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A COMPARATIVE STUDY ON THE CONTRIBUTION OF TREATED AND UNTREATED TANNERY WASTES ON ENVIRONMENT

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Abstract

Wastes, effluents, water, soil and plant samples were collected from the Hazaribagh tannery area and the Nayarhat tannery area. Analyses of the samples reveal that the environment is much more cleaner in the area where it is receiving the treated effluents. The necessity of establishing effluent treatment plants in industries, particularly in the tannery industries are essential for a friendly environment and health.

Key word: Tannery, Hazaribagh, Nayarhat, soil, plant, heavy metals

Introduction

Environmental pollution through industrialization is a serious problem throughout the world (Kabata and Pendias 1984). Soil is the primary recipient of many of the waste products and toxic chemicals or pollutants used in modern industry and thus become contaminated. The heavy metal pollutants in soil affect plants and soil biota which are associated with almost all of the soil and plant processes and affect the consumers through contamination of the food chain. (Ibekwe *et al.* 1995).

In Bangladesh pollution control issues are relatively recent. With few exceptions (only 1.75%), the industries are not equipped with pollution control systems (DoE 1992). The Ministry of Environment and Forest (1991) reported that these effluents pollute our soils as well as groundwater. Once the ground water is polluted, it is virtually impossible to purify it even in the highly technically advanced industrial countries. The rivers around the industrial belts of Dhaka, Narayanganj, Chittagong and Khulna are major receivers of the untreated effluents coming from tanneries, textiles, chemicals, pesticides, medicines, foods, engineering etc. Among all the polluting industries, tanneries- one of the most important and largest one, contribute to it to a greater extent (Imamul Huq 1998).

Land shortage has forced farmers to use the fallow lands around industries that are within the reach of waste disposal. Such a situation demands evaluation of the extent of soil and crop contamination by among others, toxic heavy metals coming out of the industrial units. Most industries in Bangladesh do not have effluent treatment plants (ETPs) and as such the contamination level around these industries is quite high. Very few of the tanneries have effluent/waste treatment plants in the country. Of late, a few

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tanneries have these plants installed. It is thus pertinent to have a comparative study on the effect of treated and non-treated waste disposal on soil characteristics and plant growth. The outcome of such a study is likely to create awareness among the public as well as the policy makers about the importance of installing ETP in the various polluting industries.

With this view in mind, a field study was performed to compare the contributions of the tannery industries having treatment plant or not, to the environmental pollution.

Materials and Methods

The study was carried out in two different areas: (1) Hazaribagh, inside the Dhaka city where most of the tanneries are situated and where not a single tannery has any ETP; and (2) Nayarhat, outskirts of Dhaka where an international tannery industry with modern ETP is operational. Solid wastes, effluents, soil and plant samples were collected from different sites of the two tannery areas:

i) ***Hazaribagh Tannery Area***- largest and worst polluted industrial area of Dhaka city is situated on the side of the Buriganga River. The tannery factories are located inside the greater Dhaka Flood Protection Embankment. The study area is around 1 Km^2 section of Hazaribagh from the main disposal spot. Solid wastes and effluents were collected from the main outfall area of three tannery factories. Five sites were selected for collecting the soil, water and plant samples in this area; one of the sites was outside the embankment and four were inside the embankment. One of the sites inside the embankment was a confined low land surrounded by tannery industries which was around 150 meters away from the tannery factories. Gazmahal, the 2^{nd} site was nearly 300 meters away and the 3^{rd} site, Sonatangor was about 500 meters away from the tannery factories. These two sites were situated at the north of the non-metalled road and the 4^{th} site, Kalinagar was situated at the south of the non-metalled road, which was about 500 meters away from the 2^{nd} site. Water samples were also collected from the main drain.

ii) ***Nayarhat Tannery industrial Area***- with waste treatment facilities, is situated at Nayarhat in Kulla Union, Dhamrai on the bank of the Bongshi river. The study area is approximately 0.5 Km^2 section of the area from the main disposal point. Solid wastes were collected from sludge drying bed. The effluent was collected from the final settlement tank. Four sites were selected for collection of soil, water and plant samples. The 1^{st} one was the effluent spraying area where the treated effluents are disposed off. The 2^{nd} site was the cultivated land area, which was about 150 meters away from the effluent spraying area. The 3^{rd} site was about 300 meters away from the 1^{st} site. It was a low-lying fallow land area. The treated effluents that are falling into the river water pass through all these sites. The last sampling site was the riverbank of the Bongshi. It is situated at a distance of about 500 meters from the effluent treatment area.

Solid wastes and effluents were collected from the source points of the industries of the two areas. Solid wastes and effluents were taken in plastic containers. Bulk soil samples were collected at a depth of 0-15 cm and were collected with the help of the spade. Water samples were collected in plastic bottles from different spots to a depth of 10-15 cm below the surface. The sampling time was between 12.00 noon and 3.00pm. Plant samples were collected from both the Hazaribagh and the Nayarhat tannery areas where soils were supposed to be affected by tannery wastes.

Each of the collected soil samples was air dried and sieved through a 2 mm sieve and then made composite. These samples were used for physical, chemical and physico-chemical analyses.

Aerial portions of the plant samples from the vicinity of the industries were collected. The plants were cut at one cm above ground level. Collected samples were washed with water, dried and then weighed and ground. It was made composite and was stored for chemical analyses.

***Analyses for Physical and Physico-Chemical Properties of Soil and Waste*:** Moisture contents of air-dried samples were determined as describe by Black (1965). The particle size analyses of soils were conducted by hydrometer method as described by Black (1965) and the textural classes was determined by Marshall's Triangular Coordinates as devised by the United States Department of agriculture (USDA 1951) The pH of soils and waste was measured electrochemically by using combined electrode digital pH meter as suggested by Jackson (1962). As the wastes absorbed more water than soil, this ratio didn't hold good. Therefore, the ratio was maintained at 1: 5 for waste to water. Electrical conductivity of the saturated extract of soils was measured at a soil :water ratio of 1:2 by an EC meter as described by USSL staff (1954).

Total potassium and sodium contents were extracted by digesting the soils and wastes with HNO_3: $HClO_4$ (2: 1). The extract was analyzed for total K and Na by a flame analyzer at 589 and 767 nm respectively (Jackson 1962).

Total contents of Fe, Mn, Zn, Cu, Pb and Cd of the samples were determined by Atomic Absorption Spectrophotometer (Jackson 1962), after digestion with HNO_3: $HClO_4$ (2: 1) acid mixture.

***Analyses of Plant Sample*:** After wet oxidation with ternary acid mixture (HNO_3: H_2SO_4: $HClO_4$= 5: 1: 2) as described by Imamul Haq and Alam (2005), the total contents of Na, Fe, Mn, Zn, Pb, Cu and Cd were determined by using the procedure as mentioned for soil.

***Analyses of Effluent and Water Sample*:** The pH of effluent and water samples were measured electrochemically by using combined electrode digital pH meter. Electrical conductivity of effluent and water samples was measured directly by an EC meter. Total dissolved solids was measured electrochemically by using TDS meter. Sodium and K were determined directly by flame photometer. Calcium and Mg were determined by EDTA (ethylene di-amine tetra acetic acid) method (Black 1965). Total Fe, Mn, Zn, Pb, Cu and Cd were determined directly by atomic absorption spectrophotometer.

***Data Analysis*:** Microsoft Excel and MINITAB (version 13) were used for data manipulation, graphing and statistical analysis.

Results and Discussion

In the field experiment a comparative study was made between an untreated tannery wastes & effluents and a treated tannery wastes & effluents to see their *in situ* effects on water, soil and plants. The average values of the analyses of the collected samples are presented in tables through 1, 2, 3, 4,5, 6 and 7.

***Wastes and Effluents*:** It was observed that pH of the untreated wastes from Hazaribagh had wide ranging values (7.8 to 11.4). The pH at the source was very strongly alkaline but decreased to moderately alkaline level at the point of disposal whereas the pH of the treated wastes from the Nayarhat area ranged from medium acidic to slightly acidic (5.6 and 6.1). The pH of the two wastes differed significantly ($t = 2.63$, $p=0.119$)

Table 1. The average value of some physico-chemical properties of wastes collected *in situ* at Hazaribagh and Nayarhat tannery industrial areas

Properties		Hazaribagh tannery industrial area	Nayarhat tannery industrial area
pH		9.03	5.85
EC (dS/m)		490	425
CEC (meq/100g)		33.24	47.52
Cations (meq/100g)	Na	6.51	1.41
	K	0.39	0.29
	Ca	4.05	1.12
	Mg	2.01	0.18

Accumulation of high amount of sodium might increase the pH of untreated wastes. Electrical conductivity (EC) of wastes from Hazaribagh was found slightly higher than wastes from Nayarhat (Table 1). From t-test no significant difference was found. (t-value= 0.74, p-value= 0.534). The CEC of wastes of both types have high value and no significant difference was found from t-test (t-value=1.27, p-value= 0.331). Among the cations Na concentration was found to be significantly higher in untreated wastes than in treated wastes (t-value=3.02, p-value=0.094). Other cations are also present in comparatively higher concentration in untreated wastes (Table 1). The t-test values for K, Ca and Mg are 1.14 (p-value=0.459), 1.85 (p-value=0.206) and 1.90 (p-value=0.198) respectively.

Table 2. The average value of some selected chemical characteristics of effluents collected *in situ* at Hazaribagh and Nayarhat tannery industrial areas.

properties	Hazaribagh tannery industrial area	Nayarhat tannery industrial area
pH	6.6	7.05
EC (dS/m)	3674	235
TDS (g/L)	17.07	9.8
Na (mg/L)	1522	376

For the untreated effluents, the pH was initially extremely acidic (3.7) but at their disposal points they were strongly alkaline (8.2). The pH values of treated effluents (Table 2) were near neutral at both the points (7.0 and 7.1). The t-value was found to be 0.71 (p-value= 0.505). EC of effluents from two types of tanneries showed distinctive variation and significant difference was found in t-test (t-value=3.13, p-value=0.02). The higher EC value was attributed to the high concentration of soluble salts in the Hazaribagh tannery effluents (Table 2). Total dissolved solid (TDS) was much higher (Table 2) in untreated effluents (ranged from 5.85 g/L to 30.56 g/L) than in treated effluents (9.8 g/L). In t-test the t-value was found to be 1.52 (p-value= 0.180).

In effluents, Na was the most dominant cation. Sodium, K, Ca and Mg ions were all found to be present in much higher concentration in untreated effluents than in treated one (Figure 1). But K concentration was found to differ significantly (t-value=2.60, p-value=0.041). For Na, Ca, and Mg the t-values were found to be 1.69 (p-value=0.143), 1.81 (p-value=0.118) and 1.52 (p-value=0.267) respectively

Table.3. Average value of some heavy metals content of wastes and effluents collected *in situ* at Hazaribagh and Nayarhat tannery industrial area.

Heavy metals	Hazaribagh tannery industrial area		Nayarhat tannery industrial area	
	Wastes	Effluents	Wastes	Effluents
Fe (ppm)	19073	2.57	2957	0.13
Mn (ppm)	351.57	1.83	19.00	0.10
Cu (ppm)	797.97	2.72	38.69	0.02
Cd (ppm)	7.54	0.03	0.001	0.0
Pb (ppm)	181.40	1.05	68.85	0.0
Zn (ppm)	99.70	1.83	16.38	0.002

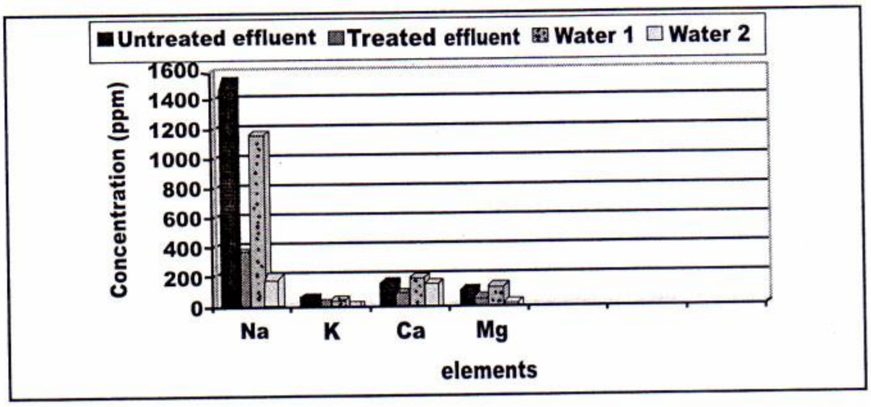

Figure 1. Elemental concentrations in untreated and treated effluents and contaminated samples. (Water 1: receiving the untreated effluent, water 2: receiving the treated effluent).

Soil Samples

Soil characteristics of the Hazaribagh and the Nayarhat tannery areas are shown in Table 4. In the Hazaribagh five locations were identified, out of which there were 15 specific spots from where soil samples were collected. Spot 1 to 11 were inside the embankments and spot 12 to 15 were outside the embankments. Particle size distribution analyses of these sites showed the textural classes to be basically loam with occasional sandy and silt loam. In the Nayarhat tannery area four locations were identified, out of which there were 12 specific spots from where soil samples were collected. The textural classes were found to be silt loam with occasional silty clay loam.

Table 4. Some physico-chemical properties of soils collected from Hazaribagh and Nayarhat tannery areas at different distance.

Properties	Hazaribagh tannery area			Nayarhat tannery area		
	150m	300m	500m	150m	300m	500m
pH	6.03	6.30	7.10	6.73	5.70	7.9
EC (dS/m)	407	773	560	46	233	233
CEC (meq%)	7.89	8.12	8.84	11.79	7.78	12.92
Exch. Na (meq/100g)	0.67	1.28	2.05	0.24	0.23	0.24

The pH values pf the soils of both the Hazaribagh and the Nayarhat tannery areas showed wide range of variation The former varied between 5.5 and 7.6 while the latter varied between 6.3 and 8.3. No significant differences were found from the t-test and also no specific relationship was found with distance in both the areas (Table 4). A significant difference was however, found for the EC values (Table 4). The electrical conductivity was higher in most of the spots of the Hazaribagh (ranging from 30 to1150) than the Nayarhat tannery area (ranging from 50 to 400).

In case of CEC, although no significant difference was found it however increased with increasing distance from the disposal point in the Hazaribagh (Table 4) area. Exchangeable Na as well as other exchangeable cations showed significant difference (t-values and p-values for Na, 2.34 and 0.034). The exchangeable cation concentrations were found higher in the Hazaribagh soils (Figure 2).

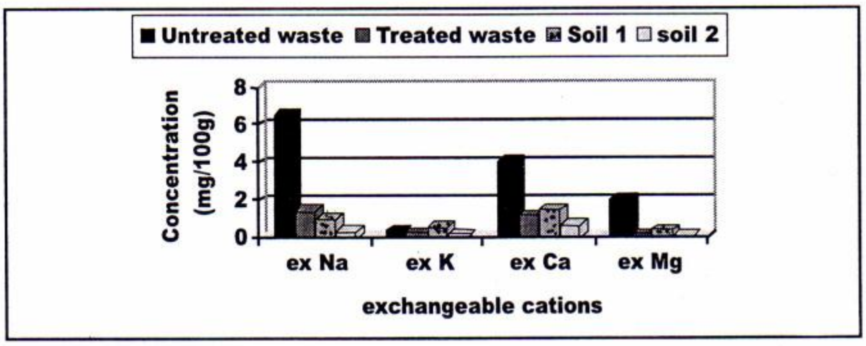

Figure 2. Exchangeable cation concentrations in untreated and treated wastes and soils contaminated by the sewage wastes. (Soil 1: receiving the untreated wastes, Soil 2: receiving the treated Wastes).

***Water Samples*:** In the Hazaribagh area five locations were identified, out of which there were 11 specific spots from where water samples were collected. Spots 1 to 7 were inside the embankments and spots 8 to 11 were outside the embankments. In the Nayarhat tannery area four locations were identified, out of which there were 9 specific spots from where water samples were collected (Shabnam 2005).

Water of the Hazaribagh tannery area showed wide range of pH value (6.6 to 8.5) and pH values of the Nayarhat tannery area showed mildly alkaline pH values (ranging from 7.4 to 8.0) throughout the sampling area. No significant difference was found from the t-test ($t= 0.47$, $p= 0.644$). Total dissolved solids (TDS) were found to be present in higher amount in water samples of the Hazaribagh than the Nayarhat area. The average value for TDS of the Hazaribagh sample was 8.98 mS/cm and for the Nayarhat samples it was 5.65. Like that in soil, a significant difference was also observed in case of EC. The electrical conductivity was found to be higher in most of the spots of the Hazaribagh (ranging from 730 to1600 dS/m) than the Nayarhat tannery area (ranging from 20 to 240 dS/m).

Total elemental concentrations were also higher in the Hazaribagh area water samples compared to that in the Nayarhat area samples. The difference was found to be highly significant (the t-value is 3.12 at $p = 0.006$). For water samples chloride, carbonate and bicarbonate anions were analyzed. Carbonate ion was found only at Sonatangor and outside the embankment where pH was found to be above 8.0. Bicarbonate ion did not show any significant difference in t-test. The Cl^- ion was found in huge concentrations in the Hazaribagh area and it differed significantly from those of the Nayarhat area ($t=2.25$, $p= 0.037$).

***Plant Samples*:** Variation in the types of plants was not observed in plants growing in the Hazaribagh areas. Mainly grass samples were collected from inside the embankment. Arum and *Bishkatali* were found scattered. Water hyacinth and grasses were collected from outside the embankment. No edible crops were found either inside or outside the embankment. On the other hand, a comparatively more variation was observed in plant species grown in the Nayarhat tannery area. Boro rice cultivation was observed just beside the tannery boundary. Water hyacinth, arum and rice were found to grow within 300 meters from effluent disposal point. Maize and Dhaincha were collected from the side of the river.

Among the heavy metals Fe, Mn, Cu, Cd, Pb and Zn were analyzed. The phyto-availability of Cr is very little and the risk to human and animal health is minimal (USEPA 1993; Naidu and Kookana 1999; Banu *et al.* 1999); for this reason Cr was not analyzed.

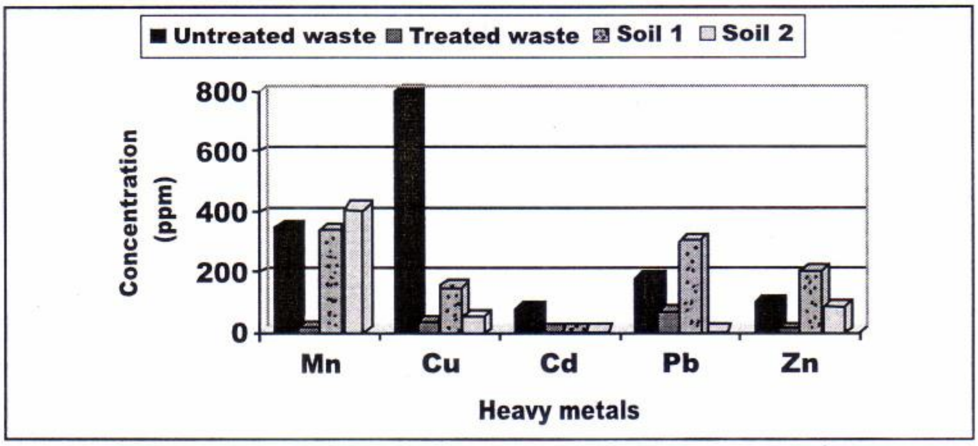

Figure 3. Heavy metals concentrations in untreated and treated wastes and soils contaminated by these wastes. (Soil 1: receiving the untreated wastes, Soil 2: receiving the treated wastes).

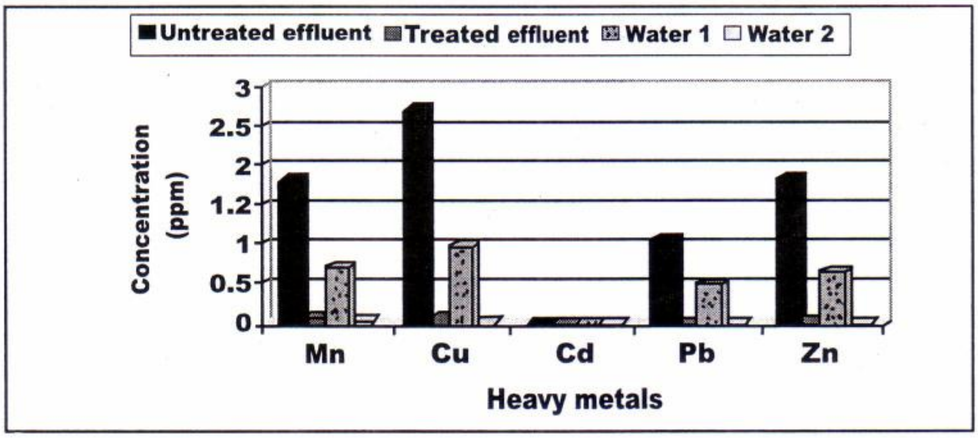

Figure 4. Heavy metal concentrations in untreated and treated effluents and contaminated water samples. (Water 1: receiving the untreated effluent, water 2: receiving the treated effluent).

The tannery industries in the Hazaribagh area discharge huge amount of heavy metals, especially Pb and Cd, which were found in significant amount in both of their wastes (Figures 3 and 4) and effluents. The t-values for Fe, Mn, Cu, Cd, Pb and Zn in wastes were 1.72 (p-value= 0.0.227), 2.07 (p-value= 0.175), 2.35 (p-value= 0.143), 15.26

(p-value=0.004), 13.34 (p-value= 0.006) and 3.56 (p-value=0.071) respectively. The t-values for Fe, Mn, Cu, Cd, Pb and Zn in effluents were 1.35 (p-value= 0.0.226), 1.56 (p-value= 0.169), 1.84 (p-value= 0.116), 5.39 (p-value=0.002), 3.58 (p-value=0.012) and 2.79 (p-value=0.032) respectively.

In soil, the average Fe and Mn concentrations were found to be higher in the Nayarhat tannery area (Figure 3). This might be due to soil variation as the wastes and effluents of the Nayarhat tannery were found to contain lower concentrations of these metals than the wastes from the Hazaribagh. Other heavy metals were found to be present in very significant concentrations in soils receiving the untreated wastes. The t-values and corresponding p-values for Cu, Cd, Pb and Zn are t= 1.91, 3.13, 2.47 and 3.06; and p= 0.077, 0.007, 0.027 and 0.009 respectively. In case of water, contrary to what was observed for soil, the Fe concentration was found significantly higher in the Hazaribagh area than the Nayarhat tannery area (Figure 4). Copper and Mn were found to be present in significant concentrations in water receiving the untreated wastes. In case of Zn, Cd and Pb the concentrations were found to be highly significant in t-test. The t-values and corresponding p-values for Zn, Cd, Pb are t= 2.21, 7.16, 3.43 and p= 0.039, 0.001, 0.003 respectively.

The plant itself can be used as an extracting agent and in this case heavy metal uptake is a direct indicator of the availability of metals in soil adjacent to various pollution sources. Considerable variation has been observed in the heavy metal accumulation in plant samples. Plant sample of the Hazaribagh area has been found to accumulate higher amount of Zn than those from the Nayarhat tannery area. In contrast, Fe concentration was found to be higher in plants of the Nayarhat tannery area; this may be due to higher Fe concentration of the soil of that area. Significant variation has been observed in the uptake of Pb and Cd by plants; which were abnormally higher in plants grown in the Hazaribagh tannery area. Copper accumulation was also found to be higher in plant samples of Hazaribagh area than those of the Nayarhat tannery area. No significant variation was observed in the Mn uptake.

From Tables 5 and 6 the variation in the accumulation of heavy metals by plants within the same distance from the disposal points in the two areas could be well apprecia-ted. From these tables it can be seen that, Fe concentration decreased with distance in the Hazaribagh tannery area. Opposite situation occurred in case of water. Plant accumulation of Fe was found to be higher at the closer distance. Mn concentrations in soil, water and plant were found to be more or less decreased with increasing distance. Zinc concentrations in soil and water were also found to be more or less decreased with distance except for plant accumulation, which was found to increase with distance in the Hazaribagh tannery area. Copper concentration in soil, water and plant was found to

Table 5. Heavy metals concentrations in soil, water and plant samples at different distance in Hazaribagh tannery area.

Distance (m)	Fe (ppm)			Mn (ppm)			Zn (ppm)			Cu (ppm)			Cd (ppm)			Pb (ppm)		
	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant
150	40071	0.00	5199.1	486.89	0.035	204.43	189.48	0.096	74.73	47.77	0.067	26.67	4.90	0.04	1.932	150.97	0.710	144.98
300	24172	0.094	3113.6	273.71	0.031	312.18	298.28	0.031	152.82	441.93	0.072	23.42	1.07	0.019	2.230	152.90	0.464	150.58
500	29571	0.11	1647.1	318.99	0.058	148.08	175.46	0.02	177.06	102.63	0.041	38.75	0.88	0.035	2.220	131.32	0.151	124.90

Table 6. Heavy metals concentrations in soil, water and plant samples at different distance in Nayarhat tannery area.

Distance (m)	Fe (ppm)			Mn (ppm)			Zn (ppm)			Cu (ppm)			Cd (ppm)			Pb (ppm)		
	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant	Soil	Water	Plant
150	26092	0.046	32899.7	375.24	0.058	344.48	78.13	0.009	35.80	49.68	0.014	9.75	0.06	0.0	0.196	9.19	0.0	0.603
300	40387	0.16	8067.3	291.44	0.058	51.51	87.35	0.008	19.97	53.47	0.018	9.14	0.00	0.0	0.012	9.00	0.0	0.0
500	48990	0.013	3141.3	296.24	0.023	111.12	88.48	0.005	27.71	59.75	0.019	13.13	0.02	0.0	0.068	4.98	0.0	0.21

increase with distance in both the areas. Caduium and Pb concentrations decreased with distance in soil and so also the plant accumulation. The variation of the heavy metal concentration with distance might be due to pH variation in soil and water. However, in most of the cases plant accumulation of the heavy metals was observed to increase with their soil contents.

From the observation of the data obtained from wastes, effluents, soil, plant and water samples analyses it can be easily said that the tanneries in the Hazaribagh area are depositing huge amount of untreated wastes and discharging untreated effluents that are significantly polluting the area and increasing the risk to environment and public health there. Whereas, the wastes and effluents from the Nayarhat tannery were found environment friendly.

The wastes and effluents of Hazaribagh area contain comparatively higher concentrations of N, S, P, Na and K. This might be responsible for presence of high amount of these elements in the adjacent soil and water bodies. Higher concentrations of N and P in water samples indicate a vitiated condition prevailing there. High concentrations of Na in soil and water samples made them not suitable for general-purpose uses. Heavy metal concentration in untreated waste samples was found at elevated levels. The Zn, Cd, Pb and Cu contents were found to be above critical limit values in soils of the Hazaribagh area. The water samples showed a similar situation.

The plants growing in the Hazaribagh tannery area were found to accumulate huge amounts of S and heavy metals that exceed toxic limits. No food crops were found to grow in this area; which is indicative of a serious environmental degradation situation of this area.

A completely opposite situation was observed in the Nayarhat tannery area. Due to using ETP, the effluents and wastes coming from the tannery was found harmless to environment.

Conclusions

Industries in Bangladesh have traditionally adhered to a minimum-cost production philosophy, overlooking social/environmental responsibilities. It naturally questions investment in effluent treatment which is considered as a non-productive activity and is responsible for causing serious environmental degradation. Whereas, if these industries had effluent treatment facilities, this could minimize the risk.

The untreated wastes and effluents from the Hazaribagh area contain comparatively higher amounts of Na and the heavy metals which are reflected in higher amounts of these metals in soil and water samples that have rendered them not suitable for general purpose uses. The Zn, Cd, Pb and Cu concentrations were found to be above critical limit values in soils of the Hazaribagh area. The water samples showed a similar situation. The plants growing in the Hazaribagh tannery area were found to accumulate huge amount of S and heavy metals that exceed toxic limits. It should be mentioned that accumulation of higher amounts of heavy metals in grasses might be threatening the animals grazing in the vicinity and also could be a risk to human beings.

A completely opposite situation was observed in the Nayarhat tannery area. Due to the using of ETP, the effluents and wastes coming from the tannery was found harmless to environment.

Protecting the environment from industrial pollution however, is not merely a problem of waste treatment. It is essentially a management problem. The city of Dhaka today is at the threshold of industrialization. More units for new products are being set up continuously. The errors of past should not be repeated; rather the lessons and knowledge from abroad should be utilized to create conditions fit for man and infrastructures fit for environment. In this respect, considering the Bangladesh situation, installing ETPs in industries is strongly recommended.

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