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Assessment of Environmental Hazard and Heavy Metal Contamination in Dhaleshwari River Sediment: A Toxicity based Study on Pollution

Md. Al Sadikul Islam[1] | Mohammad Enayet Hossain[2] | Kamrun Nahar[3] | Nehreen Majed[1]*

1. Department of Civil Engineering, University of Asia Pacific, Dhaka 1205, Bangladesh
2. Department of Soil, Water and Environment, Dhaka University, Dhaka 1000, Bangladesh
3. Department of Environmental Science and Management, North South University, Dhaka 1229, Bangladesh

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ABSTRACT

Developing nations, such as Bangladesh, face an enormous crisis in maintaining natural sustainability due to heavy metal contamination by the peripheral rivers. Frequent heavy metals discharged from tanneries, dyeing, and potential anthropogenic activities in Savar city pollute the Dhaleshwari river, which is an important river of the capital city, Dhaka. The present study aimed to assess the heavy metals contamination in the Dhaleshwari river sediment and evaluate the subsequent ecological risk indices emerging from the deposits. The contamination levels of heavy metals such as lead (Pb), cadmium (Cd), chromium (Cr), and nickel (Ni) were analyzed in the Dhaleshwari River sediment. Various environmental indices, such as Potential Enrichment Risk (PER), Geo-accumulation Index (Igeo), Enrichment factor (EF), Toxic unit analysis (TUs), etc., were observed in various compartments. The concentration of heavy metals ranged as follows: Lead (Pb), 297.3-414.6 mg/L; Cadmium (Cd), 1.5-4.4 mg/L; chromium (Cr), 97.9 -282.4 mg/L; Nickle (Ni), 85.1-264.5 mg/L; Iron (Fe), 11800-14375 mg/L. The metal concentrations were higher than the threshold effect level (TEL) and probable effect level (PEL) standards. Based on the TUs, the probability of toxicity is about 76% (TU > 2.3) at the Dhaleshwari river. Comparative evaluation of different environmental indices between present and past studies indicated progressive deterioration of sediments by heavy metals. Linear correlations of heavy metals in sediment samples demonstrated toxic accumulation of heavy metals in the surrounding ecosystem. The study outcomes emphasize the necessity of systematic investigation in the Dhaleshwari river and warranting effective prioritization to ensure river health over industrial wastewater discharge.

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INTRODUCTION

Bangladesh is a land of rivers because it is crisscrossed by hundreds of rivers, tributaries, and distributaries. The capital of Bangladesh, Dhaka, has a network of rivers - Turag to the west, Buriganga to the south, Balu and Shitalakhya to the east, Tongy Khal to the north, and Dhaleshwari to the south (Faisal et al., 1999). The Dhaleshwari river flows past Dhamrai, Savar, and is considered one of the vital rivers of Bangladesh. During the last decade, fast urbanization

*Corresponding Author Email: *nehreen-ce@uap-bd.edu*

and industrialization have given rise to heavy metal contamination globally, especially in developing nations such as Bangladesh (Bhuyan et al., 2017). In developed countries, heavy metal pollution is not severe because the regulatory bodies ensure that the industries comply with the country's laws. On the other hand, Bhuyan et al., (2019) mentioned that approximately 10% of the industries in Bangladesh installed effluent treatment plants, and the rest of the other industries are still releasing untreated effluents into the nearby water bodies (Mohanta et al., 2019). In the past decade, the Buriganga river was reported to be one of the most extensively contaminated waterways in the country. Uncontrolled discharge of untreated industrial effluents from different industries such as tanneries, pharmaceuticals, etc., and other anthropogenic pollution was responsible for the abysmal condition of the Buriganga river (Alam, 2003). The Hazaribagh tannery was founded seven decades ago with around 185 tanneries in that region (Juel et al., 2020) beside Buriganga River. On average from a recent estimate, about 22,000 m^3 of hazardous effluents were released every day in lowlands, lagoons, and waterways that eventually joined the Buriganga river (Mizan et al., 2016). The Government of Bangladesh started a program called "Hazaribagh Tannery Relocation Project" to relocate the Hazaribagh Tannery to Savar in 2017, considering the leather industry's contribution to national GDP and the simultaneous detrimental environmental impacts of the tannery waste (Mohanta et al., 2019). In this project, the tanneries from Hazaribagh were relocated to Savar on 200 acres with its Central Effluent Treatment Plant (CETP) next to the Dhaleshwari River. The newly relocated tanneries generate 25,000 m^3 of effluents on a daily basis (Hasan, 2020). Due to the inadequacies of the CETP functioning, the tanneries now discharge untreated or partially treated effluents to the river Dhaleshwari. With an increased number of industries being developed, hundreds of newly relocated tanneries are dumping a significant volume of hazardous wastes contributing heavy metals into the river consistently. The area surrounding the river is utilized for farming, and the river water is frequently utilized to irrigate the area. Anthropogenic activities in the vicinity also degrade the quality of the sediment of the Dhaleshwari River.

Leather manufacturing involves many chemical products such as chromium sulfate, tannins, fungicides, ammonia salt, etc. (Juel et al., 2020). The heavy metals may find their way into ecosystems and contribute non-degradable contaminants to nature. As a result, these heavy metals continue to exist in the ecological system and pose a risk to humans and other animals (Ayangbenro and Babalola, 2017). Whereas metals are also introduced into agricultural lands through fertilizers and pesticides (Mohanta et al., 2019). Heavy metals, including cadmium, mercury, lead, copper, and zinc, are recognized as critical marine pollutants because of their toxicity, presence in food chains, and propensity to survive in the environment for an extended period of time (Puyate et al., 2007). Accumulation of these metals in sediments and water in a significant quantity allows these metals to eventually enter the food chain via water and vegetation (Bhuyan et al., 2017). Their distribution and accumulation in the ecosystem are increasing at a worrying pace resulting in their deposition and sedimentation of aquatic organisms in water reservoirs (Mohiuddin et al., 2010).

With due consideration to all the factors mentioned above, the purpose of this research was to evaluate the contamination of sediment from the Dhaleshwari River due to the influence of the newly relocated Tannery Industrial Park. The observed sediment quality was compared with the sediment quality of certain other rivers and different sediment quality standards. Different indices, for example, Geo-accumulation Index (I_{geo}), Toxic Unit Analysis (TUs), Enrichment Factor (EF), Modified Contamination Factor (mCd), and Potential Ecological Risk (PER), etc. were utilized to evaluate the ecological risk from heavy metals in the sediments concerning their quantity, accumulations, and toxicity. This study has a specific focus on obtaining a spatial interpolation of the heavy metal accumulation in the sediments to obtain the spatial distribution of those indices and to acquire an overall notion of the progressive contamination along the selected stretch as compared with previous studies, which will pave the way for different strategies that will be required to be implemented in order to mitigate pollution.

MATERIALS AND METHODS

Study areas are described below: followed by the respective method. The Dhaleshwari river is a watercourse that stretches over 22,155 kilometers throughout the region, and its tributaries (Rahman et al., 2015). It is the primary distributary of the Jamuna River, with a total length of about 290 kilometers, a typical depth of 37.19 meters, and a maximum depth of 80.79 meters. The river takes off from the northwestern tip of the Tangail district (Majumder, 1971). The sampling stations of the sediments along the Dhaleshwari River stretch are illustrated in Figure 1. This study was conducted during the dry season near five selected outfalls along an approximately kilometers' stretch of the Dhaleshwari River covering upstream to downstream. The outfalls were selected based on the catchment characteristics and sources of input.

The sample collection procedure is described as follows followed by the relevant method. As shown in Figure 1, five different sediment samples (Savar Tannery area, Sudkhira, Tetuljhora, Akh Knitting and Dyeing and Fordnagar) have been obtained from the Dhaleshwari River near Saver District, as shown in Figure 1. Sampling was performed in the dry season. Using the grabbing technique, around 0.5 kilograms of sediment was obtained from every sampling station (Shanbehzadeh et al., 2014). Intricated sampling procedure has been followed to collect sediment samples from 0 to 10 cm deep. After collection, sediment samples were placed in bags made of polythene and then stored in an icebox at 40°C for later analysis (Shanbehzadeh et al., 2014).

Sample preparation procedure is described as follows followed by the relevant method. In a tarred silica dish, 10 – 20 g of each sample was weighed. The samples were then dried in a laboratory oven at 120°C. Then the temperature was slowly increased to 450°C at 50°C/h in the muffle furnace (Bhuyan et al., 2017). The samples were ignited at 450°C for eight hours. After cooling, the samples were processed on a hot plate in 50% nitric acid (Bhuyan et al., 2017). Finally, Using Whatman 44 Filter, materials were filtered into a 100 mL flask.

Data analysis procedure is described as follows followed by the relevant method. SPSS v.25 was used to determine Pearson's correlation coefficients. The spatial interpolation approach (Ian,

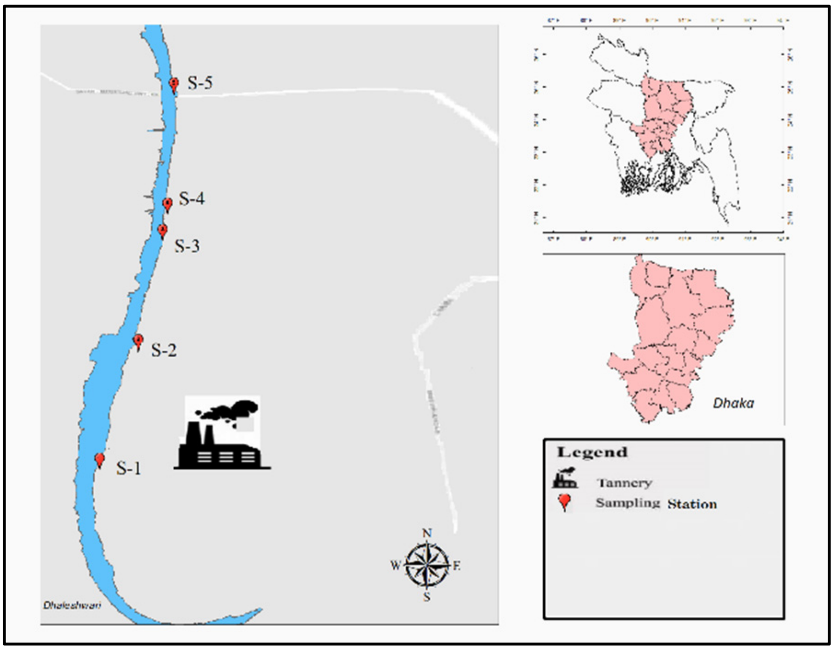

Fig. 1. Sampling Station Map of Dhaleshwari River

2010) employed in the present study demonstrates the geographical distribution of different ecological pollution indices. In Arcmap 10.3, this distribution of different ecological pollution indices has been processed by krigging method (Gao et al., 2015).

Assessments of contamination factor (CF) and modified degree of contamination (mc_d) are described: followed by the relevant method. The contamination factor (CF) has been calculated as described in Reboredo (1993) according to the following equation:

$$\mathrm{CF} = \frac{Cm\,Sample}{Cm\,Background}$$

Here, Cm Background represents the standard of background concentrations, and Cm sample represents the metal concentration in sediment (Reboredo, 1993), which which were analysed as described before. Hakanson's analysis indicated that using the given equation, the sum of the particular pollution factors (Hakanson, 1980) represented the total sediment level (Cd):

$$\mathrm{C_d} = \sum_{i=1}^{N} \mathrm{CF}_{\mathrm{i}}$$

The C_d is used to quantify the degree of contamination at sampling stations. Furthermore, the categorization and the method of calculations are based on the polychlorinated biphenyls (PCB) plus defined heavy metals in Hakanson's study. Considering these constraints, Abrahim, (2005) proposed an amended and simplified version of the Hakanson, (1980) equation for the total amount of pollution measurement. The equation below is a modified equation for determining the degree of contamination:

$${}_{\mathrm{m}}\mathrm{Cd} = \frac{1}{N} \sum_{i=1}^{N} \mathrm{CF}_{\mathrm{i}}$$

Here, the number of Heavy Metals analyzed is N and i = i th Heavy metal, and CF is the contamination factor. This simplified equation to measure mCd has the potential to integrate as many metals as a study might involve in the analysis.

Potential ecological risk (PER) is described: followed by the relevant method. The term "Potential Ecological Risk" or PER is frequently employed to evaluate the level of pollution from heavy metals in sediments. Guo et al., (2010) developed the following formulae for determining PER:

$$C_f^i = \frac{C^j}{C_n^i}, \quad C_d = \sum_{i=1}^{n} C_f^i$$

$$E_r^i = T_r^i * C_{fi}^i, \quad PER = \sum_{i=1}^{n} E_f^i$$

The potential ecological risk index (E^i_r) is utilized for characterizing an element that can potentially pose an ecological risk. The toxic biological factor of E^i_r for every element is ascribed

Table 1. Environmental indices indicating the severity of heavy metal pollution's potential ecological risk (Luo et al., 2007) and Modified Degree of Contamination (Abrahim, 2005)

Modified Degree of contamination (mC_d)	Contamination Degree	Risk Index (PER)	
$mC_d < 1.5$	Nil to very low degree of contamination	RI < 65	Low risk
$1.5 \le mC_d < 2$	Low degree of contamination	65 ≤ RI < 130	Moderate risk
$2 \le mC_d < 4$	Moderate degree of contamination	130 ≤ RI < 260	Considerable risk
$4 \le mC_d < 8$	High degree of contamination	RI ≥ 260	High risk
$8 \le mC_d < 16$	Very high degree of contamination		
$16 \le mC_d < 32$	Extremely high degree of contamination		
$mC_d \ge 32$	Ultra high degree of contamination		

Table 2. Muller's Classification for Geo-Accumulation Index (I_{geo}) and Enrichment factor (EF) categories (Mmolawa et al., 2011)

I_{geo} value	Designation of sediment quality	Enrichment Factor (EF)	Enrichment Factor (EF) Categories
≤ 0	Unpolluted	EF<2	Deficiency to minimal enrichment
0 - 1	Unpolluted to moderately polluted	2 ≤ EF < 5	Moderate enrichment
1 - 2	Moderately polluted	5 ≤ EF < 20	Significant enrichment
2 - 3	Moderately to strongly polluted	20 ≤ EF < 40	Very high enrichment
3 - 4	Strongly polluted	EF ≥ 40	Extremely high enrichment
4 - 5	Strongly to extremely polluted		
>6	Extremely		

as follows: Lead = 5, Cadmium = 30, Chromium = 2, and Nickel = 6 (Guo et al., 2010). Table 1 illustrates the environmental indices indicating the severity levels of heavy metal contamination for potential ecological risk and Modified Degree of Contamination.

Geo-accumulation index (I_{geo}) is described: followed by the relevant method. The geo-accumulation index (I_{geo}) is generally employed for determining the extent of heavy metal pollution in sediment, water, and marine ecosystems (Ozkan and Buyukisik, 2012). By comparing actual metal levels in soils with pre-industrial levels, the index provides for the assessment of pollution. I_{geo} was evaluated using the equation:

$$I_{geo} = \log_2 [C_n/1.5\ B_n]$$

Here, C_n represents the concentration of the heavy metals in the sediment, and B_n represents the background value for the same metals. Due to the possibility of fluctuations in the background information owing to the to lithological fluctuations, a factor of 1.5 is adopted (Ozkan and Buyukisik, 2012). Table 2 depicts Muller's Classification for Geo-Accumulation Index (I_{geo}).

Enrichment factor (EF) is described: followed by the relevant method. The enrichment factor is utilized to evaluate the contamination level and to take into account the dissemination of anthropogenic materials in sample sites via individual heavy metals in the sediment (Helen et al., 2016). Usually, Al or Fe are considered for the normalized concentration of heavy metals (Mucha et al., 2003). The following formulae were adopted for determining EF (Ghrefat et al., 2006):

$$\mathrm{EF} = \frac{\left(\frac{M}{Fe}\right) sample}{\left(\frac{M}{Fe}\right) background}$$

Here, the $(M/Fe)_{sample}$ is a ratio of the heavy metals to Fe concentration in samples, and the $(M/Fe)_{background}$ is a ratio of the concentration of the same metals to Fe for the background. For the purposes of geochemical normalization, iron (Fe) was employed as a component of reference. Table 2 illustrates Enrichment factor (EF) categories.

Toxic unit analysis (TU_S) is described: followed by the relevant method. To quantify the acute toxicity of pollutants in the sediments, the sum of toxic units, which can be expressed as the ratio of metal concentration to the PEL standard (Bai et al., 2011) and living organisms (MacDonald et al., 2000), Long et al., (1998) developed the following formulae for determining toxic unit:

$$\mathrm{TU} = \frac{\mathrm{Cm}}{\mathrm{PEL}}$$

PEL refers to the probable effect level standard for heavy metals in sediment, and the concentration of heavy metals in sediment is represented by Cm.

$$\Sigma TU_S = TU_1 \times TU_2 \times TU_3 \times \ldots\ldots TU_n$$

Here, the total toxic unit for heavy metals in sediments is referred to as ΣTU_S. According to the method, when TU < 0.1, probability of toxicity is 10%, 0.11 < TU < 1.5 is a 25% probability of toxicity, 1.51 <TU < 2.3 is a 50% probability of toxicity, and TU > 2.3 is a 76% probability of toxicity (Caplat et al., 2020).

RESULTS AND DISCUSSION

Assessment of heavy metals concentration in sediments is described: followed by the relevant results. Table 3 illustrates the concentrations of heavy metals in sediments, as well as the standard error of the mean (calculated for a specific heavy metal concerning the measurements of all the sampling stations) of the Dhaleshwari river and other major rivers in Bangladesh.

The concentration of Lead (Pb) in sediments ranged from BDL to 414.6 mg/kg (Table 3). The sampling station with the highest Pb level (414.6 mg/kg) was S-1, whereas Pb at S-3 and S-4 was observed below the detection level. Chromium (Cr) concentrations varied from Below Detection Limit (BDL) to 282.4 mg/kg in sediment samples, with an average of 195.8 mg/kg (Table 3). The maximum concentration of Cr was found at sampling station S-1 (282.4 mg/kg), which is Saver Tannery itself. All the sampling stations (except S-2 and S-3) were contaminated with high concentrations of chromium ranging from 195.8 to 282.4 mg/kg, which exceeded the maximum permissible concentration (MPC) even for different national standards (Such as Finland standard of 100 mg/ kg and Chinese standard of 200 mg/kg) (Juel et al., 2020). The concentration of Nickel in the sediment ranged from 85.1 to 264.5 mg/kg, with a median value of 163.8 mg/kg (Table 3). The highest concentration of Nickel (Ni) was observed at the sampling station S-1 (264.5 mg/kg) (Table 3), in contrast with the lowest concentration observed at the

Table 3. The concentration levels of heavy metals along the sampling stations of Dhaleshwari river and other major rivers in Bangladesh.

Station No.	Concentration of heavy metal (mg/kg) ± Standard Error of the Mean					References
	Pb	Cd	Cr	Ni	Fe	
S-1	414.6 ± 91.13	4.4 ± 0.79	282.4 ± 53.55	264.5 ± 33.24	14375 ± 469	This study
S-2	297.3 ± 91.13	3.2 ± 0.79	BDL	115.4 ± 33.24	14109 ± 469	This study
S-3	BDL	BDL	97.9 ± 53.55	85.1 ± 33.24	11800 ± 469	This study
S-4	BDL	1.5 ± 0.79	195.8 ± 53.55	163.8 ± 33.24	13845 ± 469	This study
S-5	379.9 ± 91.13	3.7 ± 0.79	269.9 ± 53.55	224.3 ± 33.24	14100 ± 469	This study
Range	BDL – 414.6	BDL – 4.4	BDL – 282.4	85.1 – 264.5	11800 - 14375	
Median	297.3	3.2	195.8	163.8	14100	
Buriganga River	731	7.7	97	240		Islam et al., (2018)
Shitalakshya river	20.12		55.4	48.23		Rahman et al., (2020)
Paira River	25	0.7	45	34		Islam et al., (2015)
Korotoa River	58	1.2	109	95		Islam et al., (2015)

DL_{Pb} = 0.2 μg/kg; DL_{Cd} = 0.01 μg/kg; DL_{Cr} = 0.075 μg/kg; DL_{Ni} = 0.5 μg/kg

*BDL= Below Detection Limit & DL = Detection Limit

sampling station S-3 (85.15 mg/kg). The Ni analysis revealed that all sampling sites had exceeded the allowable limit by the Environmental Protection Agency (EPA) for Ni concentrations (16 mg/kg) (US-EPA, 1989). The concentration of Cadmium (Cd) in sediments ranged from BDL to 4.4 mg/kg, with an average of 3.2 mg/kg (Table 3). The sampling station with the highest Cd level (4.4 mg/kg) was S-1, whereas Cd at S-3 was observed below below the detection. In the high Cd-containing region of the Savar District, one of the oldest and most popular wholesale fish market, is located.

The authors also purposefully compared the assessed heavy metal concentration of the present study with different major rivers in Bangladesh to quantify the current ecological status of Dhaleshwari River after the relocation of Savar Tannery. And present study observed a massive deterioration in terms of each of the assessed heavy metals in comparison with the other selected Rivers.

Pearson's correlation matrix among the metal concentrations is described: followed by the relevant method. Pearson's correlation matrix could establish interrelationships among metals, indicating familiar sources of the metals (Bastami et al., 2012). The correlation matrix of metals in the sediment under consideration was prepared accordingly, and the results are presented in Table 4. Positive correlations were observed among the contaminants, Cr and Ni (r=0.879), Cd and Ni (r=0.799), Pb and Ni (r=0.708), Pb and Cu (r=0.951). The mentioned pairs of metals had significant positive correlations ($p < 0.05$), indicating that they originated from roughly similar backgrounds (Varol, 2012). These components originate in river sediments from the discharges of nickel-based industries and are derived from lithogenic sources.

Evaluation of contamination factor (CF) and modified degree of contamination (mCd) are described: followed by the relevant results. The Contamination factor (CF) and Modified degree of contamination (mCd) were assessed for this study. Contamination factors for Pb and Cd were obtained to be the most extreme at each of the sampling stations. However, Pb was observed

Table 4. Pearson's Correlation Matrix between Heavy Metals in Sediment Samples

	Correlations			
	Pb	**Cd**	**Cr**	**Ni**
Pb	1			
Cd	**.951***	1		
Cr	.357	.431	1	
Ni	**.708**	**.799**	**.879***	1

*. Correlation is significant at the 0.05 level (2-tailed).

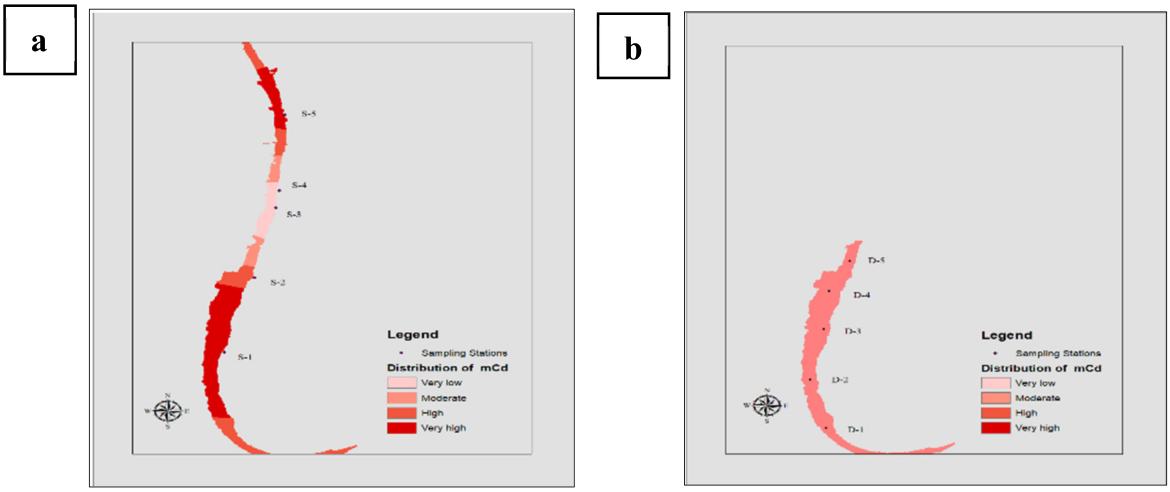

Fig. 2 : Spatial distribution of modified degree of contamination (mCd) for present and previous study in the Dhaleshwari River with a) Present Study; b) Previous Study (Mohanta et al., 2019).

at the highest CF (Pb = 20.73 and Cd = 14.66, and Ni = 3.89) at S-1, which receives municipal wastewater and mainly discharges the tannery wastewater. The contamination factors (CF) also demonstrate an anthropogenic influence on freshwater ecosystems (Förstner, 1987).

The spatial distribution of the modified degree of contamination was prepared for the present study which is shown in Figure 2 with a comparison of a previous study by Mohanta et al., (2019). Values of CF and mCd were estimated for two previous studies on the Dhaleshwari river by Ahmed et al., (2009) and Mohanta et al., (2019) which provided a comparative assessment of the indices among present and previous investigations. The values of Contamination factors analyzed by Ahmed et al., (2009) were considerable ($3 \leq CF < 6$) at every station, while those were obtained at moderate degrees ($1 \leq CF < 3$) at every station from Mohanta et al., (2019). The present study revealed that CF for all the heavy metals was the highest in the present study compared to the previous two studies. This is indicative of the continued degradation of bed sediment due to the influence of anthropogenic impact on the fresh-water ecosystems (Förstner, 1987). The revised equation was utilized to evaluate the modified degree of contamination (mCd). The modified degree of contamination for the sampling stations fell within 0.58–10.61, as demonstrated in Figure 2 for the Dhaleshwari river. Two of the five sampling stations (S-2 and S-5) exhibited mCd values above 5, while sampling station S-1 illustrated values above 9 (Figure 2a). Sampling station S-2 can be classified for a "high degree" of contamination ($4 \leq mCd < 8$), and S-1, S-5 can be classified as "very high" degree of contamination ($8 \leq mCd < 16$) respectively, while S-4 falls under "moderate degree" of contamination ($2 \leq mCd < 4$) category.

S-3 lies in the "Nil to a very low degree" of contamination (mCd < 1.5) (Figure 2a) category. At all the sampling points except S-3, the mCd values show a considerable anthropogenic effect.

Every sampling station (SH-1, SH-2, and SH-3) from Ahmed et al., (2009) shows a high degree of contamination (4 ≤ mCd < 8), while every sampling station (D-1, D-2, D-3, D-4, and D-5) calculated from Mohanta et al., (2019) lies within "moderate" degree of contamination (2 ≤ mCd < 4) (Figure 2a). In comparison with the previous study, similar sampling locations by Mohanta et al., (2019) seem to exhibit a higher degree of contamination in the present study. After considering the average mCd values, considerable accumulation could be noticed at sample stations S-1 (Savar Tannery), S-2 (Sudkhira), S-4 (Akh Knitting and Dyeing), and S-5 (Fordnagar) after comparing with baseline data. The application of antifouling paint containing lead and cadmium on the ships associated with the lower estuary is connected to this localized accumulation (Pekey et al., 2004).

Evaluation of potential ecological risk (PER) is described: followed by the relevant results. Figure 3 depicts the spatial distribution of the potential ecological factors (PER) from the Dhaleshwari river for the present and the previous studies under consideration by Ahmed et al., (2009) and Mohanta et al., (2019). The potential ecological risk approach assesses the sensitivity of an aquatic system as a function of efficiency (Islam et al., 2015), which is expressed as E^i_r. The analyzed PER values for the respective locations ranged from 9.68 to 573.07, with an average of 329.31. The lowest and the highest PER values are observed at S-3 and S-4, respectively. This highest PER (573.07) value observed in the Savar Tannery (S-1) shows High potential ecological risk (RI ≥ 260); this may have occurred due to the tanning activities. The sampling sites S-2 (Sudkhira) and S-5 (Fordnagar) also exhibit high PER values (RI ≥ 260), which indicates a high degree of ecological risk. It is an obvious speculation from the findings that every sampling station that was addressed earlier must have been contributed by leather and dying industries. On the other hand, Cd appears to pose a high risk (RI ≥ 260) in the sediments of all sampling stations except S-3. Phosphate fertilizers, non-ferrous material mining or processing, and waste disposal are anthropogenic cadmium sources in the environment (ATSDR, 2000). Numerous agricultural fields, metal processing industries, and residential buildings exist in and around the Dhaleshwari river. Cd from those sources also gets accumulated in the existing sediments,

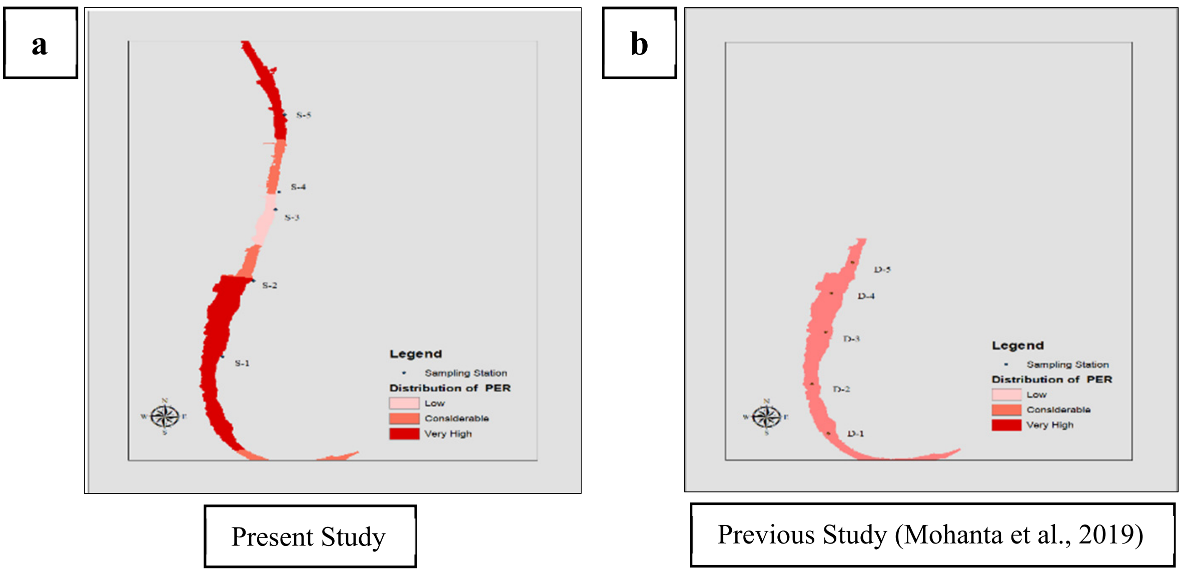

Fig. 3 (a-b): Spatial distribution of PER levels for present and previous study of Dhaleshwari River with a) Present study; b) Previous study by Mohanta et al., (2019) aquatic organisms and eventually in crops (ATSDR, 2000).

The spatial distribution for PER is illustrated in Figure 3 for the present study and a previous study by Mohanta et al., (2019). This can be further inferred from Figures 3a and 3b, respectively, that the PER values were very high at most of the stations in the present study, while the PER values were obtained at a considerable level at all sampling stations in the study by Mohanta et al., (2019). It should be noted that the sampling stations D-2 to D-3 from Mohanta et al., (2019) and S-1 are within the same area belonging to the Savar tannery. These outcomes make it evident that the PER values along the same stretch have increased, leading to elevated risk with time. Which can be attributed to the discharge of inadequately treated effluent from Savar Tannery and the other industrial activities in this area.

Evaluation of geo-accumulation index (I_{geo}) is described: followed by the applicable findings. Table 2 summarizes Muller's Sediment Classification for the Geo-Accumulation Index (I_{geo}). The comparative representation of the I_{geo} values among all three studies is provided in Figure 4. In the present study, the quality of sediments ranged from uncontaminated to strongly polluted ($0 \leq I_{geo} < 4$) for heavy metals (Figure 4a). With respect to the metal Ni, locations S-1 (Savar Tannery) and S-5 (Fordnagar) were classified as moderately polluted (Class 2), while S-2 (Sudkhira) and S-4 (Akh Knitting and Dyeing) were classified as Unpolluted to moderately polluted (Class 1) according to Table 2 classes. For Cr, locations S-1, S-4, and S-5 were classified as moderately polluted (Class 1). The sediments at sampling station S-3 could not be classified to be contaminated with Pb, Cd, Cr, and Ni, where the average I_{geo} values are associated with negative signs. Negative geo-accumulation indices indicate that heavy metal concentrations in the river sediments are lower than their background and reference levels.

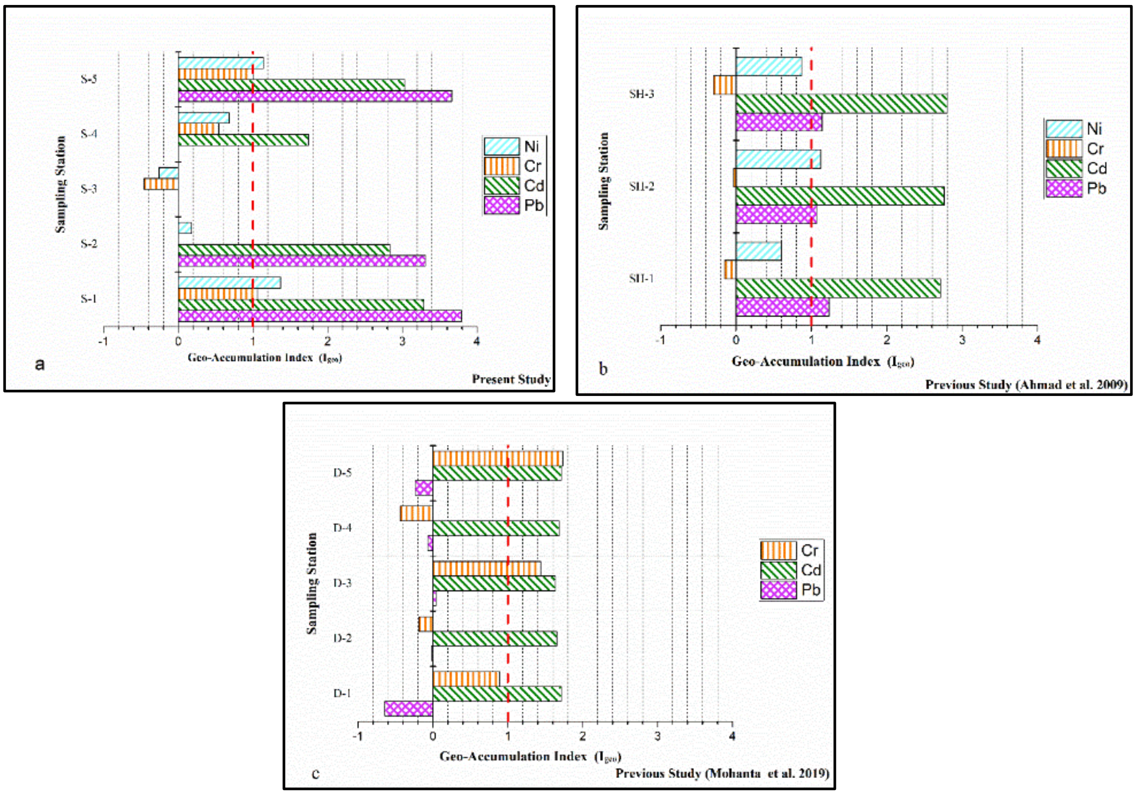

Fig. 4 (a-c): Comparison of Geo-accumulation Index (I_{geo}) for Heavy Metals in the Dhaleshwari River sediments with previous studies with a) Present study; b) Previous study by Ahmed et al., (2009); (c) Previous study by Mohanta et al., (2019). [*Red dotted line indicates the Muller's standard for Geo-accumulation Index*]

Based on the Müller's classification (Table 2), sampling stations S-1 (Savar Tannery), S-2 (Sudkhira), and S-5 (Fordnagar) could be categorized as Strongly polluted for the heavy metals Cd and Pb (Class 4) in the sediments along the Dhaleshwari river (Figure 4a). The Geo-accumulation index for Cd in most locations classified as uncontaminated to strongly polluted suggested that the surface sediments of the Dhaleshwari river are contaminated with Cd through anthropogenic activities such as tannery and dying industries. From a previous study, according to Ghrefat and Yusuf, (2006) in the middle east region, it was inferred that due to substantial industrial activities (such as tannery and dying), the sediments of the Wadi Al-Arab Dam were stable and highly polluted with Cd. These hazardous metals might have been derived from industrial waste and gasoline components used in the factories and automobiles (Mwamburi, 2003).

The geo-accumulation indices (I_{geo}) were also estimated for the heavy metals results obtained from Ahmed et al., (2009) and Mohanta et al., (2019), which are illustrated in Figure 4b and Figure 4c, respectively. Every sampling station from Ahmed et al., (2009) exhibited an uncontaminated to strongly polluted (Class 3) degree of pollution (Figure 4b), while sampling stations from Mohanta et al., (2019) exhibited an uncontaminated to moderately polluted (Class 2) degree of pollution (Figure 4c). Sampling stations from Mohanta et al., (2019) and downstream of the present study area belong to the same stretch of the Dhaleshwari river. The comparative representation of the I_{geo} values in Figure 4 exhibits that the accumulation of heavy metals has significantly increased with time.

Evaluation of the enrichment factor (EF) is described: followed by the applicable findings. Assessment of enrichment factors is presented in Figure 5(a-c) illustrating the Enrichment Factors of sediments for heavy metals in the Dhaleshwari River. Enrichment Factors were

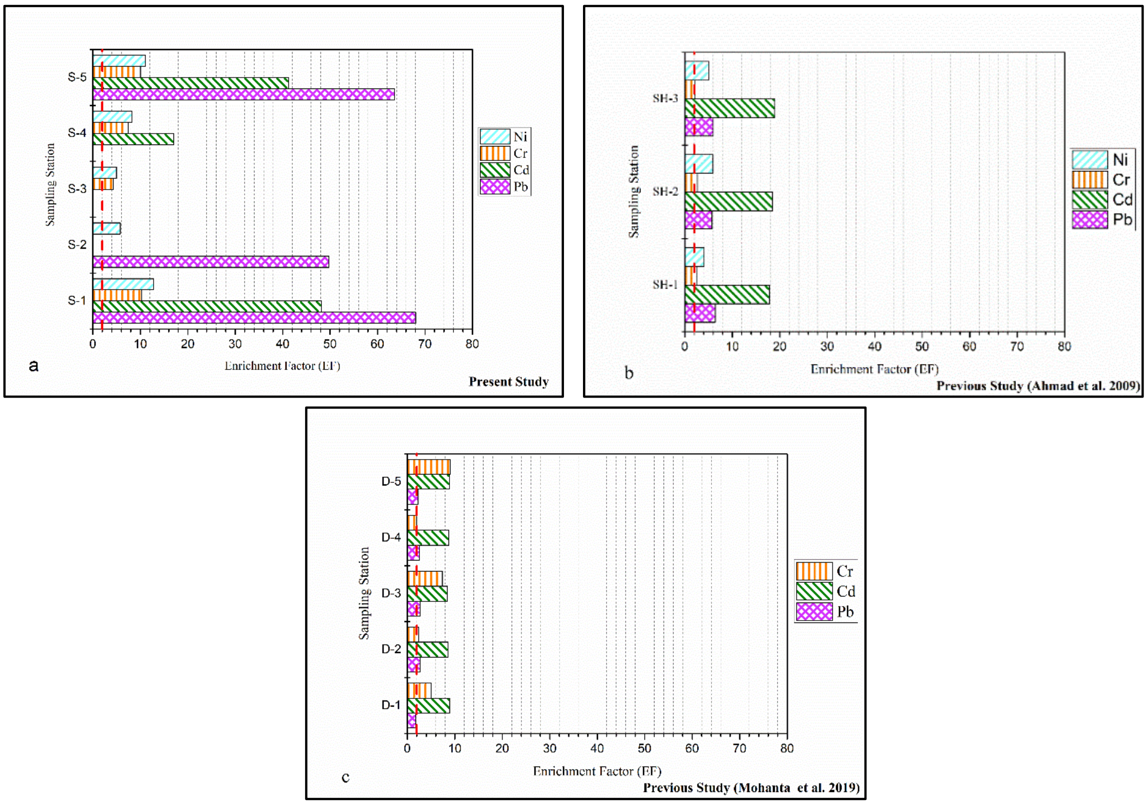

Fig. 5(a-c): Comparison of Enrichment Factor (EF) for heavy Metals in the Dhaleshwari River sediments with a) Present study; b) Previous study by Ahmed et al., (2009); (c) Previous study by Mohanta et al., (2019). [*Red dotted line indicates the EF standard (Mmolawa et al., 2011)*]

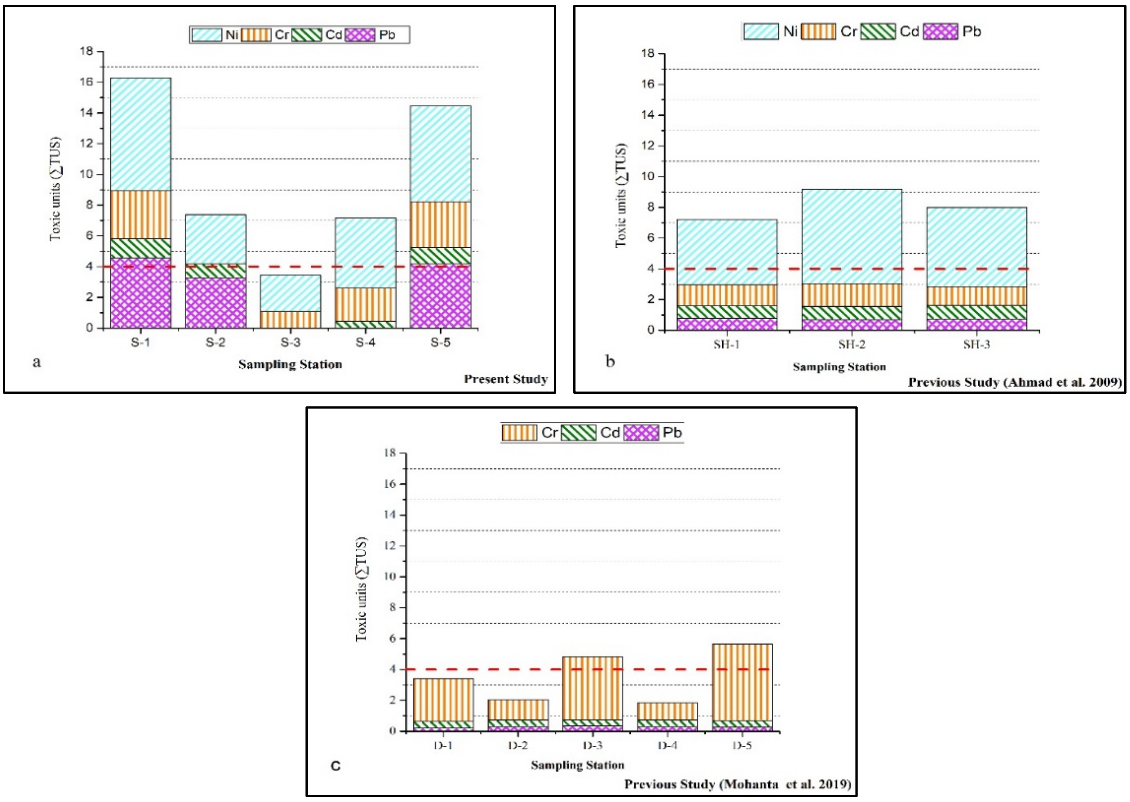

Fig. 6 (a-c): Toxic units for the heavy metals in the Dhaleshwari River sediments for a) Present study; b) Previous study by Ahmed et al., (2009); I Previous study by Mohanta et al., (2019) [*TU level at and beyond red dotted line indicates the moderate toxicity level*]

significant at sampling stations S-1, S-4 and S-5 for each metal (Figure 5a). Alternatively, minimum enrichment was observed at S-3 (Tetulijhora). Also, Deficiency in enrichment (EF < 2) was observed for Pb at S-3 and S-4, for Cd at S-3, and for Cr at S-2 (Figure 5a). The present study observed Cd in extremely high enrichment (EF ≥ 40) at the sampling stations S-1 (EF = 48.16) and S-5 (EF = 41.29) and high enrichment (20 ≤ EF < 40) at sampling station S-2 (EF = 35.68). Cadmium is a widely distributed anthropogenic metal, the accumulation of which has been attributed to human activities (Zhang and Shan, 2008). It is also related to colloidal elements in the surface runoff that are easily transmitted through river flow to a more significant extent (Wakida et al., 2008). Ni also showed significant enrichment (5 ≤ EF < 20) at the sampling stations S-1, S-2, S-4, and S-5. However, high EF values were also obtained for Pb at the sampling stations S-1 through S-5 (except S-3 and S-4), showing significant enrichment (5 ≤ EF < 20).

For comparison, Enrichment Factors (EF) were also calculated with the heavy metal data obtained from the previous studies by Ahmed et al., (2009) and Mohanta et al., (2019), and the comparative scenarios are presented in Figure 5b and Figure 5c, consecutively. Every sampling station from Ahmed et al., (2009) was found with significant enrichment (5 ≤ EF < 20) for Pb and Cd, while Cr was found at moderate enrichment (2 ≤ EF < 5) at every sampling station (Figure 5b). From the results of Mohanta et al., (2019), it was also observed that all sampling stations (except D-1) were at moderate enrichment (2 ≤ EF < 5) for Pb and at significant enrichment (5 ≤ EF < 20) for Cr at all sampling stations (except D-2 and D-4) (Figure 5c). It is evident from Figure 5 shows that metal enrichment has significantly increased at present since the onset of both of the previous studies.

The toxic unit analysis is described: followed by the applicable findings. Figure 6(a-c) shows a comparative assessment of toxic units (TUs) for the heavy metals in Dhaleshwari River sediments based on the findings from the other studies, including the present one. Heavy metal toxic units in the Dhaleshwari River showed a decreasing trend in the order Ni > Pb > Cr > Cd in the present study (Figure 6a). Toxic units for Ni in Dhaleshwari River sediments are significantly higher than the toxic units of the other heavy metals at sampling points, exceeding cumulative toxic units of Pb, Cd, and Cr. Except for S-3, the summation of toxic units (∑TUs) for all the stations was substantially higher than four, indicating that the depositional behavior in the river under investigation was exposed to moderate to severe heavy metal toxicity (∑TUs > 4, suggesting moderate to serious toxicity) (Figure 6a) (Xiao et al., 2012).

Ni exhibits a toxic impact if it is available above the safe level though not toxic in general (Das et al., 2008). It functions as a cofactor for various enzymes and is involved in urea metabolism, the hydrogen cycle, and the fixation of nitrogen, among other things (Maleva et al., 2016). Above safe level, Nickel could be neurotoxic, genotoxic, reproductively toxic, toxic to the lungs, nephrotoxic, and carcinogenic (Das et al., 2008). The elevated toxicity levels in the present study may be attributed to the presence of Organophosphate pesticides, specific chlorpyrifos (Das et al., 2008). Furthermore, the values of TUs estimated from the heavy metal data from Mohanta et al., (2019) revealed that almost 65% of sediment samples exhibited TU levels above 4, suggesting moderate toxicity (Figure 6c) (∑TUs >4, suggesting moderate toxicity). After the relocation of the tanneries from Hazaribagh next to the Dhaleshwari River, the overall concentrated and spatial deterioration of the river water quality was observed. Among the four important metals analyzed for the samples from the selected points, a higher concentration of Pb was obtained at the outfalls S-1, S-2 (Savar tannery) and S-5 (Dying area), which could have a detrimental effect. According to the spatial variation of Pb levels, pollution arises from urban runoff and industrial activities. The primary sources of Pb in the urban area include municipal runoffs, untreated or poorly treated industrial effluents, and atmospheric deposition (Varol, 2012) and similar activities were observed along the Dhaleshwari river bank in Savar city. Significant enrichment of Cr concentration was observed in each selected point indicating an alarming level of Cr discharge in both Savar tannery and Fordnagar areas. Except for S-2 (Sudkhira), all the selected areas were contaminated with significant concentrations of Cr. Textiles utilize chromium compounds such as pigments, mordants, and dyes, whereas leather uses chromium compounds as a tanning agent (Islam et al., 2014). The toxicity of trivalent and hexavalent Cr differs significantly; hexavalent Cr is substantially more hazardous than trivalent Cr (Majed and Islam, 2022). If continued discharge from point sources or upstream can be avoided, it may take 18 years for Cr to abate naturally based on its residence duration (Majed et al., 2021). Overall, Ni concentration, as reported in the present study, was also observed at a significant level over the selected stretch, which could have a significant diverse concern in the selected area.

This study also observed enrichment factor (EF) to be of "extremely high enrichment" category (EF ≥ 40) at the Savar Tannery, Fordnagar area, and of "high enrichment" (20 ≤ EF < 40) at Sudkhira area. The observed trends of EF values for Ni and Pb suggested that common sources of these metals were associated with automobiles and tannery operations (Majed et al., 2021). Two possible sources of Ni and Pb in Dhaleshwari river sediments are septic tank leaks and leaded gasoline (Sayadi et al., 2010). Long term enrichment of metals like this in river sediments could lead to significant deterioration of aquatic habitats, and the toxicity could adversely impact the ecosystem and ultimately accumulate in the food chain (Helen al., 2016). The present study also confirmed that the quality of sediments ranged from uncontaminated to strongly polluted ($0 \leq I_{geo} < 4$) with respect to heavy metals. According to the results, higher sediment toxicity was mainly distributed in the industrial zones of the Dhaleshwari river (around the Savar tannery area) and the northeast area near the industrial sector (fish market). As a result, additional sources must be controlled by lowering heavy metal concentrations in industrial discharge around the Dhaleshwari River and the tannery sector must construct buffer zones. Analysis of different environmental indices revealed that most of the locations face severe sediment pollution in terms of Pb, Cd and Cr. The relocation of tanneries with its Central Effluent Treatment Plant (CETP) from Hazaribagh next to the Dhaleshwari River is responsible for the overall concentrated and spatial deterioration of the river water quality. The inadequacy in the complete functioning, inefficiency in treating the wastewater, and lack of maintenance of the CETP result in a significantly increased pollution level. However, Majed et al., (2021) reported a relatively lower pollution level in different environmental compartments (in terms of sediment) for the Buriganga river compared to the present study after removing the tanneries from the riverside. As mentioned previously, there is no such comprehensive study after the relocation process and this study was initiated with the hypothesis of progressive deterioration of the rehabilitation. The study results confirm that the Dhaleshwari river has been progressively going through enrichment with higher levels of heavy metals than in previous after the relocation of tanneries from Hazaribagh (besides the Buriganga river) to Savar (beside Dhaleshwari). Previous studies rarely attempted to make comparative studies among the rivers and also the progression of deterioration of sediments through comprehensive and focused investigation. The correlations and the demonstrations in the present study bring out the necessity of monitoring the different compartments of a river ecosystem with respect to the toxic contaminants and addressing the gaps in inventories for an important river that emanates from infrequent investigations. The present study, finally emphasizes the significance of the preservation of a river system through prioritizing the safe discharge of toxic contaminants from industrial establishments.

CONCLUSIONS

Monitoring of sediment quality of the rivers has become a vital issue in Bangladesh, where rivers are being polluted without considering any ecological impact. It is essential to perform subsequent monitoring programs along the rivers to analyze the pollution progression and dynamics and adopt necessary actions to rehabilitate the rivers. It is also imperative to preserve the ecological balance and ensure sustainable human development by maintaining a healthy river ecosystem. Considering these facts, the present study was performed for monitoring purposes, mainly focusing on the Savar tannery area. Heavy metal levels in the sediments were compared to toxicological reference standards and some previous studies for Dhaleshwari and some of the other rivers in Bangladesh. The maximum pollution of heavy metals was observed at the Savar Tannery location. Nevertheless, sampling stations in Sudkhira, Akh Knitting and Dyeing and Fordnagar were also characterized and categorized with considerable metal concentrations due to municipal and industrial wastewater discharge. Concentration levels of Cr (282.4 mg/kg), Cd (4.4 mg/kg), Ni (264.5 mg/kg), and Pb (414.6 mg/kg) suggested that they might have a detrimental effect on sediment-dwelling microorganisms, which are likely to occur regularly. Pearson's correlation (PC) among the analyzed heavy metals (Pb, Cd, Cr, and Ni) revealed that they might be linked with anthropogenic origins. This led to a similar source input indicating that these components in the river sediment discharges have a natural origin. Evaluation of several indices such as geo-accumulation index (I_{geo}) (I_{geo} >6), enrichment factor (EF) (EF ≥ 40), and Toxic Unit (∑TUs >4) revealed highly deleterious effects for all the studied metals at Sampling Stations along Savar Tannery, Akh Knitting and Dyeing and Fordnagar which are causing alarming levels of metal-based discharge through dyeing and tanning activities. The current study provides a brief overview status of pollution associated with the Dhaleshwari river sediment and potential future ecological risks. There is a compelling necessity to monitor the treatment systems that discharge into the rivers regularly and implement various strategies to prevent heavy metals accumulation in sediment are advised, which might be helpful in the abatement and prevention of long-term ecological risk.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

LIFE SCIENCE REPORTING

The authors declare that no experiments were performed in this study involving human or animal subjects.

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