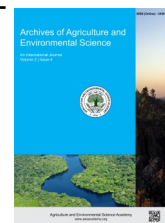This content is available online at AESA

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ORIGINAL RESEARCH ARTICLE

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Heavy metals toxicity of surface soils near industrial vicinity: A study on soil contamination in Bangladesh

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ABSTRACT

Tangail district is more vulnerable to heavy metals contamination for industrialization in Bangladesh. Present study describe six heavy metals (Cr, Ni, Cu, As, Cd, and Pb) in fifteen several sampling locations in industrial vicinity of Tangail district were determined. The concentration of Cr, Ni, Cu, As, Cd, and Pb in studied areas soils were observed 0.96–14.04, 0.71–18.39, 1.02–34.44, 1.2–11.21, 0.44–3.31 and 2.01–28.86 mg/kg, respectively. There is representing a potential risk to the environment for presenting of these heavy metals in soils. This metals are generally toxic to soil and environment. They can persist in the environment for many years and have adverse effect to ecology. Certain indices like contamination factor (CF), enrichment factor (EF), geoaccumulation index (Igeo), pollution load index (PLI), source analysis, principle component analysis (PCA), and toxic units were calculated to determine environmental hazard caused by heavy metals in studied soils. Enrichment factors for the studied metals were in the descending order of Cd > As > Pb > Cu >Ni > Cr. The PLI values for studied metals, causes the Cd contamination in soil of Tangail district. Potential ecological risk (PER) showed low to very high risk to studied vicinity.

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INTRODUCTION

Soil, a key element for the survival of human life on the planet, is expected to be the primary recipient of persistent contaminants such as toxic heavy metals (Karim *et al.*, 2014). Heavy metal soil contamination is known to be the most adverse environmental issue in the world. In Bangladesh, due to rapid industrialization, heavy metal contamination from industrial waste is now one of the burning problems. Most industries do not have waste treatment plants and the waste is thrown directly into the open environment such as soil, canal and river. These industrial wastes get mixed to soil and pollute soil. Heavy metals have

toxicity, persistent, wide sources, accumulative behaviors and non-biodegradable properties which is the result of great concern at present (Islam *et al.*, 2014a). The toxicity of heavy metals in surface soil can alter the physical, chemical, and biological characteristics of the soil. Because of these increases in heavy metals in the soil, the soil is toxic (Khan *et al.*, 2010; Kumar *et al.*, 2015). Soil pollution by heavy metals is universal issues and anthropogenic activities predispose it (Han *et al.*, 2002; Vare, 2006). In recent decades, soil pollution has occurred due to several heavy metals for further urbanization, industrialization and is more suitable for developing countries because of the indiscriminate development of these industries

without proper planning (Ahmed *et al.*, 2015). Heavy metals may originate in soils around the industrial area from numerous prime cause but industrial activities is the most important one and also generation of power, manufacturing, burning of fossil fuel and disposal of waste (Karim *et al.*, 2014; Martín *et al.*, 2014). Heavy metals have significant adverse effects on soils because of their potential environmental issues and adverse effects on soil bionetworks (Yuan *et al.*, 2014). To determine environmental risks of toxic elements in soils, several methods have been widely used, like contamination factor, enrichment factor, and geoaccumulation index (Liu *et al.*, 2014; Rashed, 2010). For determination of multiple risk of heavy metals in soil, pollution load index and potential ecological risk index have been used (Huang *et al.*, 2013). Enrichment factor of a vicinity address relative enhancement in any toxic element when pre-industrial soils are compared with studied soils in alike vicinity (Dias *et al.*, 2014). Since soil contamination derives from industry, the present area of study has been given more attention to its pollution in the environment facing threats to heavy metal toxicity contamination resulting from exponential growth, industrial activity and congestion (Islam *et al.*, 2015a). Heavy metals concentration in the industrial area soils were reported in different studies due to rapid industrial activities in Bangladesh. The main purpose of this study was to determine the degree of pollution of toxic elements in soils using enrichment factors, contamination factors, geoaccumulation index, pollution load index, potential ecological hazard, and to identify potential heavy metal sources and soil pollution determination due to ecological threat in Bangladesh's industrial areas.

MATERIALS AND METHODS

Study areas and sampling

The samples were collected from Tarutia, Tangail Sadar Upzila of Tangail district, Bangladesh (Figure 1). Tangail district area is 334.26 km² and situated at the middle part in Bangladesh. Tangail Sadar Upzila is highly densely area in Bangladesh and population density is 1,100/km^2 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 a

Figure 1. *Map showing the study areas of Tangail district, Bangladesh.*

days. There present different kinds of industries in Tangail district like garments, packaging industry, 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 are 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. Soil samples were collected during March - April, 2016. Tarutia was selected for sampling location situated near industrial area of Tangail district, Bangladesh. Fifteen soil sampling sites were selected in the industrial areas of Tangail district. Agricultural field soil samples (samples were collected from surface soil up to 10 cm) were taken and three subsamples collected which were used as composite sample by mixing it thoroughly. Soil was taken with the help of a percussion hammer corer (50–80 cm in length) for metal analysis and this samples were treated as preindustrial sample (Schottler and Engstrom, 2006). To crumble all dried soil samples, a porcelain mortar and pestle were used. Then the samples were sieved with 2 mm nylon sieve. The soil samples were stored in a clean Ziploc bag which was airtight and used for chemical analysis. Several researcher also followed the alike procedure for sampling and storing of soil samples (Oliveira *et al.*, 2012).

Physicochemical parameters analysis

Soil pH was determined by using a glass electrode pH meter (WTW pH 522; Germany). 10 g of air-dried soil from each sampling site was taken in 50 mL beakers separately and 25 mL 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. For EC determination, 5.0 g of soil was taken in 50 mL polypropylene tubes and 30 mL of Milli-Q water was added to the tube. The lid was closed properly and was shaken for 5 min. After that, EC was measured using an EC meter (WTW LF 521; Germany). For organic carbon, 1.0 g of soil was placed at the bottom of a dry 500mL conical flask (Corning/Pyrex). Then 10 mL of 1N $K_2Cr_2O_7$ was added into the conical flask and swirled a little. The flask was kept on asbestos sheet. Then 20 mL 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 200 mL of distilled water was added. After incorporation of 5.0 mL of phosphoric acid and 35 drops of diphenylamine indicator, the contents were titrated against ferrous ammonium sulfate solution till the color flashes blue-violet to green. Simultaneously, a blank titration was run without soil. Particle size was determined using the hydrometer method. The textural classes for different soil samples were then determined by plotting the results on a triangular diagram designed by Marshall followed USDA system. The percentage of sand, silt and clay were calculated as follows.
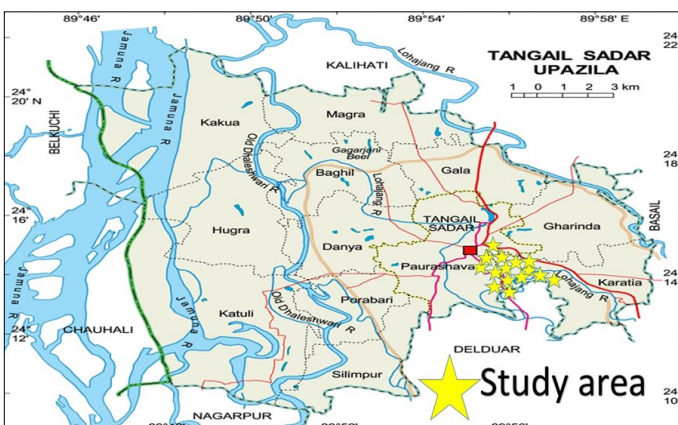
%(Silt + Clay) = (Corrected hydrometer reading at 40 seconds/ Oven dry weight of sediment) × 100　　　　　(1)

%(Clay) = (Corrected hydrometer reading after 2 hours/ Oven dry weight of sediment) × 100　　　　　(2)

Sand (%) = 100 - %(Silt + Clay)　　　　　(3)

Silt (%) = %(Silt + Clay) - % Clay　　　　　(4)

Heavy metal analysis

All chemicals were analytical grade reagents; Milli-Q water (Elix UV5 and MilliQ, Millipore, Boston, MA, USA) was used for the preparation of solutions. The Teflon vessel and polypropylene containers were cleaned, soaked in 5% HNO_3 for more than 24 h, then rinsed with Milli-Q water and dried. For metal analysis, 0.3–0.5 g of the soil sample was treated with 6 mL 69% HNO_3 (Kanto Chemical Co, Tokyo, Japan) and 2 mL 30% H_2O_2 (Wako Chemical Co, Tokyo, Japan) in a closed Teflon vessel and was digested in a Microwave Digestion System (Berghof speedwave, Eningen, Germany). The digested samples were then transferred into a Teflon beaker, and total volume was made up to 50 mL with Milli-Q water. The digested solution was then filtered by using syringe filter (DISMIC1–25HP PTFE, pore size = 0.45 mm; Toyo Roshi Kaisha, Ltd., Tokyo, Japan) and stored in 50 mL polypropylene tubes (Nalgene, New York, NY, USA). After that, the digestion tubes were then cleaned using blank digestion procedure following the same procedure of samples. For trace metals, samples were analyzed using inductively coupled plasma mass spectrometer (ICP-MS, Agilent 7700 series, Santa Clara, CA, USA). Instrument operating conditions and parameters for metal analysis are done. The detection limits of ICP-MS for the studied metals were 0.7, 0.6, 0.8, 0.4, 0.06 and 0.09 ng/L for Cr, Ni, Cu, As, Cd and Pb, respectively. Multi-element Standard XSTC-13 (Spex CertiPrep®, Metuchen, NJ, USA) solutions were used to prepare calibration curves. Multi-element solution (purchased from Agilent Technologies, Japan) was used as tuning solution covering a wide range of masses of elements. All test batches were evaluated using an internal quality approach and validated if they satisfied the defined Internal Quality Controls (IQCs). Before starting the analysis sequence, relative standard deviation (RSD, <5%) was checked by using the tuning solution purchased from Agilent Technologies. The certified reference materials INCT-CF-3 (corn flour) bought from the National Research Council (Canada), were analyzed to confirm analytical performance and good precision (relative standard deviation below 20%) of the applied method. Metals in soil samples were analyzed using an inductively coupled plasma mass spectrometer (ICP-MS).

Ecological risk assessment for soil pollution

Enrichment factor (EF): Enrichment factor (EF) is considered as an effective tool to evaluate the magnitude of contaminants in the environment (Franco-Uría *et al.*, 2009). The EF for each element was calculated to evaluate anthropogenic influences on heavy metals in sediments using the following formula (Selvaraj *et al.*, 2004).

$EF = (C_M/C_{Al})_{sample} /(C_M/C_{Al})_{background}$　　　　　(5)

Where, $(C_M/C_{Al})_{sample}$ is the ratio of concentration of heavy metal (C_M) to that of aluminum (C_{Al}) in the soil sample, and $(C_M/C_{Al})_{background}$ is the same reference ratio in the background sample. Generally, an EF value of about 1 suggests that a given metal may be entirely from crustal materials or natural weathering processes (Zhang and Liu, 2002). Samples having enrichment factor >1.5 was considered indicative of human influence and (arbitrarily) an EF of 1.5–3, 3–5, 5–10 and >10 is considered the evidence of minor, moderate, severe, and very severe modification (Birch and Olmos, 2008).

Contamination factor (C_f^i): Contamination factor means the proportion of the heavy metal concentration in the soil to that of baseline or background value.

$C_f^i = C_{heavy\ metal} /C_{background}$　　　　　(6)

Contamination factor divided into four classes ranged from 1 to 6 which are: low degree (C_f^i <1), moderate degree ($1 \leq C_f^i < 3$), considerable degree ($3 \leq C_f^i < 6$), and very high degree ($C_f^i \geq 6$) (Islam *et al.*, 2015a). This approach has been used by other researchers e.g. (Proshad *et al.*, 2017; Kumar and Thakur, 2018).

Geoaccumulation index (I_{geo}): Geoaccumulation index (Igeo) is assumed as an impressive tool to determine contamination degree from toxic metals. At present, geoaccumulation index is used globally to assess soil pollution (Bermejo *et al.*, 2003; Kumar and Thakur, 2017). The most effective objective to determine geoaccumulation index (Igeo) is to identify pollution level in soil. Geoaccumulation index (Igeo) may be assessed by applying equation given here by,

$Igeo = log2 (Cn/1.5Bn)$　　　　　(7)

Where, Cn is the determined element (n) concentration assessed from soil, Bn is the geochemical baseline value of element n in background sample (Yu *et al.*, 2008).

Pollution load index: To assess the quality of soil in terms of metal contamination, an integrated approach of pollution load index of the six metals is calculated according to Rashed (Rashed, 2010). The PLI is defined as the nth root of the multiplications of the contamination factor (C_f^i) of metals (Bhuiyan *et al.*, 2011).

$$PLI = (C_{f\ 1}^{i} \times C_{f\ 2}^{i} \times C_{f\ 3}^{i} \times ...... \times C_{f\ n}^{i})^{1/n}　　　　　(8)$$

The PLI gave an assessment of the overall toxicity status of the sample and also it is a result of the contribution of the six metals. Therefore, PLI value of zero indicates perfection, a value of one indicates the presence of only baseline level of pollutants and values above one would indicate progressive deterioration of the site and estuarine quality (Thomilson *et al.*, 1980). The PLI gave an

assessment of the overall toxicity status of the sample and also it is a result of the contribution of the six metals.

Potential ecological risk (PER): The degrees of hazardous elements contamination in agricultural soils are determined by PER index. (Guo *et al.*, 2010) and (Yu and Li, 2011) proposed equations which were used to calculate PER and are as follows:

$$C_f^i = \frac{C^i}{C_n^i} \qquad C_d = \sum_{i=1}^{n} C_f^i \qquad (9)$$

$$E_r^i = T_r^i \times C_f^i \qquad PER = \sum_{i=1}^{m} E_r^i \qquad (10)$$

Where, C_f^i is the single element contamination factor, C^i is the content of the element in samples and C_n^i is the background value of the element. The background value of Cr, Ni, Cu, As, Cd and Pb in soils were 90, 68, 45, 13, 0.3 and 20 mg/kg, respectively (pre-industrial samples of the study area) (Turekian and Wedepohl, 1961). The sum of C_f^i for all metals represent the integrated pollution degree (C_d) of the environment. E_r^i is the potential ecological risk index and T_r^i is the biological toxic factor of an individual element. The toxic-response factors for Cr, Ni, Cu, As, Cd and Pb were 2, 6, 5, 10, 30 and 5, respectively (Amuno, 2013; Hakanson, 1980; Luo *et al.*, 2007; Wu *et al.*, 2010). PER is the comprehensive potential ecological risk index, which is the sum of E_r^i. Sensitivity of the biological community is represented by it to the toxic substance and indicates the potential ecological risk caused by the overall contamination.

Toxic unit analysis: The calculation of toxic units is considered as severe toxicity of toxic metals in agricultural soils. Toxic unit analysis is the proportion of the assessed concentration of

heavy metals in soil to possible effect level (Islam *et al.*, 2014b). When the sum of toxic units for all soil samples is more than 4, moderate to serious toxicity of heavy metals remain in soil.

Statistical analysis

SPSS 20.0 (SPSS, USA) was used for statistical analysis for present study. 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

Physiochemical properties of soil

Different physical and chemical properties (Texture, pH, EC and organic carbon) of soil were shown in Table 1. Soils pH values for present study were ranged from 5.48 to 7.15 signifying that the studied soil samples were slightly acidic to neutral except S9 and S13 samples which were alkaline in nature (Table 1). Most of the studied soils were acidic to neutral because of decomposition of organic matter and subsequent formation of carbonic acid (Ahmad *et al.*, 1996). Higher soil acidity favors the availability of cations in soil. Soil pH (acidity) is of particular importance as it controls the behavior of metals and many other soil processes. Heavy metal cations (positively charged metal atoms) are most mobile in acid soils. This means that metal contaminants are more available for uptake by plants, or to move into the water supply (Adeniyi *et al.*, 2008; Oliver, 1997). Electrical conductivity (EC) value of the studied soil was non-saline (0-2 dS/m; SRDI soil salinity class) for all sampling sites which mean the salinity effect is negligible (SRDI, 2009). This condition of soil was due to organic matter decomposition with carbonic acid formation in the studied area soils. The range of organic carbon (% C) was 0.664 to 3.331. High organic carbon content is an indication that metals are more likely to be bound to organic matter to form metal chelate complexes, and this would also result in less availability of metals to plants (Yap *et al.*, 2009). According to the United States soil texture classification system (NRCS, 1993), the textural analysis revealed that the studied soil samples were loam, sandy loam, and silt loam (Table 1) according to the soil texture classes.

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

Sampling sites	pH (1:2.5 H$_2$O)	EC (dS/m)	Organic carbon (%)	Sand (% in <2 mm)	Silt	Clay	Soil type
S1	6.36	0.23	1.507	49	32.5	18.5	Loam
S2	6.04	0.54	2.661	36	51.6	12.4	Silt loam
S3	5.48	0.32	0.677	46	37.5	16.5	Loam
S4	6.24	0.36	0.660	42.6	44.1	13.3	Loam
S5	6.43	0.43	1.649	47.4	37.5	15.1	Loam
S6	6.87	0.21	0.996	48.5	39.1	12.4	Loam
S7	6.3	0.36	0.644	41.5	39.1	19.4	Loam
S8	6.35	0.3	0.650	60.1	26.6	13.3	Sandy loam
S9	7.11	0.22	1.019	54	30	16	Sandy loam
S10	6.3	0.21	1.062	49	34.1	16.9	Loam
S11	6.7	0.32	3.331	376	46.6	15.8	Loam
S12	6.43	0.25	0.933	51	36.6	12.4	Loam
S13	7.15	0.27	1.945	47.6	39.1	13.3	Loam
S14	6.54	0.47	1.341	53.5	35	11.5	Sandy loam
S15	6.11	0.19	1.402	44	36.6	19.4	Loam

[*]According to the United states Department of Agriculture soil classification system.

Heavy metals concentrations in soil

The heavy metals concentrations (Cr, Ni, Cu, As, Cd, and Pb) in soil samples were presented in Table 2. The mean concentrations of Cr, Ni, Cu, As, Cd, and Pb in soil were found 4.91, 5.86, 8.06, 4.2, 1.35, and 12.11 mg/kg, respectively (Table 3) around the industrial area of Tangail district, Bangladesh. The highest value of Cr, Ni, Cu, As, Cd and Pb were observed in soil collected from S14, S10, S13, S14, and S11 site. Hazardous element concentrations in present study soil samples were compared with other studies. The mean concentration of Cr was found 4.91 mg/kg in the present study which was lower than The Dutch Soil Quality Standard (VROM, 2000), Canadian Environmental Quality Guidelines (CCME, 2003) and Australian Guideline for Soil Quality (DEP, 2003) indicating lower contamination of Cr in soil (Table 2).

Chromium is a toxic heavy metal is discharged from several industries into the agricultural land around industrial areas and pollutes agricultural soils (Nriagu, 1988). Cr concentration was found in the study areas may be disposed of untreated tannery waste to agricultural fields since chromium salt used in tannery industries (Gowd *et al.*, 2010). The concentration of Cr in agricultural soils varies up to values as high as 350 mg/kg (Branca *et al.*, 1990). Chromium concentration in the present study was lower than other studies (Ahmad and Goni, 2010; Islam *et al.*, 2014a; Luo *et al.*, 2007; Proshad *et al.*, 2017) conducted different areas in Bangladesh and other countries. The toxicity of Cr has negative impacts on the growth of plants that interfere with some important metabolic processes (Hasnine *et al.*, 2017; Shanker *et al.*, 2009).

The solubility of nickel in soils increases with its acidity and if the acidity increases it results higher Ni in soils (Barałkiewicz and Siepak, 1999). In the present study Ni concentrations ranged between 0.71-18.39 mg/kg in the study area. The highest amount (18.39 mg/kg) was found in station 10 and the lowest value (0.71 mg/kg) in station 7 (Table 2). The elevated levels of Ni were found in station 10 which results from localized additions or accidental spillages of Ni containing materials (Krishna and Govil, 2007). The mean concentration of Ni was found 5.86 mg/kg in the present study which was lower than The Dutch Soil Quality Standard (VROM, 2000), Canadian Environmental Quality Guidelines (CCME, 2003) and Australian Guideline for Soil Quality (DEP, 2003) indicating lower contamination of Ni in soil (Table 2). Nickel (Ni) concentration in the present study was lower than other studies (Ahmad and Goni, 2010; Islam *et al.*, 2014a; Luo *et al.*, 2007; Proshad *et al.*, 2017) conducted different areas in Bangladesh and other countries.

Table 2. Metal concentration (mg/kg) in soil collected from industrial areas of Tangail district, Bangladesh.

Sampling sites	Cr	Ni	Cu	As	Cd	Pb
S1	0.964	8.058	8.192	2.128	0.447	12.102
S2	2.704	5.423	5.312	2.850	2.608	2.325
S3	4.599	3.237	2.038	1.313	0.487	11.195
S4	1.665	2.085	2.268	3.016	0.190	2.017
S5	0.414	3.349	6.110	2.149	0.692	12.120
S6	5.646	2.114	9.740	1.481	0.312	6.360
S7	5.923	0.712	4.433	1.200	0.788	9.044
S8	2.699	1.955	2.256	1.784	1.586	4.014
S9	4.214	3.656	1.028	1.365	1.309	17.124
S10	4.212	18.394	2.785	5.439	1.134	13.781
S11	5.160	8.607	7.105	3.191	0.543	28.645
S12	4.958	3.247	5.014	10.388	2.238	26.867
S13	10.532	9.036	34.440	8.049	2.416	13.503
S14	5.935	11.033	18.657	11.210	3.311	17.566
S15	14.047	7.103	11.627	7.563	2.228	5.106
Mean	4.91	5.86	8.06	4.20	1.35	12.11
Dutch standard[a]	100	35	36	29	0.80	85
Canadian guidelines[b]	64	50	63	12	1.4	70
Australian guidelines[c]	50	60	60	20	3.0	300
Background value in Tangail district	29	32	27	6.5	0.82	23

[a](VROM, 2000)   [b](CCME, 2003)   [c](DEP, 2003)

Excessive Cu concentrations are harmful to plants and highly toxic to some microorganisms (Hasnine *et al.*, 2017). Soluble soil Cu can be toxic to plants since Cu-enriched liquid dairy waste used in agricultural land as irrigation water (White and Brown, 2010). In the present study, the value of Cu ranged between 1.02 to 34.44 mg/kg (Table 2). The mean concentration of Cu was found 8.06 mg/kg in the present study which was lower than The Dutch Soil Quality Standard (VROM, 2000), Canadian Environmental Quality Guidelines (CCME, 2003) and Australian Guideline for Soil Quality (DEP, 2003) indicating lower contamination of Cu in soil (Table 2). (Alloway, 1990) provided with the regulatory standard for Cu in soil is 20-30 mg/kg. Cu concentration in the present study was compared to other studies conducted in Bangladesh and other countries. Present studied Cu concentrations were lower than other studies (Ahmad and Goni, 2010; Islam *et al.*, 2014a; Luo *et al.*, 2007; Proshad *et al.*, 2017).

In the present study, the concentration of As varied between 1.2 to 11.21 mg/kg (Table 2). A huge amount of groundwater containing As (Hug *et al.*, 2011) is being used for tanning in relation to some chemicals especially arsenic sulfide (Bhuiyan *et al.*, 2011). Moreover, emission and waste from brick fields and incineration activities might contribute to the high concentration of As (Olawoyin *et al.*, 2012). Arsenic in agricultural soils can be derived from both natural and anthropogenic sources, especially use of groundwater for irrigation and uncontrolled application of As enriched fertilizers and pesticides (Neumann *et al.*, 2010). All the concentrations of As found to below the recommended value set by Dutch Soil Quality Standard (VROM, 2000), Canadian Environmental Quality Guidelines (CCME, 2003) and Australian Guideline for Soil Quality (DEP, 2003) (Table 2). As contaminated water and As-enriched fertilizers as well as pesticides were used for irrigation in the agricultural land ((Polizzotto *et al.*, 2013). Moreover, emission and waste from brick fields and incineration activities might contribute to the high concentration of As in agricultural soil (Olawoyin *et al.*, 2012).

Cadmium concentrations were found between 0.44 to 3.31mg/kg. The mean concentration of Cd was found 1.35 mg/kg in the present study which was higher than The Dutch Soil Quality Standard (VROM, 2000) but higher than Canadian Environmental Quality Guidelines (CCME, 2003) and Australian Guideline for Soil Quality (DEP, 2003). Cadmium (Cd) concentration in the present study was compared to other studies conducted in Bangladesh and other countries. Present studied Cd concentrations were lower than other studies (Ahmad and Goni, 2010; Islam *et al.*, 2014a; Proshad *et al.*, 2017).

This level of Pb concentration present in soil due to metal processing factories release Pb into the open environment and several anthropogenic factors (Nziguheba and Smolders, 2008). In the present study, station 11 showed the elevated concentrations of Pb which can be due to the emission of Pb contaminated waste from these sites (Gowd *et al.*, 2010). The mean concentration of Pb was found 12.11 mg/kg in the present study which was lower than The Dutch Soil Quality Standard (VROM, 2000), Canadian Environmental Quality Guidelines (CCME, 2003) and

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

Region, Country	Cr	Ni	Cu	As	Cd	Pb	References
Tangail (Bangladesh)	4.91 (0.96-14)	5.86 (0.71-18.39)	8.06 (1.02-34.44)	4.2 (1.2-11.21)	1.35 (0.44-3.31)	12.11 (2.01-28.86)	Present study
Tangail (Bangladesh)	10.41 (1.57-21.91)	12.69 (4.74-25.71)	15.66 (3.08-38.56)	12.15 (2.69-28.44)	3.1 (1.03-8.06)	7.98 (2.23-18.32)	(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.*, 2007)
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.*, 2014b)
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)

Australian Guideline for Soil Quality (DEP, 2003) indicating lower contamination of Pb in soil (Table 2). Lead (Pb) concentration in the present study was lower than other studies (Ahmad and Goni, 2010; Islam *et al.*, 2014a; Luo *et al.*, 2007) conducted different areas in Bangladesh and other countries.

Source analysis of heavy metals in soil

Statistical analyses were performed to elucidate the associations among heavy metals in soils and to identify the important factors involved in controlling the transport and distribution of metal contaminants (Proshad *et al.*, 2019). Pearson's correlation (PC) matrix for analyzed soils parameters was calculated to see if some of the parameters interrelated with each other and the results are presented in Table 4. The value of EC showed significant positive correlation with silt (r=-0.524*). Sand showed significant positive correlation with organic matter (r=-0.675**). There were also showed others positive correlations like silt with organic carbon (r=0.61*) and Pb (r=0.59*), Cr with Cu (r=0.575*), Cu with As (r=0.566*) and As with Cd (r=0.762**). Considering the relationship between the combinations showed positive significant relationship which indicates the parameters were interrelated with each other and may be originated from the same source to the study area. Other relationships among the constituents of soil were not significant.

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

	pH	EC	Sand	Silt	Clay	Organic carbon	Cr	Ni	Cu	As	Cd	Pb
pH	1											
EC	-0.277	1										
Sand	0.204	-0.004	1									
Silt	-0.163	0.524*	0.334	1								
Clay	-0.271	-0.405	0.060	-0.198	1							
Organic carbon	0.216	0.318	0.675**	0.61*	-0.090	1						
Cr	0.160	-0.373	0.013	0.007	0.212	0.074	1					
Ni	0.096	-0.141	0.168	-0.053	0.100	0.327	0.196	1				
Cu	0.475	0.002	-0.030	0.079	-0.244	0.306	0.575*	0.315	1			
As	0.154	-0.004	-0.071	-0.038	-0.326	0.085	0.505	0.462	0.566*	1		
Cd	0.119	0.291	-0.220	-0.019	-0.386	0.191	0.450	0.297	0.505	0.762**	1	
Pb	0.349	-0.165	0.590*	-0.059	-0.043	0.336	0.037	0.295	0.107	0.398	0.101	1

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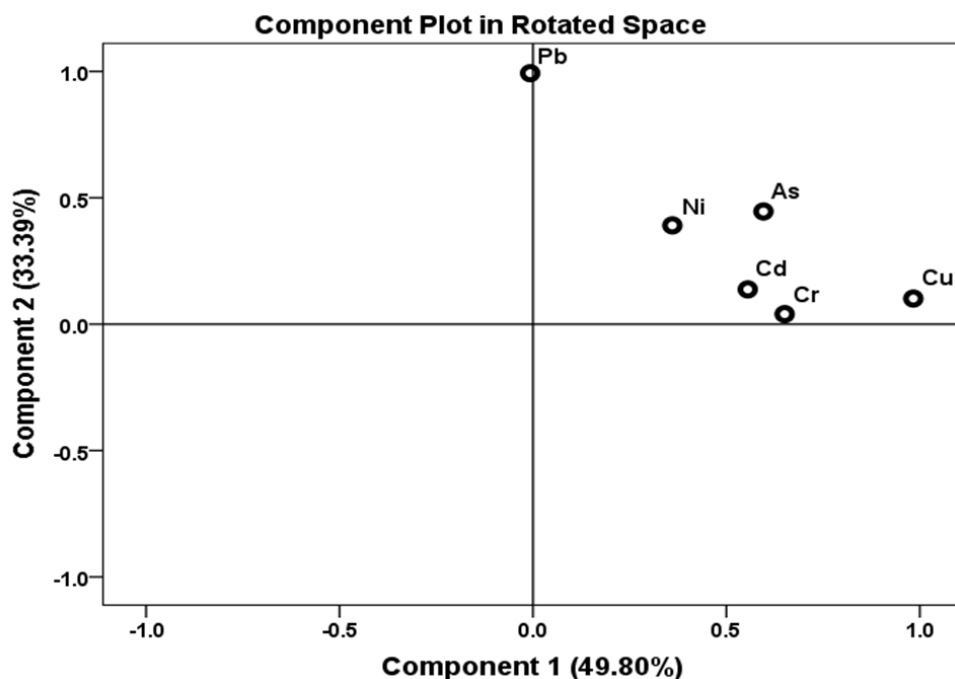

Figure 2. *Principal component analysis (PCA) of heavy metals in soil collected from Tangail district, Bangladesh. Considering the highest component loading, first PC exhibited elevated loadings of Cr, Ni, Cu, As and Cd. Second PC exhibited elevated loadings of Pb.*

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 (Anju and Banerjee, 2012). The principal component analysis was performed on the tabular and standardized forms of data set and is presented in Table 5 and Figure 2. The extraction method was performed to find out the principal components (PC) in PCA analysis that was Eigen values. In this study, two PCs were computed and the variances explained by them were 49.80% and 33.39% for soil samples in the study area (Figure 2). Overall, the PCA revealed two major groups of the metals in soils, where one group consisted of Cr, Ni, Cu, As and Cd which were predominantly contributed by anthropogenic activities (Iqbal and Shah, 2011).

Second group consisted of Pb which were contributed by lithogenic sources or by industrial emissions in the sampling sites (Proshad *et al.*, 2019).

In addition, cluster analysis (CA) with dendrogram using Ward's Method was applied to classify the heavy metals into several groups using the overall heavy metals concentration in soil samples (Figure 3). Several cluster shape were found between heavy metals which were in same cluster were of resembling in nature. In respect of metal pollution in soils exhibited strong significant correlations by building primary clusters with each other (Figure 3). The primary clusters such as Cr, As, Cd and Ni was formed and another cluster were formed with Cu and Pb within a distance of five on the scale (Figure 3).

Table 5. Total variance explained and component matrices for the hazardous elements in surface soils collected from industrial areas of Tangail district, Bangladesh.

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
1	92.613	49.802	49.802	92.613	49.802	49.802	84.396	45.383	45.383
2	62.099	33.393	83.195	62.099	33.393	83.195	70.316	37.812	83.195
3	17.271	9.287	92.482						
4	8.844	4.756	97.238						
5	4.801	2.582	99.820						
6	0.335	0.180	100.000						

Elements	Component matrix				Rotated Component Matrix			
	Raw component		Rascaled component		Raw component		Rascaled component	
Component matrix	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
Cr	2.039	-1.073	0.577	-0.304	2.300		0.651	
Ni	2.376		0.511		1.676	1.818	0.360	0.391
Cu	7.695	-3.650	0.893	-0.424	8.472		0.983	
As	2.554		0.741		2.054	1.538	0.596	0.446
Cd	0.539		0.546		0.548		0.555	
Pb	4.097	6.860	0.509	0.853		7.990		0.993

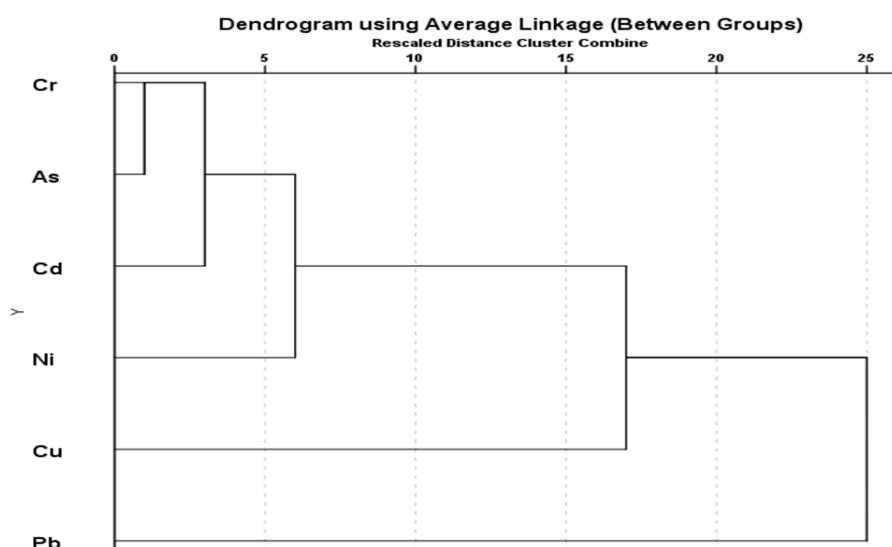**Figure 3.** *Cluster analysis of soil samples for heavy metals collected from industrial areas of Tangail district, Bangladesh.*
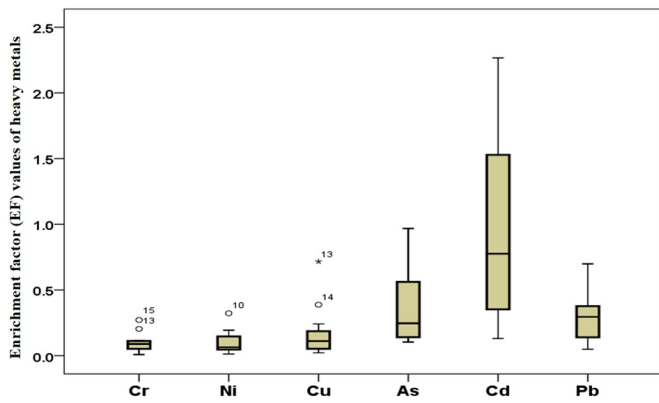
Figure 4. *Enrichment factor (EF) values for heavy metals in soils of sampling sites in Tangail district.*
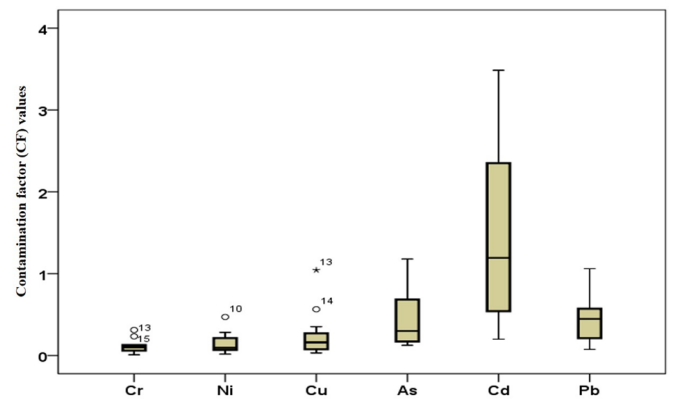

Figure 5. *Contamination Factor (CF) of heavy metals in soils collected from industrial areas of Tangail district, Bangladesh.*
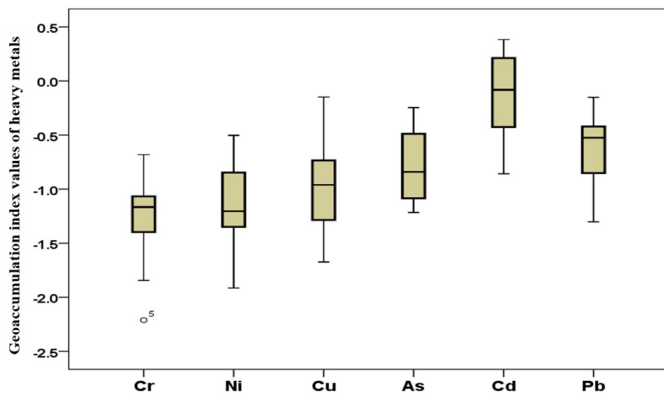

Figure 6. *Geo accumulation index (I_{geo}) value of heavy metals in soils of industrial areas in Tangail district, Bangladesh.*
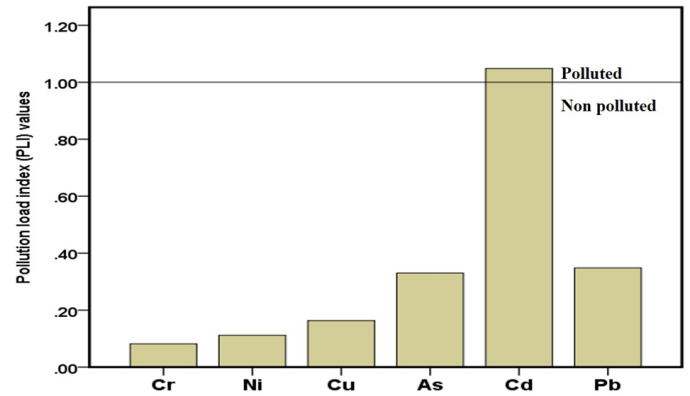

Figure 7. *Pollution load index (PLI) value of heavy metals in soils of Tangail district, Bangladesh.*

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

Sites	Potential ecological risk factor (E_r^i)						Potential Risk (PER)	Pollution degree
	Cr	Ni	Cu	As	Cd	Pb		
S1	0.171415	4.959013	4.965106	8.961436	56.40616	8.96466	84.42779	Moderate risk
S2	0.480704	3.33746	3.219426	11.99998	329.3994	1.721987	350.1589	Very high risk
S3	0.817569	1.991869	1.235273	5.527944	61.55157	8.292594	79.41682	Moderate risk
S4	0.296072	1.283189	1.374376	12.70005	24.04455	1.493711	41.19196	Low risk
S5	0.07364	2.061149	3.702827	9.046509	87.4643	8.977634	111.3261	Moderate risk
S6	1.003696	1.301099	5.902817	6.234841	39.41723	4.711326	58.57101	Low risk
S7	1.052918	0.438321	2.686423	5.054444	99.56221	6.699138	115.4935	Moderate risk
S8	0.47975	1.203172	1.367192	7.512932	200.3827	2.973274	213.919	Considerable risk
S9	0.749184	2.24997	0.623047	5.748393	165.3143	12.6848	187.3697	Considerable risk
S10	0.748717	11.3191	1.687635	22.89913	143.264	10.20823	190.1268	Considerable risk
S11	0.917281	5.296747	4.305867	13.43615	68.56799	21.21834	113.7424	Moderate risk
S12	0.881346	1.997956	3.038928	43.73748	282.7423	19.90128	352.2993	Very high risk
S13	1.872337	5.560771	20.87269	33.88869	305.2379	10.0019	377.4343	Very high risk
S14	1.055059	6.789283	11.30735	47.19896	418.2094	13.01192	497.5719	Very high risk
S15	2.497315	4.371233	7.046637	31.84236	281.4128	3.782345	330.9527	Very high risk

Table 7. Indices and grades of potential ecological risk of heavy metal pollution (Luo *et al.,* 2007).

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

Ecological risk assessment

In present experiment, the enrichment factor, contamination factor, geoaccumulation index, and pollution load index (PLI) were used as ecological risk assessment to determine toxic metal pollution in industrial vicinity soils (Table 6)

The enrichment factor values for the studied soils are presented in Figure 4. Average enrichment factor index of toxic elements assume enrichment of these metals in different sampling locations in the industrial vicinity of Tangail district, Bangladesh. For enrichment factors, cadmium and arsenic have the highest enrichment factor value which indicate soil contamination for total sampling locations. Enrichment factor for studied heavy metals showed a decreasing order of Cd > As > Pb > Cu >Ni > Cr in all sampling locations. Usually, a little enrichment values causes high contribution for crusted source in soils which were identified by several studies where anthropogenic sources have substantial contribution causes high EFs (Islam *et al.,* 2015b; Rashed, 2010).

Four types of contamination Factors (CF), four types of degree of contamination (Cd), five types of Eir and four types of PER were given by Hakanson (Hakanson, 1980) presented in Table 7. The contamination factor (CF) for individual metal were presented in Figure 5. In the studied vicinity, contamination factor was low and was considerable degree only for Cd.

Geoaccumulation index (I_geo) values were shown in Figure 6. The I_geo values presented the decreasing order of Cd>Pb>As>Cu>Ni>Cr. Average Igeo values for the studied toxic metals for studied locations causing slowly contamination of soil 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 (Islam *et al.,* 2015b; Proshad *et al.,* 2017). Present studied soils were polluted by Cd and it was observed for others metals that PLI values was less than one for all sampling sites (Figure 7).

PER index of single metal (E_r^i) with combining potential ecological risk index of the environment (PER) (Table 6) with classifications of PER (Table 7), studied area soil samples indicate the low to very high risk which must possess ecological hazard in the studied vicinity. For individual metal ecological risk assessment, cadmium showed the highest risk and the studied vicinity soils resulted from moderate, considerable and very high potential ecological risk due to combining toxic metal effects. The order of E_r^i for studied soil sample followed decreasing order of Cd> As> Pb> Cu> Ni>Cr. Cd contributes significantly higher than other metals as potential ecological risk index of the environment (PER) which can be due to the effect from anthropogenic activities such as the application of phosphate fertilizers and industrial activities (Martín *et al.,* 2013).

Sum of toxic units (ΣTUs) determine as possible heavy metal toxicity in soils (Figure 8). Toxic units may be calculated as the ratio of heavy metal concentration in soil which is measured to probable effect levels (PELs) (Islam *et al.,* 2015a). Total toxic unit
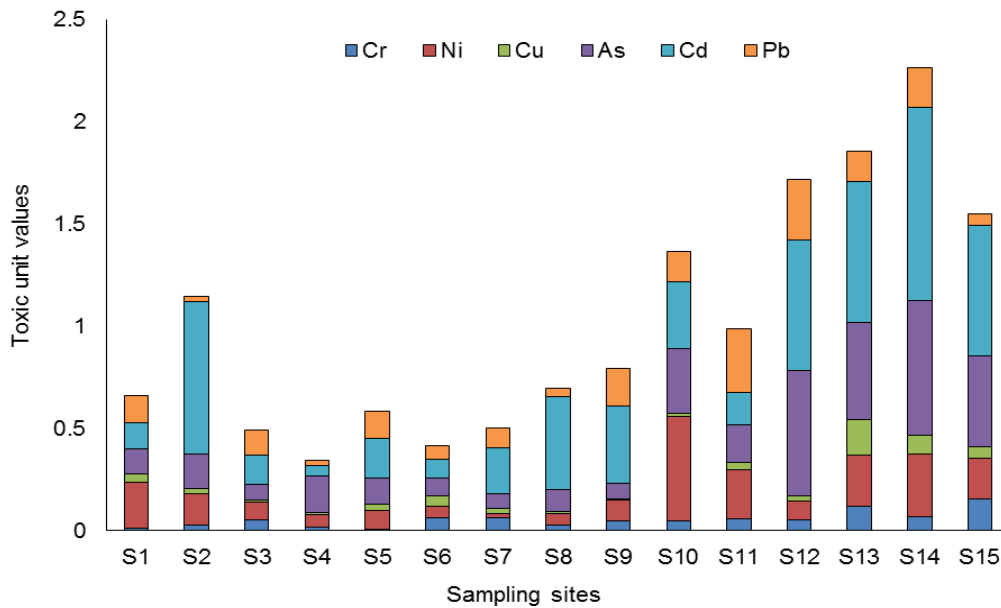
Figure 8. *Estimated sum of the toxic unit (TU) in soils of industrial areas of Tangail district, Bangladesh.*

(ΣTUs) with toxic units (TU) due to heavy metals toxicity in several soil sampling locations in industrial vicinity were presented in Figure 6. Moderate to serious toxicity of hazardous materials were resulted when sum of toxic units of studied soil samples exceed 4 and it causes serious threat to environment. Total toxic units (ΣTUs) for different sampling sites like S12, S13 and S14 were higher than other sites. In the present study, no sample was found which sum of toxic units was higher than 4.

Conclusion

Present study indicated that cadmium have the most hazardous effect to soil samples and soils were polluted by Cd (about 70% soil samples exceed the Dutch soil quality target value). Contamination factors 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 vicinity soils for Bangladesh varied in different locations. Geogenic with anthropogenic elements are the major reasons for enrichment of toxic metals in soils. Around 66% samples were polluted according to potential ecological risk (moderate to very high risk). Maximum sampling sites in the industrial vicinity of Tangail district showed cadmium toxicity with severe ecological risk for single toxic element. So in Tangail district, ecological risk indexes for toxic elements were so much high. There is urgent need to study again in present studied area and to increase public awareness not to throw industrial wastages in the open environment.

ACKNOWLEDGMENTS

Conflict of interest

No any conflict of interest was declared by the authors.

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