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An Assessment of Heavy Metal Contamination in Vegetables Grown in Wastewater-Irrigated Areas of Titagarh, West Bengal, India

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Abstract The effects of municipal wastewater irrigation on the accumulation of heavy metals (Pb, Zn, Cd, Cr, Cu and Ni) in soil and vegetables were investigated by monitoring wastewater-irrigated agricultural field of Titagarh, 24-Parganas (North), West Bengal, India. The mean concentrations of Pb, Ni, Cu in the irrigation water and the mean Cd content in soil were much above the recommended level. The concentrations of Pb, Zn, Cd, Cr and Ni in all the examined vegetables were beyond the safe limits. The study reveals that heavy metal-contaminated vegetables grown in wastewater-irrigated areas may pose public health hazards.

Keywords Wastewater · Irrigation · Heavy metal · Contamination

Wastewater irrigation is a common reality in three fourth of the cities in Asia, Africa and Latin America. Wastewater carries appreciable amounts of trace toxic metals (Pescod 1992; Yadav et al. 2002). Wastewater irrigation is known to have its significant contribution to the heavy metal content of soils (Mapanda et al. 2005; Nan et al. 2002). This loading of heavy metals often leads to degradation of soil health and contamination of food chain mainly through the vegetables grown on such soils (Rattan et al. 2002). Irrigation by sewage water and industrial effluents has been cited as the main reason for accumulation of heavy metals in the vegetables. Municipal solid waste application in agricultural land, aerial depositions from thermal power plants and abundant pesticide use are the other major sources of the toxic heavy metals. Heavy metals are the most important types of contaminants that can be found on the surface and in the tissues of fresh vegetables. Of course certain trace elements are essential in plant nutrition, but plants growing in a polluted environment can accumulate trace elements at high concentrations causing a serious risk to human health when they are consumed (Voutsa et al. 1996). Prolonged exposure to heavy metals such as cadmium, copper, lead, nickel, and zinc can cause deleterious health effects in humans (Reilly 1991). Previous studies revealed carcinogenic effects of several heavy metals such as cadmium (Cd), chromium (Cr), lead (Pb), mercury (Hg) and arsenic (As) (Trichopoulos 1997).

Titagarh (22°44′11″N; 88°22′25″E) is a suburban Industrial town located in 24 Parganas District (North), West Bengal, India. Most of the people living in this area are industrial workers and belong to the weaker section of the society. Our study area is situated in the east side of the river Hooghly (a tributary of river Ganges) 22 km North of Kolkata, the capital city of West Bengal. Titagarh Municipality has a conventional Sewage Treatment Plant. The final effluent from the treatment plant is directly taken to the adjoining agricultural land through 600 mm diameter concrete pipe. This peri-urban area is also a garbage-dumping site of Titagarh Municipality. The common vegetables grown in the study area are pudina, lettuce, spinach, cauliflower, Chinese onion etc. Most of these vegetables cultivated in this site are supplied to the wholesale vegetable market in Kolkata and the rest enter the local markets.

In this study, we investigated the concentrations of Pb, Zn, Cd, Cr, Cu and Ni in irrigation water, soil and vegetables grown in the agricultural land of Titagarh area having long term uses of the treated and untreated wastewater for irrigation. The levels of contamination were compared with the Indian standard guidelines to assess the potential hazards of heavy metals to public health.

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Materials and Methods

Samples of untreated and treated wastewater, top-soil (0–15 cm depths) and nine vegetables [lettuce (*Lactuca sativa*), pudina (*Mentha viridis*), cauliflower (*Brassica oleracea* var *botrytis*), celery (*Apium graveolens*), spinach (*Spinacia oleracea*), coriander (*Coriandrum sativum*), parsley (*Carum petroselinum*), Chinese onion (*Allium tuberosum*) and Radish (*Raphanus sativus*)] were collected randomly in triplicate from the peri-urban area of Titagarh during the months from November 2005 to June 2006 in order to estimate the total heavy metal content (Pb, Zn, Cd, Cr, Cu and Ni) in these samples.

Wastewater (untreated and treated) samples were pre-treated with concentrated HNO_3 to prevent microbial degradation of heavy metals. 50 mL of wastewater samples were digested with 10 mL concentrated HNO_3 at 80°C (APHA 1985). Soil samples were air-dried, crushed and passed through 2-mm mesh sieve and stored at ambient temperature prior to analysis. The freshly harvested mature vegetables were brought to the laboratory and washed primarily with running tap water, followed by three consecutive washings with distilled water to remove the soil particles. Samples were cut into small pieces and dried in oven at 70°C for 48 h and then ground to powder. 0.5 g each of soil and vegetable samples were digested (wet acid digestion) with concentrated HNO_3, H_2SO_4 and $HClO_4$ (5:1:1) at 80°C (Allen et al. 1986) until the solution became transparent. The digested samples of water, soil and vegetables were filtered through the Whatman No.42 filter paper and the filtrates were diluted to 50 mL with distilled water. All reagents used were Merck, analytical grade (AR) including Standard Stock Solutions of known concentrations of different heavy metals.

Heavy metal concentrations of wastewater, soil and vegetable samples were estimated by Atomic Absorption Spectrometer (Perkin Elmeyer AAnalyst 400). Blank samples were analyzed after seven samples. Concentrations were calculated on a dry weight basis. All analyses were replicated three times. The accuracy and precision of metal analysis were checked against NIST-SRM, 1570 for every heavy metal. The results were found within ±2% of certified values. To assess the contamination level of heavy metals, mean, median, minimum, maximum and standard deviation of wastewater, soil and vegetable samples were performed by using Microsoft Excel (Version 2000).

Results and Discussion

Both untreated and treated sewage water are used as irrigation water in the study area. Table 1 summarized the concentration of heavy metals in untreated and treated sewage used for irrigation. The concentrations (mg/L) of heavy metals in untreated wastewater ranged from 0.00 to 7.5 for Pb, 0.21 to 4.3 for Zn, 0.00 to 0.06 for Cd, 0.00 to 0.81 for Cr, 0.07 to 6.3 for Cu and 0.00 to 4.2 for Ni. In treated wastewater, heavy metal concentrations (mg/L) ranged from 0.4 to 6.8 for Pb, 0.1 to 3.9 for Zn, 0.00 to 0.03 for Cd, 0.00 to 0.36 for Cr, 0.01 to 5.81 for Cu and 0.00 to 2.8 for Ni. In comparison with the standard guideline of irrigation water (Pescod 1992) it was found that mean Pb, Ni and Cu concentrations of the untreated and treated wastewater exceeded the recommended level while Cr and Zn concentration of wastewater samples were within safe limit. The Cd concentration was however found to be just at the threshold level.

Table 1 Heavy metal concentrations (mg/L) in wastewater used for irrigation in peri-urban region of Titagarh, India (n = 39)

		Pb	Zn	Cd	Cr	Cu	Ni
Untreated wastewater	Mean	4.26	1.91	0.01	0.05	1.56	0.68
	Median	5.00	1.40	0.01	0.00	1.30	0.08
	Minimum	0.00	0.21	0.00	0.00	0.07	0.00
	Maximum	7.50	4.30	0.06	0.81	6.30	4.20
	Std dev	2.20	1.45	0.01	0.11	1.22	1.00
Treated wastewater	Mean	3.54	1.17	0.01	0.04	0.98	0.39
	Median	3.50	0.60	0.01	0.00	0.70	0.02
	Minimum	0.40	0.10	0.00	0.00	0.01	0.00
	Maximum	6.80	3.90	0.03	0.36	5.81	2.80
	Std dev	1.50	1.20	0.01	0.15	1.04	0.66
Safe limit[a]		0.5	2.00	0.01	0.1	0.2	0.2

[a] Source: Pescod (1992), n: No of samples

Of all the heavy metals examined, concentration of Pb was highest in both untreated and treated wastewater used for irrigation in the study area. The mean Pb, Zn, Cu and Ni concentrations of irrigation water (untreated and treated sewage) in the study area were higher than the mean metal concentrations of irrigation water in suburban area of Varanasi, India (Sharma et al. 2007). The concentration of heavy metals (mg/kg of dry soil) in soil of the study area (Table 2) ranged from 99.3 to 168.3 for Pb, 182 to 285 for

Table 2 Concentrations of heavy metals in wastewater- irrigated soil (mg/kg dry soil) in peri-urban region of Titagarh, India (n = 78)

	Pb	Zn	Cd	Cr	Cu	Ni
Mean	130.45	217.08	30.72	148.41	89.98	103.67
Median	125.25	210.05	29.35	147.05	90.65	101.30
Minimum	99.30	182.00	22.20	118.05	22.00	44.72
Maximum	168.30	285.00	51.00	190.40	166.50	133.80
Std dev	19.43	25.46	5.67	19.35	21.79	17.53
Safe limit[a]	250–500	300–600	3–6	–	135–270	75–150

[a] Source: Awashthi (2000), n: No of samples

Zn, 22.2 to 51 for Cd, 118.05 to 190.4 for Cr, 22 to 166.5 for Cu and 44.72 to 133.80 for Ni. In soil, the mean highest concentration recorded was for Zn followed by Cr, Pb, Ni, Cu and the minimum concentration was observed for Cd. The mean concentrations of Pb, Zn, Cu and Ni in soil of the study area were below the official Indian Standard (Awashthi 2000) but the mean Cd concentration in soil was several folds higher than the threshold level. The mean concentrations of Pb, Cd and Ni in soil recorded during the present study were higher than those reported in Varanasi, India (Sharma et al. 2007) and Zimbabwe (Mapanda et al. 2005). The concentration of Cr (118.05–190.4 mg/kg) in soil of the study area was lower than the value reported in Zimbabwe (Mapanda et al. 2005) and India (Sharma et al. 2007). The concentration of Cu (22–166.5 mg/kg) in soil of the study area was higher than the level reported in Zimbabwe (2.5–133.3 mg/kg) (Tandi et al. 2004), (7–145 mg/kg) (Mapanda et al. 2005) but lower than the concentration (2.55–203.45 mg/kg) reported in Varanasi, India by Sharma et al. (2007).

The comparison of the mean concentrations of heavy metals (Pb, Zn, Cd, Cr and Ni) with the safe limit of Prevention of Food Adulteration (PFA) Act 1954 (Awashthi 2000) showed that the concentrations of heavy metals such as Pb, Zn, Cd, Cr and Ni were above the recommended limit in all the examined vegetables (Table 3). The concentration of Cu was within the safe limit in all the vegetables except for Spinach. The maximum concentration of Pb was found in Radish (57.63 mg/kg) followed by Spinach (49.79 mg/kg), which exceeded the accepted tolerance level in India for Pb by 23 times and 19 times respectively. The mean Pb content in vegetables (21.59–57.63 mg/kg) was several folds higher than the values reported in China (0.18–7.75 mg/kg) (Liu et al. 2006), (1.97–3.81 mg/kg) (Liu et al. 2005) and in Varanasi, India (3.09–15.74 mg/kg) (Sharma et al. 2007) but it was significantly lower than the mean concentration of Pb (409 mg/kg) reported in Turkey by Türkdoğan et al. (2002). The highest Zn concentration was found in lettuce (171.03 mg/kg) followed by spinach (154.21 mg/kg). In both the cases the levels of contamination were about three times higher than the Indian recommended standard. The average concentration of Zn (93.00–171.03 mg/kg) in vegetables from Titagarh was higher than the vegetables from Delhi, India (46.7–91.9 mg/kg) (Rattan et al. 2005) as also from Beijing, China (32.01–69.26 mg/kg) (Liu et al. 2005), but substantially lower than the Zn concentrations (1038–1872 mg/kg) in vegetables from Harare, Zimbabwe (Tandi et al. 2004). The maximum Cd was observed in Radish (17.79 mg/kg) followed by Spinach (14.58 mg/kg), which exceeded the PFA limit, by 11 and 9 times respectively. The present study showed that the average Cd level (10.37–17.79 mg/kg) measured in vegetables from Titagarh was higher than the vegetables from China (0.03–0.73 mg/kg) (Liu et al. 2005), from Varanasi, India (0.5–4.36 mg/kg) (Sharma et al. 2007) and from Egypt (0.002–0.08 mg/kg) (Dogheim et al. 2004) but lower than the vegetables from endemic upper gastrointestinal cancer region of Turkey (25 mg/kg) (Türkdoğan et al. 2002). The mean concentrations of Cr (34.83–96.30 mg/kg) in vegetables observed in the present study were higher than concentrations reported in Varanasi, India (5.37–27.83 mg/kg) (Sharma et al. 2007) and in different parts of China (0.83–11.80 mg/kg) (Liu et al. 2005), (0.08–15.38 mg/kg) (Liu et al.2006). In the present study, the mean highest Cr content was observed in Spinach (96.30 mg/kg) which was approximately four times higher than the Indian Standard. The mean concentration of Cu in vegetables of the present study area varied between 15.66 mg/kg (in cauliflower) and 34.49 mg/kg (in spinach) which were higher than the value observed in Varanasi, India (10.95–28.58 mg/kg) (Sharma et al. 2007) but lower than the Cu content in

Table 3 Heavy metal concentration (mg/kg dry weight) in vegetables grown in waste-water irrigated agricultural land in Titagarh: Mean and (range)

	Pb	Zn	Cd	Cr	Cu	Ni
Lettuce n = 38	34.94 (33.1–38)	171.03 (149.4–193.7)	13.38 (10.3–28)	61.42 (50.4–70.5)	24.93 (17.6–37.8)	52.24 (28.7–85)
Pudina n = 40	21.59 (15.3–26.5)	139.01 (110.9–153)	10.37 (5.4–31)	67.82 (58.4–75.3)	26.25 (18.7–30.7)	53.80 (17.4–81)
Cauliflower n = 26	31.04 (28.3–35.6)	96.48 (82.6–123)	13.80 (7–30)	86.83 (74.8–103)	15.66 (13.4–18.1)	59.28 (25.5–91)
Celery n = 32	24.27 (20.8–28.6)	93.00 (87.8–98.6)	12.02 (6–22.5)	34.83 (33–36.8)	20.56 (17.9–22.5)	42.79 (20.4–62)
Spinach n = 39	49.79 (41.9–60.5)	154.21 (136.5–181)	14.58 (6.5–32)	96.30 (74–115.4)	34.49 (22.1–48.6)	69.22 (45.1–98)
Coriander n = 22	31.13 (25–35.3)	136.09 (126.5–149)	14.05 (9–26.9)	48.20 (39.7–56)	25.10 (20.8–28.6)	51.29 (24.7–68.9)
Parsley n = 34	31.30 (28–36.9)	111.65 (97–130)	12.36 (5.4–25.6)	76.34 (69.5–85.3)	29.60 (19.7–35.1)	56.07 (45.8–76)
Chinese onion n = 40	34.26 (22.8–41.4)	125.00 (110.9–150)	11.49 (5.7–25)	46.41 (40.3–53.2)	17.57 (16.3–18.9)	47.39 (22.2–70)
Radish n = 39	57.63 (50–63.5)	139.05 (125.4–157.3)	17.79 (11.8–28.6)	78.02 (69.8–100)	28.08 (18.7–35.1)	62.70 (51–77)
Safe limit[a]	2.5	50	1.5	20	30	1.5

[a] Source: Awashthi(2000), n: No of samples

vegetables (61.20 mg/kg) from Zhengzhou City, China (Liu et al. 2006). Concentrations of Cu in vegetables were within the threshold limit as recommended in PFA except for spinach, which crossed the safe limit. The concentration of Ni in the present study was highest in Spinach (69.22 mg/kg) followed by Radish (62.70 mg/kg), which were 46 and 41 times higher than the PFA limit. The present study indicates that the average Ni concentration (42.79–69.22 mg/kg) in vegetables from Titagarh were higher than the values reported in Varanasi, India (1.81–7.57 mg/kg) (Sharma et al. 2007) and in Delhi, India (8.78–21.5 mg/kg) (Rattan et al. 2005). Of the nine vegetable species, spinach (for Cu, Ni and Cr), radish (for Pb and Cd) and lettuce (for Zn) showed highest metal accumulation. The samples of soil, irrigation water and vegetables collected from the Titagarh area showed substantial level of heavy metal contamination. Besides wastewater irrigation, garbage utilization in the agricultural land is another likely source of heavy metal pollution in soil of the study area. The present study indicates that wastewater-irrigated vegetables accumulate heavy metals beyond prescribed toxic limits and may cause serious health hazards to people who consume these vegetable products regularly.

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