“Assessment of Heavy Metal Pollution in Soil of Chattogram Hill Tracts, Bangladesh and Concomitant Health Risk”

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Abstract

The increased human population and associated activities may create a risk in the ecological balance of Chattogram Hill Tracts (CHT), Bangladesh via contamination of soil with toxic heavy metals. Thus, the present study was conducted to assess the concentration of heavy metals (Lead, Cadmium, Copper, Zinc, Iron, Manganese, Chromium and Nickel) in forest soils of the CHT area by using an atomic absorption spectrometer. The degree of contamination of soil was evaluated by five indices: geo-accumulation index (I_{geo}), enrichment factor (EF), contamination factor (CF), pollution load index (PLI) and potential ecological risk index (PERI). According to these criteria, these soils can be classified as moderately contaminated with some metals. Furthermore, the identification of pollution sources based on principal component analysis and hierarchical cluster analysis have revealed that all analyzed metals are anthropogenic except Fe. Calculated hazard index >1 indicates the possibility of noncarcinogenic effect due to higher value of Fe. Carcinogenic risks through the ingestion, inhalation and dermal pathway for carcinogenic elements (Pb, Cd, Cr, and Ni) shows a non-significant risk ($CR < 10^{-6}$) for both children and adults living in the studied area. Measured data may help the policymakers to reduce the potential effects of soil contamination on eco-environment and human health.

Keywords: Chattogram Hill Tracts, soil pollution quantification, enrichment factors, Hierarchical cluster analysis, Ecological risk, Carcinogenic and Non-carcinogenic risk.

1. Introduction

Due to their potential toxicity and non-biodegradability, heavy metals (HMs) are recognized as a major inorganic pollutant for ecological and health concern, hence pose a non-negligible threat to living things (Ayangbenro and Babalola, 2017). All metals in the biosphere, regardless of essential and non-essential ones, find their ultimate

ways to soil following the life-cycle of living beings and the natural cycle of non-living things. As a result, the overall metals cycle in nature is mediated by soil (Gbadamosi et al., 2018). One of the main ways of soil pollution by inorganic pollutants is via the man-made/anthropogenic/artificial activities including the exploration and extraction of mineral resources from earth crusts. Consequently, the metals available in the ecosystem can be easily transferred to food chains via geo-accumulation and bioaccumulation process, finally causes a greater risk to human health (Pejman et al., 2015).

The Chattogram Hill Tracts has been suggested as one of the most diverting regions of Bangladesh. The land area of CHT has been classified as reserve forests (> 394,300 ha), shifting cultivation (> 905,000 ha), and unclassed (the remaining area) (Ahammad and Stacey, 2016). A total of 11 Ethnic groups of people live in this region (UNICEF Bangladesh, 2019). Due to the increasing human population in CHT area day-by-day (i.e., the migration of peoples from other places of Bangladesh to Rangamati, Khagrachari and Bandarban), it sees a large scale anthropogenic activities including deforestation. The main cause of deforestation is the shifting of traditional cultivation (Jhum cultivation), and unplanned development activities (Ahammad and Stacey, 2016; Osman, 2014). The quality of forest soil is naturally acidic, has multi-layer with forest floor, hemi-organic and mineral layers (Hassan et al., 2017; Osman, 2014). The soils of CHT are mostly sandy (37 to 67%) and the rest of them are silt and clay [6]. Though the soils of CHT is not suitable for agriculture, frequent use of the same land towards biomass production reduces soil fertility and increase the level of toxic metals and various pollutants (Biswas et al., 2010). Also, contamination of soil including metal concentration is hampered by some other factors such as soil properties, such as pH, organic matter content, minerals, cation exchange capacity of the soil phase, balancing with other metal ions etc. (Alfaro et al., 2015; Perković et al., 2017).

The higher concentration of heavy metals represents the negative influence on the forest ecosystem (Wahab et al., 2020). The high concentration of some metals such as Cu, Zn, Ni, Mn, Co, Se, Ag, Sb, and notably Cr and Ti pose toxic effects to human and animal health via the food chain (Perković et al., 2017). Based on the United States Environmental Protection Agency (USEPA, 2000) report, ingestion, inhalation and dermal contact are the main pathways for metal exposures to dwelling populace. Especially, the major cause of cancer risk is due to exposure of Arsenic, Nickel and Cadmium (Alfaro et al., 2015). Besides, Cr(VI) pose high toxicity and carcinogenic risk to human through exposure by inhalation (US EPA, 2011). Furthermore, chronic ingestion of Cd and excessive intake of Pb can cause negative health impacts like kidney dysfunction, bone fractures and hypertension etc. Therefore, excessive metal exposure is hazardous to human health.

It is a vital issue to investigate human exposure to heavy metals in CHT soils to explore the adverse effects to the dwellers and to protect human health. Generally, several steps should be followed to assess the chemical hazards like identification, characterization, exposure assessment and risks characterization (Wild Thurst of Bangladesh, 2010). However, the prior task is the accurate determination of metal levels in the soils of interest for which chemical hazards should be investigated.

Numerous studies of the three districts separately are available on the metal contamination in soils, water and sediments, however, this study has focused on the assessment of the quality of soils combinedly in Rangamati, Khagrachari and Bandarban of CHT area (Barua et al., 2019; Biswas et al., 2010; Osman, 2014; Wang et al., 2016). The study not only evaluated eight different metals such as Lead, Cadmium, Copper, Zinc, Iron, Manganese, Nickel and Chromium but also many well-defined hazards characterization parameters/indices such

as geo-accumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), enrichment factor (EF) and potential ecological risk index (PERI) were used to assess environmental risk. In the same vein, this study also assessed the non-carcinogenic and carcinogenic health risk for human beings from concerned metal elements. Besides, multivariate statistical analysis is used to understand the behaviour of trace elements in the studied areas by dividing a set of data objects into groups of similar characteristics. So, the present study is a comprehensive one where pollution parameter in soil was analysed in various ways.

To ensure a safer eco-system and health of the dwelling populace, especially the less educated ethnic peoples who depend on the natural resources of the CHT area for their livelihood, it is crucial to evaluate the quality of hilly soils w.r.t. the metal levels of carcinogenic and non-carcinogenic category. In this regards, our study was aimed to establish a benchmark of quality for Pb, Cd, Cu, Zn, Fe, Mn, Ni, and Cr in the soil of CHT that gives a base for monitoring metal levels in soils. [10].

2. Methodology

2.1. Description of the study area

The south-eastern part of Bangladesh is a wide-ranging hilly area, known as Chattogram Hill Tracts (CHT). It is occupied by 13,294 sq km of the area [5]. Most of the area of CHT is humid tropical rainforests with a high diversity of flora and fauna. It is located between 21°25' N to 23°25' N latitude and 91°54' E to 92°50' E longitude [6], [15]. The present study area consists of three distinct geographical and cultural landscape districts which are Rangamati (Rm) (22°37'60N-92°12'0E), Khagrachari (Kc) (23°04.051N-91°87.566E) and Bandarban (Bb) (22°11.121N-92°23.416E) [13]. The CHT covers 43% forest area of Bangladesh (Bangladesh Forest Department, 2016). More than 70% area in both natural and plantation forests of Bangladesh covers by the CHT areas [4].

2.2 Sample collection

A representative number of topsoil samples (n=37) (up to 20 cm deep from the top surface) were collected from the studied area. The distance between two consecutive sampling locations was 200-250 m depending primarily on local terrain and accessibility. The geographical coordinates (latitude and longitude) of the location of each sampling point were recorded using a GPS of google map. The collected soils samples are mainly of yellowish-brown to reddish-brown loams. The collected samples were brought to the laboratory of Atomic Energy Centre, Chattogram for subsequent preparation and analyses.

2.3 Sample Preparation

The collected soil samples were cleaned by removing any foreign media. A large sieve was used to remove stones, rocks, concrete, roots or other organic matter from the soil. The collected soil samples were sun-dried for one week and then kept in acid-washed containers and placed in an oven at 50°C for 48 hours. Soil samples were then fully crushed to the finest possible fraction using acid-washed pestle and mortar then clear 2 mm sieve. Later, samples were kept in zip bag separately and stored in desiccators.

2.4 Sample digestion

5 g of each soil sample was picked by a high precision analytical balance and then transferred to an acid-washed glass beaker. Then samples were weighted and added 15 mL HNO₃ (69 %). Then it was placed in an electric heater at 60°C for half an hour. The beaker covered with a watch glass. The electric hot plate was used for heating as it regulates the temperature better and distributes the temperature evenly. After then the temperature range was increased up gradually to 100°C until the concentration becomes half of the initial volume (Aga and Brhane, 2014).

After cool down the samples to room temperature, they were filtered (0.8 µm) through a glass funnel containing Whatman No.1 filter paper. Then the solutions were diluted with deionized water to a final volume of 50 mL (Lubomir Svoboda, 2016) and stored in 50ml plastic centrifuge test tube vial container.

2.5 Analytical procedure

The acid digested triplicates samples were analyzed for the determination of heavy metals (Pb, Cd, Cu, Zn, Fe, Mn, Ni and Cr) by atomic absorption spectrophotometry (AAS). Results (in ppm) are expressed on a dry weight basis of each element. A blank solution was also run under the same analytical conditions without the addition of Pb, Cd, Cu, Zn, Fe, Mn, Ni and Cr.

For digesting composts, the nitric acid procedure was used into a traditional open-vessel digestion system for the sake of recovering the analysis, saving money, efficiency and hazardousness (Material, 2017). The metal ions on prepared sample solutions were characterized by Z-2000 polarized Zeeman atomic absorption spectrophotometer to determine the amount of investigated metal ions.

2.6 Standard Solutions

Analytical grade reagents were used in this study. Purified de-ionized water was used for all sample preparation and dilution purposes throughout our study. Standard solutions of the studied metals were made based on dilution of certified standard solutions of corresponding metal ions (Khandaker et al., 2015). The standard solutions were used in preparing subsequent calibration curves after serial dilutions. After finishing the analytical work, all the glass apparatus were immersed into the dilute HNO₃ for overnight and then washed with deionized water to remove impurity repeatedly. Four standard solutions of different known concentrations were prepared and the corresponding concentration in the unknown sample was deduced by estimation from the calibration curve. Standard unit, mg/kg was used to report the concentration of the measured samples (Stihi et al., 2011).

$$\text{Concentration (mg/kg)} = \frac{(\text{mg /kg in sample solution}) \times 50}{\text{sample weight in grams}} \quad (2.1)$$

The concentration of Fe, Mn and Zn metals in soil were calculated using the formula below (Perkin Elmer, 1996).

$$\text{Concentration (mg/kg)} = \frac{\text{CM} \times \text{DF} \times \text{NV}}{\text{SV}} \quad (2.2)$$

Where: CM = Concentration of metal, DF = Dilution factor, NV = Nominal volume, SV= Sample volume (mL).

2.7 Soil pollution quantification

Several methods are available for assessing the degree of metal concentration in soil. Some common methods are used to compare the identified concentrations with a background level or with a concentration posing serious environmental risks. Five indices were used to evaluate the quality of the studied pollutants in the hilly soils.

2.7.1 Geo-accumulation Index (I_{geo})

Geoaccumulation Index (I_{geo}) is used, as an indicator, for assessing the intensity of anthropogenic contaminant level on surface soil and to evaluate the pollution in the terrestrial, aquatic and marine environment. The geo-accumulation index is expressed as follows(Zinkutė et al., 2017):

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5 B_n} \right) \quad (2.3)$$

Where, C_n : the measured concentration of heavy metal (n) in the soil,

B_n : the geochemical background value in average shale of element n,

1.5 is the background matrix correction factor detecting very small anthropogenic influence (Muzerengi, 2017). Here, the chemical composition of unaffected soil at a remote/nondisturbed site of the study area was chosen as the background value.

The I_{geo} is identified with a qualitative range of pollution intensity for seven groups of pollution from unpolluted ($I_{geo} \leq 0$) to extremely polluted ($I_{geo} > 6$) and stated for the quality of soils based on I_{geo} values. Geo-accumulation index has seven classifications (Muzerengi, 2017) depending on its value: < 0 (unpolluted), $0-1$ (no or minimally polluted), $1-2$ (moderately polluted), $2-3$ (moderately polluted to polluted), $3-4$ (polluted to strongly polluted), $4-5$ (strongly polluted), $5-6$ (strongly polluted to very strongly polluted), > 6 (very strongly polluted).

2.7.2 Contamination factor (CF)

Contamination factor (CF) is known as a single pollution index (PI). It exposes the anthropogenic input in elemental pollution (Pandey et al., 2016). It measures the overall contamination level of a sampling site by heavy metals. It can be calculated by taking the ratio of measured concentration for each metal to the background values (Muzerengi, 2017; Sana 'a Odat, 2013):

$$CF = \frac{C_{metal}}{C_{background\ value}} \quad (2.4)$$

Where, C_{metal} : the metal concentration in polluted soil,

$C_{Background\ value}$: the background value of that metal.

The CF being classified into four grades (Pandey et al., 2016) for monitoring the pollution of one single metal over a while: < 1 (low degree), 1 to < 3 (moderate degree), 3 to < 6 (considerable degree), ≥ 6 (very high degree).

2.7.3 Pollution load index (PLI)

The pollution load index (PLI) is a simple statistical technique. It provides major information about metal pollution in the soil samples. The PLI is calculated from the geometric mean of the CFs of measured elements (Abdulgaderismaeel and Kusag, 2015; Zinkutė et al., 2017).

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n} \quad (2.5)$$

Where, CF : the contamination factor,

n : the number of studied metals.

By using PLI, the degree of pollution is classified as follows: > 1 (pollution i.e. deterioration of soil quality), <1 (indicates no pollution), 1 (baseline pollution) (Abdulqaderismaeel and Kusag, 2015; Zinkutė et al., 2017).

2.7.4 Enrichment factor (EF)

The enrichment factor (EF) indicates the concentration of any metal against a reference value. A geochemically distinguishing element and consisting of a high concentration in the environment can be used as a reference element. In this study, iron (Fe) is employed as the reference element which is used for normalization (Afzal et al., 2014; Pandey et al., 2016).

$$EF = \frac{(\text{Element concentration})/(\text{Fe concentration})_{\text{Sample}}}{(\text{Element concentration})/(\text{Fe concentration})_{\text{Shale}}} \quad (2.6)$$

To determine both natural and anthropogenic metal source and the degree of anthropogenic influence, EF is widely used. On the other hand, the value of EF > 2 indicates an anthropogenic source of metal enrichment, and based on this value, EF is further classified as 2 –5 (moderate enrichment), 5 –20 (significant enrichment), > 40 (extremely high enrichment).

2.7.5 Potential ecological risk index (PERI)

The potential ecological risk caused by heavy metals in the soil can be evaluated by using the potential ecological risk index (PERI, or RI) (Gbadamosi et al., 2018). This method evaluates four parameters such as concentration, pollutant types, level of toxicity and sensitivity of the water body in the soil (Gbadamosi et al., 2018). PERI is calculated as:

$$PERI = \sum_{i=0}^n E_r^i = \sum_{i=0}^n T_r^i \times C_f^i \quad (2.7)$$

where E_r^i = the potential ecological risk factor of each heavy metals.

T_r^i = the toxic response factor of heavy metal, i. The toxic response factor for Pb, Cd, Cu, Zn, Fe, Ni and Cr are 5, 30, 5, 1, 1, 1, 5 and 2 respectively (Gbadamosi et al., 2018; Huang et al., 2020).

C_f^i = the contamination factor of heavy metal, i.

The E_r^i is described in five categories: < 40 (low), 40 to < 80 (moderate), 80 to < 160 (considerable), 160 to < 200 (high), < 320 (very high) (Fiori et al., 2013). PERI represents the total value of all measured heavy metal's potential ecological risk factors (E_r^i) in four categories: < 150 (low), 150 to < 300 (moderate), 300 to < 600 (considerable), and > 600 (very high) (Gbadamosi et al., 2018).

2.8 Multivariate statistical analysis (MSA)

With the use of MSA, one can easily determine the underlying structures of large datasets and also identify the latent factors that may help to understand the sources of metals pollution/contamination (Hou et al., 2017). It includes mainly principal component analysis (PCA) and cluster analysis (CA), was performed with IBM SPSS (Statistical Programme for Social Sciences) V26.0 software for PCA tools.

2.8.1 Principal component analysis (PCA)

The PCA is a data dimension reduction technique. It aims to interpret most of the variance in the data with a small number of independent variables (principal components) (Hou et al., 2017). It also used to identify a set of factors present in the data. Factors were rotated using the varimax method which simplified the understanding of output results by decreasing the number of variables that loaded to high loads on each component. The Kaiser-Meyer-Olkin Measure of Sampling Adequacy (KMO) and Bartlett's Test of Sphericity normalization were used where the factors are independent of each other.

2.8.2 Hierarchical Cluster analysis (HCA)

HCA was used to identify analogous behaviour among the different parameters. It is the foremost statistical method to identify homogeneous groups of cases based on the measured characteristics (Zolfaghari et al., 2019). The squared Euclidean distance is used to address how to distance the variables, it is essential to select the clustering algorithm to lead the distances of the points that are determined to specify cluster membership. Ward's method is the most frequently used algorithms to analyse the variance approach in assessing the inter-clusters distances.

2.9 Potential Health Risk Assessment

2.9.1 Non-carcinogenic risk

The US-EPA developed models to calculate the detrimental impacts of heavy metals on human health considering carcinogenic and non-carcinogenic risk (Gbadamosi et al., 2018; Jia et al., 2018; Wahab et al., 2020). Potential risk assessment posed by heavy metals from the soils through multiple routes of exposure is divided into four steps: identification of hazards, dose-response assessment, assessment of exposure, and characterization of risk (Gbadamosi et al., 2018; Gržetić and Ahmed Ghariani, 2008).

In the present case, Pb, Cd, Cu, Zn, Fe, Mn, Ni and Cr were identified as the potentially hazardous agents in the soil of CHT. By using the average daily dose (ADD), three main pathways for heavy metals exposure to human were considered here: direct ingestion of soil particles (ADD_{ing}), inhalation of suspended soil particles from the air through nose and mouth (ADD_{inh}), and absorption of heavy metals from skin adhered soil, called as dermal contact (ADD_{dermal}).

The ADD (mg kg⁻¹ day⁻¹) can be estimated by using the equations (2.8)–(2.10) (Gbadamosi et al., 2018; Jia et al., 2018; Wahab et al., 2020):

$$ADD_{ing} = \frac{C_{soil} \times R_{ing} \times EF \times ED}{BW \times AT_{noncarc}} \times 10^{-6} \quad (2.8)$$

$$ADD_{inh} = \frac{C_{soil} \times R_{inh} \times EF \times ED}{PEF \times BW \times AT_{noncarc}} \quad (2.9)$$

$$ADD_{derm} = \frac{C_{soil} \times SA \times SL \times ABS \times EF \times ED}{BW \times AT_{noncarc}} \times 10^{-6} \quad (2.10)$$

The values of all parameters in Eqs. (2.8-2.10) are given in Table 1. By using the obtained values of ADDs, the non-carcinogenic risk of heavy metals in CHT soil was evaluated via a parameter called hazard quotient (HQ) for a single substance, or (ΣHQ) for multiple substances:

$$HQ = \frac{Add_{ing+inh+derm}}{RfD} \quad (2.11)$$

Where RfD stands for reference dose, values are available in Table 3. The cumulative non-carcinogenic risk for multiple substances can be expressed as a hazard index (HI) [14] [34]:

$$HI = \sum HQ_{ing+inh+derm} \quad (2.12)$$

It is considered as 'no significant risk' for $HI \leq 1$, and indicates the probability of occurrences of non-carcinogenic effects for $HI > 1$, and it increases with an increasing value of HI (Gbadamosi et al., 2018).

2.9.2 Carcinogenic risk (CR)

The individual lifetime cancer risk can be calculated by CR. It can be obtained by multiplying lifetime average daily dose (LADD) (mg/kg-day) associated with ingestion, inhalation and dermal contact exposure for children and adult with the cancer slope factor (SF) (mg/kg-day), as follows:

$$CR = LADD \times SF \quad (2.13)$$

The following equations are applicable to determine the LADDs (Johnbull et al., 2019; US EPA, 2011; Wahab et al., 2020):

$$LADD_{ing} = \frac{C_{soil} \times R_{ing} \times EF \times ED}{BW \times AT_{carc}} \times 10^{-6} \quad (2.14)$$

$$LADD_{inh} = \frac{C_{soil} \times R_{inh} \times EF \times ED}{PEF \times BW \times AT_{carc}} \quad (2.15)$$

$$LADD_{derm} = \frac{C_{soil} \times SA \times SL \times ABS \times EF \times ED}{BW \times AT_{carc}} \times 10^{-6} \quad (2.16)$$

Table 1 listed the necessary formation of the parameters in Eqs. (2.14-2.16). SF associated with the ingestion of Pb, inhalation of Pb, Cd, Ni and Cr, and dermal of Pb. For dermal exposure, cancer slope factor (SF) is not available, but it can be found by multiplying the oral cancer slope by the gastro-intestinal absorption factor (Johnbull et al., 2019). All health risks are cumulative [14], [34], therefore the total cancer risk (lifetime carcinogenic risk) was calculated by summing the individual cancer risk as follows (Jia et al., 2018).

$$TCR = \sum CR = CR_{ing} + CR_{inh} + CR_{derm} \quad (2.17)$$

The carcinogenic risks to human health from the soil can be considered negligible ($CR < 10^{-6}$), non-negligible ($CR = 10^{-6}$ to 10^{-4}), and a high risk for the development of cancer in humans ($CR > 10^{-4}$) (Weissmannová et al., 2019).

Table 1: Variables and parameter of exposure applied in health risk assessment calculation (US EPA, 2011).

Variable	Definition	Value used		Reference
		Children	Adult	
C_{Soil} (mg/kg)	Concentration of metal in soil			
R_{ing} (mg/day)	Ingestion rate	200	100	(Wahab et al., 2020)

AT _{noncarc}	Averaging time for non-carcinogens	365 days/year/ × ED	365 days/year × ED	(Wahab et al., 2020)
BW	Average body weight	15 kg	70 kg	(US EPA, 2011; Wahab et al., 2020)
EF	Exposure frequency	350 days/year	350 days/year	(Wahab et al., 2020)
R _{inh}	Inhalation rate	7.6 m ³ /day	20 m ³ /day	(Wahab et al., 2020)
PEF	Particle emission factor	1.36×10 ⁹ m ³ /kg	1.36×10 ⁹ m ³ /kg	(Gbadamosi et al., 2018; Wahab et al., 2020)
AT _{carc}	Averaging time for carcinogens	25550 days	25550 days	(US EPA, 2011; Wahab et al., 2020)
SA	Surface area of the skin that contacts the soil	2800 cm ²	5700 cm ²	(US EPA, 2011; Wahab et al., 2020)
SL	Skin adherence factor for the dust	0.2mg/cm ²	0.07 mg/cm ²	(Wahab et al., 2020)
ED	Exposure duration	6 years	24 years	(US EPA, 2011; Wahab et al., 2020)
ABS	Dermal absorption fraction	0.001	0.001	(Wahab et al., 2020)

3. Results and Discussion

3.1. Level of heavy metals in soil samples

Accumulation of an excessive amount of heavy metals in the organic layer and topsoil over a long period eventually contaminate the soil organisms, hence affect the sustainability and productivity of terrestrial ecosystems (Chrzan et al., 2013).

In general, a pH value of > 4.0 helps in the accumulation of Pb in the organic and upper mineral layer of soil (Rademacher, 2001). The Pb concentrations (mg/kg) in the studied sample were ranged from 3.8±0.004 to 32.7±0.071 which was lower than the world average (2 - 300 mg/kg) (Alfaro et al., 2015). The concentration of Cd (mg/kg) was found between 0.03 mg/kg to 0.25 mg/kg which was lower value compared with the world average (0.01-2.0 mg/kg) (Alfaro et al., 2015). An unusual of the highest level of Cu was found 172.5±0.003 mg/kg in Rm-3 site and the smallest level of 3.9±0.1 mg/kg was found in Rm-6. The world average for Cu in the soil is 2-250 mg/kg (Alfaro et al., 2015).

The highest and significant different value was available at Rm-2 (202.60 mg/kg). The minimum value of Zn found in Kc-6 area was 13.6±0.04 mg/kg. The world average of zinc in soils is 1 – 900 mg/kg (Alfaro et al., 2015). Fe was determined in a range of 14004.6±82.63 mg/kg - 108354.6±162.53 mg/kg in the studied samples. The

literature revealed that iron concentrations vary with the depth of the soil (20,000 to 550,000 mg kg⁻¹) (Anumalla Mahender, B. P. Mallikarjuna Swamy, 2019). Manganese content in this study was in the range of 1289.9±12.14 mg/kg (sample Rm-7) - 73.4±1.05 mg/kg (sample Kc-3) in high hill soils. The world average of manganese content is 540 mg/kg (Pandey et al., 2016). A significant difference and the highest concentrations of Ni (38.12±0.206 mg/kg) was found at Bb-8 area whereas the minimum value of Ni was 1.91±0.003 mg/kg. The world average range of Ni is 2-750 mg/kg (Alfaro et al., 2015). The value of Cr in the soil samples was found to be in the range of 4.2±0.004 and 70.5±0.317 mg/kg. The world average of Cr is 48 mg/kg [11].

Overall, the soil in samples collected from three districts was less contaminant by all studied metals and less harmful to the environment (Rademacher, 2001).

3.2 Assessment of soil contamination

The degree of heavy metals contamination in the CHT soils was assessed by some indices, and they are discussed here.

3.2.1 Geo-accumulation index (I_{geo})

The value of I_{geo} indicates the presence and also the intensity of anthropogenic contaminants deposition on the soil. Overall metal contamination level in the soils shows in Table 3 was calculated by the I_{geo} . The mean I_{geo} values for all metals were found in the range of -5.74 to 1.35, which indicates a non-contamination of some soils and some are unpolluted to moderately polluted. Rm-3 is moderately polluted because of copper concentration (Muzerengi, 2017).

3.2.2 Contamination factor (CF)

Table 2 shows the calculated CFs in the range of 0.97 - 3.83, which indicates that some metals exceeded the background level. The CF showed considerable contamination by Zn and Fe, moderately contamination by Pb and Mn, and practically no contamination by Cd, Ni and Cr (CF<1.5). Based on the mean values of CF, soils were enriched with metals in the following order: Fe > Zn > Cu > Mn > Pb > Cd > Ni > Cr. This indicated that the soils at Rm-3 and most of the sites of Bandarban have been largely polluted by Fe, Zn and Cu. Overall, all calculated metals showed a moderate degree of contamination ($1 \leq CF < 3$) except CF of Cu (3.83). The calculation revealed that Fe has the highest pollution and the CF value of Cu in Rangamati district (specially Rm-3) was high (Širić et al., 2016).

3.2.3 Pollution load index (PLI)

The spatial distribution of metal contaminants in the study area was demonstrated by the pollution load index and is shown in Table 2. The PLI values were ranged from 0.27 to 0.62 in Rangamati district, from 0.15 to 0.42 in Khargachari district and from 0.32 to 0.75 in Bandarban district. Calculated values (PLI < 1) confirmed that the soil of the studied area was less contaminated by metals.

3.2.4 Enrichment Factor (EF)

The result of EFs (shown in Table 2) indicated a homogeneous distribution of the studied metals throughout the sample areas. It was found that the soil samples of most of the area were depletion to mineral enrichment with the

metals. The EF values for Cu and Zn were found not exceeding the level of moderate enrichment ($EF < 5$). The maximum EF value of Cu was 4.99, consequently signifying soil enrichment by this metal. According to the enrichment factor, high concentration of Cu in the soil of Rm-3 area is harmful to plants and can be transmitted to human beings through the food chain. So, Rm-3 was extremely enriched by Cu with significantly high enriched by Pb (1.22), Cd (1.09) and Zn (2.73). Besides, the soil of Rm-3, Kc-14 was found to be moderately enriched by Pb, Cd, Cu and Zn (Ahammad and Stacey, 2016; Miah et al., 2010).

Table 2: Descriptive statistics of Geo-accumulation Index, contamination factor, pollution load and enrichment factor index for the CHT area.

Descriptive statistics	Pb	Cd	Cu	Zn	Fe	Mn	Ni	Cr	
Geo-accumulation Index (I_{geo})									
Max.	0.12	-0.85	1.35	0.51	0.61	0.02	-1.42	-0.94	
Min.	-2.98	-4.49	-4.11	-3.39	-2.34	-4.12	-5.74	-5.08	
Mean	-1.78	-1.87	-1.76	-1.62	-0.57	-2.12	-3.39	-2.95	
Contamination factor (CF)									
Max.	1.64	0.83	3.83	2.13	2.30	1.52	0.56	0.78	
Min.	0.19	0.07	0.09	0.14	0.30	0.09	0.03	0.04	
Mean	0.52	0.47	0.58	0.59	1.11	0.43	0.17	0.27	
Pollution load index (PLI)									
Max.	...	...	...	...	...	...	...	...	0.75
Min.	...	...	...	...	...	...	...	...	0.15
Mean	...	...	...	...	...	...	...	...	0.40
Enrichment factor (EF)									
Max.	1.23	1.12	4.99	2.73	1.00	1.76	0.49	1.61	
Min.	0.25	0.04	0.10	0.15	1.00	0.10	0.06	0.05	
Mean	0.49	0.50	0.63	0.62	1.00	0.39	0.16	0.29	

According to the four pollution indices, it is observed that soil samples of Rangamati, Khagrachari and Bandarban occupied background concentrations for the studied elements, and no or unchanged by anthropogenic influences in true sense. However, some calculated values of pollution parameter for the different area were found relatively higher than others due to the influence of shifting agriculture, agricultural runoff and other anthropogenic inputs. Regular Joom (shifting) cultivation in CHT areas is also considered as one of the main reasons for forest degradation (Ahammad and Stacey, 2016; Miah et al., 2010).

3.2.5. Potential ecological risk index (PERI)

Figure 1 graphically shows the calculated potential ecological risk index of our study. Samples from Bandarban area was found to pose the highest ecological risk ($300 \leq RI < 600$), while in Rangamati and Khagrachari area posed a moderate risk ($150 \leq PERI < 300$). Based on the PERI value, Cd of the Rm, Kc and Bb area were found to have a high potential risk (E_r^i) i.e. 160, 181 and 185 respectively in the soil. The PERI of our data indicated that the potentiality of pollution of the studied heavy metals decreased by the sequence of

Cd>Cu>Pb>Fe>Ni>Zn>Cr>Mn. The sources of heavy metal pollution in Chattogram Hill Tracts might be responsible for shifting cultivation and chemical industry such as rubber factory (Ahammad and Stacey, 2016; Miah et al., 2010).

Fig1 Potential ecological risk indices (PERI) in the soil of CHT.

3.3 Multivariate statistical analysis

3.3.1 Principal component analysis (PCA)

In figure 2, the PCA with consistent data was used to interpret effectively the metal values determined in soil samples. Based on the eigenvalues greater than 1, three major components were identified, and it explains 70.850% of the system variance. As shown in figure-2, the variance of first principal component (PC-1) was 34.14% and eigenvalue was 2.731, showed strong positive loading in Fe (0.924), Ni (0.808), Mn (0.815) and moderate positive loading in Pb (0.620). Due to the highly positive loading of Fe and Mn, PC-1 could be clarified as natural sources (lithogenic activities)(Baltas et al., 2020). Ni contamination of soils resulted from the anthropogenic activities of plating works (Pandey et al., 2016).

The variance and eigenvalue of PC-2 were 21.151 % and 1.692 that showed strong positive loading in Cu (0.920) and Zn (0.738) and moderate positive loading in Pb (0.528). PC-2 was suggesting that Cu and Zn from the lithogenic origin are affirmed to show close geo-chemical dependence as the iron family (Huang et al., 2020). Animal manure in the CHT forest soils is suggested to cause a significant increase in Zn levels (Baltas et al., 2020). Moreover, Lead was distributed between the two-component (PC-1 and PC-2) which is strongly bound by the organic matter of the surface soil layer. Pb isotopes are significant tools to control the respective contribution of lithogenic and anthropogenic sources in soils (Hernandez et al., 2003).

The variance and eigenvalue of PC-3 were 15.568% and 1.245. The high loading factors detected for Cd (0.754) and Cr (0.744) in the PC-3 component. The anthropic inputs Cr showed a mixture source of both lithogenic and human origin in our study area (Baltas et al., 2020). The utmost toxic element, Cd which has high ecological risks are the source of pollutants. Extensive use of fertilizers (phosphate) is responsible for the variation of Cd (Zhang, 2016).

Overall, it is suggested that Pb, Cd, Cr and Ni are extremely affected by anthropogenic sources such as rubber factory, reverse cultivation, local combustions and applications of manure and agrochemicals in the study area

(Helena Doležalová Weissmannová , Silvie Mihočová, 2019; Rastegari Mehr et al., 2017). This study indicates a statistically significant difference between the soil samples in the investigated area.

Fig2. PCA loading plot of studied heavy metals in rotated space.

3.3.2 Cluster analysis

The dendrogram (See in Fig. 3) connected to the analyzed metals has shown four main classes. The first class has been represented by Mn-Ni, Pb and Fe, indicating a mixture of natural, lithogenic and anthropogenic sources in the studied area. Whereas lead is strongly bound by the organic matter of the surface forest soil layer (Chrzan et al., 2013) as well as nickel depends on the mineralogical structure of the soil. Nickel and lead are strongly influenced by iron oxide and hydroxides in the studies of soil samples (Chrzan et al., 2013; Kobierski and Dabkowska-Naskret, 2012).

The second class has been defined by Cu and Zn, indicating the anthropogenic sources. The third and fourth classes have been defined by Cd and Cr which were completely isolated and discriminated the anthropogenic sources (Gbadamosi et al., 2018). Cu and Zn developed a sub-cluster that illustrated the major similarity with the cluster -1, also pointing to a similar source of pollution of the soil.

The results of the HCA analysis illustrated the same pollution sources as the origin of grouped metals. The weathering of parent materials may contribute to the high concentration of Mn and Fe. The concentration of Fe and Mn indirectly indicates the presence of Fe or Mn oxides in surface soils that influenced heavy metals behaviour in soils. (Helena Doležalová Weissmannová , Silvie Mihočová, 2019). Present results of HCA and PCA are supportive of the heavy metal contents with soil physicochemical characteristics. The difference between lithogenic and anthropogenic origin might be special effectiveness in case of risk assessments, deciding on remediation or apply restrictions to use the contaminated soil, information on the differences between lithogenic and anthropogenic origin is required.

Fig3 Dendrogram showing hierarchical cluster analysis of analyzed metals of the studied area.

3.4 Health Risk Assessment

Exposure to metals might be responsible for detrimental health effects. The routes of exposure to heavy metals in soil samples of Rangamati, Khagracha and Bandarban include ingestion, inhalation and dermal contact. The calculated values of ADD ($\text{mg kg}^{-1}\text{day}^{-1}$) of three exposure pathways; ingestion, inhalation, and dermal contact were followed the Exposure Factors Handbook (Table 1) (US EPA, 2011).

3.4.1 Noncarcinogenic risk

The Reference dose of different metals mentioned in Table 3 was followed by previous literature (Afzal et al., 2014; Jia et al., 2018). In Table 3, the values of hazard quotients (HQ) via the three pathways in soils among the three districts decreased as $\text{HQ}_{\text{ingestion}} > \text{HQ}_{\text{dermal}} > \text{HQ}_{\text{inhalation}}$. It indicates that the ingestion route of exposure to these heavy metals was the main human exposure pathways. Moreover, for children HQ (via all routes of exposures to studied metals) shows higher than adults which is similar to other studies around the world (Chonokhuu et al., 2019; Gbadamosi et al., 2018; Rastegari Mehr et al., 2017). The contribution of different metals to the non-carcinogenic risk (HQ) show the decreasing order of $\text{Fe} > \text{Mn} > \text{Cr} > \text{Pb} > \text{Cu} > \text{Ni} > \text{Zn} > \text{Cd}$ for both adults and children. However, there was no health risk to the CHT environment of individual metal present in the soil (Kacholi and Sahu, 2018).

Yet, the HI values were lower than the safe level ($\text{HI} \leq 1$) except Fe element which implies that there was no unusual non-carcinogenic risk due to these heavy metals for children and adults in the soil samples. $\text{HI} > 1$ for Fe in the three pathway for both adults and children indicates a potential risk to human health. However, iron does not show direct chemical toxicity according to an ecological risk perspective, but the effect of iron was referred as the geochemistry of other potentially toxic metals and the potential physical hazard of depositing flocculent. Since the population will be exposed to all heavy metals instead of a single one, the use of integrated HI values (ΣHI) is more practical to identify the real effect (Wahab et al., 2020). ΣHI exceeded 1 due to the high concentration level of Fe. So, based on ΣHI value in the studied area, children experience a higher risk of

noncancer effects than adults. Since children directly involve themselves in hand-to-mouth activities (USEPA, 2011) (US EPA, 2011). Thus, there is a great possibility of incurring noncancerous effects and chronic diseases such as loss of appetite, nausea, and headache to the populations of CHT (Fonseca-Nunes et al., 2014; Wahab et al., 2020).

Table 3. The non-carcinogenic risk of HQs and HIs of the metal elements in the soil in children to adult.

Metal		Pb	Cd	Cu	Zn	Fe	Mn	Ni	Cr
R _f D (mg kg ⁻¹ day ⁻¹)	Ingestion	3.50E-03	1.00E-03	4.00E-02	3.00E-01	7.00E-01	4.60E-02	2.00E-02	3.00E-03
	Inhalation	3.52E-03	1.00E-03		3.00E-01	7.00E-05		2.06E-02	2.86E-05
	Dermal	5.25E-04	1.00E-05	1.20E-02	6.00E-01	7.00E-05	4.00E-02	5.40E-03	6.00E-05
HQ _{ing} (Child)		3.87E-02	1.83E-03	8.44E-03	2.44E-03	9.73E-01	1.05E-01	7.69E-03	1.03E-01
HQ _{ing} (Adult)		4.15E-03	1.96E-04	9.04E-04	2.61E-04	1.04E-01	1.12E-02	8.24E-04	1.10E-02
HQ _{inh} (Child)		1.08E-06	5.11E-08		6.82E-08	2.72E-01		2.09E-07	3.01E-04
HQ _{inh} (Adult)		6.07E-07	2.88E-08		3.84E-08	1.53E-01		1.18E-07	1.70E-04
HQ _{derm} (Child)		7.23E-04	5.12E-04	7.87E-05	3.42E-06	2.73E+01	3.38E-04	7.97E-05	1.44E-02
HQ _{derm} (Adult)		1.10E-04	7.81E-05	1.20E-05	5.21E-07	4.16E+00	5.16E-05	1.22E-05	2.20E-03
HI (Child)		3.95E-02	2.34E-03	8.52E-03	2.44E-03	2.85E+01	1.05E-01	7.77E-03	1.17E-01
ΣHI (Child)		2.88E+01							
HI (Adult)		4.26E-03	2.74E-04	9.16E-04	2.62E-04	4.42E+00	1.13E-02	8.36E-04	1.34E-02
ΣHI (Adult)		4.45E+00							

3.4.2 Carcinogenic risk (CR)

The CR for Pb was assessed through ingestion and dermal mode, and the inhalation mode was used for Pb, Cd, Ni and Cr (Table 4). Note that the risk of dermal exposure to the soil is 100 times lower than the ingestion and inhalation exposure (Gržetić and Ahmed Ghariani, 2008). Slope factor and soil exposures were used to calculate the health risk assessment (Alfaro et al., 2015; Gbadamosi et al., 2018; Johnbull et al., 2019). The result from Table 4 showed that the CR_{dermal} value of Pb for adults and children in the three districts was significantly lower, means less effective of CR (Baltas et al., 2020). CR values of ingestion, inhalation and dermal exposure, and integrated CR value (ΣCR) of all carcinogenic metals for children and adults were lower than the acceptance tolerance risk, 10⁻⁶ - 10⁻⁴. However, CR values of our results for ingestion purpose were comparatively more than inhalation and dermal exposure. Since Fe, Mn, Cu and Zn are not human carcinogens, these metals were not considered for assessment of CR, (Gbadamosi et al., 2018; Gržetić and Ahmed Ghariani, 2008; Johnbull et al., 2019). The obtained results of the potential carcinogenic risks for adults and children via the exposure to heavy metals in CHT soils show a very insignificant value.

Table 4. Carcinogenic risks of the metal elements in the soil to children to adult.

Carcinogenic Risk Assessment							
Metal	Slope factor	Child			Adult		
		Rm	Kc	Bb	Rm	Kc	Bb
CR for Ingestion							
Pb	8.50 E-03	1.01E-07	6.72E-08	1.28E-07	4.35E-08	2.88E-08	5.47E-08
CR for Inhalation							
Pb	4.20E-02	1.40E-11	9.28E-12	1.76E-11	3.16E-11	2.09E-11	3.97E-11
Cd	6.30E+00	2.81E-11	2.49E-11	2.97E-11	6.33E-11	5.63E-11	6.71E-11
Ni	8.40E-01	3.06E-10	1.89E-10	4.33E-10	6.68E-10	4.26E-10	9.76E-10
Cr	4.10E-01	2.25E-10	3.22E-10	3.60E-10	5.08E-10	7.27E-10	8.12E-10
CR for dermal							
Pb	8.50 E-06*	2.48E-15	1.64E-15	3.12E-15	7.96E-16	5.28E-16	1.00E-15
$\sum CR$		1.02E-07	6.77E-08	1.28E-07	4.47E-08	3E-08	5.66E-08

4. Conclusion

Concentration of heavy metals were determined in sequence of Fe > Mn > Zn > Cu > Cr > Ni > Pb > Cd in soils of 37 sites collected from Chittagong Hill Tracts area, Bangladesh. Essential elements (Fe, Mn and Zn) show relatively greater value than the toxic (Pb, Cd, Cu) metals. Pb, Cd, Cu and Zn indicated significant enrichment with EF > 1 in the soil of Rm-3, Kc-14 sites. Fe showed significant contamination in soil (CF > 1) in most of the analyzed sites. PLI < 1 was found almost in all sites which expressed the acceptability of soil quality except Bb-5 site. The potential ecological risk factor of Cd was shown a considerable ecological risk. PERI values indicated a moderate ecological risk, whereas, among three districts, a considerable ecological risk exists in Bandarban.

This study reveals that there is no adverse carcinogenic health risk via the use of CHT soils. However, there is a possible non-carcinogenic risk for both adults and children by the three exposure pathway from iron. Hence, regular monitoring is needed for the documentation of any alternation in the quality of soil and minimise the outbreak of health disorders and damage to the forest ecosystem. It is presumed that some of the main causes of soil pollution in CHT are shifting cultivation, clearing and burning of jungles. Other heavy metals (As, Co, Hg, etc.) as well as other parameters such as organic content, total organic carbon, oxygen demand and moisture content etc. may be considered for further analysis.

Declarations

Ethics approval and consent to participate

Not applicable

Consent for publication

Not applicable

Availability of data and materials

The datasets used and analysed during the current study are available from the corresponding author on reasonable request.

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522 **Authors' contributions**

523 **Troyee Barua:** Conceptualization, Methodology, Data curation, Formal analysis and Writing - original draft

524 **AKM Saiful Islam Bhuian :** Supervision

525 **Mayeen Uddin Khandaker:** Writing - review and editing of the manuscript

526 **Md. Abdur Rashid:** Validation, Supervision

527 **Nipa Deb:** Software

528 **Shahadat Hossain:** Formal analysis

529

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534 **References**

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Figures

Figure 1
Potential ecological risk indices (PERI) in the soil of CHT.

Figure 2

PCA loading plot of studied heavy metals in rotated space.

Figure 3

Dendrogram showing hierarchical cluster analysis of analyzed metals of the studied area.