

Heavy metal contamination of soil and vegetables with industrial effluents from sugar mill and tanneries

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Abstract

Due to scarcity of water, the use of sewage and industrial effluents for irrigating crops particularly vegetables and fodder crops in the vicinity of big cities of developing countries is increasing. On the other hand, there is a great concern about the environmental hazards associated with the use of such contaminated water for irrigation of food and fodder crops. Hence, a case study was undertaken to assess the effect of industrial effluents on heavy metal contents of soils and vegetable crops grown in the vicinity of Peshawar city of Pakistan. Charsadda road was selected where Khazana sugar mill and many tanneries are situated and discharge its effluents into irrigation water channel. Industrial effluents, soil and plant samples were collected and analyzed for heavy metal contents. Results showed that average values of heavy metals in industrial effluents were Ni (0.66 mg kg^{-1}), Cd (0.06 mg kg^{-1}), Cr (3.45 mg kg^{-1}), Cu (0.34 mg kg^{-1}), Pb (0.32 mg kg^{-1}), Fe (1.18 mg kg^{-1}), Zn (0.3 mg kg^{-1}) and Mn (0.89 mg kg^{-1}). The metal contents of samples taken before mixing with effluents were much less than at the source and were higher after mixing with effluents. Metal content was significantly different from canal water for Cd, Cr, Pb, Fe and Mn being higher in effluents than canal water. As regards soils, 27% samples were beyond safe limits in Ni, 73% in Cd, 64% in Cr, 100% in Cu, 36% in Pb, 73% in Zn and 100% in Mn. In case of vegetables, all the samples collected had higher levels of all the heavy metals in leaves as well as fruits for human consumption. The study concludes that the use of industrial waste water for irrigation has increased the contamination of heavy metals in edible portion of vegetables causing potential risks in the long run from this practice.

Key words: Heavy metals, tanneries, effluents, soils, vegetable

Introduction

Human activities such as industrial production, mining, agriculture and transportation release high amounts of heavy metals into surface and ground water, soils and ultimately to the biosphere. Accumulation of heavy metals in crop plants is of great concern due to the probability of food contamination through the soil root interface (Cieslinski *et al.*, 1996; Qadir *et al.*, 1998). Though the heavy metal viz. Cd, Pb and Ni are not essential for plant growth, they are readily taken up and accumulated by plants in toxic levels (Cieslinski *et al.*, 1996; Qadir *et al.*, 1999; Tatar *et al.*, 1999; Bhatti and Perveen, 2005; Mussarat and Bhatti, 2005).

Ingestion of vegetables irrigated with waste water and grown in soils contaminated with heavy metals poses a possible risk to human health and wildlife (Oliver, 1997; Rattan *et al.*, 2005; Bhatti and Mussarat, 2006; Otero *et al.*, 2000; Mussarat *et al.*, 2007).

Heavy metal concentration in the soil solution plays a critical role in controlling metal bioavailability to plants. Most of the studies show that the use of waste water contaminated with heavy metals for irrigation over long period of time increases the heavy metal contents of soils above safe limits. Ultimately increasing the heavy metal

content in soil increases the uptake of heavy metals by plants depending upon the soil type.

In the big cities of Pakistan, untreated sewage water and the industrial effluents are discharged directly into water channels or canals and the polluted water is used for growing crops particularly vegetables and fodder in the vicinity of big cities (Ahmad *et al.*, 1994; Wallace and Wallace, 1994; Qadir *et al.*, 1998; Khan *et al.*, 2003). City effluents (sewage and industrial) are one of the potential sources of metal pollution which is used for growing crops (mostly vegetables and fodder) in the pre-urban areas of Pakistan (Ghafoor *et al.*, 1994; Khan *et al.*, 1994; Mussarat *et al.*, 2007).

Due to industrialization in Pakistan, many different industries have also been established in Peshawar city of Pakistan. Tanning is one of the oldest industries in Pakistan and is one of the potential sources of heavy metals contamination of water, soils and plant growing on these soils. Wet process of tanning is the main source of generating waste water. Water consumption per kg of raw hides varies from tannery to tannery which should not go beyond 50 L kg^{-1} . However, the tanneries are generally consuming more water than required. Moreover, sugar

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industry is also main source of pollutants to surface water bodies.

It is alarming that most of the cities and industries in Pakistan are without waste water treatment facilities. Large quantities of untreated municipal sewage and industrial effluents are discharged directly to surface water resulting in severe pollution particularly with heavy metals.

Keeping in view the heavy metals hazards of use of untreated waste water, a study was carried out to investigate the composition of effluents of sugar and tanning industries with respect to heavy metals, located on Charsadda road, Peshawar and their effects on soils and plants irrigated with these contaminated waters.

Materials and Methods

Heavy metals hazard in irrigation water, soil and plants growing along Charsadda road, Peshawar were evaluated. Water samples were collected from a water channel at the source of discharge of effluents from Khazana sugar mill and seven tanneries. Water samples were also collected 5 m before the point of discharge and 5 m after the point of discharge along the water channel. These samples were collected in plastic bottles. One sample was also collected from the canal. All the water samples were analyzed for heavy metals (Zn, Cu, Fe, Mn, Cr, Ni, Pb and Cd) according to the standard procedures using atomic absorption spectrophotometer.

Soil samples were also collected from potentially contaminated soils at two depths (0-15 and 15-45 cm). Soil samples thus collected were air dried, crushed and passed through 2 mm sieve. These samples were stored in plastic bottles. Soil samples were analyzed for various heavy metals using AB-DTPA extractant according to the method given by Havlin and Soltanpour (1981).

Leaves and fruits of vegetables growing on these soils receiving waste water were also collected. These vegetables included tomato (*Lycopersicum esculentum*), ridge gourd (*Luffa actangula*), okra (*Hibiscus esculentus*) and bottle gourd (*Cucurbita pep*). Six samples each from tomato, ridge gourd and Okra, and three samples from bottle gourd were collected. The plants samples were washed with distilled water to remove dust and impurities, and then spread on a clean plastic sheet, air dried at room temperature and then oven dried at 72 °C for 48 hours. The dried plant samples were ground to powder form and stored in clean dry plastic jars. All the plant samples were analyzed using wet digestion method of Richards (1954) for heavy metals.

Descriptive statistics of all the water, soil and plant samples were calculated according to Steel and Torrie (1980). Comparison of effluent, at source, before mixing and

after mixing with effluents was made using ANOVA and LSD test of significance. Comparison of effluents with canal water was made using t-test of significance.

Values of heavy metals in water were compared with the standards given by EPA (2000), soils and plants by WHO (1996).

Results and Discussion

Heavy Metal Contents of Water

Heavy metal contents of water samples collected at the source i.e. point of discharge of the industrial effluent, 5 meter before the point source and 5 meters after the point source showed that both the samples collected at source from sugar mills were above the safe limits only in Ni (Table 1). Rests of the heavy metals studied were in the safe limits. As regards the seven samples collected at source from tanneries, two each in Ni, Cd, Cu and Fe, all in Cr, 3 each in Pb and Mn were above the safe limits. As regards water samples collected before the source, all the samples were within the safe limits in all the heavy metals studied (Table 2). However, one sample each in Ni, Cd and Cu, and 4 samples in Cr were found above the safe limits in the samples collected after source from tanneries, while in case of sugar mills both the samples were found in the safe limits (Table 3).

Water samples collected at source from the water channel where effluents from sugar mills and tanneries are discharged had heavy metals hazards especially for Cr, Pb and Mn. This indicates that this water used for irrigation is not suitable. Even in water samples collected at 5 m after the source when mixed with water, Cr, Cd and Cu were found above the safe limits based on the guidelines of Pak EPA (1993). Such guidelines would not allow use of this water for irrigating vegetables or fodder crops. Evaluation of quality of industrial effluents for irrigation purposes has been made by many workers and there is realization that the irrigation water passing through urban areas does not meet the criteria set by different agencies for the use of such water for growing crops particularly the vegetables. In a recent study by Mussarat *et al.* (2007), it was found that irrigation water used for growing vegetables in the vicinity of Peshawar city is not fit for growing vegetables and fodder crops. Such water has an impact on heavy metal accumulation in soils and ultimately in plants growing on these soils (Ghafoor *et al.*, 1994; Khan *et al.*, 2003; Bhatti and Perveen, 2005; Mussarat *et al.*, 2007). With the increasing population especially in big cities and expansion of industries and automobile workshops, the waste water is discharged into the irrigation water channels and canals. Use of such water for irrigation by farmers is increasing with time due to water shortage and this is likely to continue, particularly in the vicinity of urban areas (Wallace and

Table 1. Heavy metals content of effluents at source (mg L⁻¹)

Metal	No. of samples	Mean	Minimum	Maximum	CV (%)	No. samples above safe limit	
						Tanneries	Sugar Mill
Ni	9	0.66	0.13	1.06	83.18	2	2
Cd	9	0.06	0.01	0.25	146.3	2	0
Cr	9	3.45	0.002	7.29	68.49	7	0
Cu	9	0.34	0.02	1.73	180.87	2	0
Pb	9	0.32	0.08	0.72	73.51	3	0
Fe	9	1.18	0.13	3.98	116.5	2	0
Zn	9	0.30	0.03	1.79	194.5	0	0
Mn	9	0.89	0.02	2.50	95.97	3	0

Table 2. Heavy metals content of water before mixing effluents (mg L⁻¹)

Metal	No. of samples	Mean	Minimum	Maximum	CV (%)	No of samples above safe limit
Ni	4	0.30	0.01	0.86	129.32	0
Cd	4	0.03	0.003	0.09	93.40	0
Cr	4	0.05	0.03	0.09	58.86	0
Cu	4	0.05	0.04	0.06	25.88	0
Pb	4	0.09	0.04	0.13	40.76	0
Fe	4	0.09	0.02	0.13	53.84	0
Zn	4	0.05	0.01	0.09	61.25	0
Mn	4	0.24	0.01	0.93	187.43	0

Table 3. Heavy metals content of water after mixing effluents or at field (mg L⁻¹)

Metal	No. of samples	Mean	Minimum	Maximum	CV (%)	No. of samples above safe limit
Ni	8	0.48	0.02	1.25	101.74	1
Cd	8	0.39	0.01	2.61	230.80	1
Cr	8	1.52	0.05	3.47	93.93	4
Cu	8	0.21	0.01	1.20	189.22	1
Pb	8	0.17	0.12	0.32	37.81	0
Fe	8	0.40	0.12	1.60	122.68	0
Zn	8	0.15	0.03	0.67	145.73	0
Mn	8	0.50	0.02	1.07	86.77	0

Wallace, 1994; Qadir *et al.*, 1998). Similarly, heavy metals hazards in waste water from Faisalabad and Lahore have been reported by other workers as well (Ghafoor *et al.*, 1994; Khan *et al.*, 1994; Ali, 1997; Farid, 2003).

Comparison of heavy metals at different points of collection

Nickle (Ni) content differed significantly among the effluents at different points of collection (Table 4). The maximum Ni content was recorded in the effluents at source while canal water samples collected passing through the source and after passing through the source (after mixing) were at par indicating dilution of the Ni in water. The concentration of Cd did not change significantly at different locations indicating the absence or very minor amount of Cd present in the effluents. The concentration of Cr was significantly higher at source followed by the samples after

mixing with the effluents while the samples before passing through the source had negligible Cr content. The concentration of Cu, Pb and Zn did not change significantly with respect to samples collected from different locations, however, the concentration was in the order of source > after source > before source.

Differences among different points of collection regarding Fe contents were significant. Maximum Fe content was found at source followed by samples collected after the effluents mixed with the canal water which were at par with the samples collected before mixing with the effluents. Similar trends were observed for Mn.

Comparison of heavy metals in effluents with canal water

There were non significant differences between Ni contents of effluents and canal water (Table 5). The

concentration of Cd, Cr, Fe, Pb and Mn was significantly higher in effluents than in the canal water. This was attributed to the use of chemicals containing these metals in the industries. Copper, Ni and Zn contents in both canal and effluents were comparable and the differences were non significant.

Heavy metals contents of soil

Nickel content in surface soil ranged from 0.83 to 12.35 mg kg⁻¹ (Table 6) with a mean value of 4.04 mg kg⁻¹.

In subsoil, it ranged from 0.49 to 9.28 mg kg⁻¹, with a mean value of 4.02 mg kg⁻¹. In surface soil, three samples were above the safe limits of WHO (1996). For Ni, 104.38% coefficient of variation was noted which means that the samples varied considerably from one another. In subsoil, none of the sample was having Ni concentration above the safe limits of WHO (1996). It is also evident from the results that surface soil contains more Ni than the subsoil indicating its immobility. Cadmium concentration in surface soil ranged from 0.09 to 6.25 mg kg⁻¹ (Table 6) with a mean

Table 4. Comparison of effluents heavy metal

Metal (mg L ⁻¹)	Effluents at source	Before passing source	After passing source or at field
Ni	0.66 a	0.30 b	0.48 b
Cd	0.06 a	0.05 a	0.39 a
Cr	3.45 a	0.05 b	1.52 b
Cu	0.34 a	0.05 a	0.21 a
Pb	0.32 a	0.09 a	0.17 a
Fe	1.18 a	0.10 b	0.40 b
Zn	0.30 a	0.06 a	0.15 a
Mn	0.89 a	0.24 b	0.50 b

*Means followed by similar letter (s) in each row do not differ significantly from one another.

Table 5. Comparison of effluents with canal water

Metal (mg L ⁻¹)	Effluent Mean	Canal Water	t values
Ni	0.66 a	0.26 a	2.18 N S
Cd	0.06 a	0.01 b	2.50 *
Cr	3.45 a	0.01 b	4.35 *
Cu	0.34 a	0.07 a	1.28 NS
Pb	0.32 b	0.42 a	1.75 *
Fe	1.18 a	0.08 b	2.39 *
Zn	0.30 a	0.06 a	1.26 NS
Mn	0.89 a	0.06 b	2.96 *

N S Non significant at 5 % level of probability

*Means followed by similar letters in each row do not differ significantly at 5 % level of probability

Table 6. Heavy metals contents of soils (mg kg⁻¹)

Metal	No. of samples	Mean	Minimum	Maximum	CV (%)	No. of samples above safe limit
Surface soil (0-15)						
Ni	11	4.04	0.83	12.35	104.38	3
Cd	11	2.81	0.09	6.25	82.46	8
Cr	11	9.28	0.21	16.25	52.09	7
Cu	11	8.57	5.32	16.96	39.56	11
Pb	11	10.51	3.20	17.00	45.16	4
Fe	11	19.84	10.52	33.60	33.45	0
Zn	11	1.91	0.21	3.28	44.56	8
Mn	11	14.84	6.25	22.20	30.80	11
Subsoil (15-45 cm)						
Ni	11	4.02	0.49	9.28	85.46	0
Cd	11	1.24	0.06	2.73	74.77	7
Cr	11	5.32	0.12	11.00	72.33	5
Cu	11	5.85	1.69	12.28	109.21	11
Pb	11	4.59	0.28	10.14	78.63	0
Fe	11	11.41	2.05	27.55	59.93	0
Zn	11	1.12	0.19	2.18	53.48	2
Mn	11	7.03	1.69	18.25	69.61	11

Table 7. Average Heavy metals content of various vegetables

Heavy metal	Vegetable			
	Tomatoes (6)	Ridge gourd (6)	Okra (6)	Bottle gourd (3)
Ni	44.89	48.33	44.80	1820
Cd	9.84	1.95	1.66	0.59
Cr	25.90	19.63	23.59	14.02
Cu	129.20	76.27	75.18	68.83
Pb	61.98	22.43	20.28	10.49
Fe	583.40	592.80	359.90	315.40
Zn	49.65	48.90	32.12	26.97
Mn	73.51	51.49	54.27	53.09
Fruits (mg kg⁻¹)				
Ni	62.49	54.84	68.13	29.49
Cd	13.04	1.02	3.56	1.00
Cr	32.34	34.44	19.95	17.23
Cu	141.90	79.93	108.90	83.74
Pb	106.50	28.87	21.69	15.85
Fe	714.30	841.00	250.00	573.70
Zn	56.68	108.90	39.43	34.91
Mn	58.22	59.80	63.23	33.96

value of 2.81 mg kg⁻¹. In subsoil, it ranged from 0.06 to 2.73 mg kg⁻¹, with a mean value of 1.24 mg kg⁻¹. Eight samples were found above the permissible limits of WHO (1996). In subsoil, seven samples were above the permissible limits. It is clear from these results that Cd was found in higher concentration in surface soil than that of the subsoil.

Chromium concentration ranged from 0.21 to 16.25 mg kg⁻¹, with a mean value of 9.28 mg kg⁻¹ (Table 6), while in subsoil it ranged from 0.12 to 11.00 mg kg⁻¹, with mean of 5.32 mg kg⁻¹. In case of surface soil, seven samples were found above the permissible limit of WHO (1996), while five samples were observed above the safe limits in subsoil. In case of Cr high co-efficient of variation was noted in subsoil than in surface soil, which shows that there is more variability in samples of subsoil as compared to surface soil. Both the surface and subsoil contain high amount of Cr, because of high amount of Cr discharged from tanneries into irrigation water.

Copper concentration (Table 6) of surface soil ranged from 5.32 to 16.96 mg kg⁻¹, with a mean value of 8.57 mg kg⁻¹, while in subsoil it ranged from 1.69 to 12.28 mg kg⁻¹ with a mean value of 5.85 mg kg⁻¹. In all surface and subsoil samples Cu was found above the permissible limits of WHO (1996). However, high amount of Cu was determined in surface than in the subsoil. High variability was found in samples of subsoil than in surface soil.

Lead content of surface soil ranged from 3.20 to 17.00 mg kg⁻¹ with a mean value of 10.51 mg kg⁻¹. In subsoil it ranged from 0.28 to 10.14 mg kg⁻¹ (Table 6) with a mean value of 4.59 mg kg⁻¹. In surface soil, four samples were

found above threshold level of WHO (1996), while none of the samples in subsoil was found above the critical level. More Pb concentration was found in surface than in the subsoil samples. High co-efficient of variation was determined for subsoil than for surface soil.

Iron content of surface soil ranged 10.52 to 33.60 mg kg⁻¹ (Table 6) with a mean value of 19.84 mg kg⁻¹, while in subsoil it ranged from 2.05 to 27.55 mg kg⁻¹, with a mean value of 11.41 mg kg⁻¹. All the samples (surface and subsoil) were found within the safe limits of WHO (1996). However, more concentration of Fe was found in surface than in subsoil samples. More variation was found in subsoil samples.

Zinc content of surface soil (Table 6) ranged from 0.21 to 3.28 mg kg⁻¹, with a mean value of 1.91 mg kg⁻¹. While in subsoil samples, the concentration ranged from 0.19 to 2.18 mg kg⁻¹ with a mean value of 1.12 mg kg⁻¹. Eight surface soil samples were above the critical limits of WHO (1996), while two samples were found beyond the safe limits in subsoil samples. Owing to the collection of these soil samples along the roadside high Zn content was found. High co-efficient of variation was determined for subsoil samples, while high concentration of Zn was noted in surface soil samples.

Manganese content of surface soil ranged from 6.25 to 22.20 mg kg⁻¹, with a mean value of 14.84 mg kg⁻¹, while in subsoil samples, it ranged from 1.69 to 18.25 mg kg⁻¹, with a mean value of 7.03 mg kg⁻¹ (Table 6). All samples, both surface as well as subsoil were found above the safe limits of WHO (1996).

Environmental concern due to use of waste water with respect to heavy metals has been increased where as soil is considered as the major recipient of pollutants. Therefore, heavy metals accumulation in soils irrigated with sewage water needs special attention. As the use of untreated sewage water/industrial effluent is increasing for agriculture in big cities due to shortage of water, the situation is becoming alarming. Since these metals are accumulated in soil to toxic levels, would find their way into plants and ultimately move to food chain and affect the animals as well as human beings.

The soil under study showed that the surface as well as sub soils are contaminated with various heavy metals particularly Cd, Cr, Cu, Mn and Zn. Qadir *et al.* (1999) and Ghafoor (2004) reported that the urban soils of Faisalabad irrigated with city effluents for growing vegetables for more than 30 years have attained concentration of Cu, Fe and Mn above the safe limits.

Heavy metals content of plants

Heavy metals content of leaves and fruits of various vegetable (Table 7) showed that all the metals (Ni, Cd, Cr, Cu, Pb, Fe, Zn and Mn) were above the safe limits in leaves as well as in fruits of all the vegetables studied (tomato, ridge guard, okra and battle guard) based on the standard of WHO (1996). This may be the result of growing these vegetable with effluents over a long period of time. Soils contaminated with heavy metals are likely to affect the crop quality. This aspect is very important and needs special attention where food crops especially vegetables are grown on heavy metals contaminated soils (Qadir *et al.*, 1999; Ahmad and Rizvi, 2003; Ghafoor, 2004; Bhatti and Mussarat, 2006). Mussarat and Bhatti, (2005) and Damerzen and Aksoy (2005) showed that concentration of Cr, Cd, Ni, Pb and Zn in vegetable grown in the urban areas were higher than those of rural areas as a result of pollution and can be related to their concentrations in the corresponding soils.

From the present results compared with the similar work of local scientists and foreign workers, it is apparent that the soils contaminated with heavy metals due to irrigation with untreated waste water affect the quality of crops grown on these soils. This is of great concern as it is chain system i.e. untreated waste water contaminates soils, and soils in turn affect plants grown on these soils which gets their way in the food chain of human beings and animals.

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