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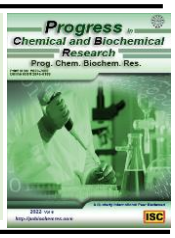

Original Research Article

Contamination and Ecological Risk Assessment of Heavy Metals in Different Land Use Urban Soils of Kathmandu District, Nepal

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ABSTRACT

Ever-increasing population, rapid urbanization, and industrialization have critically deteriorated the urban soil quality. This study was conducted to assess the ecological risk of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), and lead (Pb) in four different land-use urban soils *viz*, commercial area (CA), heavy traffic ring roadside (HT), residential area (RA), and agricultural farm (AF) of Kathmandu District, Nepal. For this purpose, concentrations of the five heavy metals (HMs) were determined by using a flame atomic absorption spectrophotometer (FAAS), in a total of 31 soil samples collected from all four land uses. Pollution indices such as contamination factor (CF), degree of contamination (CD), pollution load index (PLI), geo-accumulation index (I-geo), ecological risk factor (ER), and potential ecological risk (PER) were used to assess the ecological risk posed by the HMs. The overall mean concentrations for Cd, Cr, Cu, Ni, and Pb were 0.98, 137.05, 79.80, 100.00, and 72.27 mg/kg, respectively, and were found 2-4 times greater than the background values. The statistical analysis revealed a poor correlation of the HMs against pH and total organic carbon (TOC) suggesting little influence on HMs contamination. Results also showed the highest concentrations of the HMs in HT soils among the land use types. Ecological risk assessment revealed CF (0.42-5.06) and CD (7.83-15.72) values as indicators for low to considerable, and moderate to considerable risks respectively, in soils of all land uses under the present investigation. Whereas the PLI values (1.12-2.96) corresponded to the polluted urban soils, the Igeo values (0.08-1.02) indicated an unpolluted to the moderately polluted class of soil contamination in all the land-uses. Likewise, the ER (0.91- 114.90) and PER (113.97-170.14) values pointed out that all the land use urban soils were exposed to a class of low to considerable, and moderate to considerable risks, respectively. Among the HMs, Cd, and Pb posed a comparatively high ecological risk for soils of all land uses and the estimated indices indicated HT as the most vulnerable land use suggesting immediate control measures.

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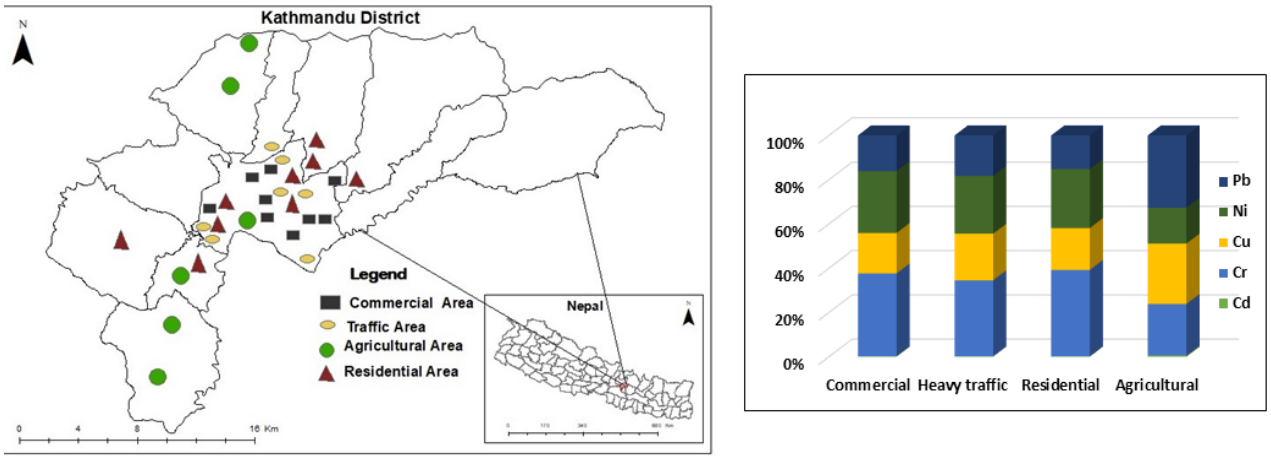
GRAPHICAL ABSTRACT



Introduction

Soil, a reservoir of water and nutrients, is a compact texture on the uppermost layer of the crust of the earth. Its origin is through weathering processes influenced by biological, climatic, geologic, and topographical factors [1]. The uppermost fertile, porous, friable, and loose layer (0-15 cm depth) of the earth, particularly rich in organic matter and plant nutrients is called surface soil. This is also enriched with both the number and activity of soil micro-organisms suitable for plant growth and has a relatively high cation exchange capacity [2]. However, several natural and anthropogenic activities have resulted in the soils being contaminated with potentially hazardous heavy metals [3].

Soil is one of the principal substrata on the earth and essential for human survival. It is the major sink for persistent heavy metal contaminants and metalloids. The hazardous elements are being continuously released into the terrestrial environment through various anthropogenic sources including industrial activity, agricultural production, traffic emissions, coal combustion, municipal waste disposal, and atmospheric deposition [3,4]. Besides, the rapid growth of population, industrialization, and urbanization are major causes of the release of significant quantities of heavy metals into the environment [5]. The United State Environment Protection Agency (USEPA) has therefore listed Ni, Cr, Cu, As, Cd, and Pb as the primary pollutants citing their toxic effects [6].

Soil contaminated with heavy metals may pose potential human health as well as ecological risks. They enter the human body through the food chain (soil-plant-human or soil-plant-animal-human) [7,8]. The contaminated soils bring about negative influences on several physiological activities of plants including photosynthetic activity, nutrient absorption, and gaseous exchange. Besides, they also retard plant growth and yield [9] and also affect food quality and safety. Moreover, some essential nutrients in the human body can be depleted causing intrauterine growth retardation along with weak psychosocial behavior, a high prevalence of upper gastrointestinal cancer, a weak immunological defense, and disabilities associated with malnutrition [10]. They can also reduce intelligence in children, affecting internal organs and damaging the central nervous system, and immune systems [11]. Besides, the toxicity and mobility of HMs in the soil also depend on metal speciation or binding condition [12].

There is growing interest in the study of ecological risk assessment of potentially hazardous heavy metals in soils citing their toxic effects. Islamd *et*

al. [13] assessed nine heavy metals (Cr, Ni, Cu, As, Cd, Pb, Fe, Mn, and Zn) in five different land use urban soils of Bangladesh with a view to studying the potential ecological risks posed by these hazardous elements. Proshad *et al.* [14] assessed the ecological risk of six heavy metals (Cr, Ni, Cu, As, Cd, and Pb) in urban soils collected from 15 sampling sites around Tangail district in Bangladesh. Huwang *et al.* [15] evaluated the severity of soil contamination by four heavy metals (As, Cr, Cu, and Pb) at 146 urban and suburban soils within the city of Xiamen, Fujian, China. Mehr *et al.* [16] assessed the ecological risk of As, Cd, Co, Cr, Cu, Ni Pb, and Zn in 83 urban surface soils of 23 cities in Isfahan province, the central part of Iran.

Contaminated soils have become a serious worldwide environmental issue because people spend most of their time in open spaces and workplaces from which they can get direct contact with heavy metals. Farmers are the most vulnerable group since they get exposed to contaminated soils almost every day [13]. Besides, children spend most of their time playing in-ground and open spaces exposed to contaminated soils. Hence, they can easily get attacked by contaminated soils and face many types of health disorders [10]. Particularly, young children are more likely to ingest a significant quantity of soil than adults because of the behavior of mouthing non-food objects and repetitive hand/finger sucking. Besides, children have a much higher absorption rate of heavy metals from the digestion system and higher hemoglobin sensitivity to heavy metals than adults [11]. It is, therefore the utmost importance to monitor the concentration of heavy metals in soils and assess ecological risk. These factors are directly related to environmental and human health issues. Kathmandu has become a densely populated city resulting in socio-economic and land use changes [17, 18]. The city has many vulnerable areas with metal contaminants resulting from construction and demolition, traffic emissions, waste disposal, industrial activities, etc. [19]. Although there is a large number of published works in the pertinent area, studies on ecological risk assessment of heavy metals in urban soils of Nepal are poorly documented. Therefore, the main objective of the present study was to evaluate physicochemical properties (pH and TOC) and concentrations of five heavy metals (Cd, Cr, Cu, Ni, and Pb) in different land use urban soils of Kathmandu District, Nepal, and to assess the potential ecological risk posed by these heavy metals. The findings of the present study may provide baseline information to the scientific community in the related area in the context of Nepal.

Experimental

Study area and sampling

Kathmandu, the bowl-shaped capital city of Nepal, is located between 27^0 45'-27^0 47' N latitude and 85^0 20'-85^0 25' E longitude. The district (Figure 1) has a population density of only 97 per square kilometer along with a density of 13, 225 per square kilometer in Kathmandu metropolitan city alone. The city is also characterized by the largest urban area which accounts for 20%. The rapid population growth, haphazard urbanization, and industrialization along with the recent development of various infrastructures are some ongoing features of the city. The present study was carried out in four land use urban soils *viz.*, commercial area, heavy traffic ring roadside, residential area, and agriculture farm of Kathmandu. Figure 1 displays the GIS-based sampling map of the study sites.

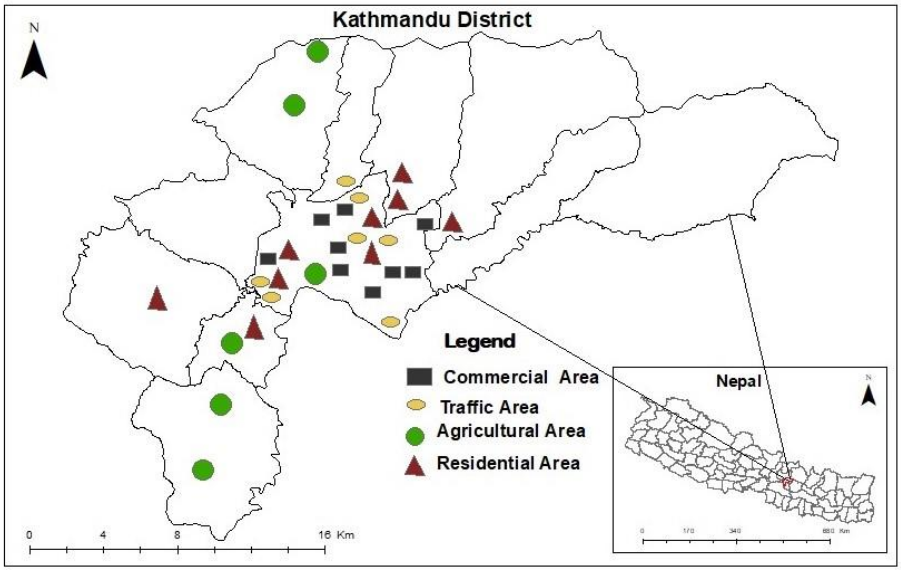


Figure 1. Map of Kathmandu District, Nepal showing study area and sampling sites

Before sample collection, an initial field survey was conducted across the study area with a view to enlisting potential sampling sites falling under these four different land use categories. Then, the potential sampling sites of each land-use type were numbered separately and consecutively. From each land use category, 30 percent of samples were selected by using a simple lottery method. Table 1 presents the description of various activities for each land use type.

Table 1. Description of sampling sites across Kathmandu District

Land use type	No. of samples (n)	Location of sampling sites	Activities at the sampling sites
Commercial area (CA)	9	Kalimati, Sinamangal, Ratnapark, Lainchaur, Bauddha, Bansbari, New Baneshwor, Battishputali, and Thapathali	High anthropogenic activities, densely populated areas, traffic load, supermarkets, hospitals, schools, colleges, departmental stores, commercial buildings, mechanical workshops, hotels, etc.
Heavy traffic ring roadside (HT)	7	Koteshwor, TIA, Chabahil, Maharajgunj, Gongabu, Kalanki, and Balkhu	Huge traffic density, business complexes, hospitals, commercial plazas, bus parks, cinema halls, mechanical and automobile workshops, etc.
Residential area (RA)	9	Tahachal, Sitapaila, Hadigaun, Baluwatar, Kirtipur panga, Gokarna, Naikap, Kapan, and Budhanilkanth	Limited commercial activities, medium traffic density, limited shopping malls, residential colonies, apartments, etc.
Agriculture farm (AF)	6	Charghare (Kirtipur), Okharbote, Bhajangal, Tarkeshowr, Kakani, and Pharping	Land cultivated with fruits and vegetables, paddy fields, negligible commercial activities, sparse residential area, the minimum traffic load, etc.

Sample collection

Thirty-one composite soil samples from four different land-use types as described were collected during the pre-monsoon season, 2021 AD. About 50 g of soil samples (to a depth of 0-15cm) from each sampling site were collected with the help of a metallic core cutter. Each soil sample was a composite mixture of 3 sub-samples from a 5 m radius in different directions [20]. The soil samples were wrapped in polythene bags with zip locks, properly labeled, and immediately taken to the laboratory. The samples were air dried at room temperature for two weeks and sieved with a mesh size of 2 mm to remove debris, stones, and pebbles. A commercial grinder was used to homogenize them into a fine powder and stored at -4 ^{o}C until chemical analysis.

Analytical reagents and chemicals

All certified standards (1000 ppm) of Cd, Cr, Cu, Ni, and Pb were purchased directly from Merck, Germany. For calibration, solutions of the standards in required concentrations were prepared by using doubly distilled water. The acids (HCl, H_2SO_4, and HNO_3) and other required chemicals such as $K_2Cr_2O_7$, $FeSO_4(NH_4)2SO_4.6H_2O$, and diphenylamine indicator (E. Merck, Germany) of analytical grade were used for the analytical purpose. The purities of all the reagents and chemicals used were over 99 %.

Analysis of physicochemical parameters

The soil samples were analyzed for physicochemical characterization as follows:

Determination of pH: pH of soil samples was determined in 1:5 soil water suspensions [21]. In each experimental procedure, 20 g of soil sample in 100 ml of distilled water was magnetically stirred over an hour. The pH of the unfiltered soil water suspension was recorded by a calibrated digital pH meter (Model 101, ESICO International).

Determination of total organic carbon (TOC): TOC in soil samples was estimated by the dichromate method [22]. In each experimental procedure, 1.0 g of soil sample was added to a solution containing a mixture of 10 ml of 1N $K_2Cr_2O_7$ solution and 20 ml of conc. H_2SO_4 in a clean and dry 500 ml conical flask and mixed by gentle swirling. The flask was placed in a hot air oven at 150 °C for half an hour till the complete reaction. Then, the content was diluted with 200 ml of distilled water and added 0.2 g of sodium fluoride. One ml of diphenylamine indicator was added to the reaction mixture and titrated with 0.4 N standard solution of ferrous ammonium sulfate till the endpoint. A blank with the same quantity of chemicals without any soil samples were further run in parallel.

Determination of heavy metals in soil samples

Preparation of sample solution

A sample solution for the determination of heavy metals in soils was prepared following the method described by Chrisroforidis and Stamatis [23]. In each experimental procedure, 1.0 g of air-dried soil sample in an acid-washed beaker was treated with 10 ml conc. HNO_3. The sample was digested over the hot plate at 90 °C and the digestion process was continued till the volume of acid was reduced to 1-2 ml. More acid was added as per necessity to carry out complete leaching of metals from the sample. After complete digestion, the solution was filtered through Whatman No. 42 filter paper collecting the filtrate into a 25 ml volumetric flask. The beaker was rinsed with distilled water and collected the filtrate into the same volumetric flask. The final volume was made with the same water through homogenous mixing.

Quality control and instrumental analysis

Before using glassware, they were treated with dilute (1:1) nitric acid by dipping it in a container for 24 hours and then rinsed with distilled water. Reagent blank was prepared by evaporating 10 ml nitric acid slowly in a clean acid-washed beaker. The slow evaporation was continued until the volume of acid was reduced to 1-2 ml. After cooling and rinsing the interior of the beaker with a small volume of distilled water, the whole of the

reagent blank was transferred into the 25 ml volumetric flask. The final volume was made up to the mark with distilled water. The potential errors in measurement were eliminated by the reagent blank.

The digested samples were analyzed for the HMs by flame atomic absorption spectrophotometer (SOLAAR M5 Dual Automizer, 180-900nm, Thermo Elemental, UK) by using air-acetylene flame [24]. Prior to sample analysis, the instrument was sufficiently warmed up and checked for the hollow cathode lamps of each specific metal along with a deuterium background correction at its specific wavelength. Blanks were also checked following the completion of a full analytical procedure without samples. At each analytical run, the screen displayed the calibration curve. The linearity and replication were visually checked afterward. Then, the FAAS instrument was introduced with three-point calibration standards of appropriate concentration prepared for each element from the stock standard solution of 1000 ppm. To obtain the linearity of the calibration curve, the stock solution was successively diluted at an order of 10^{-1}. A flow rate of 2 ml/min for each sample solution was maintained through the column as per the instruction manual. The operating wavelengths for Cd, Cr, Cu, Ni, and Pb were 228.802, 357.869, 324.754, 232.003, and 283.306 nm, respectively. To maintain a precision of the analytical experiment, all samples were analyzed in triplicates.

Recovery test

For spike analysis of each analyte, three soil samples were randomly selected from different land-uses. The analyte with a known quantity was added to 1.000 g of selected samples. Standard reference materials (SRM) traceable to NIST manufactured by Merck, Germany were used to prepare the fortified samples. The metal concentration in the fortified samples was determined by FAAS. From the reference materials, percent recovery of metal was 97.3 % (Cd), 98.2 (Cr), 97.5 % (Cu), 98.8 % (Ni), and 97.5 % (Pb). The calculated standard deviation for the pretested samples was 2.1, 2.3, 2.2, 2.8, and 2.6 % for Cd, Cr, Cu, Ni, and Pb, respectively, and can be considered satisfactory for analysis of test samples. The detection limits were 9, 5, 3, 10, and 9 µg/L for Cd, Cr, Cu, Ni, and Pb, respectively.

Assessment of ecological risk

Ecological risks posed by the heavy metals (HMs) in the urban soils of four different land uses were assessed by using single (contamination factor, CF; geo-accumulation index, I-geo, and ecological risk factor, ER) as well as integrated (the contamination degree, CD; pollution load index, PLI and potential ecological risk, PER) pollution indices.

Contamination factor (CF): The CF, initially suggested by Håkanson [25], is a measure of the degree of metal contamination relative to the measured background values from the geologically similar and uncontaminated areas and was calculated by using Equation (1):

$$CF = Cs/Cb \qquad (1)$$

Where, Cs is the measured concentration of the examined heavy metal in soil and Cb is the geochemical background concentration of the respective heavy metal. Because of the unavailability of background concentration of HMs in case of Nepal, we adopted the background value of HMs from Turekian and Wedepohl [26]. The background concentrations for Cd, Cr, Cu, Ni, and Pb are 0.3, 90, 45, 68, and 20 mg/kg, respectively.

The levels of metal contamination in soils may be classified into four categories as: low (CF < 1), moderate (1 ≤ CF < 3), considerable (3 ≤ CF < 6), and very high (6 ≤ CF).

Contamination degree (CD)*:* To assess ecological risk from more than one heavy metal, a degree of contamination (CD) is used. The CF sum for all HMs represents the integrated pollution index of the environment. This index was suggested by Håkanson [25] and calculated using Equation (2):

$$CD = \sum CF \quad (2)$$

Four categories of CD may be used to evaluate the level of metal contamination in the soil environment as: low (CD < 5), moderate (5 ≤ CD < 10), considerable (10 ≤ CD < 20), and very high (20 ≤ CD) [27].

Pollution load index (PLI): The PLI is defined as the geometric mean of the contamination factor (CF) for the nth metals [28]. To assess the soil quality in terms of the mutual effect of hazardous metals contamination, an integrated approach of the index was calculated using Equation (3).

$$PLI = \sqrt[n]{CF1\ x\ CF2\ x\ CF3\ x \dots \dots \dots \dots CFn} \quad (3)$$

Where, PLI is the pollution load index, CFs are contamination factors, and n is the number of elements. A PLI value >1 indicates polluted soil, while <1 indicates no pollution [29].

Geo-accumulation index (I-geo): The degree of HMs pollution in the soil is quantified by using geo-accumulation index (I-geo). The index was originally defined by Müller [30] and calculated using Equation (4):

$$Igeo = log2\left[\frac{Cs}{1.5\ Cb}\right] \quad (4)$$

Where, *Cs* is the concentration of heavy metal in soil sample, and *Cb* is the geochemical background concentration of the respective heavy metal. Factor 1.5 is used because of possible variations in background values for a given metal in the environment and to detect a very small anthropogenic influence.

Based on Igeo values, Müller [30] classified metal contamination levels or degree of soil pollution into the following seven categories as: Igeo ≤ 0 - Practically unpolluted (0 degree); 0 < Igeo ≤ 1 - Unpolluted to moderately polluted (the 1st degree); 1 < Igeo ≤ 2 -Moderately polluted (the 2nd degree); 2 < Igeo ≤ 3 -Moderately to strongly polluted (the 3rd degree); 3 < Igeo ≤ 4 -Strongly polluted (the 4th degree); 4 < Igeo ≤ 5 -Strongly to extremely polluted (the 5th degree), and Igeo ≥ 5 - Extremely polluted (the 6th degree).

Potential ecological risk (PER): The ecological risk factor (ER) indicates the effect of a single metal pollutant in soils, whereas the combined effect of multiple contaminants is shown by potential ecological risk (PER). The ER and PER indices were calculated by using Equations (5) and (6), respectively [31].

$$ER = BTF\ \times CF \quad (5)$$

$$PER = \sum ER = ER1 + ER2 + ER3 + \cdots \dots \dots \dots + ERn \quad (6)$$

Where, *ER, BTF*, and *CF* represent an ecological risk factor, biological toxic factor, and contamination factor of a single metal, respectively. The biological toxic factors (BFT) for Cd, Cr, Cu, Ni, and Pb are 30, 2, 5, 6, and 5, respectively [31,32].

The ecological risk factor (ER) may be classified into five grades as: ER < 40 - low risk; 40 ≤ ER < 80 –moderate risk; 80 ≤ ER < 160 –considerable risk; 160 ≤ ER < 320 –high risk and ER ≥ 320 –very high risk. Similarly, the potential ecological risk (PER) may be classified into four grade categories as: PER < 65 –low risk; 65 ≤ PER < 130 –moderate risk; 130 ≤ PER < 260 –considerable risk and PER ≥ 260 –very high risk [33].

Statistics

The descriptive statistics such as frequency, percentage, mean, and standard deviation were presented through all the recorded data preceded in an IBM computer. Correlation along with significance test among the soil parameters and heavy metals were analyzed and presented wherever applicable.

Results and Discussion

Physicochemical properties of soil

The physicochemical properties (pH and TOC) of soils measured in this study are summarized in Table 2.

Table 2. Physicochemical properties and heavy metals concentration (mg/kg) in different land-use urban soils of Kathmandu

Land uses (No. of samples)	Statistical parameter	pH	TOC (%)	Heavy metals					$\sum_5$HM
				Cd	Cr	Cu	Ni	Pb	
Commercial area (CA) (n = 9)	Mean	7.9	1.49	0.98	167.01	81.72	124.50	72.13	446.34
	Min.	7.7	1.05	0.74	91.60	54.20	78.93	50.04	275.50
	Max.	8.2	2.12	1.22	245.76	110.01	169.64	86.65	613.28
	SD	0.2	0.44	0.16	45.82	15.93	24.01	10.22	96.14
HT ring roadside (HT) (n =7)	Mean	8.0	1.12	1.15	190.05	116.87	144.30	101.21	553.58
	Min.	7.6	0.88	0.95	148.30	97.02	102.61	67.03	415.91
	Max.	8.4	1.35	1.41	242.59	146.53	196.27	120.86	707.66
	SD	0.7	0.17	0.16	32.09	14.61	31.75	16.29	94.90
Residential area (RA) (n = 9)	Mean	7.8	1.03	0.86	150.33	72.82	102.60	58.43	385.04
	Min.	7.4	0.83	0.56	73.14	46.52	72.31	40.47	233.00
	Max.	8.3	1.20	1.25	228.94	108.19	130.50	76.58	545.46
	SD	0.3	0.12	0.21	60.04	20.67	20.58	10.76	112.26
Agricultural farm (AF) (n = 6)	Mean	7.1	1.07	0.91	40.80	47.77	28.36	57.32	175.16
	Min.	5.9	0.81	0.77	30.44	42.54	20.65	44.79	139.19
	Max.	7.5	1.20	1.08	52.60	54.88	37.03	69.46	215.05
	SD	0.6	0.13	0.11	7.18	4.09	5.53	8.47	25.38
Mean of all land-uses		7.8	1.18	0.98	137.05	79.80	100.00	72.27	390.03
*Background conc.		-	-	0.3	90	45	68	20	-

* Turekian and Wedepohl (1961)

The results revealed variable mean and ranges of pH and TOC among the four land-use types. Soil pH is one of the important physicochemical properties impacting all other soil parameters. The overall mean pH (7.8) in the urban soils of the land-uses exhibited alkaline in nature (Table 2). Among the land-uses, the HT site measured the highest mean pH value (8.0) followed by CA (7.9), RA (7.8), and AF (7.1) sites. Apparently, the pH in soils of almost all study areas was found alkaline which may be attributed to carbonate and/or bicarbonate salts [34], and also the decomposition of organic matter [35].

The overall mean TOC in urban soils of Kathmandu was 1.18% indicating that soil serves as an important sink for organic matters in Kathmandu (Table 2). Among the selected land-uses, TOC values were observed in the descending order of CA (1.49%) > HT (1.12%) > AF (1.07%) > RA (1.03%) land use. The high TOC content in soils of CA and HT land uses may be attributed to organic materials from anthropogenic waste and leakage of lubricating oil or gasoline that contain hydrocarbons [36]. The results of the present study are in agreement with the findings of Herath *et al.* [37] who also reported a high organic matter content in urban soils of the Colombo metropolitan region which they attributed to several factors including leakage of lubricants and vehicle oil and waste disposal. Yadav *et al.* [20] also reported similar findings in urban soils of major cities of Nepal. The mobility and concentration of elements in the soil are often controlled by the pH and organic carbon content of the soil [35]. Organic matter helps to buffer soil pH by adsorbing ions [38]. Besides, it is more likely to bind metals forming metal chelates due to its high cation exchange capacity resulting in less availability of metals to plants [39,40].

Concentration of heavy metals (HMs) in urban soils

The statistical characteristics of Cd, Cr, Cu, Ni, and Pb measured in four different land use urban soils of Kathmandu district are reported in Table 2. The

overall mean concentration of five HMs was found to be decreased in the order of Cr (137.05 mg/kg) > Ni (100.0 mg/kg) > Cu (79.80 mg/kg) > Pb (72.27 mg/kg) > Cd (0.98 mg/kg) accounting the metals, respectively, for 35, 26, 21, 19, and 0.3% of $\sum_5$HM. This decreasing trend of metal is in agreement with the previous study from south China [41]. Besides, the mean concentrations of five HMs were 2-4 times greater than the background concentration in shale [26] suggesting either contamination or the influence of pedogenic factors [42].

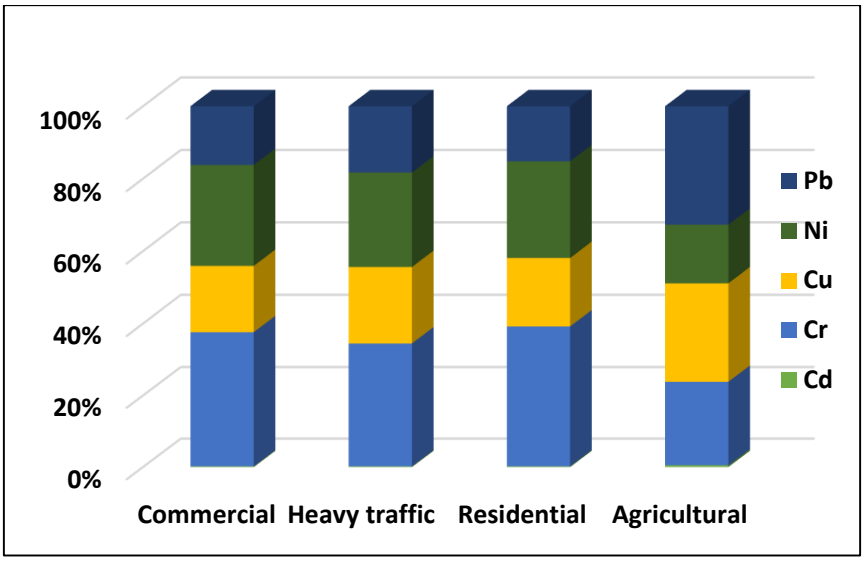


Figure 2. Profile of heavy metals showing the mean % contribution of individual metal to the total mean of $\sum_5$HM

The statistical data of HMs in soil (Table 2) showed the highest mean concentration of $\sum_5$HM at HT land-use followed by CA, RA, and AT. Accordingly, the $\sum_5$HM concentration ranged from 415.91 to 707.66 mg/kg dw (mean 553.58 mg/kg dw), 275.50 to 613.28 mg/kg dw (mean 446.34 mg/kg dw), 233.0 to 545.46 mg/kg dw (mean 385.04 mg/kg dw), and 139.19 to 215.05 mg/kg dw (mean 175.16 mg/kg dw) in HT, CA, RA, and AF land uses respectively. The selected land-uses measured their HMs concentration (mg/kg) in the soil in the following descending order as:

CA: Cr > Ni > Cu > Pb > Cd
HT: Cr > Ni > Cu > Pb > Cd
RA: Cr > Ni > Cu > Pb > Cd
AF: Pb > Cu > Cr > Ni > Cd

Likewise, the concentration of five HMs in surface soils of all land-uses followed the descending order of HT > CA > RA > AF. Among the land-uses, the HT site measured the highest concentration of Cd, Cr, Cu, Ni, and Pb in soil. The metal profile depicted in Figure 2 indicated that Cr was the most abundant metal in soils of all land-uses followed by Ni, Cu, and Pb except AF site. However, Cr, Ni, Cu, and Pb in soils of all land-uses accounted for 23-39%, 16-28%, 18-27%, and 15-32% of $\sum_5$HM, respectively. Apparently, Cd was less detected and accounted for less than 1% of $\sum_5$HM.

As indicated in Table 2, Cr was the most abundant element in surface soils of HT (190.1 mg/kg), CA (167.0 mg/kg), and RA (150.3 mg/kg) land-uses. Accordingly, the HT site recorded the highest Cr content in soils among the four land-uses and the five HMs under the present investigation except for AF land-use. The mean concentration of Cr among all land-uses was found to be 137.05 mg/kg and was recorded as the highest level among all HMs. Chromium has both geogenic and anthropogenic sources in the environment [30].

The potential sources of Cr contamination in soil include industrial activities such as chrome plating of vehicle parts and alloys making especially stainless steel. Therefore, grinding, welding, and also polishing of stainless steel can lead to the significant discharge of Cr into the soil environment [43,44]. Burning of oil, coal, petroleum from a ferro-chromate refractory material, catalyst, chromium steel, fertilizers, oil well drilling, and metal plating tanneries also deposit a significant amount of Cr in soil. Besides, Cr is further released into the environment through sewage and fertilizers [45]. In addition, Cr and Ni are important constituents of traffic emission [46]. In this study, the considerable levels of Ni were 124.50, 144.30, 102.60, and 47.77 mg/kg for CA, HT, RA, and AF land-uses, respectively. As for Cr, the HT site also recorded the highest Ni level in soils among all land-uses. Corrosion of cars, tire rims, cylinders, and pistons of a motor engine are the potential sources of Ni contamination in soil [47,48].

It was found that the intensity of Cu contamination in soils of the selected land-uses followed the decreasing order of HT (116.87 mg/kg) > CA (81.72 mg/kg) > RA (72.82 mg/kg) > AF (47.77 mg/kg). Accordingly, Cu content was found significantly high in HT land-use like those of Cr and Ni. Among the five HMs, the AF land use, however, measured a higher Cu level after Pb. Intensive fertilizing with pig slurry is one of the potential sources of elevated Cu concentrations in agricultural soil. Additional sources of Cu in soils also include sewage sludge and atmospheric deposition [49]. In the soil, Cu strongly complexes with the organic matter implying that only a small fraction of copper in solution will be available in a dissolved state as ionic copper [50]. Cu is also associated with inorganic fertilizer in the industrial area. Along with Zn and Cd, the Cu contamination in soil has also been reported from car components, tire abrasion, lubricants, leakage of oil products, and industrial and incinerator emissions [51].

Like those of Cr, Ni, and Cu, HT land-use also demonstrated a comparatively high level of Pb contamination in soils among all land-uses. The lead concentrations recorded in urban soils were 72.13, 101.21, 58.43, and 57.32 mg/kg for CA, HT, RA, and AF land-uses, respectively. A major source of Pb in the soil is from abrasion of the automobile tires, engine wares, and spills from batteries including the gasoline containing Pb [52]. Besides, the low solubility of Pb allows it to have a long residence time in the roadside soil environment [53].

Cadmium was found to be the least contaminated in soils of all land-uses under the present investigation. Table 2 showed the Cd content in the land-uses in the decreasing order of HT (1.15 mg/kg) > CA (0.98 mg/kg) > AF (0.91 mg/kg) > RA (0.86 mg/kg). The higher level of Cd at HT land-use may be attributed to paints, batteries, and catalysts from the urban activities. The used metal fittings, rubber, plastics, tires, paints, phosphate fertilizers, etc. are materials emitting Cd to the environment [54,55]. The attrition of automobile tires, car abrasion, lubrication oils, and galvanized parts of vehicles are the possible sources of Cd contamination [56]. Besides, the use of wastewater or contaminated river water for irrigation, the application of heavy loads of phosphate fertilizers, and agrochemicals for a long period may also lead to the Cd contamination in soil [55,57].

Correlation of HMs with pH and total organic carbon (TOC)

A correlation study, a valuable tool for studying the likeliness of paired data, is broadly used in HMs data analyses [58]. Table 3 shows the correlation matrix among HMs and physicochemical parameters (pH and TOC). It was found that both pH and TOC content in soil was weakly correlated with HMs except for Cr which was only moderately correlated with pH ($r = 0.594$) at $p = 0.05$. This weak correlation of HMs indicated that soil pH and TOC content have little influence on HMs contamination.

Table 3. Pearson's correlation coefficient matrix for physio-chemical properties of soils and heavy metals

	pH	TOC	Cd	Cr	Cu	Ni	Pb
pH	1.000						
TOC	0.193	1.000					
Cd	0.248	0.008	1.000				
Cr	***0.594**	0.292	0.093	1.000			
Cu	0.416	0.258	0.284	***0.761**	1.000		
Ni	0.491	0.373	0.166	***0.819**	***0.713**	1.000	
Pb	0.185	0.128	0.361	0.428	***0.751**	***0.602**	1.000

*Significant at P = 0.05

The significant information regarding sources and pathways of metals in the environment is provided by the inter-relationship of metals [59]. The results revealed that Cr was strongly and positively correlated with Cu (r = 0.761, p = 0.05) and Ni (r = 0.819, p = 0.05) indicating similar sources of origin. Cu was significantly correlated with Ni (r = 0.713, p = 0.05) and Pb (r = 0.751, p = 0.05). Similarly, Pb was strongly linked with Ni (r = 0.602, p = 0.05). Cadmium showed a weak correlation with other HMs. The significant correlation among Cr, Cu, Ni, and Pb indicated their common sources of origin and especially from anthropogenic activities. The correlations in the present study were found consistent with the findings of Yadav *et al.* [20].

Comparison of HMs in soils among various cities/countries across the world

The mean concentration of $\sum_5$HM in soils in the present study was compared with similar studies reported across cities/countries worldwide and summarized in Table 4.

Table 4. Comparison of HMs concentration (mg/kg) in surface soil in this study with previous studies across cities/countries worldwide.

Cities/ Countries	Cd	Cr	Cu	Ni	Pb	References
Kathmandu/ Nepal	0.98	137.1	79.8	100.0	72.3	Present study
Chittagong/Bangladesh	2.43	-	32.63	-	7.33	[60]
Darkhan/Mongolia	-	31.9	-	19.5	20.9	[61]
Murcia/Spain	0.22	18.0	11.0	14.0	49.0	[62]
Shanghai/ China	0.52	107.9	59.3	31.2	70.7	[63]
Beijing/ China	0.15	35.6	23.7	27.8	28.6	[64]
Isfahan/ Iran	2.17	80.6	92.8	61.7	179.9	[16]
Tehran/ Iran	0.5	41.4	74.0	39.4	64.5	[65]
Patna/ India	-	-	49.6	-	24.5	[66]

The mean concentration of Cd (0.98 mg/kg) in the present study was found to exceed the concentrations reported for Shanghai, Beijing/China, Murcia/Spain, and Tehran/ Iran, while it was found 2-3 times less than those reported for Chittagong/Bangladesh and Isfahan/Iran. Similarly, the mean concentrations of Cr (137.1 mg/kg) and Ni (100.0 mg/kg) in the present study exceeded those reported across the cities/countries under the comparative study (Table 4). With the exception of Isfahan Province/Iran, the mean concentration of Cu (79.8 mg/kg) in the present study exceeded those reported for the selected cities/countries with approximately 2 to 12 times. Likewise, the mean concentration of Pb (72.3 mg/kg) measured in soils in this study crossed the reported concentrations for Chittagong/Bangladesh,

Darkhan/Mongolia, Murcia/Spain, Beijing/China, Tehran/ Iran, Patna/India, and Shanghai/China but nearly 3 times less than reported for Isfahan/Iran. Furthermore, the comparative study revealed that the levels of Cr and Ni in the present study were several folds higher than those reported worldwide (Table 4). Cu and Pb also demonstrated similar results but with few exceptions. The HMs enrichment in urban soils may be attributed to the rapid population growth, urbanization, and poor management of industrial effluents [41]. Other potential sources of HMs contamination in soils may include land-use type, traffic volume, fuel quality, distance from the road, human habits etc. [23,67].

Ecological risk assessment

The ecological risks posed by the HMs in the urban soils of four different land-uses were assessed by using single and integrated soil pollution indices.

Contamination factor (CF) and contamination degree (CD)

Table 5 shows the CF values for individual HM and CD which is the CFs sum.

Table 5. Contamination factor (CF) and degree of contamination (CD) of heavy metals (HMs) in different land-use urban soils of Kathmandu

Land-use type	Contamination factor (CF) for HMs					Degree of contamination, CD = ∑CF
	Cd	Cr	Cu	Ni	Pb	
Commercial area (CA)	3.27	1.86	1.82	1.83	3.61	12.39
HT ring roadside (HT)	3.83	2.11	2.60	2.12	5.06	15.72
Residential area (RA)	2.87	1.67	1.62	1.51	2.92	10.59
Agricultural farm (AF)	3.03	0.45	1.06	0.42	2.87	7.83

For Pb in urban soils, the HT site showed a comparatively high CF value (5.06) followed by CA (3.61), RA (2.92), and AF (2.87). Similarly, for Cd, the HT site also showed a high CF value (3.83) followed by CA (3.27), AF (3.03), and RA (2.87). Among the selected land-uses, the HT site showed comparatively high CF values for HMs in the descending order of Pb > Cd > Cu > Ni > Cr. The AF site was found to be least contaminated with Cr and Ni. As for contamination levels based on CF values, the soil samples of the selected land-uses showed low to a considerable level of HMs contamination (Table 6). Accordingly, the RA site demonstrated a moderate level of contamination for all the selected HMs, whereas CA and HT sites showed the same degree of contamination for Cr, Cu, and Ni. On the other hand, the AF site showed a moderate level of contamination for Cu and Pb but a considerable level for Cd. Likewise, the CA and HT sites showed a considerable degree of Cd and Pb contamination in soil samples.

The contamination degree (CD) is also considered as one of the important contamination indices indicating a measure of the sum of all metals anthropogenically accumulated in soils. The CD values computed for each land-use is shown in Table 5. The results revealed variable CD values for surface soils of the selected land-uses. Accordingly, the highest value of CD (15.72) was found for HT site followed by CA (12.39), RA (10.59), and AF (7.83). Concerning the CD values, all the selected land-uses showed considerable degree of HMs contamination ($10 \leq CD < 20$) except AF site which showed a moderate degree of HMs contamination ($5 \leq CD < 10$) (Table 6). Liu *et al.* [68] considered Cr, Ni, Cd, Pb, As, and Cu as the anthropogenic and geogenic elements in the surface soils and their distribution patterns were usually invariant. The long-term exposure to these metals can have the negative impacts on the health of children, pregnant women, and the vulnerable aged people [69].

Table 6. Classification and contamination level based on contamination factor (CF) and degree of contamination (CD) in different land-use urban soils of Kathmandu

Contamination level based on CF values				
Land-use type	(CF < 1)/ Low	(1 ≤ CF < 3)/ Moderate	(3 ≤ CF < 6)/ Considerable	(6 ≤ CF)/ Very high
Commercial area (CA)	-	Cr, Cu, and Ni	Cd and Pb	-
HT ring roadside (HT)	-	Cr, Cu, and Ni	Cd and Pb	-
Residential area (RA)	-	Cd, Cr, Cu, Ni, and Pb	-	-
Agricultural farm (AF)	Cr and Ni	Cu and Pb	Cd	-
Contamination level based on CD values				
Land-use type	(CD<5)/ Low	(5 ≤ CD<10)/ Moderate	(10 ≤ CD<20)/ Considerable	(20 ≤ CD)/ Very high
Commercial area (CA)	-	-	(+)	-
HT ring roadside (HT)	-	-	(+)	-
Residential area (RA)	-	-	(+)	-
Agricultural farm (AF)	-	(+)	-	-

(+) indicates contamination level for the respective land-use.

Table 7. Pollution load index (PLI) and classification of soil pollution in different land-use urban soils of Kathmandu

Index	Land-use type			
PLI	Commercial area (CA)	HT ring roadside (HT)	Residential area (RA)	Agricultural farm (AF)
	2.36	2.96	1.90	1.12
	Classification of PLI			
Land-use type	Polluted soil (PLI >1)		Non-polluted soil (PLI < 1)	
Commercial area (CA)	(+)		-	
HT ring roadside (HT)	(+)		-	
Residential area (RA)	(+)		-	
Agricultural farm (AF)	(+)		-	

(+) indicates the polluted soil.

Pollution Load Index (PLI)

The PLI is the mutual pollution effect in soil by multiple HMs and the estimated values are presented in Table 7.

The PLI value equal to zero means perfection; a value of 1 indicates the presence of only a baseline level of pollutants and values above 1 indicate the progressive deterioration of soil by heavy metals [29]. The results showed variable PLI values in the urban soils of the selected land-uses (Table 7). Accordingly, the PLI values were found in the descending order of HT (2.96) > CA (2.36) > RA (1.90) > AF (1.12). Since the PLI value is greater than the maximum limit of 1.0, soil samples from all land-uses may be classed as polluted soil. The high PLI value for heavy traffic (2.96) and commercial area (2.36) could be attributed to the cumulative concentrations of HMs implying an alarming condition. The results of the present study are also in agreement with Islam *et al.* [32] who also reported the high PLI

value in different land-use of urban soils of Bangladesh indicating the progressive deterioration of soil due to metal contamination. Mehr *et al.* [16] also reported the high PLI values

in urban top-soils of Isfahan province, Iran indicating that the highly contaminated cities of the country were mostly affected by pollution from traffic, industries, and Pb-Zn mine.

Geo-accumulation index (Igeo)

The I-geo index is a widely accepted method for assessing the degree of metal contamination or soil pollution [70]. The estimated I-geo values are presented in Table 8 and the degree of contamination level based on the I-geo values is shown in Table 9.

The results revealed that the I-geo values were found to vary among the selected HMs and land-uses (Table 8). Among the land-uses, the HT site showed the highest I-geo values for all the selected HMs in the descending order of Pb (1.02) > Cd (0.77) > Cu (0.52) > Ni (0.43) > Cr (0.42) followed by commercial area in the same trend except for Cu. Similarly, Pb indicated predominantly the high I-geo values for the commercial area (0.72) and residential area (0.59). The agricultural farm was mainly contaminated with Cd (0.61) followed by Cu (0.21), while Cr and Ni appeared to be the least contaminated elements in this land-use type. These results are in agreement with the findings of Proshad *et al.* [71]. They also obtained a high I-geo value for Cd and low for Cr in agricultural soils of Bangladesh where they attributed to the use of polluted water for irrigation and agrochemicals in agricultural soils [72]. Based on I-geo values estimated for the selected HMs (Table 9), all land-uses except the HT site for Pb (1.02) demonstrated their I-geo range of $0 < Igeo \leq 1$ for the HMs. Accordingly, these land-uses may be graded under the 1st degree of soil pollution and classed on average as unpolluted to moderately polluted. On the other side, the I-geo value for Pb in HT site indicated moderately polluted and contaminated by the 2nd degree of soil pollution.

Table 8. Geo-accumulation index (I-geo) of HMs in different land-use urban soils of Kathmandu.

Land-use type	Heavy metals				
	Cd	Cr	Cu	Ni	Pb
Commercial area (CA)	0.66	0.37	0.36	0.37	0.72
HT ring roadside (HT)	0.77	0.42	0.52	0.43	1.02
Residential area (RA)	0.58	0.34	0.33	0.30	0.59
Agricultural farm (AF)	0.61	0.09	0.21	0.08	0.58

Table 9. Contamination degree and grade of soil quality based on I-geo value in different land-use urban soils of Kathmandu.

Igeo value	Land-use type				Degree of soil pollution	Grade of soil quality
	Commercial area (CA)	HT ring roadside (HT)	Residential area (RA)	Agricultural farm (AF)		
Igeo ≤ 0	-	-	-	-	0	Practically unpolluted
0 <Igeo ≤ 1	Cd, Cr, Cu, Ni, and Pb	Cd, Cr, Cu, and Ni	Cd, Cr, Cu, Ni, and Pb	Cd, Cr, Cu, Ni, and Pb	1	Unpolluted to moderately polluted
1 <Igeo ≤ 2	-	Pb	-	-	2	Moderately polluted
2 <Igeo ≤ 3	-	-	-	-	3	Moderately to strongly polluted
3 <Igeo ≤ 4	-	-	-	-	4	Strongly polluted
4 <Igeo ≤ 5	-	-	-	-	5	Strongly to extremely polluted
Igeo ≥ 5	-	-	-	-	6	Extremely polluted

Potential ecological risk (PER)

The estimated ecological risk factor (ER) and potential ecological risk (PER) of HMs for the selected land-uses are presented in Table 10 and their grades of ecological risk are indicated in Table 11.

Table 10. Ecological risk factor (ER) and potential ecological risk (PER) of heavy metals (HMs) in different land-use urban soils of Kathmandu

Land-use type	Ecological risk factor (ER) of individual HM					Potential ecological risk, PER = ∑ER
	Cd	Cr	Cu	Ni	Pb	
Commercial area (CA)	98.10	3.72	9.10	10.98	18.05	139.95
HT ring roadside (HT)	114.90	4.22	13.00	12.72	25.30	170.14
Residential area (RA)	86.10	3.34	8.10	9.06	14.60	121.20
Agricultural farm (AF)	90.90	0.91	5.30	2.52	14.35	113.97

Results revealed that the ER values of the selected HMs for CA and RA sites were both in the descending order of Cd > Pb > Ni > Cu > Cr, whereas both HT and AF sites followed the order of Cd > Pb > Cu > Ni > Cr. For Cd, the highest ER values were found in the descending order of HT (114.90) > CA (98.10) > AF (90.90) > RA (86.10). These results are in consistent with the findings of Islam *et al.* [73] and Proshad *et al.* [71] who also reported the highest ER value for Cd in soils showing severe ecological risk for different land-uses under different anthropogenic activities in Bangladesh.

As for Pb, the descending order of HT (25.30) > CA (18.05) > RA (14.60) > AF (14.35) was observed. It was further noted that all the selected land-uses were found to have the least ER values for Cr indicating that Cr has the least ecological impact on the surface soils. It is evident from the EF values (Table 11) that the surface soils of all the selected land-uses indicated a grade of low ecological risk (ER < 40) for Cr, Cu, Ni, and Pb except for Cd. On the other hand, Cd showed a grade of considerable ecological risk (80 ≤ ER < 160) for all the land uses.

Table 11. Indices and grades of ecological risk factor (ER) and potential ecological risk (PER) of heavy metals in different land-use urban soils of Kathmandu

ER value/Grade of ecological risk of individual HM	Ecological risk factor (ER) Land-use type			
	Commercial area (CA)	HT ring roadside (HT)	Residential area (RA)	Agricultural farm (AF)
(ER < 40)/ Low risk	Cr, Cu, Ni, and Pb	Cr, Cu, Ni, and Pb	Cr, Cu, Ni, and Pb	Cr, Cu, Ni, and Pb
(40 ≤ ER < 80)/ Moderate risk	-	-	-	-
(80 ≤ ER < 160)/ Considerable risk	Cd	Cd	Cd	Cd
(160 ≤ ER <320)/	-	-	-	-

High risk (ER ≥ 320)/ Very high	-	-	-	-

Potential ecological risk (PER)

PER value/ Grade of potential ecological risk	Land-use type			
	Commercial area (CA)	HT ring roadside (HT)	Residential area (RA)	Agricultural farm (AF)
(PER< 65)/ Low risk	-	-	-	-
(65 ≤ PER < 130)/ Moderate risk	-	-	(+)	(+)
(130 ≤ PER < 260)/ Considerable risk	(+)	(+)	-	-
(PER ≥ 260)/ Very high risk	-	-	-	-

(+) indicates ecological risk level for respective land-use.

Potential ecological risk (PER) represents the sensitivity of various biological communities to toxic substances and illustrates the potential ecological risk caused by the HMs [32]. The PER values comprising the HMs sum computed for each land-use are presented in Table 11. The results showed variable PER values for the selected land-uses. Accordingly, the HT site showed the highest PER value (170.14) followed by CA (139.95), RA (121.20), and AF (113.97). Hence, it may be inferred based on the PER values that RA and AF sites suffered from a potential ecological risk of moderate grade (65 ≤ PER < 130), whereas HT and CA showed a grade of considerable risk (130 ≤ PER < 260) for the same ecological risk parameter. Islamd *et al.* [13] also reported considerable to a very high ecological risk for Cd in soils of different land-use urban soils of Bangladesh.

Conclusion

In this study, pH, TOC, and concentrations of Cd, Cr, Cu, Ni, and Pb were measured in the urban soil samples collected from four different land-uses of Kathmandu district, Nepal with a view to assessing the potential ecological risk posed by these elements. Whereas pH values indicated the alkaline nature of the urban soils in almost all land-uses, the TOC elevated level suggested the study area as an important sink of organic materials. The overall mean concentration of HMs in soils was found in the descending order of Cr > Ni > Cu > Pb > Cd accounting, respectively, for 35, 26, 21, 19, and < 1% of $\sum_5$HM. These concentrations were 2-4 times greater than the background concentration in shale suggesting either contamination or the influence of pedogenic factors. Also, the HMs concentration in soils followed the descending order of heavy traffic ring road > commercial > residential > agricultural land use. The HMs were poorly correlated with pH and TOC suggesting that both the parameters had a very little influence on the HMs contamination. Besides, Cr and Ni concentrations in this study were several folds higher than the reported values from across several cities/countries. The ecological risk assessment indicated a different degree of contamination and a risk level in urban soils of the land-uses under the present study. Accordingly, the CF values in the range of 0.42–5.06 indicated low to considerable risk, whereas CD in the range of 7.83–15.72 corresponded to moderate to considerable risk. The PLI values (1.12-2.96) revealed that the urban soils of all land-uses were found to be polluted with HMs, while the Igeo

values in the range of 0.08–1.02 indicated a class of unpolluted to moderately polluted soil quality of the land-uses. Similarly, the ER (0.91–114.90) and PER (113.97–170.14) values indicated that all the land-use urban soils suffered from low to considerable, and moderate to considerable risks, respectively. As for HMs, Cd and Pb showed a considerably a high ecological risk for soils from all land-uses. Besides, the estimated pollution indices demonstrated the HT land-use as the most HMs enriched site suggesting the use of appropriate remediation measures for the contaminated soils. In conclusion, further study is required for investigating the potential sources of HMs in multi-environmental matrices of other land-use types in Nepal as an attempt to protect soils from long term HMs accumulation.

Conflict of Interest

The authors declare no conflict of interest regarding the publication of this paper.

Authors' Contribution

All authors contributed equally to the manuscript at every stage of research work, manuscript preparation, and critical revision of the manuscript as for important intellectual content and the final approval of the manuscript.

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