Appraisal of Pollution and Source Apportionment of Potentially Toxic Elements in Different Soils Collected around the Industrial Area

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Appraisal of pollution and source apportionment of potentially toxic elements in different soils collected around the industrial area

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Abstract

It is imperative to understand the pollution of potentially toxic elements (PTEs) in different soils in order to determine the sustainable management approaches for soils. Potentially toxic elements (Fe, Mn, As, Pb, Zn, Ni, Cu, Cn, Co and Cd), and pH and organic carbon were determined in agricultural, non-agricultural and industrial soils of Indian, Punjab. The findings of PTEs indicated that industrial soils recorded highest concentration of PTEs followed by non-agricultural and agricultural soils. The percentage change recorded from agricultural to non-agricultural soils for PTEs were 3.19% for Fe, 25.3% for Mn, 63.8% for Cu, 13.5% for Cn, 49.8% for Pb, 79.6% for Ni, 35.8% for Co and 32% for Cd. From non-agricultural to industrial soils the percentage change observed for PTEs were 89% for Zn, 2.03% for Fe, 21.9% for Mn, 68.2% Cu, 9.2% for Cn, 35.8% for Pb, 18.4% for Co, 30.4% for Cd and 43.4% for As. The results of contamination factor, enrichment factor, geoaccumulation index, pollution and modified pollution indices indicated that Cd and As showed severe contamination in all studied soil types. Ecological risk assessment results revealed that Cd exhibited very risk in different soil types. The outcomes of this study will aid in forming approaches to decline the perils allied with PTEs in soils, and produce guidelines to save the environs from long term accrual of PTEs.

Keywords: Potentially toxic elements, multivariate statistical analysis, ecological risk assessment, geoaccumulation index

1. Introduction

Soil is considered as one of the most substantive non-renewable natural resource that supports plethora of flora as well as fauna (Jansson & Hofmockel, 2019). Unfortunately, unsustainable development models are degrading agricultural soils at an alarming rate (Kopittke et al., 2019). Soil receives a lot of pollutants from diverse sources related to agriculture and other industries (Naveedullah et al., 2013; Kumar et al., 2019a). The rapid and incessant usage of inorganic pesticides, herbicides etc., has led to degradation of major agricultural edaphic system. Automobiles and other transportation systems also pollute the nearby fields (Kumar et al., 2019b). Amid other unwanted contaminants, heavy metals (HMs) form major pollutants having the starkest impact of plants. Industrial effluents rich in different kinds of heavy metals also reach soil either through water used for irrigation purposes or other anthropogenic activities.

HMs have high atomic density with atomic number more than 20. Being non-degradable, they persist in soil for very long duration of time (Mahey et al., 2020). Present global agricultural requirement pressurises producers to increase yield that induces involvement of unsustainable methods like HM based mineral phosphate fertilizers. Other HMs pollution sources are rune water from minning area, sewage disposal, smelting, and cement manufacturing etc., (Walter et al., 2006; Ohunkunle & Fatoba, 2013). These HMs are taken up by crop plants and reach humans in biomagnified levels which pose a great risk to humans as is evident from increased number of pathological issues related with kidneys, liver, bone, pancreas etc. (Kawatra & Bakhetia, 2008). Piling up of HMs in soil is a major hurdle in achieving global food safety. Most of the developing cities, due to fast development, are unable to curb this menace and face major edaphic degradation due to accumulation of HMs.

The district Ludhiana of Punjab is known as industrial hub and also famously known as Manchester of India (Sikka & Nayyar, 2016). The town is famous for bicycle, woollen, hosiery, machine tools, dying and electroplating work. These industries generate huge amount of industrial effluents that are directly poured into tributary Budda Nullah through sewers. The untreated water of Budda Nullah is used to irrigate crop fields (Sikka et al., 2009). The waste reaching soil keeps on accumulating as HMs cannot be degraded or destroyed. Both essential and on essential HMS are considered toxic for terrestrial and aquatic organisms. There abundance more than threshold level cause ecological imbalance.

Keeping this thing in mind, the present work was conducted to appraise the potentially toxic elements (Fe, Mn, As, Pb, Zn, Ni, Cu, Cn, Co and Cd) and, pH and OC in agricultural, non-agricultural and industrial soils of Ludhiana, Punjab, India. Secondly, correlation analysis, principal component analysis and non-metric multidimensional scaling were performed to find the sources of PTEs. Lastly, pollution level and risk appraisal were determined by enumerating contamination indices and ecological risk assessment indices.

2. Material and methods

2.1. Study area

Soil samples were collected in triplicate from all over Ludhiana district of Punjab, India during May 2017 to October 2019 (Supplementary Table S1). The sample collection site map can be observed in **Fig. 1.** The mean temperature and humidity during collection was 35 - 42°C & 58.1 respectively (Kumar et al., 2010). Ludhiana district has climatic conditions of humid subtropical as per koppen climatic classification (Yeotikar et al., 2019). The mean rainfall is 758 mm/yr (Hadda et al., 2020). Ludhiana is situated in the middle of Punjab and comes under Malwa sub region. The soil of Ludhiana has a loamy texture. According to 2011-20 census, the total area of Ludhiana was 3, 57, 800 hectares and total population was 34, 98, 739.

2.2. Collection of samples

Five hundred gram of soil was collected from each site in triplicates. Soil samples were dried and powdered to nullify the effect of particle size during analysis. The samples were properly coded and stored till further analysis.

2.3. Analysis of soils for pH, OC and potentially toxic elements

Soil samples were analysed for pH, OC and heavy metal content. Soil pH was measured following the method of Jackson (1967). Organic carbon from soil samples was estimated by the method given by Nelson and Sommers (1996). Heavy Metal estimation was done using Varian 20 model of atomic absorption spectrophotometer. Samples were digested using diacid mixture ($HClO_4$:HNO_3 in 4:1 ratio).

2.4. Computation of pollution indices

To determine the pollution level of PTEs in soils various indices have been proposed by different workers. The computation of indices is as:

2.4.1. Contamination factor (CF)

The CF signifies the anthropogenic inputs in the soils and computed by dividing the PTEs level in determined samples to reference environment value (Hakanson 1980). The equation used to determine CF is as:

$$CF = \frac{\text{Concentartion of each PTE}}{\text{Background value of each PTE}}$$

2.4.2. Enrichment factor (EF)

EF was used to find the natural and human impacts on PTEs concentration (Delgado et al. 2010) and computed as:

$$EF = \frac{\text{Concentartion of PTE in samples/Fe concentration in the samples}}{\text{Background values of PTE/Background value of Fe}}$$

116 The reference values for PTEs were adapted from Taylor and Mclennan (1995), and
117 the scores used to classify the pollution level on CF and EF values were provided in
118 supplementary Table S2.

119 **2.4.3. Geoaccumulation index (Igeo)**

120 Igeo index indomitable the pollution of PTEs in soils and computed by following
121 Muller et al. (1981) as:

122 $$Igeo = \log_2 \frac{Heavy\,metal\,concentration\,in\,soil\,samples}{1.5 \times heavy\,metal\,concentration\,in\,background\,environment}$$

123 The reference values of PTEs were adapted from Taylor and McLennan (1995). The constant
124 1.5 implies alterations in heavy metal concentrations in the environs (Tian et al. 2017).

125 **2.4.4. Pollution index (PI) and modified pollution index (MPI)**

126 The PI was computed by considering average and maximum value of CF for apiece
127 PTE, while MPI takes into consideration the mean and maximum value of EF for apiece PTE.
128 The PI and MPI were enumerated by ensuing Nemerow (1991):

129 $$PI = \sqrt{\frac{(CF_{average})^2 + (CF_{maximum})^2}{2}}$$

130 $$MPI = \sqrt{\frac{(EF_{average})^2 + (EF_{maximum})^2}{2}}$$

131 The grades used to categorize the pollution level are given in Supplementary Table S3.

132 **2.4.5. Ecological risk indices (RI and MRI)**

133 The ecological peril in the soils was resolute by using potential and modified
134 ecological risk indices. The potential and modified risk indices were enumerated by taking
135 into account the CF and EF values of PTEs respectively and determined as:

136 $$RI = \sum CF \times Tr$$

137 $$MRI = \sum EF \times Tr$$

Where Tr is the toxic response factor of apiece PTE adapted from (Heidari et al. 2019). The scores used to classify the ecological hazard are specified in Table S4.

2.5. Statistical analysis

All the analysis was performed in triplicates and data was presented as minimum, maximum, mean, standard error, variance, skewness, kurtosis and coefficient of variance. After that Pearson's correlation analysis was performed on the data to determine the sources among PTEs in soils using R software v3.0 (Statistical Computing, Vienna, Austria). Cluster analysis and principal component analysis was performed to find source apportionment among PTEs in soils using Minitab v.14 and SPSS v.16 software (IBM, USA). Lastly, non-metric multidimensional scaling (NMDS) was conducted and it is a data reduction and score based technique for graphical depiction of the connotations amid PTEs in a multidimensional space, and performed by using PAST v.3.21 (Hammar et al. 2001).

3. Results and discussion

3.1. Statistical analysis of potentially toxic elements in different soils and their comparison with soil guidelines

The descriptive statistical analysis of pH, organic carbon (OC) and PTEs (Zn, Fe, Mn, Cu, Cn, Pb, Ni, Co, Cd and As in agricultural, non-agricultural and industrial soils were given in **Table 1.** The pH was found in the range of 6.88 to 7.73 with average value of 7.46 in agricultural soils. In agricultural soils pH ranged from 7.4 to 7.73, while in industrial soils pH ranged from 7.42 to 7.92. The pH was found slightly acidic to alkaline in the present study which is associated with reduction of PTEs mobility in the soil (Heidari et al. 2019). Our results are in corroboration with Dogra et al. (2020) in Jalandhar, Punjab, and Keshavarzi and Kumar (2019b) in Iran, and in their findings they also reported similar results of pH. The OC ranged from 0.04 to 1.35% in agricultural soils, 0.39 to 1.11% in non-agricultural soils and 0.42 to 1.42% in industrial soils and also has pronounced provocation on the transport of PTEs in all the studied soils (Troeh and Thompson 2005). The mean concentration of PTEs in

agricultural soils followed a trend: Fe > Mn > As > Pb > Cn > Cu > Ni > Co > Cd. Similarly average values of PTEs in non-agricultural and industrial soils followed the trend as: Fe > As > Mn > Pb > Cu > Ni > Cn > Co > Cd, and Fe > As > Mn > Ni > Pb > Cu > Cn > Co > Cd respectively. After comparison of PTEs of agricultural soils with Indian background soils, it was inferred that 97.5%, 82.5%, 47.5%, 100% and 42.5% sampling sites surpassed their contents for Zn, Mn, Ni, Pb and Co respectively (Gowd et al. 2010). Arsenic concentration exceeded 75% in contrast with Poland soil guidelines, while Cd concentration found low in comparison with their limits (Wcisło 2012). Similarly comparison of PTEs in non-agricultural soils with Indian background values and Poland soil limits indicated that 100% sampling sites surpassed their values for Zn, Mn, Pb, Ni, Co and As (Gowd et al. 2010; Wcisło 2012). In industrial soils, PTEs exceeded 100% for Zn, Mn, Pb, Ni, Co and As, while 80% for Cu, and 60% for Cd in contrast with Indian background values and Poland soil limits (Gowd et al. 2010; Wcisło 2012).

The percentage increase observed from agricultural to non-agricultural soils for different PTEs were 3.19% for Fe, 25.3% for Mn, 63.8% for Cu, 13.5% for Cn, 49.8% for Pb, 79.6% for Ni, 35.8% for Co, 32% for Cd, whereas 2.02 and 2.99 times increase were found for Zn and As respectively. From non-agricultural to industrial soils the percentage change observed for different PTEs were 89% for Zn, 2.03% for Fe, 21.9% for Mn, 68.2% Cu, 9.2% for Cn, 35.8% for Pb, 18.4% for Co, 30.4% for Cd and 43.4% for As, and 4.21 times increase in Ni. The percentage change from agricultural to industrial soils were 5.2% for Fe, 52.8% for Mn, 24% for Cn, 60.8% for Co and 72.2% for Cd, while 3.8, 2.7, 2.03, 7.5 and 4.2 times increase were found for Zn, Cu, Pb, Ni and As respectively. In agricultural soils, skewness of pH, OC and all PTEs were found less than one signifying normal distribution of data (Beaver et al. 2012). Kurtosis values of Fe, Mn and As were found above one signifying leptokurtic, while pH, OC, Zn, Cu, Cn, Pb, Ni and Cd showed kurtosis values less than one reflecting

189 normal distribution of data (Beaver et al. 2012). In non-agricultural soils, skewness values of
190 Cn, Ni and Co showed strongly skewed data, while for pH, OC and other PTEs skewness
191 values were found less than one (Beaver et al. 2012). The kurtosis values of pH, OC and
192 PTEs exhibited leptokurtic kurtosis except Pb. In industrial soils, the Co, Ni, Cd and As
193 recorded skewness values less than one signifying normal distribution of data, while pH, OC
194 and other PTEs showed strongly skewed data. Except Co and Cd, kurtosis values of pH, OC
195 and other PTEs showed kurtosis values above one, representing leptokurtic (Beaver et al.
196 2012). The high kurtosis of PTEs indicates that sampling sites are gathered at comparatively
197 less values (Lu et al. 2010). High coefficient of variance values were found for PTEs,
198 signifying that anthropogenic activities have pronounced control on the concentrations of
199 PTEs (Kumar et al. 2019c).

200 **3.2. Correlation analysis of potentially toxic elements in agricultural, non-agricultural**
201 **and industrial soils**

202 The person's correlation was performed on the pH, OC and PTEs data in order to find
203 the inter-associations among the studied parameters in agricultural, non-agricultural and
204 industrial soils **(Figure 2 A, B and C).** From the results of correlation analysis in agricultural
205 soils, it was found that pH and OC is negatively correlated with all the studied PTEs. All
206 PTEs are positively correlated with each other in agricultural soils. In non-agricultural soils,
207 pH is negatively and OC is positively correlated with the studied PTEs. All PTEs are
208 positively correlated with each other in non-agricultural soils. In industrial soils, pH showed
209 positive correlation with PTEs, while OC is negatively correlated with PTEs. All PTEs
210 showed positive relationship with each other in industrial soils signifying that PTEs have
211 same source in all soil types. The positive correlation among PTEs is accredited to similar
212 type of sources accountable for PTEs concentration and anthropogenic activities such as
213 industrial activities like spring industry, iron rods factory, iron industry, cycle industry etc.,

and agricultural practices like usage of pesticides, fertilizers and herbicides are responsible for PTEs concentration (Kumar et al. 2019c; Keshavarzi & Kumar 2019a).

3.3. Principal component analysis and non-metric multidimensional scaling of potentially toxic elements in soils

Principal component analysis (PCA) was performed on the PTEs data to find source apportionment among PTEs. The first four components of PCA with Eigen values were found above one, accounts 72.9% of the variation. The loadings and loading plot of PCA are given in **Table 2** and **Figure 3.** The loadings of component matrix for PTEs revealed that PC1 is dominated by Mn, Ni, Co, Cd and As. PC2 is controlled by Fe and Cn. Zn contributes to PC3, while PC4 is influenced by Cu and Pb. After varimax rotation, PC1 is contributed by Mn, Co, Cd and As. Fe and Cn are dominated by PC2. PC3 is regulated by Pb and Ni. PC2 influenced by Zn and Cu. Industries are responsible for most of the HM pollution in Ludhiana district. The area is dominated by bicycle, woollen and hosiery industries. Further, the town is known for preparation of machine tools, dying work and electroplating (Setia et al. 2020).

The non-metric multidimensional scaling (NMDS) was also performed on the sampling sites on the basis of PTEs data. The results of NMDS scatter plot inferred that four points, i.e., 29, 40, 50 and 54 are unglued from the other sampling points (**Figure 4a**). In the sampling sites 29, 40 and 50 soils are collected adjacent to grassland, while 54 point is soil collected around the industrial zone. The stress level is less than the 0.05 (Kaur et al. 2018), indicating good fit of this data to NMDS plot (**Figure 4b**).

3.4. Appraisal of pollution level of potentially toxic elements in soils

The contamination factor (CF), enrichment factor (EF) and geoaccumulation index (Igeo) was enumerated to oversee the pollution level of PTEs in different soils (**Table 3**). On the basis of grades suggested by Hakanson (1980), the CF results for agricultural soils followed the trend as As > Cd > Pb > Ni > Co > Cu > Zn > Mn. Among PTEs, As and Cd

showed high contamination, while Pb exhibited substantial contamination. In non-agricultural soils the CF values of PTEs followed the trend as: As > Cd > Pb > Zn > Cu > Co > Ni > Mn. Amid PTEs, Cd and As exhibited high contamination, whereas Pb showed significant contamination. The Zn, Cu, Co, Ni and Mn showed low contamination in the study area. In industrial soils the CF values of different PTEs followed the trend as: As > Cd > Ni > Pb > Cu > Co > Zn > Mn. The As, Cd, Pb and Ni showed high contamination, whereas Cu, Co, Zn and Mn exhibited considerable to modest contamination in the area.

The grades given by Sutherland (2000) to categorise the pollution level on the basis of EF. The EF results of agricultural soils indicated that Cd, As, Ni and Pb exhibited extreme enrichment in the area, Co and Cu showed high enrichment, and Zn and Mn showed considerable enrichment of studied PTEs. In non-agricultural soils Cd, As, Co, Pb and Cu showed extreme, Zn and Ni showed high, and Mn exhibited considerable enrichment respectively. In industrial soils Cd, As, Co, Ni, Pb, Cu and Zn indicated extreme enrichment, while substantial enrichment showed by Mn.

Muller (1981) suggested grades to classify the pollution level on the basis of Igeo results. The Igeo results in agricultural soil indicated that Cd, As, Co, Ni, Pb, Cu and Zn showed extreme pollution, while Mn exhibited high pollution in the area. In non-agricultural and industrial soils all studied PTEs showed extreme pollution in the studied area.

Pollution index (PI) and modified pollution index (MPI) was also computed for different PTEs to find their pollution load (**Figure 5 A and B**). The grades suggested by Nemerow (1991) for PI and MPI and based upon that in agricultural soils Zn, Pb, Ni, Cd and As exhibited severe pollution, Cu and Co modest, and Mn showed slight pollution. In non-agricultural soils Cd, Pb and As showed severe pollution, Cu exhibited heavily, Ni and Co modest, and Zn and Mn showed slight pollution. In industrial soils, PI values of Cd, As, Ni, Pb and Cu showed severe pollution, Co and Zn showed modest, and Mn exhibited slight

pollution in the studied area. The MPI values for all studied PTEs in agricultural, non-agricultural and industrial soils were found above 10, signifying severe pollution of these PTEs in the area.

3.5. Ecological risk appraisal of potentially toxic elements in soils

The ecological risks were enumerated by potential ecological risk (RI) and modified potential ecological risk (MRI) for different PTEs in soils (**Table 4**). The potential ecological risk (Er) value of Cd in agricultural soils was recorded above 320, signifying very risk of this metal in the agricultural soils of study area. Er value of As showed high risk, whereas Zn, Mn, Cu, Pb, Ni and Co showed low risk in the area. In non-agricultural soils, Cd exhibited very high risk, As showed high risk, and Zn, Mn, Cu, Pb, Ni and Co exhibited low risk. In industrial soils, Cd and As showed very high risk, Ni reflected modest, and Zn, Mn, Cu, Pb and Co showed low risk in the study area. On the basis of grades used by Kumar et al. (2018) for classification of ecological risk and on the basis of these grades it was inferred that Mn, Zn and Co showed low ecological peril for all samples. Cu showed low to modest ecological risk in the area. The modified potential ecological risk (mEr) of Cd, As and Pb in agricultural soils indicated very high risk, Cu, Ni and Co showed high risk, and Zn and Mn exhibited low risk. In non-agricultural soils, Cd, As, Pb and Cu showed very high risk, Zn and Mn exhibited low risk, and Ni and Co showed high risk in the area. In industrial soils, Cd, As, Ni and Pb showed very high risk, Zn and Mn displayed low risk, and Cu and Co showed high risk in the area. The results of ecological risk indicated inferred that Cd is the vital pollutant in the area.Our results on ecological risk appraisal are in corroboration with Pandit et al. (2020) and Kumar et al. (2018). They also reported in their studies that Cd is the main contaminant responsible for polluting the soils. On the basis of RI and MRI values the trend followed by different soils are as follows: industrial soil > non-agricultural soil > agricultural soil.

289 **4. Conclusions**

290 The present study concluded that industrial soils recorded highest concentration of
291 potentially toxic elements in contrast with non-agricultural and agricultural soils. After
292 comparison of industrial soils with Indian background values and Poland soil limits, it was
293 revealed that 100% samples were surpassed their values for Zn, Mn, Pb, Ni, Co and As, while
294 80% for Cu, and 60% for Cd. 97.5%, 82.5%, 47.5%, 100% and 42.5% sampling sites
295 surpassed their contents for Zn, Mn, Ni, Pb and Co in comparison with their limits in
296 agricultural soils. Similarly, in non-agricultural soils 100% sampling sites surpassed their
297 values for Zn, Mn, Pb, Ni, Co and As in contrast with their limits. Pearson's correlation
298 analysis indicated that PTEs have same source of origin and mainly industrial activities in the
299 area contribute PTEs level in the soil. The results of CF, EF, Igeo, PI, MPI, RI and MRI
300 showed that As and Cd are the main pollutants in the soils of study area.

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308 Availability of data and materials: The datasets used and/or analysed during the current study
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Author Contribution:

The paper was conceived and designed by JS and CP. The sampling of soil and its analysis was done by FV and SS. VK and RK analyzed data and proofread the manuscript. SSD assisted in the analysis of the soil sample. The critical revisions and the final version were done by JS and CP. All authors have read and approved the final manuscript.

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Figure Captions

Figure 1 Location of study area along with different sampling sites.

Figure 2 Pearson's correlation analysis of potentially toxic elements in: (A) agricultural soils, (B) non-agricultural soils, and (C) industrial soils.

Figure 3 Loading plot of PCA showing different soil variables

Figure 4 (a) NMDS scatter plot (95% eclipse) of sampling sites on the basis of potentially toxic elements using correlation as similarity measure and (b) NMDS Shepard 2-D plot of sampling sites**,** stress = 0.03518, R2 for axis 1 = 0.6932 and axis 2 = 0.4492.

Figure 5 Pollution index (A) and modified pollution index (B) of different potentially toxic elements in soils

Table Captions

Table 1 Statistical analysis of pH, OC and potentially toxic elements (PTEs) in soils collected from study

Table 2 Principal component analysis of potentially toxic elements

Table 3 CF, EF and Igeo indices of potentially toxic elements

Table 4 Ecological risk assessment of potentially toxic elements in soils collected from study area

Supplementary Tables

Table S1 The study sites under agriculture, non-agriculture and industrial sites

Table S2 Soil grades for the contamination factor and enrichment

Table S3 Soil grades of pollution and modified pollution index

Table S4 Categories of (Er) and potential ecological index

Figures

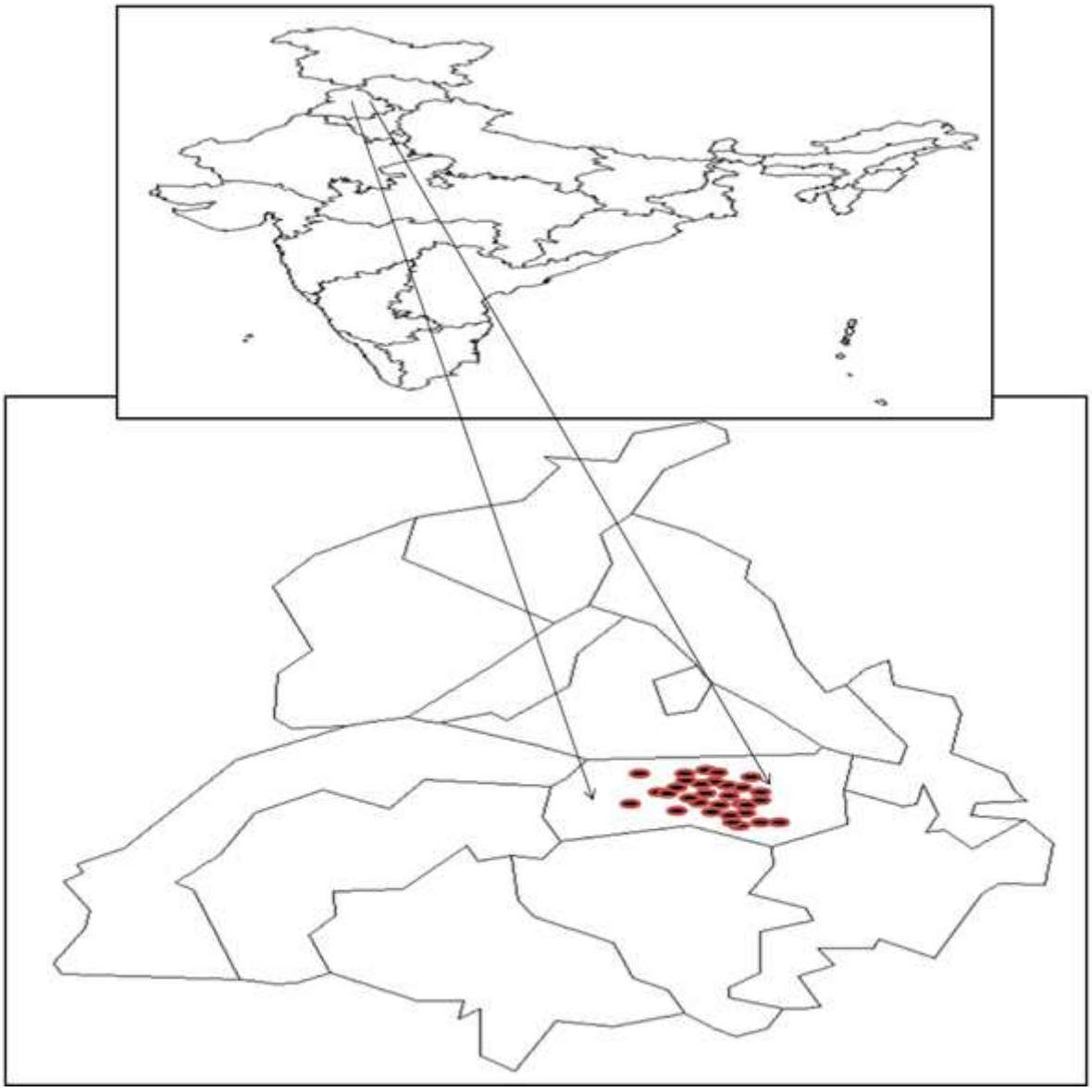

Figure 1

Location of study area along with different sampling sites. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

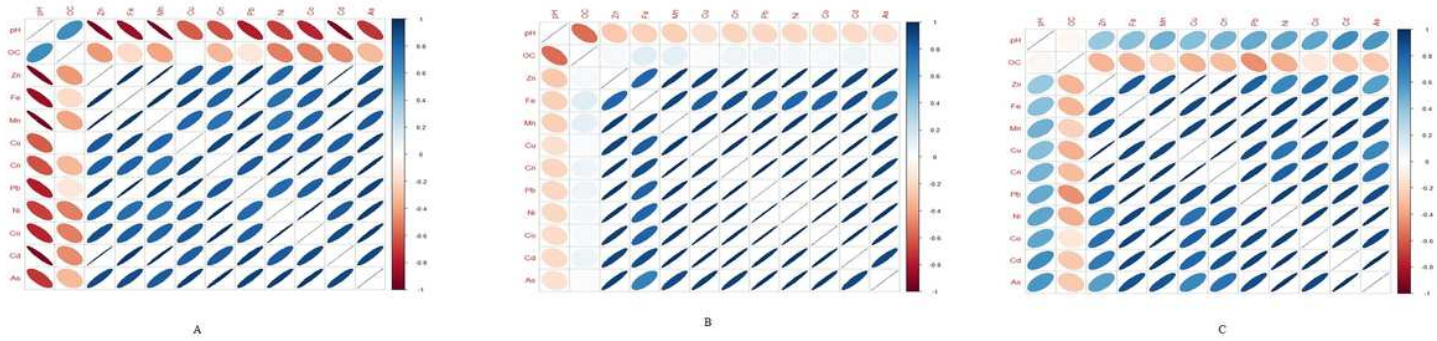

Figure 2

Pearson's correlation analysis of potentially toxic elements in: (A) agricultural soils, (B) non-agricultural soils, and (C) industrial soils.

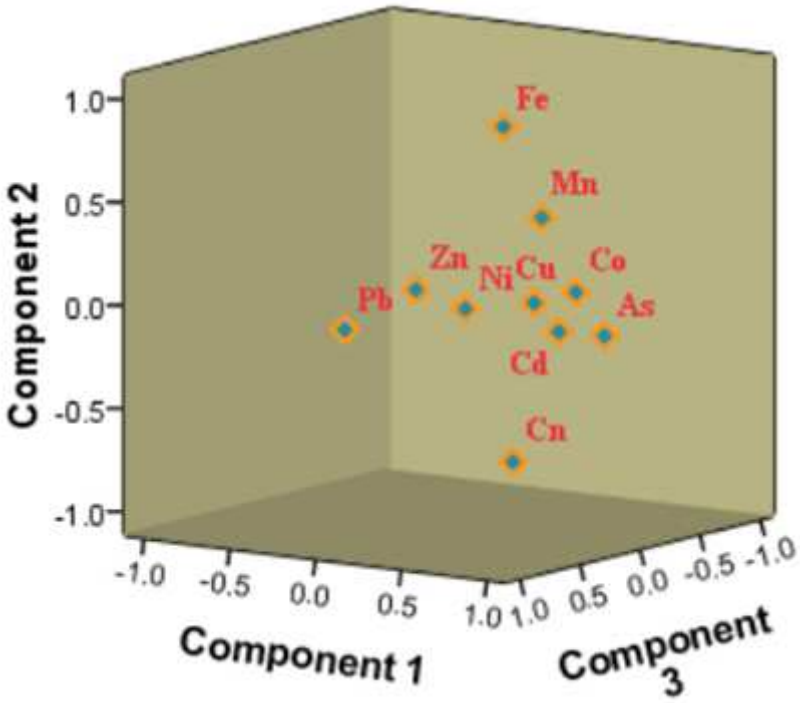

Figure 3

Loading plot of PCA showing different soil variables

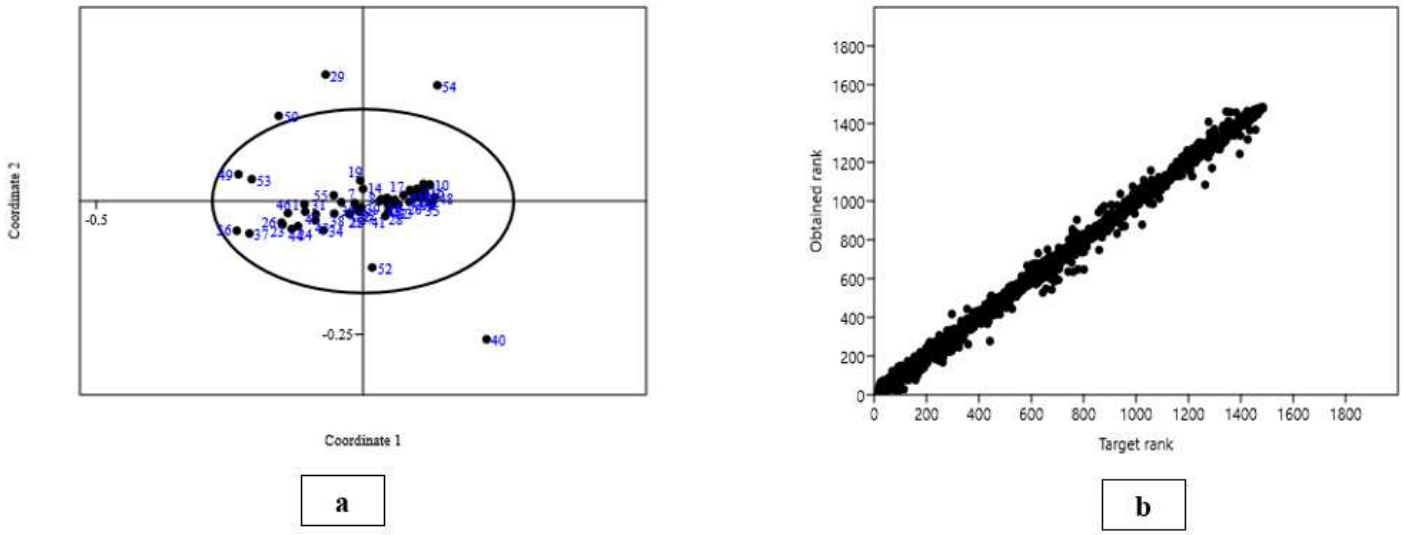

Figure 4

(a) NMDS scatter plot (95% eclipse) of sampling sites on the basis of potentially toxic elements using correlation as similarity measure and (b) NMDS Shepard 2-D plot of sampling sites, stress = 0.03518, R2 for axis 1 = 0.6932 and axis 2 = 0.4492.

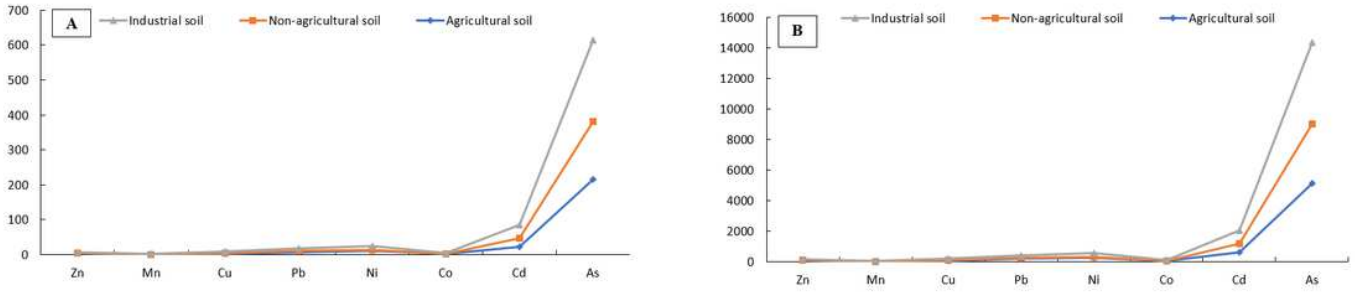

Figure 5

Pollution index (A) and modified pollution index (B) of different potentially toxic elements in soils

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- Table3.docx
- Table4.docx
- Table1.docx

- Table2.docx
- Supplementarytables.docx