CADMIUM CONCENTRATION IN VEGETABLES GROWN ON URBAN SOILS IRRIGATED WITH UNTREATED MUNICIPAL SEWAGE

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Abstract. Cadmium (Cd) is considered as a potential toxin that is principally dispersed in natural and agricultural environments through anthropogenic sources. Untreated municipal sewage, often a potential source of Cd, is generally used to irrigate urban agricultural soils in many developing countries. A study was carried out to determine Cd concentration in untreated municipal sewage and sewage-irrigated soils and vegetables. The metal ion concentration in municipal sewage was found 3-fold ($0.03\,\mathrm{mg\,L^{-1}}$) its permissible concentration in irrigation water ($\leq 0.01\,\mathrm{mg\,L^{-1}}$). Ammonium bicarbonate–diethylene triamine pentaacetic acid NH_4HCO_3–DTPA) extractable Cd concentration in top 0.15 m soil ranged between 0.25 and $0.34\,\mathrm{mg\,kg^{-1}}$. Soil Cd concentration was significantly correlated with soil clay content, pH, electrical conductivity, and cation exchange capacity. Cadmium availability index (CDI) decreased with an increase in soil depth. The metal ion was found in leaf (0.17–$0.24\,\mathrm{mg\,kg^{-1}}$ fresh weight) and fruit (0.07–$0.18\,\mathrm{mg\,kg^{-1}}$ fresh weight) portions of all the sampled vegetables: bitter gourd (*Momordica charantia* L.), cauliflower (*Brassica oleracea* L.), eggplant (*Solanum melongena* L.), fenugreek (*Trigonella foenumgraecum* L.), okra [*Abelmoschus esculentus* (L.) Moench], onion (*Allium cepa* L.), pumpkin (*Cucurbita pepo* L.), and spinach (*Spinacia oleracea* L.). Leafy tissue accumulated Cd about twice that of the fruit portion. Our results suggest that prolonged ingestion of sewage-irrigated leafy vegetables can develop such Cd levels in human body that may cause a number of illnesses.

Key words: cadmium, human health, municipal sewage, soil contamination, vegetables.

1. Introduction

Cadmium (Cd) is a pollutant that has no known function in plants and animals. Environmental release of Cd from human activities has been about 10-fold that predicted from natural sources (Nriagu, 1980). Cadmium input to agricultural environment has increased with time (Jones et al., 1992). Main sources of Cd accumulation in agricultural soils include: (1) phosphatic fertilizers, (2) atmospheric fallout from industrial and municipal activities, and (3) solid and liquid municipal sewage (Nriagu and Pacyna, 1988; Wagner, 1993; Ahumada et al., 1999; Qadir et al., 1999). Cadmium content of commercial phosphatic fertilizers, ranging from 0.1 to $200\,\mathrm{mg\,kg^{-1}}$, depends on their geographical origin (Wagner, 1993). Soils fertilized with phosphatic fertilizers have shown a small increase in Cd content after 20 years (Elinder, 1986), 50 years (Mortvedt, 1987), or as long as 70 years (Jeng and Singh, 1995). Cadmium accumulation by plants is principally from soil except in conditions

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of high atmospheric fallout (Van Bruwaene et al., 1984). The third anthropogenic source may develop higher Cd levels in soils and consequently in plants, if application of untreated municipal sewage is continued for many years.

Accumulation of Cd in plant tissue is influenced by the levels of Cd available in the soil in which the plants are grown (Bailey et al., 1995; Ahumada et al., 1999). Plant Cd concentrations, even under mild Cd pollution conditions, are generally below phytotoxic levels for most crops (Bingham, 1979). Therefore, many crops can grow on Cd-polluted soils without showing a yield depression. Ingestion of high-Cd containing crops may contribute substantial Cd to human diet (Wagner, 1993). Of primary concern is its transfer from vegetables to human body because vegetable foods contribute $\geq$ 70% of Cd intake in humans (Ryan et al., 1982).

Untreated municipal sewage, often a potential source of Cd, is generally used to irrigate urban agricultural soils in many parts of the world, particularly in developing countries (Qadir et al., 1999). In most cases, farmers grow market-ready products such as vegetables on the sewage-irrigated soils. This study was carried out in Faisalabad (Pakistan) after some known incidences of illnesses that, in addition to other contributing factors, are generally attributed to excessive accumulation of some metal ions such as Cd in human body. We report Cd concentration in untreated municipal sewage and sewage-irrigated soils and vegetables. In the end, we have calculated and compared with international standards the daily intake rate of Cd in human body as a consequence of ingestion of the sewage-irrigated vegetables.

2. Materials and methods

2.1. Sampling sites

Most urban areas of Pakistan, like many developing countries, have a large number of open and covered channels that are connected to each other and spread within and around urban premises. These channels carry municipal sewage in untreated form. Farmers divert some of the sewage to their fields for irrigation. Vegetables are generally grown on the sewage-irrigated soils. Generally, there is no check on raw sewage utilization for such purposes.

In a survey of urban agricultural soils, receiving untreated municipal sewage, 31 farmers' fields were selected in Faisalabad (Pakistan). Faisalabad is the third largest city of the country. Sampling sites were selected randomly, with at least one hectare distance from each other. The area under each sampling site was about half hectare. The sole criterion for the selection of sampling sites was that a number of vegetables had been grown with untreated municipal sewage irrigation for about 30 years.

2.2. Sample collection and analysis

Untreated municipal sewage, being used for irrigation of vegetables, was sampled from the 31 selected sites. Sewage samples were filtered and analyzed for Cd (atomic absorption spectrophotometrically), pH, electrical conductivity (EC), Na^+ and K^+ (flame photometrically), ($Ca^{2+} + Mg^{2+}$) (titration with standard versinate), CO_3^{2-} and HCO_3^- (titration with standard H_2SO_4), and Cl^- (titration with standard $AgNO_3$). Some parameters were calculated by using the values of already determined parameters, viz. total soluble salts (TSS) $= 10$ (EC), sodium adsorption ratio (SAR) $= Na^+/[(Ca^{2+} + Mg^{2+})/2]^{1/2}$, soluble $SO_4^{2-} = \text{TSS} - (CO_3^{2-} + HCO_3^- + Cl^-)$, and residual sodium carbonate (RSC) $= (CO_3^{2-} + HCO_3^-) - (Ca^{2+} + Mg^{2+})$.

Soil sampling was done from 0.00 to 0.15, 0.15 to 0.30, 0.30 to 0.60, 0.60 to 0.90, and 0.90 to 1.20 m depths. Soil samples were air dried, ground, and screened to pass a 2-mm sieve. The screened samples were stored in polyethylene bags. These samples were analyzed for Cd concentration by atomic absorption spectrophotometer using ammonium bicarbonate–diethylene triamine pentaacetic acid (NH_4HCO_3–DTPA) as the extracting solution (Soltanpour, 1985). Soil particle-size analysis was carried out by hydrometer method (Bouyoucos, 1962). Other determinations such as pH of the saturated soil paste (pH_s), electrical conductivity of the saturated soil paste extract (EC_e), and cation exchange capacity (CEC) were done according to the methods of U.S. Salinity Laboratory Staff (1954).

Samples of vegetables, viz. bitter gourd (*Momordica charantia* L.), cauliflower (*Brassica oleracea* L.), eggplant (*Solanum melongena* L.), fenugreek (*Trigonella foenumgraecum* L.), okra [*Abelmoschus esculentus* (L.) Moench], onion (*Allium cepa* L.), pumpkin (*Cucurbita pepo* L.), and spinach (*Spinacia oleracea* L.) were collected from the selected locations. Fruit (eatable portion) and leaf samples, except for fenugreek and spinach (leafy vegetables: only leaf samples), were collected separately and randomly. The plant samples were washed with 1% HCl to remove foreign material, air dried under paper bags, and finally oven dried at 70°C. The dried samples were digested in a diacid (HNO_3–$HClO_4$) mixture. Cadmium concentration in the prepared samples was determined atomic absorption spectrophotometrically.

3. Results and discussion

3.1. UNTREATED MUNICIPAL SEWAGE

Municipal sewage samples showed a location-wise variation in Cd concentration. Cadmium concentration ranged between 0.01 and 0.04 mg L^{-1} with a mean value of 0.03 mg L^{-1}. This variation may be attributed to the degree of anthropogenic perturbation and dissolved organic carbon (DOC) at different locations. Although we did not measure DOC levels in municipal sewage, Shafer et al. (1999) found a significant relationship between Cd concentration and DOC in a large number of samples from Wisconsin rivers. Average Cd concentration in sewage samples was nearly triple that recommended (≤ 0.01 mg L^{-1}) by the United Nations Food and Agricultural Organization for irrigation waters (Ayers and Westcot, 1985). The municipal sewage samples had an average pH of 7.4, with EC 1.1 dS m^{-1}, SAR 9.8, and RSC 1.2 (Table I). These values suggest that the municipal sewage was of marginal quality for irrigation regarding temporal development of salinity and sodicity problems in the soils.

TABLE I. Composition of untreated municipal sewage samples collected from different locations in Faisalabad, Pakistan.

Parameter	Unit	Value[a]	SD[b]
pH	—	7.4	0.4
EC	$dS\,m^{-1}$	1.1	0.3
TSS[c]	$mmol_c\,L^{-1}$	11.0	3.1
Na^+	$mmol_c\,L^{-1}$	8.2	1.3
K^+	$mmol_c\,L^{-1}$	0.5	0.2
$Ca^{2+} + Mg^{2+}$	$mmol_c\,L^{-1}$	1.4	0.4
CO_3^{2-}	$mmol_c\,L^{-1}$	Traces	—
HCO_3^-	$mmol_c\,L^{-1}$	2.6	0.6
Cl^-	$mmol_c\,L^{-1}$	4.5	0.8
SO_4^{2-}	$mmol_c\,L^{-1}$	3.9	0.7
SAR[d]	$(mmol\,L^{-1})^{1/2}$	9.8	2.0
RSC[e]	$mmol_c\,L^{-1}$	1.2	0.5
Cd	$mg\,L^{-1}$	0.03	0.01

[a] Each value is an average of 46 municipal sewage samples.
[b] Standard deviation for the respective parameter.
[c] Total soluble salts (TSS) = (EC) 10.
[d] Sodium adsorption ratio (SAR) $= Na^+/[(Ca^{2+}+Mg^{2+})/2]^{1/2}$.
[e] Residual sodium carbonate (RSC) $= (CO_3^{2-} + HCO_3^-) - (Ca^{2+} + Mg^{2+})$.

3.2. SEWAGE-IRRIGATED SOILS

The NH_4HCO_3–DTPA extractable Cd concentration in the upper 0.15 m soil ranged between 0.25 and 0.34 mg kg^{-1} with a mean value of 0.29 mg kg^{-1} (Table II). This depth was followed by the 0.15–0.30 m layer that had a range of Cd concentration 0.22–0.30 mg kg^{-1} with an average of 0.26 mg kg^{-1}. At most sampling locations, Cd availability index (CDI) decreased from topsoil to lower depths. There was a sharp decrease in CDI at the lowest sampling depth (0.90–1.20 m). Cadmium concentration at this depth was not detectable at some locations. Onyatta and Huang (1999) have found a similar pattern of Cd concentration in tropical soils of Kenya where CDI decreased significantly from the top soil to the lower soil depths.

In many natural soils, concentrations of Cd have been estimated to be about 0.06–0.50 mg kg^{-1}, the lower value being more often cited (Ryan et al., 1982; Wagner, 1993). Data regarding determination of plant available Cd have shown a large variation, principally because of different kinds of extractants used for Cd determination, and large genotype differences in plants for Cd uptake under similar soil Cd concentrations. Because of these factors, plant Cd concentration has been usually used for practical purposes such as Cd intake in human body from the ingestion of a given plant material of known Cd content.

Analysis of soil samples, collected from different locations, indicate that the soils were alkaline in reaction with an average pH_s as 7.7. EC_e was 3.1 dS m^{-1} with an average SAR 9.9, and CEC 114 $mmol_c\,kg^{-1}$ (Table III). Particle-size analysis reveals that the soils were sandy loam to sandy clay loam in texture. The average clay content was 178 g kg^{-1}, silt 169, and sand 653. These soil characteristics were related to soil Cd concentration. This was done because of the fact that correlation analysis is a useful tool for analyzing similarities

TABLE II. Cadmium concentration in urban agricultural soils used for growing vegetables with untreated municipal sewage as an irrigation source.

Soil under vegetable	Cadmium concentration[a] (mg kg^{-1})				
	Soil depth (m)				
	0.00–0.15	0.15–0.30	0.30–0.60	0.60–0.90	0.90–1.20
Bitter gourd	0.29[b] (0.08)[c]	0.24 (0.07)	0.16 (0.05)	0.19 (0.03)	0.06 (0.04)
Cauliflower	0.27 (0.06)	0.26 (0.06)	0.19 (0.06)	0.09 (0.05)	0.00 (0.00)
Eggplant	0.25 (0.06)	0.28 (0.05)	0.26 (0.06)	0.22 (0.04)	0.07 (0.04)
Fenugreek	0.33 (0.05)	0.27 (0.04)	0.17 (0.05)	0.21 (0.03)	0.04 (0.03)
Okra	0.29 (0.07)	0.24 (0.06)	0.27 (0.05)	0.13 (0.06)	0.04 (0.02)
Onion	0.28 (0.05)	0.26 (0.07)	0.18 (0.04)	0.19 (0.03)	0.05 (0.03)
Pumpkin	0.30 (0.04)	0.22 (0.06)	0.17 (0.04)	0.13 (0.02)	0.00 (0.00)
Spinach	0.34 (0.06)	0.30 (0.05)	0.26 (0.03)	0.15 (0.05)	0.06 (0.02)
Mean	0.29	0.26	0.21	0.16	0.04

[a] NH_4HCO_3–DTPA extractable Cd concentration (Soltanpour, 1985).
[b] Each value is an average of 31 samples.
[c] Figures in parenthesis indicate standard deviation for the respective soil depth.

TABLE III. Some physical and chemical characteristics of urban agricultural soils and their correlations with soil Cd concentration.

Characteristic	Unit	Value[a]	SD[b]	Correlation[c]
Clay	g kg^{-1}	178.0	20.3	0.48**
Silt	g kg^{-1}	169.0	17.1	0.30^{NS}
Sand	g kg^{-1}	653.0	62.3	0.22^{NS}
pH_s	—	7.7	0.9	0.38*
EC_e	dS m^{-1}	3.1	0.8	0.40*
SAR	(mmol L^{-1})$^{1/2}$	9.9	2.3	0.19^{NS}
CEC[d]	mmol$_c$ kg^{-1}	114.0	11.2	0.51**

[a] Each value is an average of 31 soil samples of the 0.15 m soil depth.
[b] Standard deviation for the respective soil characteristic.
[c] Correlation coefficient (r) between Cd concentration in soil with different soil characteristics.
[d] Cation exchange capacity.
*,** Significant at $P = 0.05$, and 0.01, respectively.
^{NS} Nonsignificant.

between paired data and is widely used in metal ion data analyses (Lee et al., 1997). Soil Cd concentration of the upper 0.15 m depth was significantly correlated with clay content ($r = 0.48$), pH ($r = 0.38$), EC_e ($r = 0.40$), and CEC ($r = 0.51$). These correlations agree well with Chen et al. (1999) who found clay content, pH, and CEC important in controlling the level and distribution of various trace elements including Cd. Correlations between soil Cd concentration and other soil properties such as silt and sand contents and SAR were not significant.

3.3. SEWAGE-IRRIGATED VEGETABLES

Cadmium was found in leaf and fruit portions of all the vegetables. The metal ion concentration was variable in different vegetables as well as their portions (Table IV). It was higher in leaf (0.17–0.24 mg kg^{-1} fresh weight) than in fruit (0.07–0.18 mg kg^{-1} fresh weight). There

TABLE IV. Cadmium concentration in vegetables grown on urban agricultural soils with untreated municipal sewage as an irrigation source.

Vegetable	Cadmium concentration[a] ($mg\,kg^{-1}$)	
	Leaf	Fruit
Bitter gourd	0.18[b] (0.04)[c]	0.08 (0.03)
Cauliflower	0.19 (0.05)	0.07 (0.05)
Eggplant	0.17 (0.04)	0.10 (0.07)
Fenugreek[d]	0.24 (0.06)	—
Okra	0.16 (0.04)	0.10 (0.05)
Onion	0.17 (0.05)	0.18 (0.04)
Pumpkin	0.18 (0.07)	0.11 (0.06)
Spinach[d]	0.23 (0.04)	—
Mean	0.19	0.08

[a] Cadmium concentration in plant material is expressed on fresh weight basis.

[b] Each value is an average of 31 samples.

[c] Figures in parenthesis indicate standard deviation for the respective portion of vegetable.

[d] Leafy vegetables: only leaves were sampled and analyzed.

was no apparent sign of a crop yield depression on all locations. This might be because of the fact that plants require substantially higher exposure levels of Cd to show a yield depression. Bingham (1979) found that the most sensitive soil-grown plants, such as spinach, required 2.4 ppm DTPA extractable Cd to produce a 25% yield decrement. This Cd level indicates that healthy crops grown on Cd-polluted soils may accumulate higher Cd levels without having a yield decline. A value of 0.3 $mg\,Cd\,kg^{-1}$ plant material, on dry weight basis, is generally considered critical (Wagner, 1993; Reiman and Caritat, 1998) although large genotype differences occur regarding Cd uptake by plants grown under similar soil Cd concentrations.

3.4. CADMIUM EFFECT ON HUMAN HEALTH

Contribution of a crop to dietary intake of Cd in human body depends on the quantity of crop consumed as food, and on Cd concentration in the consumed tissue. Our results show that leafy tissue accumulated Cd about twice that of the fruit portion. Leaves of spinach and fenugreek are used as human diet while fruit portion of the other vegetables are generally used for the same purpose. From the data given in Table IV, Cd intake levels in human body were calculated on the basis of vegetable consumption rate of 0.3 kg fresh weight per day (Table V). Cadmium intake through fenugreek was the highest (72 $\mu g\,d^{-1}$) closely followed by spinach (69 $\mu g\,d^{-1}$). World Health Organization has set a daily maximum tolerable level of Cd intake at 70 μg for adults or 1 $\mu g\,kg^{-1}$ body weight (Ryan et al., 1982; Wagner, 1993; Bailey et al., 1995). Calculated Cd intake values indicate that continuous ingestion of raw sewage-irrigated leafy vegetables, such as spinach and fenugreek, may develop Cd levels in human body closer to the upper boundary (70 $\mu g\,d^{-1}$). However, during a survey of the present study, adverse effects of Cd-contaminated leafy vegetables on human health were

TABLE V. Cadmium intake rate in humans from the ingestion of edible portions of different vegetables grown on urban agricultural soils with untreated municipal sewage as an irrigation source.

Vegetable	Cadmium intake rate in human body[a]			
	μg d^{-1}	μg wk^{-1}	mg mo^{-1}	mg yr^{-1}
Bitter gourd	24[b]	168	0.72	8.76
Cauliflower	21	147	0.63	7.66
Eggplant	30	180	0.90	10.95
Fenugreek	72	504	2.16	26.28
Okra	30	180	0.90	10.95
Onion	54	378	1.62	19.71
Pumpkin	33	231	0.99	12.04
Spinach	69	483	2.07	25.18

[a] Maximum tolerable daily Cd intake rate for adults is 70 μg or 1 μg kg^{-1} body weight.

[b] Calculations are based on an average daily vegetable consumption rate of 0.3 kg fresh weight.

not evident in most people of the area consuming these vegetables. The possible reasons may be: (1) leafy vegetables were not consumed daily, (2) higher Cd levels in vegetables could be recent since there might not be significant addition of Cd to the sampling locations through the initial years of municipal sewage application, and (3) the soils were alkaline in nature (pH > 7); soil pH values around 7 or higher lead to a diminished Cd release from chelates and clay particles (Stoeppler, 1991). Nevertheless, ingestion of sewage-irrigated vegetables may accelerate Cd accumulation in humans.

Since Cd has no known biological function, its long half life in humans (10–30 years depending on the age), and known acute toxicity make it a potential hazard of concern. The metal once taken up by the human body is excreted at a slow rate of about 0.005% of the body burden (Ryan et al., 1982). Excessive exposure to Cd has been associated with various illnesses in humans including gastroenteritis, renal tubular dysfunction, hypertension, cardiovascular disease, pulmonary emphysema, cancer, and osteoporosis (Gairola et al., 1992; Wagner, 1993).

4. Conclusions

Average Cd concentration in untreated municipal sewage samples was triple (0.03 mg L^{-1}) that permissible in irrigation water ($\leq$ 0.01 mg L^{-1}). Soil Cd concentration was significantly correlated with soil properties, including pH, clay content, EC$_e$ and CEC. Among the sampled vegetables, prolonged ingestion of sewage-irrigated leafy vegetables may accumulate more Cd in humans (69–72 μg d^{-1}) because leafy tissue was found as high Cd accumulating plant portion. Consequently, a number of illnesses may develop in human body. Our results suggest that there should be a restriction on raw sewage irrigation to grow leafy vegetables to reduce the intake rate of Cd in humans. A combination of public awareness through media, and strong legislation along with its effective implementation may help play an important role to restrict raw sewage irrigation under such conditions.

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