A CASE STUDY ON PREDICTING THE ENVIRONMENTAL IMPACTS OF UNTREATED EFFLUENT GENERATED FROM TANNERY INDUSTRIAL ESTATE IN DHAKA, BANGLADESH

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ABSTRACT. Significant amounts of heavy metals in the directly discharged wastewater released from the newly built tannery industrial site in Dhaka, Bangladesh are reported. Despite their detrimental impacts on public health and natural ecosystem, no environmental impact study is yet conducted. Therefore, information on safe discharge rates are not available. In this study, the extent of pollution around the industrial site is investigated for four representative trace metals. Temporal and spatial distributions of chromium (Cr), lead (Pb), cadmium (Cd) and arsenic (As) have been predicted using a numerical model with the aim to estimate safe discharge of these metal contaminants. From multiple simulation runs it has been estimated that a discharge of 0.026 m³ of wastewater per day can lead to high levels of Cr and Pb accumulation, exceeding the regulatory standard limits, in the study area. Whilst As and Cd concentrations remain below the advised limits in most cases at this rate. However, an order of magnitude reduction in the total discharge rate, i.e., 0.0026 m³ per day, results into the metal accumulation below the recommended guidelines in all cases. Elevated concentration of Pb is found to be limited to the top 0.5 m of the soil as compared to Cr, As and Cd, which exhibit larger spread along the depth of the soil. The relative dominance of the metal contamination follows the sequence: Pb>Cr>As>Cd as sorbed concentration in soil aggregates and Cr>Pb>As>Cd as aqueous concentration in soil porewater. Further investigations that are essential for a comprehensive environmental impact assessment have been highlighted.

KEY WORDS: tannery effluent, environmental impact, heavy metal, public health, contaminant transport, modelling

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INTRODUCTION

Discharged wastewater from leather processing industries is ranked as one of the highest pollutants among all the industrial wastes (Shen 1999). The industry uses large amount of salts and liquid chemicals to convert putrescible raw skins into stable, non-putrescible leather; a process commonly known as tanning. Tanning is a wet process which consumes large amounts of water and the bulk of which is discharged as wastewater or tannery effluent (Chowdhury et al. 2013). Direct discharge of the effluent into the environment severely damages the aquatic life of receiving water bodies, contaminates land surfaces (Cooman et al. 2003); thereby, imposes significant risks to public health and natural ecosystems.

Generally, wastewater contains large amount of heavy metals such as chromium, zinc, cadmium, lead, nickel and arsenic. These metals, at higher concentration, are toxic and cause detrimental effects on human health (Trichopoulos 2001; Kocasoy and Sahin 2007). Chromium is a carcinogen and it also damages the gastrointestinal, respiratory and immunological systems (USEPA 1989; IARC 1990; Islam et al. 2013). Cadmium, even at very small concentrations, is highly poisonous and chronic exposure to this metal can lead to anaemia, insomnia, cardiovascular diseases as well as hypertension (Sharma et al. 2006). Excessive lead exposure increases blood pressure, damages kidney, causes miscarriages and subtle abortions, hinders brain development, and leads to behavioural disruptions (Islam et al. 2013). Arsenic causes vascular diseases, hyperpigmentation, lung and skin cancer, hypertension and genotoxicity (Islam et al. 2018; Das et al. 2004). Therefore, it is essential to treat tannery effluents before discharging into the environment.

More than half of the world's leather tanning activities takes place in low and middle income countries (Blacksmith Institute 2010). These countries often lack in waste treatment facilities, appropriate environmental regulations and adequate enforcement; which consequently leads to widespread environmental pollution. The leather industry contributed more than USD 1.1 billion to the Bangladeshi economy in 2015–16 financial year (EPB 2017). The sector is experiencing rapid growth due to increasing demand in both local and international markets. More than half a million skilled and semi-skilled workers (Harris 2016) are directly employed by the industry, which is, therefore, vital for economic prosperity and employment opportunities. However, the industry poses significant threat to the local environment and the public health.

Objectives and scope

Despite the early signs of pollution, no environmental impact study of the current site has been conducted or published yet. Relevant site information, such as waste inventory and discharge rates are not available. To highlight the severity and urgency of the problem, in this study, the authors investigate the extent of contamination associated with untreated effluent discharge around the site. Temporal and spatial distributions of four heavy metals, e.g., Cr, Pb, Cd and As are predicted following a numerical model approach. These metals are chosen due to their dominant presence in the discharged tannery wastewater in Bangladesh as well as for their high toxicity to public health (Rahaman et al. 2016). In this paper, the authors aim to provide useful insights on the level of metal contamination associated with waste discharge rates and to emphasise on further investigations and information which are essential and collected immediately for a comprehensive environmental impact study and to develop effective prevention strategies.

The study area and background of tannery waste pollution in Bangladesh

For decades wastes from tanneries have polluted rivers and agricultural lands in Bangladesh. Extreme pollution of the Buriganga river in the capital, Dhaka, from the Hazaribagh tannery effluents is a widely known environmental disaster (Bernhardt and Gysi 2013). A study in 2008 funded by the European Union found the top three metres of soil at Hazaribagh were severely contaminated (Human Rights Watch 2012). The soil around Hazaribagh was contaminated with chromium (up to 37,000 mg/kg), sulphur, mineral oil, phenols and extractable organohalogen compounds (up to 1,200 mg/ kg) through percolation of the wastewater. Rahaman et al. (2016) measured concentrations of heavy metals at various depths of the riverbank soil adjacent to the Hazaribagh tannery area and compared them against the samples collected from an uncontaminated agricultural land. Their study reported significantly higher concentration of Cr, Zn, Pb, Cd (561.71, 158.23, 70.58, 2.25 mg/kg, respectively) in the riverbank soil than that of the agricultural land (27.86, 134.16, 25.76, 1.30 mg/kg, respectively). Regulatory standards for some of the heavy metals in agricultural soils are presented in Table 1 (He et al. 2015). The standards for metal concentration in Bangladeshi soil are not available. The regulatory standards of heavy metals in inland surface

and irrigation water in Bangladesh are presented in Table 2 (ECR 1997). Poor infrastructure, lack of pre-discharge waste treatment facilities, failure of administration and poor management were found responsible on that regard. In response to the mounting criticisms and pressures from the environmental activists, members of public; the High Court of Bangladesh directed the Government to relocate the industry to the Savar Tannery Industrial Estate (Khan 2017). The site is located on the banks of the Dhaleshwari river at Jhauchar village in Tetuljhara union of Savar [23°46′29.4″N, 90°14′25.9″ E] (Fig. 1a,b).

It was expected that the new site with its modern waste treatment facilities, advanced effluent discharge plants and planned development will safeguard local environment and public health as well as vitalise the industry to sustain growing demands and to compete in global markets. However, severe pollution from Savar tanneries has already been reported and it is associated with poor planning, faulty and incomplete installation of the waste treatment facilities (Khan 2017). Under the Bangladesh Environment Conservation Rules (1997), it is mandatory to install Effluent Treatment Plant (ETP) to treat tannery waste before discharging into the environment. A total of 111 tanneries have already started their production at Savar Tannery Industrial Estate, before the Central Effluent Treatment Plant (CETP) has become fully operational (RMGB 2018). Out of the four CETP modules, two are operational while the remaining two are inactive. The active CETP modules, which have the capacity to treat approximately 12,500 m³ of wastewater per day, are operating only partially (Daily Sun 2018). The tanneries are currently discharging approximately 20,000 m³ of untreated effluents per day into the Dhaleshwari river, nearby agricultural lands and wetlands (RMGB 2018). The severity of such pollution is mentioned above and there is a growing concern of similar or higher magnitude of pollution than Hazaribagh tanneries in the surrounding area. Therefore, it is vital to investigate the potential impacts of tannery effluents in the surrounding environment to avoid disastrous consequences of the Hazaribagh tanneries. The study area which has been considered in this paper is presented in Fig. 1c. The area is composed of alluvium soil of the Pleistocene period and the soil type is mainly silty-loam (Sultana et al. 2017). The area experiences a hot and humid tropical climate with annual average temperature of 30°C. The soil is dry to moist most of the year except during the monsoon season when it is saturated or oversaturated.

Table 1. Regulatory standards of heav	y metals in agricultural soils (He et al. 2015)
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Heavy metals	USA	UK	Australia	China
	Agricultural soil (mg/kg)			
Cr	11	-	50	150-300
As	0.11	43	20	20-40
Cd	0.48	1.8	3	0.3-0.6
Pb	200	_	300	80

Table 2. Regulatory standards of heavy metals in inland surface and irrigation water in Bangladesh (ECR 1997)

Heavy metals	Surface water (mg/l)	Irrigation water (mg/l)
Cr	0.1	1
As	0.2	0.2
Cd	0.5	0.05
Pb	0.1	0.1



Fig. 1. a) Geographic location of the Tannery Industrial Estate in Dhaka, Bangladesh; b) Tannery Industrial Estate enclosed in the oval; c) The study area considered in this article (Google Maps 2019)

METHODOLOGY

In this study, an advanced numerical model namely COMPASS (Code of Modelling Partially Saturated Soils) has been used to predict the spread of metal contamination in the selected site. The model investigates heat, liquid, moisture, gas and chemical flows, microbial, geochemical and biogeochemical reaction processes, and mechanical deformation under a coupled framework. The details of the model including theoretical and numerical formulation, verifications, validations and numerous applications are provided elsewhere (Thomas and He 1998, Masum et al. 2012, Masum 2012, Masum and Thomas 2018a,b). In the following sections, the theoretical basis of the model, pertinent to this study, is presented.

Assumptions

Following assumptions have been made in this work: i) contaminants flow via diffusion in the liquid phase and the advective flow is negligible; ii) transport of metal contaminants is hindered by sorption at soil particles and the process can be explained adequately by distribution coefficient or constant K_D values; iii) Metals can exist in various oxidation states i.e., Cr(III)/Cr(VI), As(III)/As(V) etc., however their total concentration is considered in this study; iv) isothermal condition prevails and the effects of plant roots and vegetation on transport processes are negligible.

Theoretical formulations

Total concentration of a solute in a unit soil volume can be expressed as,

$$C_t = \rho_b C_s + \theta_l C_l + \theta_g C_g \tag{1}$$

where C_i and C_g are concentrations of the solute in liquid phase and gas phase, respectively. ρ_b is the soil bulk density. θ_i and θ_g represent volumetric liquid content and gas content, respectively. In this work, no gas phase contaminants are considered and the 4th term of Eqn. (1) has been omitted. Therefore Eqn.(1) yields,

$$C_t = \rho_b C_s + \theta_l C_l \tag{2}$$

Sorption of solutes in soil particles i.e., the solid: solution partitioning is expressed as,

$$C_s = K_D C_l \tag{3}$$

where K_{p} is the partitioning/distribution coefficient.

The governing transport equation of a solute is given by,

$$\frac{\partial C_t}{\partial t} = \nabla J \pm S_s \tag{4}$$

here J is the contaminant flux and S_c is the sink/source term.

Since only diffusive flux of contaminants in the liquid phase has been considered, Eqn.(4) can be expressed by employing Eqn.(3) and ignoring the external sink/source as,

$$\frac{\partial(\rho_b K_D + \theta_l)C_l}{\partial t} = \nabla(\theta_l D_l \nabla C_l)$$
(5)

where D_{i} is the effective diffusion coefficient that includes path tortuosity of porous medium and is expressed by,

$$D_l = \tau_l D_0 \tag{6}$$

here τ_i is the tortuosity factor and D_o is the diffusion coefficient in free fluid. The liquid phase tortuosity factor is calculated following Millington and Quirk (1961) model as,

$$\tau_l = \frac{\theta_l^{10/3}}{n^2} \tag{7}$$

where *n* is the porosity. Eqn (5) can be rearranged

Eqn.(5) can be rearranged as,

$$R_{D}\frac{\partial C_{l}}{\partial t} = \nabla (D_{l}\nabla C_{l})$$
(8)

where $R_{\rm p}$ is the Retardation factor and it is defined as,

$$R_D = \frac{\theta_l + \rho_b K_D}{\theta_l} = 1 + \frac{\rho_b K_D}{\theta_l} \tag{9}$$

For a multicomponent system, Eqn.(8) can be written as,

$$R_D^i = \frac{\partial C_l^i}{\partial t} = \nabla (D_l^i \nabla C_l^i); i = \{1, 2, \dots, N_d\}$$
(10)

where N_d is the total number of solutes i.e., dissolved contaminant considered in this case.

Model application

The source of effluent discharge has been considered as a point-source (Fig. 2a) by approximating the location of the CETP. To proceed to numerical modelling, the domain has been discretised into 802 non-symmetric four-nodded quadrilateral elements (Fig. 2b). The model simulations have been conducted for a single moisture content, since the variation of soil water is low throughout the year (except during monsoon) and the relevant rainfall data for the area is not available.

The distance of points A, B and C are 76.2, 41.2 and 85.4 m from the source, respectively. The boundary points, B1 and B2 on x'-x' line are located 124 and 724 m away from the source, respectively.

Initial and boundary conditions

Initial and boundary conditions required for the simulations are presented in Table 3. Rahaman et al. (2016) measured the concentrations of Cr, Pb, Cd and As on the surface of an agricultural land that was not polluted by any tannery wastes, and they are considered as initial concentrations in these simulations.

The contaminant discharge rates at the discharge boundary, S, is calculated from the sludge characterisation data of Chowdhury et al. (2015) who collected heavy metal concentrations from the tannery effluents dumping area at Hazaribagh industrial zone. From their data, proportions of the contaminants in the sludge were estimated, which was then used to calculate the individual mass fluxes; based on the assumption that the ratio of the contaminants in the sludge is same as in the discharged effluent at the source. As mentioned earlier that no discharge rate information of current site is available, therefore, in this study, two effluent discharge rates, e.g., 1.0x10⁻⁷ kg/m²/s and 1.0x10⁻⁷ ⁸ kg/m²/s are considered. These values are obtained from several trial simulation-runs with the aim to identify the optimum discharge rate that results into the contaminant concentration level, in the study area, within a close



Fig. 2. a) Sketch of the study area with approximate location of the CETP or the source of effluent discharge, S. Γ_B is the boundary of the study area. A, B, C represent positions near the source and B1, B2 are locations at the boundary along the x'-x' segment;
b) Mesh geometry

proximity of the regulatory standards suggested by various countries across the world (presented in Table 1). The two discharge rates information provide an opportunity to predict the impacts for an order of magnitude variation in the wastewater discharge rates. Based on these discharge rates, two sets of contaminant flux values (Set 1 and Set 2), at the source boundary, are calculated and presented in Table 3.

RESULTS AND DISCUSSION

The parameters for the simulations are presented in Table 4.

Fig. 3 presents spread of the contaminants in the soil porewater from the discharge source during Set 1 and Set 2 simulations. Cr, which is the major contaminant in the effluent, shows significant increase in Set 1 simulation to 715 mg/l (Fig. 3a1 and Fig. 4b) within the vicinity of the source. In Set 2 simulation, the predicted concentration is (as expected) 71.5 mg/l (Fig. 3a2 and Fig. 5b). The results in Fig. 4a and Fig. 5a show the corresponding concentration of Cr sorbed into soil aggregates along the x'-x' line. Highest concentrations of 98.79 mg/kg and 9.88 mg/ kg are observed adjacent to the effluent source in Set 1 and Set 2 simulations, respectively. The difference in magnitude is due to the fact that the Set 2 discharge rate is ten times smaller than that of the Set 1. Fig. 3b shows the distribution of Pb in the study area during the simulation period. Slower diffusion and higher retardation hinder the spread of Pb in soil. In comparison to the other metals, e.g., Cr, Cd and As, distribution of Pb is more localised. Fig. 4b and 5b show that the augmentation zone is restricted to approximately 350 m radius of the source. However, the peak-sorbed concentration of Pb in soil is significantly larger than the other metal contaminants due to the large solid-liquid partitioning coefficient (Table 4). Maximum sorbed concentrations of 2441.72 mg/kg and 244.3 mg/kg are observed, in Fig. 4a and 5a, during the Set 1 and Set 2 simulations, respectively.

Distributions of Cd and As in the study area are presented in Fig. 3c and 3d respectively. The lowest retardation of As allows it to spread in the entire study area during the simulation period. The maximum concentration of sorbed Cd in Set 1 simulation is 0.19 mg/kg (Fig. 4a) and the aqueous concentration is 2.62 mg/l (Fig. 4b). Similarly, the maximum estimated concentration of As in solid phase is 0.18 mg/kg (Fig. 4a) and in the aqueous phase is 5.44 mg/l (Fig. 4b).

Evolutions of contaminants at three locations within the site, i.e., points A, B, C (Fig. 2a) are presented in Fig. 6 and at the site boundary, B1 and B2, are presented in Fig. 7. It is obvious from the results that the Point B, which is the closest of the selected points from the source, accumulates the highest amount of these metal contaminants. In this location, Cr (>60 mg/kg) and Pb (>700 mg/kg) concentrations exceed the maximum allowable limits set by a number of countries, listed in Table 1 (except Cr according to the Chinese standard). Away from the source, level of metal accumulation reduces. Boundary B1, which is relatively closer to the source than B2, shows Cr concentration of 57 mg/kg, while the B2 shows a concentration of 5.4 mg/kg for the same discharge rate. The metal concentrations along 1.75 m depth of the soil at the source are presented in Fig. 8. The results show that accumulation of high amount of Pb is limited to the top 0.5 m of the soil. In comparison, Cr, As and Cd exhibit larger spread along the depth of the soil, which is mainly due to their relatively lower retardation in the soil. Larger downward spread of heavy metals may potentially contaminate the underground water resources and, therefore, effective containment strategy should be adopted in the current site to avoid such pollution risk.

Fig. 9 presents a comparison between regulatory standard values of heavy metals in agricultural soils, recommended in Australia, China, UK and USA, and the model predicted maximum metal concentrations at the point source of the current site. The results suggest that at the rate of 1.0×10^{-7} kg/m²/s, which is equivalent to 0.026 m³ of daily discharge, concentration of Pb and Cr will exceed the recommended limit set by most of the listed countries.

Contaminants	Initial concentration (mg/kg)†	Boundary conditions		
		Discharge flux (kg/m²/s) at source, S		Domain boundary,
		Set 1	Set 2	Гв
Cr	27.867	5.9x10 ⁻⁸	5.9x10 ⁻⁹	∂Cr/∂x=0,x€Γ _β
Pb	23.60	2.8x10 ⁻⁹	2.8x10 ⁻¹⁰	∂Pb/∂x=0,x€Г _в
Cd	0.64	5.3x10 ⁻¹⁰	5.3x10 ⁻¹¹	∂Cd/∂x=0,x€Г _в
As	10.0	2.1x10 ⁻⁹	2.1x10 ⁻¹⁰	∂As/∂x=0,x€Г _в

 Table 3. Initial and boundary conditions for Set 1 and Set 2 simulation

^{\dagger} These are sorbed concentrations of the contaminants, which are converted into liquid phase dissolved concentrations using their respective K_{D} values for the simulations.

Table 4. Parameters for Set 1 and Set 2 simulation

Contaminants	$D_o(m^2/s)$	K _D (l/kg)
Cr	1.27x10 ⁻⁹ (Li et al. 2007)	140 (SKB 2009)
Pb	9.45x10 ⁻¹⁰ (Sato et al. 1996)	6,000 (SKB 2009)
Cd	7.2x10 ⁻¹⁰ (Buffle et al. 2007)	73 (SKB 2009)
As	1.15x10 ⁻⁹ (Tanaka et al. 2013)	34 (SKB 2009)
Soil properties		
Bulk density, $\rho_b = 1.33$ g/cm ³ (USDA 2019) Porosity, $n = 0.50$		



Fig. 3. Spread of contaminants in soil porewater from the effluent discharge point S at the end of simulations a1) Cr, b1) Pb, c1) Cd and d1) As for Set 1 simulation. a2) Cr, b2) Pb, c2) Cd, d2) As for Set 2 simulation. Colour scale in mg/l and Runtime 1 year



Fig. 4. Concentration profiles of the contaminants along x'-x' line (Fig. 2a) after 1-year simulation period a) Sorbed concentrations in soil aggregates and b) aqueous concentrations in soil porewater for Set 1 simulation. Dotted vertical lines represent the position of the source



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Fig. 5. Concentration profiles of the contaminants along x'-x' line (Fig. 2a) after 1-year simulation period a) Sorbed concentrations in soil aggregates and b) aqueous concentrations in soil porewater for Set 2 simulation. Dotted vertical lines represent the position of the source



Fig. 6. Evolution of contaminant concentrations at point A, B and C. Sorbed concentrations of a) Cr, Pb and b) Cd, As in Set 1 simulation



Fig. 7. Evolution of metal concentrations at the site boundary locations a) B1 and b) B2 along x'-x' line for Set 1 simulation



Fig. 8. Sorbed concentrations of the metals in the soil up to 1.75 m depth from the surface at the source



Fig. 9. Comparison between models predicted maximum concentrations and regulatory standard concentrations of heavy metals (He et al. 2015) at agricultural soils in various countries. Please be noted that the guideline values of Cr and Pb for the UK is not available

Concentration of Cd remains below the regulatory values for this discharge rate. Also, As concentration is significantly less than the recommended values of the most countries, but slightly higher than the USA standard. On the other hand, in all cases, effluent discharged at the rate of 1.0x10⁻⁸ kg/m²/s or 0.0026 m³ per day results into accumulation of the metals below the recommended regulatory standard values. This value, therefore, can be considered as a safe discharge rate with minimum environmental impacts for the studied area of the site.

It is important to mention that the discharge rates considered in this work may have been an underestimation. However, the rates presented in this paper provide guidance/suggestions on the safe discharge rates, below which the pollution associated with metal contamination in the site remains within regulatory standard limits. Adequate measures should be taken to ensure that any leakage or overflow of untreated effluent from the CETP is less than the suggested discharge rates.

For a comprehensive and accurate environmental impact assessment the following actions and further investigations are essential. Estimation of the waste inventory, wastewater discharge rates and effluent characterisation should be conducted to obtain accurate proportion of the heavy metals as well as their influx into the surrounding environment. Detailed site investigations should be carried out to establish soil morphological and geochemical baseline information. Laboratory investigations should be undertaken to measure physical, chemical, mechanical properties, sorption parameters, equilibrium and kinetic reaction rate constants, geochemical data etc. The country experiences heavy rainfall during monsoon season. The seasonal precipitation data of the current site is not available and it should be monitored and collected as well. Inspection boreholes and monitoring equipments in and around the site may provide indication of potential soil and groundwater pollution. Some of these activities are ongoing and will be presented in future publications.

Heavy metals, such as Cr, Pb, As and Cd which are carcinogenic and highly toxic to human and animals should be dealt with appropriately and adequately. Since the surrounding area is mainly agricultural lands, plants and agricultural products may act as potential pathways for direct and indirect ingestion of these toxic contaminants. Adverse impacts on the local ecological system are also expected. Therefore, a combined and comprehensive approach is essential including local authorities, utility companies, health officials, public representatives and member of public. Otherwise, a cleaner and toxicity free atmosphere in and around the tannery estate will be a serious challenge for the development of this sector without placing further burdens on the environment.

CONCLUSION

In this paper, soil pollution associated with discharged wastewater from a new tannery industrial site in Bangladesh was investigated. A study area was selected within the site to predict the extent of soil contamination by four heavy metals, e.g., Cr, Pb, Cd and As, following a numerical modelling approach. Two sets of effluent discharge rates were considered in the model simulations. The results indicated

that a daily discharge of 0.026 m³ of wastewater could exceed the safe concentration level of Pb (in all cases) and Cr (in most cases), recommended by various standards. At this rate, soil contamination by Cd and As (except in one case) were predicted to be under the regulatory values considered in this study. However, a discharge rate of 0.0026 m³ of wastewater per day maintained a safe concentration level in the soil for all cases. To date, very little information of the current site is available and that is a major challenge for an accurate estimation of environmental impacts. Therefore, to conduct a comprehensive environmental impact assessment, the further studies which are essential and should be conducted immediately are also highlighted.

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