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Assessing the health risk of heavy metals in vegetables to the general population in Beijing, China

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Abstract

A systematic survey of As, Cd, Cr, Cu, Ni, Pb and Zn concentrations in vegetables from 416 samples (involving 100 varieties) in Beijing was carried out for assessing the potential health risk to local inhabitants. The results indicated that the metal concentrations in vegetables ranged from < 0.001 to 0.479 μg/g fresh weight (fw) (As), < 0.001 to 0.101 μg/g fw (Cd), < 0.001 to 1.04 μg/g fw (Cr), 0.024 to 8.25 μg/g fw (Cu), 0.001 to 1.689 μg/g fw (Ni), < 0.001 to 0.655 μg/g fw (Pb) and 0.01 to 25.6 μg/g fw (Zn), with average concentrations of 0.013, 0.010, 0.023, 0.51, 0.053, 0.046 and 2.55 μg/g fw, respectively. The results showed that the concentrations of As, Cr, Cu, Cd, Pb and Ni in vegetables from open-fields were all significantly higher than those grown in greenhouses. In addition, in local-produced vegetables, all HMs except Zn were significantly higher than those in provincial vegetables. The estimated daily intake (EDI) of As, Cd, Cr, Cu, Ni, Pb and Zn from vegetables was 0.080, 0.062, 0.142, 3.14, 0.327, 0.283 and 15.7 μg/(kg body weight (bw)·d) for adults, respectively. Arsenic was the major risk contributor for inhabitants since the target hazard quotient based on the weighted average concentration (THQ_w) of arsenic amounted to 44.3% of the total THQ (TTHQ) value according to average vegetable consumption. The TTHQ was lower than 1 for all age groups, indicating that it was still safe for the general population of Beijing to consume vegetables.

Key words: heavy metals; health risk; estimated daily intake; Beijing; vegetable

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Introduction

Heavy metals (HMs) are known to cause deleterious effects on human health (Davydova, 2005). Excessive exposure to HMs has been shown to cause various diseases (Jarup, 2003). For most people, the main route of exposure to toxic elements is through dietary intake (Calderon *et al*., 2003; Roychowdhury *et al*., 2003). Vegetable is an important part of the human diet in China (Zheng *et al*., 2007). It is believed that vegetables can become contaminated with HMs if they grow on soils contaminated by mining, vehicular exhaust, industrial activities and other agriculture activities (Cui *et al*., 2004; Li *et al*., 2006a). The levels of HMs in vegetables and soils and their risk to people are of great public concern (Li *et al*., 2006b; Wang *et al*., 2008, 2005; Xie *et al*., 2006).

The content of HMs in soil and food has been well studied, but the attention paid mainly to mining sites or contaminated fields (Cui *et al*., 2004; Kachenko and Singh, 2006; Li *et al*., 2006b). A comprehensive regional survey of HMs in food and an assessment of their risk to the general population are lacking.

In the present study, the concentrations of As, Cr, Cd, Cu, Pb, Ni and Zn in vegetables growing in greenhouses or open-fields at different geographical regions were determined. The daily intake of these metals from vegetable consumption for children, adult and seniors was estimated. The potential risk of HMs from vegetables to human health was also evaluated with the target hazard quotient (THQ).

1 Materials and methods

1.1 Sampling method

A total of 416 fresh vegetable samples were collected. The samples involve 100 varieties, in which, 19 of them are commonly consumed by inhabitants in Beijing and account for approximately 80.8% of the total vegetable consumption (Table 1).

Among samples, 47.6% directly collected from field stalls, 13.5% from wholesaler outlets, and others from supermarkets and farmer markets. At each sampling site, 3–5 samples of each variety were collected so that they could be combined into one sample for elemental analysis

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Table 1 Percentage of typical vegetable consumption in Beijing (BMBS, 2002)

Vegetable species	Consumption (%)
Chinese cabbage (*Brassica rapa* var. *pekinensis*)	22.6
Tomato (*Lycopersicon esculentum*)	7.1
Cucumber (*Cucumis sativus*)	6.8
Radish (*Raphanus*)	6.1
Kidney bean (*Phaseolus vulgaris* Linn)	5.3
Cabbage (*Brassica oleracea* var. *capitata*)	5.1
Eggplant (*Solanum*)	4.4
Celery (*Apium graveolens*)	3.7
Scallion (*Allium fistulosum*)	2.8
Wax gourd (*Beninacasa hispida*)	2.7
Leek (*Allium tuberosum*)	2.4
Cauliflower (*Brassica oleracea* var. *acephala*)	2.0
Capsicum (*Capsicum annuum*)	1.9
Potato (*Solanum tuberosum*)	1.7
Spinach (*Spinacia oleracea*)	1.7
Garlic (*Allium sativum*)	1.4
Rape (*Brassica campestris*)	1.4
Bok Choy (*Brassica rapa* var. *chinensis*)	1.2
Lotus root (*Nelumbo adans*)	0.5
Other vegetables	19.1

(Chen *et al*., 2006). Based on the cropping patterns, samples were classified into two groups as open-field (56.7%) vegetables and greenhouse vegetables (43.3%); based on the producing area, samples were classified as locally-produced vegetables (79.3%) which cultured inside Beijing and non-locally produced vegetables (20.7%) which produced in other provinces, mainly from Shandong Province, Inner Mongolia, Guangdong Province, Tianjin and Hebei Province; and based on the edible part of the plant, samples were classified as leafy vegetables (44.0%), gourd and fruit vegetables (36.5%), and stem and root vegetables (19.5%).

1.2 Chemical analysis

The vegetable samples were cut into small pieces using a stainless steel knife after thoroughly washing with tap water and deionization water. After drying at 60°C for 3 d, and samples were ground and sieved for acid digestion. About 0.5000 g of dried sample was digested at (100 ± 5)°C with the mixture of 8.0 mL concentrated nitric acid, 2.0 mL perchloric acid and 0.5 mL concentrated sulfuric acid.

The Cu and Zn concentrations were measured using a flame atomic absorption spectrometer (AAS, Vario 6, Jena Co., Ltd., Germany). The concentrations of Cd, Cr, Pb and Ni were measured using graphite furnace atomic absorption spectrometry (GFAAS, Vario 6, Jena Co., Ltd., Germany). Arsenic (As) was measured using hydride generation atomic fluorescence spectroscopy (AFS-2202, Haiguang Instrumental Co., China) according to the method described by Yang *et al*. (1998). The standard reference material, poplar leaves (GBW07604), was obtained from the China National Center for Standard Reference Materials and was digested along with the samples for quality assurance and quality control. The recovery for Cu, Zn, Ni, Pb, Cd, Cr and As was 94.6%–105.4%, 97.3%–102.7%, 89.5%–110.5%, 86.7%–113.3%, 84.4%–115.6%, 90.9%–109.1% and 83%–113%, respectively.

1.3 Data analysis

Statistical analysis was performed using the SPSS 13.0 program. The vegetable data was not normally distributed and thus was log transformed or Box-Cox transformed prior to statistical analysis. The tests for normality of the raw and transformed data were performed prior to ANOVA analysis and a probability level of $P < 0.05$ was considered statistically significant.

1.4 Estimated daily intake of heavy metals from vegetables

Estimated daily intake (EDI) (μg/(kg body weight (bw)·d)) of HMs from vegetable consumption was obtained by Eq. (1):

$$\mathrm{EDI} = \frac{C \times F_{\mathrm{IR}}}{W_{\mathrm{AB}}} \tag{1}$$

where, C (μg/g fw) is an average weighted HM content in the edible portion of the vegetable, and is calculated by multiplying the normalization-transformed mean HM content in vegetables with their corresponding percentage of consumption. F_{IR} (g/(d·person)) is a daily vegetable consumption. As reported in literature, the average F_{IR} for all inhabitants, and for adults, children and seniors in Beijing is 335, 394, 257 and 323 g/(d·person), respectively (Ge, 1996; Ge and Zhai, 1999, 2004; Pang *et al*., 2005). W_{AB} is the average body weight (63.9 kg for adult, 32.7 kg for children, and 60.9 kg for senior) (Ge, 1996; Ge and Zhai, 1999, 2004).

1.5 Health risk assessment

The health risks associated with HMs ingested through vegetable consumption were assessed using the target hazard quotient (THQ) (Liu *et al*., 2006; USEPA, 1996, 2000; Wang *et al*., 2005).

The THQ index can be defined as the ratio of determined dose of a pollutant to the reference dose (RfD) (μg/(kg bw·d)) (Eq. (2)):

$$\mathrm{THQ} = \frac{\mathrm{EFr} \times \mathrm{ED} \times F_{\mathrm{IR}} \times C}{\mathrm{RfD} \times W_{\mathrm{AB}} \times \mathrm{ATn}} \times 10^{-3} \tag{2}$$

where, EFr is the exposure frequency (350 d/year), ED is the exposure duration (30 years for adults, 60 years for seniors, and 6 years for children), ATn is the averaged exposure time for noncarcinogens (365 d/year for ED), and 10^{-3} is the unit conversion factor. C can be defined as C_{c}, a percentile concentration of element in vegetables from 416 samples, or C_{w}, the weighted average concentration. In this study, the non-carcinogenic risk derived from C_{c} and C_{w} is defined as $\mathrm{THQ}_{\mathrm{c}}$ and $\mathrm{THQ}_{\mathrm{w}}$, respectively. THQ is a highly conservative and relative index (Wang *et al*., 2005). If THQ is less than 1, there is no obvious risk from the substance over a lifetime of exposure, while if THQ is higher than 1, the toxicant may produce an adverse effect. The higher the THQ value, the higher the probability of experiencing long term carcinogenic effects.

2 Results and discussion

2.1 Statistical analysis

In order to perform statistical analyses, it is necessary to check the probability features of the variables at first, because the model selection for variance analysis (eg., ANOVA) and the mean value type determination used to characterize a data set are both sensitive to the non-normality of the data sets (Zhang, 2006). Table 2 shows the shape parameters and results of the Kolmogorov-Smirnov (K-S) test for raw data, log-transformed data (As, Cr, Cu and Ni) and Box-Cox transformed data (Pb, Zn and Cd) (Box and Cox, 1964). The results showed that As, Cr, Cu and Ni followed a lognormal distribution and Pb, Zn and Cd followed neither a normal distribution nor a lognormal distribution, but a Box-Cox normal distribution. The raw data sets were all positively skewed and high skewness and kurtosis values were observed for As, Cr, Cu and Ni.

The statistical analysis of the raw data and the normal distribution transformed data are shown in Table 3. There were 7, 6, 14 and 10 samples below the detection limit of 0.001 μg/g for As, Cr, Cd and Pb, respectively. According to the GB2762-2005, GB 15199-94 and GB 13106-91, the Pb, As, Ni, Cr, and Cd concentration in 7.3%, 12.6%, 2.62%, 0.96% and 0.58% of vegetable samples, respectively, exceeded the maximum level (ML) of the tolerance limit of contaminants in foods from China, and only the Cu and Zn concentrations were within the limits (Table 4).

The ML of contaminants set by the Commission of the European Communities (CEC) and the Codex Alimentarius Commission (CAC) is used as guidance by many countries. In this study, two eggplant samples exceeded the ML for Cd set by the CEC and CA (FAO/WHO, 2001). And a total of 9.4% of the vegetable samples exceeded the ML for Pb set by the CEC and CAC (FAO/WHO, 2001), which including 16 stem and root vegetables, 9 leafy vegetables and 17 legume and pulse vegetable samples.

Vegetables vary in their ability to accumulate HMs. Variance analysis showed that both the Zn and Cd content in leafy vegetables were significantly higher than that in gourd and fruit vegetables ($P < 0.05$), while on average, the Cu content in stem and root vegetables and gourd and fruit vegetables were significantly higher than that in leafy vegetables (Table 3). The normalization-transformed average content of As in stem and root vegetables were significantly higher than that in leafy vegetables ($P = 0.013 < 0.05$), while there was no significant difference observed for Cr, Ni or Pb among the tested vegetables.

Through the variance analysis of normalization-transformed mean of elements, it was concluded that the As, Cr, Cu, Cd, Pb and Ni levels in open-field vegetables were significantly higher than that in greenhouse vegetables ($P < 0.05$), and no significant differences in Zn concentrations between the two groups. This result showed that vegetables grown in a greenhouse were safer for people than vegetables grown in an open-field.

The variance analysis also showed that all HMs content except Zn in locally produced vegetables are significantly higher than that in non-locally produced vegetables ($P < 0.05$, Table 3).

2.2 EDI of heavy metals from vegetable consumption

The EDI values of HMs from vegetables for different age groups in Beijing are listed in Table 5. The EDI of HMs were compared with the provisional tolerable daily intakes (PTDIs) suggested by the Joint FAO/WHO Expert Committee on Food Additives JECFA or oral reference dose (RfD) to assess the potential health risks. As a result, children had the highest EDI of each element, and followed by adults and seniors.

The EDI of Cd from vegetable consumption in Beijing was 0.079, 0.062, and 0.053 μg/(kg bw·d) for children, adults and seniors, respectively. In comparison, the EDI of Cd from vegetables was lower in Bombay, India, Santiago, Chile, but higher in Tianjin and Huludao of China, Samta of Bangladesh, the Republic of Croatia and Harare of Zimbabwe (Table 5). Approximately 24% of the total daily dietary intake of Cd comes from vegetable consumption (Zhang and Gao, 2003). The EDI of cadmium from total dietary consumption corresponded to 32.9%, 25.8% and 22.1% of the PTDI (1 μg/(kg bw·d)) for children, adults and seniors, respectively.

In comparison, the intake of Pb from vegetables in Beijing was higher than in Tianjin, and in Huludao for children, while it was lower than the EDI for adults in Huludao. Comparing with other countries, the estimated dietary exposure of Pb in vegetables from Beijing was below that of Samta, Bangladesh, the Republic of Croatiaand Harare, Zimbabwe, but above that of Bombay, India and Santiago, Chile (Table 5). A total of 35.3% of the total daily dietary intake of Pb comes from vegetable consumption in China (Zhang and Gao, 2003). At this level, the EDI of Pb from total dietary intake accounted for 28.6%, 22.5% and 19.4% of the PTDI for children, adults and seniors, respectively.

Both Cu and Zn are essential elements for humans. The EDI of Cu and Zn from vegetables for people in Beijing was similar to the values from Tianjin and Huludao. Compared with other countries, the estimated dietary exposure of Cu in vegetables from Beijing was below that from vegetables in West Bengal, India, while above that from vegetables in Bombay, India, and Harare, Zimbabwe. The estimated dietary exposure of Zn in vegetables from Beijing was below that of West Bengal, India and Samta,

Table 2 Shape parameters and results of the K-S test for element concentrations in vegetables

	Raw data							Log-transformed data				Box-Cox transformed data		
	As	Cd	Cr	Cu	Ni	Pb	Zn	As	Cr	Cu	Ni	Cd	Zn	Pb
Kurtosis	38.8	12	26.9	31.3	46.4	6.8	17.3	–0.13	–0.38	0.83	1.03	0.33	1.28	–0.27
Skewness	5.1	2.5	4.6	4.7	5.7	2.2	3.1	–0.3	0.07	0.24	0.08	–0.26	0.44	–0.14
$P_{\text{K-S}}$	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.71	0.63	0.24	0.27	0.24	0.056	0.37

Table 3 Statistical analyses of element concentrations in vegetables collected from Beijing

Vegetable species	Number	Water content (%)	As (μg/g fw)		Cd (μg/g fw)		Cr (μg/g fw)	
			Range	GM	Range	GM	Range	GM
Chinese cabbage	46	93.1	0.001–0.19	0.011	0.001–0.049	0.017	0.001–0.266	0.019
Cabbage	27	92.2	ND–0.077	0.009	0.002–0.020	0.005	ND–1.008	0.013
Rape	15	92.4	0.004–0.071	0.015	0.008–0.058	0.019	0.006–0.276	0.031
Bok Choy	13	92.4	0.003–0.223	0.020	0.012–0.034	0.021	0.015–0.698	0.089
Cauliflower	11	92.0	ND–0.032	0.003	0.004–0.021	0.014	0.004–0.113	0.018
Celery	8	94.6	0.002–0.020	0.007	0.006–0.017	0.011	0.001–0.101	0.017
Spinach	7	92.3	0.002–0.030	0.006	0.011–0.023	0.018	0.013–0.276	0.089
Leek	2	91.0	0.008–0.014	0.011	0.002–0.009	0.005	0.006–0.025	0.012
Tomato	28	93.2	0.001–0.138	0.01	0.001–0.016	0.007	0.001–0.066	0.006
Cucumber	27	96.1	0.002–0.126	0.022	ND–0.015	0.003	0.002–0.204	0.021
Capsicum	24	92.1	0.001–0.116	0.021	0.003–0.046	0.014	0.002–1.040	0.037
Kidney bean	21	86.4	0.001–0.202	0.017	ND–0.015	0.007	0.005–0.456	0.090
Eggplant	18	92.2	0.004–0.075	0.017	0.004–0.062	0.015	0.005–0.242	0.041
Wax gourd	15	90.4	ND–0.065	0.005	0.001–0.013	0.004	0.002–0.687	0.012
Radish	29	95.2	0.004–0.479	0.033	0.002–0.037	0.012	0.001–0.175	0.031
Scallion	20	85.4	0.004–0.138	0.029	ND –0.038	0.008	0.007–0.257	0.056
Potato	7	85.0	0.001–0.038	0.009	0.006–0.033	0.015	0.006–0.118	0.029
Garlic	7	89.1	0.001–0.310	0.012	0.004–0.015	0.007	0.005–0.045	0.013
Lotus root	1	86.5	0.028	0.028	0.01	0.010	0.007	0.007
Other vegetable	86	–	ND–0.127	0.01	ND–0.101	0.008	0.001–0.723	0.015
Leafy vegetable	183	–	ND–0.223	0.011	ND–0.101	0.013	ND–1.008	0.021
GFV	152	–	ND –0.202	0.013	ND–0.062	0.007	0.001–1.040	0.023
SRV	81	–	0.001–0.479	0.018	ND –0.038	0.010	0.001–0.257	0.026
LPV	330	–	ND–0.479	0.013	ND–0.101	0.010	ND–1.04	0.025
NLPV	86	–	ND–0.310	0.011	ND–0.049	0.009	0.001–0.148	0.016
Open-field vegetable	236	–	ND–0.479	0.021	ND–0.062	0.011	0.001–0.698	0.035
Greenhouse vegetable	180	–	ND–0.138	0.007	ND–0.101	0.008	ND–1.04	0.013
All vegetables	416	–	ND–0.479	0.013	ND–0.101	0.010	ND–1.04	0.023

Vegetable species	Cu (μg/g fw)		Ni (μg/g fw)		Pb (μg/g fw)		Zn (μg/g fw)	
	Range	GM	Range	GM	Range	BCM	Range	BCM
Chinese cabbage	0.13–2.16	0.29	0.012–0.157	0.048	0.003–0.370	0.045	0.37–7.42	2.52
Cabbage	0.08–1.00	0.25	0.006–0.625	0.048	ND–0.286	0.038	1.1–10.05	2.42
Rape	0.24–2.15	0.47	0.011–0.118	0.045	0.008–0.220	0.075	1.64–5.80	2.96
Bok Choy	0.22–3.47	0.56	0.011–0.339	0.071	0.023–0.350	0.098	0.86–6.23	2.92
Cauliflower	0.29–5.63	0.58	0.021–0.350	0.068	0.001–0.139	0.032	3.68–7.70	5.45
Celery	0.13–0.85	0.35	0.015–0.704	0.061	0.008–0.116	0.028	0.91–2.49	1.35
Spinach	0.44–1.05	0.70	0.046–0.109	0.073	0.008–0.061	0.031	1.53–5.94	2.99
Leek	0.48–0.53	0.51	0.033–0.164	0.073	–	–	2.15–3.39	2.72
Tomato	0.13–0.77	0.41	0.008–0.141	0.029	0.001–0.231	0.029	0.16–1.90	1.05
Cucumber	0.22–1.66	0.45	0.001–0.370	0.051	0.002–0.123	0.035	0.36–4.73	1.64
Capsicum	0.15–2.46	0.82	0.014–0.462	0.075	ND–0.447	0.085	0.12–8.91	1.98
Kidney bean	0.43–8.25	1.53	0.090–0.993	0.191	ND–0.324	0.121	0.60–12.74	4.93
Eggplant	0.20–2.71	0.77	0.007–0.196	0.036	0.012–0.367	0.060	0.01–8.97	1.70
Wax gourd	0.02–1.12	0.19	0.005–0.177	0.036	0.002–0.068	0.013	0.16–1.50	0.58
Radish	0.09–1.20	0.34	0.005–0.199	0.066	0.001–0.440	0.074	1.01–8.88	2.48
Scallion	0.32–3.96	0.61	0.007–0.114	0.04	0.002–0.261	0.068	0.30–5.37	1.81
Potato	0.52–1.88	1.03	0.022–0.087	0.054	0.003–0.286	0.067	1.62–8.86	3.77
Garlic	0.21–1.82	0.67	0.020–0.069	0.04	0.007–0.655	0.142	2.86–11.15	5.11
Lotus root	1.62	1.62	0.127	0.127	–	–	3.33	3.33
Other vegetable	0.14–6.34	0.67	0.006–1.689	0.051	ND–0.218	0.018	0.06–25.6	3.66
Leafy vegetable	0.08–5.63	0.42	0.006–0.966	0.05	ND–0.370	0.038	0.37–16.05	3.02
GFV	0.02–8.25	0.60	0.001–1.689	0.059	ND–0.447	0.051	0.01–25.6	2.02
SRV	0.09–3.96	0.55	0.005–0.316	0.048	ND –0.655	0.057	0.06–11.15	2.58
LPV	0.02–8.25	0.52	0.001–0.966	0.053	ND–0.655	0.059	0.01–16.05	2.42
NLPV	0.09–6.34	0.45	0.01–1.689	0.053	ND–0.286	0.024	1.07–25.6	3.11
Open-field vegetable	0.09–8.25	0.55	0.001–1.689	0.060	ND–0.447	0.054	0.01–25.6	2.64
Greenhouse vegetable	0.02–5.63	0.45	0.005–0.966	0.045	ND–0.655	0.029	0.06–16.05	2.45
All vegetables	0.024–8.25	0.51	0.001–1.689	0.053	ND–0.655	0.046	0.01–25.6	2.55

GM: geometrical mean; BCM: Box-Cox mean.
GFV: gourd and fruit vegetable; SRV: stem and root vegetable; LPV: local-produced vegetable; NLPV: non-locally produced vegetable.
ND: concentration < 0.001 μg/g.

Bangladesh, while above that of Bombay, India (Table 5).

The EDI of Cu from vegetable consumption accounted for 0.8%, 0.6% and 0.5% of the PTDI for children, adults and seniors, respectively. The EDI of Zn from vegetables accounted for 2.0%, 1.6% and 1.4% of the RfD for children, adults and seniors, respectively. Research on the vegetable consumption from Huludao showed that the estimated Cu and Zn daily intake via vegetable consumption

Table 4 Tolerance limit of heavy metals in vegetables

	Vegetable type	As[a]	Cd[a]	Cr[a]	Cu[b]	Ni[c]	Pb[a]	Zn[d]
Maximum level (μg/g fw)	Legumes and pulses	0.1	0.2	1.0	20	3.0	0.2	100
	Leafy vegetables	0.05	0.2	0.5	10	0.3	0.3	20
	Stem and root vegetables	0.05	0.1	0.5	10	0.3	0.1	20
	Other vegetables	0.05	0.05	0.5	10	0.3	0.1	20
Samples exceeded the limit (%)		12.6	0.58	0.96	0	2.62	17.3	0

[a] GB2762-2005; [b] GB 15199-94; [c] Yang *et al.*, 1998; [d] GB 13106-91.

Table 5 Comparison of estimated dietary intake of heavy metals through vegetable consumption in different areas

Site		Vegetable intake (g/d)	EDI of HMs (μg/(kg bw·d))							Reference
			As	Cd	Cr	Cu	Ni	Pb	Zn	
Beijing, China	Adults	394	0.080	0.062	0.142	3.14	0.327	0.283	15.7	This study
	Children	257	0.102	0.079	0.181	4.01	0.416	0.361	20.0	
	Seniors	323	0.069	0.053	0.122	2.70	0.281	0.245	13.5	
Tianjin, China	Adults	345	–[a]	0.14	0.75	3.2	–	0.12	12	Wang *et al.*, 2005
	Children	–		0.16	0.9	4.4	–	0.13	15	
Huludao, China	Adults	242	–	0.42	–	4.1	–	0.43	17.2	Zheng *et al.*, 2007
	Children	109	–	0.32	–	3.1	–	0.33	13.2	
Bombay, India	Adults	105	–	0.009	–	1.14	–	0.035	4.2	Tripathi *et al.*, 1997
West engal, India	Adults	500	0.174	–	–	13.3	3.0	–	44.1	Roychowdhury *et al.*, 2003
Samta, Bangladesh	Adults	130	0.463	0.158	–	–	–	1.25	59.5	Alam *et al.*, 2003
Republic of Croatia	Adults	284	0.002	0.11	–	–	–	0.46	–	Sapunar-Postruznik *et al.*, 1996a; Sapunar-Postruznik *et al.*, 1996b
Santiago, Chile	Adults	327	0.038	0.021	–	–	–	0.15	–	Munoz *et al.*, 2005
Harare, Zimbabwe	Adults	219	–	0.33–0.66	0.8–1.6	0.7–0.8	0.8–1.6	0.8-1.5	10–55	Mapanda *et al.*, 2007
RfD			0.3	1	3	40	20	3.57[b]	300	
PTDI			2.14	1	–	500	–	3.57	1000	

[a]: no data; [b]: RfD for Pb not available, JECFA value 3.57 (μg/(kg bw·d)) was used.
RfD: obtained from the Integrated Risk Information Systems (IRIS)(2003), USEPA (http://www.epa.gov/iris) (USEPA, 2007a, 2007b); PTDI: provisional tolerable daily intake suggested by FAO/JECFA.

was responsible for 8.4% and 7.8% of the total dietary intake for adults, respectively (Zheng *et al.*, 2007). As the general population of Beijing has a similar diet, the EDI of Cu and Zn from the total dietary intake accounted for 9.5% and 25.6%, 7.1% and 20.5%, 6.0% and 17.9% of the PTDI for children, adults and seniors, respectively. The daily dietary intake of Cu and Zn was far below the recommended PTDI value.

The EDI of Cr and Ni from vegetable consumption in Beijing was lower than that in West Bengal, India and Harare, Zimbabwe. For Cr, the EDI for inhabitants from Beijing was lower than that in Tianjin (Table 5). As PTDI was not available for Cr and Ni, the oral reference dose (RfD) was used for comparison. The EDI of Cr and Ni from vegetable consumption corresponded to 6.0% and 2.1%, 4.7% and 1.6%, 4.1% and 1.4% of the RfD for children, adults and seniors, respectively.

Arsenic is one of the most important global environmental toxicants (Das *et al.*, 2004). The health problems arising from As intake are found throughout the world (Hossain, 2006; Yost *et al.*, 2004). The results from this study showed that the estimated total As (AsT) daily intake from vegetable consumption was 0.080, 0.102, and 0.069 μg/(kg bw·d) for adults, children and seniors, respectively. Thus the As intake from vegetables in Beijing was higher than 0.038 μg/(kg bw·d) for adults in Santiago, Chile and 0.002 μg/(kg bw·d) for adults in Republic of Croatia, but the level was lower than 0.463 μg/(kg bw·d) that for adults in Bangladesh and 0.174 μg/(kg bw·d) for adults in West Bengal, India (Table 5).

It is known that inorganic arsenic (AsI) is much more toxic than organic arsenic (Edmonds and Francesconi, 1993) and inorganic arsenic is a potent human carcinogen (Yost *et al.*, 1998, 2004). An intake of AsI of 10–50 μg/(kg bw·d) contributes to vascular problems, which may ultimately lead to necrosis and gangrene of the hands and feet (Larsen and Berg, 2001). Thus it is preferred to assess the risk of As to humans through measurement of the intake of AsI.

Based on the rate of AsI to AsT in vegetables of 83% (Diaz *et al.*, 2004), the daily intake of AsI was determined to be 0.085, 0.066, 0.057 μg/kg bw for children, adults and seniors in Beijing, which corresponded to 3.96%, 3.1% and 2.68% of 2.14 μg AsI/kg bw (WHO, 1989), respectively, from the PTDI established by the FAO/WHO. In China, a total of 4.8% of the total daily dietary intake of As comes from vegetable consumption (Zhang and Gao, 2003). The EDI of AsI accounted for 64.6%, 82.4% and 55.8% of the PTDI for adults, children and seniors, respectively, which was close to the PTDI limits suggested by JECFA, but was within a level that was considered safe.

The EDI of Cd, Cu, Pb and Zn obtained through Beijing vegetable consumption was higher than that in Bombay, India. In Beijing, the EDI of As, Cd and Zn was also higher

Table 6 Target hazard quotient (THQ) values of heavy metals from vegetables

Element	Cumulative probability percentile of THQ_c								THQ_w
	Min	25	50	75	85	95	99	Max	
Adults									
As	0.007	0.103	0.275	0.65	1.03	1.78	4.38	9.45	0.268
Cd	0.001	0.029	0.059	0.10	0.13	0.21	0.32	0.60	0.063
Cr	0.001	0.015	0.042	0.13	0.23	0.50	1.40	2.05	0.051
Cu	0.004	0.046	0.069	0.12	0.17	0.28	0.79	1.22	0.079
Ni	0.000	0.008	0.016	0.03	0.04	0.09	0.22	0.50	0.017
Pb	0.000	0.027	0.085	0.19	0.26	0.46	0.73	1.08	0.074
Zn	0.000	0.029	0.044	0.08	0.10	0.15	0.30	0.50	0.053
TTHQ	–	–	–	–	–	–	–	–	0.605
Children									
As	0.000	0.130	0.350	0.82	1.30	2.27	5.58	12.02	0.341
Cd	0.001	0.037	0.076	0.13	0.16	0.26	0.40	0.76	0.080
Cr	0.001	0.019	0.054	0.17	0.30	0.64	1.79	2.61	0.065
Cu	0.005	0.059	0.088	0.16	0.22	0.36	1.01	1.55	0.101
Ni	0.000	0.011	0.021	0.04	0.05	0.12	0.28	0.64	0.022
Pb	0.000	0.034	0.108	0.24	0.33	0.59	0.93	1.38	0.095
Zn	0.000	0.037	0.056	0.10	0.13	0.19	0.38	0.64	0.067
TTHQ	–	–	–	–	–	–	–	–	0.771
Seniors									
As	0.006	0.089	0.237	0.56	0.88	1.53	3.76	8.13	0.230
Cd	0.001	0.025	0.051	0.09	0.11	0.18	0.27	0.52	0.054
Cr	0.001	0.013	0.036	0.11	0.20	0.43	1.21	1.76	0.044
Cu	0.003	0.040	0.059	0.11	0.15	0.24	0.68	1.05	0.068
Ni	0.000	0.007	0.014	0.02	0.03	0.08	0.19	0.43	0.015
Pb	0.000	0.023	0.073	0.16	0.22	0.40	0.63	0.93	0.064
Zn	0.000	0.025	0.038	0.07	0.09	0.13	0.26	0.43	0.045
TTHQ	–	–	–	–	–	–	–	–	0.520

THQ_c: the THQ based on the percentile concentration of element in vegetables; THQw: the THQ based on the weighted average concentration of element in vegetables; TTHQ: total THQ of heavy metals.
–: no data.

than that in Santiago, Chile. The EDI of As, Cd, Pb and Zn obtained via Beijing vegetable consumption was lower than that in Samta, Bangladesh (Table 5).

2.3 Health risk from heavy metals in vegetables

The THQs of HMs in vegetables for the inhabitants in Beijing are listed in Table 6. Among those 7 elements, the THQ of As was the highest, and was higher by 2–14 folds than that of the other elements. The contribution of As from vegetable consumption to THQ_W was 44.3% of total THQ (TTHQ) value. The THQ_c for As at the 85th percentile exceeded the acceptable safety value of unity for adults and children. The results indicated that the non-carcinogenic risks from oral As exposure from vegetables was significant at the 85th percentile or the higher end of the range and the risk is especially high for children because they are more sensitive to pollutants. The THQ for Cd, Ni or Zn was generally less than 1 at all percentiles for all age groups in Beijing (Table 6), suggesting that it is not risky for the inhabitants to consume these single elements in vegetables. For adults, and seniors, the THQ_c of Cu and Pb was less than 1 at the 99th percentile or below, and the THQ_c of Cr was less than 1 at the 95th percentile or below (Table 6), indicating that there would be a higher probability of experiencing carcinogenic effects for those ingest vegetables containing higher concentrations of HMs.

As shown in Table 6, except the minimum of THQ_c, the THQ_c and THQ_W for children were higher than that for adults and seniors suggesting that the exposure risk of HMs to children through vegetable consumption was higher than to adults and seniors. Children are very sensitive to Pb, for it could jeopardize their nerve and mental development (Jin *et al*., 2006; Sovcikova *et al*., 1996). Table 6 shows that the THQ_c value for Pb was very close to or exceeded the level considered to be non-carcinogenic at the 99th percentile or higher. The potential health risks to children ingesting Pb from vegetables was higher than that for other two groups.

It has been reported that exposure to two or more pollutants may result in additive and/or interactive effects (Hallenbeck and William, 1993). In this study, the TTHQ was treated as the mathematical sum of each individual THQ from each vegetable species according to its C_W. For all three age groups, the TTHQ was lower than 1, indicating that it is still safe for the general population with vegetable consumption pattern of Beijing at present.

3 Conclusions

The Cu and Zn levels in all samples were within the Chinese National Standards, but the Pb, As, Ni, Cr and Cd concentration in 17.3%, 12.6%, 2.62%, 0.96% and 0.58% of samples surpassed the standards. All elements except Zn in open-field vegetable and local-produced vegetable were all significantly higher than those in greenhouse vegetable and non-locally produced vegetable, respectively. The EDI of As, Cd, Cr, Cu, Ni, Pb and Zn from vegetables, respectively, was 0.080, 0.062, 0.142, 3.14, 0.327, 0.283 and 15.7 μg/(kg bw·d) for adults, 0.102, 0.079, 0.181, 4.01, 0.416, 0.361 and 20.0 μg/(kg bw·d) for children, and 0.069, 0.053, 0.122, 2.70, 0.281, 0.245 and 13.5 μg/(kg bw·d) for seniors in Beijing. Arsenic was the major risk contributor to inhabitants from vegetable consumption, and the THQ_c of As at the 85th percentile was close to or exceeded the acceptable safety value of unity for all age groups in Beijing. In conclusion, it is still safe for the population of Beijing to consume vegetable as the TTHQ were all lower than 1 for studied elements for all age groups.

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