# Health risk assessment of heavy metals in vegetables grown around battery production area

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ABSTRACT: Battery production is one of the main sources of heavy metals that present great harm to human health even in low concentrations. Chromium (Cr), Cadmium (Cd) and Lead (Pb) were measured in edible portions of vegetables and soils around a battery production area in China, and the potential health risk of heavy metal contamination to the local population via vegetable consumption was evaluated. Their concentrations in edible portions of vegetables were 2.354 (0.078-14.878), 0.035 (0.003-0.230) and 0.039 (0.003-0.178) mg kg$^{-1}$, respectively. Approximately 3 % of the Cd in the vegetable samples exceeded the maximum concentration allowable by national food safety criteria, although Pb content in all samples were within the criteria. Transfer factors (TF) from soils to vegetables were dependent on vegetable species. Leguminous vegetables were more likely to accumulate Cr, while leaf vegetables tended to show higher levels of concentration of Cd and Pb. Melon vegetables demonstrated a relatively low capacity for accumulating the heavy metals studied. TF were positively correlated with soil organic matter and negatively correlated with soil pH. The mean estimated daily intake of Cr, Cd and Pb via dietary consumption of vegetables was 0.011, $1.65 \times 10^{-4}$ and $1.84 \times 10^{-4}$ mg kg$^{-1}$ of body weight per day, respectively, levels that were much lower than the reference doses recommended by USEPA (U.S. Environmental Protection Agency) and JECFA (Joint FAO/WHO Expert Committee on Food Additives), indicating that the potential health risk of Cr, Cd and Pb exposure via vegetable consumption to the local population around this battery production area could be negligible.

## Introduction

Heavy metals can be very harmful to the human body even in low concentrations as there is no effective excretion mechanism (Ghosh et al., 2012). Lead (Pb) gives rise to adverse effects in several organs and systems in all known animal species, such as the blood, central nervous system, kidneys, reproductive and immune systems (NCM, 2003). Cadmium (Cd) is persistent and accumulates mainly in the kidney and liver of vertebrates, producing severe diseases in these organs (UNEP, 2006). Chromium (Cr) can cause skin ulcers and nasal septum perforations (USEPA, 1980).

While both Cr (III) and Cr (VI) can be toxic to plants and animals, Cr (III) toxicity occurs in higher concentrations, and this form is actually an essential nutrient to human and other animals. Cr (VI), on the other hand, is toxic in much lower concentrations and also tends to be more mobile and bioavailable than Cr (III) in surface and subsurface environments (Adriano, 2001). Compared with other pathways such as inhalation and dermal contact, dietary intake is the main route of exposure to heavy metals for most people (Tripathi et al., 1997; Qian et al., 2010; Yeganeh et al., 2012).

Battery production is one of the main sources of heavy metals, yet some battery production areas have not been carefully monitored. In this study, the heavy metal pollution around Changxing County, a famous lead-acid battery production base located in the north of Zhejiang Province, China, was investigated. Started

in 1970s, lead-acid battery production brought about serious environmental problems, especially heavy metal pollution. Extensive studies have focused on the assessment of potential health risks associated with exposure to heavy metals via vegetable consumption for residents in the vicinity of mining and smelting sites (Li et al., 2006; Sipter et al., 2008; Cao et al., 2010; Liu et al., 2010; Dong et al., 2011; Wang et al., 2011; Xiao et al., 2011; ).

Some studies have been carried out to assess the health risk of heavy metals through consuming vegetables from sewage-irrigated areas (Avci, 2012; Ghosh et al., 2012; Wang et al., 2012; Weldegebriel et al., 2012; Xue et al., 2012). To our knowledge. few studies have been carried out that have analyzed heavy metals in agro-products and evaluated potential health risks to inhabitants around battery production areas . Therefore, the main objective of this work is to determine the contamination levels of Cr, Cd and Pb in selected vegetables and to evaluate the health risk to the local population via consumption of vegetables.

## Materials and Methods

### Study area

Changxing County (30º43'-30º11' N, 119º33'-120º06' E) is located in the north of Zhejiang Province, China. The samples were collected from four locations including Meishan Town (MS), the suburb of the County's urban area (SC), Hongqiao Town (HQ) and Lvshan Town (LS), respectively (see Figure 1). A large industrial

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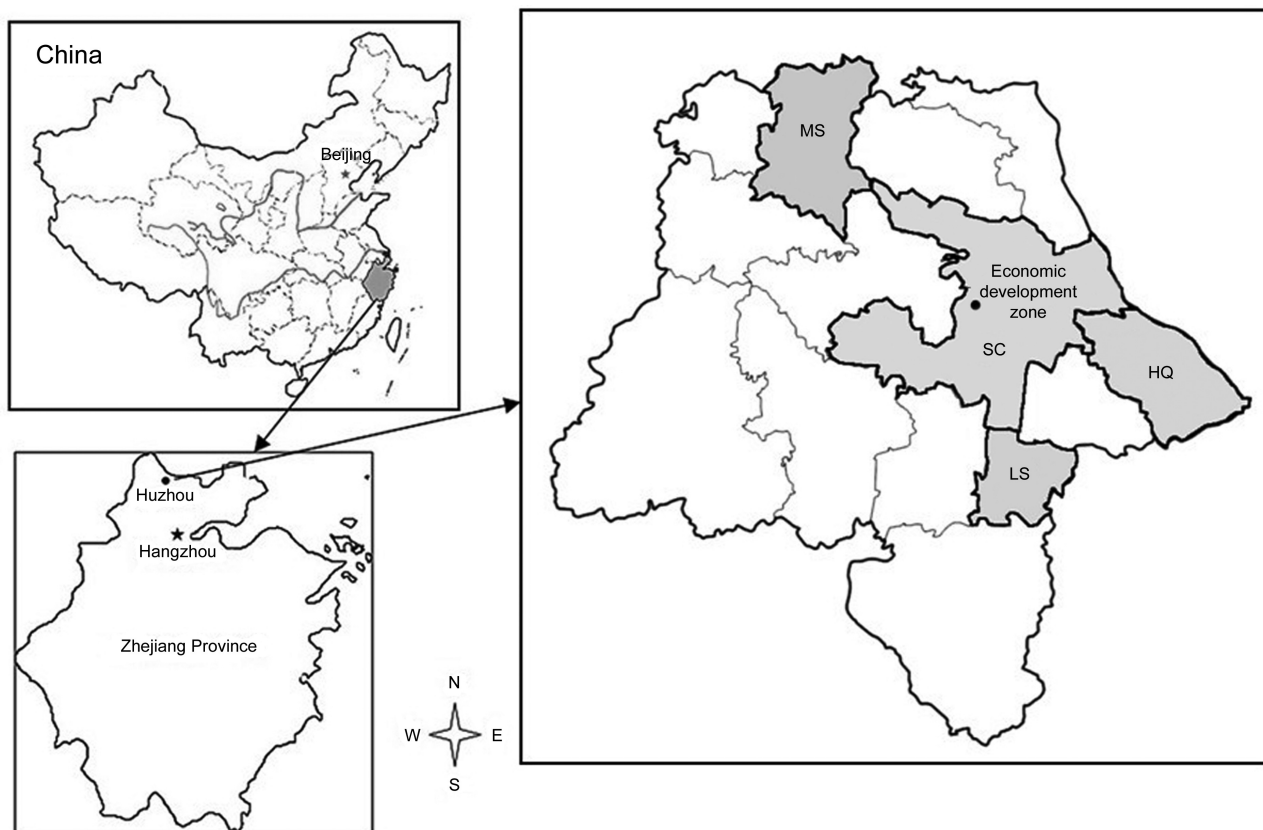Chen et al.                                                    Health risk assessment of heavy metals



Figure 1 – Illustration of sampling sites in Changxing County, Zhejiang, China. MS: Meishan Town; SC: the suburb of the County's urban area; HQ&LS: Hongqiao Town and Lvshan Town.

park consisting mainly of battery productions and re-fractory manufacturers was located in MS, representing areas seriously impacted by Pb and Cr. SC was near a residential area and the economic development zone of Changxing, mainly polluted by domestic waste and small-scale industries. On the other hand, HQ and LS (called "HQ & LS" henceforward) were both character-ized as having "pollution-free vegetable production dem-onstration base", representing areas with relatively less heavy metal contamination.

**Collection and preparation of samples**

From Nov. 2011 to May 2012, eleven vegetable species, *Brassica chinensis L.* (green vegetable), *Brassica campestris L.* (Chinese cabbage), *Spinacia oleracea* (spin-ach), *Allium tuberosum Rottler* (garlic chives), *Brassica oleracea L.var.capitata L.* (cabbage), *Capsicum annuum* (pepper), *Solanum lycopersicum* (tomato), *Solanum mel-ongena L.* (eggplant), *Cucumis sativus Linn* (cucumber), *Pisum sativum Linn* (garden pea), *Vicia faba Linn* (horse-bean) were randomly collected. They were considered as representative of vegetable species commonly con-sumed in the area under study.

A total of 96 vegetable samples were gathered from three locations. The number of vegetable samples

for each kind ranged from four to nine, depending on the amount of vegetables planted in the study area. Soil and vegetable samples were paired collected. Vegeta-bles' rooted soil samples were taken at 0-20 cm in depth. A composite sample was composed of five subsamples at each sampling site for both vegetables and soils. All vegetables and soil samples were individually sealed into polyethylene bags to avoid cross-contamination and transferred to the laboratory on the same day of sam-pling (CMEP, 2004).

Only the edible part of each vegetable sample was used for analysis. The vegetable samples were washed with ultrapure water three times. After the water had evaporated, the vegetable samples were weighed, oven-dried at 65 °C for 48 h, weighed again and then crushed into powder. The heavy metal concentrations in edible portions of vegetables were determined on a wet weight basis.

All soil samples were naturally air-dried until con-stant weight was reached. The dried soil samples were homogenized with pestle in a mortar, and then passed through standard sieves No.20 (Φ 0.9 mm), No.60 (Φ 0.3 mm), No.100 (Φ 0.15 mm) for analysis of pH, organic matter (OM) and heavy metal contents, respectively. Soil pH was measured using a pH electrode and the ratio

of solid: water was 1:2.5 (SFA, 1987). OM contents of soil samples were determined using the loss on ignition method (Boyle, 2004; Heiri et al., 2001; Dean, 1974).

### Analysis

For digestion of soils, a 0.2 g dried sample was digested with 9 mL concentrated $HNO_3$ in a high borosilicate glass (HBG) vessel. The heating conditions were: 90 °C for 30 min, 110 °C for 30 min, 125 °C for 1 h. After cooling, 1 mL $HClO_4$ was added, and the mixture was again subjected to heating at 110 °C for 30 min and at 130 °C for 1 h (Huang et al., 2007; USEPA, 1996). This method was not a total digestion technique. However, it was a very strong acid digestion that would dissolve almost all elements that were environmentally available.

Solutions were diluted to 25 mL with ultrapure water and transferred to a bottle after filtration. For vegetables, a 0.5 g dried sample was digested in $HNO_3$-$HClO_4$ mixture (9: 1) and the heating conditions were: 60 °C for 2 h, 100 °C for 2 h, 125 °C for 1 h, and 175 °C for 1 h (Ghosh et al., 2012). Total Cr, Cd and Pb contents in the vegetable and soil samples were analyzed by inductively coupled-mass spectrometry (ICP-MS). The limits of detection (LOD) for vegetables and soils were as follows: 0.137 µg $L^{-1}$ for Cr, 0.024 µg $L^{-1}$ for Cd, and 0.350 µg $L^{-1}$ for Pb. The coefficient of variation for triplicate analysis was ≤ 10 %. For quality control, certified reference material (GBW 10043) was used. The recovery rates ranged from 98 % to 102 %.

### Statistical analysis

Statistical analysis of data was carried out using Microsoft Office Excel 2010 and the SPSS 16.0 statistical package program. The Pearson correlation test was used to check for correlations between transfer factor (TF) values of heavy metals and soil properties (pH, organic matter). Calculations of the distribution of dietary exposure to heavy metals via consumption of vegetables for the local population were performed using the commercially available software package @risk (Version 5.0,

for excel professional edition, Palisade). The estimated daily intake dose (EDI) was calculated as follows:

$$EDI = \frac{C_i \times IR}{BW} \tag{1}$$

where $C_i$ (mg $kg^{-1}$) is the concentration of heavy metals in edible portions of vegetables ($i$ = Cr, Cd or Pb), $IR$ (g per day) the average daily consumption of vegetables, and $BW$ (kg) the body weight.

## Results and Discussion

All the analyzed vegetable samples contained detectable concentrations of total Cr, Cd and Pb (Table 1). Mean concentrations (and range) of total Cr, Cd and Pb in edible portions of vegetables were 2.354 (0.078-14.878), 0.035 (0.003-0.230), and 0.039 (0.003-0.178) mg $kg^{-1}$, respectively. The average concentrations of heavy metals in all vegetable samples were in the order of Cr > Pb > Cd. The coefficients of variation for the three heavy metals in vegetables also followed the same order, suggesting that Cr is more susceptible to the interference of vegetable species, pollution and other external factors.

According to Chinese food safety criteria, the maximum allowable concentrations (MAC) of Cd were 0.2 mg $kg^{-1}$ for leaf vegetables and leguminous vegetables and 0.05 mg $kg^{-1}$ for melon vegetables and solanaceous vegetables (CMH, 2005). About 3 % of the samples were above the MAC for Cd and the samples with elevated Cd levels were all found in MS. Additionally, the Pb concentrations observed in vegetable samples were all within the permissible limit.

The total Cr levels in edible portions of vegetables were much higher than those found in presumably less contaminated areas, such as Jiangsu, China (0.67 mg $kg^{-1}$) (Cao et al., 2010) and in Brazil (0.01-0.6 mg $kg^{-1}$) (Guerra et al., 2012). Higher concentrations of total Cr in this region may be expected due to the mass production of refractory materials in Changxing County, especially in MS, where there are many refractory factories. For the concentrations of Cd in edible portions of vegetables,

Table 1 – Concentrations of heavy metals in edible portions of vegetables.

| Sampling areas | Sample number | Cr | | Cd | | Pb | |
|---|---|---|---|---|---|---|---|
| | | Mean ± SD | Range | Mean ± SD | Range | Mean ± SD | Range |
| | | mg $kg^{-1}$ | | | | | |
| MS | 31 | 1.645 ± 1.902 | 0.213 - 6.992 | 0.059 ± 0.060 | 0.006 - 0.230 | 0.047 ± 0.049 | 0.005 - 0.178 |
| SC | 30 | 2.871 ± 4.239 | 0.078 - 14.868 | 0.030 ± 0.045 | 0.003 - 0.195 | 0.040 ± 0.041 | 0.005 - 0.155 |
| HQ&LS | 35 | 2.505 ± 4.244 | 0.177 - 14.878 | 0.020 ± 0.016 | 0.005 - 0.061 | 0.031 ± 0.028 | 0.003 - 0.104 |
| Standards | | Cr | | Cd | | Pb | |
| | | mg $kg^{-1}$ | | | | | |
| WHO/EU, 1983 | | 0.1 | | 0.01 | | 5.0 | |
| FAO/WHO, 2001 | | - | | 0.02-0.2 | | 0.5-1.0 | |
| CMH, 2005 | | 0.5-1.0 | | 0.05-0.2 | | 0.1-0.3 | |

MS: Meishan Town; SC: the suburb of the County's urban area; HQ&LS: Hongqiao Town and Lvshan Town; WHO: World Health Organization; EU: European Union; FAO: Food and Agriculture Organization; CMH: Chinese Ministry of Health; SD: Standard Deviation.

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the mean value of the three locations was 0.035 mg kg$^{-1}$, which was in the same range as the results from Jiangsu, China (Cao et al., 2010) and Gejiu City, Yunnan Province, China (Xiao et al., 2011) and lower than the findings from Gaziantep, Turkey irrigated with municipal and industrial wastewaters (Avci, 2012).

Concentrations of Cd in vegetables grown in uncontaminated soils generally varied from 0.005 to 0.5 mg kg$^{-1}$, with levels being somewhat higher in leafy vegetables and in belowground tissues than in other plant tissues (Avci, 2012). The range of Pb concentrations (0.003-0.178 mg kg$^{-1}$) in edible portions of vegetables was comparable to the values found in Jiangsu, China (Cao et al., 2010). Plants grown on polluted sites could have a considerably high level of Pb. Weldegebriel et al. (2012) reported Pb levels in vegetables varying from 0.11 to 0.89 mg kg$^{-1}$, which was almost ten times higher than those in the present study.

The average total Cr concentrations in edible portions of vegetables of the three sampling locations were in the order SC > MS > HQ&LS, while the average concentrations of Cd and Pb in edible portions of vegetables were in the following order: MS > SC > HQ&LS. The differences in heavy metal concentrations in edible portions of vegetables from the three sampling locations were tested by One-Way ANOVA analysis ($p$ = 0.05). There was no difference among the three locations for total Cr and Pb ($p$ > 0.05) though the concentrations of Cd in edible portions of vegetables from MS were higher than those from HQ&LS ($p$ < 0.05). Meanwhile, no differences in Cd values were observed between MS and SC in this study ($p$ > 0.05).

The maximum concentration of total Cr observed in horsebean surpassed the MAC by about 11 times, showing that the vegetables in the study area were severely contaminated by Cr (Figure 2). The concentrations of total Cr in horsebean and garden pea were higher than those in the other nine species of vegetables ($p$ < 0.05). The mean total Cr values (mg kg$^{-1}$) in the vegetables were in the following order: horsebean (10.644) > garden pea (7.509) > eggplant (1.315) > garlic chives (1.045) > pepper (1.044) > cabbage (0.951) > tomato
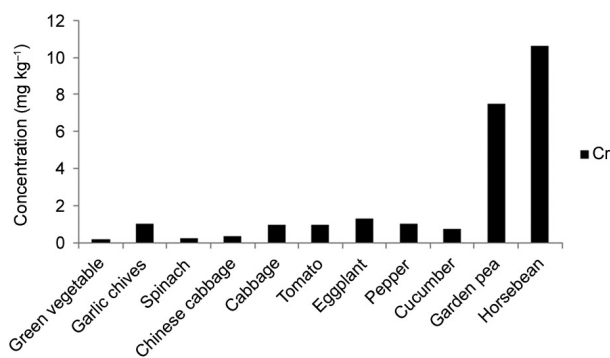
(0.947) > cucumber (0.740) > Chinese cabbage (0.327) > spinach (0.243) > green vegetable (0.190). However, unlike Cr, the maximum Cd and Pb levels were both found in spinach (Figure 3).

The average Cd content in spinach was 1.84 time the MAC. Cd values (mg kg$^{-1}$) in the vegetables were in the following order: spinach (0.092) > green vegetable (0.073) > garlic chives (0.055) > cabbage (0.052) > Chinese cabbage (0.035) > eggplant (0.033) > garden pea (0.023) > horsebean (0.022) > pepper (0.014) > tomato (0.012) > cucumber (0.005), while Pb values (mg kg$^{-1}$) were in the following order: spinach (0.113) > green vegetable (0.106) > Chinese cabbage (0.059) > garlic chives (0.054) > garden pea (0.054) > horsebean (0.041) > cabbage (0.025) > eggplant (0.012) > tomato (0.009) > pepper (0.007) > cucumber (0.006).

In general, the level of acceptance of Pb in vegetables was more consistent with those of Cd than Cr. Leguminous vegetables were more likely to accumulate Cr, while leaf vegetables tended to show higher concentrations of both Cd and Pb. Melon vegetables displayed a relatively tendency to accumulate these three heavy metals. This was similar to the finding that more metals accumulated in leaf vegetables, namely, Cd and Pb (Ghosh et al., 2012).

The trends of the total heavy metals in soils were in the following descending order: Cr > Pb > Cd (Table 2). The mean total Cr value in soils from HQ&LS was a little higher than those from SC and MS. In only one of the 96 soil samples was Cr found where concentration exceeded the maximum permissible level of 150 mg kg$^{-1}$ set by Chinese Environmental Quality for Soils (NEPAC, 1995). The contents of Cd varied in a range of 0.52-1.61, 0.18-1.41 and 0.23-0.57 mg kg$^{-1}$ for MS, SC and HQ&LS, respectively, with 68 % of the samples in the three locations higher than the threshold value (when pH ≤ 7.5, 0.3 mg kg$^{-1}$ for Cd; pH > 7.5, 0.6 mg kg$^{-1}$ for Cd). The mean (and range) concentrations of Pb in soil samples were 71.63 (39.20-340.54) mg kg$^{-1}$ in MS, 31.74 (17.74-87.82) mg kg$^{-1}$ in SC, 29.03 (19.72-67.19) mg kg$^{-1}$ in HQ&LS, respectively. The concentrations of Pb in all the soil samples were below the permissible limit.



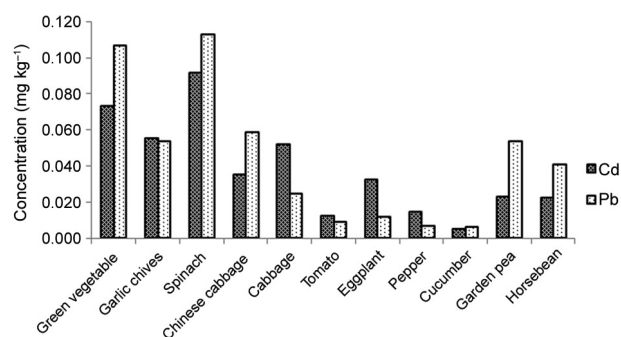Figure 2 – Concentrations of Cr in edible portions of vegetables.



Figure 3 – Concentrations of Cd and Pb in edible portions of vegetables.

The transfer of heavy metals from soils to vegetables was dependent on the vegetable species (Figures 4, 5 and 6). The highest average TF values of Cr, Cd and Pb were observed in horsebean, green vegetables and green vegetables, respectively. Cr had the highest TF values followed by Cd and then Pb, suggesting that Cr was the most bio-available. Heavy metals in soils can be

absorbed and utilized by plants in two forms: (i) dissolving in soil solution, and (ii) exchangeable in organic and inorganic components (Aydinalp and Marinova, 2003). Furthermore, plant uptake of heavy metals depends mainly on the mobility and availability of heavy metals in soils. Among soil properties, soil pH had the greatest impact on the desorption and bioavailability of heavy metals, because of its strong effects on solubility and speciation of heavy metals both in the soil as a whole and particularly in the soil solution (Müehlbachová et al., 2005).

Apart from soil pH, OM was also one of the most important properties affecting heavy metal availability in soils for retaining heavy metals in an exchangeable form (Zeng et al., 2011). Therefore, the effect of soil pH and OM on the uptake of heavy metals by vegetables was considered in this study (Table 3); TF values for three heavy metals were all negatively correlated with soil pH and positively correlated with soil OM. The mobility and uptake of heavy metals to vegetables increased with decreasing soil pH and increasing OM. A negative correlation between heavy metal mobility and availability and soil pH has been reported in numerous studies (Sukreeyapongse et al., 2002; Bang and Hesterberg, 2004; Wang et al., 2006; Ghosh et al., 2012). It has also been reported that heavy metal adsorption declined with decreasing OM in soils (Almas and Singh, 2001; Xue et al., 2012).
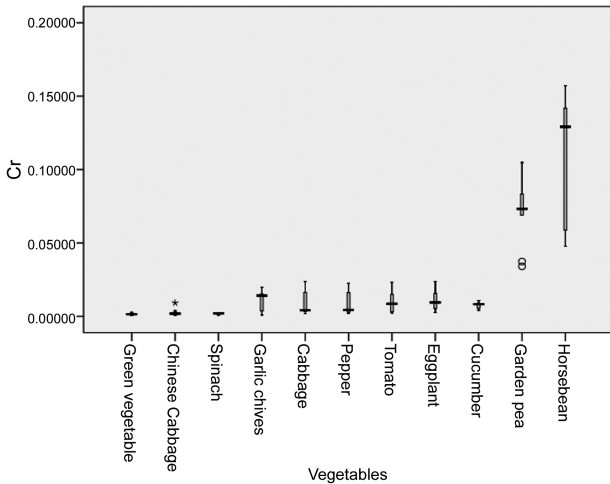


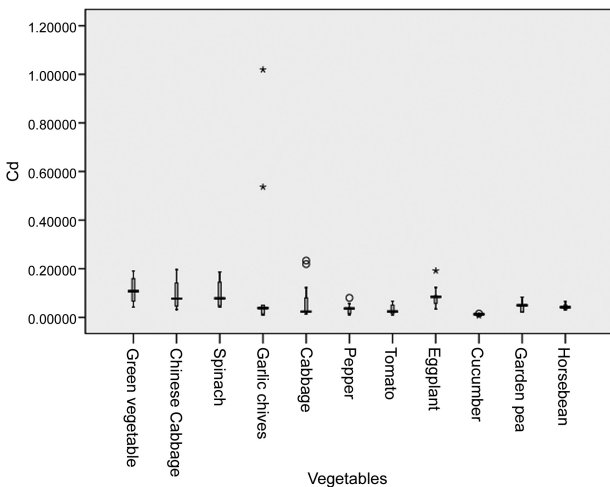Figure 4 – The transfer factors (TF) values of Cr in vegetables.



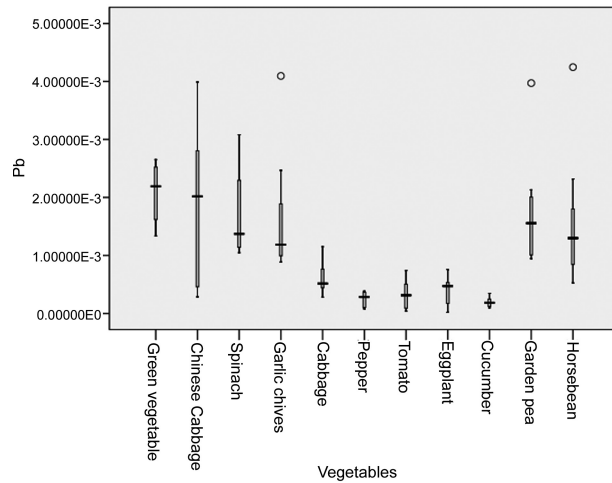Figure 5 – The TF values of Cd in vegetables.



Figure 6 – The TF values of Pb in vegetables.

Table 2 – Chemical and physical attributes of soil.

|  | MS | | SC | | HQ&LS | |
|---|---|---|---|---|---|---|
|  | Mean ± SD | Range | Mean ± SD | Range | Mean ± SD | Range |
| Cr (mg kg⁻¹) | 104.12 ± 17.90 | 76.95 - 142.32 | 109.41 ± 17.56 | 78.73 - 156.92 | 118.96 ± 16.03 | 96.73 - 143.85 |
| Cd (mg kg⁻¹) | 0.89 ± 0.27 | 0.52 - 1.61 | 0.40 ± 0.27 | 0.18 - 1.41 | 0.37 ± 0.10 | 0.23 - 0.57 |
| Pb (mg kg⁻¹) | 71.63 ± 55.21 | 39.20 - 340.54 | 31.74 ± 19.80 | 17.42 - 87.82 | 29.03 ± 7.49 | 19.72 - 67.19 |
| pH | 6.06 ± 1.17 | 4.61 - 8.34 | 6.85 ± 1.06 | 5.11 - 9.62 | 6.15 ± 1.09 | 4.27 - 8.13 |
| OM (%) | 5.11 ± 1.23 | 2.44 - 7.52 | 6.39 ± 1.68 | 4.12 - 11.22 | 7.14 ± 0.78 | 5.96 - 9.74 |

MS: Meishan Town; SC: the suburb of the County's urban area; HQ&LS: Hongqiao Town and Lvshan Town; OM: Organic Matter; SD: Standard Deviation.

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Table 3 – Correlations between TF values of heavy metals and soil pH, OM values.

|     | TF for Cr | TF for Cd | TF for Pb |
| --- | --- | --- | --- |
| pH  | -0.101 | -0.097 | -0.185 |
| OM  | 0.321  | 0.072  | 0.157  |

TF: Transfer factor; OM: Organic Matter.

According to the Agro-geology Environment Survey in Zhejiang, the soil acidification trend and the decrease in soil organic matter in the northern part of Zhejiang were bound to increase the accumulation of heavy metals in agro-products (Pan et al., 2007). Therefore, much attention should be paid to the excessive accumulation of heavy metals in agro-products caused by soil environment change (Pan et al., 2007).

Based on the Chinese nutrition and health survey (Wang, 2002), the average daily consumption of vegetables was 276.2 g per day for adults, and their average body weight was 58.7 kg. The average concentrations of total Cr, Cd, and Pb in edible portions of vegetables in this study were 2.354, 0.035, 0.039 mg kg$^{-1}$, respectively. Then the EDI values of Cr, Cd and Pb were calculated to be 0.011, $1.65 \times 10^{-4}$ and $1.84 \times 10^{-4}$ mg kg$^{-1}$ per day, respectively. The daily intake of heavy metals (Pb, Cd and Cr) depends on both the concentration of heavy metals in edible portions of vegetables and the associated consumption data. Additionally, body weight can influence the tolerance of pollutants.

The reference dose (RfD) for Cr and Cd were set by the USEPA (USEPA, 1998) at 1.5 and $1 \times 10^{-3}$ mg kg$^{-1}$ per day, respectively and the RfD for Pb was set by JECFA (JECFA, 2003) at $3.6 \times 10^{-3}$ mg kg$^{-1}$ per day. The EDIs of Cr, Cd and Pb were all far below the RfDs, suggesting that the consumption of vegetables posed negligible health risk to the local population. However, vegetables only accounted for a part of the human diet. Other food, such as grain, seafood and livestock, might contain higher levels of heavy metals, which could significantly increase the risk of ingestion of heavy metals by the local population. Moreover, considerable attention should also be paid to the potential health risk of heavy metals via other exposure pathways.

## Conclusions

Pb levels in all samples were within national food safety criteria, while Cd concentration in 3 % of vegetable samples exceeded MAC according to the criteria. The transfer of heavy metals from soils to vegetables was dependent on vegetable species. Leguminous vegetables were more likely to accumulate Cr, while leaf vegetables tended to demonstrate higher concentrations of Cd and Pb. Melon vegetables had a relatively low tendency to accumulate these three heavy metals. Transfer factors from soils to vegetables were positively correlated with soil OM and negatively correlated with soil pH. Thus, the potential health risks posed by Cr, Cd and Pb were

considered negligible based on the results of EDI, indicating that it remained safe for the local population in Changxing County to consume vegetables.

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