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Metals concentration in the industrial wastewater in Bangladesh and their removal by low cost byproducts

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Abstract

The present study was conducted to assess the contamination of heavy metals in the industrial waste waters collected from Dhaka city, Bangladesh. Three different types of industries (tannery, dye chemical and textiles) have been considered as the major sources of untreated effluents. Metals were analyzed using inductively coupled plasma mass spectrometer (ICP-MS) and compared with different international standard values. The ranges of Cr, Ni, Cu, As, Cd and Pb were 387–1806, 0.77–9.1, 0.69–7.8, 3.6–13, 0.17–5.1, and 0.22–4.5 mg/L for tannery, 1.1–9.1, 0.11–8.6, 0.88–11, 1.0–9.8, 0.01–13 and 0.19–14 mg/L for dye-chemical and 0.93–4.6, 1.1–4.5, 1.9–5.2, 1.5–5.1, 0.01–3.9 and 0.19–4.4 mg/L for textiles industry. The concentration of heavy metals in the industrial wastewater exceeded the acceptable environmental quality standards that pose severe threat to the aquatic environment and human health in the urban area of Dhaka City. Results showed that egg shell can be fruitfully used for the removal of heavy metals from industrial wastewater. All possible sources of agro-based inexpensive adsorbents should be explored and their feasibility for the removal of heavy metals should be studied in detail.

Keywords: Wastewater, Heavy metals, Rice husk, Egg shell, Bangladesh

1. Introduction

Globally, the contamination of heavy metal in water bodies ranks among the major environmental problems with many issues accompanying rapid economic development in both developed and developing countries like Bangladesh [1, 2]. Excessive release of heavy metals into the environment due to industrialization and urbanization has posed a great problem worldwide. As a direct consequence, untreated wastewater from different industries is being discharged into the channels around city which may result in heavy metal accumulation in water, soils and other environmental media that deserve more attention [3]. Thus, the removal of heavy metals from contaminated wastewater has received increasing attention [4]. Additionally, remediation of heavy metals contaminated waters using some environmental friendly and low cost sorbents is a potentially applicable option [5].

Rapid urbanization is enlarging the size of Dhaka City in terms of geographical area and population with about 12 million people in an area of 815.8 km2 [6,7]. Population growth and economic development in the urban area can accelerate the consumption of commodities and increase the waste generation in developing countries like Bangladesh [8]. Wastewater that is disposed from different industries contains heavy metals, organic compounds, macronutrients, micronutrients, organic micro pollutants, microorganisms and eggs of parasitic organisms [9]. So, the accumulation of heavy metals in the industrial wastewater poses a growing environment problem and the disposal of these wastewaters with toxic metals may result in secondary environmental pollution. Thus, contamination of aquatic environment by toxic metals from untreated wastewater of various industries is a worldwide environmental problem. Metals such as chromium, nickel, copper, arsenic, cadmium and lead have been documented as hazardous elements [7, 10]. Unlike organic wastes, heavy metals are non-biodegradable and they can be accumulated in living tissues, causing various diseases and disorders; therefore, they must be removed before discharge to the environment.

Treatment processes for heavy metal removal from wastewater include precipitation, membrane filtration, adsorption, ion exchange and co-precipitation/adsorption. Studies on the treatment of effluent bearing heavy metals have revealed adsorption to be a highly effective technique for the removal of heavy metals from waste stream and activated carbon has been widely used as an adsorbent [11]. Despite its extensive use in water and wastewater treatment industries, activated carbon remains an expensive material. In recent years, the need for safe and economical methods for

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the elimination of heavy metals from contaminated waters has necessitated research Low cost agricultural waste by-products such as sugarcane bagasse [12], Rice husk [13], sawdust [14], egg shell [15], neem bark [16] etc., for the elimination of heavy metals from wastewater have been investigated by various researchers. Literature on heavy metals in the industrial wastewater in Bangladesh is scarce. Therefore, the objectives of the present study are to evaluate the contamination levels of trace metals in the industrial wastewater as compared to international standards and to assess the removal efficiency of heavy metals by low-cost byproducts such as rice husk and egg shell.

2. Materials and methods

2.1 Sampling and pre-treatment

Sampling stations were selected by site survey during August and September, 2016. Thirty wastewater samples were collected from the disposal channels of three different types of industries such as tanneries, dye chemicals and textiles in Dhaka City, Bangladesh (Figure 1). Wastewater samples were filtered immediately after collection using ADVANTEC® 0.45 µm size sterile syringe filter and were transferred immediately into acid cleaned 100 mL polypropylene bottles. After filtration samples were preserved in the refrigerator until other chemical analysis was carried out. The physicochemical parameters such as pH, electrical conductivity (EC) and ash were estimated within the same day of sampling [17, 18]. Analytical grade reagents and Milli-Q (Elix UV5 and MilliQ, Millipore, USA) water was used for solution preparation. For metal analysis, 20 mL of wastewater sample was treated with 1.5 ml 69% HNO_3 (Kanto Chemical Co, Japan) and 4.5 mL concentrated HCl (Kanto Chemical Co, Japan) in a closed Teflon vessel and was digested in a Microwave Digestion System (Berghof-MWS2, Berghof Speedwave® Germany). The digested samples were then transferred into a Teflon beaker and total volume was made up to 50 mL with Milli-Q water. The digested solution was then filtered by using syringe filter (DISMIC® - 25HP PTTF, pore size = 0.45 µm) Toyo Roshi Kaisha, Ltd., Japan and stored in 50 mL polypropylene tubes (Nalgene, New York).

2.2 Batch experiment and instrumental analysis

Adsorption batch experiments were carried out by shaking a series of bottles containing various amounts of rice husk, egg shell and heavy metal ions at normal pH condition. The pH of the slurry was adjusted to a desired value 6.5 and was agitated (agitation rate 200 rpm) in a shaking bath at (28 ±3 °C) for 20 min until the pH was stabilized. Then, Cr, Ni, Cu, A, Cd and Pb ions in the form of chromium chloride, nickel chloride, copper chloride, arsenic chloride, cadmium chloride and lead chloride were added to the bottles to make initial concentrations of 5–30 mg/L and the bottles were further agitated for 2 h until equilibrium was obtained [15,19]. For metals, samples were analyzed by using Inductively Coupled Plasma mass spectrometer (ICP-MS, Agilent 7700 series). Multi-element Standard XSTC-13 (Spex CertiPrep® USA) solutions was used to prepare calibration curve. The calibration curves with $R^2 > 0.999$ were accepted for concentration calculation. Internal calibration standard solutions containing 1.0 mg/L of indium, yttrium, beryllium, tellurium, cobalt and thallium were purchased from Spex CertiPrep® USA. Working standards were prepared daily in 5% (v/v) HNO_3 at 69% ultrapure grade and were used. Multielement solution (Agilent Technologies, USA) 1.0 µg/L was used as tuning solution covering a wide range of masses of elements. A blank also carried out in the sequential extraction experiment. A run included blank and samples were analyzed in duplicate to eliminate any batch-specific error. Before starting the analysis sequence, relative standard deviation (RSD, <5%) was checked by using tuning solution purchased from the Agilent Technologies.

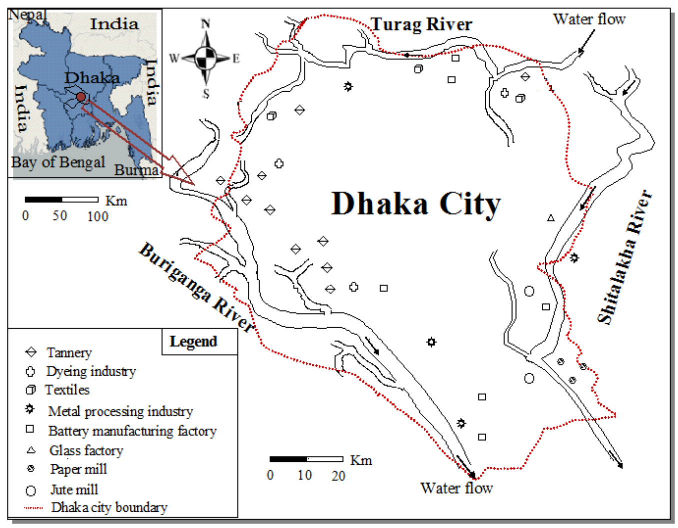

Figure 1: Map of the study area in Dhaka city, Bangladesh

3. Result and Discussion

3.1. Physicochemical properties and heavy metals in wastewater samples

The physicochemical properties of wastewater samples are presented in Table 1. Among the selected industries, the highest mean value of ash was observed for tannery industry (mean: 4.5 g/L and range: 0.91–8.4 g/L) and the lowest value was observed for textile industry (mean: 0.56 g/L and range: 0.21–1.8 g/L). Among the industries the pH ranged from 4.2 to 8.9. The average values of pH of wastewater samples of the studied industries were slightly acidic (Table 1). Due to the lower pH, the wastewater of studied industries might. Detrimental effects and react synergistically with other materials such as heavy metals causing toxicity [20]. The concentrations of heavy metals viz. Cr, Ni, Cu, As, Cd and Pb in wastewater samples are presented in Table 1.

Table 1: Physicochemical parameters and heavy metals (mg/L) of wastewater samples collected from different industries in Dhaka City, Bangladesh

Industries		Ash (g/L)	pH	EC (µS/cm)	Cr	Ni	Cu	As	Cd	Pb
Tannery	Mean±SD	4.5±2.5	6.2±1.3	3.0±2.2	891±436	3.6±2.8	4.5±2.4	5.5±2.9	2.3±1.7	2.0±1.4
	Range	0.91–8.4	4.2–7.9	0.55–6.3	387-1806	0.77-9.1	0.69-7.8	3.6-13	0.17-5.1	0.22-4.5
Dye	Mean±SD	1.4±1.5	6.8±0.91	2.0±1.9	4.9±2.9	3.0±2.5	6.5±3.5	4.7±3.3	4.0±4.3	2.8±4.4
	Range	0.55–4.9	5.1–7.5	0.22–5.2	1.1-9.1	0.11-8.6	0.88-11	1.0-9.8	0.01-13	0.19-14
Textile	Mean±SD	0.56±0.47	7.1±1.0	2.2±0.73	2.5±1.1	2.4±1.5	3.8±0.92	3.4±1.3	1.3±1.4	1.8±1.6
	Range	0.21–1.8	5.6–8.9	1.1–3.8	0.93-4.6	1.1-4.5	1.9-5.2	1.5-5.1	0.01-3.9	0.19-4.4

Note: SD=Standard deviation

Table 2: Permissible limits and health effects of various toxic heavy metals

Metals	Permissible limits for industrial effluents discharge (mg/L)							Permissible limits for potable water (mg/L)					
	Indian standard[33]				Japan[34]		WHO[35]						
	Inland surface water	Public sewers	Marine coastal areas		Inland surface water		Inland surface water	Bangladesh[36]	India[33]	Japan[34]	WHO[35]	USEPA[37]	EU[38]
Cr	2	2	2		2		2	0.05	0.05	0.1	0.05	0.1	0.05
Ni	3	3	5		NA		3	0.1	0.02	0.01	0.02	0.1	0.02
Cu	3	3	3		3		0.05-1.5	1	1.5	1.3	2	1.3	2
As	0.2	0.2	0.2		0.1		0.2	0.05	0.01	0.01	0.01	0.01	0.01
Cd	2	1	2		0.1		0.1	0.005	0.01	0.01	0.003	0.005	0.005
Pb	0.1	1	2		0.1		0.1	0.05	0.05	0.01	0.01	0.015	0.01

[33] Indian standard drinking water-specification (First Revision) IS-10500. New Delhi, India: BIS; 1991.
[34] General standards for discharge of environment pollutants: effluent. Gazette Notification of MoEF, May, 1993.
[35] Clesceri, 1998.
[36] ECR, 1997.
[37] http://www.gemswater.org
[38] http://www.lenntech.com

Table 1 showed a wide variation in the concentration of heavy metals. This difference in concentration of heavy metals among industries may due to the nature of raw materials required and the components used in the operation process [21]. The highest concentration of Cr was observed in the wastewater of tannery industry (891±436 mg/L) followed by dye industry (mean: 4.9±2.9 mg/L). This study has provided the evidence that wastewater discharged from the tanneries and dyeing industries were the main sources of Cr in the wastewater of Dhaka City [22]. Chromium compounds are used as pigments, mordents and dyes in the textiles and as a tanning agent in the leather [23]. The concentration of Cr in the wastewater samples of the industries was very much higher than the permissible limit for portable water of 0.05, mg/L set by WHO, EU, India and Bangladesh (Table 2), indicating the disposable wastewater might create adverse health effects.

The highest concentration of Ni was observed in the wastewater of tannery industry (3.6±2.8) followed by dye industry (3.0±2.5 mg/L). The average concentration of Ni in wastewater samples collected from different industries was very much higher than the maximum permissible limit of WHO, USEPA and EU (Table 2). A considerable amount of Cu was observed in the wastewater of the studied industries which might be attributed to the use of Cu containing chemicals for tanning or battery manufacturing process [7, 24]. The highest concentration of as was observed in the wastewater of tannery industry (5.5±2.9 mg/L) followed by dye-chemical industry (4.7±3.3 mg/L). High level of As in wastewater samples might be attributed to the treatment of wood by using copper arsenate [25] and tanning in relation to some chemicals especially arsenic sulfide [26]. The highest concentration of Cd was observed in the wastewater of dye-chemical industry (4.0±4.3 mg/L) followed by tannery industry (2.3±1.7). Cadmium is contributed to the surface water from the effects of paints, pigments, glass enamel, deterioration of the galvanized pipes etc. [23]. The level of Cd in the wastewater was very much higher than the permissible limit set by WHO, USEPA, EU and other standards (Table 2), indicating the industrial wastewater might create significant health effects due to Cd contamination. Lead is a well-known metal toxicant and it is gradually being phased out of the materials that human beings regularly use. The highest concentration of Pb was observed in the wastewater of dye-chemical industry (2.8±4.4 mg/L) followed by tannery industry (2.0±1.4 mg/L). High level of Pb in wastewater of dye-chemical industry might be attributed to the leachates from Cd-plated items and lead smelting and lead products manufacturing at the sampling sites [6, 7, 22]. However, the metal ions are being added to the water stream at a much higher concentration than the prescribed limits by industrial activities, thus leading to cause health effects and environmental degradation (Table 2).

Table 3: Total variance explained and component matrices for the heavy metals in the industrial wastewater collected from Dhaka City, Bangladesh.

Component	Initial Eigen values			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	2.5	41.8	41.8	2.5	41.8	41.8	2.2	36.4	36.4
2	1.6	27.1	69.0	1.6	27.1	69.0	1.6	27.5	63.9
3	0.91	15.2	84.2	0.91	15.2	84.2	1.2	20.3	84.2
4	0.38	6.4	90.6						
5	0.32	5.4	96.0						
6	0.24	4.0	100						
Elements	Component matrix				Rotated Component Matrix				
	PC1	PC2	PC3		PC1	PC2	PC3		
Component Matrix									
Cr	0.83	-0.26	-0.33		0.93	-0.03	0.056		
Ni	0.56	0.03	0.79		0.16	0.02	0.95		
Cu	0.86	0.01	0.12		0.70	0.16	0.48		
As	0.82	-0.22	-0.29		0.90	-0.006	0.089		
Cd	0.18	0.86	-0.28		0.06	0.91	-0.14		
Pb	0.25	0.88	0.08		-0.03	0.89	0.22		

3.2. Sources of heavy metals

Principal component analysis (PCA) was employed to identify the source of heavy metals in the wastewater of different industries, a principal component analysis (PCA) was performed, which has been considered to be an effective tool for source identification [27,28]. Three principal components were obtained (Table 3 and Figure 2), and those accounted for 84.2% of all the total variation. In PCA analysis, first three principal components were computed and the variance explained by them was 41.8%, 27.1% and 15.2% (Table 3). Overall, PCA revealed three major groups of the studied metals for the samples where one group comprised of Ni for wastewater, which was predominantly contributed by lithogenic effects [29]. Second group showed Cd and Pb, which were contributed by the industrial emissions and third group revealed the loadings of Cr, Cu and As for samples indicated that these were mostly contributed by the anthropogenic activities.

3.3. Heavy metals removal by rice husk and egg shell

Rice husk consists of cellulose (32.2%), hemicelluloses (21.3%), lignin (21.4%) and mineral ash (15.05%) [30] as well as high percentage of silica in its mineral ash, which is approximately 96.34% [31]. Rice husk and egg shell are insoluble in water, has good chemical stability, has high mechanical strength and possesses a granular structure, making it a good adsorbent material for treating heavy metals from wastewater.

The concentrations of heavy metals in the wastewater decreased after the treatment by rice husk and egg shell. The effect of the amount of adsorbent on the removal of heavy metals by rice husk and egg shell is depicted in Table 4 for varied adsorbent doses of 20, 30, 40 and 50 mg/L. The results revealed that the removal efficiency of rice husk and egg shell differed widely depending on type and concentration of metals for different industries. The removal of Cr using rice husk was ranged 35–80%, 29–84% and 22–74% for tannery, dye-chemical and textile industries, respectively and for egg shell the removal of Cr was ranged 41–82%, 37–86% and 16–77% for tannery, dye-chemical and textile industries, respectively (Table 4).

The removal of Ni using rice husk was ranged 39–83%, 53–93% and 38–86% for tannery, dye-chemical and textile industries, respectively and for egg shell the removal of Ni was ranged 42–83%, 60–91% and 46–87% for tannery, dye-chemical and textile industries, respectively (Table 4).

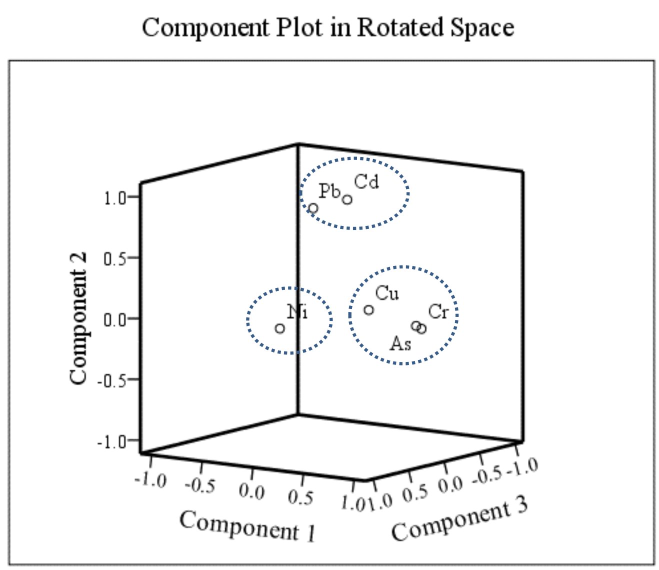

Figure 2: Principal component analysis (PCA) of heavy metals in wastewater collected from different industries in Dhaka City, Bangladesh

Table 4: Removal efficiency of heavy metals for different adsorbent doses

Metals	Adsorbent Dose (mg/L)	Tannery					Dye-chemical					Textiles			
		After treatment %removal					After treatment %removal					After treatment %removal			
		RH	ES	RH	ES		RH	ES	RH	ES		RH	ES	RH	ES
Cr	20	578	523	35	41		3.5	3.1	29	37		2.0	2.1	22	16
	30	349	328	61	63		2.5	2.3	49	53		1.2	1.1	52	56
	40	297	265	67	70		2.1	2	57	59		0.88	0.78	65	69
	50	179	162	80	82		0.79	0.69	84	86		0.65	0.57	74	77
Ni	20	2.2	2.1	39	42		1.4	1.2	53	60		1.5	1.3	38	46
	30	2.1	1.8	42	50		0.68	0.66	77	78		1.2	1.2	50	50
	40	0.94	0.91	74	75		0.33	0.43	89	86		0.46	0.43	81	82
	50	0.62	0.61	83	83		0.21	0.27	93	91		0.33	0.31	86	87
Cu	20	3.3	3.2	27	29		4.2	4.1	35	37		2.5	2.4	34	37
	30	2.6	2.3	42	49		3.4	3.1	48	52		1.8	1.9	53	50
	40	1.9	1.8	58	60		2.5	2.2	62	66		1.3	1.2	66	68
	50	0.54	0.51	88	89		1.3	1.1	80	83		1.1	0.68	71	82
As	20	4.3	3.8	22	31		3.1	3.3	34	30		2.6	2.4	24	29
	30	3.3	3.1	40	44		2.4	2.3	49	51		2.1	2.1	38	38
	40	2.2	2.1	60	62		1.3	1.1	72	77		1.5	1.3	56	62
	50	1.1	0.69	80	87		0.84	0.74	82	84		0.82	0.62	76	82
Cd	20	1.6	1.4	30	39		2.6	2.5	35	38		0.88	0.84	32	35
	30	1.1	1.1	52	52		2.2	2.1	45	48		0.71	0.62	45	52
	40	0.64	0.53	72	77		1.1	0.91	73	77		0.45	0.35	65	73
	50	0.33	0.31	86	87		0.85	0.55	79	86		0.33	0.22	75	83
Pb	20	1.2	1.4	40	30		1.3	1.7	54	39		0.99	1.1	45	39
	30	0.89	0.78	56	61		1.1	1.1	61	61		0.81	0.77	55	57
	40	0.44	0.33	78	84		0.63	0.55	78	80		0.48	0.41	73	77
	50	0.22	0.21	89	90		0.36	0.26	87	91		0.35	0.26	81	86

Among the industries, the ranges of percent reduction of Cu, As, Cd and Pb were 27–88%, 22–82%, 30–86% and 40–89%, respectively after applying rice husk treatment and 29–89%, 29–87%, 35–87% and 30–91%, respectively after applying egg shell treatment (Table 4). The egg shell showed higher sorption affinity than rice husk for the studied metals (Table 4). The relatively high ability of egg shell metal sorption may be due to its high content of $CaCO_3$ (96%) as reported also by Ahmad et al. [32]. The sorption mechanism could be the ion exchange between metal ions in the aqueous solutions and Ca^{2+} on the surfaces of calcite ($CaCO_3$) in the egg shell. The precipitation mechanism may become important for the removal of heavy metals by egg shell, which maintained their strong affinity with increasing surface loading. It was found that the percentage removal of heavy metals was dependent on the dose of rice husk, egg shell and adsorbent concentration. As a case study rice husk and egg shell was used to see the removal efficiency of heavy metals from different industrial wastewater. However, dynamics involved i.e. effect of dilution, pH, and temperature and contact time on the rice husk removal capacity which was not considered for this study. Therefore, our future study will be addressed this environment issue and also for providing the economically feasible solution to remove contaminants from effluents for safe disposal.

4. Conclusion

Our aim was to assess the efficiency of rice husk and egg shell for the removal of toxic metals from contaminated wastewaters of three types of industries in Dhaka City, Bangladesh. The concentrations of heavy metals in wastewater varied widely among the industries and exceeded the permissible limits for industrial effluents, indicating high risk to the surrounding ecosystems. The results showed that egg shell was effective for the removal of heavy metals from industrial wastewater. In this scenario it is essential to individuate a simple and cost effective remediation technique that allows the removal of heavy metals from contaminated waters. The removal of metals by rice husk and egg shell proposed in this study demonstrated to meet those requirements. This highlights their potential as promising sorbents for the effective removal of toxic metals from the environment and offered the potential for low cost effective media for post treatment of metal contaminated wastewaters. However, for future studies, elucidating the possibility and efficiency of some approaches to treat safely the resultant sludge i.e., metal desorption, bioleaching, and stabilization will offer a range of scientific opportunities for a comprehensive understanding of the remediation processes.

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