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Geospatial Visualization and Ecological Risk Assessment of Heavy Metals in Rice Soil of a Newly Developed Industrial Zone in Bangladesh

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Abstract: With rapid industrialization in Gazipur areas of Bangladesh, untreated industrial effluents have been polluting rice soils which could exert potential ecological risk. Therefore, four different types of industries including chemical (SL), textile and paints (MIX), dyeing (CK), and sweater and dyeing (RD) were selected to monitor the intensity of heavy metal pollution in rice soils and ecological risk assessment. The di-acid digestion method was used for the determination of Pd, Cd, and Ni, and the DTPA extraction method was used for Fe, Zn, and Cu. ArcGIS was used to visualize the spatial patterns of heavy metal pollution, and different pollution indices were calculated to assess the ecological risk. The highest concentration (mg kg^{-1}) of Cd (0.72), Pb (104.20), and Ni (5.02) was found in soils of the MIX industrial area. The highest concentration (mg kg^{-1}) of Fe (147.65) and Zn (11.27) was found in the SL industry, while the highest Cu (7.67) was found in the CK industry. It was evident from the spatial distribution that the soils of paddy fields adjacent to the different industries are more contaminated than background soil. Although the potential ecological risk of heavy metal was low, different pollution indices indicated low to high pollution. Thus, the adjacent rice field soil of different industries is being contaminated by different heavy metals which may raise ecological risk.

Keywords: paddy soil; contaminants; anthropogenic; pedogenic; di-acid digestion; DTPA extraction; pollution index; spatial distribution

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1. Introduction

In recent years, much care has been paid to environmental pollution by means of rapid urbanization and industrialization throughout the world, and Bangladesh is part of this [1,2]. Owing to industrialization, the most serious environmental problem is the pollution of agricultural soil. A huge amount of wastewater generated from the industries has been discharged into the environment without appropriate treatments [3,4]. The spreading of these untreated heavy metals and contaminated effluents used for irrigation purposes could contaminate agricultural soils. Thus, heavy metal accumulation in agricultural soils has received serious attention due to potential health risks and their adverse effects on soil health, toxicity and persistence in the environment, and food safety issues as food chain contaminant through plant uptake [5–9].

Anthropogenic activities such as mining, transportation, waste disposal, industrialization, and social and agricultural activities have significant impact on ecological pollution and the total ecosystem [10,11]. Different factors like climate, parent materials, mineralogy, and soil texture and type are involved in distributing heavy metals in nature [12,13]. However, various sources of untreated industrial wastewater are the main anthropogenic origin of heavy metals in the soil of Bangladesh [14–16]. Irrigation with different sources of contaminated water such as industrial, municipal, and sewage sludge wastewater and the dumping of solid waste directly into farmland result in the spreading of untreated effluent throughout the farmland, and plants are exposed to poisonous metal ponds without any treatment [17–19].

Many industries have been identified in the country by the Department of Environmental Promotion (DoE) which do not have sewage and waste treatment facilities [20,21]. This highly toxic wastewater discharges directly into the adjacent soil and rivers and includes lead (Pb), cadmium (Cd), nickel (Ni), iron (Fe), zinc (Zn), and copper (Cu), which are mainly responsible for environmental pollution [1,2,4,22]. In addition, floods can cause arable land to be inundated with industrial wastewater through the discharge of untreated industrial wastewater [23]. Therefore, the crops grown in the vicinity of the industrial zone contain higher concentrations of heavy metals and enter into the edible and non-edible parts. The high quantities of heavy metal-containing food may cause toxicity for all living beings including human beings because the appropriate mechanisms are not available to eliminate them. In many cases, they are considered as contaminated bio-monitor loads [24].

The history of industrialization in Bangladesh is very brief, but the impact of contamination from untreated wastewater from dyeing, textiles, and pharmaceuticals is recognized in industrial zones [7,19,25]. Some sporadic research on heavy metal pollution has been conducted [1,2,15,25], but no systematic studies have been attempted that are urgently needed to know the causes and sources of different heavy metal loads in the environment of the newly developed industries. As rice is the principal cereal in Bangladesh, heavy metal accumulation in paddy soils is the greatest concern for human and animal health. However, a methodical study for quantifying heavy metal pollution in agricultural soils and their sources has not yet been conducted in Bangladesh. Therefore, this study aimed (1) to investigate the contamination level of heavy metals in the command areas of paddy field soil adjacent to an industrial zone, (2) to identify the sources of heavy metals using a spatial distribution map, and (3) to evaluate the ecological risk of heavy metals in paddy soil using chemometric approaches. The outcomes of this study will provide significant information on the impacts of urban activities of the newly developed industrial zone of Bangladesh and safety of soil ecology, and furthermore, it could provide a reference guideline to conduct studies across other urban regions of Bangladesh.

2. Materials and Methods

2.1. Study Area

This study covered four different types of industries and associated agricultural sites. They were the chemical industry (SL), textile and paints industry (MIX), dyeing industry (CK), and sweater and dyeing industry (RD). They are situated at South Donua of Sreepur, Jangaliapara of Gazipur Sadar, and Kewa and Nayanpur of Sreepur in the Gazipur District of Bangladesh located between 24.095° and 24.309°N and between 90.373° and 90.388°E (Figure 1). The age of the industries ranges between 15 and 20 years (Figure 2).

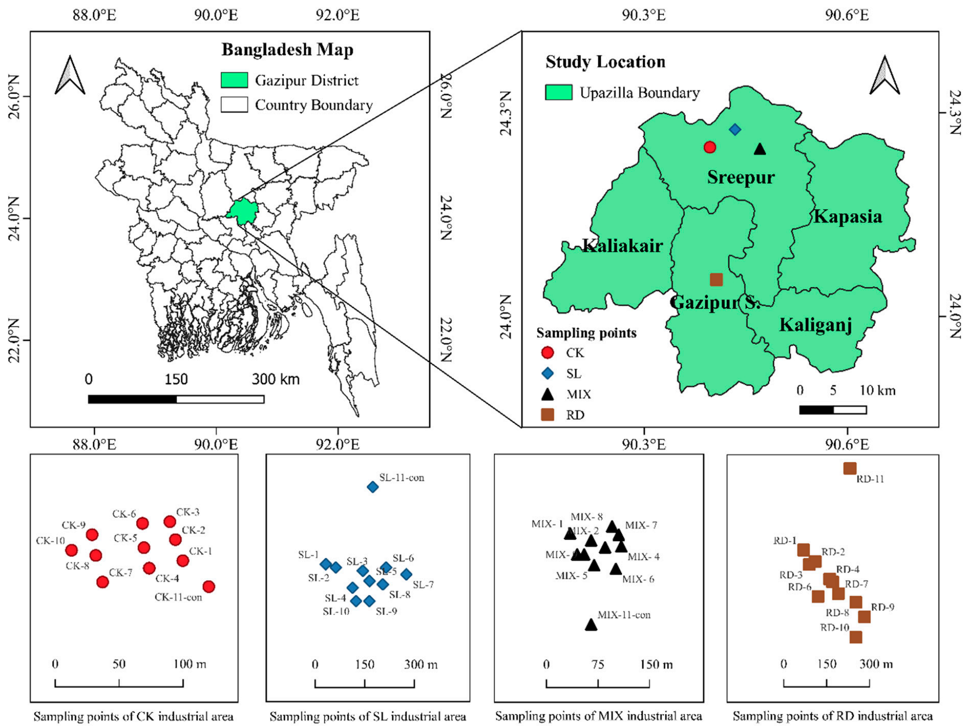

Figure 1. Map showing composite sampling points in four industrial areas in the Gazipur district of Bangladesh.

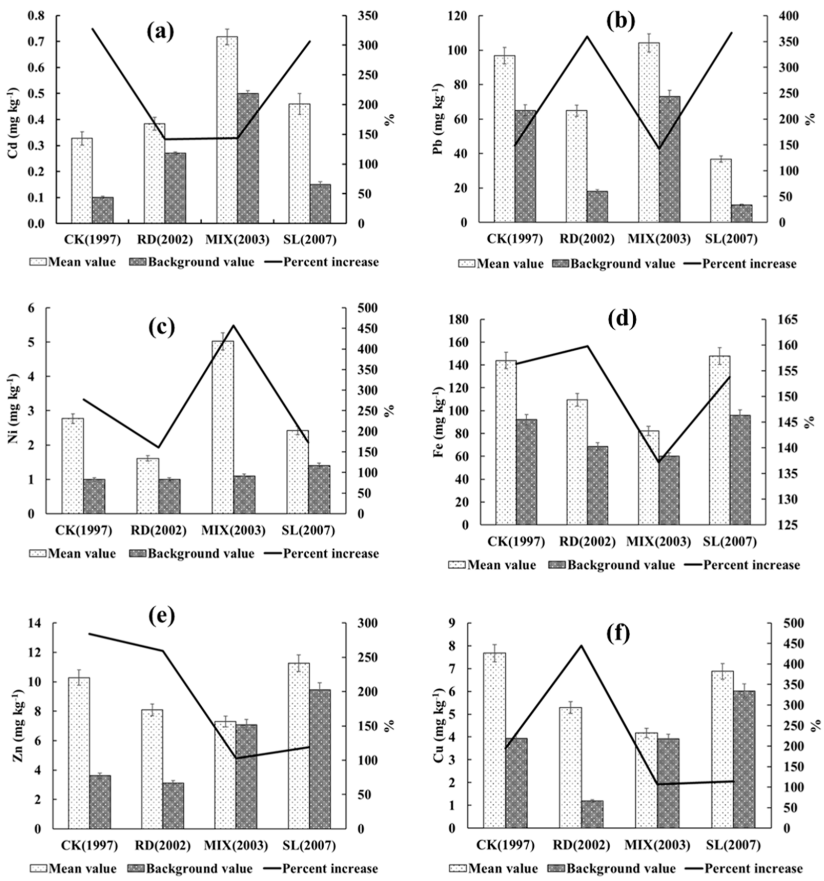

Figure 2. Percent increase of heavy metal concentrations compared to the industry establishment year; (**a**) Cd, (**b**) Pb, (**c**) Ni, (**d**) Fe, (**e**) Zn, and (**f**) Cu (in parenthesis year of establishment).

2.2. Soil Sample Collection

The area of a paddy field soil in the selected study sites is considered as a command area, which has been contaminated through the effluents discharged from respective industries. Representative soil samples (total of 44 samples including 4 controls) were collected from the selected study sites during January–July 2019. Control samples were collected from an uncontaminated field where effluent does not enter. Soil samples were collected from the whole command area maintaining 15 m distance from one sampling point to another sampling point. Each sample was the composite of 5 sub-samples within 2 m surrounding a specific sampling location. Around 2.5 kg of soil for each sample were collected. The longitudes and latitudes of the sampling points were recorded using a portable GPS (global positioning system, Garmin 60CSx, Kansas City, MO, USA) machine.

2.3. Sample Processing and Heavy Metal Analyses

After collection, soil samples were air dried and cleaned to remove the stones, gravels, and plant roots. Prior to chemical analysis, the soil samples were homogenized with a vibrating sample mill (HEIKO TI-200, Tokyo, Japan) and sieved using a 2 mm sieve. For the heavy metal (Cd, Ni, and Pb) determination, the soil sample was digested by di-acid HNO_3–$HClO_4$ (2.5:1) [26,27] in a block digester (Model behrotest K24 Digestion Unit) [28]. To ensure the analysis quality at the time of digestion, the secondary reference material (SRM) was used to compare the certified value; blank and duplicate samples were used to compare the recovery percentage of digestion where 90% and above was accepted. The DTPA extraction method was used for the determination of Fe, Cu, and Zn [29]. The concentrations of Cd, Ni, Pb, Mn Fe, Cu, and Zn were determined by an atomic absorption spectrophotometer (AAS, VGP 210 Buck Scientific, Norwalk, CT, USA). The BUCK Scientific standard (BS-AQ) for individual metals was used for the calibration of the AAS.

2.4. Evaluation Method for Soil Pollution Indices

Different pollution indices were calculated based on the analysis data to evaluate the extent of the heavy metal contamination. The following indices are described here.

2.4.1. Single Factor Pollution Index (SFPI)

The Single Factor Pollution Index (SFPI) method was applied by the China Green Food Development Center [30] for the first time to evaluate the heavy metal pollution. They suggested the following equation to determine the pollution index (Pi) (Equation (1)):

$$Pi = \frac{Ci}{Si} \tag{1}$$

where P_i is the pollution index of pollutant i; C_i is the measured value of i; and S_i is the guideline value of i. The P_i value > 1 indicates metal pollution whereas <1 indicates no pollution. For calculating the SFPI, the background value was used as a guideline value.

2.4.2. Nemerow's Multi-Factor Pollution Index

For the determination of the individual heavy metal pollution in soils, SFPI was used, whereas to measure the overall heavy metal pollution, Nemerow's multi-factor pollution index was used by following the equation described below (Equation (2)):

$$I = \sqrt{(P_{iMax}^2 + P_{iAve}^2)}/2 \tag{2}$$

where I is Nemerow's multi-factor pollution index at location i; P_{iMax} is the maximum; and P_{iAve} is the average value of SFPI. According to Nemerow's multi-factor pollution index, environmental quality is divided into five levels, viz., heavy pollution level ($I > 3.0$), moderate level ($2.0 < I \leq 3.0$), light pollution level ($1.0 < I \leq 2.0$), precaution level ($0.7 < I \leq 1.0$), and clean level ($I \leq 0.7$) [30].

2.4.3. Contamination Factor (*CF*)

The magnitude of soil heavy metal contamination was assessed by using the contamination factor (*CF*). To measure the contamination factor (*CF*), the following equation (Equation (3)) was used [31]:

$$CF = \frac{Cn(sample)}{Bn} \tag{3}$$

where *Cn* is the metal concentration of the studied soil sample, and *Bn* is the geochemical background value.

2.4.4. Geo-Accumulation Index (I_{geo})

The Geo-accumulation index (*Igeo*) was used to determine the degree of metal pollution by following Equation (4) [32]:

$$Igeo = \log 2\left(\frac{Cn}{1.5Bn}\right) \tag{4}$$

where *Cn* is the metal concentration of the studied sample, and *Bn* is the background value.

2.4.5. Potential Ecological Risk Index (*PERI*)

To calculate the potential ecological risk index (*RI*) of heavy metals, the following equations (Equations (5) and (6)) were used [31]:

$$E^r{}_i = T^r{}_i \times CF \tag{5}$$

where $E^r{}_i$ is the potential ecological risk for metals; $T^r{}_i$ is the toxic response factor; and the $T^r{}_i$ value is 5 for Cu, Pb, and Ni, 30 for Cd, and 1 for Zn [33].

$$RI = \sum_{i=1}^{\infty} E_i^r \tag{6}$$

where the comprehensive potential ecological risk index of the metals is expressed as RI.

2.5. Statistical and Geostatistical Analysis

The statistical analyses of experimental data were performed using Statistix 10. An analysis of variance (ANOVA) test was performed to identify the significant results between means of heavy metals in the studied area [34]. For geostatistical analysis and visualization of map outputs, the QGIS software was used. The spatial pattern maps of Cd, Ni, Pb, Fe, Cu, and Zn in the command area were generated by using the Inverse Distance Weighted (IDW) method in a QGIS environment.

3. Results

3.1. Heavy Metal Concentration in Paddy Soil

The mean concentrations (mg kg^{-1}) of different heavy metals (Cd, Pd, Ni, Fe, Zn, and Cu) in rice soil of all selected industries are presented in Table 1. The concentrations of all heavy metals were significantly different from each other among all the selected industries. Significantly higher concentrations of Cd (0.72 mg kg^{-1}), Pb (104.20 mg kg^{-1}), and Ni (5.02 mg kg^{-1}) were found in the MIX industry compared to other industries, while significantly higher concentrations of Fe (147.65 mg kg^{-1}) and Zn (11.27 mg kg^{-1}) were recorded in the SL industry. On the other hand, a higher concentration of Cu (7.67 mg kg^{-1}) was detected in the CK industry which was statistically similar to the SL industry (6.88 mg kg^{-1}).

The trend of Cd, Pb, and Ni loading in rice field soils in different industries was MIX > SL > RD > CK, MIX > CK > RD > SL, and MIX > CK > SL > RD, respectively, whereas for Fe, Zn, and Cu SL > CK > RD > MIX, SL > CK > RD > MIX, and CK > SL > RD > MIX, respectively. (Table 1).

Table 1. Mean concentration (mg kg^{-1}) of different heavy metals in industrial contaminated paddy field soils.

Industry	Cd	Pb	Ni	Fe	Zn	Cu
SL	0.46 ± 0.08 b	36.70 ± 3.85 c	2.42 ± 0.32 b	147.65 ± 10.64 a	11.27 ± 1.33 a	6.88 ± 0.48 a
MIX	0.72 ± 0.06 a	104.20 ± 4.59 a	5.02 ± 0.81 a	82.31 ± 5.38 c	7.29 ± 0.62 b	4.18 ± 0.42 b
CK	0.33 ± 0.05 b	96.80 ± 5.25 b	2.77 ± 0.73 b	143.86 ± 10.29 a	10.28 ± 1.33 ab	7.67 ± 0.76 a
RD	0.38 ± 0.05 b	64.80 ± 6.74 b	1.61 ± 0.25 b	109.58 ± 8.6 b	8.09 ± 0.80 b	5.29 ± 0.33 b
Min	0.33 ± 0.05	36.70 ± 3.85	1.61 ± 0.25	82.31 ± 5.38	7.29 ± 0.62	4.18 ± 0.42
Max	0.72 ± 0.06	104.20 ± 4.59	5.02 ± 0.81	147.65 ± 10.64	11.27 ± 1.33	7.67 ± 0.76
CV%	39.77	21.81	62.37	23.47	36.47	27.55
SE	0.08	7.38	0.82	12.69	1.50	0.74

SL, MIX, CK, and RD indicate chemical industry, textile and paints industry, dyeing industry, and sweater and dyeing industry, respectively. Values having different letters indicate significant differences among the industries.

3.2. Percent Increase of Heavy Metals Concentration

The studied paddy soils adjacent to different industrial areas showed higher heavy metals concentration compared to their background values (Figure 2). The highest Cd concentration increase (327.20%) was found in the study area of CK industry as compared to the background value (0.1 mg kg^{-1}) which was close (306.40%) to the SL industrial area, where background value was 0.15 mg kg^{-1} (Figure 2a and Table 2).

Table 2. Background value of different industry and PTE-MPC value* of different heavy metals.

Heavy Metal	Background Value (mg kg^{-1})				PTE-MPC Value *
	SL	MIX	CK	RD	
Cd	0.15	0.50	0.10	0.27	0.30
Pb	10.00	73.00	0.65	18.00	300.00
Ni	01.40	01.10	1.00	01.00	50.00
Fe	96.00	60.00	92.0	68.60	-
Zn	09.46	07.08	3.62	03.12	250.00
Cu	06.02	03.92	3.94	01.19	100.00

SL, MIX, CK, and RD indicate chemical industry, textile and paints industry, dyeing industry, and sweater and dyeing industry, respectively. * PTE-MPC= "maximum permissible concentrations of potential toxic elements" for agricultural soils of China [35].

The percent increases of Cd concentration for the MIX and RD industries were 143.46% and 142.00%, respectively, as compared to their background concentration (0.50, 0.27 mg kg^{-1}) (Figure 2a and Table 2). It was observed that the maximum increase of Pb was 367.00% in the SL industry followed by the RD industry (360.00%) compared to the background values 10.00 mg kg^{-1} and 18.00 mg kg^{-1} (Table 2), respectively. However, the increasing percentages of Pb in the CK and MIX industries were 148.92 and 142.74%, respectively, compared to their background concentration (Figure 2b). In the case of Ni, the maximum increase (456.36%) was observed in the MIX industry followed by the CK (277.00%), SL (172.86%), and RD (161.00%) industries where the background concentrations were 1.10, 1.00, 1.40, and 1.00 mg kg^{-1}, respectively (Figure 2c and Table 2).

The percent increases of Fe concentration were 159.74, 156.36, 153.80, and 137.00% in the RD, CK, SL, and MIX industries compared to their background concentrations of 68.60, 92.00, 96.00, and 60.00 mg kg^{-1}, respectively (Figure 2d, and Table 2). The percent increase of Zn concentration was the highest (284.06%) in the CK industrial area containing a background value of 3.62 mg kg^{-1}, where the lowest value (102.98%) was observed in the MIX industrial area having a background value of 7.08 mg kg^{-1} (Figure 2e and Table 2). The highest percent increase of Cu was observed in the RD industry (444.29%) followed by the CK (194.75%), SL (114.30%), and MIX (106.58%) industries, respectively, as compared with their background concentrations (1.19, 3.94, 6.02, and 3.92 mg kg^{-1}) (Figure 2f and Table 2).

The ordering of the percent increase of heavy metals in different industrial areas compared to their background values is Ni > Cu > Pb > Cd > Zn > Fe (Figure 2).

3.3. Assessment of Pollution Indices

3.3.1. Single Factor Pollution Index (SFPI)

The single factor pollution index (SFPI) of Cd by the RD, CK, SL, and MIX industry were 3.72, 3.27, 3.13, and 1.43, respectively (Table 3). The highest amount of Cd was released by RD while MIX released the lowest amount of Cd. On the other hand, the highest mean value of single factor pollution index of Pb (3.67), Ni (4.56), Fe (1.56), Zn (2.84), and Cu (2.08) was found in the SL, MIX, CK, and RD industries, respectively. It can be said that Fe released from the CK industry was similar to the SL industry. In the cases of CK and RD, the released amount Zn and Cu were almost similar and higher than the other industries (Table 3).

Table 3. Single factor pollution index (SFPI) for Cd, Pb, Ni, Fe, Zn, and Cu in industrial contaminated paddy field soils.

Industry		**Cd**			**Pb**			**Ni**	
	Mean	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**
SL	3.13	1.28	6.37	3.67	1.20	5.50	1.73	0.71	2.79
MIX	1.43	0.87	1.93	1.43	1.16	1.73	4.56	1.00	8.00
CK	3.27	1.58	6.81	1.49	1.15	1.92	2.77	0.90	8.20
RD	3.72	1.50	5.64	1.20	0.56	1.80	1.79	0.56	3.11
		Fe			Zn			Cu	
SL	1.54	1.05	2.03	1.19	0.22	1.58	1.14	0.46	1.33
MIX	1.37	1.01	1.71	1.03	0.61	1.39	1.07	0.31	1.35
CK	1.56	1.07	2.06	2.84	1.07	4.64	1.94	0.81	2.79
RD	1.37	0.99	1.88	2.59	1.30	4.14	2.08	1.14	2.57

SL, MIX, CK, and RD indicate chemical industry, textile and paints industry, dyeing industry, and sweater and dyeing industry, respectively.

3.3.2. Nemerow's Multi-Factor Pollution Index

From Nemerow's multi-factor pollution index of heavy metals in the study areas, it was observed that, in the SL industry, heavy pollution occurred from Cd (5.02) and Pb (4.68) (Table 4), while Ni (2.32) was responsible for moderate pollution, and light pollution was found for Fe (1.80), Zn (1.40), and Cu (1.24). In the case of the MIX industry, heavy pollution was observed only for Ni (6.51), and light pollution was observed for other elements. However, in the CK industry heavy pollution was found for Ni (6.12), Cd (5.34), and Zn (3.85). Moreover, in the RD industrial area heavy pollution was revealed for Cd (4.78) and Zn (3.45), whereas Ni (2.54) and Cu (2.34) were accountable for moderate pollution. The pollution intensity of individual heavy metals followed the order of Ni > Cd > Pb > Zn > Cu > Fe (Table 4).

Table 4. Nemerow's multi-factor pollution index (*I*) of heavy metals in soils.

Metals	**Industry**							
	SL		**MIX**		**CK**		**RD**	
	I	***PL***	***I***	***PL***	***I***	***PL***	***I***	***PL***
Cd	5.02	Heavy	1.70	Light	5.34	Heavy	4.78	Heavy
Pb	4.68	Heavy	1.59	Light	1.72	Light	1.53	Light
Ni	2.32	Moderate	6.51	Heavy	6.12	Heavy	2.54	Moderate
Fe	1.80	Light	1.55	Light	1.83	Light	1.64	Light
Zn	1.40	Light	1.22	Light	3.85	Heavy	3.45	Heavy
Cu	1.24	Light	1.22	Light	2.40	Light	2.34	Moderate

I and *PL* indicate Nemerow's multi factor pollution Index and pollution level. SL, MIX, CK, and RD indicate chemical industry, textile and paints industry, dyeing industry, and sweater and dyeing industry, respectively.

3.3.3. Contamination Factor (*CF*)

The contamination factor (*CF*) of heavy metals in the study areas was found < 1 for Ni, Fe, Zn, and Cu, while Pb exhibited significantly ($p < 0.05$) the highest *CF* value (5.21) followed by Cd (2.39). The highest *CF* value of Pb (> 3 but < 6) was recorded in the MIX industrial area which indicates the considerable contamination in the MIX industry of Pb. Likewise, the CK (4.84) and RD (3.24) industrial areas were considerably contaminated with Pb. However, moderate contamination from Pb (1.84) was found in the SL industrial area. The range of *CF* values for Cd was 1.90–2.39 which showed that moderate contamination ($1 < CF < 3$) occurred from Cd in all industrial areas (Table 5).

Table 5. Contamination factor (*CF*)* of heavy metals in different industrial contaminated paddy soils.

Industry	Pb	Cd	Ni	Fe	Zn	Cu
RD	3.24 b	1.28 b	0.02 b	2.32×10^{-3} b	0.09 b	0.12 b
CK	4.84 a	1.09 b	0.05 b	3.05×10^{-3} a	0.11 ab	0.17 a
SL	1.84 c	1.53 b	0.04 b	3.13×10^{-3} a	0.12 a	0.15 a
MIX	5.21 a	2.39 a	0.07 a	1.74×10^{-3} c	0.08 b	0.09 b
Min	1.84	1.09	0.02	1.74×10^{-3}	0.08	0.09
Max	5.21	2.39	0.07	3.13×10^{-3}	0.12	0.17
CV%	21.81	39.74	62.37	23.47	36.47	27.55
SE	0.37	0.28	0.01	2.688×10^{-3}	0.02	0.02

SL = chemical industry, MIX = textile and paints industry, CK = dyeing industry, RD= sweater and dyeing industry. Values having different letters indicate significant differences among the industries. **CF*s were classified as: low contamination: $CF < 1$; moderate contamination: $1 < CF < 3$; considerable contamination: $3 < CF < 6$ and very high contamination at $CF > 6$ [36].

3.3.4. Geo-Accumulation Index (I_{geo})

Geo-Accumulation Index (I_{geo}) of studied industrial areas is presented in Table 6. According to I_{geo}, all industrial areas were moderately polluted ($1 < I_{geo} < 2$) except SL (0.19), which can be considered as uncontaminated to moderately contaminated. Among them, the maximum I_{geo} value (1.78) of Pb was found in the MIX industrial area. In the case of Cd, only the MIX industrial area showed uncontaminated to moderately contaminated ($0 < I_{geo} < 1$) levels while other industrial areas showed practically no contamination. The I_{geo} values of other metals (Ni, Zn, Fe, and Cu) were less than 0, which indicates that all industrial areas are practically uncontaminated (Table 6).

Table 6. Geo-Accumulation Index (*Igeo*)* of heavy metals in industrial contaminated paddy field soil.

Industry	Pb	Cd	Ni	Fe	Zn	Cu
RD	1.03 b	−0.37 b	−6.18 b	−9.38 b	−4.20 a	−3.70 bc
CK	1.67 a	−0.59 b	−5.58 b	−8.98 a	−3.92 a	−3.22 a
SL	0.19 c	−0.15 b	−5.52 b	−8.94 a	−3.83 a	−3.34 ab
MIX	1.78 a	0.62 a	−4.59 a	−9.78 c	−4.34 a	−4.12 c
Min	0.19	−0.59	−6.18	−9.78	−4.34	−4.12
Max	1.78	0.62	−4.59	−8.94	−3.83	−3.22
CV%	37.36	−533.74	−16.46	−3.64	−15.85	−14.38
SE	0.20	0.28	0.40	0.15	0.29	0.23

SL, MIX, CK, and RD indicate the chemical industry, textile and paints industry, dyeing industry, and sweater and dyeing industry, respectively. *I_{geo} has seven grades: Grade 0 (practically uncontaminated); Grade 1 (uncontaminated to moderately contaminated): $0 < I_{geo} < 1$; Grade 2 (moderately contaminated): $1 < I_{geo} < 2$; Grade 3 (moderate to heavily contaminated): $2 < I_{geo} < 3$; Grade 4 (heavily contaminated): $3 < I_{geo} < 4$; Grade 5 (heavy to extremely contaminated): $4 < I_{geo} < 5$; Grade 6 (extremely contaminated): $I_{geo} > 5$ [32]. Values having different letters indicate significant differences among the industries.

3.3.5. Potential Ecological Risk Index (PERI)

The Potential Ecological Risk Index (PERI) of six heavy metals in all industrial areas is summarized in Table 7. The findings of this study showed that the intensity of pollution by industrial wastewater in all industrial areas was low. The PERI of Cd (32.72–71.73) was found to be highest compared to other elements. The descending order of PERI in all industrial areas was Cd > Pb > Zn > Ni > Fe > Cu (Table 7).

Table 7. Potential Ecological Risk Index (PERI) of heavy metals in industrial contaminated paddy field soil.

Industry	Pb	Cd	Ni	Fe *	Zn	Cu
RD	16.20 b	38.34 b	0.12 b	-	0.59 b	0.02 a
CK	24.20 a	32.72 b	0.20 b	-	0.85 a	0.01 b
SL	09.18 c	45.96 b	0.18 b	-	0.76 a	0.01 b
MIX	26.05 a	71.73 a	0.37 a	-	0.46 b	0.01 c
Min	09.18	32.72	0.12	-	0.46	0.01
Max	26.05	71.73	0.37	-	0.85	0.02
CV%	21.81	39.74	62.37	-	27.55	13.15
SE	1.84	3.89	0.06	-	0.08	6.953×10^{-4}

SL, MIX, CK, and RD indicate chemical industry, textile and paints industry, dyeing industry, and sweater and dyeing industry, respectively. RI has four categories viz. very high ≥ 600, high pollution = RI = 300–600, considerable pollution = RI = 150–300, and low pollution = RI < 150 (Hakanson 1980). Values having different letters indicate significant differences among the industries. * N.B. $T^r{}_i$ value for Fe was not found which is why PERI is not calculated for Fe.

3.3.6. Relationship among Heavy Metals Concentration

The Pearson correlation coefficient was calculated to observe the relationship among heavy metal concentrations. The positive and significant ($p < 0.05$, <0.01, <0.001) correlations were observed between Ni and Cd (0.4278), between Ni and Pb (0.4272), between Zn and Fe (0.4106), between Cu and Fe (0.6702), and between Cu and Zn (0.4951) (Tables 8 and 9).

Table 8. The Pearson's correlation matrix of Cd, Pb, and Ni in surface soils of the contaminated areas.

	Cd	Pb	Ni
Cd	1		
Pb	0.2832	1	
Ni	0.4278 **	0.4272 **	1

** indicate significance at $p < 0.01$.

Table 9. The Pearson's correlation matrix of Fe, Zn, and Cu in surface soils of contaminated area.

	Fe	Zn	Cu
Fe	1		
Zn	0.4106 **	1	
Cu	0.6702 ***	0.4951 **	1

** indicate significance at $p < 0.01$; *** indicate significance at $p < 0.001$.

3.3.7. Spatial Variation of Heavy Metals

The spatial distribution maps illustrate the visual display of heavy metal distribution in the command areas (Figures 3 and 4). The increasing trend of heavy metal contamination was observed compared to its background value. The highest concentrations of heavy metals were found in the source of the effluent deposal area by industrial outlets, and then the concentrations gradually spread to the other areas with decreasing concentration.

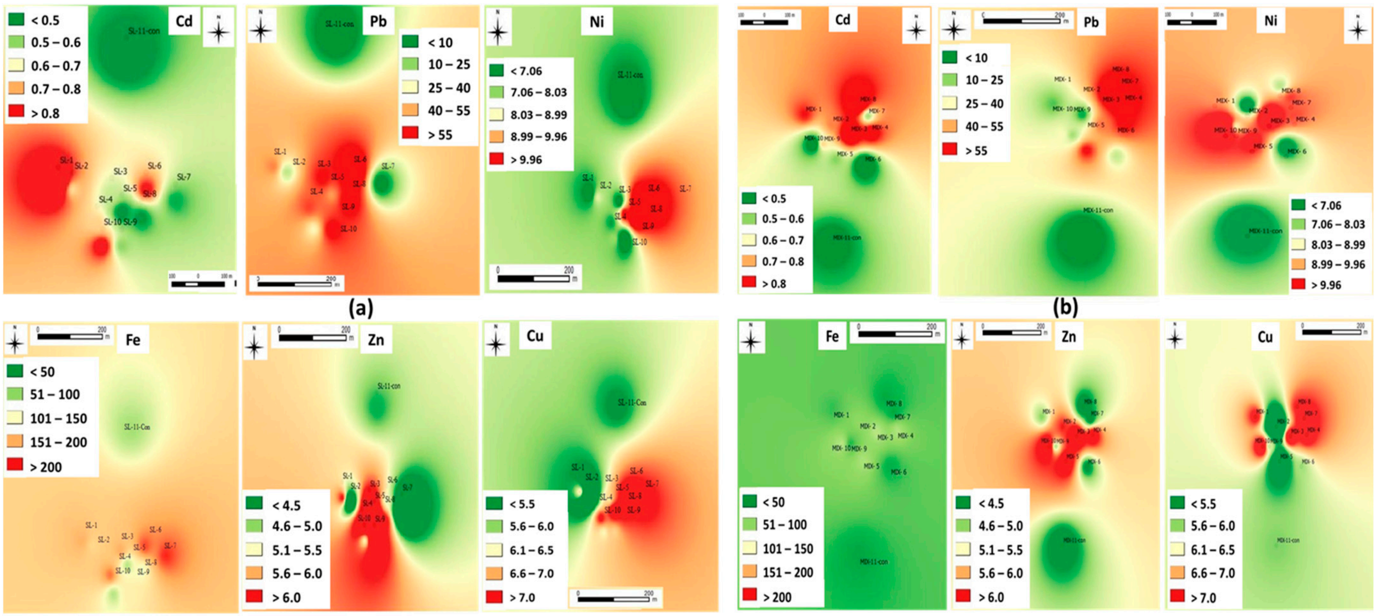

Figure 3. Spatial distributions of (**a**) chemical (SL) industrial and (**b**) textile and paints (MIX) industrial area soils.

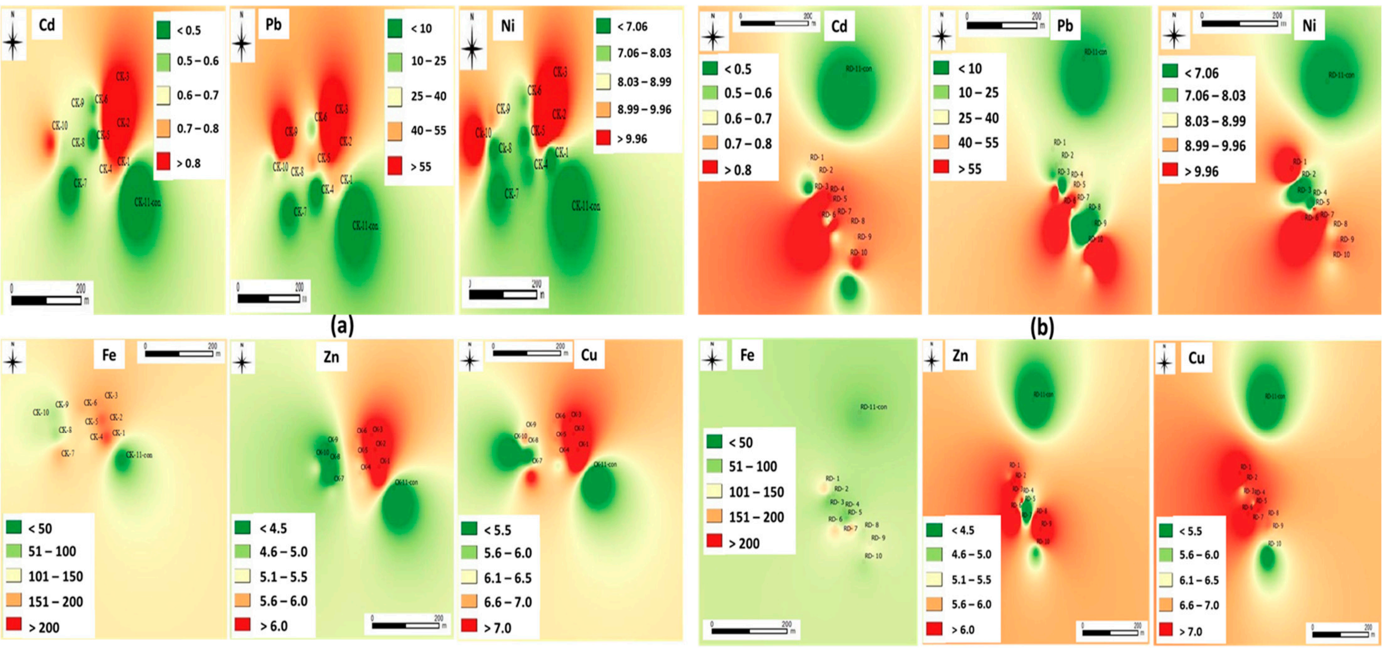

Figure 4. Spatial distributions of (**a**) dyeing (CK) industrial and (**b**) sweater and dyeing (RD) industrial area soils.

4. Discussion

Due to rapid industrialization, more industries are developed day by day in Bangladesh without proper planning [37]. However, most of the industries do not have appropriate treatment plants for wastewater, and effluents are discharged directly into the agriculture fields, canals, and rivers. As a consequence, the untreated industrial wastewater carries different types of contaminants including heavy metals to the adjacent areas of each industry, and ultimately the soils become contaminated. Thus, the intrusion of and irrigation with this untreated industrial wastewater is the main source and cause of increasing heavy metal buildup in the adjacent area soils of an industrial zone. Previous research revealed that short-term sewage irrigation increases heavy metal concentration both in soil and crops [1,2,38]. The results from the present study revealed that heavy metal contamination increases in the vicinity of the industrial-contaminated rice field soils. This agrees with the outcomes of the studies conducted by [39,40]. They observed that the physical, chemical, and biological properties of the surface soil were altered due to the toxicity of heavy metals, and increasing levels of heavy metals polluted the area day by day from the adjacent industries.

Based on the findings of the present study, the mean values of Cd, Pb, Ni, Fe, Cu, and Zn in command area soils were higher than the background values which indicates an increasing trend of heavy metal contamination in paddy field soils through contaminated wastewater from the different industries of the selected industrial zone of Bangladesh (Table 1). Proshad et al. [41] also found almost similar outcomes from their study. The determined values of different heavy metals were higher due to the probability of anthropogenic activities for their distribution. The findings of Liu et al. [42] also matched our findings that anthropogenic sources of heavy metal gradually created soil pollution which ultimately contaminated the food chain. Karim et al. [43] also reported that heavy metal accumulation occurs mainly due to unplanned industrial activities and the discharge of untreated industrial wastewater in the soil environment.

Focusing on individual industries, the heavy metal loading pattern was not similar in all of the industries. For example, significantly higher concentrations of Cd, Pb, and Ni were found in MIX industry compared to other industries while significantly higher concentrations of Fe and Zn were recorded in the SL industry. Furthermore, a higher concentration of Cu was detected in the CK industry which was statistically similar to the SL industry. In the cases of maximum contamination, the effluents are not treated before being disposed of in the industrial adjacent areas. This phenomenon might be due to the cost involved and the lack of knowledge about the toxicity of soil pollution and ultimately contaminating the food chain which may cause serious health hazards [44–47]. As a result, heavy metals remain for a long time in the deposited areas, and the areas become more highly contaminated than the uncontaminated areas. It can also be observed that heavy metal pollution loading is not dependent on the year of establishment of the industry (Figure 2). The intensity of heavy metal accumulation in paddy field soils varied due to the different types discharged wastewater from the different industries (Table 1). The major sources of Cd and Pb are different types of dye complexes that are used in textile and dyeing industries [48]. This could be the reason for the higher concentrations of Cd, Pb, and Ni in the MIX industry where the effluents were discharged from the textile and paint industries. However, higher concentrations of Fe and Zn in the SL industry could be attributed to the use of different Fe- and Zn-rich raw materials for manufacturing of different chemicals. A relatively higher concentration of Cu in the CK industry could be due to the use of Cu for dyeing purposes [1,2].

The mean SFPI values of Cd, Pb, Ni, Fe, Zn, and Cu were higher than the value of 1.0, compared with the background values in the study area indicating that the soils of the study areas are being contaminated on a day by day basis through the discharge or overflow of paddy soils with industrial wastewater containing heavy metals. From this study, it is observed that if the SFPI value of any heavy metal of an agricultural soil exceeded 1, the soil is not suitable for agricultural production due to the possibility of food contamination [49]. The I_{geo} values indicate that there is moderate contamination of Pb in all industries except the SL industry due to a different source and industrial activities. According to [50], there is moderate pollution from Pb compared to other metals which exhibit a I_{geo} value of less than zero. The highest PERI value of Cd, Pb, and Ni were found in the MIX industrial area which suggests that the textiles and paints are more responsible for increasing the pollution level than the dyeing and sweater industries. According to the study of Martin et al., it can be suggested that the PERI values of Cd, Pb, and Ni were significantly higher than other metals which could be due to different types of anthropogenic activities such as the application of phosphate fertilizers and industrial activities [51]. A positive and significant correlation was observed (Table 8) among heavy metals, which indicates the common origin of these heavy metals However, a weak and negative correlation among other metals denotes the different sources of each metal, indicating that heavy metal occurrence might be either natural or anthropogenic.

The highest concentrations of heavy metals were found in the source of effluent disposal areas by industrial outlets, and then the concentrations gradually spread to other areas (Figures 3 and 4). This is due to the initial deposition of effluents with higher concentrations of heavy metals near the source, and with increasing time and distance the concentrations decrease. It was reported that there is a decreasing trend in the concentration of heavy metals with increasing distance from contaminated areas [52]. The results of Faccinelli et al. and Martin et al. also revealed the similar spatial distribution of heavy metals in paddy field soil [53–55].

5. Conclusions

This study identified the heavy metal contamination in the vicinity of different industries located in Gazipur district of Bangladesh. Based on the background value used as a guideline value, the surface soils of rice fields adjacent to the different industries are being contaminated with different heavy metals. The spatial pattern of heavy metals indicated a higher contamination at the adjacent area of industries as compared to the distal soils. Although the ecological risk factor was not high enough yet to be threat to the environment, the pollution loading of different heavy metals varies from low to high which could include the potential rise of ecological risk in the future if the discharge of industrial effluents continues. The outcomes of this study will provide background information on soil quality status due to heavy metal contamination and can also be used as a soil quality data set for further comparisons in Bangladesh. We suggest that the GOB should require each industry to remove heavy metals from their effluent before disposing of it in nearby fields or rivers. Future studies should focus on heavy metal accumulation in rice paddies and other crops and their associated human health risk.

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