

Accumulation of heavy metals in the vegetables grown in wastewater irrigated areas of Dehradun, India with reference to human health risk

A. K. Chopra · Chakresh Pathak

Received: 31 December 2014 / Accepted: 1 June 2015 / Published online: 20 June 2015 © Springer International Publishing Switzerland 2015

Abstract The present study on accumulation of heavy metals in the vegetables viz. Beta vulgaris, Phaseolus vulgaris, Spinacea oleracea, and Brassica oleracea var. botrytis grown in the wastewater-irrigated soil near the Bindal river. Dehradun, has shown the maximum accumulation of metals for Pb (196.91 \pm 8.13 mg/kg), Cu $(36.75 \pm 6.19 \text{ mg/kg})$, Zn $(305.54 \pm 14.30 \text{ mg/kg})$, Ni $(125.48 \pm 5.97 \text{ mg/kg})$, Cd $(29.58 \pm 4.26 \text{ mg/kg})$, and Cr $(93.06 \pm 3.25 \text{ mg/kg})$ in agricultural soil irrigated with wastewater. The enrichment factor of soil was maximum for Cr(8.74) and minimum for Cu(0.88). In case of vegetables, the concentrations of heavy metals were maximum for Pb (86.69 ± 6.69) in the flower of B. oleracea var. botrytis, Cu (33.49 \pm 2.09) and Zn (161.86 ± 17.79) in the leaves of S. oleracea, Ni (80.72 ± 8.40) and Cd (23.19 ± 2.76) , and Cr (57.18 ± 8.16) in the root of *B. vulgaris* grown in wastewater (WW)-irrigated soil. The bioaccumulation factor (BAF) for Cu (0.911) was maximum in S. oleracea and minimum for Pb (0.440) in B. vulgaris. The maximum daily intake of metals was found for Zn (0.059) in S. oleracea and minimum for Cd (0.008) in B. vulgaris. The human health risk index was found to be more than 1 for Pb and Cd. The long-term wastewater irrigation resulted in accumulation of heavy metals in vegetables which may cause potential health

A. K. Chopra $(\boxtimes) \cdot C$. Pathak (\boxtimes)

Department of Zoology and Environmental Science,

Gurukula Kangri University, Haridwar, Uttarakhand 249404, India

e-mail: profakchopra@yahoo.co.in

e-mail: chakreshpathak@yahoo.co.in

risks to consumers as these vegetables are sold in local markets of Dehradun city.

Keywords Heavy metals · Enrichment factor · Bioaccumulation factors · Daily intake of metals · Health risk index

Introduction

Heavy metals are ubiquitous in the environment, and as a result of both natural and anthropogenic activities, humans may be exposed to them through various pathways, especially food chain. Wastewater irrigation, solid waste disposal, sludge application, vehicular exhaust, and industrial activities are the major sources of soil contamination with heavy metals. An increased metal uptake by food crops, vegetables, and fruits grown on such contaminated soils is often observed worldwide (Harmanescu et al. 2011). The occurrences of heavy metal-enriched ecosystem components arise from rapid industrial growth, advances in agricultural chemicalization and/or the urban activities of human beings. This has led to metal dispersion in the environment and consequently impaired health of the population by the ingestion of victuals contaminated by harmful elements (Orisakwe et al. 2012).

Many industrial plants in developing countries operate without any or a nominal wastewater treatment and routinely discharge their waste into drains that either contaminate rivers and streams or add to the contaminant load of sewage sludge. The contaminants may enter the food chain in addition to the low water quality of the area. The heavy metal contamination of agricultural soils and crops is causing concerns due to the probable effects on food production and human health in affected areas (Hassan et al. 2013). Reuse of domestic and industrial wastewater in agriculture for irrigating crops appears to be a lucrative option.

The concentration of metals in vegetables mainly depends on the texture of soil or media on which they are grown but this also depends on the type and nature of plants being grown on these contaminated soils (Kabata-Pendias and Pendias 1984; Khan et al. 2009). Heavy metals are often toxic if accumulated beyond the permissible limits of BIS standards. Pb and Cd are known to be the most toxic and are the most abundant metals in food (Khan et al. 2009; Korfali et al. 2013). A number of serious health problems can develop as a result of excessive uptake of dietary heavy metals. Furthermore, the consumption of heavy metalcontaminated food can seriously deplete some essential nutrients in the body causing a decrease in immunological defenses, intrauterine growth retardation, impaired psycho-social behaviors, disabilities associated with malnutrition, and a high prevalence of upper gastrointestinal cancer (Orisakwe et al. 2012).

Food safety is a major public concern worldwide. The increasing demands for food and food safety have drawn the attention of researchers to the risks associated with consumption of contaminated foodstuffs, i.e., pesticides, heavy metals, and/or toxins in vegetables (Khan et al. 2009). Thus, the present study was carried out to assess the concentration of metals viz. Pb, Cu, Zn, Ni, Cd, and Cr in the vegetables viz. *Beta vulgaris*, *Phaseolus vulgaris*, *Spinacea oleracea*, and *Brassica oleracea* var. *botrytis* grown in wastewater (WW)-irrigated area close to the Bindal river, Dehradun.

Materials and methods

Study area and sampling Dehradun city, also called Doon valley, in Uttarakhand state of India is located on 30°19' north latitude and 78° 04' east longitude. The long-term WW-irrigated area (site I) near the Bindal river situated at Haridwar bypass road, Dehradun, was selected for the present study. For the last 15–20 years, WW being drained from the Bindal river is being used for irrigation of various vegetables crops like *B. vulgaris* (beet), *P. vulgaris* (French bean), *S. oleracea* (spinach),

B. oleracea var. *botrytis* (cauliflower). The WW, soil, and vegetables were collected from three different plots of the nearby Bindal river area. Tube-well water (TWW)-irrigated area of Gulerghati about 10 km away from the study area was taken as control site (site II) where the crops like *Raphnus sativus*, *P. vulgaris*, and *S. olercea* were being grown (Fig. 1). The study was made in the summer season of the year 2009.

Sample preparation The samples of vegetable crops were washed with double-distilled water to remove airborne pollutants and dust particles. The edible parts (flower, leaves, and root) of the samples were air-dried to constant weight to remove water. All samples were then oven-dried at 70–80 °C for 24 h to remove remaining moisture. Samples were then grounded into powder state and stored into polythene bags for further analytical use. The soil samples of each site were composited separately for analysis. WW and TWW samples were collected in 1-1 plastic bottles, immediately acidified with HNO₃ (2 ml), and transported to the laboratory where samples were filtered through Whatman 42 filter paper and stored in a refrigerator at 4 °C.

Digestion of samples The WW, TWW, soil, and vegetable samples were digested with a mixture of HNO_3 and $HClO_4$ acid (2:1, v/v) individuals per the method described in AOAC (1990) for wet ashing. WW (10 ml) and 0.5 g of powdered samples of vegetables and soil were taken using indigestion tubes separately, and then nitric acid and perchloric acid were added to each. The digestions were completed on digestion blocks (Make FOSS) following standard procedures as described by Chaturvedi and Sankar (2006). After digestion, all the samples were diluted, filtered through Whatman 42 filter paper, and volume was made up to 50 ml in volumetric flasks.

Analysis of metals Heavy metals such as Pb, Ni, Zn, Cd, Cu, and Cr in the digested aliquot were determined by atomic absorption spectrophotometer (AAS) (ECIL, model no. 4129). Appropriate quality procedures and precautions were carried out to assure the reliability of the results. Quality control measures were taken to assess contamination and reliability of data, the variations were found to be less than 10 %.



Fig. 1. Study area at Dehradun: site I wastewater (WW) and site II tube-well water (TWW) irrigated

Data analysis

Enrichment factor Enrichment factor (EF) was used to assess the contamination degree of heavy metals in the soil and was calculated following the formula given by Kim and Kim (1999).

Bioaccumulation factors Bioaccumulation factor (BAF), an index of the ability of the uptake of particular metal from soil to root then upper part of vegetable crops, was calculated as per Zhuang et al. 2009.

Daily intake of metals and health risk index The daily intake of heavy metals was calculated as per Arora et al. (2008). The average body weights were considered to be 65 kg, while average daily vegetable intake was considered to be 0.300 kg/person/day as reported by Arora

et al. (2008) and Singh et al. (2010). Health risk index (HRI) was calculated as the ratio of estimated exposure of vegetables and oral reference dose (Cui et al. 2004; Singh et al. 2010). An HRI value of greater than 1 (HRI > 1) was considered as not safe for human health (USEPA 2002) and is likely to cause health hazards in consumers.

Results and discussion

Metal concentrations in wastewater and tube-well water The concentrations of heavy metals in WW and TWW used for irrigation of soil and vegetable crops are summarized in Table 1. The concentrations of the heavy metals (Pb 0.88 ± 0.13 mg/l, Cu 0.45 ± 0.09 mg/l, Zn 0.83 ± 0.11 mg/l, Ni 0.94 ± 0.09 mg/l, Cd

 Table 1
 Heavy metal contents (mg/l) in WW (site I) and TWW (site II) used for irrigation

Heavy metals	Site I	Site II	Permissible limits
Pb	$0.88 \pm 0.13^{**}$	0.11 ± 0.01	0.5
Cu	$0.45 \pm 0.09^{**}$	0.04 ± 0.01	0.2
Zn	$0.83 \pm 0.11^{**}$	0.17 ± 0.02	2.0
Ni	$0.94 \pm 0.09^{**}$	0.13 ± 0.03	0.2
Cd	$0.13\pm0.04*$	0.04 ± 0.01	0.01
Cr	$0.58 \pm 0.09^{**}$	0.09 ± 0.01	0.1

Significance: p < 0.05(*) and p < 0.01(**); permissible limits, Pescod (1992)

 0.13 ± 0.04 mg/l, and Cr 0.58 ± 0.09 mg/l) in WW used for irrigation in nearby areas of the Bindal river were observed to be above the standard levels proposed by Pescod (1992), while the Zn concentration was found to be within safe limit. The paired two-sample t test showed that there was a significant level of Pb, Cu, Zn, Ni, Cr (P < 0.01), and Cd (P < 0.05) concentrations in WW as compared to TWW. The values of the metal concentrations of WW of the Bindal river area were higher than the values observed by Sharma et al. (2007) for Pb (0.08–0.10 mg/l), Cu (0.04–0.11 mg/l), Zn (0.09-0.23 mg/l), Ni (0.03-0.05 mg/l), Cd (0.01-0.02 mg/l), and Cr (0.03–0.09 mg/l) at different sites in WW used for irrigation in suburban region of Varanasi, India, and also by Chopra and Pathak (2012) for Pb $(0.36 \pm 0.06 \text{ mg/l})$, Cu $(0.15 \pm 0.01 \text{ mg/l})$, Zn $(0.55 \pm 0.02 \text{ mg/l})$, Ni $(0.32 \pm 0.01 \text{ mg/l})$, Cd $(0.03 \pm 0.00 \text{ mg/l})$, and Cr $(1.37 \pm 0.09 \text{ mg/l})$ in WW used for irrigation near the Bindal river, Dehradun, during winter season 2008. However, the values of Pb, Cu, Zn, Ni, Cd, and Cr were lower than that of Cu $(2.17 \pm 0.46 \text{ mg/l})$ and Zn $(0.95 \pm 0.40 \text{ mg/l})$ as reported by Ahmad and Goni (2010) in effluent-contaminated waters used for irrigation in the industrial areas of Dhaka, Bangladesh, and also for Pb $(4.26 \pm 2.20 \text{ mg/l})$, Zn $(1.91 \pm 1.45 \text{ mg/l})$, and Cu $(1.56 \pm 1.22 \text{ mg/l})$ in WW used for irrigation in periurban region of Titagarh, India (Gupta et al. 2008).

Metal concentrations in soil The concentrations of Pb, Cu, Zn, Ni, Cd, and Cr in the agricultural soils collected from the Bindal river area were found to be $196.91 \pm 8.13, 36.75 \pm 6.19, 305.54 \pm 14.30,$ 125.48 ± 5.97 , 29.58 ± 4.26 , and 93.06 ± 3.25 mg/kg, respectively, which were below the permissible limits of Indian standards except that of Cd. However, these values of Zn, Ni, and Cd were above the permissible limits of European Union Standards (EU 2006). There was a notable increase of Pb (+116.03 %), Cu (+115.54 %), Zn (+263.83 %), Ni (+264.66 %), Cd (+168.42 %), and Cr (+230.35 %) in WW-irrigated soil in comparison to TWW-irrigated soil. The paired twosample t test for the metals showed that the concentration of Pb, Zn, Ni, Cr (P < 0.001), Cu (P < 0.05), and Cd (P < 0.01) were significantly higher in WW-irrigated soil than that observed in TWW soil (Table 2).

The concentrations of heavy metals in the soil of the study area were higher than those reported by Ahmed

Heavy metals	Site I	Site II	EF	Limits ^a	Limits ^b
Pb	$196.91 \pm 8.13^{***}$ (+116.03 %)	91.15 ± 11.61	2.16	300	250-500
Cu	$36.75 \pm 6.19*$ (+115.54 %)	17.05 ± 1.41	2.16	140	135–270
Zn	$305.54 \pm 14.29^{***}$ (+263.83 %)	83.98 ± 5.25	3.64	300	300-600
Ni	$125.48 \pm 5.97^{***}$ (+264.66 %)	34.41 ± 4.69	3.65	75	75–150
Cd	$29.58 \pm 4.26^{**}$ (+168.42 %)	11.02 ± 1.76	2.69	3.0	3–6
Cr	93.06 ± 3.25*** (+230.35 %)	28.17 ± 1.68	3.30	150	NA

Table 2 Heavy metals contents (mg/kg) in WW-irrigated soil (site I) and TWW-irrigated soil (site II)

% increase/decrease in comparison to control site. Limits, (a) European Union Standards (EU 2006), (b) Indian Standard (Awashthi 2000) *EF* enrichment factor

Significance: *p* < 0.05(*), *p* < 0.01(**), and *p* < 0.001(***)

Table 3	Heavy metals content	s (mg/kg) in vege	tables grown in WW	(site I) and TWW	(site II) irrigated soil
---------	----------------------	-------------------	--------------------	------------------	--------------------------

Vegetables	Dry wt.%	Pb	Cu	Zn	Ni	Cd	Cr
WW (site I)							
B. vulgaris	21.75	49.54 ± 6.26	26.78 ± 2.91	96.43 ± 0.60	80.72 ± 8.40	23.19 ± 2.76	57.18 ± 8.16
P. vulgaris	14.85	59.39 ± 2.21	23.65 ± 2.02	72.09 ± 12.99	66.57 ± 8.08	19.48 ± 2.68	15.3 ± 3.56
S. oleracea	12.96	68.17 ± 4.96	33.49 ± 2.09	161.86 ± 17.79	65.69 ± 3.72	21.35 ± 2.12	55.74 ± 5.55
B. oleracea var. botrytis	20.78	86.69 ± 6.69	29.35 ± 1.39	48.12 ± 5.83	68.95 ± 7.55	17.51 ± 3.71	46.19 ± 6.86
TWW (site II)							
R. sativus	7.80	45.18 ± 3.11	17.54 ± 1.18	59.41 ± 8.03	66.53 ± 3.03	9.07 ± 0.73	19.11 ± 2.86
P. vulgaris	17.48	33.59 ± 3.58	12.44 ± 0.63	47.91 ± 4.38	35.87 ± 6.62	6.96 ± 1.39	30.07 ± 2.99
S. oleracea	12.78	29.69 ± 1.95	13.01 ± 0.72	57.54 ± 8.24	48.62 ± 4.62	12.69 ± 1.74	24.61 ± 8.70

and Goni (2010) for Pb (49.71 \pm 3.32 mg/kg), Zn (115.43 \pm 7.75 mg/kg), Ni (58.16 \pm 18.15 mg/kg), Cd (11.42 \pm 4.63 mg/kg), and Cr (53.70 \pm 12.37 mg/kg) except Cu (39.14 \pm 5.36 mg/kg) in WW-irrigated soil in industrial areas of Bangladesh, Chopra, and Pathak (2012) for Pb (70.49 \pm 10.54 mg/kg), Zn (244.15 \pm 7.19 mg/kg), Ni (27.77 \pm 1.50 mg/kg), and Cd (17.47 \pm 1.20 mg/kg) except Cu (54.48 \pm 9.63 mg/kg) and Cr (129.52 \pm 9.71 mg/kg) in WW-irrigated soil near the Bindal river, Dehradun, India, during winter season in the year 2008 and lower than the metal concentrations of Pb (278 mg/kg), Cu (502 mg/kg), and Zn (498 mg/kg) in paddy soils of Dabaoshan mine in Guangdong, China, reported by Zhuang et al. (2009).

The EF for the WW-irrigated soil was maximum for Ni (3.65) and minimum for Pb and Cu (2.16) (Table 2). The present EF values for Pb (2.16), Cu (2.16), Zn (3.64), Ni (3.65), Cd (2.69), and Cr (3.30) were found to be lower than the values for Zn (4.8), Cu (6.5), and Pb (15.5) observed by Mishra and Tripathi (2008) in WWirrigated soil of Varanasi, than the Cu (9.62) and Zn (7.96) reported by Pathak et al. (2011) in sewageirrigated soil, than the Zn (4.09), and Cr (7.09) reported by Chopra and Pathak (2012) in WW-irrigated soil during winter season in the year of 2008. The present EF values for contamination of Pb, Cu, Zn, Ni Cd, and Cr were in moderate enrichment category when compared with contamination categories established by Sutherland (2000).

Heavy metal concentrations in vegetables The average concentrations of heavy metals (mg/kg, on dry weight basis) in the selected vegetables (edible parts) grown in the nearby Bindal river are given in Table 3. The concentrations of heavy metals were maximum for Pb (86.69 ± 6.69) in flower of *B. oleracea* var. *botrytis*, for Cu (33.49 ± 2.09) and Zn (161.86 ± 17.79) in leaves of S. oleracea and for Ni (80.72 ± 8.40), and Cd (23.19 ± 2.76) and Cr (57.18 ± 8.16) in root of B. vulgaris grown in WW irrigated soil. It was noticeably observed that the concentrations of all the heavy metals were higher in WW-irrigated vegetables than TWW-irrigated. The levels of Pb, Cu, Zn, Ni, Cd, and Cr except that of Cu in these vegetables were above the prescribed safe values proposed by Prevention of Food Adulteration Act and rules (PFA) (Awashthi 2000).

In the present study, the concentration of metals (Pb, Cu, Zn, Ni, Cd, and Cr) in vegetables grown in WWirrigated soil were higher than that reported by Gupta et al. (2008) for Pb (57.63 mg/kg), Zn (139.05 mg/kg),

Table 4 Bioaccumulation factor (BAF) for vegetables grown in WW-irrigated soil

Vegetables Pb Cu Zn Ni Cd Cu Beta vulgaris 0.252 0.729 0.316 0.643 0.784 0.4 Phaseolus vulgaris 0.302 0.644 0.236 0.531 0.659 0.							
Beta vulgaris 0.252 0.729 0.316 0.643 0.784 0. Phaseolus vulgaris 0.302 0.644 0.236 0.531 0.659 0.	Vegetables	Pb	Cu	Zn	Ni	Cd	Cr
Phaseolus vulgaris 0.302 0.644 0.236 0.531 0.659 0.	Beta vulgaris	0.252	0.729	0.316	0.643	0.784	0.614
	Phaseolus vulgaris	0.302	0.644	0.236	0.531	0.659	0.164
<i>Spinaceaoleracea</i> 0.346 0.911 0.530 0.524 0.722 0.	Spinaceaoleracea	0.346	0.911	0.530	0.524	0.722	0.599
Brassica oleracea var. botrytis 0.440 0.799 0.157 0.549 0.592 0.592	Brassica oleracea var. botrytis	0.440	0.799	0.157	0.549	0.592	0.496

Vegetables	Pb	Cu	Zn	Ni	Cd	Cr
Beta vulgaris	0.019	0.011	0.038	0.032	0.009	0.022
Phaseolus vulgaris	0.023	0.009	0.028	0.026	0.008	0.006
Spinaceaoleracea	0.027	0.013	0.063	0.026	0.008	0.022
Brassica oleracea var. botrytis	0.034	0.012	0.019	0.027	0.007	0.018

Table 5 Daily intake of metals (DIM) via consumption of vegetables grown in WW-irrigated sites

Cd (17.79 mg/kg), Cu (28.08 mg/kg), and Ni (62.70 mg/kg) except Cr (78.02 mg/kg) in radish, Pb (49.79 mg/kg), Zn (154.21 mg/kg), Cd (14.58 mg/kg), Cu (34.49 mg/kg), and Ni (69.22 mg/kg) except Cr (96.30 mg/kg) in spinach, and Pb (31.04 mg/kg), Zn (96.48 mg/kg), Cd (13.80 mg/kg), Cu (15.66 mg/kg), and Ni (59.28 mg/kg) except Cr (86.83 mg/kg) in cauliflower grown in WW-irrigated soil at Tetagarh, West Bengal, and were also higher than the values reported by Chopra and Pathak (2012) for Pb ($22.42 \pm 2.59 \text{ mg/kg}$), Cu $(9.17 \pm 2.78 \text{ mg/kg})$, Zn $(31.38 \pm 3.54 \text{ mg/kg})$, and Cd $(15.79 \pm 3.09 \text{ mg/kg})$ except Ni $(128.21 \pm 19.85 \text{ mg/kg})$ and Cr $(105.43 \pm 8.50 \text{ mg/kg})$ in R. sativus, for Pb (10.44 \pm 1.85 mg/kg), Cu $(11.61 \pm 2.71 \text{ mg/kg})$, Zn $(30.58 \pm 4.39 \text{ mg/kg})$, Cd $(16.73 \pm 1.19 \text{ mg/kg})$, and Cr $(54.86 \pm 6.37 \text{ mg/kg})$ except Ni (156.33 \pm 9.02 mg/kg) in *P. vulgaris*, for Pb $(30.76 \pm 1.77 \text{ mg/kg})$, Cu $(11.26 \pm 0.81 \text{ mg/kg})$, Zn $(47.51 \pm 19.64 \text{ mg/kg})$, Cd $(7.59 \pm 2.10 \text{ mg/kg})$ except Ni $(147.34 \pm 19.02 \text{ mg/kg})$ and Cr $(102.88 \pm 16.01 \text{ mg/kg})$ in *B. juncea*, for Pb $(12.07 \pm 2.16 \text{ mg/kg})$, Cu $(10.85 \pm 2.34 \text{ mg/kg})$, Zn $(31.97 \pm 3.04 \text{ mg/kg})$, and Cd $(16.89 \pm 4.12 \text{ mg/kg})$ except Ni $(123.30 \pm 17.28 \text{ mg/kg})$ and Cr $(85.73 \pm 9.89 \text{ mg/kg})$ in *B. oleracea* var. *botrytis* grown in WW-irrigated soil near the Bindal river, Dehradun, during winter season in the year of 2008.

Bioaccumulation factors from soil to food crops The BAF for vegetables grown in WW- and TWW- irrigated soil are given in Table 4. According to Cluis

(2004), hyperaccumulating plants have a BAF more than 1.0. Mishra et al. (2009) stated that BAF values differ significantly among locations and the plant species. The difference in BAF between locations may be related to soil nutrient management and soil properties. The plants differ in the uptake of heavy metals due to different binding capacities of the soils for these metals, also to plant root and metal interactions which may vary with the type of metals (Korboulewsky et al. 2002). Bioavailability of many metallic elements increases when these become associated with labile or soluble organic compounds (Antoniadis and Alloway 2002).

In the present study, the maximum BAF value was for Pb (0.440), Ni (0.643), Cd (0.784), and Cr (0.614) in *B. vulgaris*, Cu (0.911) and Zn (0.530) in *S. oleracea* grown on WW-irrigated soil. The BAF values were less than 1, thereby showing that the plants absorbed heavy metals but these were not accumulated. In case of control site, the BAF value was observed to be 0.496 for Pb, 1.029 for Cu, 0.707 for Zn, 1.934 for Ni, 2.639 for Cd in *R. sativus*, and Cr (1.068) in *P. vulgaris* grown on TWW-irrigated soil. The BAF value is greater than 1 which showed that higher efficient accumulation of these metals from soil to the edible parts of these vegetables.

Daily intake of metals and health risk assessment The daily intake of metals (DIMs) of Pb, Cu, Zn, Ni, Cd, and Cr through consumption of vegetables are given in Table 5. To calculate approximately health risk, estimation of DIM of metals and risk index were

Table 6 Health risk index (HRI) of heavy metals via consumption of vegetables grown in WW-irrigated sites

Pb	Cu	Zn	Ni	Cd	Cr
2.330	0.008	0.002	0.871	5.095	0.001
2.164	0.007	0.002	0.808	4.731	0.000
2.674	0.011	0.004	0.859	5.584	0.001
3.401	0.010	0.001	0.902	4.580	0.001
	Pb 2.330 2.164 2.674 3.401	Pb Cu 2.330 0.008 2.164 0.007 2.674 0.011 3.401 0.010	Pb Cu Zn 2.330 0.008 0.002 2.164 0.007 0.002 2.674 0.011 0.004 3.401 0.010 0.001	Pb Cu Zn Ni 2.330 0.008 0.002 0.871 2.164 0.007 0.002 0.808 2.674 0.011 0.004 0.859 3.401 0.010 0.001 0.902	Pb Cu Zn Ni Cd 2.330 0.008 0.002 0.871 5.095 2.164 0.007 0.002 0.808 4.731 2.674 0.011 0.004 0.859 5.584 3.401 0.010 0.001 0.902 4.580

calculated by formula given by Arora et al. (2008) and Cui et al. (2004), respectively. The metals enter human body through two routes inhalation and ingestion of contaminated vegetables. The maximum DIM was found for Pb (0.034) in *B. oleracea* var. *botrytis*, Cu (0.013) and Zn (0.063) in *S. oleracea*, Ni (0.032) and Cd (0.009) in *B. vulgaris*, and Cr (0.022) in *B. vulgaris* and *S. oleracea* grown in WW-irrigated soil.

HRI differed for various metals in different vegetable plants. These were in the order of Cd > Pb > Ni > Cu > Zn > Cr for *B. vulgaris*, Cd > Pb > Ni > Cu > Zn for *P. vulgaris*, Cd > Pb > Ni > Cu > Zn > Cr for S. oleracea, and Cd > Pb > Ni > Cu > Zn = Cr for *B. oleracea* var. botrytis grown at WW-irrigated agricultural soil. The present data revealed that Pb and Cd values greater than 1 in B. vulgaris (Pb 2.330 and Cd 5.095), P. vulgaris (Pb 2.164 and Cd 4.731), S. oleracea (Pb 2.674 and Cd 5.584), and B. oleracea var. botrytis (Pb 3.401 and Cd 4.580) in all vegetables grown in WWirrigated soil (Table 6), which indicated their greater potential to pose health risk to the consumers. Higher HRI for Pb, Cd, Cr, Cu, and Zn were also reported by Singh et al. (2010) in foodstuffs from the WW-irrigated site of a dry tropical area of India. However, consumption of the contaminated vegetables with metals would lead to potential health risks for human especially in children, since HRI for heavy metals due dietary intake were higher than 1. Prolonged consumption of unsafe concentrations of heavy metals through vegetables may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney, and bone diseases. Pb and Cd are considered potential carcinogens and are associated with etiology of a number of diseases especially cardiovascular, kidney, nervous system, blood as well as bone diseases.

Conclusion

This present study concluded that concentrations of Pb, Cu, Ni, Cd, and Cr in the wastewater used for irrigation of vegetable crops were higher than the permissible limits of WW standards used for irrigation set by Pescod (1992). Zn was found to be the only exception to this. The Cd concentration was higher than permissible limits of Indian standards in soil. The EF values for Pb, Cu, Zn, Ni, Cd, and Cr showed moderate enrichment category. The concentrations of metals in edible parts were above the Indian permissible limits in all vegetables grown in WW-irrigated area. The BAF less than 1 for Pb, Cu, Zn, Ni, Cd, and Cr indicated that the *B. vulgaris*, *P. vulgaris*, *S. oleracea*, and *B. oleracea* var. *botrytis* plants only absorbed heavy metals but did not accumulate them. HRI >1 for Pb and Cd suggests that their contamination in the vegetables had potential threat for human health risk from such irrigation practices and thus, there is a need to monitor to avoid any casualty that may happen due to long-term WW irrigation.

Acknowledgments The University Grant Commission, New Delhi, India, is acknowledged for providing the financial support in the form of UGC research fellowship (F.7–70/2007 BSR) to Mr. Chakresh Pathak.

References

- Ahmad JU, Goni MA (2010). Heavy metal contamination in water, soil, and vegetables of the industrial areas in Dhaka, Bangladesh. Environmental Monitoring and Assessment., 166:347–357
- Antoniadis V, Alloway BJ (2002). The role of dissolved organic carbon in the mobility of Cd, Ni and Zn in sewage sludgeamended soils. Environmental Pollution, 117: 515–521. doi: 10.1016/S0269-7491(01)00172–5.
- AOAC (1990). Official methods of analyses. Method no. 975.03 metal in plants, AAS method (15th ed., p. 42). Arlington: Association of Official Analytical Chemists, Inc.
- Arora M, Kiran B, Rani S, Rani A, Kaur B, Mittal N (2008). Heavy metal accumulation in vegetables irrigated with water from different sources. Food Chemistry, 111:811–815
- Awashthi, S. K. (2000). Prevention of Food Adulteration Act No. 37 of 1954. Central and state rules as amended for 1999 (3rd ed.,). New Delhi: Ashoka Law House.
- Chaturvedi RK, Sankar K. (2006). Laboratory manual for the physico-chemical analysis of soil, water and plant. Wildlife Institute of India, Dehradun, pp 97.
- Chopra A. K, Pathak C (2012). Bioaccumulation and translocation efficiency of heavy metals in vegetables grown on long-term wastewater irrigated soil near Bindal river, Dehradun. Agricultural Research. DOI 10.1007/s40003-012-0016-8.
- Cluis C. (2004). Junk-greedy greens: phytoremediation as a new option for soil decontamination. *BioTech Journal*, 2, 60–67.
- Cui YJ, Zhu YG, Zhai RH, Chen DY, Huang YZ, Qui Y, Liang JZ (2004). Transfer of metals from near a smelter in Nanning, China. Environment International. 30: 785–791.
- EU (2006). Commission regulation (EC) No. 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. Official Journal of European UnionL364/5.

- Gupta N., Khan D. K., & Santra S. C. (2008). An assessment of heavy metal contamination in vegetables grown in wastewater-irrigated areas of Titagarh, West Bengal, India. *Bulletin of Environmental Contamination and Toxicology*, 80, 115–118.
- Hassan NU, Mahmood Q, Waseem A, Irshad M, Faridullah AP (2013) Assessment of heavy metals in wheat plants irrigated with contaminated wastewater. Polish Journal of Environmental Studies., 22(1):115–123.
- Harmanescu M, Maria Alda L, Maria Bordean D, Gogoasa I, Gergen I. (2011). Heavy metals health risk assessment for population via consumption of vegetables grown in old mining area; a case study: Banat County, Romania. Chemistry Central Journal, 5:64
- Kabata-Pendias A, Pendias H. (1984). Trace elements in soils and plants. CRC, Press Boca Raton.
- Khan S, Farooq R, Shahbaz S, Aziz Khan M, Sadique M (2009). Health risk assessment of heavy metals for population via consumption of vegetables. World Applied Sciences Journal, 6 (12): 1602-1606.
- Korboulewsky N, Bonin G, Massiani C. (2002). Biological and ecophysiological reactions of white wall rocket (*Diplotaxiserucoides* L.) grown on sewage sludge compost. Environmental Pollution, 17: 365–370. doi:10.1016/S0269-7491(01)00165–8.
- Korfali S. I., Hawi T., & Mroueh M. (2013). Evaluation of heavy metals content in dietary supplements in Lebanon. *Chemistry Central Journal*, 7, 10.
- Kim KH, Kim, SH (1999). Heavy metal pollution of agricultural soils in central regions of Korea. Water, Air, and Soil Pollution, 111:109–122
- Orisakwe O. E., KanayochukwuNduka J., NwadiutoAmadi C., Onyekachi Dike D., & Bede O. (2012). Heavy metals health risk assessment for population via consumption of food crops and fruits in Owerri, South Eastern, Nigeria. *Chemistry Central Journal*, 6, 77.

- Pathak C., Chopra A. K., Kumar V., & Sharma S. (2011). Effect of sewage-water irrigation on physico-chemical parameters with special reference to heavy metals in agricultural soil of Haridwar city. *Journal of Applied and Natural Science*, 3(1), 108–113.
- Pescod MB. (1992). Wastewater treatment and use in agriculture. FAO irrigation and drainage paper 47. Food and Agriculture Organization of the United Nations, Rome
- Mishra V. K., Upadhyay A. R., & Tripathi B. D. (2009). Bioaccumulation of heavy metals and tow organochlorine pesticides (DDT and BHC) in crops irrigated with secondary treated wastewater. *Environmental Monitoring and Assessment*, 156, 99–107.
- Mishra A, Tripathi BD (2008). Heavy metal contamination of soil, and bioaccumulation in vegetables irrigated with treated waste water in the tropical city of Varanasi, India. Toxicological and Environmental Chemistry., 90(5):861–871
- Sharma RK, Agarwal M, Marshall F (2007). Heavy metals contamination of soil and vegetables in suburban areas of Varanasi, India. Ecotoxicology and Environmental Safety, 66: 258–266
- Singh A., Sharma R. K., Agrawal M., & Marshall F. M. (2010). Health risk assessment of heavy metals via dietary intake of foodstuffs from the wastewater irrigated site of a dry tropical area of India. *Food and Chemical Toxicology*, 48, 611–619.
- Sutherland R. A. (2000). Bed sediment-associated trace metals in an urban stream, Oahu, Hawaii. *Environmental Geology*, 39, 611–627.
- USEPA (US Environmental Protection Agency) (2002). Region 9, Preliminary Remediation Goals. http://www.epa.Gov/ region09/waste/sfund/prg.
- Zhuang P, Zou B, Li NY, Li ZA (2009). Heavy metal contamination in soils and food crops around Dabaoshan mine in Guangdong, China: implication for human health. Environmental Geochemistry and Health. doi:10.1007/ s10653-009-9248-3