegetables

	Pb	Cd	Cr
Cabbage	0.638**	0.668**	0.597*
Pakchoi	0.668**	0.749**	0.622*
Celery	0.616*	0.723**	0.697*
Tomato	0.547*	0.530*	0.493
Cucumber	0.588*	0.485*	0.394
Cowpea	0.575*	0.526*	0.381
Pumpkin	0.471	0.524*	0.414
Eggplant	0.484	0.499*	0.411

(r=0.749**), celery (r=0.723**), and cabbage (r=0.668**).

It should be pointed out that soil metals were not all correlated with metal concentrations in vegetables. Lead is strongly bound with soil particles, which results in a significant soil Pb fraction being insoluble, displaying low phytoavailability (Wang et al. 2004). However, Pb contents in some vegetables had positive correlations with those of soil Pb (i.e., r=0.638** for cabbage; r=0.668** for pakchoi; r=0.616* for celery; r=0.547* for tomato; r=0.588* for cucumber; r=0.575* for cowpea). Therefore, the vegetables cabbage, pakchoi, celery, tomato, cucumber, and cowpea have relatively high health risks when cultivated on Pb-contaminated soils.

The potential health risk of Cr is the lowest, which may be ascribed to its low phytoavailability. The toxicity of Cr is directly dependent on its valence state (i.e., Cr(VI) and Cr(III)). The solubility and mobility of Cr(III) in soils are minimal in comparison to Cr(VI), which exhibits a high degree of solubility and mobility in soil (Robson 2003). Cr(VI) can be reduced to Cr(III) in soils by various redox reactions with aqueous inorganic species, electron transfer at mineral surfaces, reactions with nonhumic organic substances such as proteins and carbohydrates, or reduction by soil humic substances (Kozuh et al. 2000). The reduction of Cr(VI) dominates over the possible oxidation of Cr(III); thus, the major form of Cr is Cr(III) in most soils, which results in the low phytoavailability of Cr. Only the vegetables cabbage, pakchoi, and celery have positive correlations with soil Cr contents (i.e., r=0.597* for cabbage; r=0.622* for pakchoi; r=0.697* for celery).

Bio-concentration factor (BCF) values were generally used to relate pollutant residues in plants to the pollutant concentration in the environment. The BCF value can be calculated based on the ratio of heavy metal concentrations in plants to soil. We considered heavy metals in vegetable edible parts as the heavy metal residues in plants (Krishnamurti and Naidu 2002). BCF values of heavy metals are shown in Figs. 1, 2, and 3. The BCF values for Cd, Pb, and Cr in the sampled vegetables are respectively below 0.5, 0.01, and 0.002. In our study, the levels of heavy metals in analyzed vegetables were much lower than the amounts recorded in the soils in which the vegetables were grown, leading to low BCF values. The low BCF values indicate that only limited quantities of heavy metals are absorbed into the upper parts of the vegetables, probably because the roots act as a barrier to the translocation of metals within the vegetables (Davies and White 1981). This may also reflect the small percentage of soluble heavy metals in the soil because of the subalkaline environment. pH and organic matter play an important role in the retention of heavy metals in the soil (Hernandez et al. 2003).

The BCF values for Cd is higher than those for Pb and Cr, which indicates that Cd is more readily absorbed by vegetables than Pb and Cr. Xue et al. (2005) also reported that Cd had maximal BCF values among Cd, Pb, Cr, Cu, and Zn. In most cases, the BCF values decrease with increasing heavy metal concentrations in the soil, especially for Cd. Similar result was also reported by Samsoe-Petersen et al. (2002) and Huang et al. (2007).

Conclusion

The present study examined heavy metal pollution in vegetables and the corresponding soils in the main vegetable production regions of Zhejiang province. The average levels of Cd, Pb, and Cr in 97 vegetable samples (Chinese cabbage, pakchoi, celery, tomato, cucumber, cowpea, pumpkin, and eggplant) were 0.020, 0.048, and 0.043 mg kg^{-1}, respectively. The Pb and Cr concentrations in all the vegetable samples were below the threshold levels of the Food Quality Standard (0.3 and 0.5 mg kg^{-1}, respectively), except that two eggplant samples exceeded the threshold levels for Cd concentrations (0.05 mg kg^{-1}). As and Hg contents in these vegetables were below the detection level (0.005 and 0.002 mg kg^{-1}, respectively).

A total of 50 and 68 soil samples from the investigated sites, respectively, exceeded the maximum allowable contents for Cd and Hg according to the Chinese

Soil Quality Criterion (GB15618-1995, Grade II). The Cd–Hg accumulation possibly resulted from the long-term use of heavy doses of agrochemicals. The As–Pb in soils may be derived from atmospheric deposition associated with anthropogenic activity. The Cr in soil originated from the weathering of parent material and subsequent pedogenesis.

There were significant positive correlations between metal concentrations in vegetables and the corresponding soils, especially for the leafy and stem vegetables, such as pakchoi, cabbage, and celery. The BCF values are higher for Cd than for Pb and Cr. Therefore, more attention should be paid to the pollution of heavy metals in leafy and stem vegetables, especially Cd.

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