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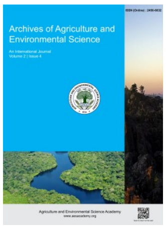

ORIGINAL RESEARCH ARTICLE

CrossMark

Chronic exposure assessment of toxic elements from agricultural soils around the industrial areas of Tangail district, Bangladesh

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ABSTRACT

The present research was conducted to evaluate the potential ecological and human health risk of toxic elements (Cr, Ni, Cu, As, Cd and Pb) from agricultural soils around the industrial areas of Tangail district in Bangladesh. Potential ecological and human health risk were assessed through enrichment factor (EF), contamination factor (C^i_f), geoaccumulation index (I_{geo}), pollution load index (PLI), toxic unit analysis, exposure pathway, hazard quotient and hazard index. The mean concentration of Cr, Ni, Cu, As, Cd and Pb in the studied soils were 5.88, 13.92, 18.07, 5.90, 2.19 and 8.08 mg/kg, respectively. The mean values of enrichment factor, geoaccumulation index, contamination factor, pollution load index and toxic units were found low for all metals excluding Cd. Considering the severity of potential ecological risk factor for single metal (E^i_r), the descending order of pollutants was Cd > As > Cu > Ni > Pb > Cr. In the perspective of potential ecological risk (PER), soils from all sampling sites indicated moderate to very high PER. Chronic daily intake values were higher in children than the adult for both ingestion and dermal contact as body weight of children was lower than the adult. The non-cancer health risks related to individual element exposure through soil ingestion, dermal contact and inhalation was low for all investigated elements resulted in a HQ < 1, indicating low risk for both adults and children. Considering the total exposure of hazard index of ingestion, dermal contact and inhalation, there was no chance of having non-cancer risk for the inhabitants of the studied industrial area. Carcinogenic risks for both adult and children lying between an acceptable ranges.

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INTRODUCTION

Soil contamination by toxic elements is considered as the most adverse environmental issue in the world (Islam *et al*., 2015a, 2018; Proshad *et al*., 2017a). Soil is a vital component for human life to survive on the earth which is anticipated as principal receiver of persistent pollutants such as toxic trace elements (Luo *et al*., 2007; Karim *et al*., 2014; Islam *et al*., 2015b). Heavy metal pollutions in soils are of great concern due to their wide sources, toxicity, non-biodegradable nature and toxicity to human and other organisms (Yuan *et al*., 2011; Zhao *et al*., 2014; Islam *et al*., 2015a, 2018; Bhuyan and Bakar, 2017; Bhuyan *et al*., 2017). In the last few decades, there has been a significant concern regarding soil contamination by various trace elements due to rapid industrialization and development, especially in developing countries like Bangladesh (Khan *et al*., 2008; Chen *et al*., 2010; Sun *et al*., 2010; Islam *et al*., 2015a, 2018). In the industrial areas, trace elements may originate in soils from numerous activities such as emissions from vehicular exhaust, generation of power, manufacturing, burning of fossil fuel, wastewater irrigation and disposal of waste (Rodríguez *et al*., 2014; Islam *et al*., 2016, Proshad *et al*., 2018a). Hazardous elements toxicity changes surface soil physical, chemical, and biological features that have a significant negative consequence on the productivity of land (Khan *et al*., 2010).

The contaminations of heavy metals in soil have exerted long-term ecological and health effects (Needleman, 1980; Mclaughlin *et al*., 1999). Crops which are being cultivated in the contaminated agricultural soils may cause serious carcinogenic and non-carcinogenic risks to the human body (Man *et al*., 2010; Proshad *et al*., 2018b). Heavy metals (chromium, copper, cadmium and lead) and metalloid (arsenic) are of particular concern because of very well-known detrimental health effects on humans in excessive quantities (Shaheen *et al*., 2016, Islam *et al*., 2018). In the industrial areas, heavy metal polluted soil can pose significant human health risks due to soil ingestion, inhalation of volatiles and fugitive soil particulates, and dermal contact, especially in the public parks and playgrounds (Siciliano *et al*., 2009; Luo *et al*., 2011; Okorie *et al*., 2011; Li *et al*., 2011). Therefore, exposure to heavy metal pollutants is of utmost concern for children in their primary developmental years and also for the adult (Lee *et al*., 2013; Rachwał *et al*., 2017). However, research on possible health risk due to heavy metals pollution in soil of the industrial area is very essential. Tangail district is an industrialized area of Bangladesh that is supposed to be highly contaminated by heavy metals. This area is well-known for agricultural production and it provides a large portion of agricultural products all over the country (Huq and Shoaib, 2013). Although several studies have conducted for assessing human health risk due to heavy metal contamination from soil in the urban and industrial regions of the world (Chen *et al*., 2005, Luo *et al*., 2007, Man *et al*., 2010, Proshad *et al*., 2017b), but there is very limited research has been conducted so far on heavy metals in soil and its adverse effects on the environment as well as human health especially the industrial area like Tangail district. Therefore, the present research was conducted to address the following questions: i) what are the concentration of heavy metals in soils of the studied industrial area? ii) Is the concentration of heavy metals is alarming for our environment? iii) Is it possesses potential health risk?

MATERIALS AND METHODS

Study area and sampling

This study was conducted in Bangladesh Small and Cottage Industries Corporation (BSCIC) areas of Tangail district, Bangladesh (Figure 1). It is one of the densely populated (1,100/square Km) district of the country having an area of 334.26 Km^2. Tangail Sadar Upazila is one of the most densely polluted area in Bangladesh where the density of population is 1,100/Km^2 (2011 census) (BBS, 2011). The study area is situated between at 24.20° N to 89.58° E. Tangail is an industrial growing site of Bangladesh, which is highly susceptible to environmental pollution over the last decade (Proshad *et al*., 2018c). There are several types of industrial units including garments, tannery industries packaging industry, dyeing, brick kiln, metal workshops, battery manufacturing industries, tanneries, textile industries, pesticide and fertilizer industries, different food processing industries and other factories of BSCIC industrial areas produce huge volumes of effluents that contain trace metals. The untreated wastes and effluents from these industries are discharged randomly to river and canals. Then that wastes are mixed with soils and the soil is continuously polluted by heavy metals.

Soil samples were collected during March-April, 2016. Ten agricultural soil sampling locations were selected in the industrial areas of Tangail district. Agricultural soil samples (up to 10 cm) were collected in the form of three subsamples. These sub-samples were thoroughly mixed to form a composite sample. Samples were air-dried at room temperature for two weeks, then ground and homogenized. The dried soil samples were crumbled with a porcelain mortar and pestle and sieved through 2 mm nylon sieve and stored in an airtight clean Ziploc bag and kept frozen until chemical analysis (Oliveira *et al*., 2012; Arenas-Lago *et al*., 2013, 2014).

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.

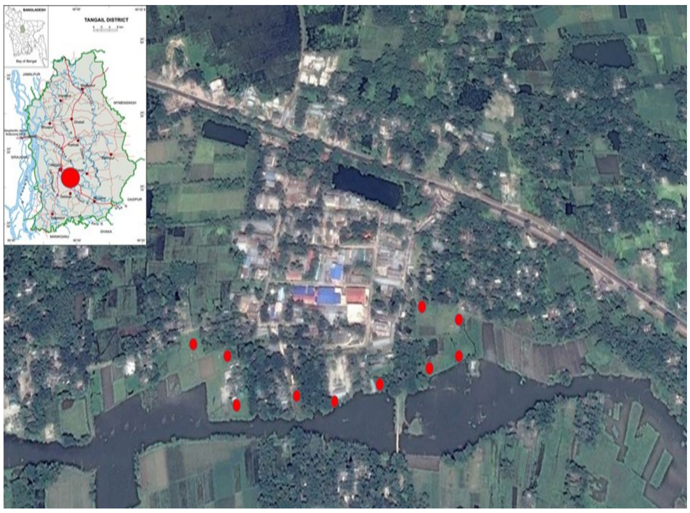

Figure 1. *Map of the sampling sites of industrial areas in Tangail district, Bangladesh (red circle indicate sampling locations).*

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 soil) × 100 (1)

%(Clay) = (Corrected hydrometer reading after 2 hours/ Oven dry weight of soil) × 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 were 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 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.

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-Uria *et al*., 2009). The EF for each element was calculated to evaluate anthropogenic influences on heavy metals in soils using the following formula (Selvaraj *et al*., 2004):

$$EF = (C_M/C_{Al})_{Sample} / (C_M/C_{Al})_{Background} \quad (5)$$

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^i_f)

Contamination factor means the proportion of the heavy metal concentration in the soil to that of baseline or background value:

$$C^i_f = C_{Heavy\ metal} / C_{Background} \quad (6)$$

Contamination factor divided into four classes ranged from 1 to 6 which are: low degree (C^i_f <1), moderate degree ($1 \le C^i_f < 3$), considerable degree ($3 \le C^i_f < 6$), and very high degree ($C^i_f \ge 6$) (Islam *et al*., 2015c). This approach has been used by other researchers (e.g. Proshad *et al*., 2017a).

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 (2010). The PLI is defined as the n^{th} root of the multiplications of the contamination factor (C^i_f) of metals (Bhuiyan *et al*., 2011).

$$PLI = (C_f{}^i{}_1 \times C^i{}_{f\,2} \times C_f{}^i{}_3 \times ...... \times C_f{}^i{}_n)^{1/n} \quad (7)$$

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. 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. Proposed equations which were used to calculate PER and are as follows (Luo *et al*., 2007; Guo *et al*., 2010).

$$C_f^i = \frac{C^i}{C_n^i}, \quad C_d = \sum_{i=1}^{n} C_f^i \quad (8)$$

$$E_r^i = T_r^i \times C_f^i, \quad PER = \sum_{i=1}^{m} E_r^i \quad (9)$$

Where, C^i_f is the single element contamination factor, C^i is the content of the element in samples and C^i_n 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^i_f for all metals represent the integrated pollution degree (C_d) of the environment. C^i_r is the potential ecological risk index and $T^i_r$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 (Håkanson, 1980; Luo *et al*., 2007; Wu *et al*., 2010; Guo *et al*., 2010; Jintao *et al*., 2011; Amuno, 2013). *PER* is the comprehensive potential ecological risk index, which is the sum of E^i_r. 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 sum of toxic units (ΣTUs) is considered as potential acute toxicity of hazardous elements in agricultural soil samples. Toxic unit analysis is stated as the ratio of the assessed concentration of hazardous elements in soil to probable effect level (PELs) (Zheng *et al*., 2008). A moderate to serious toxicity of hazardous elements remain in soil when the sum of toxic units for all soil samples is more than 4 (Bai *et al*., 2011).

Health risk assessment from polluted soil

Daily intake of heavy metals through exposure pathway from soil

Ingestion and dermal absorption of heavy metals from polluted agricultural soils have great importance in potential exposure pathways (Fryer *et al*., 2006; Qu *et al*., 2012). Out of several exposure pathways, ingestion of metals from soil is the most common exposure pathway for Cr, Ni, Cu, As, Cd and Pb (Ordóñez *et al*., 2011). Chronic daily intake (CDI) (mg/kg/day) of metals was determined from ingestion ($CDI_{ingest\text{-}soil}$) and dermal contact ($CDI_{dermal\text{-}soil}$) in the present study for both adult and children were estimated using the following formulas:

Ingestion from soil: $CDI_{ingest\text{-}soil} = \frac{CS \times IRS \times EF \times ED}{BW \times AT} \times CF$ (10)

Dermal contact from soil: $CDI_{dermal\text{-}soil}$=

$$\frac{CS \times SA \times AF \times ABS \times IRS \times EF \times ED}{BW \times AT} \times CF \quad (11)$$

Inhalation from soil: $CDI_{inhalation\text{-}soil}$=

$$\frac{CS \times InhR \times EF \times ED}{BW \times AT} \times CF \quad (12)$$

Where, CDI = chronic daily intake; CS — exposure-point concentration: mg/kg; IRS—ingestion rate: 100 and 200 mgd^{-1}for adult and children (USEPA, 2011); EF — exposure frequency: 350 d/a (USEPA, 2011); ED — exposure duration: 30 years for adult, 6 years for children (USEPA, 2011); CF—units conversion factor: 10^{-6} kg mg^{-1} (USEPA, 2002); SA — exposure skin area: 5700 and 1600 cm^2 for adult and children (USEPA, 2011); AF — adherence factor: 0.07 and 0.02 mg·cm^{-2} for adult and children (USEPA, 2011); ABS —dermal absorption fraction: 0.01 for adult and 0.001 for children (USEPA, 2011); BW — body weight: 70 kg for adult, 15 kg for children (USEPA, 2001); AT — averaging time for non-carcinogens: 365 × ED (USEPA, 2002); InhR— Inhalation rate 20 m3/d for both adult and child (USEPA, 1997).

Hazard Quotient (HQ)

The non-carcinogenic risks for each individual heavy metal (Cr, Ni, Cu, As, Cd and Pb) through ingestion, dermal and inhalation were assessed by the target hazard quotient (THQ) (USEPA, 1989). The methodology for the estimation of non-carcinogenic risks was applied in accordance with that provided by the U.S. Environmental Protection Agency (USEPA) Region III's risk-based concentration table (USEPA, 2011). Hazard quotient (HQ) was determined on the basis of chronic daily intake from ingestion (CDI_{ingest}) dermal (CDI_{dermal}) and inhalation ($CDI_{inhalation}$), it was calculated by dividing the average daily dose to a specific reference dose (RfD) (USEPA, 1989). The equation used for estimating the target hazard quotient is as follows:

$$HQ_{ingest} = (CDI_{ingest})/RfD \quad (13)$$

$$HQ_{dermal} = (CDI_{dermal})/RfD \quad (14)$$

$$HQ_{inhalation} = (CDI_{inhalation})/RfD \quad (15)$$

Where, THQ is the target hazard quotient, CDI is the chronic daily intake of heavy metal (mg/kg) and RfD is reference dose (mg/kg/day). The RfD for Cr, Ni, Cu, As, Cd and Pb were 0.003, 0.02, 0.04, 0.0003, 0.0005 and 0.0035 mg/kg/day, respectively (USDOE, 2011; USEPA, 2002). The reference dose (RfD) (mg/kg/day) is an estimation of maximum permissible risk on human population through daily exposure, taking into consideration sensitive group (children) during the lifetime. If the CDI is higher than RfD (HQ>1), there will be a severe health hazard to human, whereas CDI is less than RfD (HQ ≤ 1), there will be no severe human health effects (USEPA, 1989; USEPA, 2001). The health risk guidelines determination of chemical mixtures defined that "simultaneous sub-threshold exposures to several chemicals may result in an adverse health effect" and "the magnitude of the adverse effect will be proportional to the sum of the ratios of the sub-threshold exposures to acceptable exposures" (USEPA, 1986). Again, hazard index (HI) can be generated from the hazard quotient to calculate the combined risk of individual heavy metals in the form of mix contaminates (USEPA, 1989).

Hazard Index (HI)

In order to assess the overall potential for non-carcinogenic effects from more than one heavy metal, a hazard index (HI) has been formulated based on the guidelines for health risk assessment of chemical mixtures (USEPA, 1999). The hazard index (HI) from THQs is expressed as the sum of the hazard quotients (USEPA, 2011). The equation used for estimating the hazard index is as follows:

$$HI=\Sigma TTHQ_n \tag{16}$$

$$HI=TTHQ_{\text{element 1}} + TTHQ_{\text{element 2}} + \ldots\ldots\ldots + TTHQ_{\text{elements n}} \tag{17}$$

$$HI= \Sigma TTHQ= Hi_{ingest} + HQ_{dermal} + HQ_{inhalation} \tag{18}$$

The guidelines also state that any single metal with an exposure level greater than the toxicity value will cause the hazard index to exceed unity, for multiple metal exposures the HI can also exceed unity even if no single metal exposure exceeds its RfD.

Carcinogenic risk

Carcinogenic risk is considered as the probability of an individual developing any type of cancer in the whole lifetime due to exposure to carcinogenic hazards (Li *et al.*, 2014). Carcinogenic risk expressed as the total cancer risk Eq. (22).

$$CR_{ingest\text{-}soilt}=\{(CS \times AF\times IngR\times EF\times ED)/(BW\times AT)\}\times CF\times CSF_{ingest} \tag{19}$$

$$CR_{dermal\text{-}soil}=\{(CS\times SA\times AF\times ABS_d\times EF\times ED)/(BW\times AT)\}\times CF \times CSF_{ingest} \times ABS_{GI} \tag{20}$$

$$CR_{inhalation\text{-}soil}=\{(CS\times ET\times EF\times ED)/(PEF\times 24\times AT)\}\times IUR\times 10^3 \tag{21}$$

$$\text{Total cancer risk}=\Sigma\text{Cancer risk} =Risk_{ingestion} +Risk_{dermal}+ Risk_{inhalation} \tag{22}$$

Where, $CR_{ingest\text{-}soil}$— cancer risk of metals from ingestion of soil $CR_{dermal\text{-}soil}$— cancer risk of metals from dermal contact of soil; CS — heavy metal concentration in soil: mg/kg; AF — soil -to-skin adherence factor: 0.7 mg/cm^2 for adult and 0.2 mg/cm^2 for children (USEPA, 2011); IngR—ingestion rate of soil: 100 and 200 mgd−1 for adult and children respectively (USEPA, 2011); EF — exposure frequency: 350 days/year (USEPA, 2011); ED — exposure duration: 30 years for adult and 6 years for children (USEPA, 2011); BW — body weight: 70 kg for adult and 15 kg for children; AT — averaging time for non-carcinogens: 365 × ED (USEPA, 2011); CF—units conversion factor: 10^{-6} kg/mg (USEPA, 2002); CSF_{ingest}—Chronic oral slope factor: 1.5 for As and 8.5×10^{-3} for Pb (USEPA, 2010; USDOE, 2011); SA — exposure skin surface area available for contact: 5700 cm^2 for adult and 1600 cm^2 for children (USEPA, 2011); ABS_d — dermal absorption fraction: 0.01 for adult and 0.001 for children (USEPA, 2011); ET— Exposure time: 1 for residents for the site specific (USDOE, 2011); ABS_{GI} —Gastrointestinal absorption factor: 0.41 and 1 for As and Pb respectively (USEPA, 2011); PEF—Particle emission factor: 1.36 $\times10^9$ (USDOE, 2011; USEPA, 2011); IUR—Chronic inhalation unit risk: 4.30×10^{-3} for As, 1.20×10^{-5} for adult (USDOE, 2011).

In present study, we calculated carcinogenic risk for arsenic and lead as they are classified as probably carcinogenic to humans (ASTDR, 2007; ATSDR, 2012). The excess cancer risks lower than 10^{-6} (a probability of 1 chance in 1,000,000 of an individual developing cancer) are considered to be negligible, cancer risks above 10^{-4} are considered unacceptable by most international regulatory agencies (USEPA, 1989; Guney *et al.*, 2010) and risks lying between 10^{-6} and 10^{-4} are generally considered an acceptable range, depending on the situation and circumstances of exposure (Hu *et al.*, 2012). The value 10^{-6} is also considered the carcinogenic target risk (USEPA, 2011).

Statistical analysis

The data were statistically analysed using the statistical package, SPSS 20.0 (SPSS, USA). The means of the hazardous element concentrations in soils were calculated. Other calculations were performed by Microsoft Excel 2013.

RESULTS AND DISCUSSION

Heavy metals pollution in agricultural soils of industrial area is a great concern and affects soil health. Polluted soils in the industrial areas are greatly responsible for environmental pollution with human health inferences. Heavy metals are too toxic to affect soil health as well as human health. Crop production may be affected by the presence of heavy metals in soils, their storage in soil and transformation. Heavy metals affect human, animal and plant health (VROM, 2000). The concentration of heavy metals for present the study was lower than the Dutch standard (VROM, 2000), Australian guidelines (DEP, 2003) and

Canadian guidelines (CCME, 2003) except cadmium. Cadmium concentration for the present study was higher than the Dutch standard (VROM, 2000) and Canadian guidelines (CCME, 2003). Environmental action level demonstrates that the low risk to environment and human health.

Physicochemical properties and heavy metals concentration in soils

The studied soils pH values were ranged from 5.58 to 6.67 indicating that soils were slightly acidic (Table 1). 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 (Oliver, 1997; Adeniyi *et al.*, 2008). 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). The range of organic carbon (% C) was 0.504 to 4.310, where the highest value was observed in soil collected from the S10 site and lowest value observed in S1 site. 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 (Soil Survey Division Staff, 1993), the textural analysis revealed that the studied soil samples were loam (Table 1).

The mean concentrations of Cr, Ni, Cu, As, Cd, and Pb in agricultural soils were found 5.88, 13.92, 18.07, 5.9, 2.19, and 8.08 mg/kg, respectively (Table 2) around the industrial vicinity of Tangail district, Bangladesh. The highest Cr concentration was observed at 13.41 and 10.95 mg/kg at S_1 and S_4 sampling sites in the present study. A considerable amount of Cr was observed in soil collected from the agricultural field near industrial areas of Tangail district which might be due to the use of tannery waste for the supplement of organic matter for crop production. Agricultural field may receive Cr from the unplanned activities of tannery industries in Tangail City. The mean concentration of Cr was found 5.88 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 3). 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 (Srinivasa *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 (Tokalıoğlu and Kartal, 2006; Bhagure and Mirgane, 2011; Acosta *et al.*, 2011; Islam *et al.*, 2014; Islam *et al.*, 2015a, 2017; Proshad *et al.*, 2018b) 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 (Panda and Patra, 2000; Panda, 2007; Yu *et al.*, 2008; Shaker *et al.*, 2009; Hasnine *et al.*, 2017).

Nickel can cause dermatitis, lung fibrosis, cardiovascular and kidney diseases and cancer of the respiratory tract in the human body (Hasnine *et al.*, 2017). The solubility of nickel in soils increases with its acidity and if the acidity increases it results higher Ni in soils (Baralkiewicz and Siepak, 1999). In the present study Ni concentrations ranged between 3.01-25.92 mg/kg in the study area. The highest amount (25.92 mg/kg) was found in station 1 and the lowest value (3.01 mg/kg) in station 9 (Table 2). The elevated levels of Ni were found in station 1 which results from localized additions or accidental spillages of Ni containing materials (Krishna and Govil, 2007). The mean concentration of Ni was found 13.92 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 (Tokalıoğlu and Kartal, 2006; Bhagure and Mirgane, 2011; Acosta *et al.*, 2011; Islam *et al.*, 2014; Islam *et al.*, 2015a, 2017; Proshad *et al.*, 2018b) conducted different areas in Bangladesh and other countries. USPHS (1997), Alloway (1990) reported that the typical concentration of Ni in soil is 50 mg/kg. Hasnine *et al.* (2017) reported average Ni concentration in the surface agricultural soil at DEPZA was found to be 655.53 ± 979.73 mg/kg. Dojlido and Best (1993) found approximately 26,000 mg/kg Ni of highly developed nickel smelting in Canada. 250 mg/kg Ni was determined in a highly polluted area contaminated by galvanization plant sewage (Dojlido and Best, 1993). The concentration of Ni in the agricultural soils of Ontario varied between 1.3 to 6,560 mg/kg (Frank *et al.*, 1976).

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 3.86 to 78.11 mg/kg (Table 2). The elevated concentration of Cu was observed in soil from waste disposal sites which can be due to the emission of Cu from the uncontrolled industrial and waste burning activities (Kashem and Singh, 1999; Srinivasa *et al.*, 2010; Luo *et al.*, 2011). The mean concentration of Cu was found 18.07 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 (Tokalıoğlu and Kartal, 2006; Bhagure and Mirgane, 2011; Acosta *et al*., 2011; Islam *et al*., 2014; Islam *et al*., 2015a, 2017; Proshad *et al*., 2018b). Frank *et al*. (1976) documented the value of Cu ranged from 2.1 to 664 mg/kg in agricultural soils of Ontario. Sonmez *et al*. (2006) reported decrease height in plant, total yield, number of fruit, and dry root weight with increasing Cu application. Yu *et al*. (2008) found 17.10 mg/kg Cu in arid agricultural soil in central Gansu Province, China. The threshold value for Cu is ≤ 60 mg/kg for arid agricultural soils in China (NEPA, 1995). Hasnine *et al*. (2017) reported average Cu concentration in the surface agricultural soil at DEPZA was found to be 91.06 ± 152.70 mg/kg.

In the present study, the concentration of As varied between 1.56 to 28.30 mg/kg (Table 2). A huge amount of groundwater containing As (Neumann *et al*., 2010; Hug *et al*., 2011) is being used for tanning in relation to some chemicals especially arsenic sulfide (Asaduzzaman *et al*., 2002; 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 (Renner, 2004; Neumann *et al*., 2011). All the concentrations of As found to below the recommended value set by The Dutch Soil Quality Standard (VROM, 2000) (Table 2). Present studied As concentrations were lower than other studies (Proshad *et al*., 2017a; Islam *et al*., 2014, 2015a, 2017). Frank *et al*. (1976) estimated 6.21± 2.67 mg/kg As in agricultural soils of Ontario while Yu *et al*. (2008) recorded 8.80 mg/kg As in arid agricultural soil in central Gansu Province, China. The threshold value for As is ≤20 mg/kg for arid agricultural soils in China (NEPA, 1995). As contaminated water and As-enriched fertilizers as well as pesticides were used for irrigation in the agricultural land (Alam *et al*., 2003; 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 0.36 to 7.53 mg/kg. The mean concentration of Cd was found 2.19 mg/kg in the present study which was lower than The Dutch Soil Quality Standard (VROM, 2000) and Canadian Environmental Quality

Table 1. Physiochemical properties of soil collected from agricultural field in the industrial areas of Tangail district, Bangladesh.

Sampling sites	pH (1:2.5 H_2O)	EC(dS/m)	Organic carbon (%)	Sand (% in <2 mm)	Silt	Clay	Soil type[a]
S_1	6.62	0.08	0.504	37.6	46.6	15.8	Loam
S_2	5.58	0.15	0.506	34.9	47.5	17.6	Loam
S_3	6.11	0.12	0.506	44.7	40	15.3	Loam
S_4	5.82	0.33	0.522	36.5	45	18.5	Loam
S_5	6.87	0.15	2.582	37.6	44.1	18.3	Loam
S_6	6.38	0.13	0.578	31.5	46.6	21.9	Loam
S_7	6.38	0.21	0.746	42.2	37.5	20.3	Loam
S_8	6.54	0.08	0.750	37.5	47.4	15.1	Loam
S_9	6.24	0.11	0.820	41.5	41.6	16.9	Loam
S_{10}	6.2	0.09	4.310	43.5	44.1	12.4	Loam

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

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

Sampling sites	Cr	Ni	Cu	As	Cd	Pb
S_1	13.41	25.92	2.91	2.64	2.53	2.18
S_2	6.05	9.40	78.11	28.30	7.53	17.93
S_3	9.40	27.69	5.13	2.32	1.15	8.54
S_4	10.95	18.95	26.64	13.22	3.05	18.32
S_5	1.67	7.09	3.86	2.48	0.88	3.82
S_6	1.09	3.35	8.66	2.38	3.58	7.37
S_7	1.93	3.47	21.54	1.56	0.36	6.65
S_8	7.04	26.77	19.21	1.59	1.88	10.84
S_9	5.07	3.01	8.66	2.15	0.63	4.03
S_{10}	2.24	13.63	6.03	2.38	0.37	1.19
Mean	5.88	13.92	18.07	5.90	2.19	8.08
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

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

Guidelines (CCME, 2003) but higher than Australian Guideline for Soil Quality (DEP, 2003). Cd pollution has been reported from areas surrounding smelters in many countries (Martley *et al.*, 2004; Rawlins *et al.*, 2006). 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 (Tokalıoğlu and Kartal, 2006; Bhagure and Mirgane, 2011; Acosta *et al.*, 2011; Islam *et al.*, 2014; Islam *et al.*, 2015a, 2017; Proshad *et al.*, 2018b). Frank *et al.*, (1976) documented 0.5±0.69 mg/kg Cd in agricultural soils of Ontario. 0.5±0.69. The soil is considered clean if any heavy metal concentration in soil is below its respective Dutch Target Value. The soil is regarded to be slightly to moderately contaminated if the concentration level lies between the target values and intervention values. In contrast, if the value is above the Dutch Intervention Value, the soil is considered detrimental to humans, plants, and animals. About 70% of the studied soil samples exceeded the Dutch target value assuming that Cd in soil might pose a severe risk to the surrounding ecosystems.

The highest concentration of Pb was 18.32 mg/kg found on station 4. This level of Pb concentration present in soil due to metal processing factories release Pb into the open environment and several anthropogenic factors (Karim *et al.*, 2008; Nziguheba and Smolders, 2008). In the present study, station 4 showed the elevated concentrations of Pb which can be due to the emission of Pb contaminated waste from these sites (Srinivasa *et al.*, 2010). The mean concentration of Pb was found 8.08 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 Pb in soil (Table 2). Lead (Pb) concentration in the present study was lower than other studies (Tokalıoğlu and Kartal, 2006; Bhagure and Mirgane, 2011; Acosta *et al.*, 2011; Islam *et al.*, 2014; Islam *et al.*, 2015a, 2017; Proshad *et al.*, 2018b) conducted different areas in Bangladesh and other countries. Yu *et al.*, (2008) recorded 23.30 mg/kg Pb in arid agricultural soil in central Gansu Province, China. The threshold value for Pb is ≤50 mg/kg for arid agricultural soils in China (NEPA, 1995). Frank *et al.* (1976) recorded value for Pb that ranged between 1.5 to 888 mg/kg in agricultural soils of Ontario.

Correlation coefficient matrix for physicochemical properties of soil and heavy metals

The results highlighted close association among correlation coefficient matrix for physiochemical properties of soil and heavy metals collected from industrial vicinity of Tangail district (Table 4). The value of pH showed significant negative correlation with Cu (r = -0.73*), As (r = -0.78*) and Pb (r = -0.72*). Electrical conductivity, organic carbon, clay, nickel and cadmium didn't show any significant positive and negative correlations. Sand showed a significant negative correlations with silt (r = -0.75*) and cadmium (r = -0.69*). There were also showed others positive correlations like silt with Cd (r = 0.63*), Cr with Ni (r = 0.78*), Cu with As (r = 0.93**), Cu with Cd (r = 0.83**), Cu with Pb (r = 0.77**), As with Cd (r = 0.88**), As with Pb (r = 0.77**). 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.

Ecological risk assessment

Ecological risk assessment for heavy metals contamination in soil was performed following the methodology developed by

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

District (Country)	Cr	Ni	Cu	As	Cd	Pb	References
Tangail, Bangladesh	5.88	13.93	18.08	5.9	2.2	8.09	Present study*
Tangail, Bangladesh	10.41	12.69	15.66	12.15	3.1	7.98	Proshad *et al.*, 2017a
Tangail, Bangladesh	8.31	16.49	20.64	5.06	2.2	16.9	Proshad *et al.*, 2018b
Dhaka, Bangladesh	158–1160	104–443	157–519	41–93	3.9–13	84–574	Islam *et al.*, 2014
Dhaka, Bangladesh	2.4–1258	8.3–1044	9.7–823	8.7–277	1.8–80	13–842	Islam *et al.*, 2017
Bogra (Bangladesh)	6.3–256	8.3–271	13–279	7.5–87	0.09–29	5.3–624	Islam *et al.*, 2015a
Maharashtra (India)	164	171	155	2.8	30	42	Bhagure and Mirgane, 2011
Murcia (Spain)	18	14	11	NA	0.22	49	Acosta *et al.*, 2011
Kayseri (Turkey)	29	45	37	NA	2.5	75	Tokalıoğlu and Kartal, 2006
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

Hakanson (1980). In the present study, enrichment factor (EF), contamination factor (CF), degree of contaminations (C_d), pollution load index (PLI), potential ecological risk (PER) and toxic units have been applied to assess the contamination of heavy metals in soil of Tangail district.

For all sampling sites, enrichment factors of Cr, Ni and Pb in soils were less than 1.5 (Figure 2, 3). About 10% of soil samples for Cu and As and 40% of Cd were higher than 1.5 indicating strong human influence from industrial pollution (Rashed, 2010). This research addressed that crusted source to the soil was the main reason of low enrichment of heavy metals and great contribution from anthropogenic sources resulting from high enrichment factors in soils (Rashed, 2010). The mean enrichment factors of Cr, Ni, Cu, As, Cd and Pb were 0.113, 0,244, 0.375, 0.509, 1.503 and 0.197 respectively. Here only Cd exceeds the standard value of enrichment and Cd have strong human influence from industrial contamination on soils.

Contamination factors of heavy metals for the present study were presented in Table 5. Present study indicates four types of contamination factors (C^i_f) and four types of degree of contamination (Cd) (Håkanson, 1980). The contamination factors (C^i_f) and four types of degree of contamination (Cd) were presented in Table 6. The contamination level for the present study was found low to considerable indicating low to considerable contamination of heavy metals in soil. According to the contamination factor, Cr, Ni, and Pb showed low contamination. Cu and As showed low to moderate contamination. Only Cd showed low to considerable contamination (Table 5). In the present study, contamination factor values (C^i_f) existed in the decreasing order of Cd > As > Cu > Ni > Pb > Cr in soils of different sampling sites in Tangail district.

Pollution load index (PLI) value equal to zero indicates non-polluted; value of unity indicates the presence of only baseline level of pollutants and values above unity indicates progressive deterioration due to trace element pollution (Rashed, 2010; Suresh *et al.*, 2011). The extent of pollution increases with the increase of numerical PLI value. According to above grade, only cadmium (Cd) exceeds the standard value (Figure 4). Other metals showed less pollution load index indicating low contamination. The main reason for high cadmium pollution may be waste from different industries in the agricultural soil, tannery and dyeing industry had caused some extent risk of the studied area (Bhuiyan *et al.*, 2010). The pollution load index values of the present study were in the decreasing order of Cd > As > Cu > Ni > Pb > Cr (Figure 4).

Potential ecological risk for the present study was calculated on the basis of five categories of risk index of individual metal (E^i_r) and potential ecological risk index of the environment (PER) (Table 7) with their grade classifications (Luo *et al.*, 2007). Studied area soil samples indicate the moderate 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. Cd contributes significantly to the potential ecological risk index of the environment (PER) which can be due to the effect from anthropogenic activities such as application of phosphate fertilizers and industrial activities (ATSDR, 2008; Mass *et al.*, 2011; Rodríguez Martín *et al.*, 2013). Considering the potential ecological risk factor (E^i_r) for the individual element, Cd showed very high potential ecological risk with the E^i_r factor ranging between 56.73 to 1189.67 (Table 6). The order of E^i_r for studied soil sample followed the decreasing order of Cd > As > Cu > Ni > Pb > Cr. Potential risk for present study ranged from 87.80 to 1422.97.

Potential acute toxicity of hazardous elements in soil samples can be estimated as the sum of toxic units (ΣTUs), considered as ecological risk. Toxic unit determines how much the soils were toxic by the accumulation of heavy metals (Zheng *et al.*, 2008).

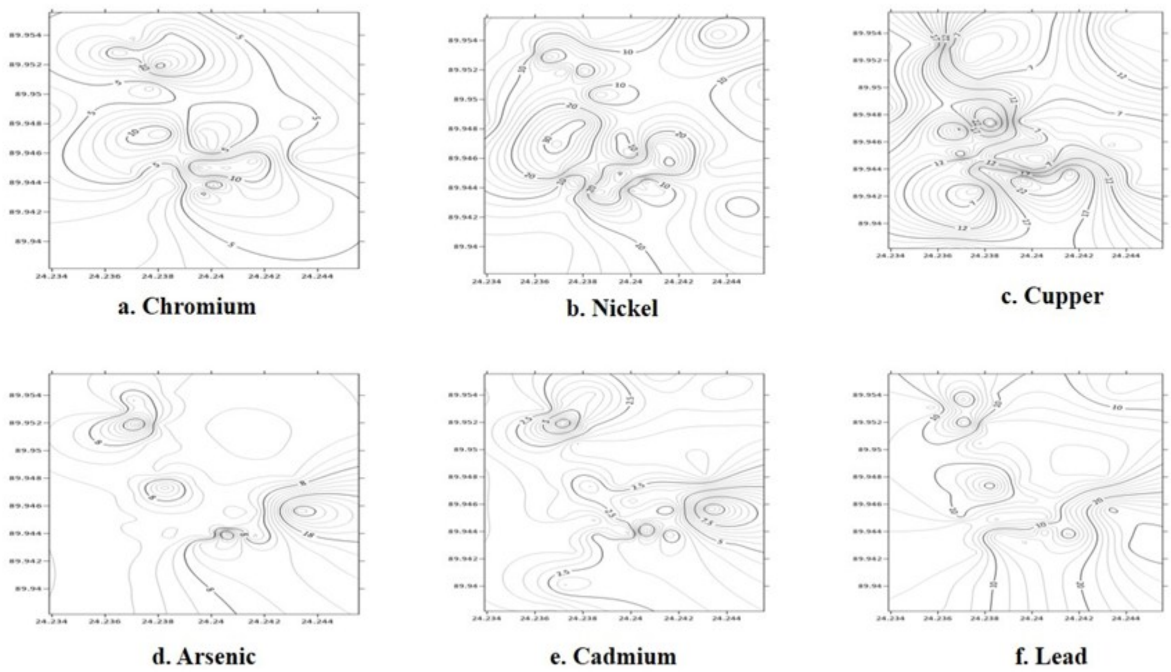

Figure 2. Distribution of heavy metals concentration in the soil samples of the study area.

Table 4. Correlation coefficient matrix for physiochemical properties of soil and heavy metals.

	pH	EC	Organic carbon	Sand	Silt	Clay	Cr	Ni	Cu	As	Cd	Pb
pH	1											
EC	-0.42	1										
Organic carbon	0.24	-0.24	1									
Sand	0.032	-0.16	0.35	1								
Silt	-0.061	-0.204	-0.25	-0.75*	1							
Clay	0.27	0.49	-0.49	-0.58	-0.097	1						
Cr	-0.22	0.105	-0.48	-0.032	0.22	-0.32	1					
Ni	0.007	-0.17	-0.16	0.19	0.24	-0.59	0.78*	1				
Cu	-0.73*	0.28	-0.301	-0.35	0.302	0.16	0.043	-0.18	1			
As	-0.78*	0.36	-0.24	-0.39	0.39	0.109	0.18	-0.077	0.93**	1		
Cd	-0.606	0.14	-0.42	-0.69*	0.63*	0.25	0.21	-0.015	0.83**	0.88**	1	
Pb	-0.72*	0.61	-0.52	-0.41	0.28	0.27	0.29	0.13	0.77**	0.77**	0.705	1

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

Table 5. Contamination factors, degree of contamination and contamination level in soil.

Sites	Contamination factors (C^i_f)						Degree of contamination (C_d)	Contamination level
	Cr	Ni	Cu	As	Cd	Pb		
S_1	0.30	0.66	0.09	0.28	2.66	0.08	4.07	Low
S_2	0.13	0.24	2.37	2.98	7.93	0.66	14.31	Considerable
S_3	0.21	0.71	0.16	0.24	1.21	0.32	2.85	Low
S_4	0.24	0.49	0.81	1.39	3.21	0.68	6.82	Moderate
S_5	0.04	0.18	0.12	0.26	0.93	0.14	1.66	Low
S_6	0.02	0.09	0.26	0.25	3.77	0.27	4.66	Low
S_7	0.04	0.09	0.65	0.16	0.38	0.25	1.57	Low
S_8	0.16	0.69	0.58	0.17	1.98	0.40	3.97	Low
S_9	0.11	0.08	0.26	0.23	0.66	0.15	1.49	Low
S_{10}	0.05	0.35	0.18	0.25	0.39	0.04	1.27	Low

Table 6. Potential ecological risk factor, risk index and pollution degree of heavy metals in soil.

Sites	Potential ecological risk factor (E^i_r)						Potential Risk (PER)	Pollution degree
	Cr	Ni	Cu	As	Cd	Pb		
S_1	2.98	19.94	2.20	13.89	400.19	2.02	441.21	Very high risk
S_2	1.34	7.23	59.18	148.95	1189.67	16.60	1422.97	Very high risk
S_3	2.09	21.30	3.89	12.23	182.26	7.91	229.68	Considerable risk
S_4	2.43	14.57	20.19	69.60	481.89	16.96	605.65	Very high risk
S_5	0.37	5.45	2.92	13.03	139.01	3.53	164.32	Considerable risk
S_6	0.24	2.58	6.56	12.51	564.95	6.82	593.66	Very high risk
S_7	0.43	2.67	16.32	8.20	56.73	6.15	90.50	Moderate risk
S_8	1.56	20.59	14.56	8.35	296.44	10.04	351.53	Very high risk
S_9	1.13	2.31	6.56	11.32	98.94	3.73	123.99	Moderate risk
S_{10}	0.50	10.49	4.57	12.51	58.63	1.10	87.80	Moderate risk

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

Contamination factor (C^i_f)	Contamination degree of individual metal	Degree of contamination (C_d)	Contamination degree of the environment	E^i_r	Grade of ecological risk of individual metal	Risk index (PER)	
$C^i_f<1$	Low	$C_d<5$	Low contamination	$E^i_r<40$	Low risk	RI<65	Low risk
$1\leq C^i_f<3$	Moderate	$5\leq C_d<10$	Moderate contamination	$40\leq E^i_r<80$	Moderate risk	65≤RI < 130	Moderate risk
$3\leq C^i_f<6$	Considerable	$10\leq C_d<20$	Considerable contamination	$80\leq E^i_r<160$	Considerable risk	130 ≤RI < 260	Considerable risk
$C^i_f\geq6$	High	$C_d\geq20$	High contamination	$160\leq E^i_r<320$	High risk	RI ≥ 260	Very high risk
				$E^i_r\geq320$	Very high risk		

Table 8. Chronic daily intake (CDI) of heavy metals through ingestion of soil.

Sampling sites	Cr		Ni		Cu		As		Cd		Pb		Total (Adult)	Total (Children)
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Chil-dren	Adult	Children		
S_1	7.8E-06	2.7E-05	1.5E-05	5.3E-05	1.7E-06	5.9E-06	1.5E-06	5.4E-06	1.4E-06	5.2E-06	1.2E-06	4.4E-06	2.8E-05	1.0E-04
S_2	3.5E-06	1.2E-05	5.5E-06	1.9E-05	4.5E-05	1.6E-04	1.6E-05	5.8E-05	4.4E-06	1.5E-05	1.0E-06	3.6E-05	7.5E-05	3.0E-04
S_3	5.5E-06	1.9E-05	1.6E-06	5.6E-05	3.0E-06	1.0E-05	1.3E-06	4.7E-06	6.0E-07	2.3E-06	5.0E-06	1.7E-05	1.7E-05	1.0E-04
S_4	6.4E-06	2.2E-05	1.1E-06	3.8E-05	1.5E-05	5.4E-05	7.7E-06	2.7E-05	1.7E-06	6.2E-06	1.0E-05	3.7E-05	4.1E-05	1.8E-04
S_5	0.9E-06	3.4E-06	4.1E-06	1.4E-05	2.2E-06	7.9E-06	1.4E-06	5.0E-06	5.0E-07	1.8E-06	2.2E-06	7.8E-06	1.1E-05	3.9E-05
S_6	0.6E-06	2.2E-06	1.9E-06	6.8E-06	5.0E-06	1.7E-05	1.4E-06	4.8E-06	2.1E-06	7.3E-06	4.3E-06	1.5E-05	1.5E-05	5.3E-05
S_7	1.1E-06	3.9E-06	2.0E-06	7.1E-06	1.2E-05	4.4E-05	9.1E-07	3.2E-06	2.0E-07	7.0E-07	3.9E-06	1.3E-05	2.0E-05	7.1E-05
S_8	4.1E-06	1.4E-05	1.5E-05	5.5E-05	1.1E-05	3.9E-05	9.3E-07	3.2E-06	1.1E-06	3.8E-06	6.3E-06	2.2E-05	3.8E-05	1.3E-04
S_9	2.9E-06	1.04E-05	1.7E-06	6.1E-06	5.0E-06	1.7E-05	1.2E-06	4.4E-06	3.0E-07	1.2E-06	2.3E-06	8.2E-06	1.1E-05	4.7E-05
S_{10}	1.3E-06	4.6E-05	8.0E-06	2.8E-05	3.5E-06	1.2E-05	1.4E-06	4.8E-06	2.0E-07	7.0E-07	7.0E-07	2.4E-06	1.5E-05	9.3E-05

Table 9. Chronic daily intake (CDI) of heavy metals through dermal contact of soil.

Sampling sites	Cr		Ni		Cu		As		Cd		Pb		Total (Adult)	Total (Children)
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children		
S_1	4.4E-06	1.5E-05	8.6E-06	3.0E-05	9.7E-07	3.4E-06	2.6E-06	9.2E-06	8.5 E-07	2.9E-06	7.3 E-07	2.5E-06	1.6E-05	6.3E-05
S_2	2.0E-06	7.0E-06	3.1E-06	1.1E-05	2.6E-07	9.1E-05	2.8E-05	9.9E-05	2.5E-06	8.8E-06	6.0E-06	2.1E-05	4.1E-05	2.3E-04
S_3	3.1E-06	1.1E-05	9.2E-06	3.2E-05	1.7E-06	6.0E-06	2.3E-06	8.1E-06	3.9E-07	1.3E-06	2.8E-06	1.0E-05	1.9E-05	6.8E-05
S_4	3.6E-06	1.2E-05	6.3E-06	2.2E-05	8.9E-06	3.1E-05	1.3E-05	4.6E-05	1.0E-06	3.5E-06	6.1E-06	2.1E-05	3.8E-05	1.3E-04
S_5	5.6E-07	1.9E-06	2.3E-06	8.3E-06	1.2E-06	4.5E-06	2.4E-06	8.7E-06	2.9E-07	1.0E-06	1.2E-06	4.4E-06	7.9E-06	2.8E-05
S_6	3.6E-07	1.2E-06	1.1E-06	3.9E-06	2.9E-06	1.0E-05	2.3E-06	8.3E-06	1.2E-06	4.1E-06	2.4E-06	8.6E-06	1.0E-05	3.6E-05
S_7	6.4E-07	2.2E-06	1.1E-06	4.0E-06	7.2E-06	2.5E-05	1.5E-06	5.4E-06	1.2E-07	4.2E-07	2.2E-06	7.7E-06	1.2E-05	4.4E-05
S_8	2.3E-06	8.2E-06	8.9E-06	3.1E-05	6.4E-06	2.2E-05	1.5E-06	5.5E-06	6.3E-07	2.2E-06	3.6E-06	1.2E-05	2.3E-05	8.0E-05
S_9	1.6E-06	5.9E-06	1.0E-06	3.5E-06	2.9E-06	1.0E-05	2.1E-06	7.5E-06	2.1E-07	7.3 E-07	1.3E-06	4.7E-06	9.1E-06	3.1E-05
S_{10}	7.5E-07	2.6E-06	4.5E-06	1.5E-05	2.0E-06	7.0E-06	2.3E-06	8.3E-06	1.2E-07	4.3E-07	4.0E-07	1.3E-06	1.0E-05	3.4E-05

Table 10. Chronic daily intake (CDI) of heavy metals through inhalation of soil.

Sampling sites	Cr		Ni		Cu		As		Cd		Pb		Total (Adult)	Total (Children)
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children		
S_1	3.7E-06	3.4E-06	7.1E-06	6.6E-06	8.0E-07	7.4E-07	7.2E-07	6.8E-07	6.9E-07	6.5E-07	6.0E-07	5.6E-07	1.4E-05	1.3E-05
S_2	1.7E-06	1.5E-06	2.6E-06	2.4E-06	2.1E-05	2.0E-05	7.8E-06	7.2E-06	2.1E-06	1.9E-06	4.9E-06	4.6E-06	4.0E-05	3.8E-05
S_3	2.6E-06	2.4E-06	7.6E-06	7.1E-06	1.4E-06	1.3E-06	6.4E-07	5.9E-07	3.2E-07	2.9E-07	2.3E-06	2.2E-06	1.5E-05	1.4E-05
S_4	3.0E-06	2.8E-06	5.2E-06	4.8E-06	7.3E-06	6.8E-06	3.6E-06	3.4E-06	8.4E-07	7.8E-07	5.0E-06	4.7E-06	2.5E-05	2.3E-05
S_5	4.6E-07	4.3E-07	1.9E-06	1.8E-06	1.1E-06	9.9E-07	6.8E-07	6.3E-07	2.4E-07	2.3E-07	1.0E-06	9.8E-07	5.4E-06	5.1E-06
S_6	3.0E-07	2.8E-07	9.2E-07	8.6E-07	2.4E-06	2.2E-06	6.5E-07	6.1E-07	9.8E-07	9.2E-07	2.0E-06	1.9E-06	7.2E-06	6.8E-06
S_7	5.3E-07	4.9E-07	9.5E-07	8.9E-07	5.9E-06	5.5E-06	4.3E-07	4.0E-07	9.9E-08	9.2E-08	1.8E-06	1.7E-06	9.7E-06	9.1E-06
S_8	1.9E-06	1.8E-06	7.3E-06	6.8E-06	5.3E-06	4.9E-06	4.4E-07	4.1E-07	5.2E-07	4.8E-07	3.0E-06	2.8E-06	1.8E-05	1.7E-05
S_9	1.4E-06	1.3E-06	8.2E-07	7.7E-07	2.4E-06	2.2E-06	5.9E-07	5.5E-07	1.7E-07	1.6E-07	1.1E-06	1.0E-06	6.5E-06	6.0E-06
S_{10}	6.1E-07	5.7E-07	3.7E-06	3.5E-06	1.7E-06	1.5E-06	6.5E-07	6.1E-07	1.0E-07	9.5E-08	3.3E-07	3.0E-07	7.1E-06	6.6E-06

Table 11. Target hazard quotient (THQ) of heavy metals through ingestion, dermal and inhalation of soil.

Sampling sites	Cr		Ni		Cu		As		Cd		Pb		Total (Adult)	Total (Children)
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children		
S_1	0.0053	0.0151	0.0015	0.0045	0.0001	0.0003	0.0161	0.0509	0.0042	0.0175	0.0005	0.0021	0.0277	0.0904
S_2	0.0024	0.0068	0.0006	0.0016	0.0017	0.0068	0.1727	0.5473	0.0180	0.0514	0.0034	0.0176	0.1987	0.6316
S_3	0.0037	0.0108	0.0009	0.0048	0.0002	0.0004	0.0141	0.0446	0.0026	0.0078	0.0029	0.0083	0.0244	0.0767
S_4	0.0043	0.0123	0.0006	0.0032	0.0008	0.0023	0.0810	0.2547	0.0071	0.0210	0.0060	0.0179	0.0999	0.3113
S_5	0.0006	0.0019	0.0004	0.0012	0.0001	0.0003	0.0149	0.0478	0.0021	0.0061	0.0013	0.0038	0.0194	0.0610
S_6	0.0004	0.0012	0.0002	0.0006	0.0003	0.0007	0.0145	0.0457	0.0086	0.0246	0.0025	0.0073	0.0264	0.0802
S_7	0.0008	0.0022	0.0002	0.0006	0.0006	0.0019	0.0095	0.0300	0.0008	0.0024	0.0023	0.0064	0.0141	0.0435
S_8	0.0028	0.0080	0.0016	0.0046	0.0006	0.0016	0.0096	0.0304	0.0045	0.0130	0.0037	0.0105	0.0226	0.0681
S_9	0.0020	0.0059	0.0002	0.0005	0.0003	0.0007	0.0130	0.0415	0.0014	0.0027	0.0013	0.0040	0.0181	0.0553
S_{10}	0.0009	0.0164	0.0008	0.0023	0.0002	0.0005	0.0145	0.0457	0.0008	0.0025	0.0004	0.0011	0.0176	0.0685

Table 12. Carcinogenic risk of adult due to ingestion, dermal contact and inhalation of arsenic and lead in soil.

Sampling sites	Arsenic (As)				Lead (Pb)			
	Ingestion	Dermal contact	Inhalation	Total risk	Ingestion	Dermal contact	Inhalation	Total risk
S_1	3.79E-09	8.57E-07	3.33E-13	8.60E-07	1.77E-11	1.01E-08	7.68E-14	1.01E-08
S_2	4.07E-08	9.19E-06	5.57E-12	9.23E-06	1.46E-10	8.33E-08	6.32E-13	8.34E-08
S_3	3.33E-09	7.53E-07	2.93E-13	7.56E-07	6.96E-11	3.96E-08	3.01E-13	3.96E-08
S_4	1.90E-08	4.29E-06	1.67E-12	4.30E-06	1.49E-10	8.51E-08	6.45E-13	8.52E-08
S_5	3.56E-09	8.05E-07	3.13E-13	8.08E-07	2.02E-11	1.77E-08	1.34E-13	1.77E-08
S_6	3.42E-09	7.73E-07	3.00E-13	7.76E-07	6.00E-11	3.42E-08	2.59E-13	3.42E-08
S_7	2.24E-09	5.06E-07	1.97E-13	5.08E-07	5.42E-11	3.08E-08	2.34E-13	3.08E-08
S_8	2.28E-09	5.16E-07	2.00E-13	5.18E-07	8.83E-11	5.03E-08	3.82E-13	5.03E-08
S_9	3.09E-09	6.98E-07	2.71E-13	7.01E-07	3.28E-11	1.87E-08	1.42E-13	1.87E-08
S_{10}	3.42E-09	7.73E-07	3.00E-13	7.76E-07	9.69E-12	1.10E-08	4.19E-14	1.10E-08

Table 13. Carcinogenic risk of children due to ingestion, dermal contact and inhalation of arsenic and lead in soil.

Sampling sites	Arsenic (As)				Lead (Pb)			
	Ingestion	Dermal contact	Inhalation	Total risk	Ingestion	Dermal contact	Inhalation	Total risk
S_1	1.01E-08	3.21E-08	3.33E-13	4.22E-08	4.73E-11	3.79E-10	7.69E-14	4.26E-10
S_2	1.08E-07	3.44E-07	3.57E-12	4.52E-07	3.89E-10	3.11E-09	6.32E-13	3.49E-09
S_3	8.86E-09	2.82E-08	2.93E-13	3.70E-08	1.85E-10	1.48E-09	3.01E-13	1.66E-09
S_4	5.07E-08	1.60E-07	1.67E-12	2.10E-07	3.98E-10	3.18E-09	6.45E-13	3.57E-09
S_5	9.51E-09	3.01E-08	3.13E-13	3.96E-08	8.30E-11	6.64E-10	1.34E-13	7.47E-10
S_6	9.12E-09	2.89E-08	3.00E-13	3.80E-08	1.60E-10	1.28E-09	2.59E-13	1.44E-09
S_7	5.98E-09	1.89E-08	1.97E-13	2.48E-08	1.44E-10	1.15E-09	2.34E-13	1.29E-09
S_8	6.09E-09	1.93E-08	2.00E-13	2.53E-08	2.35E-10	1.88E-09	3.82E-13	2.11E-09
S_9	8.24E-09	2.61E-08	2.71E-13	3.43E-08	8.75E-11	7.00E-10	1.42E-13	7.87E-10
S_{10}	9.12E-09	1.44E-08	2.75E-13	2.35E-08	2.58E-11	2.09E-10	4.19E-13	2.35E-10

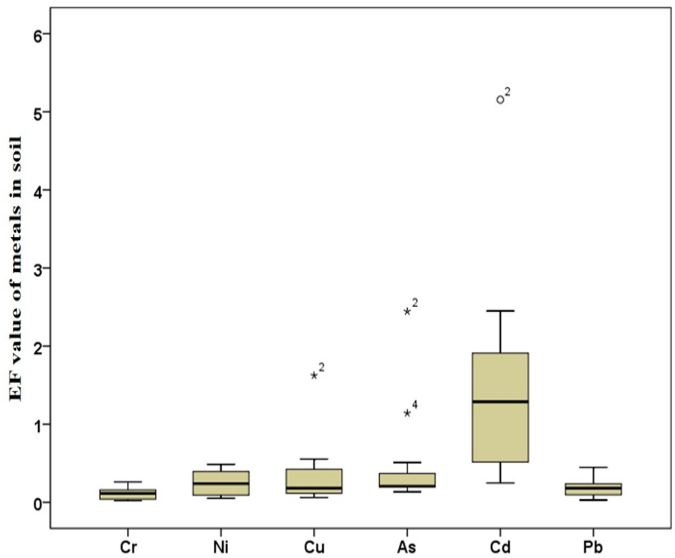

Figure 3. *Enrichment factor values of heavy metals in soil.*

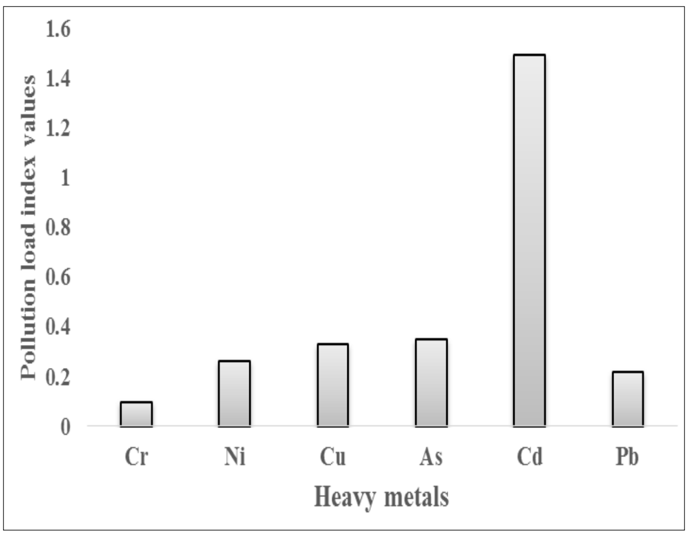

Figure 4. *Pollution load index values of heavy metals in soil.*

Toxic unit analysis of the present study was shown in Figure 5. A moderate to serious toxicity of hazardous elements remain in soil when the sum of toxic units for all soil samples is more than 4 (Bai *et al.*, 2011). In the present study, only sampling site 2 exceeds the standard value indicating serious toxicity of hazardous elements in soil.

Health risk assessment

Heavy metals present in soils may have an impact on human health (Okorie *et al.*, 2011). In the industrial areas, the risks of hazardous elements in industrial, waste burning sites, waste thronging sites and brick fields are important for the exposure through ingestion and dermal contact (Bright *et al.*, 2006; De Miguel *et al.*, 2007; Zheng *et al.*, 2010). According to the risk assessment approach, non-carcinogenic risks of trace metals through two exposure pathways were characterized in this study. In order to evaluate the risk, the chronic daily intakes (CDIs), hazard quotients (HQs), hazard index (HI) and carcinogenic risk of the studied metals were estimated for adults and children and the results are presented hereby.

Chronic daily intake (CDI) of heavy metals through ingestion, dermal contact and inhalation for adult and children was presented in Tables 8, 9 and 10. On the basis of ingestion, the chronic daily intake of total metals ranged from 1.1E-05 to 7.5E-05 for adult and 3.90E-05 to 3.00E-04 for children. According to dermal contact, chronic daily intake of total metals ranged from 9.1E-06 to 4.10E-05 for adult and 2.80E-05 to 2.30E-04 for children. Due to inhalation, CDI of heavy metals ranged from 5.40E-06 to 4.00E-05 for adult and 2.30E-05 to 3.80E-05 for children. Chronic daily intake was higher in children than the adult for ingestion, dermal contact and inhalation as body weight of children was lower than the adult.

The Hazard quotients (HQs) of individual metal for the present study were shown in Table 11. Hazard quotients were calculated from according to ingestion, dermal contact and inhalation concentration of metals. The non-cancer health risks related to individual element exposure through soil ingestion, dermal contact and inhalation was low for all investigated elements resulted in a HQ < 1, indicating low risk for both adults and children.

The combined effects of exposed metals and metalloids were calculated as hazard index (HI) and the data indicated that the HI values were also lower than one. However, when considering the total exposure HI of ingestion, dermal contact and inhalation there was no chance of having non-cancer risk at all of the sites on adults and children health. The total hazard index for children and adult was 0.0176 and 0.0685, respectively (Figure 6). The hazard risk index values for children were higher than that of adult inhabitants indicating children may pose non-cancer risk in the future. The hazard index value for children was higher in children than adult on the basis of ingestion, dermal contact and inhalation. The total target hazard quotients (TTHQ) for children was higher due to touching and mouthing of dust contaminated particles, direct ingestion by hand to mouth activities (Mielke *et al.*, 1999). The ingestion of greater amounts of small particles may have greater impact on children because of their small body weight than adult (Beamer *et al.*, 2008). Children are exposed to higher amount of soil than the adult due to pica and play behavior (CDC, 2005).

The carcinogenic risk of As and Pb for adults are presented in Table 12 and 13. The carcinogenic risks from As and Pb at all sites via ingestion, dermal contact and inhalation were in acceptable ranges. The cancer risk of As and Pb ranged from 5.18E -07 to 9.23E-06 and 1.01E-08 to8.34E-08 for adult. The range of carcinogenic risk of children for As was 2.35E-08 to 4.52E-07 and for Pb was 2.35E-10 to 3.57E-09. For all sampling sites, carcinogenic risk posed by As and Pb was lower than 10^{-6} through different exposure pathways. The carcinogenic risks of As and Pb due to exposure from studied soil via ingestion, dermal contact and inhalation pathways can be negligible in the industrial areas of Tangail district, Bangladesh, as Cancer risk value for all sites were lower than target value 10^{-6}(USEPA, 2011). Among the three exposure pathways, the ingestion of soil seems to be the major pathway of exposure to hazardous elements followed by dermal contact and inhalation. Hazardous elements could be accumulated in human for a long time and especially non-cancer adverse effects of these toxic metals to the tissues of adult population can become more serious. According to the result of present study, health risk for adult and children due to heavy metal exposure through soil could not be overlooked.

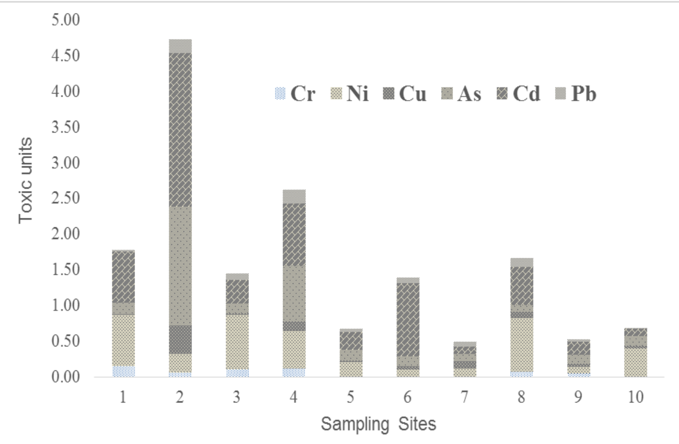

Figure 5. *Toxic unit analysis of heavy metals in soil.*

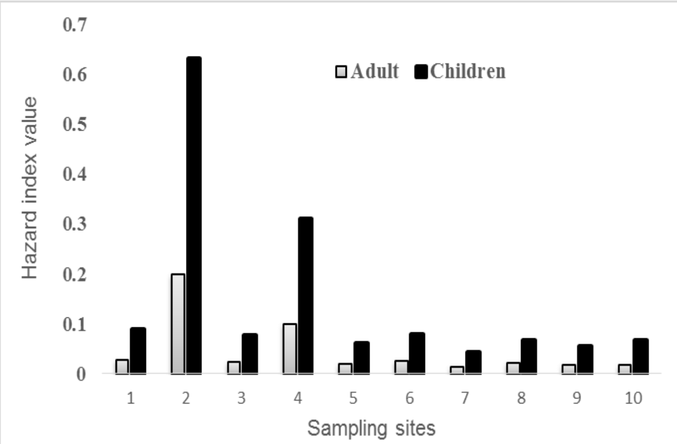

Figure 6. *Hazard index (HI) of heavy metals due to ingestion, dermal contact and inhalation of soil.*

Conclusion

The major findings of the study revealed that Cd concentrations in some sampling sites exceeded the Dutch standard and Canadian quality guidelines values, representing that the studied soils were heavily polluted by Cd. The enrichment factor, geoaccumulation index, contamination factor, pollution load index and toxic unit analysis values were found low for all metals except Cd. Toxic elements in different sampling sites showed moderate to very high degree of contamination. The severity of potential ecological risk factor for single metal (E^i_r), only Cd had very severe ecological risk for most of the sampling sites in the study area. Ingestion and dermal contact of the toxic elements in adult and children body in the study area have no probability to pose the non-cancer risk. But the concern is that long term exposure of these toxic elements can pose cancer both in child and adult population around the industrial vicinity of Tangail district in Bangladesh.

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Conflict of interest

No any conflict of interest is declared by the authors.

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