

Accumulation of heavy metals in leaf vegetables from agricultural soils and associated potential health risks in the Pearl River Delta, South China

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Abstract This study investigated the extent of heavy metal accumulation in leaf vegetables and associated potential health risks in agricultural areas of the Pearl River Delta (PRD), South China. Total concentrations of mercury (Hg), cadmium (Cd), lead (Pb), chromium (Cr) and arsenic (As) were determined in 92 pairs of soil and leaf vegetable (flowering Chinese cabbage, lettuce, pakchoi, Chinese cabbage, loose-leaf lettuce, and Chinese leaf mustard) samples collected from seven agricultural areas (cities). The bioconcentration factors (BCF) of heavy metals from soil to vegetables were estimated, and the potential health risks of heavy metal exposure to the PRD residents through consumption of local leaf vegetables were assessed. Results showed that among the six leaf vegetables, pakchoi had the lowest capacity for heavy metal enrichment, whereas among the five heavy metals, Cd had the highest capacity for transferring from soil into vegetables, with BCF values 30-fold those of Hg

and 50-fold those of Cr, Pb and As. Sewage irrigation and fertilization were likely the main sources of heavy metals accumulated in leaf vegetables grown in agricultural areas of the PRD region. Different from previous findings, soil pH had no clear effect on metal accumulation in leaf vegetables. Despite a certain degree of metal enrichment from soil to leaf vegetables, the PRD residents were not exposed to significant health risks associated with consumption of local leaf vegetables. Nevertheless, more attention should be paid to children due to their sensitivity to metal pollutants.

Keywords Pearl River Delta · Leaf vegetables · Heavy metals · Bioconcentration factor · Target hazard quotient

Introduction

Heavy metals can accumulate and migrate in soil environments. Due to their cumulative effects and long-term interactions, accumulation of heavy metals in soil negatively affects regional eco-safety and poses a threat to relevant animals and plants. Additionally, heavy metals can enter human bodies through the food chain, leading to an increased incidence of chronic diseases such as deformity and cancer (Müller and Anke 1994; Ramadan and Al-Ashkar 2007; Tembo et al. 2006). Studies have shown that fruit and vegetable consumption is the primary pathway of human exposure to heavy metals (Adamsa et al. 2004; McLaughlin et al. 1999). Therefore, it is of practical significance to assess the extent of heavy metal accumulation from soil into plants such as fruits and

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vegetables, and relevant research has gained increasing attentions.

The behaviors of metal in soils are very complex, which involve adsorption–desorption, complexation–dissociation, oxidation–reduction, ion exchange, and other carrier transport role. The former two reactions mainly affect metal activity in soil, while oxidation–reduction can also change metal valence (Swartjes et al. 2007). These chemical mechanisms can work together and affect one another, which formed a complex interaction system controlling metal fates. Heavy metals enter the vegetable tissues mainly through the roots and foliage, of which root uptake was the dominant pathway. Metals can be transferred from soil pore water into the plants through the roots in the form of dissolved ions (e.g., Cd^{2+}) (McLaughlin et al. 2011). Series of complex processes occur in the soil pore water and crop rhizosphere.

The degree of metal enrichment in crops can be described with enrichment factor, which is defined as the ratio of a particular element content in a plant to that in soil. The concept of enrichment factor was first proposed by Zoller et al. (1974) for source diagnosis of atmospheric particulate matter in the Antarctic. In the following year, enrichment factor was used for investigating high-altitude atmospheric substances in the North Atlantic Ocean (Duce et al. 1975). Recently, the application of enrichment factor in the form of transfer factor, bioconcentration factor (BCF), and plant uptake factor has been expanded to research on soil, water system, and sediment, as well as assessment of heavy metal pollution in environmental geochemistry (Wang et al. 2006a, b; Khan et al. 2010; Brioschi et al. 2013; Delgado et al. 2012). In soil research, BCF is defined as the ratio of the content of a particular element in a plant to that in soil. BCF is an important quantitative indicator of crop contamination and has commonly been used for estimating metal transfer from soil into plants (García et al. 2009; Melgar et al. 2009; USEPA 2005). BCF-based research shows that the extent of metal enrichment in vegetables is highest in leaf vegetables, followed by tubers and fruit vegetables (Liu et al. 2012; Pandey and Pandey 2009). Regarding metal concentrations, cadmium (Cd) and lead (Pb) commonly occur at high levels in leaf vegetables while the Zn content of tubers is higher than other metal contents (Ngole 2011).

The bioavailability and toxicity of metals in soil are significantly influenced by pH condition (Badawy et al. 2002; Wang et al. 2006b). Soil pH is considered to be one of the most important factors that influence the transfer of

Cd and Pb from soil to plants, and higher pH values have been found to reduce the bioavailability and toxicity of Cd and Pb (McBride et al. 1997; Gray et al. 1999). There are many researchers who devote themselves to the research on the relation between soil pH and Cd uptake (Costa and Morel 1993; Hart et al. 2002; Kim et al. 2002; Tudorean and Phillips 2004; McLaughlin et al. 2011). The consistent conclusion is that the adsorption between soil particles and Cd increases with the increase of soil pH, while Cd bioavailability decreased. The mechanism for this phenomenon can contribute to the increases of solubility and ion competition. As the soil pH decreases, the concentrations of Fe^{2+} , Mn^{2+} , Zn^{2+} , and Ca^{2+} increase in soil solution, which enhance the competition of free ions and reduce the adsorption to soil particles (Tudorean and Phillips 2004). Additionally, Speir et al. (2003) reported that soil pH is the greatest determinant of the solubility and mobility of chromium (Cr), Pb and Zn in a sandy soil. Due to the close relation between soil pH and heavy metal properties, correlation analysis of pH value and heavy metal accumulation is commonly applied in research on accumulation of heavy metal from soil to vegetables.

The Pearl River Delta (PRD) region in South China covers an area of 54,733 km², with 13,357 km² of agricultural land and 4,829 km² of vegetable area (Fig. 1) (Guangdong Statistics Bureau 2012; Lu 1988). The PRD region is the largest vegetable production base in Guangdong Province and produced 11,291,987 tons of vegetable in 2011. It mainly exports to Hong Kong and Macao, which receive more than 50 % of the total export volume of vegetables from the PRD (Guangdong Statistics Bureau 2012). Being one of the most developed regions in China, the PRD region has experienced substantial industrial pollution in recent decades. Associated soil and air pollution can enhance pollutant accumulation in vegetables, further increasing potential human health risks. However, previous studies on heavy metal accumulation in soil and/or vegetable in the PRD region were mainly focused on polluted soils in suburb, industrial or mining areas. Presently, there is a lack of report on heavy metal uptake by vegetables in large agricultural bases in the PRD region (Luo et al. 2011; Zhao et al. 2012).

To this end, the present study assessed the extent of heavy metal accumulation in leaf vegetables commonly grown in agricultural areas in the PRD region. Total concentrations of mercury (Hg), Cd, Pb, Cr and arsenic (As) were determined in 92 pairs of leaf vegetable and soil samples. The accumulation of heavy metals in leaf

vegetables was interpreted using BCF, and the relation between BCF values and soil pH was examined by principal component analysis (PCA) and Pearson correlation analysis. The potential human health risks of heavy metal exposure to the PRD residents through consumption of local leaf vegetables were evaluated using the target hazard quotient (THQ). The results will provide reference data for further investigation of heavy metal accumulation in the PRD region, thereby improving the regional food safety.

Materials and methods

Soil and vegetable sampling and processing

Ninety-two pairs of soil and vegetable samples were collected in the PRD region during September–November 2011 (Fig. 1). The sampling locations were chosen from large vegetable bases and national basic farmland protection zones (>30 ha) distant from urban and industrial areas. Six kinds of leaf vegetables were collected during the harvest period, including flowering Chinese cabbage (*Brassica campestris* L. ssp. *chinensis* var. *utilis* Tsen et Lee), lettuce (*Lactuca sativa* L. var. *romana* Hort), pakchoi (*Brassica chinensis* L.), Chinese cabbage (*Brassica pekinensis* (Lour.) Rupr), loose-leaf lettuce (*Lactuca sativa* L.), and Chinese leaf mustard (*Brassica juncea* Coss). Soil samples were taken from the surface layer (0–20 cm) using a bamboo shovel and gently shaken off from the vegetables roots. All samples were sealed in polyethylene bags and transported to the laboratory within 6 h of collection.

The soil samples were air-dried at room temperature, with impurities manually removed. Then, the soils were ground and sieved through 80 meshes (0.2 mm). For vegetable samples, the decay and withered tissues were removed and the edible parts were washed with tap water to remove surface dirt. The edible parts of vegetables were repeatedly rinsed with deionized water and dried at 60 °C to a constant weight. The dry vegetable samples were crushed with a wooden hammer in a carnelian mortar and then passed through an 80-mesh sieve.

Heavy metal and soil pH analyses

The concentrations of heavy metals in soil and vegetable samples were determined according to the methods

of Rasmussen et al. (1991). Briefly, 600 mg of air-dried soil was mixed with 6 mL of concentrated HNO₃–HClO₄ (87:13, v/v) and 6 mL of concentrated HCl. The mixture was digested and then dissolved in 2 % HCl solution. The vegetable samples were digested in a mixture of HNO₃–HClO₄–H₂O₂ (87:13:10, v/v/v). The Hg and As concentrations of digestion solutions were determined using an atomic fluorescence morphological analyzer (SA-10, Titan, Beijing, China), Pb and Cr concentrations were determined using a WFX-130 flame atomic adsorption spectrophotometer (Braic, China), and the Cd content was determined using a Z-2700 graphite furnace atomic absorption spectrophotometer (Hitachi, Japan). Standard reference materials of soil samples (GBW 07418 and GBW07429 (GSS-15)) and citrus leaves (GBW10020 (GSB-11)) were assayed during sample analysis for quality control. Reagent blanks were included to ensure detection precision (<5 %). Soil pH was measured in soil slurries using a 1:2.5 soil-to-water ratio with a PHS-3C pH meter (Sartorius, China).

Data analyses

BCF calculation

BCF was calculated as follows:

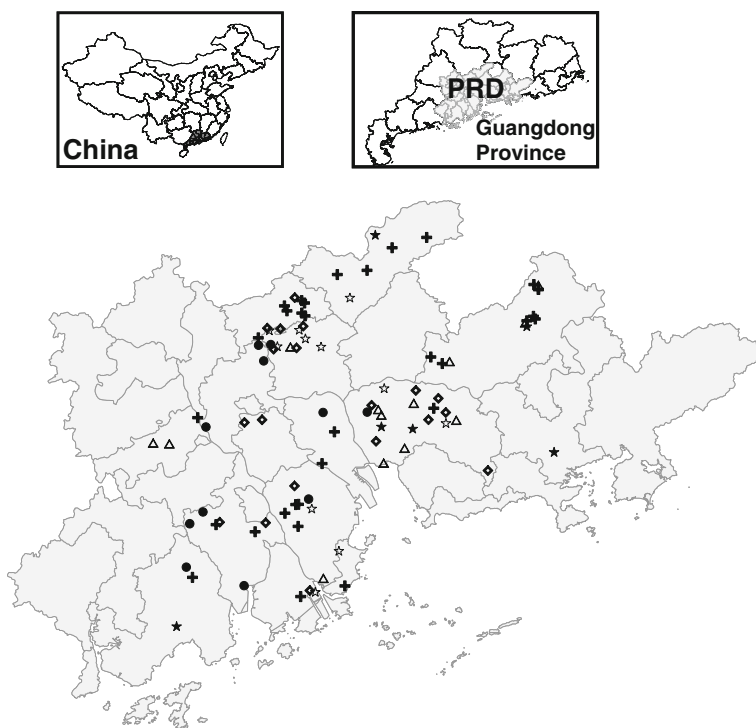
$$\text{BCF} = \frac{C_{\text{vegetable}}}{C_{\text{soil}}} \quad (1)$$

where $C_{\text{vegetable}}$ is the total concentration of a particular heavy metal in the vegetable (mg kg⁻¹ dw), and C_{soil} is the corresponding heavy metal concentration in the soil habitat of the vegetable (mg kg⁻¹).

Vegetable consumption-associated health risk assessment

Potential health risks of heavy metal exposure to PRD residents through local leaf vegetable consumption were assessed using THQ that was first proposed by the United States Environmental Protection Agency (USEPA) for assessing the potential health risks of pollutant exposure to human health (USEPA 2002; Liu et al. 2006; Song et al. 2009; Wang et al. 2005). THQ is defined as the ratio of the body intake dose of a pollutant to the reference dose (Eqs. 1 and 2). If THQ > 1, there will be a potential risk associated with this

Fig. 1 Agricultural soil and leaf vegetable samplings locations in the Pearl River Delta region, South China (*plus sign*, flowering Chinese cabbage; *diamond*, lettuce; *black circle*, pakchoi; *triangle*, Chinese cabbage; *white star*, loose-leaf lettuce; *black star*, Chinese leaf mustard)



pollutant. If $THQ < 1$, there will be no obvious potential risk associated with this pollutant.

$$\text{Single pollutant: } THQ = \frac{C \times IR_{\text{vegetable}} \times EF \times ED}{BW \times AT \times RfD} \times 10^{-3} \quad (2)$$

$$\text{Total pollutants: } THQ_{\text{total}} = \sum THQ_{(\text{single pollutant})} \quad (3)$$

where C is the mean concentration of a particular metal in a vegetable, IR is the daily vegetable intake by the PRD residents (including local and extraneous vegetables), EF is the exposure frequency (exposure days per year), ED is the exposure duration, BW is the average weight of local residents, AT is the average exposure time for non-carcinogens (exposure days within whole lifetime), and RfD is the reference dose. It must be noted that the present study concerns metal health risks due to the consumption of local leaf vegetables. Thus, parameters of f_v (local vegetables/total vegetables consumption), f_a (leaf vegetable/local vegetable), and f_b (fruit vegetable/local vegetable) were introduced in the estimates of daily local leaf vegetable intake.

Statistical analyses

Statistical analyses were performed with the SPSS 13.0 software package. Charts and graphs were produced using Origin 8.1. The map of regional soil and vegetable sampling locations (Fig. 1) was constructed in ArcGis 10.0. The relation between BCF values of heavy metals in vegetables and associated soil pH was examined by Pearson correlation with the levels of significance at $P < 0.05$ and $P < 0.01$ (two-tailed) as well as PCA.

Results and discussion

Heavy metal distribution in agricultural soils and leaf vegetables in the PRD region

Total concentrations of Hg, Cd, Pb, Cr, and As in agricultural soils showed large variations in the PRD region (Table 1). In particular, the ranges of Pb, Cr, and As concentrations were 3.42–140, 3.57–117, and 0.68–105 mg kg^{-1} , respectively. The average Hg, Cd, Pb, and As concentrations of soil samples were higher than the background metal levels in Guangdong (Soil Survey Office of Guangdong Province 1993), whereas

Table 1 Concentration (mg kg^{-1} dw, mean \pm standard deviation) of heavy metals in soils and vegetables in the Pearl River Delta region, South China

| Sample type | Index | Sample number | Hg | Cd | Pb | Cr | As |
|---------------------------|---|---------------------|---------------------|-------------------|-------------------|-------------------|-------------------|
| Soil | Mean \pm SD | 92 | 0.29 \pm 0.26 | 0.17 \pm 0.13 | 42.5 \pm 25.6 | 46.7 \pm 27.7 | 20.0 \pm 20.8 |
| | Range | | 0.038–1.49 | 0.01–0.69 | 3.42–140 | 3.57–117 | 0.68–105 |
| | Background level of Guangdong ^a | | 0.09 | 0.09 | 35.9 | 56.5 | 13.5 |
| | Chinese standard for agricultural soil ^b | | \leq 0.3 | \leq 0.3 | \leq 250 | \leq 150 | \leq 40 |
| Vegetable (mean \pm SD) | Flowering Chinese cabbage | 32 | 0.0017 \pm 0.0013 | 0.060 \pm 0.060 | 0.085 \pm 0.053 | 0.16 \pm 0.11 | 0.035 \pm 0.039 |
| | Lettuce | 19 | 0.0019 \pm 0.0014 | 0.049 \pm 0.052 | 0.18 \pm 0.20 | 0.13 \pm 0.09 | 0.033 \pm 0.034 |
| | Pakchoi | 11 | 0.0014 \pm 0.001 | 0.029 \pm 0.033 | 0.11 \pm 0.12 | 0.095 \pm 0.068 | 0.051 \pm 0.067 |
| | Chinese cabbage | 13 | 0.0019 \pm 0.0017 | 0.027 \pm 0.012 | 0.11 \pm 0.12 | 0.23 \pm 0.10 | 0.047 \pm 0.061 |
| | Loose-leaf lettuce | 11 | 0.0018 \pm 0.0012 | 0.037 \pm 0.027 | 0.26 \pm 0.32 | 0.19 \pm 0.13 | 0.063 \pm 0.058 |
| Chinese leaf mustard | 6 | 0.0026 \pm 0.0019 | 0.020 \pm 0.011 | 0.055 \pm 0.041 | 0.17 \pm 0.077 | 0.033 \pm 0.023 | |

^a Metal background value in the soil of Guangdong province (Soil Survey Office of Guangdong Province 1993)

^b Environmental Quality Standards for soil (GB 15618–1995, National Environmental Protection Agency of China)

soil Cr concentration had no significant increases compared to the background Cr level in Guangdong (Table 1). Compared with the values in the Environmental Quality Standards for soil (GB 15618–1995, National Environmental Protection Agency of China), the mean soil Pb and Cr concentrations did not exceed the limits. Although the mean soil Hg, Cd, and As concentrations were below the national standard limits (Table 1), a few samples were moderately enriched with one or more than one of these heavy metals (19/92 for Hg, 11/92 for Cd, and 10/92 for As). Overall, these results suggest that Hg, Cd, Pb, and As were accumulated in agricultural soils in the PRD region.

Regarding regional distribution, Foshan and Zhongshan had relatively higher Cd, Pb, and Cr concentrations than other sampling areas (cities) in the PRD region (Fig. 2a). Among the seven cities, Jiangmen and Foshan had the highest average soil Hg concentration, whereas Huizhou had the highest average soil As concentration. The metal (Hg, Cd, and As) in some samples exceed the limit values recommended by the Environmental Quality Standards for soil (GB 15618–1995, National Environmental Protection Agency of China), while no samples contained Pb and Cr exceeding their limit values, respectively. The soil samples with excessive metals (Hg, Cd, and As) mainly came from Guangzhou (7/32), Zhongshan (4/8), and Huizhou (9/13), suggesting that more attention should be paid to Hg, Cd, and As pollution in Guangzhou, Zhongshan and Huizhou, respectively.

As compared to the soils, leaf vegetables contained less heavy metal contents with obvious variations among different species (Table 1). The ranges of vegetable Pb concentration were 0.009–0.22 mg kg^{-1} (flowering Chinese cabbage), 0.006–0.65 mg kg^{-1} (lettuce), 0.008–0.37 mg kg^{-1} (pakchoi), 0.022–0.43 mg kg^{-1} (Chinese cabbage), 0.028–0.79 mg kg^{-1} (loose-leaf lettuce), and 0.008–0.13 mg kg^{-1} (Chinese leaf mustard), respectively. Of the 92 vegetable samples, there were 10 samples (11 %) containing excessive levels of Pb concentration (0.3 mg kg^{-1}) and 27 samples (30 %) containing excessive levels of As (0.05 mg kg^{-1}) referred to the Codex Alimentarius Commission (CAC) standard values. In contrast, the Hg, Cd and Cr concentrations of all vegetable samples were below the CAC (FAO/WHO 2001) standard values (0.01, 0.2, and 0.5 mg kg^{-1} , respectively). These results demonstrate that Pb and As are the dominant metal pollutants accumulated in leaf vegetables grown in agricultural soils in the PRD region.

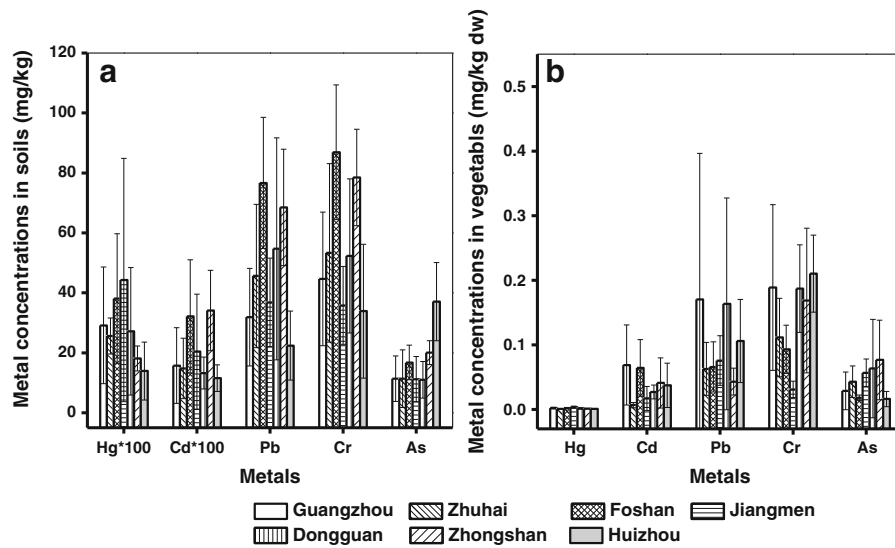


Fig. 2 Metal concentrations in soils (a) and vegetables (b) from different agricultural areas of the Pearl River Delta region, South China. The numbers of samples in Guangzhou, Zhuhai, Foshan, Jiangmen, Dongguan, Zhongshan and Huizhou were 32, 5, 5, 10, 19, 8, and 13, respectively

Regarding regional distribution, the five metals accumulated in leafy vegetables showed different trends from those in soils. The vegetable samples with excessive levels of heavy metals (10/92 for Pb and 27/92 for As) were mainly from Guangzhou (5/32 for Pb and 5/32 for As) and Dongguan (5/19 for Pb and 8/19 for As) (Fig. 2b), possibly due to the high degree of industrialization and relatively high pollutant concentrations in the atmosphere in these two cities (Wong et al. 2003). No vegetables with excessive levels of heavy metals were found in Foshan or Huizhou.

Bioaccumulation of heavy metals from soil to vegetables

The BCF values of Hg in vegetables ranged from 0.0005 to 0.0716 and averaged 0.010, and those of Pb, Cr and As in vegetables varied in similar ranges, i.e., 0.0001–0.0648, 0.0002–0.027, and 0.0001–0.103, respectively. By comparison, the BCF values of Cd in vegetables were significantly higher, varying from 0.010 to 3.10 and averaging 0.374. The BCF values of Cd were approximately 30-fold those of Hg and 50-fold those of Cr, Pb and As (Fig. 3). Since a greater BCF value indicates a higher accumulation potential of metals in vegetables (Chumbley and Unwin 1982; Cui et al. 2004), the above results indicate that Cd has higher capacity for transferring from soil to the edible parts of

vegetables than the other four heavy metals in agricultural soil of the PRD region. This can be attributed to the competition between Cd^{2+} and Ca^{2+} . It is easier for Ca^{2+} to be replaced by Cd^{2+} than other metals because of their same ionic radius and valence (Kim et al. 2002; Hart et al. 2002). In addition, Ca is an essential element for crops and it can enter plant tissues by active transport, while most heavy metals (as nonessential elements) can only enter plant tissues through passive methods (e.g., concentration diffusion and permeation) (Costa and

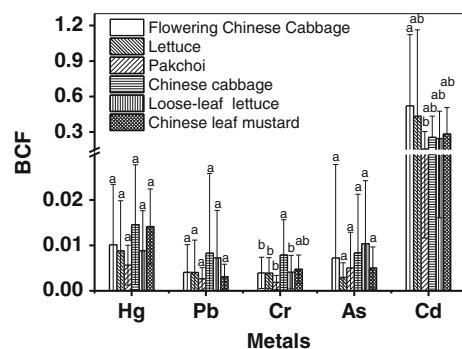


Fig. 3 Bioconcentration factor (BCF) values of five heavy metals in six kinds of leaf vegetables collected from agricultural soils in the Pearl River Delta region, South China. Average value within the same treatment that has different lowercase letters (i.e., a or b) is significantly different at $P < 0.05$, whereas average value within the same treatment that has the same lowercase letters is not significantly different at $P = 0.05$

Table 2 The bioconcentration factor values of vegetables in previous studies

| Country | Samplings site description | Hg | Cd | Pb | Cr | As | References |
|----------|----------------------------------|--------------------------|------------|-------------|------------|--------------|-----------------------------|
| Spain | Slopes of a hill | Mean ± SD 0.103±0.011 | | | | | Millán et al. (2006) |
| | Near a mine | Mean ± SD 0.036±0.003 | | | | | |
| Pakistan | Near a former mine | Mean | 0.58–2.01 | 0.352 | | 0.184 | Álvarez-Ayuso et al. (2012) |
| | Agricultural land | Range | 0.007 | | 0.065 | | Khan et al. (2010) |
| China | Wastewater-irrigated soil | Mean | 0.005–0.6 | 0.01–0.2 | 0.001–0.02 | | Jan et al. (2010) |
| | Vegetable yards | Range | 0.84–4.26 | 0.64–1.96 | 0.47–1.19 | 0.15–1.14 | Wang et al. (2004) |
| | Agricultural soil | Range | 0.045–12.3 | 0.0007–0.17 | | | Cai et al. (2012) |
| | The main crop production sources | Range | 0.010–3.10 | 0–0.065 | 0–0.027 | 0.00015–0.10 | Zhang et al. (2011) |
| | Vegetable soil | Range | | | | | This study |

Morel 1993). Differently, Cd can also enter plant tissues through Ca channels (Tester 1990).

Although there was no significant difference between different kinds of vegetables for most heavy metals, the BCF values of different heavy metals showed large variations in the six kinds of vegetable (Fig. 3). In particular, flowering Chinese cabbage had a higher BCF average value of Cd (0.521) and lower values of Hg (0.010), Pb (0.004), Cr (0.004), and As (0.007) than other kinds of vegetables. Compared with flowering Chinese cabbage and lettuce, Chinese cabbage, loose-leaf lettuce and Chinese leaf mustard had lower BCF average values of Cd and higher values of Hg, Pb, Cr and As. Among the six leaf vegetables, pakchoi had the lowest average BCF values of heavy metals (except for As). Thus, there is a relatively low potential for metal accumulation in pakchoi in the PRD region.

The obtained BCF values were compared with previously published data of heavy metal accumulation in vegetables from soils associated with agricultural land use, wastewater irrigation, and mining activity (Table 2). Despite showing great variability, the BCF values of heavy metals in vegetables from soils impacted by mining, hillslope landform, or wastewater irrigation were obviously higher than those obtained in the present study. This suggests that there is a relatively low potential for heavy metal accumulation in agricultural soils in the PRD region, possibly due to their distant locations from urban centers and industrial areas.

Leaf vegetables generally grow faster with higher transpiration rates than non-leaf vegetables (Luo et al. 2011). Thus, metal uptake by plant roots can be enhanced in leaf vegetables, resulting in the translocation of metals from roots to other vegetable tissues (Stalikas

Table 3 Matrix of principal component analysis of the bioconcentration factor values of heavy metals in vegetables

| | PC1 | PC2 | PC3 |
|--------------------------|--------|-------|--------|
| Hg | 0.833 | 0.127 | 0.221 |
| Cd | -0.300 | 0.593 | 0.621 |
| Pb | 0.298 | 0.587 | -0.638 |
| Cr | -0.266 | 0.727 | -0.061 |
| As | 0.839 | 0.108 | 0.209 |
| Eigenvalue | 1.65 | 1.25 | 0.89 |
| Variation (%) | 33.0 | 25.1 | 17.8 |
| Cumulative variation (%) | 33.0 | 58.1 | 75.9 |

Table 4 Coefficients of correlations between soil pH and bioconcentration factor values

| | Flowering Chinese cabbage | Lettuce | Pakchoi | Chinese cabbage | Loose-leaf lettuce | Chinese leaf mustard |
|---------------|---------------------------|--------------------|---------|-----------------|--------------------|----------------------|
| Sample number | 32 | 19 | 11 | 13 | 11 | 6 |
| Hg | -0.412 ^a | 0.156 | -0.374 | -0.370 | 0.448 | -0.193 |
| Cd | 0.371 ^a | 0.533 ^a | 0.222 | -0.267 | 0.307 | -0.690 |
| Pb | 0.022 | 0.156 | -0.111 | 0.086 | 0.727 ^a | -0.520 |
| Cr | 0.126 | 0.574 ^a | 0.272 | 0.142 | 0.054 | 0.437 |
| As | -0.476 ^b | -0.174 | -0.304 | 0.191 | 0.290 | -0.648 |

^a Correlation is significant at the 0.05 level (two-tailed)

^b Correlation is significant at the 0.01 level (two-tailed)

et al. 1997; Zheng et al. 2007). Due to the broad leaf area, leaf vegetables are more susceptible to pollutant accumulation by the dust from soil and rainwater. To alleviate metal pollution in local vegetables, it is suggested to plant leaf vegetables in soil with low metal content and non-leaf vegetables in soil with relatively high metal content in large agricultural areas (Kucharski et al. 1994; Luo et al. 2011). According to results of the present study, pakchoi can be planted with priority in Cd-enriched zones, followed by loose-leaf lettuce, Chinese cabbage, Chinese leaf mustard, lettuce and flowering Chinese cabbage. However, metal accumulation in vegetable tissues can be strongly influenced by multiple factors, including soil properties (e.g., soil pH, organic matter, clay content and metal concentration), plant factors (e.g., plant type and planting mode), and other environmental conditions (e.g., atmosphere and industrial pollution) (Twining et al. 2004). Therefore, caution must be taken when making specific agricultural planting plans.

Identification of heavy metal sources

PCA has commonly been used for investigating metal sources, anthropogenic activities, or soil parent materials (Facchinelli et al. 2001; Loska and Wiechula 2003; Borůvka et al. 2005; Cai et al. 2012). In the present study, three principal components (PC) were extracted from the BCF values of heavy metals in leaf vegetables (Table 3). The first principal component (PC1) explains 33.0 % of the total variance and loaded heavily on Hg (0.833) and As (0.839). Studies have shown that average Hg concentration in wastewater-irrigated areas is approximately 6.9-fold the concentration of non-wastewater-irrigated area and that Hg contamination is serious in wastewater-irrigated agricultural

soils in the PRD region (Zhu et al. 2005; Chen et al. 2005). Additionally, a study of Hg emission in Guangdong has shown that atmospheric Hg fluxes have no strong correlation with soil Hg concentration (Fu et al. 2012). Based on the above findings, we speculate that PC1 reflects the impacts of sewage irrigation.

The second principal component (PC2) accounts for 25.1 % of the total variance and is dominated by Cd (0.593), Pb (0.587), and Cr (0.727). Because the mean soil Cr concentration (46.7 mg kg⁻¹) was lower than the background level in Guangdong (56.5 mg kg⁻¹), the Cr accumulated in vegetables could mainly come from natural source(s). Based on Pb isotope analysis, Wong et al. (2002) reported that soil Pb in the PRD region mainly came from vehicle exhaust emissions. Whether the PC2 is derived from automobile exhaust emissions or natural source(s) in agricultural areas in the PRD region needs to be further investigated.

The third principal component (PC3) represents 17.8 % of the total variance and is dominated by Cd (0.621) and negatively correlated with Pb (-0.638). Due to acidic soil pH condition and active phosphorus leaching, phosphate fertilizers are commonly used in the red-soil areas in the PRD region (Zhang and Ke 2004; Wong et al. 2002). Because phosphate fertilizers contain a large amount of Cd, the application of phosphate fertilizers could result in substantial increases in the soil Cd concentration (Li et al. 2004). Therefore, we assign the PC2 to a fertilizer source.

The effect of soil pH on heavy metal accumulation in vegetables

In target agricultural areas of the PRD region, soil pH ranged from 4.51 to 7.85 and averaged 6.31 (median, 6.45). Of the 92 soil samples, 63 had pH values less

Table 5 The parameters used in the target hazard quotient calculation of heavy metals via vegetable consumption for the residents in the PRD region, South China

| Meaning | Unit | Value | | Reference |
|----------------|------------|-------------|------------------------------|--|
| | | Adults | Children | |
| EF | d/a | 365 | 365 | USEPA (2002) |
| ED | A | 60 | 10 | Bennett et al. (1999), China (2012) |
| IR | g/person/d | 290.8 | 210 | Ma et al. (2005) |
| f _v | Unitless | 0.85 | | Ma et al. (2005), Guangdong Statistics Bureau (2012) |
| f _a | Unitless | 0.7 | | Ma et al. (2005), Guangdong Statistics Bureau (2012) |
| f _b | Unitless | 0.3 | | Ma et al. (2005), Guangdong Statistics Bureau (2012) |
| BW | kg | 55.9 | 32.7 | Wang et al. (2005) |
| AT | d | 21,900 | 3,295 | USEPA (2002) |
| C | mg/kg | See Table 1 | | This study |
| θ | % | 85 | | Baes et al. (1984) |
| RfD | mg/(kg·d) | | | USEPA (2002) |
| | | | RfD(Hg)=3×10 ⁻⁴ | |
| | | | RfD(Cd)=1×10 ⁻³ | |
| | | | RfD(Pb)=3.5×10 ⁻³ | |
| | | | RfD(Cr)=3×10 ⁻³ | |
| | | | RfD(As)=3×10 ⁻⁴ | |

than 7.0. Results of frequency analysis of soil pH showed that there were 24, 25, 37 and 6 soil samples with soil pH ranges of <5.5, 5.5–6.5, 6.5–7.5 and >7.5, respectively. Integrating these results indicates that the PRD agricultural soils was slightly to severely acidic.

Results of Pearson correlation analysis showed that there were significant correlations between soil pH and BCF values of Hg, Cd and As but not Pb or Cr in flowering Chinese cabbage (Table 4). Thus, soil pH condition may play an important role in Hg, Cd and As accumulation while showing minor or no effect on Pb and Cr uptake by flowering Chinese cabbage grown in agricultural areas in the PRD region. The BCF values of Cd and Cr in lettuce and the BCF values of Pb in loose-leaf lettuce were positively correlated to soil pH values (*r*=0.533, 0.574, and 0.727, respectively; *P*< 0.05). In contrast, the BCF values of the five metals in pakchoi, Chinese cabbage and Chinese leaf mustard had no correlations with soil pH values.

Our findings regarding the positive correlation between soil pH and the BCF value of Cd in flowering Chinese cabbage as well as lettuce in agricultural area of the PRD region are inconsistent with the conclusions of Zhao et al. (2009). Moreover, Dayton et al. (2006) reported that pH had no significant effect on the accumulation of Pb from soil into vegetables, while Zeng et al. (2011) showed that there was a significantly negative correlation between soil pH values and the EDTA-extractable Cr, Cu, Fe, Mn, Pb and Zn concentrations. Overall, the effect of pH on the availability of heavy metals in soil and their accumulation behaviors in vegetables has long been controversial (Wang et al. 2006a, b; Eriksson 1989; McBride 2002), which needs further investigations.

In addition to soil pH values, finer soil particles (clays) have a greater cation exchange capacity (CEC) and hence a greater ability to retain cationic metals than sandy soils. Soil organic matter (OM) plays an important role in affecting metal bioavailability in soils as OM is a major contributor to the pH-dependent negative charge in soils which gives rise to the soils' ability to retain cationic metals (McLaughlin et al. 2011). Additionally, adsorption and complexation can also influence metal bioavailability in the soil. In the case of exogenous contamination, heavy metals are preferentially adsorbed and fixed in soil components with high specific surface areas, mainly oxides, clay minerals, and OM present in fine particles (Chen et al. 2009; McLaughlin et al. 2011).

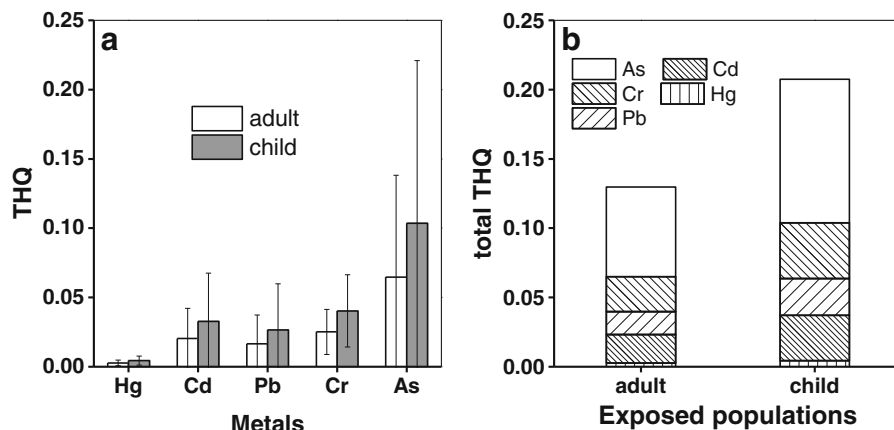
Table 6 Target hazard quotient values (mean \pm standard deviation) of heavy metals via vegetable consumption for the residents in the Pearl River Delta region, South China

| | | Hg | Cd | Pb | Cr | As | Total THQ |
|-----------------------------|-------|---------------------|---------------------|---------------------|--------------------|-------------------|-------------------|
| Flowering Chinese cabbage | Adult | 0.0026 \pm 0.0020 | 0.028 \pm 0.028 | 0.011 \pm 0.0071 | 0.025 \pm 0.017 | 0.055 \pm 0.061 | 0.122 \pm 0.060 |
| | Child | 0.0041 \pm 0.0031 | 0.045 \pm 0.045 | 0.018 \pm 0.011 | 0.041 \pm 0.027 | 0.088 \pm 0.097 | 0.195 \pm 0.096 |
| Lettuce | Adult | 0.0029 \pm 0.0021 | 0.023 \pm 0.024 | 0.023 \pm 0.027 | 0.021 \pm 0.014 | 0.051 \pm 0.052 | 0.117 \pm 0.040 |
| | Child | 0.0047 \pm 0.0034 | 0.037 \pm 0.039 | 0.038 \pm 0.043 | 0.033 \pm 0.022 | 0.082 \pm 0.084 | 0.188 \pm 0.064 |
| Pakchoi | Adult | 0.0022 \pm 0.0013 | 0.013 \pm 0.015 | 0.014 \pm 0.016 | 0.015 \pm 0.011 | 0.079 \pm 0.104 | 0.124 \pm 0.117 |
| | Child | 0.0036 \pm 0.0020 | 0.022 \pm 0.025 | 0.023 \pm 0.026 | 0.024 \pm 0.017 | 0.127 \pm 0.166 | 0.198 \pm 0.188 |
| Chinese cabbage | Adult | 0.0029 \pm 0.0027 | 0.013 \pm 0.0055 | 0.015 \pm 0.016 | 0.036 \pm 0.016 | 0.073 \pm 0.094 | 0.140 \pm 0.111 |
| | Child | 0.0047 \pm 0.0043 | 0.020 \pm 0.0088 | 0.024 \pm 0.025 | 0.058 \pm 0.026 | 0.117 \pm 0.151 | 0.224 \pm 0.177 |
| Loose-leaf lettuce | Adult | 0.0027 \pm 0.0019 | 0.017 \pm 0.012 | 0.035 \pm 0.043 | 0.029 \pm 0.020 | 0.098 \pm 0.090 | 0.167 \pm 0.086 |
| | Child | 0.0044 \pm 0.0030 | 0.028 \pm 0.020 | 0.056 \pm 0.069 | 0.047 \pm 0.032 | 0.157 \pm 0.143 | 0.268 \pm 0.138 |
| Chinese leaf mustard | Adult | 0.0040 \pm 0.0030 | 0.0093 \pm 0.0053 | 0.0073 \pm 0.0055 | 0.027 \pm 0.0012 | 0.051 \pm 0.035 | 0.098 \pm 0.039 |
| | Child | 0.0063 \pm 0.0048 | 0.0015 \pm 0.0085 | 0.012 \pm 0.0088 | 0.043 \pm 0.019 | 0.082 \pm 0.056 | 0.158 \pm 0.063 |
| Mean THQ of leaf vegetables | Adult | 0.0028 \pm 0.0021 | 0.020 \pm 0.022 | 0.017 \pm 0.021 | 0.025 \pm 0.016 | 0.065 \pm 0.073 | 0.128 \pm 0.077 |
| | Child | 0.0044 \pm 0.0033 | 0.033 \pm 0.035 | 0.027 \pm 0.033 | 0.040 \pm 0.026 | 0.104 \pm 0.117 | 0.204 \pm 0.123 |

Potential health risk associated with consumption of leaf vegetables

The potential health risks were assessed using THQ, which were calculated according to Eqs 1 and 2. The parameters used in the equations were listed in Table 5. The potential health risk to local residents associated with vegetable consumption was assessed with the THQ index (Tables 6). Of the five heavy metals, As posed the greatest health risk to adults and children in the PRD study areas, followed by Cr, Cd, Pb and Hg ($Hg < Pb < Cd < Cr < As$) (Fig. 4a). For both children and adults, the THQs of heavy metals in the

six leaf vegetables were less than 1.0 (Table 6), indicating that the residents in the PRD region were not exposed to significant health risks associated with consumption of local leaf vegetables. In addition, the health risks of heavy metal exposure to PRD residents varied in different leaf vegetables. Of the five metals, Hg and Cr had the lowest THQs in pakchoi, whereas the other metals (Cd, Pb and As) had the lowest THQ values in Chinese leaf mustard. In the six kinds of leaf vegetables, the THQs of Hg, Cd, Pb, Cr and As were highest in Chinese leaf mustard, flowering Chinese cabbage, loose-leaf lettuce, Chinese cabbage and loose-leaf lettuce, respectively (Table 6).

**Fig. 4** The target hazard quotient (THQ) (a) and total THQ (b) of five heavy metals to children and adults via vegetable consumption in the Pearl River Delta region, South China

The total THQs of health risk to children and adults in the PRD region with consumption of leaf vegetables were 0.208 and 0.130, respectively (Fig. 4b). Thus, the residents in the PRD region were exposed to minimal risks associated with consumption of leaf vegetable. Although the children's average daily intake of vegetable only accounts for 72.4 % of the adults' intake, the total THQ of heavy metals for children was approximately 1.6-fold that for adult. Thus, the children are more sensitive to heavy metal exposure through local vegetable consumption than adults in agricultural areas in the PRD region.

The THQ values of As, Cr, Cd, and Pb accounted for 50.1, 19.3, 16.0, and 12.4 % of the total THQs, respectively, while that of Hg accounted for a small portion only, ~2.17 %. These results indicate that As, Cr, Cd and Pb primarily impose health risks to the resident in the study area, while Hg has a minor contribution to human health risks. The total THQs of metals in leaf vegetables for children and adults in the PRD region (0.208 and 0.130) were far below those of market vegetables in Chongqing where even a single metal's THQ has exceeded the total THQ (e.g., THQ of Pb for adults were 0.45–0.52) (Yang et al. 2011) and Beijing (0.771 and 0.605) (Song et al. 2009).

It should be noted that the present study only estimated the health risk of heavy metals to the residents via consumption of local leaf vegetables, which accounts for 70 % of total vegetable consumption in the PRD region. That is, this work only takes into account part but not the total risks to PRD residents associated with consumption of vegetables. In addition, the total risks of heavy metals are related to the main polluted metals, and their long-term interactions which can impose distinct ecological or health risks via antagonistic, additive, and/or synergistic effects (Menzie et al. 2009; Nordberg et al. 2005) were not considered. Thus, the potential health risks of heavy metal exposure to local residents through consumption of leaf vegetables might be underestimated.

Conclusions

In the agricultural areas of the PRD region, pakchoi had the lowest capacity for metal enrichment among the six leaf vegetables under study. Of the five heavy metals, Cd was more easily transferred from soil unto leaf vegetables, with BCFs ~30-fold those of Hg and 50-fold those of Cr, Pb and As. The main metal

sources for leaf vegetables in the PRD agricultural areas might include sewage irrigation and chemical fertilization. Despite the significant correlation between BCF and soil pH values in specific samples, there is no clear relationship between these two parameters in all samples as a whole. The characteristics of heavy metal accumulation in crops and associated agricultural soil conditions need to be comprehensively considered for appropriate formulation of agricultural planting plans. Despite a certain degree of metal enrichment in soils and vegetables, the PRD residents were not experiencing significant health risk of heavy metal exposure through consumption of local leaf vegetables in agricultural areas. Still, more attentions should be paid to children due to their higher sensitivity to heavy metal exposure than that of adults.

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