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Health risk assessment of heavy metals for edible parts of vegetables grown in sewage-irrigated soils in suburbs of Baoding City, China

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Abstract With the long-term application of wastewater to vegetable production fields, there is concern about potential health risks of heavy metals contaminating the edible parts of vegetables grown in contaminated soils in the suburban areas of Baoding City, China. The average concentration of elemental Zn in sewage-irrigated soil was the highest (153.77 mg kg^{-1}), followed by Pb (38.35 mg kg^{-1}), Cu (35.06 mg kg^{-1}), Ni (29.81 mg kg^{-1}), and Cd (0.22 mg kg^{-1}) which were significantly higher ($P<$ 0.05) than those in the reference soil. The results showed that long-term sewage irrigation had led to a growing accumulation of heavy metals in the soils, especially for Cd, Zn, and Pb. Furthermore, the concentrations of elemental Cd, Zn, and Ni in vegetables (e.g., *Beassica pekinensis* L., *Allium fistulosum* L., *Spinacia oleracea* L.) collected from the wastewater-irrigated soils exceeded the maximum permissible limits, and this also increased the daily intake of metals by food. However, compared with the health risk index of <1 for heavy metals, the ingestion of vegetables from the soils irrigated with sewage effluent posed a low health risk. Nevertheless, heavy metal concentrations should be periodically monitored in vegetables grown in these soils together with the implementation effective remediation technologies to minimize possible impacts on human health.

Keywords Assessment · Health risk · Heavy metals · Sewage irrigation · Vegetables

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Introduction

Wastewater has been widely used for agricultural irrigation in many countries around the world (Friedel et al. 2000; Barbagallo et al. 2001), including cities or provinces in China, such as Beijing (Liu et al. 2005), Shanghai (Chen et al. 2007), Liaoning (Sun et al. 2008), Hebei (Wu et al. 2003). However, long-term application of partially treated or untreated wastewater could result in the accumulation of heavy metals in the soil (van de Graaff et al. 2002; Kirpichtchikoval et al. 2003; Rattan et al. 2005; Dère et al. 2007; Qishlaqi et al. 2008; Li et al. 2009). The rate at which heavy metals are accumulated in the soil depends on the physiochemical properties of the soil and the relative efficiency of crops to remove the metals from the soil. Heavy metals accumulated in cultivated soils can be transferred to humans through various exposure pathways causing adverse effects on human health (Qishlaqi et al. 2008). One major exposure pathway of heavy metals to humans is through the consumption of crops grown in the soils which are contaminated by heavy metals from wastewater irrigation (Liu et al. 2005; Qishlaqi et al. 2008; Chary et al. 2008; Khan et al. 2008).

Contaminated vegetables by heavy metals have been documented in many provinces of China (Dong et al. 2001). Sewage irrigation is one of the main causes for vegetable contamination by heavy metals. The use of treated and untreated wastewater for irrigation increased the contamination of Cd, Pb, and Ni in the edible portion of vegetables, thus posing potential health risks for humans (Sharma et al. 2007). The excessive accumulation of dietary heavy metals such as Cd, Cr, and Pb in the human body can result in serious systemic health problems (Oliver 1997). Therefore, the Food and Agriculture Organization and World Health Organization (WHO), US Environmental Protection Agency (US-EPA), and other regulatory bodies of various countries have established the maximum permitted concentrations of heavy metals in foodstuffs or soils (Qishlaqi et al. 2008; Chary et al. 2008). The level of health risks posed by wastewater with heavy metals was determined using different indices, including the transfer factor (TF), daily intake of metals (DIM), and health risk index (HRI) or health quotient (HQ) (Liu et al. 2005; Chary et al. 2008; Khan et al. 2008).

Earlier studies by authors in Baoding City of Hebei province reported concentrations of heavy metals in soils and vegetables (Xie et al. 2002; Wang 2005). There are no reported studies on possible health risks associated with the consumption of contaminated vegetables grown in sewage-irrigated soils. This study was conducted to determine the concentration of heavy metals in sewage-irrigated soils, uptake by vegetables grown in the contaminated soils, and the transfer to human body through the food chain. This knowledge is important for assessing the health risks associated with consuming vegetables grown in soils irrigated with wastewater.

Materials and methods

Description of study area

The study areas are located in the suburbs of Baoding of Hebei province, which is situated between latitude of 113°40′–116°20′ E and longitude of 38°10′–40°00′ N and borders upon Beijing and Tianjin cities in China. The area has a continental monsoon climate, characterized by a wide seasonal variation in average annual rainfall (547 mm), cold-dry winters (average −4.5°C) and hot-wet summers (average 26.5°C) (Wang 2005). The soil was formed on the flood alluvium of the Taihang Mountains with alluvial fans in the Baoding City. The soil classification is aquic cinnamon soils (Argosols) in Chinese Soil Taxonomy, which corresponds to Udic haplustalf (Alfisols) in the US Soil Taxonomy. The surface area of suburban region is close to 2,000 ha, of which approximately 1,100 ha is occupied by vegetable crops. The cultivation system of vegetables is a rotation system with wheat–vegetable or vegetable–vegetable annually. Owing to deficiency in groundwater resource, nearly 900 ha of vegetable crops have been irrigated with wastewater from domestic and industrial effluent which was partially treated or untreated since 1970s.

The average volumes of river water applied in the studied area, estimated from the questionnaires, were 24 ML ha^{-1} $year^{-1}$ with a range from 19 ML ha^{-1} $year^{-1}$ for wheat–vegetable rotation system to 29 ML ha^{-1} $year^{-1}$ for vegetable–vegetable rotation system. The average concentrations of heavy metals in the wastewater were obtained from a monitoring study by Bureau of Environmental Protection of Baoding City during the period of the Tenth Five-Year Plan. Then, they were used to estimate the annual loading rates of heavy metals in the sewage-irrigated soils.

Soil sampling and analysis

Soil samples used in this experiment were collected from sewage-irrigated soils as well as from the groundwater-irrigated (reference) soils in the suburbs of Baoding on October 2009. Global positioning system was used to locate every sampling site. Five surface soil (0–20 cm) samples were collected using a Dutch auger within a distance of 20 m surrounding a specific sampling location and then were mixed. For each location, a total of 1 kg of soil was taken from the mixed samples using a quartile method. According to the earlier sampling site of soil in suburbs of Baoding (Xie et al. 2002; Wang 2005), a total of 19 samples including 14 from wastewater-irrigated soils (W1–W14) and 5 from control soils (C1–C5) were collected from suburban areas in representative wastewater- and groundwater-irrigated soils with common vegetable cultivars grown (Fig. 1).

The samples were air-dried, ground, mixed, passed through a 2-mm sieve and then sealed in Kraft paper envelopes for further analysis. Soil pH was measured with a glass electrode using soil–water suspension method (Datta et al. 1997). Soil organic matter was determined using the potassium dichromate oxidation method (Bao 2000). The particle size distribution was determined by the hydrometer method (Day 1965). Total N, available P, and K were determined using Uygur et al. (2010) method. Total concentrations of Cu, Zn, Ni, Pb, and Cd were determined by high-resolution inductively coupled plasma mass spectrometry (HR-ICP-MS, Element 2, Finnigan Co., Waltham, MA, USA) (Bao 2000).

Vegetable sampling and analysis

Although 15 kinds of vegetables were found in the sewage-irrigated soils, the most common vegetables were Chinese cabbage (*Beassica pekinensis* L.), cauliflower (*Brassica oleracea* L.), scallion (*Allium fistulosum* L.), and spinach (*Spinacia oleracea* L.), which covered 32%, 23%, 18%, and 15% of planted area, respectively. When they were harvested, edible parts of these vegetables were collected from the same sites where soils were collected. Vegetable samples were washed with distilled water for three to four times to remove foreign material, and then were prepared according to the procedure by Arora et al. (2008). The heavy metal concentrations were determined with HR-ICP-MS.

Quality assurance

Soil and vegetable samples were digested in triplicate along with blanks to minimize error. The instrument was calibrated with a series of standard solutions supplied by the manufacturer. All determinations were replicated three times.

Data analysis

Pollution load index

The following modified equation was used to assess the pollution load index (PLI) level in soils (Liu et al. 2005).

$$PLI = \frac{C_{soil}(Samples)}{C_{reference}(Reference)}$$

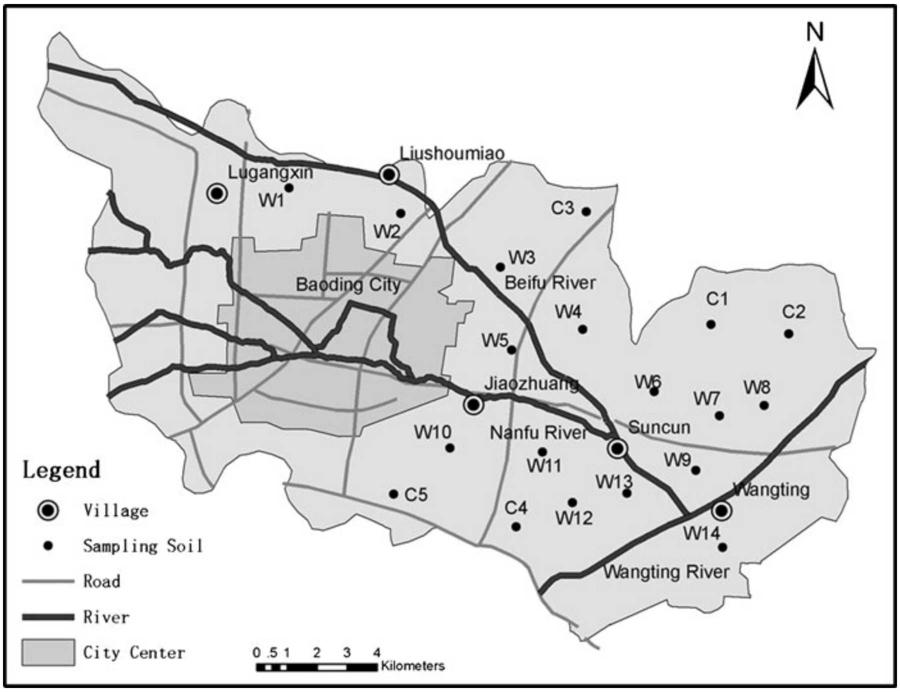

Fig. 1 Sketch map of soil sampling sites in the studied area. *W1–W14* and *C1–C5* are representative soil sites irrigated with wastewater and groundwater, respectively

where C_{soil} (samples) and $C_{reference}$ (reference) represent the concentrations of heavy metal in the sewage-irrigated and reference soils, respectively.

Transfer factor from soil to vegetable

In order to assess the transfer of metals from soil to vegetables, the TF values of metals were calculated as follows.

$$TF = \frac{C_{vegetable}}{C_{soil}}$$

where $C_{vegetable}$ and C_{soil} represent the concentration of heavy metal in extracts of vegetables and soils on dry weight (DW) basis, respectively.

Daily intake of metals

The exposure pathway of heavy metals to human through ingestion of vegetables grown on sewage-irrigated soils was focused by many researchers (Arora et al. 2008; Khan et al. 2008; Qishlaqi et al. 2008; Chary et al. 2008). Moreover, the DIM in this exposure pathway was determined by the following equation.

$$DIM = \frac{C_{vegetable} \times C_{factor} \times D_{food\ intake}}{B_{average\ weight}}$$

where $C_{vegetable}$, C_{factor}, $D_{food\ intake}$ and $B_{average\ weight}$ represent the concentrations of heavy metal in vegetables (milligrams per kilogram) on dry weight basis, conversion factor for fresh to dry weight of vegetables (0.085) (Rattan et al. 2005), daily intake of vegetables (0.345 kg per person per day for adults, 0.232 kg per person per for children) and average body weight (55.90 kg for adults, 32.70 kg for children), respectively (Ge 1992; Wang et al. 2005).

Health risk index

Health risk assessment of consumers from the intake of metal-contaminated vegetables was characterized by using HRI. If HRI is less than 1, there will not be obvious risk for exposed population and vice verse. The HRI was calculated by using the equation below.

$$HRI = \frac{DIM}{RfD}$$

Where reference oral doses (RfD) for Cu, Zn, Ni, Pb, and Cd are 4E-2, 3E-1, 2E-2, 3.5E-3, and 1E-03 mg kg^{-1} day^{-1} in this study, respectively (US-EPA, IRIS 2002).

Statistical analysis

All the data obtained in the study were analyzed using a statistical package SPSS 16.0 and subjected to statistical analysis of independent samples *T* test. Differences at the $P<0.05$ level were considered significant. The figures were constructed using the SigmaPlot 10.0.

Results

Physicochemical properties of soils

The physicochemical properties of the sewage-irrigated and reference soils are shown in Table 1. The decrease in pH by 0.28 U accompanied with an increase in organic matter by 59.17%, total nitrogen by 48.21%, and available phosphorus by 68.61% in sewage-irrigated soils compared to reference soils. This suggested that the selected properties were significantly affected by sewage irrigation ($P<0.05$). There was no significant difference in clay content and available potassium between sewage-irrigated and reference soils ($P>0.05$), although clay content in the sewage-irrigated soil was, on average, 7.69% higher than in reference soils.

Heavy metal concentrations of soils

According to the description of heavy metal concentrations in wastewater, only Zn and Cd concentrations did not meet the maximum recommended standard for vegetable irrigation in China (Table 2). Correspondingly, the estimated annual loading rates were above their maximum permissible limits (MPLs), being 4.10-fold for Zn and 1.60-fold for Cd higher, respectively. Furthermore, the average concentrations of Cu, Zn, Ni, Pb, and Cd exceeded those of background values of Hebei province by 1.62, 2.26, 1.02, 1.92, and 2.75-fold for the corresponding metals for sewage-irrigated soils and 1.26, 1.21, 0.82, 1.27, 1.38-fold for the corresponding metals for reference soils, respectively. The elemental Zn, Cu, and Pb concentrations in the sewage-irrigated soils were higher than the concentration limits established by

Table 1 Physicochemical properties of soils irrigated with sewage and groundwater in the studied area

Physicochemical parameters	Sewage-irrigated soils		Reference soils	
	Range	Average±SD	Range	Average±SD
pH (H_2O)	7.40–8.18	7.84±0.22	8.02–8.33	8.12±0.14
Organic matter (g kg^{-1})	14.89–33.13	22.65±5.17	9.88–18.54	14.23±3.40
Clay (%)	14.04–22.07	15.55±2.75	14.04–16.05	14.44±0.90
Silt (%)	30.09–64.20	39.65±7.77	36.11–40.12	37.71±1.68
Sand (%)	39.82–53.86	44.80±8.12	45.84–49.85	47.84±2.01
Total N (g kg^{-1})	0.48–1.25	0.83±0.21	0.22–0.81	0.56±0.22
Available P (mg kg^{-1})	23.65–81.01	50.55±14.76	14.73–48.00	29.98±14.95
Available K (mg kg^{-1})	78.74–83.45	81.29±1.35	80.94–83.54	82.57±1.13

WHO. Because of the higher background values of elemental Zn, Cu, and Pb in Hebei province, the concentrations of corresponding heavy metals in reference soils were also closed to the limits proposed by WHO. Moreover, the PLI for Cu, Zn, Ni, Pb, and Cd were 1.29, 1.86, 1.24, 1.51, and 2.00 (Fig. 2), respectively. The results indicated that long-term sewage irrigation had led to an accumulation of heavy metals in soils, especially for Cd, Zn, and Pb.

Heavy metal concentrations of vegetables

The average concentrations of heavy metals in the dry matter of edible parts of vegetables grown in sewage-irrigated soils showed significant differences as shown in Fig. 3. In comparison with different edible parts of vegetables cultivated in sewage-irrigated soils, the edible parts of spinach (*S. oleracea* L.) accumulated the highest concentration of Cu (average 9.23 mg kg^{-1} DW), Pb (average 3.68 mg kg^{-1} DW), and Cd (average 0.27 mg kg^{-1} DW), while the highest concentrations of Zn (average 86.47 mg kg^{-1} DW) and Ni (average 9.35 mg kg^{-1} DW) were recorded in Chinese cabbage (*B. pekinensis* L.). It is clear that edible parts of vegetable species showed variations in the accumulation of metals. According to SEPA (2005), the MPLs of Cu, Zn, Ni, Pb, and Cd for vegetables and fruits are 20, 100, 10, 9, and 0.2 mg kg^{-1}, respectively, on a dry weight basis (Khan et al. 2008). Apart from cauliflower (*B. oleracea* L), the heavy metal concentrations of Cd, Zn, and/or Ni in edible parts of all the other vegetables exceeded these MPLs (Fig. 3); therefore, they were not fit for human consumption. It indicated that some edible parts of vegetables (e.g., *B. pekinensis* L., *A. fistulosum* L., *S. oleracea* L.) were heavily contaminated with elemental Cd, Zn, and/or Ni in sewage-irrigated soils. Consequently, close monitoring of the concentrations of Cd, Zn, and/or Ni in edible parts of vegetables should be followed for leafy vegetables irrigated with wastewater.

Transfer factors of metals from sewage-irrigated soils to vegetables

TFs of heavy metals varied greatly among the selected vegetables grown in sewage-irrigated soils (Table 3). The trend of TF for different metals in the same edible parts of vegetables grown in sewage-irrigated soils were in the order of Cd>Zn>Ni>Cu>Pb for *B. pekinensis* L., Zn>Ni>Cd>Cu>Pb for *B. oleracea* L., Cd>Ni>Zn>Cu>Pb for *A. fistulosum* L., and Cd>Zn>Ni>Cu>Pb for *S. oleracea* L., respectively. It is seen that elemental Cd had the greatest bioavailability and Pb had the lowest bioavailability for different vegetables in this studied area, although the concentration of Pb was almost 170-fold bigger than that of Cd in the sewage-irrigated soils (Table 2). The observed results indirectly proved that uptake of metals by vegetables mainly depended on the bioavailability of the metals. As far as the TF of the same metal in different edible parts of vegetables was concerned, relative efficiency of vegetables to uptake metals from sewage-irrigated soil were found in the following order: *S. oleracea* L.>*B. pekinensis* L.>*B. oleracea* L.>*A. fistulosum* L. for Cu and Zn, *A. fistulosum* L.>*B. pekinensis* L.>*S. oleracea* L.>*B. oleracea* L. for Ni, *S. oleracea* L.>*B. pekinensis* L. >*A. fistulosum* L.>*B. oleracea* L. for Pb, and *S. oleracea* L.>*B. pekinensis* L.>*A. fistulosum* L.>*B.*

Table 2 Heavy metal concentrations in water (milligrams per liter) and in soil (milligrams per kilogram), as well as estimated annual loading rates (kilograms per hectare per year) in the studied area

Heavy metals	Water					Soil			
	Average (range) concentrations in wastewater[a]	Average (range) concentrations in groundwater[a]	Maximum concentrations for irrigation, China[b]	Estimated annual loading rate	Annual loading rate limits[c]	Average (range) concentrations in sewage irrigated soil	Average (range) concentrations in reference soil	Background value, Hebei province[d]	WHO limits[e]
Cu	0.08 (0.03–0.11)	0.03 (0.00–0.03)	1.00	1.92	7.50	35.06 (26.11–44.01)	27.26 (25.30–29.64)	21.70	30.00
Zn	2.56 (1.75–3.11)	0.03 (0.01–0.04)	2.00	61.44	15.00	153.77 (112.70–250.00)	82.49 (72.60–93.00)	68.00	90.00
Ni	–	0.01(0.00–0.01)	0.20	–	3.00	29.21 (26.03–38.24)	23.47 (21.30–26.14)	28.60	–
Pb	0.03 (0.01–0.05)	ND	0.10	0.72	15.00	38.35 (27.41–57.91)	25.46 (21.50–33.14)	20.00	30.00
Cd	0.01 (0.005–0.015)	ND	0.005	0.24	0.15	0.22 (0.17–0.40)	0.11 (0.10–0.14)	0.08	0.35

ND not detected

[a] BEPBC (2005)

[b] Wang et al. (2007)

[c] DoE (1996)

[d] DEPH (2010)

[e] Tembo et al. (2006)

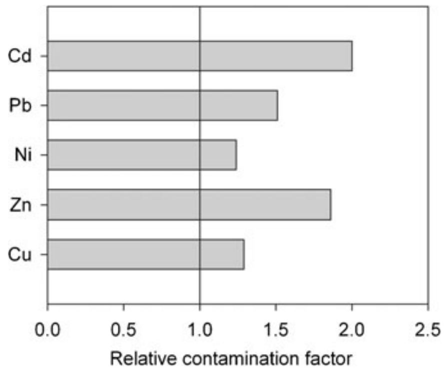

Fig. 2 Pollution load index for heavy metal in sewage-irrigated soils

oleracea L. for Cd, respectively. This information will be very useful in selecting the vegetable, e.g., *B. oleracea* L. to be grown on sewage-irrigated soils to reduce the health risks of metal transfer to humans.

Health risk assessment

There was a substantial buildup of Cu, Zn, Ni, Pb, and Cd in sewage-irrigated soils. They will pose a threat to human health in the food chain. Hence, the DIM and HRI for adults and children through consumption of vegetables grown in sewage-irrigated soils were estimated (Table 4). The highest DIM of Cu, Zn, Ni, Pb, and Cd for both adults and children were from the consumption of *S. oleracea* L., *B. pekinensis* L., *B. pekinensis* L., *S. oleracea* L., and *S. oleracea* L., respectively. This indicated that both adults and children consuming *S. oleracea* L. and *B. pekinensis* L. grown in sewage-irrigated soils ingested greater amount of the metals studies. The HRI of Cu varied from 5.52E-2 to 1.21E-1, Zn 8.63E-2 to 1.51E-1, Ni 1.84E-1 to 2.45E-1, Pb 5.65E-2 to 5.52E-1, and Cd 2.19E-2 to 1.44E-1, respectively, for adults; while Cu varied from 6.34E-2 to 1.39E-1, Zn 9.92E-2 to 1.74E-1, Ni 2.11E-1 to 2.82E-1, Pb 6.50E-2 to 6.35E-1, Cd 2.52E-2 to 1.65E-1, respectively, for children. HRI of less than 1 indicates a relative absence of health risks associated with the ingestion of contaminated vegetables. In addition, it was apparent that intake of a single metal through consumption of edible parts of vegetables posed a relatively higher potential health risks to children as compared with adults. Hence, consumption of these contaminated vegetables, e.g., *S. oleracea* L. and *B. pekinensis* L. for children is likely to induce health problems with special reference to Zn and Cd in the vegetables.

Discussion

It has often been observed that physicochemical properties of soils were changed after long-term

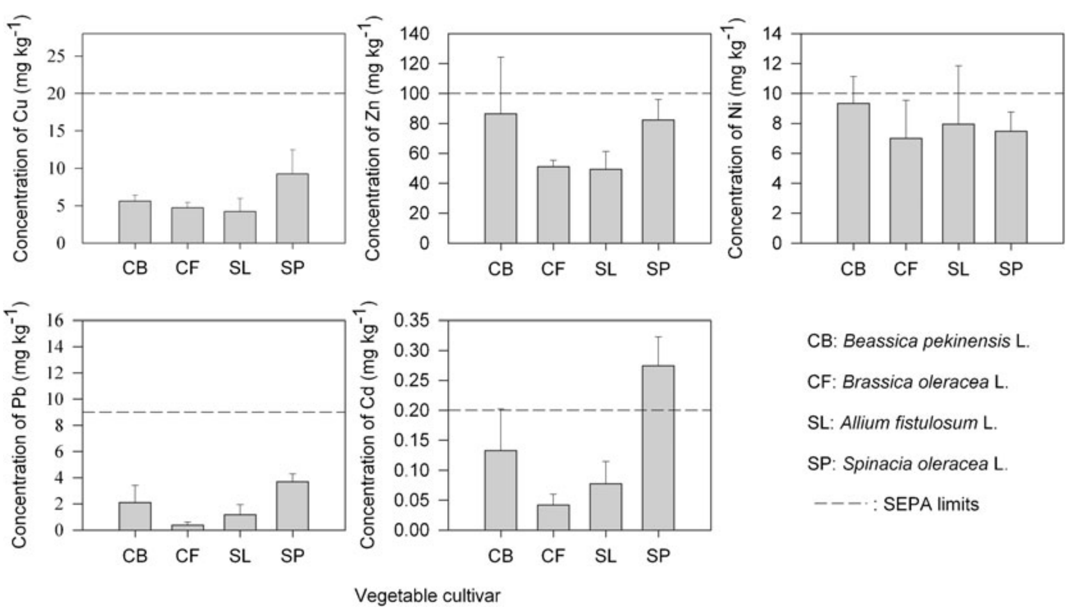

Fig. 3 Heavy metal concentrations (on dry weight basis) in the edible parts of vegetables cultivated in sewage-irrigated soils

Table 3 Transfer factors (TF) of heavy metals for vegetables grown on sewage-irrigated soils (on dry weight basis)

Transfer factor	*Beassica pekinensis* L.		*Brassica oleracea* L.		*Allium fistulosum* L.		*Spinacia oleracea L.*	
	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD
TF_{Cu}	0.13–0.25	0.18±0.05	0.10–0.15	0.12±0.03	0.06–0.12	0.10±0.03	0.13–0.36	0.25±0.11
TF_{Zn}	0.35–0.66	0.52±0.13	0.33–0.43	0.38±0.05	0.27–0.36	0.32±0.05	0.44–0.78	0.67±0.20
TF_{Ni}	0.19–0.50	0.32±0.13	0.17–0.33	0.23±0.08	0.18–0.60	0.33±0.23	0.23–0.28	0.26±0.03
TF_{Pb}	0.01–0.11	0.07±0.05	0.002–0.016	0.009±0.006	0.01–0.07	0.04±0.02	0.07–0.10	0.08±0.02
TF_{Cd}	0.30–0.78	0.57±0.20	0.16–0.26	0.20±0.06	0.28–0.49	0.36±0.11	1.30–1.37	1.34±0.04

wastewater application. The results from this study showed that sewage irrigation significantly reduced the pH values of soils. However, the result from this study is inconsistent with the data reported by Mapanda et al. (2005). This may be related to the different buffer capacity of soils, which depends on the clay and organic matter content among other soil properties (Shomar et al. 2005). Moreover, sewage irrigation provides a valuable source of essential nutritional elements, such as N, P, and organic matter for agricultural soils, and this has been shown in the results here (Table 1) and other studies (Siebe 1998; Mapanda et al. 2005). The increase in organic matter, total nitrogen, and available phosphorus after application of wastewater would be beneficial for soil fertility, but the associated decline in pH values of the soils may affect the mobility and bioavailability of heavy metals and/or loss of nutrients.

For the sewage-irrigated soils, greater amounts of heavy metals were observed, especially for Cd, Zn, Pb, Cr, and Hg (Liu et al. 2005; Singh and Kumar 2006). The distribution of metals in cultivated soils was not only affected by physiochemical properties of soils, but also by irrigation times (Mapanda et al. 2005; Lucho-Contantino et al. 2005; Sun et al. 2008). In this study area, examined vegetable field had been irrigated by sewage for more than 30 years until 2003 (Wu et al. 2003), which showed the higher level of contamination by heavy metals compared to reference soils (Table 2; Fig. 2). Similar results of contamination by heavy metals derived from sewage-irrigation were also found in the previous studies (Xie et al.

Table 4 DIM and HRI (on dry weight basis) for individual heavy metals caused by the consumption of selected vegetables grown on sewage-irrigated soils

Vegetables	Individuals		Cu	Zn	Ni	Pb	Cd
Beassica pekinensis L.	Adults	DIM	2.94E-3	4.54E-2	4.91E-3	1.10E-3	6.97E-5
		HRI	7.34E-2	1.51E-1	2.45E-1	3.15E-1	6.97E-2
	Children	DIM	3.38E-3	5.21E-2	5.64E-3	1.27E-3	8.01E-5
		HRI	8.44E-2	1.74E-1	2.82E-1	3.62E-1	8.01E-2
Brassica oleracea L.	Adults	DIM	2.48E-3	2.68E-2	3.68E-3	1.98E-4	2.19E-5
		HRI	6.19E-2	8.92E-2	1.84E-1	5.65E-2	2.19E-2
	Children	DIM	2.85E-3	3.08E-2	4.23E-3	2.27E-4	2.52E-5
		HRI	7.12E-2	1.03E-1	2.11E-1	6.50E-2	2.52E-2
Allium fistulosum L.	Adults	DIM	2.21E-3	2.59E-2	4.17E-3	6.15E-4	4.07E-5
		HRI	5.52E-2	8.63E-2	2.08E-1	1.76E-1	4.07E-2
	Children	DIM	2.54E-3	2.98E-2	4.79E-3	7.07E-4	4.68E-5
		HRI	6.34E-2	9.92E-2	2.40E-1	2.02E-1	4.68E-2
Spinacia oleracea L.	Adults	DIM	4.84E-3	4.32E-2	3.93E-3	1.93E-3	1.44E-4
		HRI	1.21E-1	1.44E-1	1.96E-1	5.52E-1	1.44E-1
	Children	DIM	5.57E-3	4.97E-2	4.52E-3	2.22E-3	1.65E-4
		HRI	1.39E-1	1.66E-1	2.26E-1	6.35E-1	1.65E-1

2002; Wang 2005). Wang et al. (2007) reported that the quality of sewage used for irrigation was in the middle to serious pollution level by Cd, Pb, Cu, Zn, and As in Baoding. Moreover, the concentrations and estimated annual loading rates of Cd and Zn in wastewater significantly exceeded the limits of irrigated water for vegetables and UK annual loading rates, respectively (Table 2). High concentrations of Zn, Cu, Pb, and Cd in wastewater may come from chemical fiber plants, plating factories, paper mills, and battery industries located in the west of Baoding City. The application of unacceptable quality of wastewater was the major cause for the buildup of heavy metals in sewage-irrigated soils in this study area. Aydinalp et al. (2005) showed that water being extensively used for irrigation in the Bursa plain of Turkey was seriously polluted by industrial wastewater and then its use for irrigation caused soil pollution with heavy metals. The heavy metals were of little concern for irrigation when the wastewater from industrial sewage was treated (Toze 2004). However, the quantity of sewage generated from domestic and industrial activities was many times more than the design capacity of the sewage treatment plant due to the rapid development of Baoding. This resulted in a high percentage of raw sewage directly entering the receptor of Fu River, and then the drainage water was pumped from the river for irrigation during the agricultural production season. Consequently, the periodic monitoring of water and soil quality, together with taking into account of local soil texture and agricultural practices, was required to prevent potential health hazards of long-term irrigation with wastewater for a given set of local conditions.

In general, the level of metal contamination in leafy vegetables was higher than in non-leafy ones. The results of our study showed that the edible parts of spinach (*S. oleracea* L.) and cabbage (*B. pekinensis* L.) accumulated a higher amount of Cu, Pb, Cd, Zn, and Ni, while a lower amount of heavy metals for cauliflower (*B. oleracea* L.) and scallion (*A. fistulosum* L.) was recorded in the sewage-irrigated soils (Fig. 3). The difference in the level of heavy metal contamination among vegetable species was due to their physiological differences in terms of heavy metal uptake, exclusion, accumulation, as well as foliage deposition and retention efficiency (Carlton-Smith and Devis 1983). In our study, with the exception of *B. oleracea* L., the other vegetables were heavily contaminated with elemental Cd, Zn, and/or Ni in sewage-irrigated soils (Fig. 3). The significant contamination of leafy vegetables was observed in Beijing, where the leafy vegetables contaminated with Cd, Cr, and Ni were well excess of the permissible limits set by SEPA and WHO in sewage-irrigated soils (Khan et al. 2008). On one hand, the heavily contaminated vegetables by Cd, Cr, and Ni were related to their greater TF (Liu et al. 2005; Rattan et al. 2005). On the other hand, the foliar deposition of leafy vegetables contaminated with heavy metals was another reason (Singh and Kumar 2006). The traffic and industrial activities have been increasing in urban and suburban areas of Beijing, resulting in greater deposition of heavy metals on vegetables. This led to a greater contamination of leafy vegetables by heavy metals in sewage-irrigated soils of Beijing. According to the results, the non-leafy vegetables or vegetables with a lower efficiency of metal uptake (e.g., *B. oleracea* L.) from soil should be cultivated in sewage-irrigated soils to reduce health risks of heavy metals. In other words, restrictive measures for cultivation of vegetables may be taken on long-term sewage-irrigated soils. Furthermore, the uncontaminated vegetables cultivated in gardens and in groundwater-irrigated soil, which were practiced by half of households surrounding Jiaozhuang, Suncun, and Wangting villages and is situated away from Fu River, respectively (Fig. 1), may be consumed to abate the health risks.

With the contamination of vegetables grown in wastewate-irrigated soils, there has been an increase in public awareness of the health hazards posed by heavy metals derived from consumption of contaminated vegetables (Liu et al. 2005; Sharma et al. 2007; Arora et al. 2008; Khan et al. 2008; Chary et al. 2008), because heavy metals can accumulate in human bodies producing toxic, neurotoxic, carcinogenic, mutagenic, or teratogenic illness (Duruibe et al. 2007). The greater concentrations of metals in vegetables with higher HQ were stated by Agrawal (1999). Then, the HQ was also used as a valuable data for evaluation of health risks associated with the consumption of metal-contaminated crops grown in sewage-irrigated soils (Rattan et al. 2005; Chary et al. 2008). In the studied area, the values of HRI for all examined vegetables were less than 1 (Table 4), which indicated that there was a relative absence of health risks associated with the ingestion of contaminated vegetables grown in sewage-irrigated soils. The result is in concurrence with the conclusions of Khan et al. (2008). However, the greater HQ values (more than 1) of vegetables (e.g., *S. oleracea* L.) for Cd, Zn, and Cr were observed by Qishlaqi et al. (2008) and Chary et al. (2008). The difference between HQ and/or HRI may be related to the estimated reference oral doses (RfD) (US-EPA, IRIS 2002). Although the overall confidence in this RfD assessment varied from low for Cd to high for Zn, the RfD using the calculation of HRI or HQ by consumption of vegetable must be high because the vegetable is a component of daily intake foodstuffs via food chain. Other food such as grains, fruit, meat, eggs, and milk all play important roles in metal exposures to health hazard. In addition, the absence of health risks for children was lower than for adults, especially consumption of leafy vegetables (e.g., *S. oleracea* L. and *B. pekinensis* L.) (Table 4). However, there are some other sources of metal exposure for children, like dermal contact, dust inhalation, and ingestion of metal-contaminated soils, which were pointed out and were not involved in their studies (Khan et al. 2008; Arora et al. 2008). Therefore, further detailed studies are required to completely understand the health risks for children.

Conclusions

Compared with groundwater irrigation, the long-term wastewater irrigation has led to moderate accumulation of Cu and Ni and significant buildup of Zn, Cd, and Pb accompanied with an enrichment of organic matter and some nutrients in suburban soils of Baoding City. Moreover, the leafy vegetables were contaminated heavily by Zn, Cd, and/or Ni and were not safe for human consumption. However, HRI values of less than 1 indicated a relative absence of health risks associated with ingestion of the contaminated vegetables.

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