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Risk assessment of heavy metal toxicity through contaminated vegetables from waste water irrigated area of Varanasi, India

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Abstract: In certain areas of Varanasi city, waste water from Dinapur sewage treatment plant is used for irrigating vegetable plots. We quantified the concentrations of heavy metals, viz. Cd, Cr, Cu, Ni, Pb and Zn in soil, vegetables and the waste water used for irrigation. The waste water used for irrigation had the highest concentration of Zn followed by Pb, Cr, Ni, Cu and Cd. Continuous application of waste water for more than 20 years has led to accumulation of heavy metals in the soil. Consequently, concentrations of Cd, Pb and Ni have crossed the safe limits for human consumption in all the vegetables. Percent contribution of fruit vegetables to daily human intake for Cu, Ni, Pb and Cr was higher than that of leafy vegetables, while the reverse was true for Cd and Zn. Target hazard quotient showed health risk to the local population associated with Cd, Pb and Ni contamination of vegetables. Therefore, to reduce the health risk and the extent of heavy metal contamination, steps must be taken for efficient treatment of sewage. Regular monitoring of heavy metals in the vegetables grown in waste water irrigated areas is also necessary.

Resumen: En ciertas áreas de la ciudad de Varanasi, el agua residual de la planta de tratamiento de drenaje de Dinapur es usada para regar cultivos de verduras. Cuantificamos las concentraciones de metales pesados, i.e. Cd, Cr, Cu, Ni, Pb y Zn en el suelo, las verduras y el agua residual usada para el riego. El agua residual usada para regar tuvo la mayor concentración de Zn, seguida de Pb, Cr, Ni, Cu y Cd. La aplicación continua de aguas de desecho por más de 20 años ha conducido a una acumulación de metales pesados en el suelo, por lo que las concentraciones de Cd, Pb y Ni han rebasado los límites seguros para el consumo humano en todas las verduras. La contribución porcentual de las verduras frutales a la ingestión diaria en los humanos de Cu, Ni, Pb y Cr fue mayor que la de las verduras hojosas, mientras que lo contrario fue cierto para el Cd y el Zn. El Cociente de riesgo para blancos (Target hazard quotient) mostró un riesgo para la salud de la población local asociado con las verduras contaminadas con Cd, Pb y Ni. Por lo tanto, para reducir este riesgo y la magnitud de la contaminación por metales pesados se deben tomar pasos para el tratamiento eficiente del drenaje. Asimismo, es necesario monitorear regularmente los metales pesados en las verduras cultivadas en áreas irrigadas con aguas de desecho.

Resumo: Em certas áreas da cidade de Varanasi, as águas residuais da estação de tratamento de esgotos é utilizada parta a irrigação de hortas. Quantificaram-se as concentrações em metais pesados, i.e. Cd, Cr, Cu, Ni, Pb e Zn no solo, nos vegetais e na água residual usada para irrigação. A água residual usada na irrigação apresentava a maior concentração em Zn seguida pelo Pb, Cr, Ni, Cu e Cd. A aplicação continuada de água residual por mais de 20 anos conduziu a uma acumulação de metais pesados no solo. Consequentemente, as concen-

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trações de Cd, Pb e Ni ultrapassaram os limites de segurança para o consumo humano em todos os vegetais. A contribuição percentual dos frutos para a ingestão humana diária para Cu, Ni, Pb e Cr foi maior do que a das folhas, enquanto o reverso foi verdadeiro para o Cd e o Zn. O quociente do risco projectado mostrou riscos para a saúde da população associados com a contaminação dos vegetais em Cd, Pb e Ni. Assim, para reduzir os riscos para a saúde e a extensão da contaminação em metais pesados, são necessárias medidas para o tratamento eficiente dos esgotos. A monitorização regular dos metais pesados nos vegetais produzidos nas áreas irrigadas com águas residuais é também necessária.

Key words: Contamination, daily intake, heavy metal, health risk, target hazard quotient, waste water.

Introduction

Long term waste water irrigation may lead to the accumulation of heavy metals in agricultural soils and plants. Food safety issues and potential health risks make this as one of the most serious environmental concerns (Cui *et al.* 2004). Vegetables accumulate heavy metals in their edible and non edible parts. Although some of the heavy metals such as Zn, Mn, Ni and Cu act as micro-nutrients at lower concentrations, they become toxic at higher concentrations. Health risk due to heavy metal contamination of soil has been widely reported (Eriyamremu *et al.* 2005; Muchuweti *et al.* 2006; Satarug *et al.* 2000). Crops and vegetables grown in soils contaminated with heavy metals have greater accumulation of heavy metals than those grown in uncontaminated soil (Marshall *et al.* 2007; Sharma *et al.* 2006, 2007). Intake of vegetables is an important path of heavy metal toxicity to human being. Bioavailability of Cd, Cu, Zn and Mn in the human gastrointestinal tract from the edible part of vegetables using an *in vitro* gastrointestinal (GI) extraction technique was assessed by Intawongse & Dean (2006). Lettuce and radish were found to be more responsible than other vegetables for the accumulation of heavy metals in humans through the edible portion (Intawongse & Dean 2006). Absorption capacity of heavy metals depends upon the nature of vegetables and some of them have a greater potential to accumulate higher concentrations of heavy metals than others.

Dietary intake of heavy metals through contaminated vegetables may lead to various chronic diseases. Duruibe *et al.* (2007) suggested that biotoxic effects of heavy metals depend upon the concentrations and oxidation states of heavy metals, kind of sources and mode of deposition. Severe exposure of Cd may result in pulmonary effects such as emphysema, bronchiolitis and alveolitis. Renal effects may also result due to subchronic inhalation of Cd (European Union 2002; Young 2005). Pb toxicity causes reduction in the haemoglobin synthesis, disturbance in the functioning of kidney, joints, reproductive and cardiovascular systems and chronic damage to the central and peripheral nervous systems (Ogwuegbu & Muhanga 2005). Higher concentration of Zn can cause impairment of growth and reproduction (Nolan 2003).

The present work deals with the quantification of heavy metal concentrations in soil and vegetables grown in a suburban area of Varanasi, a medium sized city of India, having long term uses of treated and untreated sewage water for irrigation. Health risk caused by the daily intake of heavy metals through contaminated vegetables was also assessed. In earlier studies at the same area, vegetable samples were collected once in a year to quantify the heavy metal concentrations (Sharma *et al.* 2006; Singh *et al.* 2004), whereas in the present study samplings were done through out the year and health risk was ascertained through calculation of different hazard quotients.

Materials and methods

Experimental site

The experimental site covers the area around Dinapur sewage treatment plant (DSTP). Dinapur, a suburban area in the north east of Varanasi (25º 18' N latitude, 83º 01' E longitude and 76.19 m above the mean sea level) city, is situated in the eastern Gangetic plains of India (Fig. 1). Dinapur sewage treatment plant (DSTP) has a treatment

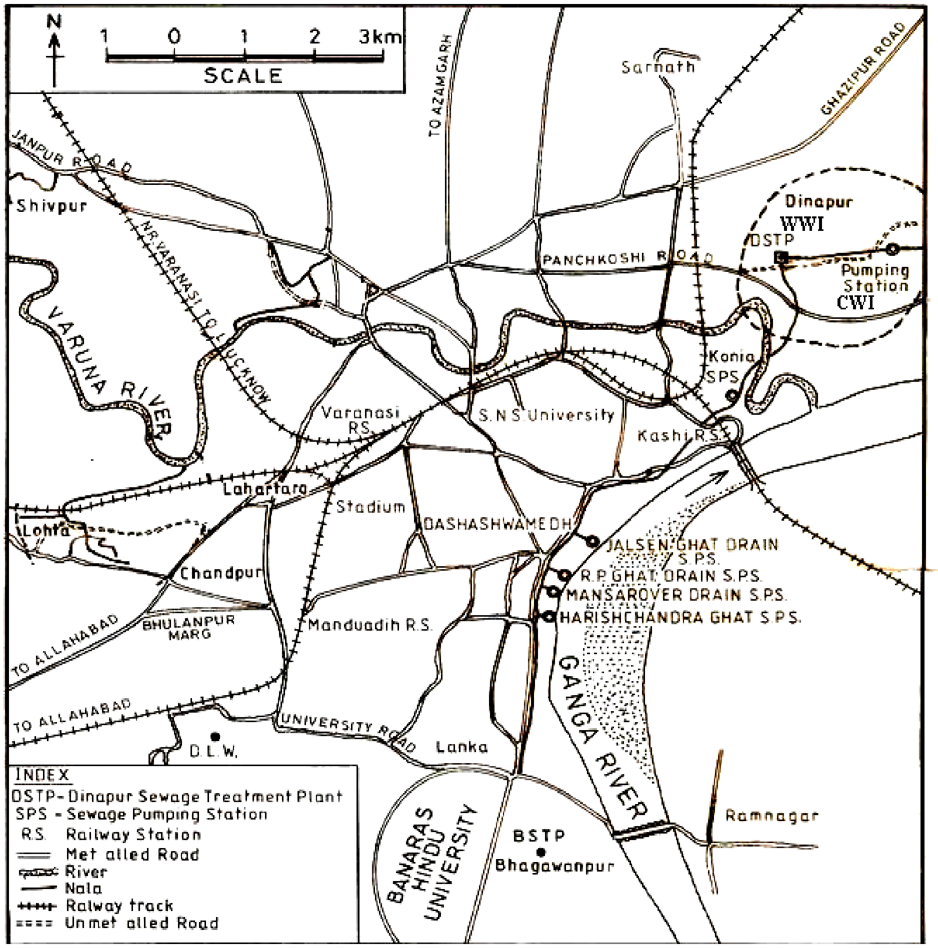

Fig. 1. Map of the study site. WWI: Waste water irrigated site; CWI: Clean water irrigated site.

capacity of 80 million litres per day (MLD) and is operating since 1994. Effluents from various small scale industries situated in the city are also discharged along with sewage for treatment at DSTP. Two areas following different irrigation sources were demarcated at the experimental site. Clean water from deep bore well is used for irrigation at clean water irrigated site designated as CWI, whereas waste water from DSTP is used for irrigation at the other site designated as WWI (Fig. 1). During power failures, untreated waste water from DSTP is also used for irrigation.

Soil, water and plant sampling

Both clean and waste water samples used for irrigation were collected along with the blank (distilled water) in a 100 ml pre acid - washed polypropylene bottle and 1 ml of concentrated HNO_3 was added to the sample to avoid microbial activity. Soil samples were collected in triplicate by digging out a monolith of 10 x 10 x 15 cm size from 5 different fields from CWI and WWI sites. Samples were air dried, crushed, passed through a 2 mm mesh sieve and stored at ambient temperature for analysis.

Edible parts of different vegetables were collected from the experimental sites. Leafy vegetables included palak (*Beta vulgaris* L. cv. All green), amaranthus (*Amaranthus caudatus* L.) and cabbage (*Brasssica oleracea* L. var. *capitata*). Inflorescence vegetable included cauliflower (*Brassica oleracea* L. var. *botrytis*). Fruit vegetables included lady's finger (*Abelmoschus esculentus* L.), brinjal (*Solanum melongena* L.), tomato (*Lycopersicon esculentum* L.), bottle gourd (*Lagenaria siceraria* Mol.), sponge gourd (*Luffa cylindrica* L.), bitter gourd (*Momordica charantia* L.), pumpkin (*Cucurbita maxima* Duch.), pointed gourd (*Tricosanthes* *dioica* Roxb.) and root vegetable included radish (*Raphanus sativus* L.). These are major vegetables grown in the experimental area for own consumption of the farmers and for supply to retail and wholesale markets of Varanasi. Vegetables were collected randomly from 5 x 5 m area of five different fields, each from CWI and WWI sites at 15 d interval for one year from March 2004 to February 2005. Only edible portions of the test vegetables were collected. After washing with clean tap water to remove the soil particles, vegetable samples were oven dried at 80 ºC to constant weight. The dried samples were ground, passed through a 2 mm sieve and stored at room temperature before analysis.

Heavy metal analysis

For heavy metal extraction, 1 g dried sample of plant or soil was digested in 15 ml of HNO_3, H_2SO_4 and $HClO_4$ mixture (5:1:1) at 80 ºC until a transparent solution was obtained (Allen *et al.* 1986). Water samples (50 ml) were digested with 10 ml of concentrated HNO_3 at 80 ºC until the solution became transparent (APHA 2005). These transparent solutions were then filtered through Whatman number 42 filter paper and diluted to 50 ml with distilled water. The concentrations of Cd, Cu, Pb, Zn, Ni and Cr in the filtrate were determined by using atomic absorption spectrophotometer (Model 2380, Perkin Elmer, Inc. Norwalk, CT, USA), fitted with a specific lamp of particular metal using appropriate drift blanks.

Quality control analysis

Quality control measures were taken to assess contamination and reliability of data. Blank and drift standards (Sisco Research Laboratories Pvt. Ltd., India) were run after five determinations to calibrate the instrument. The coefficients of variation of replicate analysis were determined for precision of analysis; the variations were found to be less than 10 %. Precision and accuracy of analysis was assured through repeated analysis of samples against National Institute of Standard and Technology, Standard Reference Material (SRM 1570) for all the heavy metals. The results were found within ± 2 % of the certified values.

Data analyses

Transfer factor (TF)

Transfer factor (TF) was calculated to understand the extent of risk and associated hazard due to waste water irrigation and consequent heavy metal accumulation in edible portion of test vegetables following Cui *et al.* (2004):

TF= concentration of metal in edible part/concentration of metal in soil

Daily intake rate (DIR)

Daily intake was calculated by the following equation:

$$DIR= (C_{metal} \times D_{food\ intake})/ B_{average\ weight}$$

where, C_{metal}, $D_{food\ intake}$ and $B_{avereage\ weight}$ are the heavy metal concentrations in plants (μg g^{-1}), daily intake of vegetables (kg $person^{-1}$) and average body weight (kg $person^{-1}$), respectively. The average daily vegetable intake rate was calculated by doing a survey in which 100 people having average body weight of 60 kg were asked for their daily intake of particular vegetable from the experimental site in each month of sampling (Ge 1992; Wang *et al.* 2005).

Target hazard quotient (THQ)

For the assessment of health risks through consumption of vegetables by the local inhabitants, THQ was calculated following the methodology described by USEPA (USEPA 2000). THQ was determined based on the formula given by Chien *et al.* (2002):

$$THQ= 10^{-3} (E_F F_D F_{IR} C/ R_{FD} W_{AB} T_A)$$

where, E_F is exposure frequency (365 days $year^{-1}$); E_D is the exposure duration (70 years), equivalent to the average lifetime (Bennett *et al.* 1999); F_{IR} is the food ingestion rate (g $person^{-1}$ day^{-1}); C is the metal concentration in food (μg g^{-1}); R_{FD} is the oral reference dose (mg kg^{-1} day^{-1}); W_{AB} is the average body weight (55.9 kg) and T_A is the average exposure time for noncarcinogens (365 days $year^{-1}$ x number of exposure years, assuming 70 years in this study).

Results and discussion

Levels of heavy metals in water and soil samples

The concentration (μg ml^{-1}) of heavy metals in waste water was highest for Zn followed by Pb, Cr, Ni, Cu and Cd. Heavy metal concentrations in clean irrigation water were below the detectable limits (Table 1). Rattan *et al.* (2005) have also found higher concentrations of heavy metals in sewage effluents as compared to the ground water. Many small scale industries such as dyeing, electroplating, metal surface treatment, fabric printing, battery and paints discharge their effluents in sewage water, which may be the cause of elevated

Table 1. The range (μg g^{-1}) of heavy metals in water and soil of clean (CWI) and waste water (WWI) sites.

Heavy metals		Water		Soil	
		Clean water	Waste water	Clean water	Waste water
Cd	Mean	Nd	0.016***	1.49	3.12**
	SD		0.002	0.094	0.190
	Min – Max		0.010 – 0.034	0.80 – 2.80	1.80 – 4.80
Cu	Mean	0.003	0.056***	8.39	21.13***
	SD	0.001	0.002	0.35	0.45
	Min – Max	0.001 – 0.004	0.019 – 0.098	6.00 – 12.00	18.00 – 26.00
Pb	Mean		0.090***	8.15	21.95***
	SD	Nd	0.010	0.90	0.40
	Min – Max		0.043 – 0.150	15.00 – 17.60	19.80 – 24.20
Zn	Mean	0.009	0.130***	44.19	58.13***
	SD	0.002	0.008	1.25	1.45
	Min – Max	0.002 – 0.013	0.043 – 0.193	37.90 – 51.00	52.30 - 66.00
Ni	Mean		0.060***	9.06	23.65***
	SD	Nd	0.012	0.50	1.20
	Min – Max		0.019 – 0.86	7.00 – 12.29	19.89 – 29.00
Cr	Mean		0.053***	9.07	19.21
	SD	Nd	0.005	0.45	0.60
	Min – Max		0.033 – 0.090	7.20 – 11.37	17.56 – 22.00

Nd = not detectable, Level of significance for the differences between WWI & CWI sites, ** = $P \leq 0.01$; *** = $P \leq 0.001$.

heavy metals in treated waste water of DSTP. The concentrations (μg ml^{-1}) of Cd (0.02), Pb (0.0.09), Zn (0.13), Ni (0.06) and Cr (0.05) in waste water recorded during the present study were lower than concentrations of Cd (0.03), Pb (0.26), Zn (0.30), Ni (0.070) and Cr (0.09) in waste water collected from Dinapur area earlier by Sharma *et al.* (2006). Concentrations of Cu, Zn, Pb, Ni and Cr in the waste water were below the permissible limits of heavy metals allowed in the irrigation water, but that of Cd was near the limit (FAO 1985; Pescod 1992).

Continuous application of treated and untreated sewage water to the soil led to higher concentrations of heavy metals in the soil at WWI site as compared to CWI site (Table 1). As compared to the values at CWI site, concentrations of heavy metals were higher by 109 % for Cd, 152 % for Cu, 25 % for Pb, 32 % for Zn, 161 % for Ni and 52.8 % for Cr (Table 1). Singh *et al.* (2004) also reported percent increment for the concentration of Cu by 40.29 %, Pb by 2.05 %, Zn by 41.42 % and Cr by 15.7 % in soil of Dinapur site irrigated by treated and untreated waste water of DSTP as compared to those in soil at clean water irrigated site.

Among all the heavy metals, Zn was found to be maximum and Cd was minimum (Table 1). Singh *et al.* (2004) and Sharma *et al.* (2007) have also found similar trends of highest and lowest concentrations of Zn and Cd in soil of Dinapur area. Mapanda *et al.* (2005) have reported maximum concentration (μg g^{-1}) of 3.4 for Cd, 145 for Cu, 228 for Zn, 21 for Ni, 59 for Pb and 225 for Cr in the waste water irrigated soil of Harare, Zimbabwe. In the present study, the maximum concentrations for Cu, Pb, Zn and Cr were lower and for Cd and Ni were higher than the values reported by Mapanda *et al.* (2005). Not withstanding the above, all the heavy metals in the soil were below the permissible limits of Indian and EU standards at WWI site (Table 2). Continuous cultivation and regular absorption by plants, possibly keep the concentrations of heavy metals in soil within safe limits at WWI site.

Levels of heavy metals in vegetables

Heavy metal concentrations varied among different vegetables (Fig. 2), which may be attributed

Table 2. Guideline for safe limits of heavy metals.

Sample	Standards	Cd	Cu	Pb	Zn	Mn	Ni	Cr
Soil (μg g^{-1})	Indian Standard (Awashthi 2000)	3-6	135-270	250-500	300-600	-	75-150	-
	WHO/FAO (2007)	-	-	-	-	-	-	-
	European Union Standards (EU 2002)	3.0	140	300	300	-	75	150
Water ((μg ml^{-1})	Indian Standard (Awashthi 2000)	0.01	0.05	0.10	5.0	0.10	-	0.05
	FAO (1985)	0.01	0.20	5.0	2.0	0.20	0.20	0.10
	European Union Standards (EU 2002)	-	-	-	-	-	-	-
Plant (μg g^{-1})	Indian Standard (Awashthi 2000)	1.5	30.0	2.5	50.0	-	1.5	20.0
	WHO/FAO (2007)	0.2	40.0	5.0	60.0	-	-	-
	Commission regulation (EU 2006)	0.2	-	0.30	-	-	-	-

to differential absorption capacity of test vegetables for different heavy metals (Zurera *et al.* 1989). All the heavy metal concentrations were several fold higher in the vegetables at WWI site as compared to CWI site receiving clean water for irrigation (Fig. 2). Arora *et al.* (2008) have also found higher concentrations of heavy metals in radish, spinach, turnip, brinjal, cauliflower and carrot grown under waste water irrigation as compared to those at clean water irrigated site.

Among fruit vegetables, Zn concentration (μg g^{-1}) was highest in lady's finger (122.3 - 132.7). The observed range of Zn concentration recorded during the present study was higher than the range (1.3 - 3.7 μg g^{-1}) reported by Sridhara Chary *et al.* (2008) for lady's finger grown in waste water irrigated areas of Hyderabad, Andhra Pradesh. Sharma *et al.* (2006), however, have reported similar ranges (127.30 - 132.70) for lady's finger collected from the agricultural field of Dinapur area during late autumn. The concentration of Cu was found maximum (17.94 μg g^{-1}) in tomato (Fig. 2). As compared to Cu concentration recorded in tomato at WWI site, Liu *et al.* (2006) found several fold higher concentration (201.75 μg g^{-1}) in tomato collected from waste water irrigated area of Zhengzhou city, China. The range of Cd concentration in brinjal (1.55 - 13.80 μg g^{-1}) recorded in this study was higher than the range (1.10 - 9.20 μg g^{-1}) reported by Sharma *et al.* (2006) from Dinapur area and Radwan & Salama (2006) 0.008 - 0.034 μg g^{-1} from Egyptian market. The ranges of Zn and Cu concentrations were 9.00 - 30.05 μg g^{-1} and 4.55 - 17.00 μg g^{-1}, respectively in bottle gourd and 1.40 - 1.70 μg g^{-1} and 4.60 - 5.55 μg g^{-1}, respectively in bitter gourd (Fig. 2). The concentrations of Zn and Cu obtained during the present study were lower than those collected from agricultural field receiving treated tannery waste water for irrigation at Jajmau, Kanpur (Sinha *et al.* 2006).

Among leafy vegetables (palak, cabbage and amaranthus), range of Ni concentration was highest in palak (10.45 - 39.25 μg g^{-1}). These values were higher than the range (5.55 - 15.00 μg g^{-1}) reported by Sharma *et al.* (2006) in palak from Dinapur area as well as the range (0.2 - 3.0 μg g^{-1}) in palak from waste water irrigated areas of Hyderabad reported by Sridhara Chary *et al.* (2008). In cabbage and amaranthus, concentrations of Zn, Cr, Cu, Ni and Pb were higher during the present study as compared to those obtained by Sridhara Chary *et al.* (2008). The present concentration (μg g^{-1}) of 2.19 for Cd, 12.20 for Pb, 3.69 for Cr and 13.75 for Cu in radish were lower than the values obtained for radish collected from a suburban area of Zhengzhou city, Henan Province, China (Liu *et al.* 2006). Khan *et al.* (2008) have reported higher concentrations of Cd, Cr, Cu, Ni and Pb in radish plants grown at waste water irrigated areas of Beijing than the clean water irrigated ones.

Among all the heavy metals, Zn concentration was maximum and Cd was minimum in all the vegetables. Radwan & Salama (2006) have also found highest concentration of Zn and lowest of Cd in vegetables collected from Egyptian markets. Due to variations in absorption of metals in plants through roots and their further translocation within the plant parts, edible parts of vegetables showed

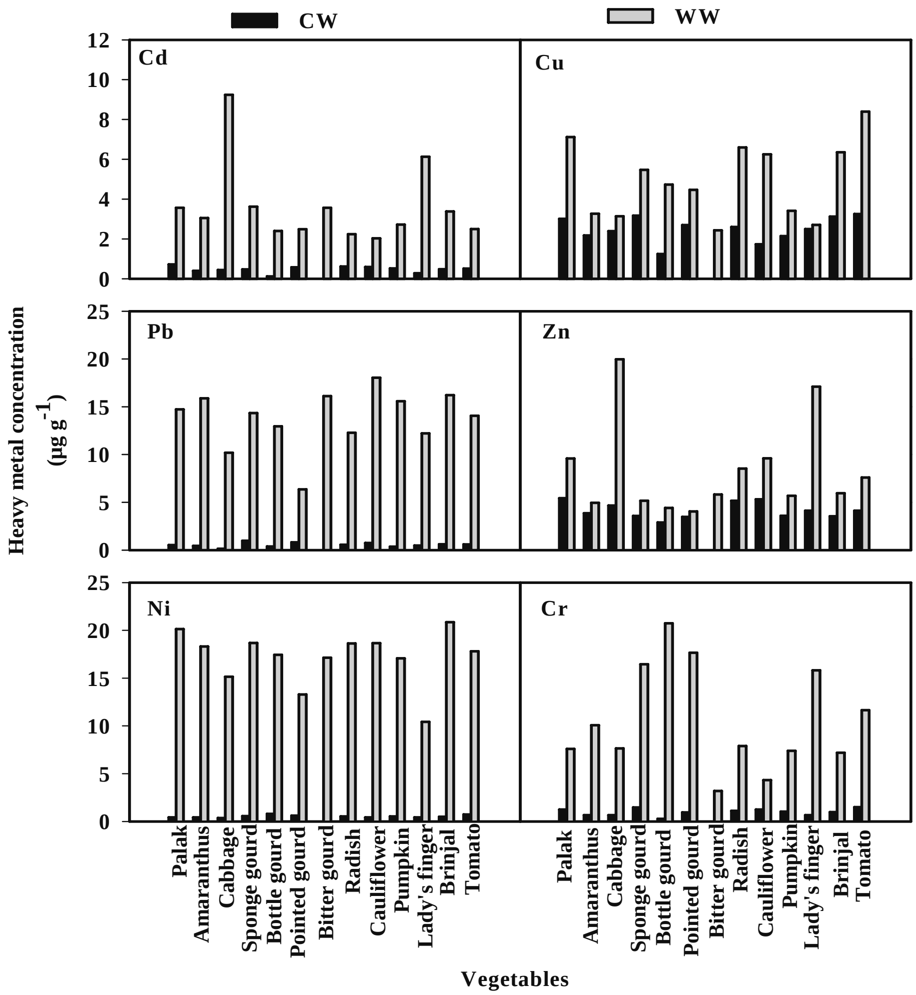

Fig. 2. Mean concentrations of heavy metals (µg g^{-1}) in different vegetables, CW = clear water, WW = waste water.

variations in heavy metal concentrations (Vousta *et al.* 1996).

When the present concentrations of metals were compared with permissible limits of Indian Standard (Awashthi 2000) and safe limits given by WHO/FAO (WHO/ FAO 2007), it was found that at WWI site Cd, Pb and Ni concentrations were higher in all the vegetables, whereas Zn concentration was higher in lady's finger and cabbage. The present concentration of Cd and Pb were also higher in all the vegetables as compared to the safe limits given by commission regulation (EU 2006) (Table 2).

Transfer factor

At control site (CWI), transfer factor for all the metals was lower as compared to waste water irrigated site (WWI) except for Cu (Table 3). Among different metals, Cd showed maximum values for transfer factor, which ranged from 0.61 (cauliflower) to 2.96 (cabbage) at WWI site and it was minimum for Cr, ranging from 0.073 (bitter gourd) to 0.356 (bottle gourd). Transfer factor for Cu, Pb, Zn and Ni was highest for lady's finger (2.08) followed by cauliflower (1.12), cabbage (1.36) and brinjal (0.890) (Table 3). Variations in transfer factor among different vegetables may be attributed to differences in the concentration of metals in the soil and differences in element uptake by different vegetables (Cui *et al.* 2004; Zheng *et al.* 2007). Among all the vegetables, transfer factor of Cd was highest except for radish and tomato at WWI site, which showed that Cd is more mobile than other metals. Lokeshwari & Chandrappa (2006) have

Table 3. Transfer factor of heavy metals through different vegetables at clean (CWI) and waste water irrigated (WWI) sites.

	Cd		Cu		Pb		Zn		Ni		Cr	
Vegetables	CWI	WWI	CWI	WWI	CWI	WWI	CWI	WWI	CWI	WWI	CWI	WWI
Palak	0.392	0.996	0.690	0.641	0.061	0.673	0.421	0.660	0.054	0.827	0.070	0.194
Cabbage	0.288	2.959	0.445	0.261	0.018	0.460	0.445	1.358	0.034	0.575	0.034	0.175
Amaranthus	0.379	0.919	0.621	0.344	0.056	0.673	0.326	0.317	0.041	0.669	0.031	0.229
Brinjal	0.297	1.257	0.723	0.565	0.037	0.714	0.319	0.411	0.058	0.890	0.053	0.200
Sponge gourd	0.454	1.440	0.915	0.523	0.117	0.544	0.320	0.337	0.057	0.693	0.068	0.146
Bottle gourd	0.043	1.051	0.105	0.457	0.015	0.581	0.082	0.300	0.023	0.688	0.004	0.356
Pumpkin	0.515	1.153	0.587	0.341	0.515	0.700	0.295	0.355	0.050	0.656	0.049	0.196
Lady's finger	0.060	0.904	0.340	2.083	0.060	0.904	0.340	2.083	0.037	0.375	0.030	0.355
Pointed gourd	0.218	0.650	0.278	0.425	0.034	0.242	0.107	0.289	0.024	0.488	0.014	0.273
Bitter gourd	Nd	1.140	Nd	0.202	Nd	0.726	Nd	0.395	Nd	0.656	Nd	0.073
Tomato	0.230	0.684	0.687	0.726	0.074	0.643	0.375	0.514	0.085	0.782	0.085	0.304
Radish	0.316	0.670	0.568	0.628	0.075	0.589	0.470	0.610	0.071	0.803	0.069	0.230
Cauliflower	0.279	0.610	0.391	0.597	0.090	1.116	0.474	0.657	0.059	0.811	0.077	0.115

Table 4. Daily intake rate (g person^{-1}day^{-1}) of heavy metals through consumption of contaminated vegetables.

Vegetables	Cd	Cu	Pb	Zn	Ni	Cr
Palak	98.92	416.50	415.18	1091.71	560.94	103.34
Brinjal	36.96	152.59	185.23	270.90	235.56	37.98
Sponge gourd	52.53	172.57	158.66	302.38	272.22	116.51
Amaranthus	79.80	188.24	409.52	515.30	483.57	125.12
Radish	12.21	76.41	67.82	186.97	103.33	20.51
Cauliflower	23.26	150.77	299.48	442.78	216.31	23.68
Bottle gourd	26.86	115.10	136.73	188.23	187.37	123.50
Pumpkin	34.21	93.54	192.73	301.52	218.28	41.90
Lady's finger	58.61	54.91	202.74	1228.30	99.93	66.14
Pointed gourd	12.73	48.30	34.60	84.28	71.07	41.93
Cabbage	126.96	89.83	140.25	1098.85	208.31	50.42
Bitter gourd	26.75	38.00	120.88	174.50	128.50	11.50
Tomato	18.35	137.11	107.90	237.73	135.52	40.83
PTDI*	60	300	214	60	-	-

* Joint FAO/WHO Expert Committee on Food Additives, 1999; PTDI; Potential tolerable daily intake.

Table 5. Target hazard quotient for different heavy metals due to consumption of vegetables collected from WWI site.

Vegetables	Cd	Cu	Pb	Zn	Ni	Cr
Palak	5.87	2.60	2.38	2.38	9.44	0.004
Brinjal	2.02	0.86	0.36	0.36	4.11	0.001
Sponge gourd	3.15	0.85	0.35	0.35	4.23	0.010
Amaranthus	4.00	0.56	0.56	0.56	7.35	0.007
Radish	0.45	0.44	0.35	0.35	1.60	0.001
Cauliflower	0.78	0.82	0.94	0.94	3.38	0.001
Bottle gourd	1.11	0.51	0.18	0.18	2.70	0.016
Pumpkin	1.53	0.29	0.40	0.40	3.11	0.002
Lady's finger	5.99	0.13	4.68	4.68	0.87	0.005
Pointed gourd	0.53	0.19	0.08	0.08	0.82	0.004
Cabbage	19.54	0.24	4.88	4.88	2.63	0.002
Bitter gourd	1.59	0.08	0.23	0.23	1.83	0.000
Tomato	0.73	1.03	0.41	0.41	2.00	0.002

reported that Cd is retained less strongly by the soil and hence it is more mobile than other metals.

Daily intake rate

The degree of toxicity of heavy metals to human being depends upon their daily intake. Heavy metals intake through consumption of vegetables grown in Dinapur area showed large variations (Table 4). The standard of FAO/WHO (1999) has established a reference value for tolerable daily intake. Our estimated daily intake rate for all the metals were below the tolerable daily intake rates except for Cd in palak, amaranthus and cabbage and for Pb in palak and amaranthus. Radwan & Salama (2006) and Khan *et al.* (2008) have also observed no risk due to consumption of common foodstuff grown under waste water irrigated areas. In the present study, Cd is mainly responsible for causing human health risk. Zheng *et al.*

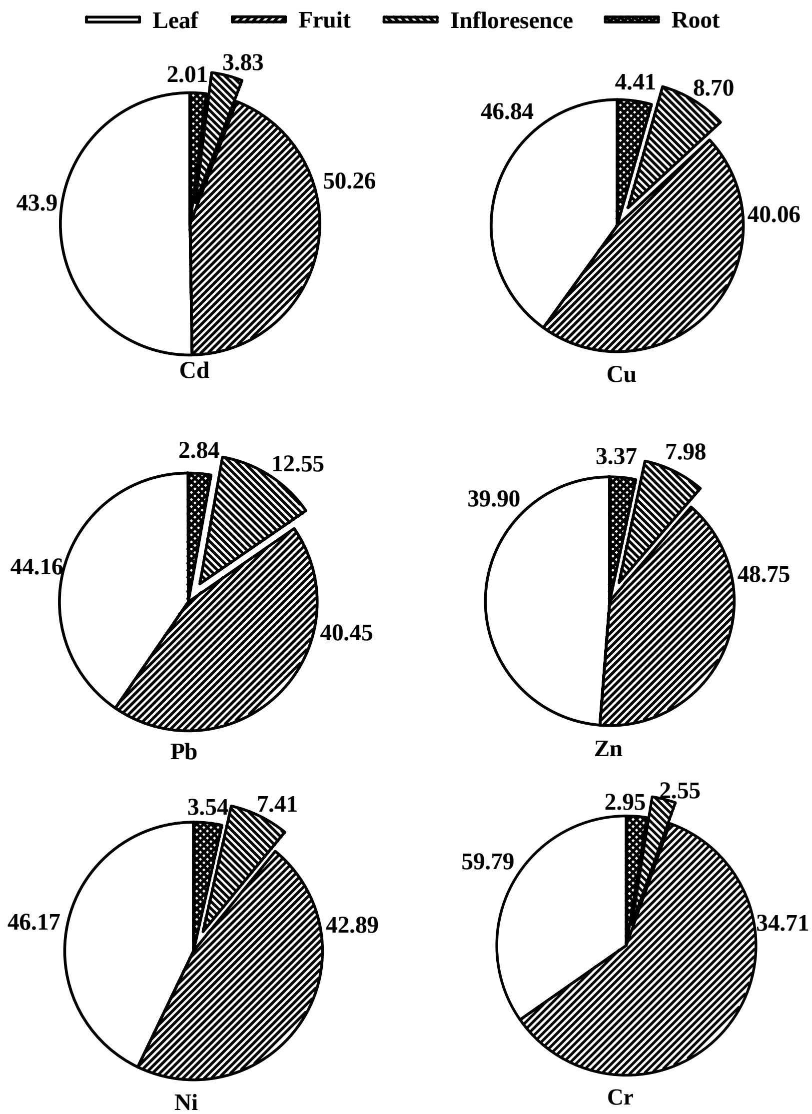

Fig. 3. Percent contribution of vegetables to daily intake of heavy metals.

(2007) have observed that daily intake of Cd was higher than the tolerable daily intake limit around Huludao Zinc Plant, thus causing a threat to the inhabitants of the area.

Percent contribution of heavy metals by vegetables in daily intake varied with the edible part of vegetables (Fig. 3). Fruit vegetables showed highest contribution to the daily intake of Cu, Pb, Ni and Cr, whereas Cd and Zn were maximally contributed by leafy vegetables (Fig 3). Higher availability of fruit and leafy vegetables during the sampling period in the study area may also be contributing to the higher percentages of daily intake of heavy metals in humans.

Target hazard quotient (THQ)

The results of target hazard quotient calculations showed that Cd, Pb and Ni contamination in plants had potential to pose health risk to the local population (Table 5). Cd THQ was more than 1 in most of the vegetables except for radish, cauliflower, pointed gourd and tomato. Pb THQ was more than 1 in palak, lady's finger and cabbage, whereas for Ni it was higher in all except lady's finger and pointed gourd (Table 5). Higher THQ for Cd and Pb were also reported by Zheng *et al.* (2007) in vegetables collected from Huludao Zinc Plant in Huludao City, China. Higher THQ for Cd and Pb in an area near a lead (Pb) and antimony (Sb) smelter in Nanning, China, was also reported by Cui *et al.* (2004). For Cu, the values of THQ were more than 1 in palak and for Zn in palak, lady's finger and cabbage (Table 5). Consumption of such vegetables may have a risk to the local population due to higher target hazard quotient. In the present study, Cr is least responsible for causing risk to the local population as the value of THQ was below 1 for all the vegetables (Table 5).

Conclusions

Waste water irrigation led to the accumulation of heavy metals in soil and consequently into the vegetables. Heavy metal concentrations varied among the test vegetables, which reflect the differences in their uptake capabilities and their further translocation to edible portion of the plants. Cd, Pb and Ni concentrations were above the national and international permissible limits in all the vegetables. Target hazard quotient of heavy metals also suggests that Cd, Pb and Ni contamination in most of the test vegetables had potential for human health risk due to consumption of plants grown in the area having long term uses of treated and untreated waste water for irrigation. Percent contribution to daily intake rate of Cu, Ni, Pb and Cr was higher by fruit vegetables, whereas that of Cd and Zn was higher by leafy vegetables. Consumption of these vegetables with elevated levels of heavy metals may lead to high level of body accumulation causing related health disorders. Thus regular monitoring of heavy metal contamination in the vegetables grown at waste water irrigated area is necessary and consumption of contaminated vegetables should be avoided in order to reduce the health risk caused by taking the contaminated vegetables. The waste water treatment technology should involve steps to remove heavy metals causing risk to human health.

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