

Original Research Paper

Seasonal Variation of Heavy Metal Concentrations in Farm Soils of Sreepur Industrial Area of Gazipur, Bangladesh: Pollution Level Assessment

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Abstract: A study was conducted to measure the seasonal variation of different heavy metal contents in farm soils of Sreepur Upazila of Gazipur district and to assess their pollution level. Industrial affected and non-affected soil samples were collected from 5 locations in three seasons viz. pre-monsoon (April), monsoon (August), and dry season (January), thus a total of 30 soil samples were analyzed for this study. Concentrations of different heavy metals (Cd, Pb, Cr, Ni, Cu, and Zn) in soil samples were determined by an Atomic Absorption Spectrophotometer (AAS). The mean total concentration of Cd, Pb, Cr, Ni, Cu and Zn in industrial affected soils were trace, 12.05, 101.10, 51.32, 20.79 and 55.40 mg kg⁻¹, respectively in the pre-monsoon, trace, 28.54, 40.96, 22.70, 2.46 and 7.72 mg kg⁻¹, respectively in the monsoon and 0.27, 10.49, 39.45, 20.69, 1.85 and 4.11 mg kg⁻¹, respectively in the dry season. All heavy metal concentrations in industrial affected soil samples were higher than the corresponding non-affected sites. The study results revealed that Cd contents in the dry season and Pb contents in the monsoon season for industrial affected soils were higher compared to the average earth's crust abundance value. The geoaccumulation index (I_{geo}) values for Pb of industrial affected soils ranged from 0.30-0.85 in the monsoon that means I_{geo} class 1, indicating unpolluted/moderately polluted soil quality. Furthermore, the calculated I_{geo} value for one industrial affected location revealed moderate soil pollution by Cr in the pre-monsoon season and unpolluted/moderately polluted soil quality by Ni and Cd in the pre-monsoon and the dry season, respectively. The measured pollution load index (PLI) values were also higher in all industrially polluted sites than corresponding non-polluted sites in three seasons, which indicates pollution load due to anthropogenic sources.

Keywords: Soil Pollution, Industrialization, Geoaccumulation Index, PLI

Introduction

The world has been technologically improved as a result of industrialization. It helps to create the employment opportunity and eventually leads to eliminate the poverty of a country and build a better quality of life. Despite having various positive effects of industrialization, if it does not grow in a systematic and regulated way it eventually produces many environmental and social hazards in a society (Zakir *et al.*, 2016; Hossain *et al.*, 2017; Patnaik, 2018). It is well known that in the last two decades, the Asian region has

undergone a rapid transition from a traditional agricultural-based economy to an increasingly industrial and technology-based economy. Bangladesh has also followed the same strategy. A large number of different types of industries have developed all over the country. Simultaneously, a huge number of polluting industries also thrived, particularly in Dhaka, Chattogram, Khulna, and Gazipur areas, either ignoring the regulatory norms by the entrepreneurs or absence of strict enforcement activities of the relevant agencies. But the fact is that at the early stage of industrialization in Bangladesh the issue of environmental pollution was

not well familiar to all stakeholders and this important matter was unattended for a long time.

It is a fact that currently most of the rivers and canals around the big cities like Dhaka, Chattogram, Khulna, and Gazipur are already polluted due to absence or irregular operations of the effluent treatment facility. As a result, both industrial and domestic untreated sewage is being disposed into the wetlands and natural streams and rivers, causing a serious environmental problem and bringing pollution of rivers, streams, and wetlands (Hossain *et al.*, 2015; Zakir *et al.*, 2015a; 2017a; 2017b; Yesmeen *et al.*, 2018). When this polluted water comes in contact with the soil through irrigation or surface runoff, quality of the soil also deteriorated and crops grown in this soil affects the quality of food and eventually, it comes into the food chain (Aysha *et al.*, 2017; Haque *et al.*, 2018; Zakir *et al.*, 2018).

A couple of years back the Sreepur area of Gazipur district was famous for its diverse scenic view. It had a pristine natural condition with a multi-type eco-system with rich biodiversity. Rivers and canals were full of clean water. People used to catch fish from the river and canals and crop production was quite satisfactory. But unfortunately, now green fields became squeezed, different types of industries have been flourishing, acres of farmlands kept idle inside the boundary wall or fencing having erection of signboard of a company or industry indicating the land will no longer be used for agriculture or any other farming purposes. A survey estimated a total of 120 medium to large industries in this area, among those 52 were in the red category, 53 were in the orange-B category, 13 were in the orange-A and only 2 were in the green category. Furthermore, among the surveyed industries about 33% didn't have any Effluent Treatment Plant (ETP) (Hossain *et al.*, 2019). Thus, clean water containing canals being converted to the carrier of dark, filthy and foul smelled den. Canals became narrowed down and the polluted water is spreading over the adjacent lands especially when overflow the banks during heavy rain in the rainy season. Moreover, the farmers in most places of the study area use the polluted canal water in their land for irrigation. In such a situation, industrial effluent is a serious concern for all environmental compartments along with the farm soils of the study area. Considering the fact stated above this work was conducted to measure the seasonal variation of different heavy metal concentrations in farm soils of the Sreepur industrial area of Gazipur district and to assess their pollution level.

Materials and Methods

Soil Sampling

The surface soil (0-15 cm depth) samples were collected in three seasons [pre-monsoon (April), monsoon (August), and dry (January)] both from industrially affected and non-affected sites. Ten (10) samples (5 from

industrially affected and the other 5 from non-affected sites) were collected from each season, thus a total of 30 soil samples were analyzed for this study. For obtaining a representative sample, soils were collected from a number of points and mixed together to make a composite sample. Both sites were selected based on visual observation and local people's opinions during the field visits. Care was taken so that both the sites of the same area fall under the same soil series. Care has also been taken during selecting the sites seeing the drainage pattern and irrigation practice and the possibility of entering the wastewater into both land areas. The sampling location along with soil series are shown in Fig. 1.

Preparation of Soil Samples for Analysis

After collection, each composite soil sample was placed in a thin layer on a clean piece of brown paper on a shelf in the laboratory and left until it was air-dried. Visible garbage, stones, fragments of weeds and roots, etc. were removed from the soil samples and discarded. Then the samples were ground and subsequently sieved by using a 2 mm stainless steel sieve. Each sample was then kept in a separate clean polythene bag with an appropriate marking for physical and chemical analyses. The chemical analyses of soils were accomplished in the laboratory of the Soil Resource Development Institute (SRDI), Dhaka, Bangladesh.

Analytical Methods

Soil pH was determined by glass electrode benchtop pH meter (SensION™ + EC5, HACH, USA) as described by Ghosh *et al.* (1983) and the result was reported as soil pH measured in water (soil: Water ratio being 1:2.5). For the determination of total heavy metal concentration, exactly 1.00 g of powdered soil sample was digested with 10 mL of conc. nitric acid (HNO₃) in a 100 mL digestion tube and heated at 130°C temperature until the volume was reduced to about 1 mL. After cooling, 5 mL of 1% HNO₃ was added to the sample and then the solution was filtered with Whatman No. 42 filter paper (Hseu, 2004). Finally, concentrations of different heavy metals viz. Cu, Zn, Pb, Cr, Ni, and Cd in aqueous extracts were measured by an atomic absorption spectrophotometer (AAS) (Shimadzu, AA7000, Japan).

Assessment of Soil Pollution

Geoaccumulation Index (I_{geo})

The index of geoaccumulation (I_{geo}) is a quantitative measure of metal pollution, using the relationship between the concentration of the element in soil/sediment (fraction <2 μm) and the background as introduced by Muller (1969), which can be calculated by the following formula:

$$I_{geo} = \log_2(Cn / 1.5 \times Bn)$$

Where C_n is measured concentration of metal in the soil and B_n is the geochemical background for the same element. The average earth's crust value of the metal described by Taylor (1964) was used as the background value. Factor 1.5 is introduced by Muller (1969) to include possible variations of the background values that are due to lithologic variations. There are seven I_{geo} classes based on the numerical value of the index viz.

class 0 ($0 \geq I_{geo}$) unpolluted; class 1 ($0 \leq I_{geo} \leq 1$) uncontaminated/ moderately polluted; class 2 ($1 \leq I_{geo} \leq 2$) moderately polluted; class 3 ($2 \leq I_{geo} \leq 3$) moderately/strongly polluted; class 4 ($3 \leq I_{geo} \leq 4$) strongly polluted; class 5 ($4 \leq I_{geo} \leq 5$) strongly/ extremely polluted and the class 6 is an open class and comprises all values of I_{geo} higher than 5, which indicates extremely polluted soil quality (Muller, 1969).

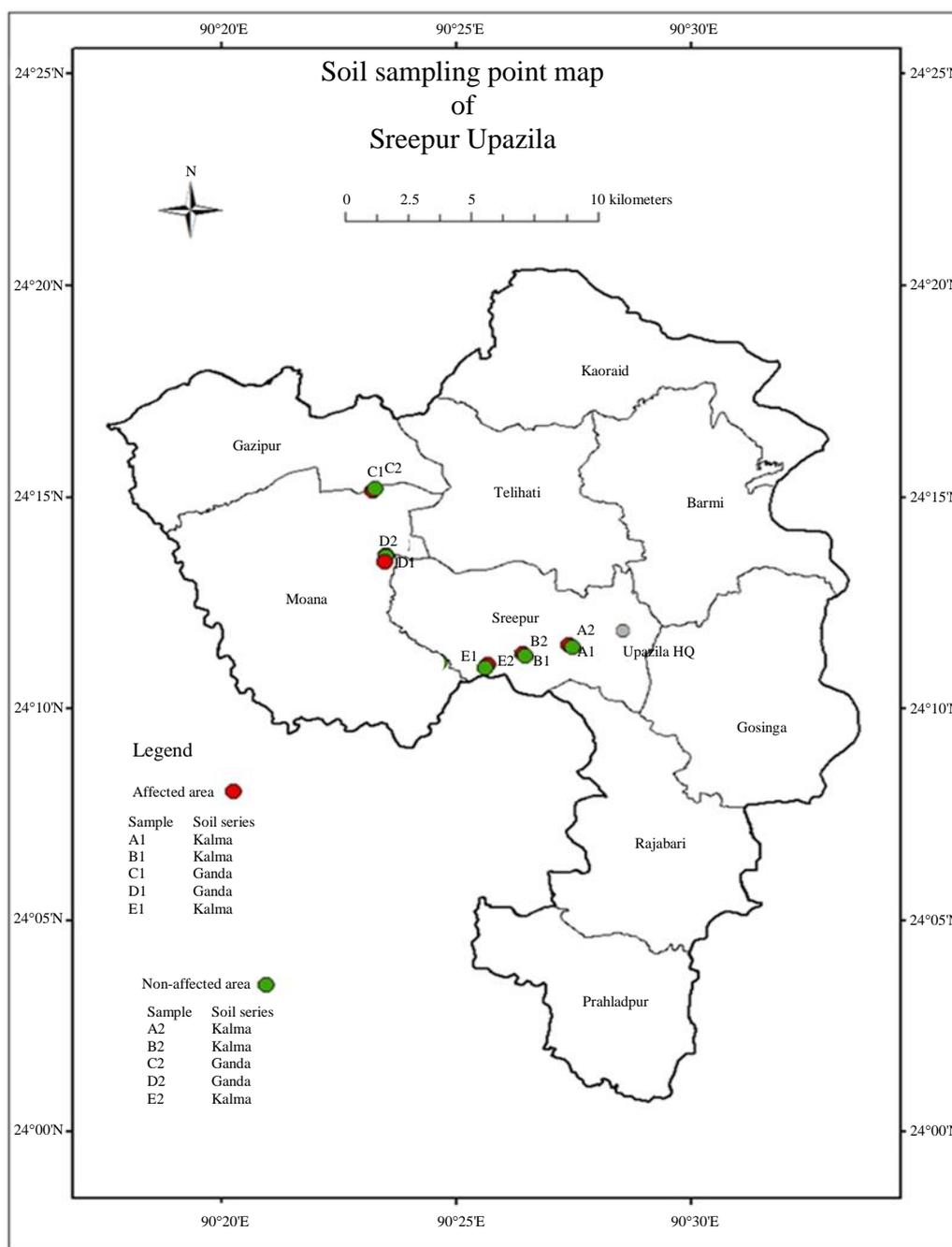


Fig. 1: Soil sampling locations (industrially affected and non-affected) along with soil series in the map of the Sreepur Upazila, Gazipur, Bangladesh

The Pollution Load Index

The Pollution load index (PLI) proposed by Tomlinson *et al.* (1980) has been calculated both for industrially affected and non-affected soils of Sreepur, Gazipur. The PLI is used to know the pollution level for a single site, which is the *n*th root of *n* number of multiplied together Contamination Factor (CF) values. The CF is measured as follows:

$$CF = C_{\text{Metal concentration}} / C_{\text{Background concentration of the same metal}}$$

and

$$PLI \text{ for a site} = \sqrt[n]{CF_1 \times CF_2 \times \dots \times CF_n},$$

where, *n* = the number of contamination factors and sites, respectively. First a number of contamination factors are derived for different metals at each site and then a site pollution index is calculated by taking the five highest contamination factors and deriving the fifth root of the five factors multiplied together (Tomlinson *et al.*, 1980).

Results and Discussion

Status of pH in Soils of the Study Area

The average pH values at the industrially affected sites were slightly acidic in all three seasons (pre-monsoon-6.26; dry season-6.22 and monsoon-6.26) and the ranges were 4.7-7.7; 5.5-7.2 and 5.3-6.9 in the pre-monsoon, dry and monsoon seasons, respectively (Table 1). On the other hand, at the non-affected sites, the average pH values were moderately acidic in the dry (5.78) and pre-monsoon seasons (5.66); but near neutral in the monsoon season (6.74) and the ranges were 5.2-6.2; 5.3-6.4 and 6.2-7.1 in the pre-monsoon, dry and monsoon seasons, respectively. The terrace and hill soils

are strongly to very strongly acidic. In Kalma series medium high land soils are strongly to slightly acidic (pH 4.5-5.9) and medium low land soils are strongly acidic (pH 4.8-5.5) (SRDI, 2000). As the soil sampling sites are within Kalma and Genda series, therefore naturally the soil reaction is acidic. Thus, the acidic character of soil agrees with the soil series characteristics of the study area. According to (SRDI, 2000) pH range, 5.6-7.3 is suitable for the better production of most of the crops.

Heavy Metal Status in Soils of the Study Area

Cadmium (Cd)

The average Cd concentrations at both the industrially affected and non-affected sites were trace (below detectable limit) in two seasons (pre-monsoon and monsoon). But the mean concentrations were 0.27 and 0.13 mg kg⁻¹ in the dry season at the industrially affected and non-affected sites, respectively (Table 2). Soils collected from three different industrial areas of Gazipur reported the mean Cd concentrations 0.41 mg kg⁻¹ with a range of 0.08 to 1.60 mg kg⁻¹ and stated that about 35% of sampling sites showed higher Cd concentration than the average shale value (Zakir *et al.*, 2015b). On the other hand, the distribution of Cd in the sediments of the Karatoa river of Bangladesh was more than a hundred times higher compared to standard continental crust (Zakir *et al.*, 2013). It is evident from Table 2 that Cd content in all industrially affected sites was higher than the corresponding non-affected sites and in most cases, Cd concentration was higher than the average earth's crust abundance value. This might be due to the deposition of Cd in soil from different types of human activities at the study area viz. industrial effluents, construction activities, fuel or battery from the near motor terminal, and others.

Table 1: Soil pH of both industrially affected and non-affected sites of the study area in different seasons

Sampling ID	Soil pH in different seasons			Mean ± SD	Range	Soil Series	Permissible limit
	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)				
AS-1	7.70	5.60	6.00	6.43±1.11	5.60-7.70	Kalma	5.60 -7.30 (SRDI, 2000)
AS-2	6.70	5.30	6.80	6.26±0.83	5.30-6.80	Kalma	
AS-3	4.70	6.60	5.50	5.60±0.95	4.70-6.60	Genda	
AS-4	5.20	6.90	5.60	5.90±0.88	5.20-6.90	Genda	
AS-5	7.00	6.90	7.20	7.03±0.15	6.90-7.20	Kalma	
Mean ± SD	6.26±1.26	6.26±0.76	6.22±0.75	-	-	-	
Range	4.70-7.70	5.30-6.90	5.50-7.20	-	-	-	
NAS-1	5.20	6.30	5.30	5.60±0.60	5.20-6.30	Kalma	
NAS-2	6.00	6.20	5.30	5.83±0.47	5.30-6.20	Kalma	
NAS-3	5.00	7.30	6.00	6.10±1.15	5.00-7.30	Genda	
NAS-4	6.20	7.10	6.40	6.56±0.47	6.20-7.10	Genda	
NAS-5	5.90	6.80	5.90	6.20±0.52	5.90-6.80	Kalma	
Mean ± SD	5.66±0.52	6.74±0.48	5.78±0.48	-	-	-	
Range	5.00-6.20	6.20-7.30	5.30-6.40	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

Table 2: Total concentration of Cd in soils of both industrially affected and non-affected sites of the study area in different seasons

Soil Cd (mg kg ⁻¹) in different seasons							
Sampling ID	Soil Cd (mg kg ⁻¹) in different seasons			Mean ± SD	Range	Soil series	Earth's Crust average
	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)				
AS-1	Trace	Trace	0.43	0.14±0.24	Trace-0.43	Kalma	0.2 mg kg ⁻¹ (Taylor, 1964)
AS-2	Trace	Trace	0.28	0.09±0.16	Trace-0.28	Kalma	
AS-3	Trace	Trace	0.28	0.09±0.16	Trace-0.25	Genda	
AS-4	Trace	Trace	0.19	0.06±0.10	Trace-0.19	Genda	
AS-5	Trace	Trace	0.18	0.06±0.10	Trace-0.18	Kalma	
Mean ± SD	-	-	0.27±0.10	-	-	-	
Range	-	-	0.18-0.43	-	-	-	
NAS-1	Trace	Trace	0.31	0.10±0.17	Trace-0.31	Kalma	
NAS-2	Trace	Trace	0.03	0.01±0.01	Trace-0.03	Kalma	
NAS-3	Trace	Trace	0.25	0.08±0.14	Trace-0.28	Genda	
NAS-4	Trace	Trace	0.03	0.01±0.01	Trace-0.03	Genda	
NAS-5	Trace	Trace	0.03	0.01±0.01	Trace-0.03	Kalma	
Mean ± SD	-	-	0.13±0.14	-	-	-	
Range	-	-	0.03-0.31	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

Table 3: Total concentration of Pb in soils of both industrially affected and non-affected sites of the study area in different seasons

Soil Pb (mg kg ⁻¹) in different seasons							
Sampling ID	Soil Pb (mg kg ⁻¹) in different seasons			Mean ± SD	Range	Soil series	Earth's Crust average
	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)				
AS-1	13.50	23.13	8.95	15.19±7.24	8.95-23.13	Kalma	12.50 mg kg ⁻¹ (Taylor, 1964)
AS-2	9.00	27.74	9.54	15.42±10.66	9.00-27.24	Kalma	
AS-3	6.00	26.20	8.35	13.51±11.04	6.00-26.20	Genda	
AS-4	22.50	33.88	5.37	20.58±14.35	5.37-33.88	Genda	
AS-5	9.25	31.83	20.23	15.99±5.90	9.25-20.23	Kalma	
Mean ± SD	12.05±6.42	28.54±4.32	10.49±5.68	-	-	-	
Range	6.00-22.50	23.13-33.88	5.37-20.23	-	-	-	
NAS-1	4.15	3.65	4.80	4.20±0.57	3.65-4.80	Kalma	
NAS-2	8.00	6.67	8.94	7.87±1.14	6.67-8.00	Kalma	
NAS-3	3.00	2.88	3.89	3.25±0.55	2.88-3.89	Genda	
NAS-4	1.25	0.96	1.12	1.11±0.14	0.96-1.25	Genda	
NAS-5	5.76	4.89	5.11	5.25±0.45	4.89-5.76	Kalma	
Mean ± SD	4.43±2.59	3.81±2.14	4.80±2.81	-	-	-	
Range	1.25-8.00	0.96-6.67	1.12-8.94	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

Lead (Pb)

The average Pb values at the industrially affected sites during the pre-monsoon, monsoon and dry seasons were 12.05, 28.54 and 10.49 mg kg⁻¹, respectively and the ranges were 6.00-22.50, 23.13-33.88 and 5.37-20.23 mg kg⁻¹, respectively. On the other hand, at the non-affected sites, the average Pb values during the pre-monsoon, monsoon and dry seasons were 4.43, 3.81 and 4.8 mg kg⁻¹, respectively and the ranges were 1.25-8.00, 0.96-6.67 and 1.12-8.94 mg kg⁻¹, respectively (Table 3). Therefore, the average Pb values in industrially affected soil sites were higher than corresponding non-affected sites in all three seasons. It is evident from Table 3 that in all industrially affected sites, Pb content was comparatively higher during the monsoon season than the other two seasons, which might be due to highly mobile characteristics of Pb containing organic

compounds in nature (Mengel *et al.*, 2001). Another reason might be the vehicular movement and construction activities. The average Pb content in canal water of this study area during the monsoon season was 0.734 mg L⁻¹, which was beyond DoE recommended irrigation water quality limit (Hossain *et al.*, 2019). Therefore, the value of Pb both in canal water and affected soil of the study area showed higher amounts of Pb in the monsoon season than the other two seasons. Lead is a common inorganic pigment and it is found in textile wastewater (Slater, 2003). Zakir *et al.* (2015b) reported that Pb concentrations in soils of different industrial areas of Gazipur ranged between 0.44 to 127.45 mg kg⁻¹ with an average value of 27.95 mg kg⁻¹.

Chromium (Cr)

Chromium concentrations at the industrially affected sites varied between 16.75-404.70, 18.24-86.13

and 34.70-51.25 mg kg⁻¹ in the pre-monsoon, monsoon and dry seasons, respectively with the mean values of 101.10, 40.96 and 39.45 mg kg⁻¹, respectively (Table 4). On the other hand, at the non-affected sites, the average Cr values during the pre-monsoon, monsoon and dry season were 20.11, 21.83 and 21.14 mg kg⁻¹, respectively and the ranges were 16.46-25.58, 16.80-26.75 and 18.83-25.69 mg kg⁻¹, respectively (Table 4). Therefore, the average Cr concentration in most soils was within the limit of earth's crust abundance (100 mg/kg) (Taylor, 1964). This result agrees with the report published earlier (Zakir *et al.*, 2015b), who stated that the level of Cr in soils collected from different industrial areas of Gazipur varied from

trace to 80.78 mg kg⁻¹. But among all the sites only one affected site (AS# 4) showed several times higher (404.70 mg kg⁻¹) Cr concentration during the pre-monsoon season, which might be due to the curvature nature and downstream point of the *Labandaha* canal. Furthermore, a wool wear factory is also close to this site, which usually uses Cr and Cu. Copper, Cr, Ni and Pb are common in inorganic pigments and these are found in textile wastewater (Slater, 2003; World Bank, 1998). Metals in textile industrial effluent are produced during the dyeing process, which usually contributes to Cr, Pb, Zn and Cu to wastewater (Deepali and Gangwar, 2009).

Table 4: Total concentration of Cr in soils of both industrially affected and non-affected sites of the study area in different seasons

Sampling ID	Soil Cr (mg kg ⁻¹) in different seasons			Mean ± SD	Range	Soil series	Earth's Crust average
	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)				
AS-1	28.34	35.01	37.49	33.61±4.73	28.34-37.49	Kalma	100.00
AS-2	34.06	32.22	36.59	34.29±2.19	32.22-36.59	Kalma	mg kg ⁻¹
AS-3	16.75	33.22	37.23	29.06±10.85	16.75-37.23	Genda	(Taylor, 1964)
AS-4	404.70	86.13	51.25	180.7±194.8	51.25-404.7	Genda	
AS-5	26.13	18.24	34.70	26.35±8.23	18.24-34.70	Kalma	
Mean ± SD	101.1±169.3	40.96±26.11	39.45±6.68	-	-	-	
Range	16.75-404.7	18.24-86.13	34.70-51.25	-	-	-	
NAS-1	25.58	26.02	20.98	24.19±2.79	20.98-26.02	Kalma	
NAS-2	22.96	26.75	25.69	25.13±5.54	22.96-26.75	Kalma	
NAS-3	16.46	22.14	19.45	19.35±2.84	16.46-22.14	Genda	
NAS-4	18.54	17.44	20.77	25.23±1.96	17.44-20.77	Genda	
NAS-5	17.01	16.80	18.83	17.54±1.11	16.80-18.83	Kalma	
Mean ± SD	20.11±3.98	21.83±4.65	21.14±2.70	-	-	-	
Range	16.46-25.58	16.80-26.75	18.83-25.69	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

Table 5: Total concentration of Ni in soils of both industrially affected and non-affected sites of the study area in different seasons

Sampling ID	Soil Ni (mg kg ⁻¹) in different seasons			Mean ± SD	Range	Soil series	Earth's Crust average
	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)				
AS-1	18.60	26.25	19.37	21.4±4.21	18.60-26.25	Kalma	75.00
AS-2	15.25	22.88	18.10	18.74±3.85	15.25-22.88	Kalma	mgkg ⁻¹
AS-3	9.00	11.96	18.01	12.99±4.59	9.00-18.01	Genda	(Taylor, 1964)
AS-4	198.25	67.28	31.52	99.01±87.78	31.52-198.25	Genda	
AS-5	15.50	7.64	16.45	13.19±4.83	7.64-16.45	Kalma	
Mea ± SD	51.32±82.21	22.70±23.67	20.69±6.14	-	-	-	
Range	9.00-198.25	7.64-67.28	16.45-31.52	-	-	-	
NAS-1	13.00	14.63	14.15	13.92±0.83	13.00-14.63	Kalma	
NAS-2	19.50	17.88	18.10	18.49±0.87	17.88-19.50	Kalma	
NAS-3	6.50	7.78	8.45	7.57±0.99	6.50-8.45	Genda	
NAS-4	9.50	7.21	8.72	8.47±1.16	7.21-9.50	Genda	
NAS-5	10.25	9.64	11.82	10.57±1.12	9.64-11.82	Kalma	
Mean ± SD	11.75±4.91	12.96±4.64	12.25±4.02	-	-	-	
Range	6.50-19.50	7.21-17.88	8.45-18.10	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

Nickel (Ni)

The average Ni contents in the soil at the industrially affected sites during the pre-monsoon, monsoon, and dry seasons were 51.32, 22.70 and 20.69 mg kg⁻¹, respectively and the ranges were 9.00-198.25, 7.64-67.28, and 16.45-31.52 mg kg⁻¹, respectively (Table 5). At the non-affected sites, the average Ni concentrations during the pre-monsoon, monsoon and dry seasons were 11.75, 12.96 and 12.25 mg kg⁻¹, respectively and the ranges were 6.50-19.50, 7.21-17.88 and 8.72-18.10 mg kg⁻¹, respectively. So, it is apparent from Table 5 that in all affected sites Ni contents were higher than corresponding non-affected sites. However, in all sites, the average Ni values were within the average earth's crust abundance limit (75 mg kg⁻¹) (Taylor, 1964) except site AS# 4. The higher concentration of Ni at this site might be due to the curvature nature of the *Labandaha* canal at this point and a wool wear factory, which uses different types of dyeing chemicals. Nickel is common in inorganic pigment and this element is found in textile wastewater (Slater, 2003; World Bank, 1998). The major sources of nickel contamination in the soil are metal plating industries, combustion of fossil fuels, and nickel mining and electroplating. It is released into the air by power plants and trash incinerators and settles to the ground after undergoing precipitation reactions (Wuana and Okieimen, 2011).

Copper (Cu)

The average Cu contents in soil at the industrially affected sites during the pre-monsoon, monsoon, and dry seasons were 20.79, 2.46 and 1.85 mg kg⁻¹, respectively while the ranges were 9.30-41.50, 0.87-5.12 and 0.58-2.66 mg kg⁻¹, respectively (Table 6). On the other hand, at the non-affected sites, the average Cu contents during the pre-monsoon, monsoon and dry seasons were 0.73, 0.65 and 0.77 mg kg⁻¹, respectively and the ranges were 0.52-0.82, 0.44-0.76 and 0.60-0.96 mg kg⁻¹, respectively. It is evident from Table 6 that Cu concentrations in all polluted sites were also higher than the corresponding non-polluted sites. According to Domingo and Kyuma (1983), the Cu status of selected Bangladesh paddy soils ranged from 6-48 mg kg⁻¹. There was quite a variation in Cu status of soils collected from 3 different industrial areas of Gazipur district and concentration of Cu in soils ranged between 5.92 to 74.76 mg kg⁻¹, having an average value of 36.19 mg kg⁻¹ (Zakir *et al.*, 2015b). Cu concentration obtained from the present study is a little bit lower than that of the reported value. However, the average Cu concentration was within the limit of earth's crust abundance (45 mg kg⁻¹) (Taylor, 1964) in all sites and all seasons. But, Cu concentration was very close to this limit only one affected site (AS# 4) in the pre-monsoon season (Table 6). Dye wastewaters are highly colored and may contain heavy

metals such as Cu and Cr. Heavy metals such as Pb, Cr, Cd and Cu are widely used for the production of colour pigments of textile dyes (Halimoon and Yin, 2010). The level of Cu in the study soils was less than the maximum acceptable concentration of 100 mg kg⁻¹ for crop production (Kabata-Pendias and Pendias, 1992).

Zinc (Zn)

The average Zn concentrations in soil at the industrially affected sites during the pre-monsoon, monsoon and dry seasons were 55.40, 7.72 and 4.11 mg kg⁻¹, respectively and the ranges were 35.25-81.75, 1.94-11.05 and 0.82-8.21 mg kg⁻¹, respectively (Table 7). On the other hand, at the non-affected sites, the average Zn contents during the pre-monsoon, monsoon and dry seasons were 2.80, 2.52 and 2.35 mg kg⁻¹, respectively and the ranges were 2.06-4.12, 1.74-3.86 and 0.94-3.96 mg kg⁻¹, respectively. Therefore, it is apparent from Table 7 that all the polluted sites contained a higher amount of Zn than the corresponding non-polluted sites. Zinc concentrations obtained from this study were within the average earth's crust abundance (70 mg kg⁻¹) (Taylor, 1964) in all sites. Similar to other heavy metals, Zn content in one polluted site (AS# 4) was also higher than this limit in the pre-monsoon season (81.75 mg kg⁻¹). The higher value of Zn in this site might be due to the dumping of poultry and municipal wastes and different construction activities. According to Kabata-Pendias and Pendias (1992), the maximum acceptable concentration of Zn is 150 mg kg⁻¹ for crop production and the level of Zn in the study soils was far below than of this limit. The high Zn concentration in soil could be attributed to the use of Zn during different industrial activities, such as coal and waste combustion, steel processing and others. Furthermore, agricultural use of sewage sludge and the use of agro-chemicals such as fertilizers and pesticides might also contribute to Zn enrichment in soil. Large concentrations of Zn in the soil have adverse effects on crops, livestock and humans (Parth *et al.*, 2011).

Assessment of Heavy Metal Pollution Level in Soils

Geoaccumulation Index (I_{geo})

The geo-accumulation index (I_{geo}) is a common criterion used for quantifying the intensity of heavy metal contamination in terrestrial, aquatic and marine environments, which has been used by many researchers in order to determine the extent of the metal accumulation in the soil and sediments (Gaur *et al.*, 2005; Zakir *et al.*, 2015b; 2017c; Hossain and Zakir, 2016; Islam *et al.*, 2020). The geo-accumulation index (I_{geo}) introduced by Muller (1969) was used to assess heavy metal pollution in different industrial soils of Sreepur Upazila of Gazipur district. The calculated I_{geo} values and their corresponding contamination levels of different metals in soils are given in Fig. 2 for both soils.

Table 6: Total concentration of Cu in soils of both industrially affected and non-affected sites of the study area in different seasons

Soil Cu (mg kg ⁻¹) in different seasons							
Sampling ID	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)	Mean ± SD	Range	Soil Series	Earth's Crust average
AS-1	14.50	1.68	1.42	5.87±7.48	1.42-14.50	Kalma	45.00 mgkg ⁻¹ (Taylor, 1964)
AS-2	18.89	2.49	2.14	7.84±9.57	2.14-18.89	Kalma	
AS-3	9.30	5.12	0.58	5.00±4.36	0.58-9.30	Genda	
AS-4	41.50	2.14	2.47	15.37±22.63	2.14-41.50	Genda	
AS-5	19.75	0.87	2.66	7.76±10.42	0.87-19.75	Kalma	
Mean ± SD	20.79±12.30	2.46±1.60	1.85±0.85	-	-	-	
Range	9.30-41.50	0.87-5.12	0.58-2.66	-	-	-	
NAS-1	0.82	0.76	0.89	3.82±5.19	0.76-0.89	Kalma	
NAS-2	0.75	0.65	0.78	3.59±5.21	0.65-0.78	Kalma	
NAS-3	0.75	0.68	0.60	1.87±2.13	0.60-0.75	Genda	
NAS-4	0.52	0.44	0.61	1.51±1.94	0.44-0.61	Genda	
NAS-5	0.82	0.72	0.96	3.56±4.71	0.72-9.60	Kalma	
Mean ± SD	0.73±0.12	0.65±0.12	0.77±0.16	-	-	-	
Range	0.52-0.82	0.44-0.76	0.60-0.96	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

Table 7: Total concentration of Zn in soils of both industrially affected and non-affected sites of the study area in different seasons

Soil Zn (mg kg ⁻¹) in different seasons							
Sampling ID	Pre-monsoon (Mar.-May)	Monsoon (Jun.-Oct.)	Dry (Nov.-Feb.)	Mean ± SD	Range	Soil Series	Earth's Crust average
AS-1	35.25	1.94	0.82	12.67±19.56	0.82-35.25	Kalma	70.00 mgkg ⁻¹ (Taylor, 1964)
AS-2	56.25	10.66	2.56	23.15±28.94	2.56-56.25	Kalma	
AS-3	32.75	11.05	1.02	14.94±16.21	1.02-32.75	Genda	
AS-4	81.75	9.87	8.21	32.27±41.98	8.21-81.75	Genda	
AS-5	71.00	5.08	7.96	28.01±37.25	5.08-71.00	Kalma	
Mean ± SD	55.40±21.55	7.72±4.02	4.11±3.69	-	-	-	
Range	35.25-81.75	1.94-11.05	0.82-8.21	-	-	-	
NAS-1	2.06	1.74	0.94	10.22±15.39	0.94-2.06	Kalma	
NAS-2	3.31	3.15	2.88	13.32±19.23	2.88-3.31	Kalma	
NAS-3	2.25	1.95	2.16	6.86±9.87	1.95-2.25	Genda	
NAS-4	2.25	1.92	1.83	6.11±8.78	1.83-2.25	Genda	
NAS-5	4.12	3.86	3.96	18.25±24.84	3.86-4.12	Kalma	
Mean ± SD	2.80±0.89	2.52±0.93	2.35±1.13	-	-	-	
Range	2.06-4.12	1.74-3.86	0.94-3.96	-	-	-	

AS = Affected Soil; NAS = Non-Affected Soil

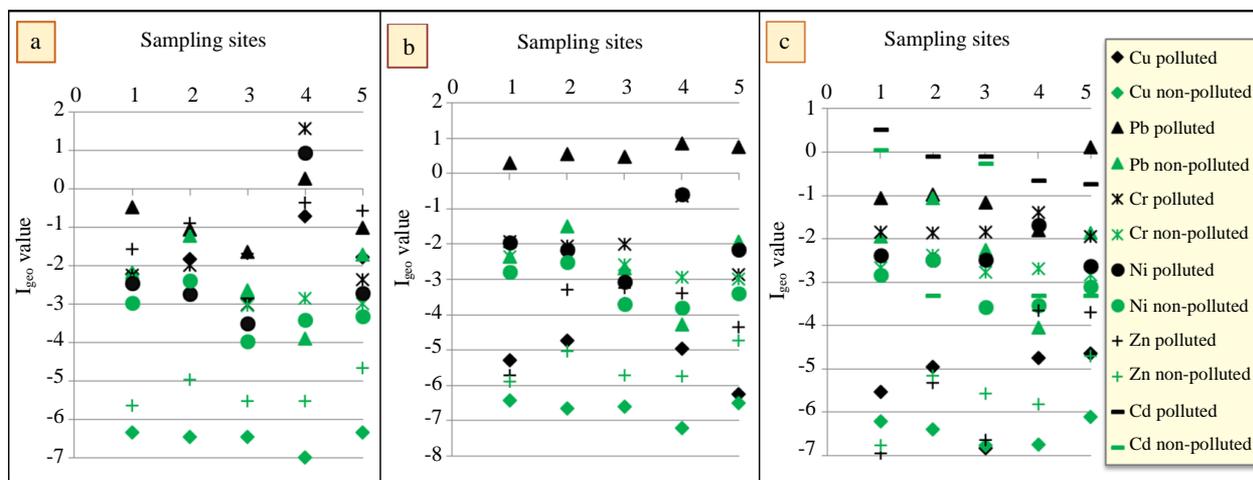


Fig. 2: Geoaccumulation index (I_{geo}) values for soils collected from different industrial areas of Sreepur, Gazipur during pre-monsoon (a), monsoon (b) and dry (c) seasons

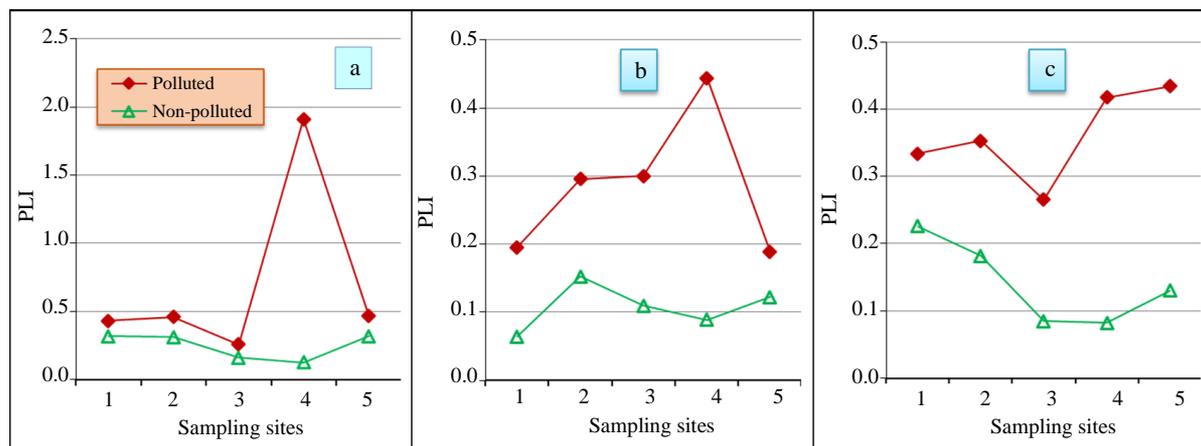


Fig. 3: Pollution load index (PLI) values for soils collected from different industrial areas of Sreepur, Gazipur during pre-monsoon (a), monsoon (b) and dry (c) seasons

The I_{geo} values for Cu and Zn exhibited zero class in all five locations in all three seasons, indicating unpolluted soil quality with respect to these two metals. Considering the values of Pb, all locations of industrially affected soil exhibited positive values within the range of $0 < I_{geo} < 1$, that means I_{geo} class-1 in monsoon, indicating unpolluted/ moderately polluted soil quality by Pb. Similarly, one site in the pre-monsoon and dry season also exhibited positive values within the same range that means I_{geo} class-1 indicating unpolluted/ moderately polluted soil quality by Pb.

In the case of Cr and Ni for industrially affected soils, one site (AS# 4) showed positive values within the range of $1 < I_{geo} < 2$, which indicating moderately polluted soil quality by these metals during the pre-monsoon time, but all other seasons the exhibited I_{geo} classes were zero. In the dry season, Cd showed positive I_{geo} value for one site (AS# 1) within the range of $0 < I_{geo} < 1$, that means I_{geo} class-1, indicating unpolluted/moderately polluted soil quality by Cd (Fig. 2). Finally, the I_{geo} calculations of soils of one location of Sreepur Upazila of Gazipur district revealed moderate pollution level in soils by Cr and unpolluted/moderately polluted soil quality by Pb, Ni and Cd from different anthropogenic sources preferably different industrial activities at the study area. On the other hand, the geo-accumulation index (I_{geo}) for non-polluted sites showed negative values in all sites and all seasons except for Cd in one site (NAS# 1) at the dry season. According to Zakir *et al.* (2015b), the I_{geo} calculations of soils of three different industrial areas of Gazipur district revealed moderate pollution levels in soils by Pb, Zn and Cd from different anthropogenic sources preferably different industrial activities, which is almost at par with the present study.

Pollution Load Index (PLI)

The PLI has been used in this study to measure PLI of farm soils of five locations of Sreepur Upazila of

Gazipur district at different seasons. The concept of a baseline is a fundamental issue to the formation of a PLI (Tomlinson *et al.*, 1980). The PLI represents the number of times by which the metal content in the soil exceeds the average natural background concentration and gives a summative indication of the overall level of heavy metal toxicity in a particular sample that has been widely used (Zakir *et al.*, 2015b; 2017c; Hossain *et al.*, 2017; Zakir and Arafat, 2020). The PLI values ranged from 0.26-1.91 and 0.12-0.32 for polluted and non-polluted soil sampling sites, respectively during the pre-monsoon season (Fig. 3a). During the monsoon season, the PLI values varied from 0.19-0.44 and 0.06-0.15 for polluted and non-polluted soil sampling sites, respectively (Fig. 3b), while in the dry season the PLI values ranged from 0.27-0.43 and 0.08-0.23 for polluted and non-polluted soil sampling sites, respectively (Fig. 3c).

The index as presented provides a simple, comparative means for assessing a site quality: A value of zero indicates perfection, a value of one that only baseline levels of pollutants are present and values above one would indicate progressive deterioration of the site quality (Tomlinson *et al.*, 1980). Out of 5 locations, only one location in pre-monsoon season had value >1 indicates pollution load in the site (AS# 4), but all other sites had value <1 , which indicates pollution load in those sites are not alarming yet. But it is apparent from Fig. 3 that the PLI values were higher in all polluted sites than the corresponding non-polluted sites in all three seasons, which indicates pollution load due to anthropogenic sources. Moreover, the sites are polluted with the particular toxic metal element, especially in site AS# 4 is more polluted than others. Similar to I_{geo} , the contamination factor for Pb, Cr, and Ni were several times higher compared to Cu, Zn, and Cd, which indicates that Pb, Cr, and Ni were the major pollutants in soils of different industrial areas of Sreepur Upazila in

Gazipur district giving rise to PLI values for the study area. So, to protect ecology and the environment as a whole, the present status should not let continue that may get alarming in the near future.

Conclusion

This study was conducted to measure the seasonal variation of different heavy metal contents and to assess pollution levels in industrial affected and non-affected farm soils of Sreepur Upazila of Gazipur district, Bangladesh. All heavy metal concentrations in industrial affected soil samples were higher than the corresponding non-affected sites. The measured I_{geo} and PLI values were also higher in all industrially polluted sites than the corresponding non-polluted sites in three seasons, which indicates pollution load due to anthropogenic sources. Finally, the study results inferred that the soil quality in the industrial affected areas is deteriorating progressively throughout the year in respect of different heavy metal contents. Therefore, the following points should be taken into consideration for future research and for the agencies responsible to monitor the area as well:

- (a) Assessment of heavy metal contamination levels in the water, soil, and sediment samples should be carried out at regular intervals and study the relationships among the compartments
- (b) In Bangladesh, heavy metals containing chemicals are used in the textile, dyeing, washing, and printing industries although there was no clear report about the composition of those chemicals. Therefore, the heavy metal-containing specific chemical should be taken into consideration so that it could be ascertained about the metal(s) presents in those chemicals
- (c) Finally, different industries have been using different types of ETP, but their capacity and capability regarding removing different toxic substances including heavy metals is a burning question. Therefore, the inclusive technological study is necessary for the capacity and performance of ETP's.

Author's Contributions

Md. Billal Hossain: Contributed to the design and implementation of the experiments, data processing and analysis of the results. Wrote the first draft of the manuscript.

Md. Nurul Islam: Contributed to the design and implementation of the experiments. Supervised the whole study.

Mohammad Shamsul Alam: Contributed to the design and implementation of the experiments. Co-supervised the whole study.

Md. Zakir Hossen: Made considerable contributions in the interpretation of obtained data. Helped to prepare the final draft of the manuscript.

Disclaimer

Authors have declared that no competing interests exist. The products used for this research are commonly and predominantly use products in our area of research and country. This article is original and contains unpublished material. The corresponding author confirms that all of the other authors have read and approved the manuscript and no ethical issues involved.

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