Research Article

Assessment of potentially toxic elements' contamination in surface soils of Kulsi River Basin in North East India

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Abstract

The assessment of potentially toxic elements' contamination in surface soils of Kulsi River Basin in North East India has been studied based on the analysis of 50 soil samples collected from the basin. The average concentrations of Co (2.30 mg/kg), Ni (7.26 mg/kg), Pb (9.41 mg/kg), and Zn (22.7 mg/kg) surpassed the background levels prescribed for sedimentary rocks which indicated anthropogenic contribution of these metals. The average values of enrichment factors for different elements under study followed the order as Co > Ni > Zn > Pb > As > Fe > Cr. Results of multi-element indices revealed significant contamination at many sites; however, computed values for potential ecological risk index indicated only low ecological risk at all the sampling sites. The contribution of individual toxic elements toward the potential ecological risk followed the order as Co > Ni > As > Pb > Zn > Cr. The study recommends that suitable measures need to be taken for checking any further contamination in the area.

Keywords Hierarchical clustering analysis · Kulsi River Basin · North East India · Pollution indices · Potentially toxic elements · Principal component analysis

1 Introduction

Potentially toxic elements' contamination of surface soils has emerged as a significant environmental issue throughout the world due to persistent, toxic, non-biodegradable, and bio-accumulative nature of toxic elements [1–3]. The toxic elements present in the soil can be either natural or anthropogenic in origin [4–6]. Generally, toxic elements' content of anthropogenic origin surpasses the geochemical background levels in soil [4, 7, 8]. The condition is becoming more severe in developing countries due to rapid increase in population, industrialization, and modern practices of agriculture [9, 10]. After accumulation in soil, these elements can deteriorate the soil quality and disrupt the common biochemical processes taking place in the soil–water–air continuum and ultimately leads to

reduction in crop yield and affect the quality of agricultural products [5, 11]. Further, these elements may also affect the health of human beings and animals by entering into food chain via crop grown on the soil affected by such toxic elements [7, 12].

Keeping in view the severe environmental and ecological impacts of these elements, various indices like geo-accumulation index (I_{geo}) [13], enrichment factor (EF) [14], contamination factor (CF) [15], pollution load index (PLI) [16], Nemerow pollution index ($PI_{Nemerow}$) [17], ecological risk factor, and potential ecological risk index [15] have been developed to assess the extent of contamination and ecological risks. These indices are effective tools for processing, analyzing, and converting raw environmental data to valuable information based on which the decision makers, managers, and technicians can rank the contaminated

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areas for further investigations and improvements [18, 19]. Caeiro et al. [18] classified the indices into three categories: contamination indices, background enrichment indices, and ecological risk indices, whereas Qingjie et al. [20] classified the commonly used pollution indices into two categories, i.e., single indices and integrated indices. Single indices are used to calculate elemental contamination by only single element, and these include contamination factor, ecological risk factor, enrichment factor, and index of geo-accumulation, etc. On the other hand, integrated indices are used to calculate the extent of contamination by more than one element and are calculated based on the values determined for single indices.

Kulsi River Basin is one of the important sub-basins of the Himalayan river system which has demonstrated active river migration in the past as well as present times. Kulsi river watershed is surrounded by the River Brahmaputra in the north, hills of Meghalaya plateau in the south, the watersheds of Deosila and Rani in the east and west, respectively. Administratively, this watershed is bordered by Nalbari district in the north; west Khasi hill district of Meghalaya in the south; and Kamrup district in west and east side. Since this watershed is located in an arsenic- and fluoride-rich belt [10], it seems necessary to evaluate the soil and water characteristics of the area. In earlier papers [21, 22], groundwater quality of Kulsi River Basin has been reported. In the present study, an attempt has been made to study the potentially toxic elements' contamination in surface soils of this river basin. The objective is to profile the accumulation of potentially toxic elements (Fe, Ni, Cr, Pb, Zn, As, Co) and the associated ecological risk in the region with main focus in terms of their toxicity and enhancement strategies. For comprehensive evaluation of the existing state of soil in the area, the extent of toxic elements contamination/enrichment in the soil was assessed by employing several single and multi-element indices. The results obtained can provide baseline information for environmental management in the region.

2 Methodology

2.1 Study area

The Kulsi River Basin is a part of the Brahmaputra Basin and is situated on the south bank of the mighty River Brahmaputra between 25°30′N–26°10′N latitude and 89°50′E–91°50′E longitude with an elevation of 100–1900 m above mean sea level. The total length of the river is 220 km. The river originates in Meghalaya from the northern slope of the West Khasi hills and flows toward north and enters Kamrup district of Assam and drains out a total area of 2806 km². The basin covers some part of

Kamrup District of Assam as well as part of West Khasi Hills and Ribhoi Districts of Meghalaya (Fig. 1).

The geology of the river basin consists mostly of gneiss and sandstones overlain by deep to moderately deep soil layer. Much of the terrain is rough, rolling to steeply sloping. Under saturated conditions, such a formation is highly conducive to rapid subsurface storm flow. The rock types in the Kulsi basin vary from Precambrian stage to recent. The surface Geological formation is newer alluvium sand, gravel, clay, and silt. The Assam part of the basin that falls in Kamrup District has two distinct groups of rock formations, i.e., consolidated and unconsolidated. The soil data show distribution of clay loam type of soil in the plain areas, sandy clay soil in forest area and sandy loam in the hilly areas (Master Plan of Kulsi-Deosila Sub Basin [23]).

2.2 Sampling and chemical analysis

Fifty soil samples (0–20 cm) were collected from Kulsi River Basin on grid pattern with size of the grid as 5 km × 5 km. Samples were packed into polyethylene bags and brought to the laboratory for further processing and analysis. The distribution of all the sampling locations in the study area is presented in Fig. 1. All the collected soil samples were left for air-drying and then passed through a sieve of 2 mm pore size to remove coarse particles. The particle size of the soil samples for microwave digestion was less than 200 µm. The pH of the soil suspension (1:5) was measured and found to vary from 6.8 to 7.7. Sieved samples (0.3 g each) were digested using microwave digestor (Anton Paar Model Multiwave PRO) with a mixture of concentrated acids (5 mL HNO_3, 2 mL HF, and 1 mL $HClO_4$). Concentrations of eight elements (Fe, Ni, Cr, Pb, Zn, As, Co, and Al) were determined using ICP-MS (Perkin Elmer Model Elan DRC 6100). The detection limit range for iron, nickel, chromium, zinc, and aluminum is 1–10 µg/L, whereas, for lead detection, limit range is 0.1–1 µg/L. Standard solutions of metal ions were procured from Merck, Germany. Accuracy and precision of the analytical results were within 5%. Ultrapure water was used throughout the study.

2.3 Statistical analysis

Statistical analysis was performed using SPSS 16.0 software. For a general description of the obtained results, the descriptive statistical analysis of the data (mean, minimum, maximum, standard deviation (SD), and coefficient of variation (CV) was done. The multivariate statistical tools like principal component analysis (PCA) and hierarchical clustering analysis (HCA) were employed with the objective to recognize associations and common origin among elements. HCA classify elements into different geochemical groups by clustering
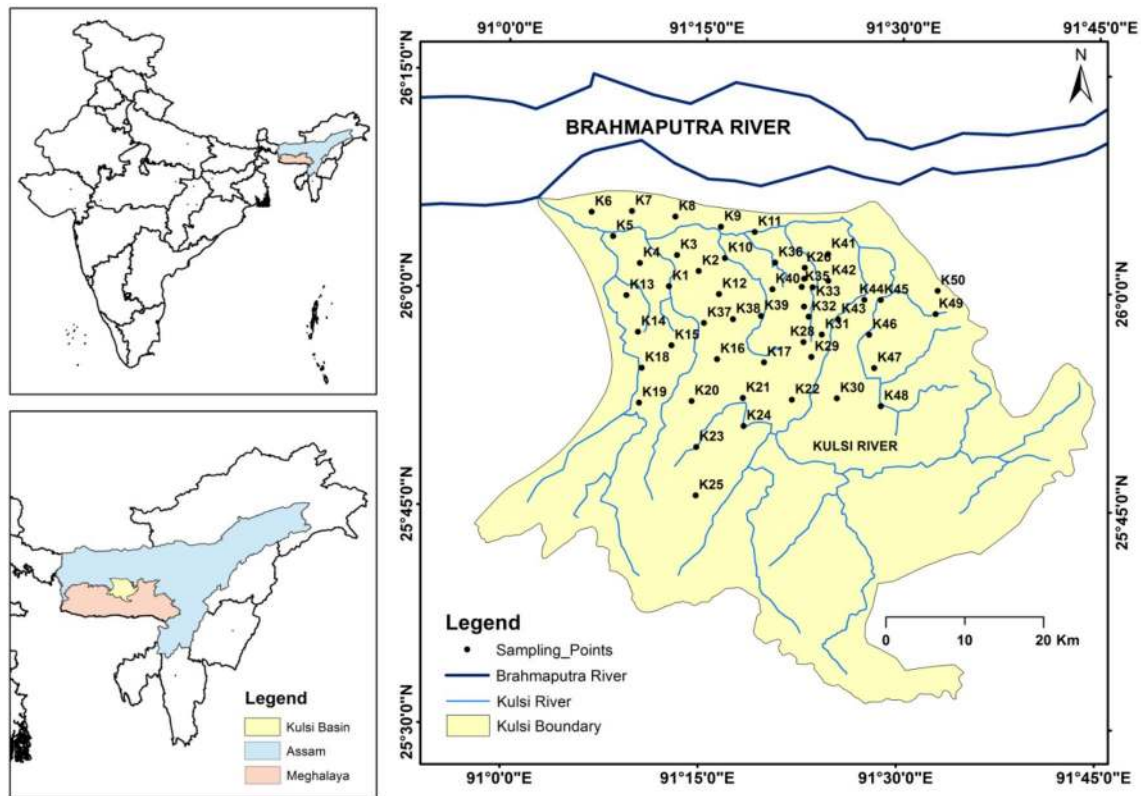
Fig. 1 Kulsi River Basin showing location of sampling points

them and the results were obtained in the form of a dendrogram which provide a visual summary of the clusters. For further confirmation about source of toxic elements, PCA was used with Varimax rotation to minimize the number of variables with a high loading on each component.

2.4 Soil quality with pollution indices

Soil quality was assessed by applying various single- and multi-element indices to the results obtained from elemental analysis of all the samples. Single-element indices like geo-accumulation index (I_{geo}), enrichment factor (EF), and contamination factor (CF) were applied for assessment of contamination by individual elements, while multi-element indices like degree of contamination, modified degree of contamination, pollution load index, and Nemerow pollution index ($PI_{Nemerow}$) were applied to calculate the extent of contamination by more than one element and calculated based on the values determined for single indices. Formulas and categories of contamination based on the value of each index are summarized in Table S1 (Supplementary Information).

2.5 Ecological risk assessment

In order to assess the ecological risk posed by individual and combined effect of the elements under consideration, ecological risk factor and potential ecological risk index were computed. Formulas and categories of risk for both the indices are given in Table S1 (Supplementary Information).

3 Results and discussion

3.1 Concentration of potentially toxic elements in soil samples

The concentrations of various elements in the soil samples collected from different locations of the Kulsi River Basin were determined and descriptive statistics of the data like average, minimum, maximum, standard deviation, and coefficient of variation (%) are presented in Table 1. The average concentrations of Ni, Pb, Zn, and Co surpassed the background levels prescribed for sedimentary rocks. However, when compared with the world normal averages, it has been observed that the toxic element concentrations (Ni, Cr, Pb, Zn, As and Co) in the analyzed samples were

Table 1 Potentially toxic elements' concentration in soil samples of Kulsi River Basin (mg/kg)

Sample ID	Fe	Ni	Cr	Pb	Zn	As	Co	Al
Average	2189.38	7.26	5.52	9.41	22.70	0.90	2.30	22,986.52
Min.	101.65	2.08	0.26	3.81	8.13	0.06	0.48	7578.35
Median	2084.75	6.47	5.69	8.53	19.94	0.88	2.18	23,197.82
Max.	4325.61	14.03	13.10	31.66	69.48	2.05	5.40	45,541.20
SD	1177.34	3.07	3.32	5.23	11.95	0.46	1.12	10,475.50
CV (%)	53.78	42.28	60.12	55.65	52.63	51.38	48.7	45.57
World soil average[a]	–	29	59.5	27	70	6.83	11.3	–
Background values[b]	9800	2	35	7	16	1	0.3	25,000

SD Standard deviation, *CV* coefficient of variation

[a]Mean values compiled from [24]

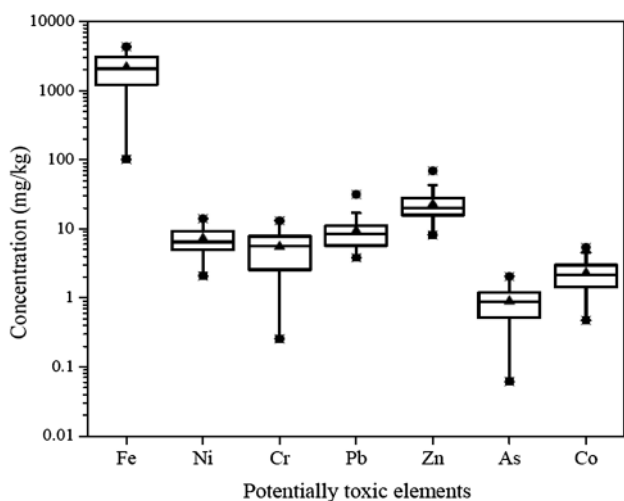[b]Values given by [25] for sedimentary rocks (sandstone)

Fig. 2 Box and whisker plot of concentrations of potentially toxic elements in soil samples

very low. Only, concentration of Pb was slightly higher than the world normal average concentration at site Bagdoba (K2) and Sautara (K7) (Fig. 2). Moreover, comparison with other studies from India, notably in Sukinda (Odisha),

Zaheerabad (Telangana), and Singhbhum (Jharkhand) also showed that the average concentration of toxic elements in Kulsi River Basin is very low [26, 27].

Further, CV (%) was calculated for all the elements under consideration and the values indicated that the variation was least in case of Al content and highest in case of Cr content. Therefore, Al has been used as normalization/reference element in the study [28].

3.2 Identification of source of potentially toxic elements

Pearson's correlation coefficient matrix among the selected toxic elements in the soil samples of the study area is presented in Table 2. A strong linear correlation was evident between Fe and Cr ($r = 0.81$), which indicated a common origin of these elements. Cr, As, Ni, and Co also formed highly correlated pair with a correlation coefficient of 0.74 and 0.70, respectively, suggesting that these may be originated from some common sources. Industrial wastes, fertilizers/pesticides, and disposal of sewage sludge might be some of the causes for the elevated concentration of these correlated elements [29–31]. Both Fe and Ni exhibited strong positive correlations with As. This

Table 2 Pearson's correlation matrix for potentially toxic elements in soil samples

	Fe	Ni	Cr	Pb	Zn	As	Co
Fe	1.00						
Ni	0.17	1.00					
Cr	**0.81**	0.55	1.00				
Pb	− 0.10	0.15	− 0.03	1.00			
Zn	0.09	0.32	0.16	− 0.11	1.00		
As	**0.67**	**0.60**	**0.74**	0.07	0.01	1.00	
Co	0.30	**0.70**	0.48	0.13	0.30	0.49	1.00

Bold values represent strong correlation between the associated elements; for example, r = 0.81 indicates strong correlation between Fe and Cr. Similarly, Fe and As; Ni and Co; Ni and As etc. are strongly correlated
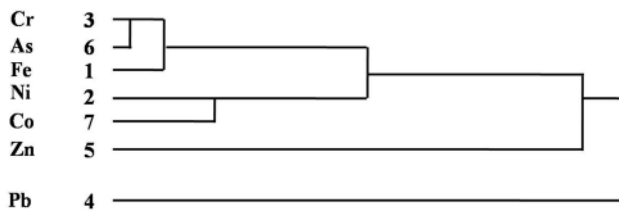
Fig. 3 Dendrogram of hierarchical cluster analysis

Table 3 Varimax rotated factor loadings of potentially toxic elements in soil samples

Element	Components[a]		
	1	2	3
Fe	**0.908**		− 0.181
Ni	0.368	**0.779**	0.277
Cr	**0.894**	0.290	
Pb			**0.891**
Zn	− 0.118	**0.771**	− 0.491
As	**0.854**	0.246	0.193
Co	0.375	**0.735**	0.226
% of variance	37.84%	27.01%	16.69%
Cumulative %	37.84%	64.85%	81.54%

[a]Values in bold depict the corresponding clusters in Fig. 3. For example Component 1 (in Table 3) corresponds to cluster 1 (in Fig. 3) viz. Fe, Cr, As. Similarly, components 2 and 3 correspond to clusters 2 and 3, respectively

is due to the fact that arsenic usually occurs in combination with a number of elements, such as Ca, Fe, Mn, in the form of calcium arsenate ($Ca_3(AsO_4)_2$) and Fe- and Mn-oxi/hydroxides [10, 32]. Negative correlation between Fe and Pb, Cr and Pb, and Pb and Zn suggested that their sources were quite different from those of the others.

Both principal component analysis (PCA) and hierarchical cluster analysis (HCA) were carried out to group the variables (i.e., elements) based on the similarities of their sources. Similar kind of studies has also been performed earlier in India as well as other regions of the world [26–28]. The elements were grouped into three clusters in HCA (Fig. 3), which also correlated with the three principal components (PCs) in the PCA (Table 3). The three PCs explained a total variance of 81.54% in the data set and allowed the tentative grouping of elements as per sources. The first component in PCA (Table 3) explained 37.84% of the data variance and correlated with the first cluster in HCA (Fig. 3, comprising of Fe, Cr, and As). The second factor in PCA with 27.01% of variance comprises Ni, Zn, and Co showing resemblance to Cluster 2 of HCA in Fig. 3. The third PCA factor explained 16.69% of data variance with elevated loadings of Pb. This can be linked to cluster 3 in

the HCA comprising of Pb only (Fig. 3). The strong correlation among Zn, Ni, and Co could be attributed to anthropogenic activities [33]. Zn, Ni, and Co are the markers for diesel and lubricant oil combustion and tire and brake abrasion [33–35]. Pb could be linked to traffic emissions [35, 36].

3.3 Assessment of soil quality by single pollution index

Index of geo-accumulation (I_{geo}) as proposed by Müller [13] was used to describe the elemental contamination by comparing the current concentrations of selected elements (Fe, Ni, Cr, Pb, Zn, As, and Co) with their pre-industrial levels. The comparison was based on seven classes of qualification [20]. Figure 4a represents box and whisker plot for I_{geo} values for Fe, Ni, Cr, Pb, Zn, As, and Co. Figure 4a depicts that the average I_{geo} values for Fe, Cr, Pb, Zn, and As were less than 0 which indicate no pollution. On the other hand, average I_{geo} values for Ni was 1.15 which indicated slight pollution, whereas, average I_{geo} value for Co was 2.16 which pointed toward moderate pollution. Ni and Co were also found to be higher as compared to the results reported by Giri et al. [27] for the soil samples of the Singhbhum region of India.

Enrichment Factors (EF) were determined to assess the degree of anthropogenic contributions of elements to the soils of area under study [26–28]. As in the results obtained from elemental analysis, Al content showed low variability, hence, Al was taken as a reference element in order to assess the expected effect of anthropogenesis on elemental accumulation. The average values of EF for Fe, Ni, Cr, Pb, Zn, As, and Co were 0.26, 5.21, 0.19, 1.87, 2.02, 1.11 and 10.66, respectively, with the ranges of 0.02–0.57, 1.09–19.71, 0.02–0.52, 0.37–8.81, 0.30–6.82, 0.20–2.87 and 2.11–33.56, respectively. Box and whisker plot (Fig. 4b) showed that the EF values of Fe and Cr were less than 1 which indicated depletion to minimal enrichment or natural origin of these two elements. The values for As were between 0.20 and 2.87, suggesting minimal to moderate enrichment. In addition, with the highest EF value at greater than 5, Ni, Pb, Zn, and Co showed significant enrichment. These findings suggested that the soils in the area under study were affected by anthropogenic activities like diesel and lubricant oil combustion, tire and brake abrasion and traffic emissions [27, 33–37].

Contamination factor (CF) is also a single index indicator proposed by Håkanson [15] and was used to evaluate contamination by individual elements. This provided information regarding how the elements under study were concentrated at a particular site relative to the background site. Figure 4c summarizes the contamination factors of elements in the soil samples of Kulsi River Basin. Results
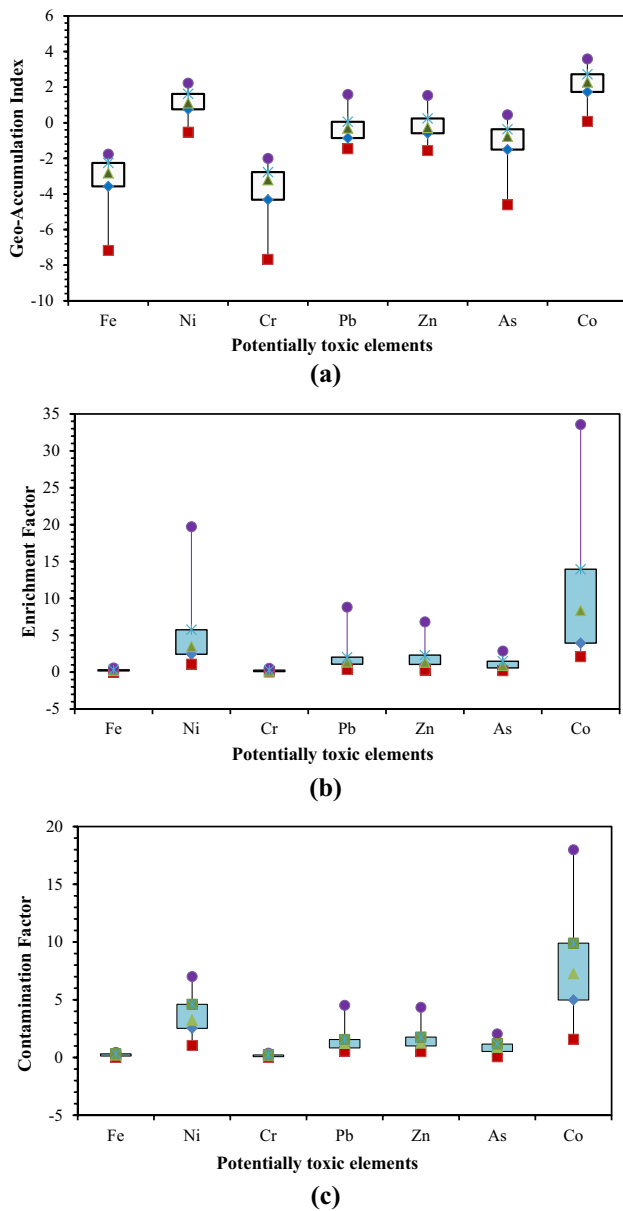
Fig. 4 Box and whisker plot displaying **a** Geo-accumulation index, **b** enrichment factor and **c** contamination factor for individual elements

Fig. 5 Graphical representation of pollution indices at different sites

showed that the average CF values for Fe, Ni, Cr, Pb, Zn, As, and Co were 0.22, 3.63, 0.16, 1.34, 1.42, 0.90 and 7.66, respectively, with the ranges of 0.01–0.44, 1.04–7.01, 0.01–0.37, 0.54–4.52, 0.51–4.3, 0.06–2.05 and 1.58–17.99, respectively. From the four contamination categories enumerated by Qingjie et al. [20], average values for all the elements showed low to moderate contamination except of Ni and Co which showed considerable and very high average contamination, respectively. CF values in case of Ni indicated moderate contamination at 20 sites, consid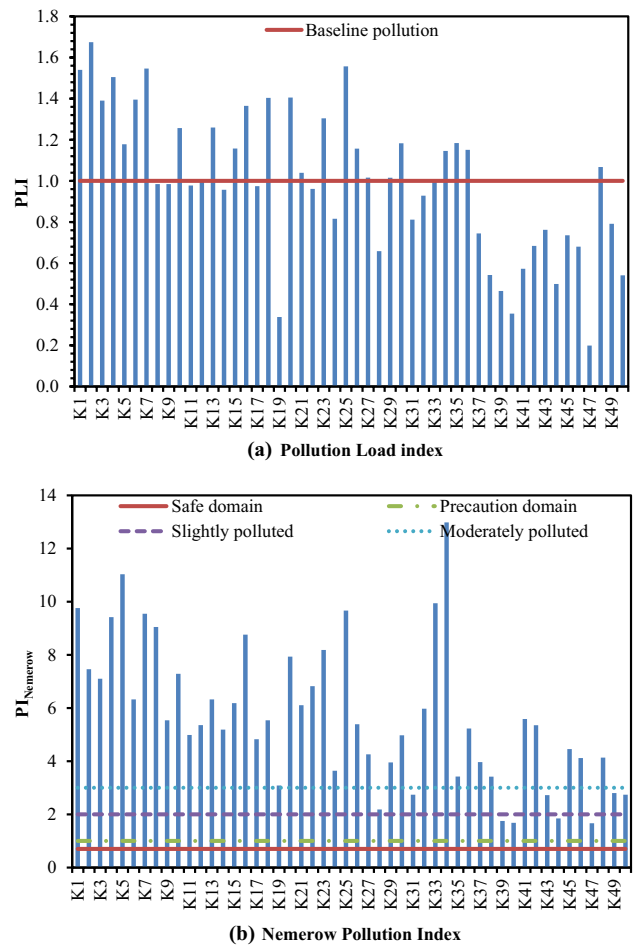erable contamination at 24 sites, and very high contamination at 6 sites. However, CF values in case of Co indicated moderate contamination at 5 sites, considerable at 13 sites, and very high contamination at rest 32 sites. The contamination factor for different elements followed the order: $Cr < Fe < As < Pb < Zn < Ni < Co$.

3.4 Assessment of soil quality by multi-element index

The status of contamination by elements under consideration was evaluated using the pollution load index (PLI). The pollution load index (PLI) ranged from 0.20 to 1.67 (Fig. 5a) with average PLI value 1. It is to be noted that a PLI value = 1 depicts toxic elements' load near to the background level, while PLI > 1 indicates the pollution due to toxic elements [38]. Out of 50 sites, PLI value for 26 sites was ≥ 1 indicating increased pollution, while rest of the sites were non-polluted with PLI value less than 1. However, Nemerow pollution index (PI_Nemerow) showed that the Kulsi River Basin is heavily polluted. For PI_Nemerow, only 4
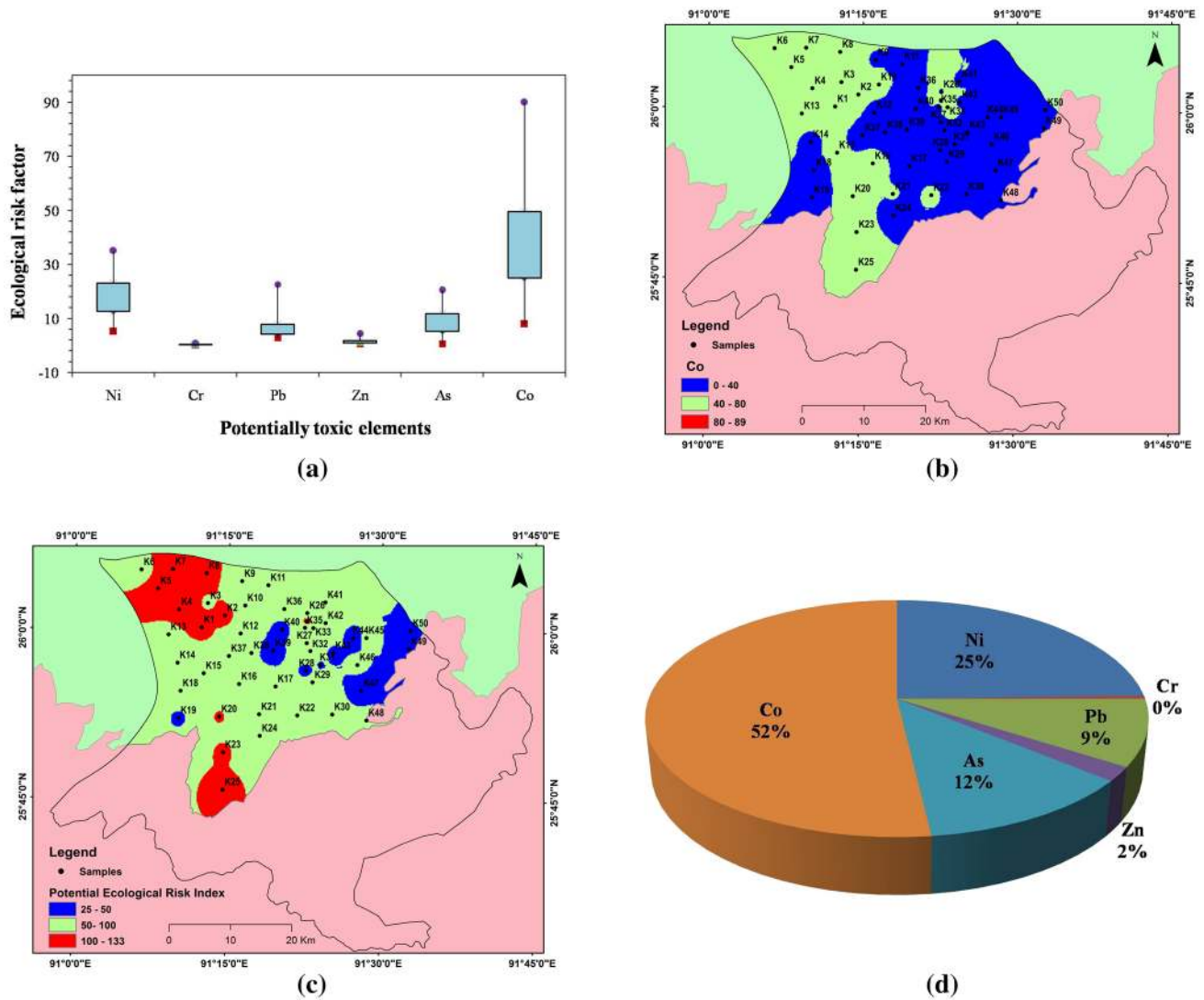
Fig. 6 Ecological risk assessment through **a** ecological risk factor for individual elements, **b** at different sites and **c** potential ecological risk index at different sites with **d** contribution of different elements toward potential ecological risk index

sites (K39, K40, K44 and K47) were slightly polluted, and four sites (K28, K43, K49 and K50) were moderately polluted while all the remaining sites were severely polluted (Fig. 5b). This variation in results may be due to the fact that Nemerow pollution index is basically used for assessing the overall quality of soil which does not take into account of the weighing factors [39].

3.5 Ecological risk assessment

Both single- and multi-element indices calculated above deal only with anthropogenic impacts of elements. However, in addition to the anthropogenic impacts of elements present in the soil, further assessment of the potential ecological risks posed by the multiple elements is also required. Hence, potential ecological risk index (RI) was

calculated to ascertain the ecological risk posed by the elements present in the soils.

Box and whisker plot for ecological risk posed by the individual element (E_r) computed with contamination factors (C_f) is presented in Fig. 6a. The average E_r values for Ni, Cr, Pb, Zn, As, and Co were 18.14, 0.32, 6.72, 1.42, 8.96 and 38.31, respectively, with the ranges of 5.19–35.06, 0.01–0.75, 2.72–22.61, 0.51–4.34, 0.61–20.47 and 7.92–89.95, respectively. All elements under consideration (except Co) pose low ecological risk at all sites with E_r value less than 40. The E_r values for Co revealed that Co present some form of ecological risk at some sites. Cobalt (Co) poses low risk at 31 sites, moderate risk at 18 sites, and considerable risk at one site (K34) only (Fig. 6b). Comparison of this result with the soil samples of another site in India (Singhbhum, Jharkhand) shows that all the elements

pose low risk except Ni (18.14 in Kulsi basin vs. 13.2 in Singhbhum) [27].

However, the computed values of potential ecological risks (RI) posed by the combined effect of all the elements under consideration was less than 150 at all the sites suggesting that all the sites exhibited low ecological risk (Fig. 6c). The usage of ecological risk index is of great value here, which suggests that any contamination level cannot cause ecological risk.

The contribution of individual elements toward the potential ecological risks is summarized in Fig. 6d. It is evident from Fig. 6d that the major contributor to average potential ecological risk is cobalt (52%) followed by nickel (25%), Arsenic (12%), lead (9%) and zinc (2%). Cr has negligible or no contribution toward average potential ecological risk.

4 Conclusion

Anthropogenic activities and the often-associated contamination of soils have become a major environmental problem during recent years. The results of the present study revealed marked variations in distribution of selected elements in the Kulsi River Basin. Enrichment factor values show that average concentration of Ni, Pb, Zn and Co surpassed the background levels which indicate the anthropogenic contribution of these elements. Elemental contamination in the soils of this region can be attributed to diffuse pollution sources like traffic, agriculture, and frequent inundation of floodplain areas with contaminated river water. The anthropogenic contribution of above mentioned elements is further supported by the results of multivariate analysis along with the average EF value greater than 5. Results of various indices show that the Kulsi River Basin has significant contamination of potentially toxic elements. Therefore, effective environmental management is needed for ameliorative measures as well as to check any further contamination from potential sources.

Compliance with ethical standards

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