

SF Journal of Nanochemistry and Nanotechnology

Contamination of Heavy Metals in Agricultural Soils: Ecological and Health Risk Assessment

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Abstract

In recent years, heavy metal contamination in agricultural soils calls for significant concerns due to the rapid urbanization and industrialization. The present research was conducted to assess the ecological and health risk of heavy metals (Cr, Ni, Cu, As, Cd and Pb) from agricultural soils in the industrial areas of Tangail district, Bangladesh. Heavy metals were investigated utilizing an Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The mean concentrations of Cr, Ni, Cu, As, Cd and Pb in different soil sampling sites were found 6.73, 29.74, 24.69, 4.79, 2.50 and 19.90 mg/kg, respectively. The mean concentration of the studied heavy metals were found underneath as far as possible set by the Dutch standard, Canadian guidelines and Australian guidelines with the exception of Cd. Principal Component Analysis (PCA) demonstrates that the vast majority of the metals in agricultural soil may originate from the anthropogenic sources. The geo-accumulation index, contamination factor and toxic unit analysis were discovered low contamination for all metal with the exception of Cd. Potential ecological risk (PER) of soils from all sampling sites showed low to very high risk. Add up to Total Target Hazard Quotients (TTHQ) for every single concentrated metal in all-out testing sites were <1 and cancer risk values were under 10⁻⁶ demonstrating low non-cancer-causing and cancer risk in grown-up and youngsters for a few exposure pathways.

Keywords: Heavy metals; PCA; Potential ecological risk; Health risk; Bangladesh

Introduction

Heavy metals contamination in agricultural soils has been becoming a global concern. Heavy metal contamination is harmful to plants, animals and human health [1,2]. Soil can be contaminated by natural and human causes, and anthropogenic activities are the main contributors to the pollution of heavy metals in agricultural land [3,4]. The source of toxic metals can be identified by using Principal Components Analysis (PCA), whether it is formed natural or anthropogenic sources [5-7]. Heavy as well as toxic metals like Nickel (Ni), Chromium (Cr), Cadmium (Cd), Lead (Pb) and Arsenic (As) are highly toxic reported by the USEPA [8].

The toxicity and sources of heavy metals are of burning question nowadays, which are non-biodegradable in nature and biomagnification in food chain [9,10-18]. In recent decades, due to rapid industrialization and urbanization, soil pollution from several toxic elements has been a matter of considerable concern, particularly in undeveloped countries [19,20-22]. Soil pollution that occurs by different heavy metals is regarded as the most adverse effect on the environment as well as global ecology [23].

In Bangladesh, heavy metals pollution from industrial wastages is one of the burning issues nowadays due to rapid industrialization [15-17]. Untreated effluents discharge from industries

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Received Date: 05 Feb 2019

Accepted Date: 08 Mar 2019

Published Date: 11 Mar 2019

Citation: Proshad R, Islam MS, Kormoker T, Bhuyan MS, Hanif MA, Hossain N, et al. Contamination of Heavy Metals in Agricultural Soils: Ecological and Health Risk Assessment. *SF J Nanochem Nanotechnol.* 2019; 2(1): 1012.

ISSN 2643-8135

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in open water bodies like lakes, canals, and the rivers that finally finds their way into the ocean [15-17]. However, this polluted water mostly uses by farmers for their irrigation purposes in agricultural field intentionally or unintentionally and subsequently contaminate the soils. The contamination has long-term negative effects on the ecology and human health [24-26].

Crops which are being cultivated in contaminated agricultural soils may cause serious health hazards to the human body resulting from their consumption [18,27]. Exposure to contaminants from heavy metals is extremely alarming for children in their first developmental stage and also for adults [28-30]. Chromium, copper, arsenic, cadmium and lead are of particular concern because of much known detrimental health effects on humans through food consumption in extra amounts [31,32]. Incorporation of toxic elements of soils come to the body by food implies a recognized pathway to elements pollutant exposure for children [33,34]. Chronic heavy metal exposure has a harmful effect on humans and other animals [35,36]. Cr and Cu can be responsible for non-carcinogenic health hazards such as neurological involvement, headache with liver disease if they exceed their safe threshold values [37].

The risk of lifetime lung cancer death that can occur in any age resulted from harmful exposure to dust and mists for containing hexavalent chromium [38]. Acute and chronic arsenic exposure also responsible for several human health problems like cardiovascular, carcinogenic (such as liver cancer), dermal, developmental, gastrointestinal, genotoxic, hematological, hepatic, immunological, neurological, respiratory, renal, reproductive, and mutagenetic effects [39].

The industrial area is regarded as a great source of pollutants (e.g. heavy metals) that accumulated into the environmental matrices and enter plants, animals and human bodies [40-42]. Tangail district is an industrialized area of Bangladesh and soil is being contaminated by heavy metals due to rapid industrialization in this area. It is famous for agricultural products and it provides a large portion of agricultural products for the whole country. Unfortunately, there is no or very limited research was carried out to assess the toxic elements contained in this area, their contamination level in soil along with their environmental and human health effects. As a result, the current research was conducted to determine toxic elements (Cr, Ni, Cu, As, Cd, and Pb) concentrations in agricultural soils and to evaluate the potential ecological risk of toxic elements to the environment and the human body.

Materials and Methods

Study area

The soil samples were collected from Tarutia, a place situated in Tangail Sadar Upazila of Tangail District, Bangladesh (Figure 1). Tangail region is arranged at the centrepiece of Bangladesh with a zone of 334.26km², which is a densely populated zone. Recently, because of quick industrialization Tangail region has been subjected to heavy contamination. As there have various types of enterprises (e.g. garment, knitwear, footwear, and tanneries) in Tangail region produces colossal amounts of toxic wastes which comprises distinctive substantial elements. Those enterprises also released squanders haphazardly to stream and trenches without any treatment [23] that at last blended with soils and defiled by poisonous substantial metals there. Ten soil-examining locales were chosen from Tarutia (Figure 1) from the region zone of the automated region of Tangail locale,

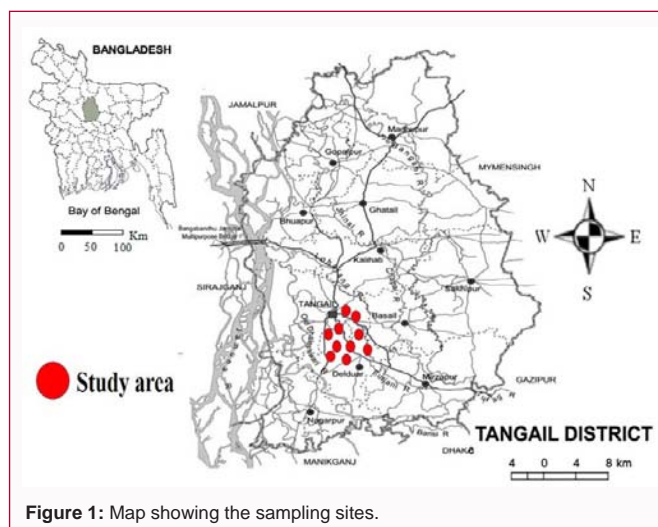


Figure 1: Map showing the sampling sites.

Bangladesh.

Sample collection

Agricultural soil samples were collected from the surface soil (0-10 cm) and composite examining procedure was carried out (three subsamples were gathered, blended all together and made one example from each side) by using a percussion pound corer (length was 50-80 cm) for toxic elements investigation. After collection, air-drying collection, air-drying samples were prepared for about 14 days in the research center. A mortar of porcelain and a pestle were utilized to break the blocks. Samples were sieved using a 2mm nylon sifter for the homogeneity and expulsion of pointless substances (roots, garbage, and stones). The sieved soil tests were put away in a spotless, sealed shut Ziploc sack for compound investigations [43].

Physicochemical parameters analysis

Soil particle estimation was dictated by hydrometer technique [44]. Textural classes were controlled by plotting the % sand, % residue and % earth on a triangular graph planned by Marshall pursued USDA framework. pH value of soil was resolved in a proportion of 1:2.5 (w:v) soil: water suspension with glass terminal pH meter (WTW pH 522; Germany). Soil Electrical conductivity (EC) was dictated by utilizing an EC meter (WTW LF 521; Germany) [45]. Natural carbon (C) was dictated by wet-oxidation technique [46].

Heavy metal analysis

All synthetic substances were investigative review reagents; Milli-Q water (Elix UV5 and Milli-Q, Millipore, Boston, MA, USA) was utilized for the planning of arrangements. The Teflon vessels and polypropylene holders were cleaned by absorbing 5% HNO₃ for in excess of 24 h, at that point washed with Milli-Q water and dried. For elements investigation, 0.3-0.5 g of the air-dried soil test was treated with 6mL 69% HNO₃ (Kanto Chemical Co, Tokyo, Japan) and 2mL 30% H₂O₂ (Wako Chemical Co, Tokyo, Japan) in a shut Teflon vessel and was processed in a system of Microwave Digestion (Berghof speedwave1, Eningen, Germany). The processed examples were then moved into a Teflon receptacle, and added up to 50mL volume with Milli-Q water. Then by utilizing a channel of the syringe (DISMIC1-25HP PTFE, pore estimate = 0.45mm; Toyo Roshi Kaisha, Ltd., Tokyo, Japan) the processed arrangement was separated and put the polypropylene in 50mL tubes (Nalgene, New York (NY), USA) for toxic elements investigation. A clear assimilation methodology was pursued without a soil test. For heavy metal, assurance tests

were investigated by utilizing Inductively Coupled Plasma Mass Spectrometer (ICP-MS, Agilent 7700 arrangement, Santa Clara, CA, USA). The farthest ICP-MS discovery ranges for the metals examined were 0.7, 0.6, 0.8, 0.06 and 0.09 ng/L for Cr, Ni, Cu, As, Cd and Pb respectively. Arrangements for Multi-component Standard XSTC-13 (Spex CertiPrep®, Metuchen, NJ, USA) have been utilized to prepare adjustment bends. Spex CertiPrep1 (Metuchen, NJ, USA) produced standard internal alignment arrangements containing 1.0mg/L of indium (In), yttrium (Y), beryllium (Be), tellurium (Te), cobalt (Co) and thallium (Tl), respectively. Also, 10mg/L inside a typical arrangement was set up from the essential standard and added to the processed examples. Multiple component arrangements (bought from Agilent Technologies, Japan) were utilized as a tuning arrangement and covered with an extensive variety of components. Whole experiment bunches were evaluated using an internal quality methodology and approved if the characterized Internal Quality Controls (IQCs) were carried out. At first beginning the examination grouping, relative standard deviation (RSD, <5%) was checked by utilizing a tuning arrangement acquired from the technology of Agilent. The affirmed reference materials INCT-CF-3 (corn flour) purchased from the National Research Council (Canada), were examined to affirm expository execution and great correctness (comparative standard deviation beneath 5%) of the linked approach.

Assessment of ecological hazard for soil pollution

A methodology was developed by Hakanson [47] for ecological risk assessment from toxic metals is now widely used.

Contamination factor (C_f^i)

Contamination factor means the proportion of the concentration of metals (heavy metals) in the soil to the baseline or background value:

$$C_f^i = C_{\text{heavy metal}} / C_{\text{background}}$$

Pollution calculates partitioned four classes ran from 1 to 6 which are: low degree ($C_f^i < 1$), direct degree ($1 \leq C_f^i < 3$), significant degree ($3 \leq C_f^i < 6$), and high degree ($C_f^i \geq 6$) [48]. This methodology has been utilized by different scientists (e.g. Proshad et al. [18]).

Geo-accumulation index (I_{geo})

(I_{geo}) is accepted as a noteworthy apparatus to decide sully degree from dangerous metals. At present, this technique is utilized all inclusive to evaluate soil contamination [49]. The best target to decide geo-gathering list (I_{geo}) is to recognize the contamination level in the dirt. Geo-aggregation list (I_{geo}) might be evaluated by applying condition given below by,

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

where C_n is the determined element (n) concentration measured from the soil, B_n is the geochemical baseline value of metal n in the background sample [50]. Geoaccumulation index (I_{geo}) values were interpreted as: $I_{\text{geo}} \leq 0$ – practically uncontaminated; $0 \leq I_{\text{geo}} \leq 1$ – uncontaminated to moderately contaminated; $1 \leq I_{\text{geo}} \leq 2$ – moderately contaminated; $2 \leq I_{\text{geo}} \leq 3$ – moderately to heavily contaminated; $3 \leq I_{\text{geo}} \leq 4$ – heavily contaminated; $4 \leq I_{\text{geo}} \leq 5$ – heavily to extremely contaminated; and $5 < I_{\text{geo}}$ – extremely contaminated.

Potential ecological risk (PER)

The degrees of substantial contamination of metals in farming soils is controlled by PER index. Guo et al. [51] proposed conditions which were utilized to compute PER and are as per the following:

$$C_f^i = \frac{C^i}{C_n^i}, C_d = \sum_{j=1}^n C_f^j$$

$$E_r^i = T_r^i \times C_f^i, PER = \sum_{j=1}^m E_r^j$$

where C_f^i is the sully variable of individual metal, C^i is component content in soils tests and C_n^i is metal gauge esteems. The reference estimations of Cr, Ni, Cu, As, Cd and Pb in soils were 90, 68, 45, 13, 0.3 and 20 mg/kg, separately [52]. The coordination of C_f^i for aggregate components speaks to the general level of contamination (C_d). E_r^i represent PER index and T_r^i is the biological toxic factor of a single element. The dangerous reaction factors for Cr, Ni, Cu, As, Cd and Pb were 2, 6, 5, 10, 30 and 5, separately [51, 53-57]. Environmental hazard index is the exhaustive potential natural index, which is the entirety of E_r^i . It refers to the impact of the natural network on the dangerous substances and demonstrates the environmental hazard caused by the general sully.

Analysis of toxic unit

The count of lethal units is considered as extreme harmfulness of dangerous elements in agricultural soils. The harmful unit examination is the extent to which substantial metals in the soil are centralized to a conceivable level of impact [58]. At the point when the whole of lethal units for all dirt examples is more than 4, direct to the genuine poisonous quality of substantial metals stay in the dirt.

The Carcinogenic and non-carcinogenic risk from the soil heavy metal exposure pathway

In this examination, danger records for both cancer-causing and non-cancer-causing impacts were connected to every presentation pathway in private situations. The models utilized in this investigation to decide introduction danger of youngsters and grown-ups to substantial metals in soil tests allude to the suggested models sourced from the United States Environmental Protection Agency (USEPA) [59] and the Dutch National Institute of Public Health Agency. The accompanying suspicions created by USEPA are: (1) human creatures are presented to the dirt through four fundamental pathways: ingestion of residue, an inward breath of residue particles through mouth and nose, dermal contact retention and introduction through the inward breath of Hg (vapor); (2) consumption rates and molecule emanation can be approximated by those produced for soil; (3) the presentation that related with parameters of kids and grown-ups in the considered zones are like those of reference populaces and (4) the generally speaking, cancer-causing and non-cancer-causing danger of every component can be ascertained by summing up the individual hazards derived from the initial methods. The wellbeing hazard evaluation exhibited in this investigation may have a few vulnerabilities and impediments. For example, the distinctions in presentation term, age, body weight, gender, and ingestion rate of every individual during a time gathering could be a wellspring of vulnerability. The relating dosage got through each of the pathways was assessed separately for non-cancer-causing hazard [8,59]. For cancer-causing agents, the lifetime normal day-by-day measurements (LADD) ($\text{mgkg}^{-1}\text{day}^{-1}$) was utilized for disease chance evaluation.

Assessment of non-carcinogenic risk

$$D_{\text{ing}} = C \times \frac{\text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CF}$$

$$D_{\text{inh}} = C \times \frac{\text{InhR} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{BW} \times \text{AT}}$$

$$D_{\text{dermal}} = C \times \frac{\text{SL} \times \text{SA} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CF}$$

Table 1: Physicochemical properties of soil collected from Tangail industrial area.

Sampling sites	pH (1:2.5 H ₂ O)	EC (dS/m)	Organic carbon (%)	Sand (% in <2mm)	Silt	Clay	Soil type ^a
S1	6.08	0.07	1.282	38.5	46.6	14.9	Loam
S2	6.04	0.06	0.22	37.6	46.6	15.8	Loam
S3	6.05	0.09	0.63	37.2	42.5	20.3	Loam
S4	6.22	0.11	0.657	41	47.5	11.5	Loam
S5	6.11	0.06	0.727	36.5	47.5	16	Loam
S6	5.87	0.18	2.617	43.5	45	11.5	Loam
S7	5.96	0.07	0.617	46.5	36.5	16.9	Loam
S8	6.06	0.09	2.627	44	39.1	16.9	Loam
S9	6.48	0.15	0.974	40.1	46.6	13.3	Loam
S10	6.54	0.21	0.831	39.1	40.9	20	Loam

^aAccording to the United States Department of Agriculture soil classification system.

Assessment of carcinogenic risk

$$LADD = \frac{C \times EF}{PEF \times AT} \times \frac{InhR \times ED}{BW}$$

where C-Exposure point fixation (mg/kg); IngR-Ingestion rate (100mg/d for kids and 50mg/d for grown-up) [60,61]; EF-Exposure recurrence (320d/y for kids and 220 d/y for grown-up) [62]; ED-Exposure length (6 years for youngsters and 24 years for grown-up) [8]; CF-units transformation factor: 10⁻⁶ kg mg⁻¹ [61]; BW-Average body weight (18.6 kg for kids and 80kg for grown-up) [60]; AT (for non-cancer-causing)-Average presentation time (ED 365 d) [61,63]; AT (for cancer-causing)-Average introduction time (25,550) [61]; InhR-Inhalation rate (7.6m³/d for kids and 20m³/d for grown-up) [8]; PEF-Particle discharge factor (4.59 × 10⁸); SL-Skin adherence factor (0.2mg/cm²/d for kids and 0.07mg/cm²/d for grown-up); SA-Exposed skin territory (2699cm² for kids and 3950cm² for grown-up) [60]; ABS-Dermal assimilation factor (0.001) [60].

Statistical analysis

Multivariate measurable strategies like chief segment investigation (PCA) was executed to get definite data of the informational collection and gain understanding into the appropriation of substantial metals by distinguishing likenesses or dissimilarities of soil tests. The PCA was dissected by applying Varimax-standardized pivot with Ward's strategy and Microsoft Excel 2013 was utilized for different computations.

Results and Discussion

Heavy metals contamination is detrimental for the agricultural soil that ultimately affects the health of the soils. Thereafter, contaminated soil is largely liable for environmental deterioration and human health problems. Crop production may be affected by exposure of heavy metals in soils as well as their storage and transformation and its effects on human, animal and plant health [64]. From the findings of this research, it is found that the concentration level of all soil metal concentration was lower than Dutch standard [64], Australian guidelines [65] and Canadian guidelines [66] except Cu and Cd. Copper and Cadmium concentrations were recorded higher in the present study compared to the Dutch standard. Environmental action level demonstrates that the low risk to the environment and human health.

Physicochemical properties and concentration of toxic elements in soils

According to the soil texture classification system in the United States (Soil Survey Division Staff) [67], the textural analysis showed

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

Sampling sites	Cr	Ni	Cu	As	Cd	Pb
S1	1	8.75	4.78	1.57	0.41	5.29
S2	0.22	14.69	6.37	2.18	0.37	10.14
S3	3.99	10.18	16.81	0.99	0.19	2.83
S4	5.93	16.5	17.97	3.43	5.14	81.43
S5	11.99	87.72	37.24	5.87	1.65	20.37
S6	5.43	12.21	41.6	6.57	9.04	3.36
S7	5.98	11	37.79	6.44	0.83	37.42
S8	13.22	31.35	15.46	3.99	5.33	14.57
S9	13.26	84.44	57.69	9.01	0.83	12.4
S10	6.3	20.65	11.23	7.89	1.23	11.24
Mean	6.73	29.74	24.69	4.79	2.5	19.9
Dutch standard ^a	100	35	36	29	0.8	85
Canadian guidelines	64	50	63	12	1.4	70
Australian guidelines	50	60	60	20	3	300

^aVROM (2000); ^bCCME (2003); ^cDEP (2003).

that the soil samples studied were loam (Table 1). The studied soils pH values for different sampling sites were found slightly acidic to neutral possibly due to decomposition of organic matter and subsequent carbonic acid formation [68] (Table 1). Higher soil acidity favours the presence of cations in soil [69]. According to SRDI soil salinity class, electrical conductivity (EC) value of the soil was non-saline (0-2 dS/m) for all sampling sites which mean the salinity effect is negligible [70]. The highest percentage value of organic carbon was observed in soil that collected from the S8 site and lowest value observed in the S2 site. High organic carbon content indicates that metals are more likely to form metal chelate complexes in organic matter, which would also lead to fewer metals being available to plants [71].

The heavy metals' (Cr, Ni, Cu, As, Cd, and Pb) concentrations in soil samples of different sampling sites are presented in Table 2. In this research, the mean concentrations of Cr, Ni, Cu, As, Cd and Pb in different soil sampling sites were found 6.73, 29.74, 24.69, 4.79, 2.50 and 19.90 mg/kg, respectively around the industrial area of Tangail district, Bangladesh (Table 2).

Chromium (Cr) is an important contaminant that is discharged from industries into the agricultural land [72]. The concentration of Cr in agricultural soils varies up to values as high as 350 mg kg⁻¹ [73]. In the present research, the highest Cr concentration was

Table 3: Total variance explained and component matrices for the hazardous elements in soils.

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	1093.07	59.24	59.24	1093.07	59.24	59.24	1076.28	58.33	58.33
2	569.37	30.86	90.1	569.37	30.86	90.1	586.16	31.77	90.1
3	164.2	8.9	99						
4	11.11	0.6	99.6						
5	4.37	0.23	99.84						
6	2.89	0.15	100						
Elements	Component matrix				Rotated Component Matrix				
	Raw component		Rescaled component		Raw component		Rescaled component		
	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC2
Cr	3.71		0.79		3.78		0.8		
Ni	29.94		0.98		29.6		0.97		
Cu	13.22		0.74		13.35		0.75		
As	1.69		0.61		1.68		0.61		
Cd									
Pb		23.75		0.99			23.75		0.99

Table 4: Contamination factors, degree of contamination and contamination level in soils.

Sites	Contamination factors (C _i)						Degree of contamination (C _d)	Contamination level
	Cr	Ni	Cu	As	Cd	Pb		
S1	0.022	0.224	0.145	0.165	1.344	0.265	2.165	Low
S2	0.005	0.377	0.193	0.229	1.213	0.507	2.524	Low
S3	0.089	0.261	0.509	0.104	0.623	0.142	1.728	Low
S4	0.132	0.423	0.545	0.361	16.852	4.072	22.384	High
S5	0.266	2.249	1.128	0.618	5.41	1.019	10.69	Considerable
S6	0.121	0.313	1.261	0.692	29.639	0.168	32.193	High
S7	0.133	0.282	1.145	0.678	2.721	1.871	6.83	Moderate
S8	0.294	0.804	0.468	0.42	17.475	0.729	20.19	High
S9	0.295	2.165	1.748	0.948	2.721	0.62	8.498	Moderate
S10	0.14	0.529	0.34	0.831	4.033	0.562	6.435	Moderate

observed at 13.22mg/kg and 13.26mg/kg at S8 and S9 sampling sites. 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 [74]. Yu et al. [75] recorded 40.10mg/kg Cr in the arid agricultural soil in central Gansu Province, China. The threshold value for Cr is ≤ 120 mg/kg for arid agricultural soils in China [76]. Hasnine et al. [77] reported average Cr concentration in the surface agricultural soil at DEPZA was found to be 2753.2 ± 4598.86 mg/kg. Cr amount was recorded 5 to 1,500 mg/kg in Canadian soils with a mean of 43mg/kg [78]. Frank et al. [79] recorded 14.3 ± 8.5 mg/kg Cr in agricultural soils of Ontario. The toxicity of Cr has negative impacts on the growth of plants that inhibit different important metabolic systems [80-82].

Nickel (Ni) can cause cardiovascular disease, dermatitis, kidney disease, lung fibrosis and respiratory cancer, and in the human body [77]. The solubility of nickel in soils increases with its acidity and if the acidity increase it results in higher Ni in soils [83]. In this research, Ni concentrations ranged between 8.75-87.72 mg/kg in the study area. Highest amount (87.72mg/kg) was found in site 5 and the lowest value (8.75mg/kg) in site 1 (Table 2). Ni concentration was

recorded in a lower amount in all site compared to Dutch standard, Canadian guidelines and Australian guidelines except in site 5 & 9 (Table 2). Hasnine et al. [77] reported average Ni concentration in the surface agricultural soil at DEPZA was found to be 655.53 ± 979.73 mg/kg. Dojlido and Best [84] found approximately 26,000mg/kg Ni of highly developed nickel smelting in Canada. 250mg/kg Ni was measured in a highly polluted area contaminated by galvanization plant sewage [84]. The concentration of Ni in the agricultural soils of Ontario varied between 1.3 to 6,560mg/kg [79].

Excessive copper (Cu) concentrations are harmful to plants and highly toxic to some microorganisms [77]. Soluble soil Cu can be toxic to plants since Cu-enriched liquid dairy waste used in agricultural land as irrigation water [85]. Alloway [86] provided with the regulatory standard for Cu in soil is 20-30 mg/kg. In this research, the value of Cu ranged between 4.78mg/kg to 57.69mg/kg (Table 2). The maximum amount 57.69mg/kg was found at site 9 while minimum 4.78mg/kg was found at site 1. The concentration of Cu in site 5, 6, 7 and 9 were found above the allowable limit set by the Dutch standard and recommended value of Alloway [86]. The value of Cu except these sites found below the standard value set by Canadian guidelines

Table 5: Potential ecological risk factor, risk index and pollution degree of heavy metals in soils.

Sites	Potential ecological risk factor (E_i^p)						Potential Risk (PER)	Pollution degree
	Cr	Ni	Cu	As	Cd	Pb		
S1	0.044	1.346	0.724	1.653	40.328	1.323	45.418	Low
S2	0.01	2.26	0.965	2.295	36.393	2.535	44.458	Low
S3	0.177	1.566	2.547	1.042	18.689	0.708	24.729	Low
S4	0.264	2.538	2.723	3.611	505.574	20.358	535.067	Very high
S5	0.533	13.495	5.642	6.179	162.295	5.093	193.237	Considerable
S6	0.241	1.878	6.303	6.916	889.18	0.84	905.359	Very high
S7	0.266	1.692	5.726	6.779	81.639	9.355	105.457	Moderate
S8	0.588	4.823	2.342	4.2	524.262	3.643	539.858	Very high
S9	0.589	12.991	8.741	9.484	81.639	3.1	116.545	Moderate
S10	0.28	3.177	1.702	8.305	120.984	2.81	137.257	Considerable

Table 6: Indices and grades of the potential ecological risk of heavy metal pollution [53].

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

and Australian guidelines (Table 2). Frank et al. [79] documented the value of Cu ranged from 2.1 to 664 mg/kg in agricultural soils of Ontario. Yu et al. [75] found 17.10mg/kg Cu in the arid agricultural soil in central Gansu Province, China. The threshold value for Cu is ≤ 60 mg/kg for arid agricultural soils in China [76]. Hasnine et al. [77] reported average Cu concentration in the surface agricultural soil at DEPZA was found to be 91.06 ± 152.70 mg/kg. Sonmez et al. [87] reported a decreased height in the plant, total yield, fruit, and dry root weight with increasing Cu application.

Arsenic (As) concentration varied between 0.99 mg/kg to 9.01 mg/kg in this present research. The highest amount of 9.01mg/kg was recorded from site 9 while the lowest amount of 0.99mg/kg was found on site 3 (Table 2). All the concentrations of As found far below the recommended value set by the Dutch standard, Canadian guidelines and Australian guidelines (Table 2). Frank et al. [79] estimated 6.21 ± 2.67 mg/kg As in agricultural soils of Ontario while Yu et al. [75] recorded 8.80mg/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 [76]. As contaminated water and As-enriched fertilizers, as well as pesticides, were used for irrigation in the agricultural land [88,89]. In addition, emission and waste from brickfields and incineration activities could contribute to the high concentration of As in agricultural soil [90].

Cadmium (Cd) concentrations were found between 0.19mg/kg to 9.04mg/kg. Maximum amount (9.04mg/kg) was recorded at site 6 and minimum (0.19mg/kg) was recorded at site 3. Cd concentrations at site 4 (5.14mg/kg), 6 (9.04mg/kg) and 8 (5.33mg/kg) were recorded above the permissible limit set by the Dutch standard (0.80mg/kg), Canadian guidelines (1.4mg/kg) and Australian guidelines (3.0mg/kg) (Table 2). The amount of Cd at site 7 (0.83mg/kg) and 9 (0.83mg/kg) found above the allowable limit set by the Dutch standard (0.80mg/kg) (Table 2). Frank et al. [79] documented 0.5 ± 0.69 mg/kg

Cd in agricultural soils of Ontario. 0.5 ± 0.69 . About 70% of studied soil samples exceeded the Dutch target amount assuming that Cd in soil might pose a serious and harmful risk to the surrounding ecosystems.

Lead (Pb) concentration was ranged between 2.83mg/kg to 81.43mg/kg in the study area (Table 2). The highest amount of Pb (81.43mg/kg) was found at site 4 that exceeded the allowable limit set by Canadian guidelines (Table 2). This excess concentration of Pb found in soil may be due to several anthropogenic factors like metal processing factories [91,92]. But the all other value at different sites recorded below the standard limit set by the Dutch standard, Canadian guidelines and Australian guidelines (Table 2). Yu et al. [75] recorded 23.30mg/kg Pb in the arid agricultural soil in central Gansu Province, China. The threshold value for Pb is ≤ 50 mg/kg for arid agricultural soils in China [76]. Frank et al. [79] recorded value for Pb that ranged between 1.5 to 888 mg/kg in agricultural soils of Ontario. Chrastný et al. [93] measured 21.3 ± 2.1 mg/kg of Pb in agricultural soil close to the vicinity of a shooting range.

Source analysis of heavy metals

Multivariate statistical analyses are as often as possible and broadly used in source allotment of natural contaminations in urban conditions [94,95]. Principal component analysis (PCA) is utilized to recognize the wellspring of harmful metals in soils around mechanical regions. PCA has been expected a successful instrument for source identification [96,97-99]. From the present examination, two essential segments were discovered executing PCA investigation (Table 3 and Figure 2). Two principal components were represented by 90.1% of the aggregate variety. From the present examination PCA investigation, the initial two main parts were figured and the fluctuation clarified by them was 58.33% and 31.77% after the revolution (Table 3). The metals Cr, Ni, Cu and As were processed in the primary chief part (PC1) bringing about the most elevated change (58.33%) where Cd and Pb were incorporated into the second vital

Table 7: Non-carcinogenic risk (D_{ing} , D_{inh} , and D_{dermal}) of heavy metals for children.

D_{ing}							
Sampling sites	Cr	Ni	Cu	As	Cd	Pb	Total (Hazard index)
S1	4.70E-06	4.10E-05	2.20E-05	7.4 E-06	1.9 E-06	2.40E-05	9.17E-05
S2	1.00E-06	6.90E-05	3.00E-05	1.00E-05	1.7 E-06	4.70E-05	1.57E-04
S3	1.80E-05	4.80E-05	7.90E-05	4.7 E-06	9.0 E-07	1.30E-05	1.58E-04
S4	2.80E-05	7.70E-05	8.40E-05	1.60E-05	2.4 E-05	3.80E-04	5.85E-04
S5	5.60E-05	4.10E-04	1.70E-04	2.70E-05	7.8 E-06	9.60E-05	7.59E-04
S6	2.50E-05	5.70E-05	1.90E-04	3.10E-05	4.20E-05	1.50E-05	3.60E-04
S7	2.80E-05	5.10E-05	1.70E-04	3.00E-05	3.9 E-06	1.70E-04	4.49E-04
S8	6.20E-05	1.40E-04	7.20E-05	1.80E-05	2.50E-05	6.80E-05	3.85E-04
S9	6.20E-05	3.90E-04	2.70E-04	4.20E-05	3.9 E-06	5.80E-05	8.22E-04
S10	2.90E-05	9.70E-05	5.20E-05	3.70E-05	5.8 E-06	5.30E-05	2.68E-04
D_{inh}							
S1	1.00E-08	9.00E-08	4.90E-08	1.60E-08	4.20E-09	5.40E-08	2.20E-07
S2	2.30E-09	1.50E-07	6.50E-08	2.20E-08	3.80E-09	1.00E-07	3.50E-07
S3	4.10E-08	1.00E-07	1.70E-07	1.00E-08	2.00E-09	2.90E-08	3.60E-07
S4	6.10E-08	1.70E-07	1.80E-07	3.50E-08	5.30E-08	8.40E-07	1.30E-06
S5	1.20E-07	9.00E-07	3.80E-07	6.00E-08	1.70E-08	2.10E-07	1.70E-06
S6	5.60E-08	1.30E-07	4.30E-07	6.70E-08	9.30E-08	3.50E-08	8.00E-07
S7	6.10E-08	1.10E-07	3.90E-07	6.60E-08	8.50E-09	3.80E-07	1.00E-06
S8	1.40E-07	3.20E-07	1.60E-07	4.10E-08	5.50E-08	1.50E-07	8.60E-07
S9	1.40E-07	8.70E-07	5.90E-07	9.30E-08	8.50E-09	1.30E-07	1.80E-06
S10	6.50E-08	2.10E-07	1.20E-07	8.10E-08	1.30E-08	1.20E-07	6.00E-07
D_{dermal}							
S1	2.50E-08	2.20E-07	1.20E-07	4.00E-08	1.00E-08	1.30E-07	5.45E-07
S2	5.60E-08	3.70E-07	1.60E-07	5.50E-08	9.40E-09	2.60E-07	9.10E-07
S3	1.00E-07	2.60E-07	4.30E-07	2.50E-08	4.80E-09	7.20E-08	8.92E-07
S4	1.50E-07	4.20E-07	4.60E-07	8.70E-08	1.30E-07	2.10E-06	3.35E-06
S5	3.10E-07	2.20E-06	9.50E-07	1.50E-07	4.20E-08	5.20E-07	4.17E-06
S6	1.40E-07	3.10E-07	1.10E-06	1.70E-07	2.30E-07	8.50E-08	2.04E-06
S7	1.50E-07	2.80E-07	9.60E-07	1.60E-07	2.10E-08	9.50E-07	2.52E-06
S8	3.40E-07	8.00E-07	3.90E-07	1.00E-07	1.40E-07	3.70E-07	2.14E-06
S9	3.40E-07	2.10E-06	1.50E-06	2.30E-07	2.10E-08	3.20E-07	4.51E-06
S10	1.60E-07	5.30E-07	2.90E-07	2.00E-07	3.10E-08	2.90E-07	1.50E-06
Total	3.16E-04	1.39E-03	1.15E-03	2.13E-04	6.79E-05	9.31E-04	4.07E-03

segment (PC2) and clarifying the fluctuation of 31.77%.

Ecological risk assessment

In the present examination, pollution factor was in the diminishing request of $Cd > Pb > Ni > Cu > As > Cr$ in soils of various inspecting destinations (Table 4). The level of defilement of soils was resolved in light of the degree of contamination (Cd). The contamination level ranged from low to high level of contamination. As indicated by individual inspecting destinations, the contamination factor was in the dropping request of $S6 > S4 > S8 > S5 > S9 > S7 > S2 > S1 > S3$. On account of individual component tainting, all metals aside from Cd demonstrated the low level of sullyng. The scope of defilement for the present investigation was 1.728 to 32.193 (Table 4). Out of all inspecting locales, Cd contamination was the most astounding than other metal sullyng demonstrating that these metals may represent a

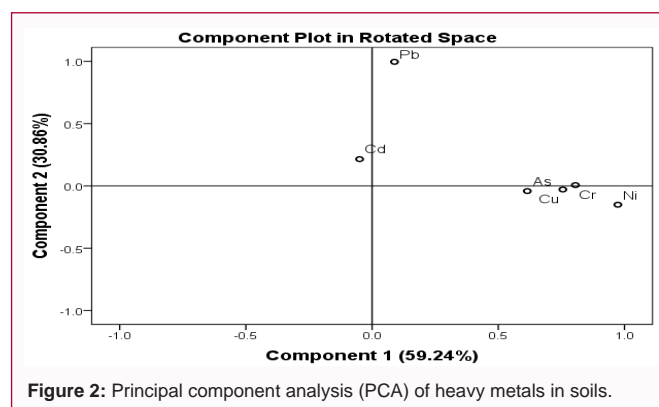


Figure 2: Principal component analysis (PCA) of heavy metals in soils.

potential hazard to the encompassing environments [100].

Table 8: Non-carcinogenic risk (D_{ing} , D_{inh} , and D_{dermal}) of heavy metals for an adult.

D_{ing}							
Sampling sites	Cr	Ni	Cu	As	Cd	Pb	Total (Hazard index)
S1	4.00E-07	3.30E-06	1.80E-06	6.00E-07	2.00E-07	2.00E-06	8.30E-06
S2	1.00E-07	5.50E-06	2.40E-06	8.00E-07	1.00E-07	3.80E-06	1.27E-05
S3	1.50E-06	3.80E-06	6.30E-06	4.00E-07	1.00E-07	1.10E-06	1.32E-05
S4	2.20E-06	6.20E-06	6.80E-06	1.30E-06	1.90E-06	3.00E-05	4.84E-05
S5	4.50E-06	3.30E-05	1.40E-05	2.20E-06	6.00E-07	7.70E-06	6.20E-05
S6	2.00E-06	4.60E-06	1.50E-05	2.50E-06	3.40E-06	1.30E-06	2.88E-05
S7	2.30E-06	4.10E-06	1.40E-05	2.40E-06	3.00E-07	1.40E-05	3.71E-05
S8	5.00E-06	1.10E-05	5.80E-06	1.50E-06	2.00E-06	5.50E-06	3.08E-05
S9	5.00E-06	3.10E-05	2.10E-05	3.40E-06	3.00E-07	4.70E-06	6.54E-05
S10	2.40E-06	7.80E-06	4.20E-06	3.00E-06	5.00E-07	4.20E-06	2.21E-05
D_{inh}							
S1	8.20E-10	7.20E-09	3.90E-09	1.30E-09	3.40E-10	4.30E-09	1.80E-08
S2	1.80E-10	1.20E-08	5.20E-09	1.80E-09	3.00E-10	8.30E-09	2.80E-08
S3	3.30E-09	8.40E-09	1.40E-08	8.10E-10	1.60E-10	2.30E-09	2.90E-08
S4	4.90E-09	1.40E-08	1.50E-08	2.80E-09	4.20E-09	6.70E-08	1.10E-07
S5	9.80E-09	7.20E-08	3.10E-08	4.80E-09	1.40E-09	1.70E-08	1.40E-07
S6	4.50E-09	1.00E-08	3.40E-08	5.40E-09	7.40E-09	2.80E-09	6.40E-08
S7	4.90E-09	9.00E-09	3.10E-08	5.30E-09	6.80E-10	3.10E-08	8.20E-08
S8	1.10E-08	2.60E-08	1.30E-08	3.30E-09	4.40E-09	1.20E-08	6.90E-08
S9	1.10E-08	6.90E-08	4.70E-08	7.40E-09	6.80E-10	1.00E-08	1.50E-07
S10	5.20E-09	1.70E-08	9.20E-09	6.50E-09	1.00E-09	9.20E-09	4.80E-08
D_{dermal}							
S1	2.10E-09	1.80E-08	1.00E-08	3.30E-09	8.50E-10	1.10E-08	4.53E-08
S2	4.60E-10	3.10E-08	1.30E-08	4.50E-09	7.70E-10	2.10E-08	7.07E-08
S3	8.30E-09	2.10E-08	3.50E-08	2.10E-09	4.00E-10	5.90E-09	7.27E-08
S4	1.20E-08	3.40E-08	3.70E-08	7.10E-09	1.10E-08	1.70E-07	2.71E-07
S5	2.50E-08	1.80E-07	7.80E-08	1.20E-08	3.40E-09	4.20E-08	3.40E-07
S6	1.10E-08	2.50E-08	8.70E-08	1.40E-08	1.90E-08	7.00E-09	1.63E-07
S7	1.20E-08	2.30E-08	7.90E-08	1.30E-08	1.70E-09	7.80E-08	2.07E-07
S8	2.80E-08	6.50E-08	3.20E-08	8.30E-09	1.10E-08	3.00E-08	1.74E-07
S9	2.80E-08	1.80E-07	1.20E-07	1.90E-08	1.70E-09	2.60E-08	3.75E-07
S10	1.30E-08	4.30E-08	2.30E-08	1.60E-08	2.60E-09	2.30E-08	1.21E-07
Total	2.56E-05	1.11E-04	9.20E-05	1.82E-05	9.47E-06	7.49E-05	3.31E-04

Geo-accumulation index (I_{geo}) of toxic elements in soils delineated in Fig. 3. Among the contemplated metals, the I_{geo} esteems were in the lessening request of $Cd > Cu > Ni > Pb > As > Cr$. For all testing locales, the scope of I_{geo} for Cr, Ni, Cu, As, Cd and Pb were -2.487 to -0.707, -0.827 to 0.175, -1.006 to 0.075, -1.3 to -0.341, -0.858 to 0.819 and -6.171 to -1.436 individually coming about uncontaminated to tolerably sullied status of the dirt. The most noteworthy I_{geo} esteem was found in Cd (0.574) in S4 of the examination territory, which may be because of the higher focus in the soil.

Potential ecological risk factor (E^i), risk index and degree of pollution of heavy metals in soils for the present study were documented in Table 5. The potential ecological risk factor of toxic elements in soils was in the descending order of $Cd > As > Pb > Ni > Cu > Cr$. As an individual metal, cadmium demonstrated the most

elevated ecological risk factor (E^i) for the present examination.

The scope of potential ecological risk factor (E^i) for Cd was 18.689 to 889.180. The individual potential natural hazard for different metals was low. Examining sites S4, S6, and S8 demonstrated the most elevated potential natural hazard factor. These sites demonstrated the most noteworthy measure of hazard factor because of Cd presentation in the open condition or utilization of phosphate manures to the rural fields [101]. The potential ecological risk index value went from 44.458 to 905.359 demonstrating low to high hazard to the dirt. Contamination factor (C^i), a degree of contamination (C_d), ecological risk (E^i) and risk index (PER) for heavy metals were reported in Table 6. From the present examination, it was seen that potential natural hazard for Cd was higher than different metals.

The probable lethality of toxic elements in soils can be assessed as

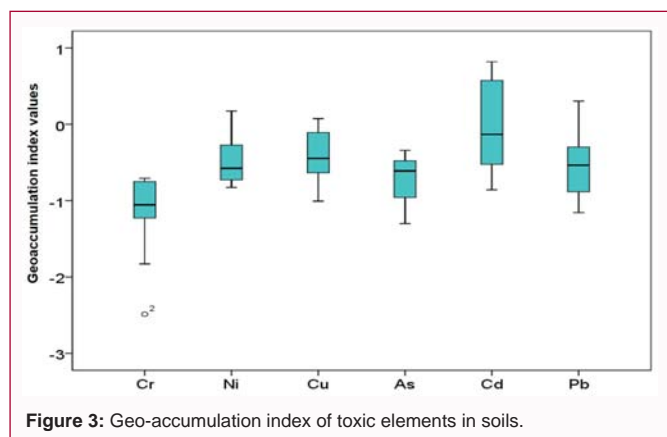


Figure 3: Geo-accumulation index of toxic elements in soils.

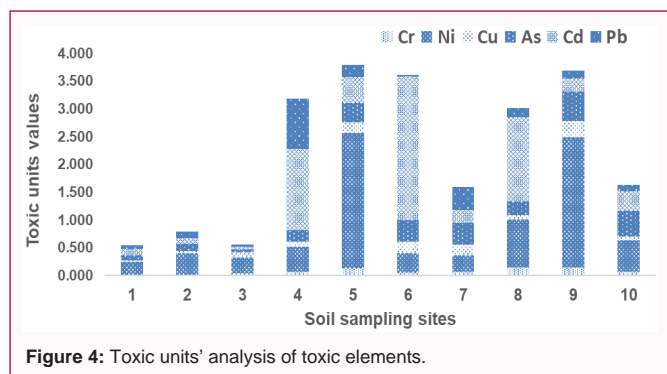


Figure 4: Toxic units' analysis of toxic elements.

the summation of toxic units (Σ TUs), characterized as the proportion of the decided centralization of toxic elements in the dirt to probable effect levels (PELs) [102]. Toxic unit (TU) and Σ TUs for harmful metals for the present investigation was showed in Figure 4. Toxic units of toxic elements in the present examination were in the decreasing order of Ni> Cd> As> Pb> Cu> Cr. The entirety of lethal units for all inspecting locales was lower than four coming about the low poisonous quality of toxic elements in soils [97].

Health risk assessment for toxic elements exposure from soil

Dangerous metals present in soil may make genuine serious health problem to human. In the present investigation, human health risk was evaluated based on ingestion, dermal contact and inward

breath of substantial metals introduction in the modern zones [103,104]. Non-carcinogenic and carcinogenic risk, the cumulative hazard index, multiple pathway exposure risks, and combined toxic elements were resolved by health risk assurance approaches.

Total non-carcinogenic risk (D_{ing} , D_{inh} , and D_{dermal}) and hazard index of heavy metals for youngsters and grown-up were displayed in Table 7 and Table 8. The non-cancer-causing hazard was assessed for knowing the non-malignancy chance status on human due to dermal contact, ingestion and inward breath of soils in the mechanical territory. For all examining locales, the non-carcinogenic health risks identified with individual component presentation through soil ingestion, inward breath, and dermal contact was low for all explored components in kids and grown-up (Table 7 and Table 8).

The consolidated impacts of uncovered metals and metalloids were estimated as hazard index (HI) and the information showed that the HI values were likewise lower than one. Be that as it may while considering the aggregate introduction HI of ingestion, inward breath and dermal contact; there was no way of having a non-malignancy chance at all of the locales for grown-ups and kids. The aggregate danger file for kids and grown-up was 4.07E-03 and 3.31E-04 separately. The hazard risk index values for youngsters were higher than that of grown-up occupants showing kids may represent a genuine non-disease chance later on. The hazard index value for kids was higher than a grown-up based on ingestion, inward breath and dermal contact. The total target hazard quotients (TTHQ) for kids was higher because of contacting and mouthing of residue tainted particles, guide ingestion by hand to mouth exercises [105].

The ingestion rate of metals was higher in youngsters than a grown-up because of little body weight than grown-up [106]. Through ingestion, youngsters have a tendency to be presented to more prominent measures of soil than grown-ups because of pica and play conduct [106-108]. The cancer-causing danger of As and Pb for grown-ups was displayed in Table 5. The Carcinogenic risk {lifetime average daily dose (LADD) ($mgkg^{-1}day^{-1}$)} of toxic elements in soils for both grown-up and kids were an adequate level. The aggregate growth hazard estimation of Cr, Ni, Cu, As, Cd and Pb for grown-up were 7.55E-09, 3.35E-08, 2.79E-08, 5.39E-09, 2.81E-09, and 2.24E-08, separately. For youngsters, the cancer-causing hazard estimation of Cr, Ni, Cu, As, Cd and Pb were 4.50E-09, 1.99E-08, 1.65E-08, 3.21E-09, 1.67E-09, and 1.33E-08 separately.

Table 9: Carcinogenic risk {lifetime average daily dose (LADD) ($mg kg^{-1} day^{-1}$)} of heavy metals in soils.

Sampling sites	Cr		Ni		Cu		As		Cd		Pb		Total	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children
S1	1.10E-10	6.70E-11	9.80E-10	5.80E-10	5.40E-10	3.20E-10	1.80E-10	1.00E-10	4.60E-11	2.70E-11	6.00E-10	3.50E-10	2.50E-09	1.50E-09
S2	2.50E-11	1.50E-11	1.70E-09	9.80E-10	7.20E-10	4.30E-10	2.50E-10	1.50E-10	4.20E-11	2.50E-11	1.10E-09	6.80E-10	3.80E-09	2.30E-09
S3	4.50E-10	2.70E-10	1.10E-09	6.80E-10	1.90E-09	1.10E-09	1.10E-10	6.60E-11	2.10E-11	1.30E-11	3.20E-10	1.90E-10	3.90E-09	2.30E-09
S4	6.70E-10	4.00E-10	1.90E-09	1.10E-09	2.00E-09	1.20E-09	3.90E-10	2.30E-10	5.80E-10	3.40E-10	9.20E-09	5.40E-09	1.50E-08	8.70E-09
S5	1.30E-09	8.00E-10	9.90E-09	5.90E-09	4.20E-09	2.50E-09	6.60E-10	3.90E-10	1.90E-10	1.10E-10	2.30E-09	1.40E-09	1.90E-08	1.10E-08
S6	6.10E-10	3.60E-10	1.40E-09	8.20E-10	4.70E-09	2.80E-09	7.40E-10	4.40E-10	1.00E-09	6.00E-10	3.80E-10	2.20E-10	8.80E-09	5.20E-09
S7	6.70E-10	4.00E-10	1.20E-09	7.40E-10	4.30E-09	2.50E-09	7.20E-10	4.30E-10	9.30E-11	5.50E-11	4.20E-09	2.50E-09	1.10E-08	6.60E-09
S8	1.50E-09	8.80E-10	3.50E-09	2.10E-09	1.70E-09	1.00E-09	4.50E-10	2.70E-10	6.00E-10	3.60E-10	1.60E-09	9.70E-10	9.40E-09	5.60E-09
S9	1.50E-09	8.90E-10	9.50E-09	5.60E-09	6.50E-09	3.90E-09	1.00E-09	6.00E-10	9.30E-11	5.50E-11	1.40E-09	8.30E-10	2.00E-08	1.20E-08
S10	7.10E-10	4.20E-10	2.30E-09	1.40E-09	1.30E-09	7.50E-10	8.90E-10	5.30E-10	1.40E-10	8.20E-11	1.30E-09	7.50E-10	6.60E-09	3.90E-09
Total	7.50E-09	4.50E-09	3.30E-08	1.90E-08	2.70E-08	1.60E-08	5.30E-09	3.20E-09	2.80E-09	1.60E-09	2.20E-08	1.30E-08	1.00E-07	5.90E-08

The outcomes demonstrated that the growing dangers of cancer-causing components were largely inside the globally acknowledged prudent paradigm (10^{-6} to 10^{-4}) [60]. The cancer-causing dangers of contaminated metals introduction from mechanical zones soil by means of ingestion, dermal contact, and inward breath pathways were lower than the acceptable limit. Along these lines, the present examination uncovered that there was no such kind of cancer risk in both grown-up and youngsters.

Conclusions

From the findings, it was found that heavy metals are most probably industrial origin. All the metals were recorded below the contamination level suggested by different international organizations (e.g. Dutch standard, Canadian guidelines, and Australian guidelines). Cu and Cd was the exception that was found above the safe limit. Different analysis (e.g. contamination factor and potential ecological risk) demonstrated that metals were below the pollution level. Ingestion, inhalation and dermal contact of the metals in adult and children in the study area have no possibility to pose a cancer risk. But the concern is that long-term exposure of these metals can pose cancer both in child and adult.

Acknowledgements

Authors are grateful to the Yokohama National University, Japan and Patuakhali Science and Technology University (PSTU), Bangladesh for providing laboratory facilities to complete this study.

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