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Ecological Risk Analysis of Heavy Metals Toxicity from Agricultural Soils in the Industrial Areas of Tangail District, Bangladesh

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Abstract

The aim of this research was to analyze ecological risk of heavy metals from agricultural soils in the industrial areas of Tangail district in Bangladesh. In this research, six heavy metals (Cr, Ni, Cu, As, Cd and Pb) were assessed in 10 different sampling locations around the industrial areas of Tangail district. Certain indices, including the toxic unit analysis, principal component analysis (PCA), enrichment factor (EF), contamination factor (C_f), geoaccumulation index (I_{geo}) and pollution load index (PLI) were used to assess the potential ecological risk posed by heavy metals in soils. The ranges of Cr, Ni, Cu, As, Cd and Pb in studied soils were 0.22–40.1, 0.71–92.1, 1.03–72.7, 0.99–11.7, 0.14–9.04 and 1.19–81.4 mg/kg, respectively. The enrichment factor of all the studied metals for all sampling sites were in the descending order of Cd>Cu>As>Pb>Cr>Ni. The contamination factor (CF) values of Cd ranged from 0.74 to 18.9 revealed that the studied soils were highly impacted by Cd. The pollution load index (PLI) values of Cd were higher than 1, indicating the progressive deterioration of soil due to Cd contamination. Potential ecological risk (PER) of soils from all sampling sites showed considerable to very high potential ecological risk.

Keywords: Heavy metal; Potential ecological risk; Industrial areas; Bangladesh

Introduction

Soil contamination by heavy metals is a universal problem that is greatly predisposed by anthropogenic activities [1,2]. Soil a key element for human life to survive on the planet which is assumed as prime receiver of persistent pollutants such as toxic heavy metals [3-5]. Nowadays, soil pollution by heavy metals is regarded as the most adverse environmental issue in the universe [4]. According to the US Environment Protection Agency (EPA), heavy metals such as Cr, Ni, Cu, As, Cd and Pb have been considered as the most heavy metals in the environment [6,7]. Heavy metals are of great concern due to their wide sources, toxicity, non-biodegradable properties and accumulative behaviors [4,8]. In recent decades, there has been a major concern regarding soil pollution by various heavy metals due to rapid industrialization and urbanization, especially in developing countries [4,9,10]. Heavy metals may originate in soils around the industrial area from numerous prime sources but industrial activities are the most important one and also generation of power, manufacturing, burning of fossil fuel and disposal of waste [11,12]. The accumulations of heavy metals in soils are a great concern due to their potential environmental risk and have adverse effects on soil bionetworks [4,13,14]. To assess the ecological risks of heavy metals in soil different methods have been widely used, such as enrichment factor (EF), contamination factor (CF), and geoaccumulation index (I_{geo}) [2,15]. Enrichment factor of a vicinity address relative enhancement in any toxic element when pre-industrial soils are compared with studied soils in alike vicinity [16,17]. Since contamination of soil derive from industry, present study area have got more attention for its pollution in the environment facing threats for heavy metals toxicity contamination derived from the exponential development, industrial activities and congestion [2,18]. Numerous studies have stated the concentration of heavy metals in the industrial area soils in Bangladesh [8,19]. Therefore, the objectives of this study were to investigate the variations of heavy metals in soils of different soil sampling sites and to assess ecological risk of heavy metals in soil in the industrial areas of Tangail

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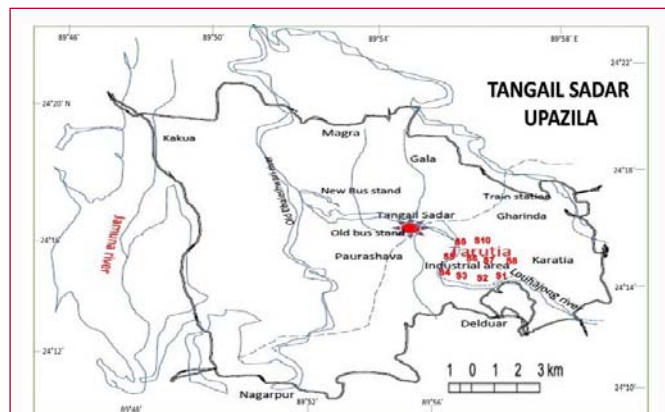


Figure 1: Map of the sampling sites of industrial areas in Tangail district, Bangladesh.

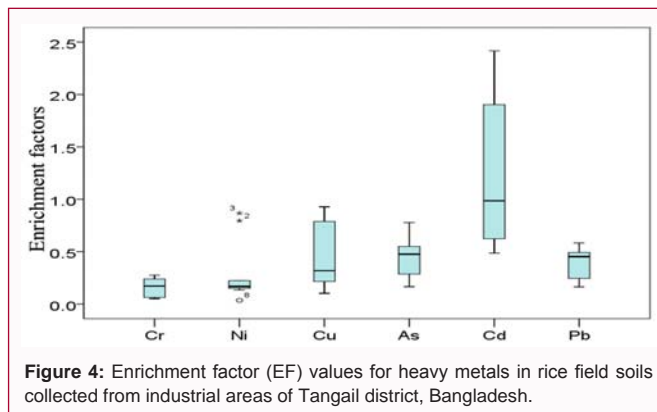


Figure 4: Enrichment factor (EF) values for heavy metals in rice field soils collected from industrial areas of Tangail district, Bangladesh.

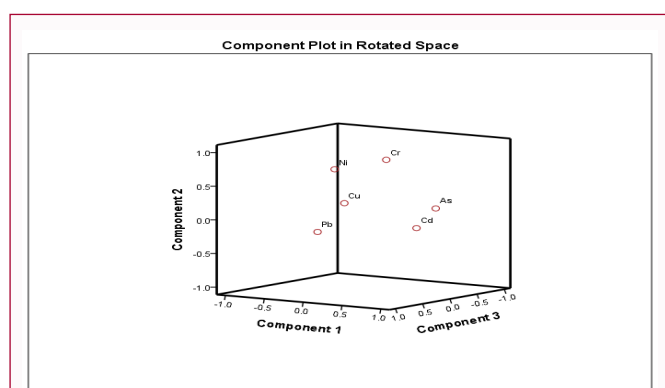


Figure 2: Principle component analysis (PCA) of heavy metals in rice field soils of industrial areas of Tangail district, Bangladesh.

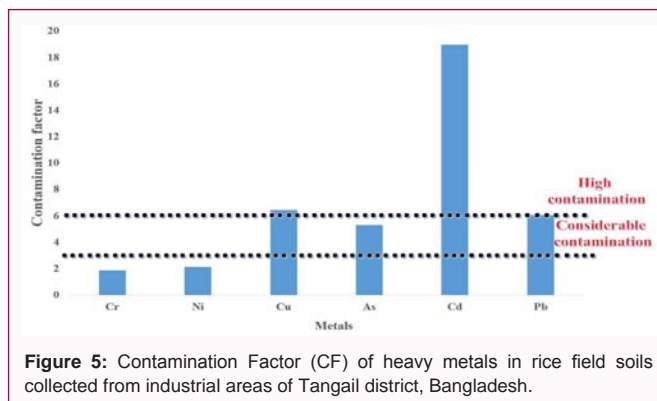


Figure 5: Contamination Factor (CF) of heavy metals in rice field soils collected from industrial areas of Tangail district, Bangladesh.

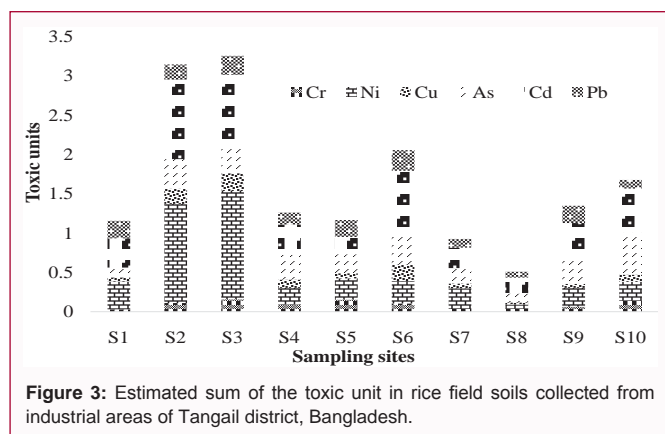


Figure 3: Estimated sum of the toxic unit in rice field soils collected from industrial areas of Tangail district, Bangladesh.

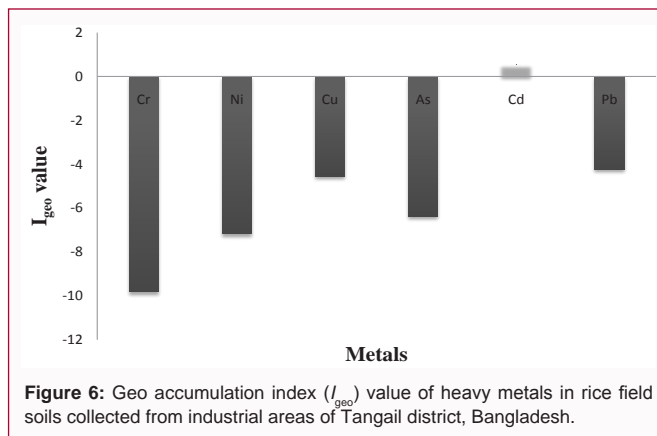


Figure 6: Geo accumulation index (I_{geo}) value of heavy metals in rice field soils collected from industrial areas of Tangail district, Bangladesh.

district in Bangladesh.

Materials and Methods

Study area and sampling

The samples were collected from Tarutia, Tangail Sadar Upzila of Tangail district, Bangladesh (Figure 1). Tangail district area is 334.26km² and situated at the middle part in Bangladesh. Tangail Sadar Upzila is highly densely area in Bangladesh and population density is 1,100/km² in Tangail district. The study area is situated between Tangail Sadar is located at 24.2500°N to 89.9167°E. Tangail as an industrial vicinity of Bangladesh possess highly vulnerable to environmental pollution now days. There present different kinds of industries in Tangail district like garments, packaging industry,

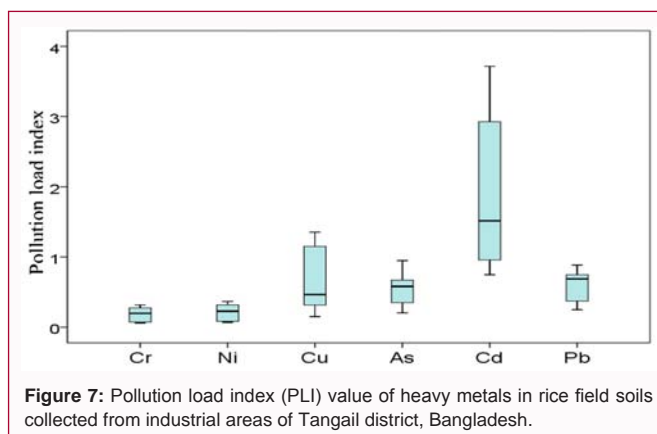


Figure 7: Pollution load index (PLI) value of heavy metals in rice field soils collected from industrial areas of Tangail district, Bangladesh.

dyeing, brick kiln, metal work-shops, battery manufacturing industries, tanneries, textile industries, pesticide and fertilizer

industries, different food processing industries and other factories produce huge volumes of effluents that contain trace metals. These industries discharged untreated wastes randomly to river and canals. Then that wastes are mixed with soils and the soil is continuously polluted by toxic elements in the industrial areas of Tangail district in Bangladesh. Ten different soil sampling sites were selected in the industrial areas of Tangail district during March-April, 2017. About 50 soil samples were collected from 10 different sampling stations in the industrial areas of Tangail district during March-April, 2017. Agricultural soil samples (up to 10cm) were collected in the form of three subsamples. These sub-samples were thoroughly mixed to form a composite sample. Samples were air-dried at room temperature for two weeks, then ground and homogenized. For metal analysis, soil was taken with the help of a percussion hammer corer (50–80 cm in length) for metal analysis and these samples were treated as preindustrial sample [20]. To crumble all dried soil samples, a porcelain mortar and pestle were used. Then the samples were sieved with 2mm nylon sieve. The soil samples were stored in a clean Ziploc bag which was airtight and used for chemical analysis. Several researches also followed the alike procedure for sampling and storing of soil samples [21-23].

Soil physiochemical properties measurements

Soil pH was determined by using a glass electrode pH meter (WTW pH 522; Germany). Ten grams of air dried soil from each sample site was taken in 50mL beakers separately and 25mL of distilled water was added to each beaker. The suspension was stirred well for 20 minutes and allowed to stand for about 30 minutes. Then each sample was stirred again for 2 minutes before taking the reading. The position of the electrode was immersed into the partly settled soil suspension and pH was measured. The result was reported as soil pH measured in water (soil: water ratio was 1:2.5). The electrical conductivity (EC) of collected soil samples were determined electrometrically (soil water ratio was 1:5) by a conductivity meter (WTW LF 521; Germany). Twenty gram of air dried soil was taken in a plastic container and 100mL of distilled water was added to it. The suspension was stirred for 30 minutes intermittently and then allowed to stand for 30 minutes. Then the electrical conductivity was determined by an electrical conductivity meter (Calibrated with 0.01N KCl solutions). The results of EC were expressed in desi Siemens per meter (dS/m). For organic carbon measurement, at first the soil was ground and completely passed through a 0.2mm sieve, and then 1.00g of that soil was placed at the bottom of a dry 500mL conical flask (Corning/Pyrex). Then 10mL of 1N $K_2Cr_2O_7$ was added into the conical flask and swirled a little. The flask was kept on asbestos sheet. Then 20mL of concentrated H_2SO_4 was added into the conical flask and swirled again 2-3 times. The flask was allowed to stand for 30 minutes and thereafter 200mL of distilled water was added. After incorporation of 5mL of phosphoric acid and 40 drops of diphenylamine indicator the contents were titrated against ferrous ammonium sulphate solution till the color flashes blue-violet to green. Simultaneously, a blank titration was run without soil. The amount of organic carbon in the sample was calculated by using the following formula-

$$\text{Organic carbon (\%)} = \left\{ \frac{10(B - T)}{B} \right\} \times 0.003 \times (100/\text{weight of soil})$$

where, B=Volume (in mL) of standard ferrous ammonium sulphate solution.

Required for blank titration and,

T=Volume (in mL) of standard ferrous ammonium sulphate

solution.

Needed for soil sample.

The organic matter was calculated by multiplying the content of organic carbon by Van Bemmelen factor, 1.73 and formula is as follows:

$$\text{Organic matter (\%)} = \text{OC (\%)} \times 1.73$$

Hydrometer method was used for texture analysis of soil. Fifty grams of oven dried soil was taken in a dispersion cup and 10mL of 5% calgon solution was added to the samples and allowed to soak for 15 minutes. Then 90mL distilled water was added to the cup. The suspension was then stirred with an electrical stirrer for 10 minutes. The content of the dispersion cup was then transferred to a liter sedimentation cylinder and distilled water was added to make the volume up to the mark. A cork was placed on the mouth of the cylinder and the cylinder was inverted several times until the whole sediment mass appeared in the suspension. The cylinder was set upright and the hydrometer readings were taken at 40 seconds and 2 hours of sedimentation. The corrections of hydrometer readings were made as the hydrometer was calibrated at 68°F. The percentage of sand, silt and clay were calculated as follows:

$$\% (\text{Silt} + \text{Clay}) = (\text{Corrected hydrometer reading at 40 seconds/ Oven dry weight of soil}) \times 100$$

$$\% (\text{Clay}) = (\text{Corrected hydrometer reading after 2 hours/ Oven dry weight of soil}) \times 100$$

$$\text{Sand (\%)} = 100 - \% (\text{Silt} + \text{Clay})$$

$$\text{Silt (\%)} = \% (\text{Silt} + \text{Clay}) - \% \text{Clay}$$

Sample analysis

Analytical grade reagents were used for sample analysis and for the preparation of the solution Milli-Q (Elix UV5 and MilliQ, Millipore, USA) water was used. The Teflon vessel and polypropylene containers were cleaned, soaked in 5% HNO_3 for more than 24h, then rinsed with Milli-Q water and dried. For metal analysis, 0.3g of the soil sample was treated with 4.5mL 35% HCl (Kanto Chemical Co, Tokyo, Japan) in a closed Teflon vessel added with 1.5mL 69% HNO_3 (Kanto Chemical Co, Tokyo, Japan) and was digested in a Microwave Digestion System (Bergh of speed wave®, Eningen, Germany). The digested solution was then filtered using a syringe filter (DISMIC®-25HP PTFE, pore size= 0.45 μ m) and 50mL polypropylene tubes (Nalgene, New York, NY, USA) were used for storing the filtrate solution.

Instrumental analysis and quality control

For sample analysis of heavy metals, inductively coupled plasma mass spectrometer (ICP-MS, Agilent 7700 series, Santa Clara, CA, USA) was used. For metal analysis, instrument operating conditions and parameters are done. The ICP-MS detection limits for the studied metals were 0.7, 0.6, 0.8, 0.4, 0.06 and 0.09ng/L for Cr, Ni, Cu, As, Cd and Pb, respectively. Calibration curve was made by using Multi element Standard XSTC-13 (Spex CertiPrep®, Metuchen, USA) solutions. Internal calibration standard solutions containing 1.0mg/L of indium, yttrium, beryllium, tellurium, cobalt and thallium were purchased from Spex CertiPrep1 (Metuchen, NJ, USA). During the procedure, 10mg/L internal standard solution was prepared from the primary standard and added to the digested samples. 1.0 μ g/L Multi-

Table 1: Physiochemical properties of rice field soils collected from industrial areas of Tangail district, Bangladesh.

Land types		pH (1:2.5 H ₂ O)	EC (dS/m)	Organic matter (%)	Sand (% in <2mm)	Silt	Clay	Soil type ^a
S1	Range	6.04-6.22	0.06-0.11	1.09-7.46	37.2-43.5	42.5-47.5	11.5-20.3	Loam
	Mean±SD ^b	6.11±0.19	0.08±0.02	4.88±3.44	39.6±2.65	45.4±2.08	14.9±3.45	
S2	Range	5.87-6.48	0.06-0.18	1.07-4.54	36.5-46.5	36.5-47.5	11.5-16.9	Loam
	Mean±SD ^b	6.1±0.23	0.11±0.05	2.62±1.77	42.1±3.88	42.9±4.86	14.9±2.41	
S3	Range	6.69-7.27	0.07-0.70	0.38-5.38	49.7-74.0	9.1-27.5	11.5-22.8	Sandy loam
	Mean±SD ^b	6.92±0.23	0.32±0.28	1.93±1.97	63.9±9.08	19.8±7.6	16.2±4.21	
S4	Range	6.24-7.23	0.2-0.48	0.26-2.67	46.5-71.0	19.1-37.5	15.1-23.3	Sandy loam
	Mean±SD ^b	6.73±0.41	0.31±0.11	1.11±1.14	55.9±9.91	29.6±8.0	15.1±5.92	
S5	Range	7.45-8.01	0.41-1.46	0.26-2.49	38.5-61.5	21.6-49.1	12.4-20.1	Loam
	Mean±SD ^b	7.77±0.21	0.95±0.38	1.38±0.83	49.6±8.52	34.1±9.84	16.2±3.06	
S6	Range	6.96-7.91	0.09-2.11	0.32-3.75	37.5-61.0	25.0-44.9	14.0-22.8	Loam
	Mean±SD ^b	7.45±0.46	1.07±0.88	2.27±1.54	48.9±9.76	33.9±7.99	17.1±3.45	
S7	Range	5.48-6.36	0.23-0.64	1.14-4.6	36.0-51.0	32.5-51.6	11.5-18.5	Loam
	Mean±SD ^b	6.0±0.34	0.42±0.17	2.41±1.41	44.9±5.91	40.6±7.38	14.4±2.95	
S8	Range	6.3-7.11	0.21-0.43	1.11-2.84	41.5-60.1	26.6-39.1	12.4-19.4	Loam
	Mean±SD ^b	6.61±0.36	0.30±0.09	1.72±0.71	50.3±7.05	34.4±5.79	15.2±2.72	
S9	Range	6.3-7.15	0.21-0.47	1.61-5.76	47.6-50.12	34.1-46.6	11.5-16.9	Loam
	Mean±SD ^b	6.62±0.33	0.30±0.1	2.98±1.69	48.3±2.12	38.2±5.02	13.9±2.29	
S10	Range	5.99-6.65	0.17-0.31	1.11-2.43	44.0-49.0	31.0-36.6	19.4-22.0	Loam
	Mean±SD ^b	6.32±0.27	0.21±0.06	1.66±0.49	46.2±1.92	32.8±2.22	20.9±1.42	

^aAccording to the United states Department of Agriculture soil classification system; ^bSD: Standard Deviation.

Table 2: Heavy metals (mg/kg) in rice field soils collected from industrial areas of Tangail District, Bangladesh.

Sites		Cr	Ni	Cu	As	Cd	Pb
S1	Range	0.22-5.93	8.75-16.5	4.78-17.9	0.99-3.43	0.19-5.14	1.19-81.4
	Mean±SD ^a	2.68±2.31	12.7±3.21	10.3±6.43	2.11±0.92	1.3±2.15	20.1±34.4
S2	Range	5.43-13.2	11.0-87.7	15.4-57.6	3.99-9.01	0.83-9.04	3.36-37.4
	Mean±SD ^a	9.98±3.93	45.3±38.1	37.9±15.0	6.37±1.8	3.53±3.59	17.6±12.6
S3	Range	5.63-33.6	15.3-92.1	15.7-70.3	2.5-9.93	0.5-8.47	14.8-31.7
	Mean±SD ^a	14.2±11.2	49.4±29.4	44.6±25.7	5.89±3.48	3.15±3.53	22.1±6.65
S4	Range	4.59-13.4	0.82-15.7	4.48-61.8	3.03-8.7	0.6-2.23	6.94-16.6
	Mean±SD ^a	8.2±3.81	7.64±7.12	20.5±23.5	5.34±2.17	1.35±0.58	13.7±3.93
S5	Range	3.06-40.1	1.87-21.2	6.94-21.9	3.24-4.97	0.29-1.3	16.3-24.7
	Mean±SD ^a	13.9±15.8	9.67±8.04	12.9±7.54	4.1±0.63	0.77±0.36	19.5±3.39
S6	Range	3.72-20.7	3.69-15.4	3.35-72.7	1.0-11.74	0.14-6.57	1.79-36.1
	Mean±SD ^a	9.52±6.68	10.2±5.86	41.4±30.7	6.72±4.2	2.78±2.53	23.8±14.1
S7	Range	0.96-5.05	3.01-27.7	2.04-28.6	1.31-5.75	0.41-2.61	2.32-14.7
	Mean±SD ^a	3.19±1.66	9.5±10.4	11.7±10.5	3.32±1.81	0.91±0.95	10.0±4.68
S8	Range	0.41-5.92	0.71-3.35	2.26-9.74	1.2-3.02	0.19-1.59	2.02-12.1
	Mean±SD ^a	3.27±2.44	2.04±0.93	4.96±3.12	1.92±0.70	0.71±0.55	6.71±4.0
S9	Range	4.21-10.5	3.25-18.4	1.03-34.4	1.37-10.3	0.52-2.44	13.5-28.6
	Mean±SD ^a	5.81±2.67	8.59±6.11	10.0±13.8	5.69±3.63	1.53±0.79	19.9±7.26
S10	Range	5.93-16.3	7.1-11.0	11.6-21.2	7.56-11.2	1.23-3.31	5.11-17.5
	Mean±SD ^a	12.3±3.9	8.77±1.67	17.7±3.72	9.01±1.42	1.97±0.83	9.92±4.64
Dutch Standard ^b		100	35	36	29	0.8	85
Canadian Guidelines ^c		64	50	63	12	1.4	70
Australian Guidelines ^d		50	60	60	20	3	300
Environmental Standard ^e	Class 1	90	40	35	15	0.2	35
	Class 2	250	60	100	25	1	350
Background value in Tangail district		29	32	27	6.5	0.82	23

^aSD: Standard Deviation; ^bVROM (2000); ^cCCME (2003); ^dDEP (2003); ^eSEPA 1995. Class 1 is the natural background level; class 2 is for the need of agricultural production and human health.

Table 3: Comparison of metal concentration (mg/kg) in soil of present study with other study.

District (Country)	Cr	Ni	Cu	As	Cd	Pb	References
Tangail, Bangladesh	8.31 (0.22-40.1)	16.4 (0.71-92.1)	20.6 (1.03-72.7)	5.06 (0.99-11.7)	2.20 (0.14-9.04)	16.9 (1.19-81.4)	Present study
Tangail, Bangladesh	10.4 (1.57-21.9)	12.6 (4.74-25.7)	15.6 (3.08-38.6)	12.1 (2.69-28.4)	3.1 (1.03-8.06)	7.98 (2.23-18.3)	Proshad et al., (2017)
Guandong (China)	12.3 (9.66-19)	8.83 (7.04-10.3)	324 (210-450)	NA	0.9 (0.26-1.17)	96 (73-134)	Luo et al., (2011)
Bogra (Bangladesh)	41 (6.6-87)	45 (15-95)	42 (6.4-107)	10 (2.0-36)	4.2 (0.7-10)	44 (13-96)	Islam et al., (2014)
Dhaka (Bangladesh)	54 (34-68)	58 (36-74)	39 (31-45)	NA	11 (6-16)	50 (44-52)	Ahmad and Goni (2010)
Dutch soil quality standard (Target Value)	100	35	36	29	0.8	85	VROM (2000)
Dutch soil quality standard (Intervention Value)	380	210	190	55	12	530	VROM (2000)
Canadian Environmental Quality Guidelines	64	50	63	12	1.4	70	CCME (2003)
Department of Environmental Protection, Australia	50	60	60	20	3	300	DEP (2003)

Table 4: Correlation coefficient matrix for physiochemical properties of soils and heavy metals collected from rice field around industrial areas of Tangail district, Bangladesh.

	pH	EC	Sand	Silt	Clay	Organic matter	Cr	Ni	Cu	As	Cd	Pb
pH	1											
EC	0.66**	1										
Sand	0.088**	0.037	1									
Silt	-0.36**	-0.248	-0.056	1								
Clay	0.017	-0.03	0.038		1							
Organic matter	-0.18	-0.101	0.173	0.354**	-0.228	1						
Cr	0.28*	0.337**	-0.03	-0.259*	-0.037	-0.172	1					
Ni	0.005	-0.094	0.049	-0.138	-0.008	-0.064	0.361**	1				
Cu	0.26*	0.125	-0.041	-0.358**	0.154	-0.094	0.114	0.461**	1			
As	-0.016	-0.123	-0.039	0.09	0.061	-0.049	0.298*	0.028	0.207	1		
Cd	-0.046	-0.199	-0.034	-0.073	-0.034	-0.012	0.064	0.093	0.275*	0.485**	1	
Pb	0.24	0.226	0.002	-0.056	-0.008	-0.142	-0.011	0.133	0.291*	-0.036	0.087	1

*Correlation is significant at the 0.05 level (two-tailed); **Correlation is significant at the 0.01 level (two-tailed).

element solution (Agilent Technologies, USA) was used as tuning solution covering a wide range of masses of elements. For evaluation of every test batches an internal quality system was used and once defined Internal Quality Controls (IQCs) was satisfied than it was validated. Relative standard deviation (RSD, <5%) was checked by using tuning solution purchased from Agilent Technologies before starting the analysis sequence. After that, the vessels were washed by Milli-Q water and dried with air.

Ecological risk assessment for soil pollution

Enrichment factor (EF): Enrichment factor assumed an impressive tool and used for determining toxic element magnitude of environment. In soil, anthropogenic influences of toxic metals were assessed by enrichment factor and following formula was used.

$$EF = (C_M/C_{Al})_{\text{sample}} / (C_M/C_{Al})_{\text{background}} \quad (1)$$

where, $(C_M/C_{Al})_{\text{sample}}$ is assumed as proportion of hazardous element concentration of (C_M) to that of aluminum (C_{Al}) in the soil sample, and $(C_M/C_{Al})_{\text{background}}$ is the same reference ratio in the background sample. Enrichment factor value of toxic element is equal to 1 indicate that toxic elements arise due to natural weathering processes in the environment [24]. Enrichment factor effects of metals known as minor, moderate, severe, and very severe modification when enrichment factor value are 1.5–3, 3–5, 5–10 and >10 respectively [25].

Contamination factor (C_i^i): Contamination factor means the proportion of the heavy metal concentration in the soil to that of baseline or background value. Contamination factor was calculated by using the following equation:

$$C_i^i = C_{\text{heavy metal}} / C_{\text{background}} \quad (2)$$

Contamination factor divided on four classes ranged from 1 to 6 which are: low degree ($C_i^i < 1$), moderate degree ($1 \leq C_i^i < 3$), considerable degree ($3 \leq C_i^i < 6$), and very high degree ($C_i^i \geq 6$) [2].

Geoaccumulation index (I_{geo}): Geoaccumulation index (I_{geo}) is assumed as an impressive tool to determine contamination degree from toxic metals. At present, geoaccumulation index is used globally to assess soil pollution [26]. The most effective objective to determine geoaccumulation index (I_{geo}) is to identify pollution level in soil. Geoaccumulation index (I_{geo}) may be assessed by applying equation given here by,

$$I_{\text{geo}} = \log_2(C_n / 1.5B_n) \quad (3)$$

where, C_n is the determined element (n) concentration assessed from soil, B_n is the geochemical baseline value of element n in background sample [13]. Geoaccumulation index (I_{geo}) values were categorized as: $5 < I_{\text{geo}}$ – extremely contaminated; $4 \leq I_{\text{geo}} \leq 5$ – heavily to extremely contaminated; $3 \leq I_{\text{geo}} \leq 4$ – heavily contaminated; $2 \leq I_{\text{geo}} \leq 3$ – moderately to heavily contaminated; $1 \leq I_{\text{geo}} \leq 2$ –

Table 5: Total variance explained and component matrices for the hazardous elements in rice field soils collected from Tangail district, Bangladesh.

Initial Eigen values				Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
Component	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
1	2.009	33.84	33.48	2.009	33.48	33.48	1.54	25.81	25.81
2	1.286	21.43	54.92	1.286	21.43	54.92	1.43	23.92	49.73
3	1.11	18.49	73.41	1.11	18.49	73.41	1.42	23.67	73.41
4	0.741	12.35	85.76						
5	0.498	8.3	94.07						
6	0.356	5.93	100						
Elements	Component matrix			Rotated Component Matrix					
	PC1	PC2	PC3		PC1	PC2		PC3	
Component matrix									
Cr	0.518		-0.697			0.836		0.432	
Ni	0.63	0.488	-0.362			0.758		0.719	
Cu	0.724	0.341				0.329			
As	0.598	-0.656			0.861				
Cd	0.603	-0.485	0.4		0.839				
Pb	0.316	0.511	0.532					0.793	

Table 6: Potential ecological risk factor, risk index and pollution degree of heavy metals in rice field soils collected from industrial areas of Tangail district, Bangladesh.

Sites	Potential ecological risk factor (E ⁱ)						Potential Risk (PER)	Pollution degree
	Cr	Ni	Cu	As	Cd	Pb		
S1	3.21	7.35	10.2	85.4	688	7.85	802	Very high risk
S2	2.08	11.3	9.99	34.6	294	5.79	357	Very high risk
S3	2.59	9.81	7.87	11.1	205	18.6	254	Considerable risk
S4	2.22	34.8	28.7	33.5	558	16.3	673	Very high risk
S5	3.17	38	33.8	31	497	20.5	623	Very high risk
S6	3.82	5.88	15.5	28.1	213	12.7	279	Very high risk
S7	3.09	7.44	9.81	21.6	122	18	181	Considerable risk
S8	2.12	7.86	31.4	35.3	438	22.1	537	Very high risk
S9	1.29	6.61	7.63	29.9	241	18.5	305	Very high risk
S10	2.75	6.74	13.4	47.4	311	9.19	391	Very high risk

moderately contaminated; $0 \leq I_{geo} \leq 1$ – uncontaminated to moderately contaminated; and $I_{geo} \leq 0$ – practically uncontaminated.

Pollution load index (PLI): Pollution load index can be determined for six toxic metals like Cr, Ni, Cu, As, Cd, and Pb. Pollution load index is the result of total toxicity level of hazardous metals in soil. Pollution load index was measured by using the following formula:

$$PLI = (CF1 \times CF2 \times CF3 \times \dots \times CFn)^{1/n} \tag{4}$$

Potential ecological risk (PER): The degrees of hazardous elements contamination in agricultural soils are determined by PER index. The equations which were used to calculate PER proposed by Guo and are as follows [27]:

$$C_j^i = \frac{C_j}{C_n^i}, \quad C_d = \sum_{i=1}^n C_j^i \tag{5}$$

$$E_r^i = T_r^i \times C_j^i, \quad PER = \sum_{i=1}^m E_r^i \tag{6}$$

where, C_j^i is contamination factor of individual metal, C_j is element content in soils samples and C_n^i is metal baseline values. The baseline value of Cr, Ni, Cu, As, Cd, and Pb in soil samples were 45, 39, 33, 9.5, 0.95 and 27 mg/kg respectively. The integration of C_j^i for total

elements represents the overall degree of pollution (C_d). E_r^i Represent PER index and T_r^i is the biological toxic factor of single metal.

Toxic unit analysis

The calculation of toxic units is considered as severe toxicity of toxic metals in agricultural soils. Toxic unit analysis is stated as the ratio of the assessed concentration of hazardous elements in soil to probable effect level (PELs) [28]. A moderate to serious toxicity of hazardous elements remain in soil when the sum of toxic units for all soil samples is more than 4 [29].

Statistical analysis

SPSS 20.0 (SPSS, USA) was used for statistical analysis for present study. The means and standard deviations of the metal concentrations in agricultural soils were calculated. To address the sources of heavy metals in soil, principal component analysis (PCA) were applied. Microsoft Excel 2013 was used for other calculations.

Results and Discussion

Physicochemical properties and metal concentrations in soil

The physicochemical properties of soil are presented in Table 1.

The studied soils pH values for different sampling sites was slightly acidic to neutral excluding the S3, S4, S5, S6, and S9 sites that were alkaline (Table 1) which was due to the decomposition of organic matter and subsequent formation of carbonic acid [7]. The highest mean values of soil pH 7.77 and 7.45 were observed in S5 and S6 sites and lowest mean values of soil pH 6 and 6.1 were observed in S7 and S2 sites. The highest mean value of soil EC 1.07dS/m was observed in S6 site and lowest mean value of soil EC 0.08dS/m was observed in S1 site. According to SRDI soil salinity class, electrical conductivity (EC) value of the soil was non-saline (0-2dS/m) for all sampling sites which mean the salinity effect is negligible. The highest mean value of organic carbon (%C) 4.88 was observed in soil collected from the S1 site and lowest value 0.31 observed in S4 site. The organic matter content in Bangladesh soil is 0.7-1% and the organic matter status is low in Bangladesh soils. In this study, it was obtained that the range of organic matter content was very high than Bangladesh soils. There were some reasons behind the increase of organic matter. Firstly, the maximum samples were collected from boro rice field soils and these fields were remained as alternate wetting and drying. For that, there was present less amount of aerobic microorganism which was responsible for matter decomposition. Secondly, in the industrial area soils presence of heavy metals in soils. The highest percentage of mean values of sand, silt and clay 63.9, 45.4 and 20.9 were observed in S3, S1 and S10 sites. The studied soil samples were loam and sandy loam (Table 1) according to the United States soil texture classification system.

The heavy metals concentrations (Cr, Ni, Cu, As, Cd and Pb) in soil samples were presented in Table 2 and 3. The mean concentrations of Cr, Ni, Cu, As, Cd and Pb in different soil sampling sites were found 8.31, 16.49, 20.64, 5.06, 2.20 and 16.9 mg/kg, respectively (Table 3) around the industrial vicinity of Tangail district, Bangladesh. The maximum value of Cr, Ni, Cu, As, Cd and Pb were observed in soil collected from the S3, S10, S2 and S6 sites. The levels of heavy metals were varied among the sampling sites and followed the descending order of S3 > S2 > S6 > S5 > S10 > S4 > S9 > S1 > S7 > S8. Heavy metals in different soils sampling sites were compared with the other studies in Bangladesh and other countries. Ni, Cu, As and Pb concentrations of the present study were higher than those of the study conducted in Bangladesh and China (Table 3). The mean concentrations of Cd were above the Dutch soil quality standard, Canadian Environment Quality Guidelines and background value in Tangail district. The Dutch soil quality standard is considered appropriate protocol assuming total probable exposure pathways for saving plants, humans, and animals [7]. The soil is considered clean, when any heavy metal concentration does not exceed respective Dutch Target Value. The soil is regarded to be slightly to moderately contaminated, when heavy metal concentration is in between intervention values and target values. According to Table 3, Cd was in the worst situation among the studied metals as the mean concentration of Cd was higher than the Dutch Target Value. The highest concentration of Cr was found in S4 sampling sites followed by S5 where tannery industries were situated in the studied area which can be resulted from the dispose and application of untreated tannery waste (chromate smelters) to agricultural fields and generally chromium salt use in tannery industries [30]. There were also variations for Ni concentrations in soils of BSCIC areas of Tangail district and S3 sampling sites contained highest Ni concentration due to localize additions or accidental spillages of highly concentrated materials that contain Ni. Elevated level of Cu was found in S3 sampling site followed by S5 which might

be resulted from emission from the electric waste burning and metal workshop activities [30]. Arsenic concentration in the BSCIC areas shows differences and considerable As concentrations were found in S10, S6, S2, S3 sampling sites (Table 1). Generally effluent amount of underground water with As contaminated were used for irrigation purposes in the studied area agricultural soils [31]. Again, waste from brick fields and incineration activities might contribute to the high concentration of As in the study areas. In smelters surrounding areas of several countries, there occur severe Cd pollution in soil. Due to concentration Cd in soil, about 80% studied soil samples surpassed the Dutch target value assuming that Cd in soil might pose a severe risk to the surrounding ecosystems. In this study, it was observed that some heavy metals displayed higher standard deviation, and such deviation may be indicative of the lack of consistency of the elemental distribution across the sites [21,32].

Source analysis of heavy metal in soil

Source of toxic elements in soils were assessed in the form of principal component analysis (PCA) in different soil sampling sites of Tangail district. Principal component analysis is determined for sources identification [33]. There prevalent three principal components (Table 5 and Figure 2) due to analysis of source for toxic elements. Total variation was computed for 73.41% of source analysis. First principal component (PC1) described the largest variance (33.48%); second principal component (PC2) which expound 21.43% of the variance. Third principal component (PC3), described 18.49% of the total variance.

Toxic unit analysis

Sum of toxic units (Σ TUs) determine as possible heavy metal toxicity in soils. Toxic units may be calculated as the ratio of heavy metal concentration in soil which is measured to probable effect levels (PELs) [4]. Total toxic units (Σ TUs) with toxic units (TU) due to heavy metals toxicity in several soil sampling locations in industrial vicinity were presented in Figure 3. The sum of toxic units for the studied metals for the sites S2, S3, S6, and S10 was higher than the other sites, which were in the similar trends of metal concentrations in soils. Moderate to serious toxicity of hazardous materials were resulted when sum of toxic units of studied soil samples exceeded 4 and it caused serious threat to environment. In the present study, no samples were found which sum of toxic units was higher than 4.

Ecological risk assessment

In this study, the enrichment factor, contamination factor, geoaccumulation index, and pollution load index (PLI) were used to determine toxic metal pollution in industrial vicinity soils. The enrichment factor values for different soil sampling sites are presented in Figure 4. For enrichment factors, Cd and Cu have the highest enrichment factor value which indicate soil contamination for total sampling locations As a whole, the enrichment factor of all the studied metals for all sampling sites were in the descending order of Cd>Cu>As>Pb>Cr>Ni. Usually, a little enrichment values cause high contribution for crusted source in soils which were identified by several studies where anthropogenic sources have substantial contribution causes high enrichment factors [4,15]. Hakanson, (1980) delineates four types of contamination factors (CF), four types of degree of contamination (Cd), five types of E_r^i and four types of PER [34], as presented in Table 7. The contamination factor for individual metal was presented in Figure 5. In the studied area, contamination factor was low for Cr and Ni, considerable for As and Pb, and higher degree for Cu and Cd. I_{geo} values of the present study

Table 7: Indices and grades of potential ecological risk of heavy metal pollution [6].

Contamination factor (C_i^j)	Contamination degree of individual metal	Degree of contamination (Cd)	Contamination degree of the environment	E_i^j	Grade of ecological risk of individual metal	Risk index (PER)	
$C_i^j < 1$	Low	$Cd < 5$	Low contamination	$E_i^j < 40$	Low risk	$RI < 65$	Low risk
$1 \leq C_i^j < 3$	Moderate	$5 \leq Cd < 10$	Moderate contamination	$40 \leq E_i^j < 80$	Moderate risk	$65 \leq RI < 130$	Moderate risk
$3 \leq C_i^j < 6$	Considerable	$10 \leq Cd < 20$	Considerable contamination	$80 \leq E_i^j < 160$	Considerable risk	$130 \leq RI < 260$	Considerable risk
$C_i^j \geq 6$	High	$Cd \geq 20$	High contamination	$160 \leq E_i^j < 320$	High risk	$RI \geq 260$	Very high risk
				$E_i^j \geq 320$	Very high risk		

were shown in Figure 6. For all heavy metals in the studied samples for different sampling sites, the I_{geo} values presented the decreasing order of $Cd > Pb > Cu > As > Ni > Cr$. The mean of I_{geo} values for all the studied metals for all sampling sites indicating the soils were slowly contaminated with heavy metals. Pollution load index (PLI) value is zero means accurate; PLI value is one means there only present baseline level of contaminants where PLI values above 1 means successive contamination by heavy metals in soils [2,7,15]. As per above grade, studied soils were highly contaminated by Cd and it was observed that pollution load index (PLI) values of all others heavy metals for all sampling sites were not more than one (Figure 7).

PER index of single metal (E_i^j) with combining potential ecological risk index of the environment (PER) (Table 6) with classifications of PER (Table 7), studied area soil samples indicate the considerable to very high risk which must possess ecological hazard in the studied area. The order of E_i^j in soils was in the following descending order of $Cd > As > Cu > Pb > Ni > Cr$. On the whole, the range of PER for studied area soils is 182–802, indicating considerable to very high potential ecological risk. The maximum value of PER (802 in soil at S1 in Table 6) denotes a very high potential ecological risk of soil. Among the sampling sites, about 80% soil samples possess very high potential ecological risk in the studied area.

Conclusions

This study revealed that soil samples from different sites were polluted by heavy metals. Concentrations of Cd in some sampling sites exceeded background values, as well as the Dutch and Canadian quality guidelines values, indicating that the studied soils were highly polluted by Cd. The contamination factor, enrichment factor, and pollution load index of Cd was higher than other metals in the studied areas. It was also observed from the study that heavy metal concentration in industrial area soils for Bangladesh varied with different sampling sites. Heavy metals in soil for different sampling sites showed considerable to very high degree of contamination. For individual heavy metal, Cd had severe ecological risk for most of the sites, whereas, the study area comprises high potential ecological risk according to the ecological risk indexes of heavy metals. There is urgent need to study again in present studied areasto find out the causes for the higher potential ecological risk caused mainly by Cd and to increase public awareness not to throw industrial wastages in the open environment.

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