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RESEARCH ARTICLE

HEAVY METAL CONCENTRATIONS IN CONTAMINATED SOIL AND VEGETABLES OF TANNERY AREA IN DHAKA, BANGLADESH

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ABSTRACT

The present investigation was conducted to determine levels of heavy metal pollution in tannery area of Dhaka city. Concentration of Fe, Cr, Cu, Cd, Pb, Ni and As have been estimated in soil and vegetables grown around the leather industrial area. The investigated soil and vegetables samples were collected in open place of different distances from the tannery area. The order of metal contents was found Fe > Cr > Pb > Cu > Ni > As > Cd both in soil and vegetables. The results were compared with the recommended maximum tolerable levels proposed by the joint FAO/WHO Expert Committee on Food Additives. The experimental results indicate that the concentration of Pb, Cu, Ni, As and Cd both in soil and vegetables were within the limit however the concentration of Cr and Fe were extremely higher than the tolerance limit.

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INTRODUCTION

During the last few decades Leather industries have been attracted a considerable attention in regarding environmental pollution. Hides and skins are important by-product of meat industry. The tanning processes consist of converting raw hides and skins into leather. During operations a lot of organic and inorganic chemicals/compounds are used. Many of them are not fully consumed by processed leather. Moreover, after completion of process without any further treatment they are discharged from the industries. Nearly 90% of all leather produced is tanned using chromium salts (Stein and Schwedt 1994). Generally 8% of the basic chromium sulphate salt is used for conventional tanning. It binds with the collagenous protein to convert to leather. About 60% to 70% chromium compound are consumed by hides and skins and rest amount of chromium discharge directly from the industry which pollutes the environment (Adeel *et al.*, 2012). Other metals pollutant of concern within the tanning industry includes cadmium, cobalt, copper, barium, arsenic, nickel, lead, mercury, iron etc. (Mwinyihija 2010). Several ways soil of lowland are contaminated by tannery waste water. Heavy metal pollution of agricultural soil and vegetable is one of the most ecological problems on a world scale and also in Bangladesh. Industrial wastewater carries an appreciable amount of toxic heavy metals which create a problem for safe rational utilization of agricultural soil (Singh *et al.*, 2004; Yadav *et al.*, 2002; Chen *et al.*, 2005). Sources of metal pollution in soil and plants include rapid urbanization, industrialization with improper environmental planning, irrigation with wastewater application of excessive agrochemicals and sewage sludge in farming and other human activities (Arora *et al.*, 2008; Khan *et al.*, 2008). Crops growing in polluted water and soil usually absorb an excessive level of metals which ultimately enter into the edible parts of plants without showing visible symptoms of damage (Manapanda *et al.*, 2005; Sharma *et al.*, 2008). Deposition and translocation of metals in different parts of crop plants depend upon the availability and concentration of metals as well as particular plant species and its population (Goni *et al.*, 2014; Chandra Sekhar *et al.*, 2001; Liu *et al.*, 2007). Again, the uptake of metal concentration, soil characteristics and type of plant species (Lin *et al.*, 2003). A number of studies have been reported on the deposition of metals in soil, crops, and vegetables grown in the vicinity of industrial areas around the world (Ahmad *et al.*, 2010; Ayari *et al.*, 2010; Gupta *et al.*, 2008; Smith 2009). However, there is limited published information on the contamination of vegetables and crops in the vicinity of tanning industrial area of Bangladesh.

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The objective of this study was to determine the concentration of Cr, Cu, Fe, Pb, Ni, Cd and As in soil and vegetables (root and edible parts) of the Hazaribagh tannery industrial area of Bangladesh and to identify the interaction of metals between soil and vegetables. It is mentioned that compounds of Cr, Cu, Pd, Cd, and Ni are used in tanning industries and unconsumed metal compounds discharge from the industry without any treatment which pollutes the local land.

MATERIALS AND METHODS

The present study dealt with the soils and vegetables grown around Hazaribagh leather industrial area. Leather industry discharges a significant amount of metals and others toxic material with their effluents through the open drains, thereby contaminating water. This wastewater pollutes lowland. Sometimes during heavily rain or blockage in drain, effluents overflow and pollutes nearby land where people cultivate vegetables. Four samples of the identical species (soil, vegetable, spinach) were collected from different places of the sample collecting area by hand using vinyl gloves, then carefully packed into polyethylene bags. The removal of soil from the crop samples was performed by shaking. Then the root and edible parts of the vegetable plants were separated. Plant organs washed with tap water for several times and subsequently dipped into 0.001 M HCl for 5 min followed by thoroughly washing with distilled and de-ionized water to remove airborne pollutants. Samples were cut into 2 cm pieces and dried in a hot air oven at 70-80°C until a constant weight was achieved. Dried samples were grounded in a stainless steel blender and then passed through a 2-mm-size sieve. Soil samples from the surface to a depth of 10-15 cm around each were collected. It was dried at room temperature and finally at 105°C for 3 hrs to remove the moisture content. Dried samples were homogenized and wet digested.

Digestion procedure: 0.2 g of soil, root, and branch samples were taken in a vessel of microwave. Digestions were performed according to ISO-3050, 8 mL of concentrated HNO_3, 1 mL of concentrated HCl and 1 mL of H_2O_2 were added into the vessel. The vessels were kept in the microwave and it was run for one hour. Samples were filtrated using Whitman no. 42 filter paper and the filtrate transferred into the volumetric flux and finally diluted to 50 mL with de-ionized water. All samples were analyzed using ICP-OES (Optima-7000 DV, USA). All reagents used were Merck, analytical grade including standard stock solutions of known concentrations of different metals.

RESULTS AND DISCUSSION

Heavy metal in tannery wastewater contaminated soil

Out of eight metals examined in tannery effluent contaminated soil in Dhaka leather industrial area, concentration of heavy metals (mg kg^{-1}) were found (Table 1) ranged from 994 to 1120 for Cr, 34.35 to 39.66 for Cu, 46.70 to 55.16 for Pb, 24.10 to 26.73 for Ni, 0.32 to 0.54 for Cd, 1.49 to 2.21 for As, 0.44 to 1.10 for Sb and 20812 to 21216 for Fe. Besides Fe, the mean highest concentrations recorded in soil was for Cr followed by Pb, Cu, Ni, As, Sb and Cd. Concentrations of Pb, Cu and Ni in soil of the present study were very similar to the results reported in previous study for agricultural soil in Bangladesh by Ahmed *et al.,* (2010) and by Ahsan *et al.,* (2009) however Fe and Cr content were much higher than those agricultural soils. Chromium concentration was found ranged from 815 to 1120 mg kg^{-1} which was extremely high for cultivation. The Cr concentration observed in the tannery industrial area in the present investigation exceeded the permissible level reported elsewhere (Ahmad *et al.,* 2010, Gupta *et al.,* 2008, Sharma *et al.,* 2007 and Pendias *et al.,* 1992). Tanning industry in Bangladesh uses deep underground water and highest deposition of Fe in soil might be due to the long-term use of this ground water which is heavily contaminated with Fe as well as production of metallic tools, use of dyes and pigments in tanning industries of the studied area contaminates the soil.

The comparison of mean concentration of heavy metals in soil of presently studied study area with the official Indian standard (Awashthi 2000) and maximum allowable concentration (MAC) of elements in agricultural soil in China showed that all the metals are within the limits except Cr and Fe. However, Cu concentration (34.35 to 39.66) in soil was lower than the previous results obtained by Tandi *et al.,* (2004) (2.5-133.3 mg kg^{-1}), Manapanda *et al.,* (2005) (7-145 mg kg^{-1}) in Zimbabwe and by Ahmad *et al.,* (2010) (31.35-45.16 mg kg^{-1}) in Bangladesh.

The higher standard deviation reveals higher variations in heavy metals distribution from the point source of emission to the adjacent areas. Among the eight heavy metals examined in soil, concentration of Fe was maximum (21,575 mg kg^{-1}) and also showing more standard deviation value (383).

Concentration of heavy metals in different parts of vegetables

Concentration of Cr, Pb, Cu, Ni, As, Sb, Cd and Fe of root and edible parts of vegetable of present studied area were investigated and the results are shown in table 2. The concentrations of observed metals were within the safe limits (FAO/WHO Standard 1984, Indian Standard Awashthi 2000, SEPA 2005) except Cr and Fe. The range of Cr concentration in present study found from 31.08 to 56.06 mg kg^{-1} in edible part which was lower than the Cr contents (34.83-96.30 mg kg^{-1}) found in the vegetables from Titagarh, West Bengal, India by Gupta *et al.,* 2008 but higher than the results (0.086-2.93 mg kg^{-1}) reported by Singh *et al.,* (2011) and (4.16-6.45 mg kg^{-1}) reported by Goni *et al.,* (2014). The Cu content in vegetables (10.27 - 29.10 mg kg^{-1}) was very similar to the reported result in DEPZ, Bangladesh (8.63-27.94 mg kg^{-1}) (Goni *et al.,* 2014) and in Titagarh, West Bengal, India (15.66-34.49 mg kg^{-1}) (Gupta *et al.,* 2008) but lower than the Cu content in vegetables (61.20 mg kg^{-1}) from Zhengzhou city, China (Liu *et al.,* 2006). The obtained results of Cu concentration of present study also in good agreement with the values observed in Varanasi, India (10.95 – 28.58 mg kg^{-1}) by Sharma *et al.,* (2007). The Pb concentration in edible parts of spinach examined in the present study ((7.32-17.00 mg kg^{-1}) were above the permissible levels recommended by FAO/WHO, India (Awashthi 2000) and China (SEPA 2005).

Table 1. Concentration of heavy metals in tannery wastewater contaminated soil (mg kg^{-1} dry soil) in Dhaka tanning industrial area, Bangladesh (n = 4)

Metal	World Limit[a]	MAC of elements in agricultural soil in China[b]	Threshold of elements in natural background soil in China[b]	Safe Limit of India[c]	Non contaminated Soil		Contaminated soil (Present study)				
					Soil[d]	Soil[e]	Max.	Median	Min.	Mean	SD
Cr	70	200	90	-	100	100	1120	994	815	976	153
Cu	30	100	35	135-270	20	30	39.66	38.7	34.35	37.57	2.83
Pb	35	350	35	250-500	100	50	55.16	49.1	46.7	50.32	4.36
Ni	50	60	40	75-150	40	30	26.73	25.32	24.1	25.38	1.32
Cd	0.35	0.6	0.20	3-6	0.06	1	0.537	0.49	0.318	0.45	0.11
As							2.21	2.12	1.49	1.94	0.39
Sb							1.10	0.51	0.44	0.68	0.36
Fe	1000	-	-	-	-	1000	21575	21262	20812	21216	383

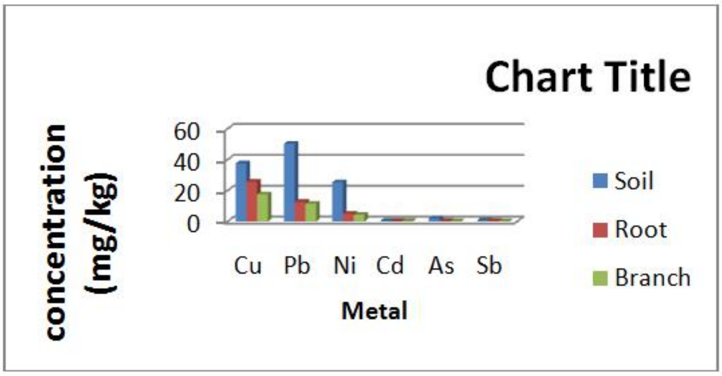
[a]Coskun et al. (2006); [b] National Environmental Protection Agency of China, GB 15618 (1995); [c]Awashthi (2000); [d]Temmerman *et al*. (1984); [e]Pendias *et al.* (1992)

Table 2. Metals concentration (mg kg^{-1} dry weight) in vegetables (spinach) in the tanning industrial area in Dhaka

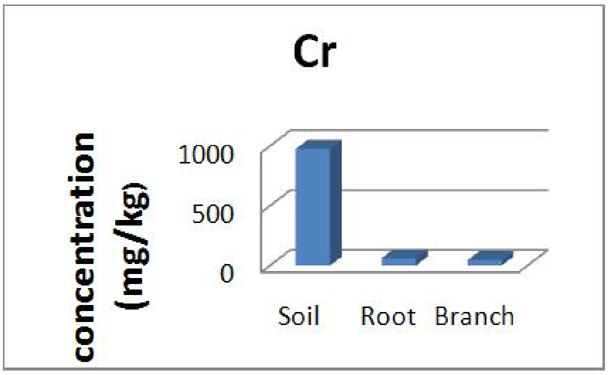
Metal	Safe limit[a]	Safe limit[b]	Safe limit[c]	Root					Edible part				
				Max.	Median	Min.	Mean	SD	Max.	Median	Mini.	Mean	SD
Cr	5	20	0.5	72.56	59.01	51.4	60.99	10.72	56.06	46.3	31.08	44.48	12.59
Cu	40	30	20	30.63	23.68	22.8	25.70	4.29	29.1	13.52	10.27	17.63	10.07
Pb	5	2.50	9	23.56	8.40	6.14	12.70	9.47	17	10.12	7.32	11.48	4.98
Ni	20	1.50	10	6.88	4.12	4.09	5.03	1.60	4.80	4.15	3.95	4.30	0.45
Cd	0.3	1.50	0.20	0.395	0.39	0.3	0.36	0.052	0.40	0.35	0.216	0.32	0.094
As				0.73	0.29	0.29	0.44	0.25	0.51	0.16	0.113	0.26	0.22
Sb				0.72	0.36	0.36	0.48	0.21	0.496	0.122	0.106	0.24	0.22
Fe	425	-	-	3608	3308	3294	3403	177	1209	1146	1126	1160	43.32

[a]FAO/WHO Standard (Codex Alimentarius Commission) 1984, [b]Indian standard (Awashthi 2000); [c]SEPA (2005)

Summary of the concentration of heavy metals in soil and vegetables



Metals	Soil (mg/kg)	Root (mg/kg)	Branch (mg/kg)
Cr	976	61	44.5
Cu	37.6	25.7	17.6
Pb	50.3	12.7	11.5
Ni	25.4	5.03	4.3
Cd	0.45	0.36	0.32
As	1.94	0.44	0.26
Sb	0.68	0.48	0.24
Fe	21216	3403	1160



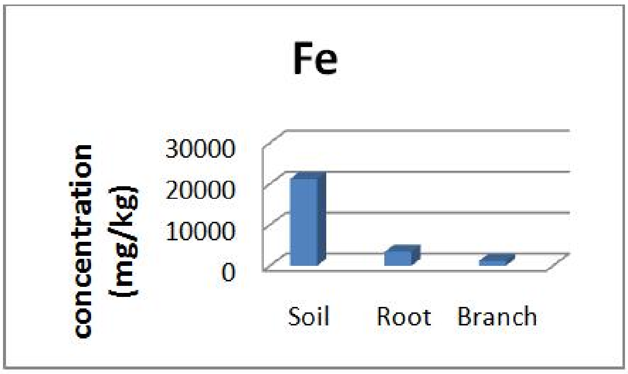


Table 3. Correlation of metals found in the edible parts of spinach

Metals	Cr	Cu	Pb	Ni	Cd	As	Sb	Fe
Cr	1							
Cu	.284	1						
Pb	.399	.993	1					
Ni	-.985	-.444	-.550	1				
Cd	.790	.812	.877	-.884	1			
As	.232	.999*	.985	-.395	.779	1		
Sb	-.907	-.661	-.748	.966	-.975	-.620	1	
Fe	.636	-.560	-.455	-.494	.029	-.604	-.252	1

*. Correlation is significant at the 0.05 level (2-tailed), N=4, No. of samples

The mean Pb content in vegetable (7.32-17.00 mg kg $^{-1}$) was lower than the values reported in Titagarh, West Bengal, India (21.59-57.63 mg kg $^{-1}$) (Gupta *et al.*, 2008), in DEPZ, Bangladesh (24.08-27.35 mg kg $^{-1}$) reported by Goni *et al.*, 2014) but comparatively higher than the Pb level reported in China (0.18-7.75 mg kg $^{-1}$, Liu *et al.*, 2006; 1.97-3.81 mg kg $^{-1}$, Liu *et al.*, 2005) and in Varanasi, India (3.09-15.74 mg kg $^{-1}$) (Sharma *et al.*, 2007). The Ni content in spinach in the current study was found 3.95- 4.80 mg kg $^{-1}$ which was lower than the permissible levels recommended by FAO/WHO and China (SEPA 2005). However, it was very similar to the findings of Sharma *et al.*, (2007) (1.87-7.57 mg kg $^{-1}$) in Varanasi, India. Concentration of Cd, As, Sb were found in the vegetable of study area in tress amount 0.218 – 0.40 for Cd, 0.113-0.51 for as and 0.106 - 0.496 for Sb.

Heavy metals transfer from soil to vegetables

The transportation of investigated heavy metals in the study area was found in the order of soil > root > edible part. Bio-concentration factor (BCF) or plant concentration factor (PCF) is a parameter used to describe the transfer of trace elements from soil to plant body. It is calculated as the ration between the concentration of heavy metals in the vegetables and that in the corresponding soil (all based on dry weight) for each vegetable (spinach) separately (Liu *et al.*, 2006). BCF or PCF = C_{plant}/C_{soil}, where C_{plant} and C_{soil} represent the heavy metal concentration in extracts of plants and soils on a dry weight basis respectively. The PCF values of the investigated heavy metals in current study for the root of spinach was found Cr 0.06, Cu 0.60 – 0.79, Pb 0.13 – 0.43, Ni 0.15 – 0.27, Cd 0.74 – 0.94, As 0.13 – 0.34, Sb 0.65 – 0.82 and Fe 0.15 – 0.17. Metal uptake by the root of spinach was in the following order: Cd > Sb > Cu > Pb > As > Ni > Fe > Cr. Uptake of Cr is lowest among the investigated metals. This result which was supported by the investigation done by Liu *et al.* (2007). On the other hand, the BCF values of the investigated metals of edible parts of spinach was Cr 0.03 – 0.06, Cu 0.26 – 0.75, Pb 0.13 – 0.35, Ni 0.15 – 0.19, Cd 0.40 – 1.10, As 0.07 – 0.24, Sb 0.21 – 0.45 and Fe 0.05 – 0.06. Metals uptake by the edible parts of spinach was in the following order: Cd > Cu > Sb > Pb > As > Ni > Fe > Cr. The BCF value of Cd in the edible parts of spinach was higher than 1 which indicates that bioavailability of this metal in spinach was very high. Statistical analysis to establish the association among the parameters was performed by using the software MS Excel and Pearson's correlation (r) by SPSS (18.0 version) (Table-3). The accumulation of Cd in edible parts of spinach was higher than other metals and was also above the safe limit of FAO/WHO. The similar result was found in our previously published article, Goni *et al.*, (2014). The obtained results shows that the transportation of the investigated metals of the studied area followed the order of soil > root > edible parts of spinach. Variation on the translocation of the metals from roots to the edible parts might be due to the interaction between different metals occurring at the root surface and also within the plant, which ultimately affects uptake and mobilization of metals (Ashraf *et al.*, 2011; Sharma *et al.*, 2007).

Conclusion

Heavy metals in the soil of tannery effluents affected area in Dhaka are higher than the acceptable limits. In particular, the Cr and Fe content in the soil of experimental area are significantly high. Source of Cr content is basic chromium sulphate which is one of the most fundamental compounds used in tanning industry. About 60-70% of the said Cr compound consumes in leather and rests of the amount remain in wastewater which is directly discharge to the low land without any further treatment. Deep machine water is used in the tanning industries which are highly contaminated with Fe. Concentration of Cr, Fe, Cd in root and edible parts of spinach are higher than the accepted level. Variation was observed in the uptake and mobilization of metals from soil to different parts of vegetables. The accumulation and absorption of all metals were higher in roots compared to edible part of spinach. But the translocation of the metals especially Cr from soil to edible part is not significant.

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