

Distribution and Statistical Source Identification of Heavy Metals and Radionuclides in Soil: A Case Study from a Proposed Uranium Mining Site, Jharkhand, India



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Abstract Singhbhum Thrust Belt is well recognized for its uranium and copper deposits and thus radionuclides as well as metals can be anticipated in its environs inclusive of soil. The enrichment of metals and radionuclides in the vicinity of the uranium mining areas can be attributed to their release due to mining activities and waste disposal. As a part of the baseline study of the Bagjata mining area, 10 soil samples were collected from the villages in proximity of the mining area and analyzed for radionuclides (U(nat), ^{226}Ra , ^{230}Th , ^{210}Po) and metals (Fe, Mn, Zn, Pb, Cu, and Ni). The geometric mean activity of ^{226}Ra , ^{230}Th and ^{210}Po was 23.24, 65.54 and 55.15 Bqkg^{-1} , respectively while geometric mean concentration of U (nat), Fe, Mn, Zn, Pb, Cu and Ni was calculated to be 2.37, 26,106, 267.6, 53.2, 37, 159.6 and 141.4 mgkg^{-1} , respectively. Cu and Ni surpassed the Maximum Allowable limits (MAL) in the majority of locations. Owing to uneven and sporadic mineralization of the region, widespread spatial variation in the heavy metals and radionuclides has been observed. The Principal Component Analysis (PCA) depicted the geogenic contribution as the main source of radionuclides and metals. The secondary sources of metals were illustrated to be anthropogenic sources like mining and related vehicular load. The association of the radionuclides with the Fe is also depicted from the study.

Keywords Radionuclides · Heavy metals · Soil · Principal component analysis · Uranium mining

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1 Introduction

In recent years, owing to rapid industrial growth, one of the major environmental problems that have surfaced is land degradation. This is further aggravated by the immense increase in the mining activities. As in case of other mining activities such as metal mining or coal mining, uranium mining can also be apprehended to pose risk to the adjoining environment due to the release of enormous amount of waste rich in metals and radioactive elements. However, the extent of the influence is dependent on a number of factors such as the method and quantity of uranium excavation, infrastructure for the treatment of uranium, facility of waste disposal etc. [1, 2].

In the contaminant transport system, soil-plants-food-man, the soil forms the first line of contamination and thus it is important to know activity of the soil. The background radiation in soil is extensively contributed by the gamma radiations from U and Th series elements and radioactive K [3]. A significant amount of radiation exposure to the human population is contributed by these natural radionuclides of the soil [4, 5].

The transport and buildup of the pollutants in the soil involves intricate with processes like absorption, adsorption, leaching, plant uptake, runoff, etc. [6, 7]. Small amount of U and Th with their daughter radionuclides are naturally present in soil however, their levels are dependent on the lithology of the area and the type of the rocks which forms the soil [8].

The multivariate statistical tools such as principal component analysis (PCA) help in source apportionment of the contaminants with data reduction and identification of the factors which explains the most of the data variance. The factors are extracted in such a manner so as to protect the associations in the original data [9–11].

In several parts of eastern Singhbhum uranium mining have started subsequent to the discovery of low-grade uranium deposits in the region. With the increase in demand of nuclear energy, new mining sites have been excavated for uranium. One such underground uranium mine site was at Bagjata. Mining activities indiscriminately affect the environmental status of the mining and adjoining areas and thus baseline studies are important to ascertain the pre-existing environmental condition [12]. The study presented here was a part of baseline study conducted for the Bagjata mining area of which soil was also a component. The soils from the mining and surrounding areas were studied for their content of radionuclides and heavy metals.

2 Materials and Methods

2.1 Study Area

Singhbhum Thrust Belt (STB) is known worldwide for its rich deposits of copper, apatite, magnetite and kyanite. STB is an EW trending belt of approximately 160 km

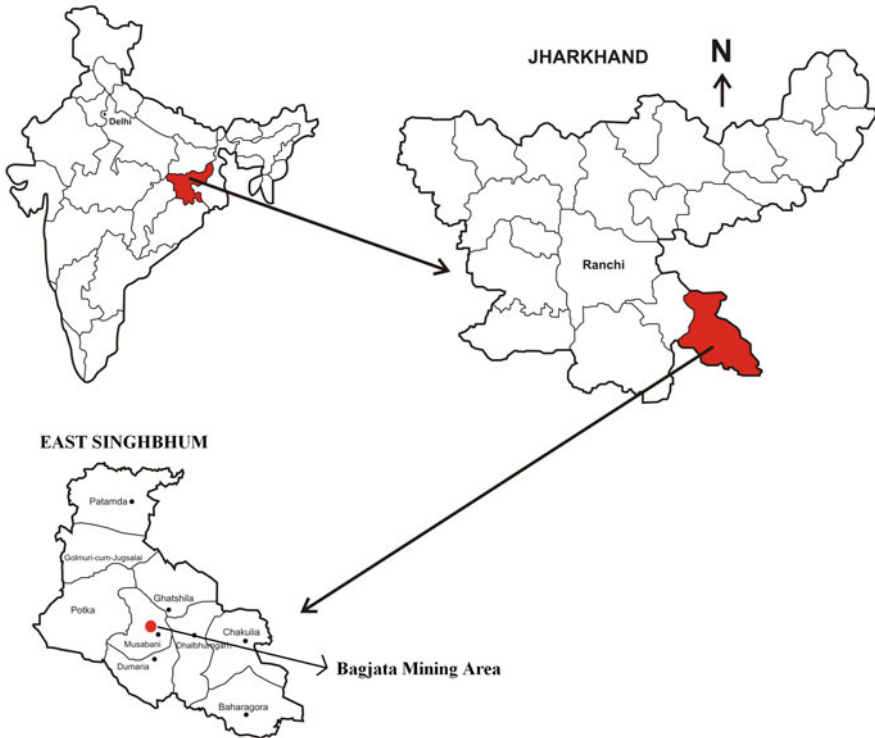
POLITICAL MAP OF INDIA

Fig. 1 Locations detail of the study area (adapted from [12])

length in which the mineralization occurred in 2 phases. The first phase was the deposition of apatite and magnetite and subsequently uranium. The second phase included the deposition of sulphides including chalcopyrites. U-Cu mineralization coexists in the area besides other minerals of Fe, Mn, etc. The study area under consideration i.e. Bagjata mining area is situated in the Bhalki-Kanyaluka deposit [13] and has latitude of $22^{\circ}26'07''\text{N}$ to $22^{\circ}28'34''\text{N}$ and longitude of $86^{\circ}25'16''\text{E}$ to $86^{\circ}31'29''\text{E}$. The key minerals of U of the region consist of uraninite and pitchblende along with autonite as the secondary mineral. Sulphides of various metals like Cu, Ni, Co, Bi, As, etc. are also associated to the U minerals [14].

Figure 1 depicts the location map of the study area.

2.2 Sample Collection and Laboratory Analysis

The soil samples were collected from approximately 0–20 cm below the ground surface. The grassroots and other debris which were present in the soil were removed

firstly. Then the samples were mixed homogenously using coning and quartering technique. Subsequently 100 g of soil was taken and oven dried for 24 hours at 110 °C in porcelain dishes. The soil samples were further pulverized using mortar-pestle and sieved through of 100 mesh size. After this the samples were acid digested in hotplate using HNO₃ [15]. Subsequent to the elimination of organic matter from the samples, repeated leaching was done using 8N HNO₃. The samples were then filtered using Whatmann filter paper and the final volume was made up to 100 ml using double distilled water. Further, the aliquots were preserved after proper labeling for the detection of heavy metals and radionuclides.

Electrochemical exchange method followed by alpha counting was used for evaluation of ²¹⁰Po [16]. Determination of U(nat) and ²²⁶Ra was done using fluorimetric and radon emanation techniques [17, 18], respectively. ²³⁰Th was analyzed by screening it by use of anion exchange resin and subsequent alpha counting [19]. The evaluation of heavy metals was done with the help of Atomic Absorption Spectrophotometer (GBC Avanta).

2.3 Analysis of the Data

The data distribution of radionuclides and metals for the soil samples were confirmed by the cumulative frequency curves [20]. The concentrations were found to follow log-normal distribution which is quite common for the environmental samples. Thus the central tendency was characterized by geometric mean [21].

2.4 Principal Component Analysis (PCA)

Principal component analysis (PCA) is a statistical source apportionment tool which replaces a larger number of interrelated variables with smaller number of independent variables called principal components (PCs). In this method, the covariance matrix of original variables is used to obtain the eigenvalues and eigenvectors [22]. The PCs are weighted linear groupings of original variables that give idea about the most dominant factors and in the process there is least loss of original information [23–26]. Further reduction of the effect of the less important variables is achieved by factor analysis which is done by extracting the varifactors obtained by rotation of axis as defined by PCA [27, 28].

2.5 *Enrichment Factor (EF) and Geo-accumulation Index (I_{geo})*

For the assessment of anthropogenic influence on the metal concentration of the soil of the study area enrichment factors (EF) were evaluated by the equation:

$$EF = (C_X/C_{Fe})_s / (C_X/C_{Fe})_c$$

Where, C_X and C_{Fe} refer to the concentration of metal X and Fe in the soil (s) and earth's crust (c), respectively. Iron is taken as the reference metal.

The degree of metal pollution in the soil of the study area was quantitatively evaluated using the Index of Geoaccumulation (I_{geo}) [29] as:

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

Where, C_n is the measured concentration of the heavy metal 'n' in the soil, and B_n is the geochemical background value in average shale [30].

3 Results and Discussion

3.1 Radionuclides

Table 1 depicts the descriptive statistics of the analytical results of radionuclides and heavy metal in the soil samples of the study area and Table 2 provides the average concentration of the radionuclides in the soil with respect to sampling locations. It can be observed from the Tables that Uranium varied from 0.46 to 12.73 mg kg⁻¹, the highest being at Bagjata village. The activities of ²²⁶Ra, ²³⁰Th, and ²¹⁰Po vary from 4.1 to 145, 14.3 to 407.1 and 7.8 to 172.2 Bqkg⁻¹, respectively. The geometric

Table 1 Descriptive statistics of the activity of radionuclides and concentration of heavy metals in soil samples (unit: ²²⁶Ra, ²³⁰Th, ²¹⁰Po in Bqkg⁻¹; all other parameters in mgkg⁻¹)

Parameters	N	Minimum	Maximum	Geometric mean
U(nat)	40	0.46	12.73	2.37
²²⁶ Ra	40	4.1	145.0	23.24
²³⁰ Th	40	14.29	407.14	65.64
²¹⁰ Po	40	7.78	172.22	55.15
Fe	40	13,132	64,798	26,106
Mn	40	104	586	267.6
Zn	40	26	115.1	53.2
Pb	40	16.6	81.0	37.0
Cu	40	70.1	392.2	159.6
Ni	40	64.0	296.0	141.4

Table 2 Average activity/concentration of radionuclides in soil samples with respect to different locations (unit: U (nat) in mgkg^{-1} ; other parameters in Bqkg^{-1})

Code	Location	U(nat)	^{226}Ra	^{230}Th	^{210}Po
1	Bagjata	11.0	74.2	204.2	104.8
2	Bhaduya	2.3	20.6	110.0	100.5
3	Phuljhari	3.9	16.0	54.0	89.5
4	Manajhari	0.9	9.0	66.3	48.8
5	Balidungri	1.3	16.0	52.8	25.2
6	Bakra	3.3	69.5	43.1	50.6
7	Katsakra	1.9	4.7	74.8	31.8
8	Gohala	1.5	23.4	62.7	59.4
9	Nimdih	7.4	66.4	52.7	128.7
10	Surda	1.8	16.1	61.0	39.6

mean concentration of U(nat) was calculated to be 2.37 mgkg^{-1} while the geometric mean activity of ^{226}Ra , ^{230}Th and ^{210}Po were analyzed to be 23.24, 65.54 and 55.15 Bqkg^{-1} , respectively. The highest activity of all the considered four radionuclides was found in the same location i.e. Bagjata village. This may be attributed to the location being in close proximity to the mining site.

An uneven distribution of the specific activity of the considered radionuclides in the soil is observed in the study area. There is widespread uranium mineralization in this area, and accordingly wide variation in the concentration of radionuclides is expected [31]. This may be due to the intense weathering of the regolith following precipitation. The chemical characteristics of various radionuclides are different and so the extent of leaching. Apart from this, organic complexation of uranium and adsorption of polonium over ferric hydroxides may also lead to disequilibrium between the radionuclides of uranium series.

3.2 Heavy Metals

Owing to uneven mineralization, widespread spatial variation in the heavy metals has been observed. Table 1 depicts the descriptive statistics of heavy metal concentrations in the soil samples of the study area while the comprehensive results are presented in Tables 3. Taking into consideration all the four sampling episodes, the concentration of Fe was found in the range of 13,132 to 64,798 mgkg^{-1} ; with the geometric mean of 26,106 mgkg^{-1} . The concentration of Mn and Zn varied from 104 to 586 and 26 to 115.1 mgkg^{-1} , respectively. The geometric mean concentration taking into account all the locations and all the sampling periods for Zn was found to be 53.2 mg kg^{-1} . The Pb concentration was evaluated to be in between 16.6 to 81.0 mgkg^{-1} . None of the locations exceeded the Maximum Allowable limits for Zn and Pb of 300 and 100 mgkg^{-1} , respectively as stated by Kloke [32].

However, both Cu and Ni surpassed the Maximum Allowable limits given by Kloke [32] i.e. 100 and 50 mgkg^{-1} for Cu and Ni, respectively. The area falls in the uranium-copper-nickel deposits of Singhbhum Thrust Belt, with extensive mining of

Table 3 Average concentration of heavy metals in soil samples for different locations (Unit: mgkg⁻¹)

Location	Fe	Mn	Zn	Pb	Cu	Ni
Bagjata	52,531	482.8	50.2	64.5	323.5	256.0
Bhaduya	22,467	274.2	64.9	41.8	91.3	131.8
Phuljhari	31,947	393.5	38.0	49.4	122.3	100.0
Manajhari	18,267	171.9	57.5	25.4	145.0	131.1
Balidungri	30,177	309.0	95.9	45.4	194.0	91.3
Bakra	22,818	311.0	85.0	46.8	198.3	176.0
Katsakra	21,478	251.0	39.4	28.7	150.1	135.4
Gohala	21,281	393.8	40.0	26.8	231.2	141.3
Nimdih	42,356	189.7	55.8	33.8	112.6	177.7
Surda	20,784	145.8	47.8	45.6	188.7	162.5

Table 4 Principal Component Analysis (PCA) for the elements in the soil of the study area (n = 40)

Elements	PC1	PC2	PC3
U(nat)	0.935	-0.117	-0.002
²²⁶ Ra	0.691	0.376	0.002
²³⁰ Th	0.573	-0.416	-0.008
²¹⁰ Po	0.695	0.002	-0.488
Fe	0.855	-0.006	0.276
Mn	0.588	-0.009	0.048
Zn	0.005	0.831	0.237
Pb	0.191	0.675	0.437
Cu	-0.114	0.108	0.615
Ni	-0.126	-0.006	0.877
Eigen values	2.99	2.36	1.48
% Total variance	29.7	20.0	18.6
Cumulative variance	29.7	49.7	68.3

Cu. This may be attributed for the widespread spatial variation and elevated concentration of Cu and Ni in the soil samples of the study area. The concentration of Cu and Ni varied between 70.1 to 392.2 and 64 to 296 mg kg⁻¹, respectively. The level of Cu and Ni surpassed the Indian limits for soil [33] also at some of the locations. The highest concentration of Cu and Ni were found in the Bagjata village.

This is an obvious observation that the maximum concentration of heavy metals and radionuclides was found in the same location (Bagjata village) which is in very close proximity to Bagjata mining site. Though the concentration of metals and radionuclides were found to be highest in the same location no definite trend could be established in the spatial variation with respect to distance from the site.

3.3 Principal Component Analysis (PCA)

The results of PCA which is a statistical source apportionment tool based on inter-element correlations are presented in Table 4. The PCs obtained corresponds to

cluster of metals that are linked together and so they may be associated with a common source and similar behavior [34]. The PCA of soil of Bagjata area depicts that 3 PCs obtained after varimax rotation explain the 68.3% of data variance. The number of principal components having eigenvalue greater than 1 are selected which is based on the Kaiser criterion [35, 36].

The first factor explaining 29.7% of variance and having high loading for U(nat), ^{226}Ra , Th(α), ^{210}Po , Fe and Mn appears to be linked to the geological setting of the region. The factor may also be related to the linkage of Fe with other radionuclides since it is a well known fact that iron oxides and hydroxides adsorb radionuclides inclusive of Uranium, Radium and Thorium [37–40]. The second component (PC2) depicted a strong association of Pb and Zn justifying 20% of variance and may be linked to vehicular pollution particularly from the mining related vehicles. Pb is known to be associated to vehicle related emissions [41]. Zn is used in the tires of the vehicles as vulcanization agent [42] which has strong probability of getting weared out due to high temperature of the region and thus contribute in increase in Zn concentration of the soil. The third factor (PC3) explains 18.6% of variance and is emphasized by the metals Cu and Ni. This factor can be allied to the widespread Copper mining activities of the region.

3.4 Enrichment Factor and Geo-accumulation Index

The Enrichment factor calculated for the metals of soil of the study area depicted moderate contamination ($\text{EF} = 2$ to 5) [43, 44] for Zn, Pb and Ni and significant contamination ($\text{EF} = 5$ to 20) [43, 44] with respect to Cu (Table 5). This may be attributed to the anthropogenic activities i.e. copper mining and vehicular pollution of the area which is also depicted from the PCA results.

The Geoaccumulation index (I_{geo}) values were calculated and classified as per Muller [29] (Table 6). The I_{geo} values suggested an unpolluted status (Class 0) of the soil with respect to Fe, Mn, and Zn. Ni and Pb falls in Class 1 indicating an unpolluted to moderately polluted status in the soil. However, the I_{geo} values for Cu were found to be in between 1 and 2 (Class 2), thus suggesting a moderate pollution with respect to Cu in the soil samples. This is in accordance to the calculated enrichment factors.

Table 5 Enrichment Factor of elements in the study area

	Fe	Mn	Zn	Pb	Cu	Ni
Average background shale [30]	47,200	850	95	20	45	68
Geometric mean	26,106	267.7	53.2	37	159.6	141.4
Enrichment Factor	1	0.569	1.012	3.345	6.412	3.760

Table 6 Geo-accumulation Index of Elements in the study area

I_{geo} Value	I_{geo} Class [29]	Sediment Quality (Muller [29])	Element
>5	6	Very highly polluted	
4–5	5	Highly polluted to very highly polluted	
3–4	4	Highly polluted	
2–3	3	Moderately polluted to highly polluted	
1–2	2	Moderately polluted	Cu
0–1	1	Unpolluted to moderately polluted	Pb, Ni
<0	0	Unpolluted	Fe, Mn, Zn

4 Conclusions

The present study which was a part of the baseline study of the Bagjata mining areas revealed a natural background concentration with respect to radionuclides in the soil. Amongst the studied metals, Cu and Ni surpassed the Maximum Allowable limits for the majority of the locations. The radionuclides and metals depicted an uneven distribution in the area which may be accredited to the widespread U-Cu mineralization in the region. The PCA suggested that the major source of radionuclides and metals in the soil is geogenic; however, some anthropogenic sources like mining and vehicular load also contribute to the metal content in the soil. The association of the radionuclides with Fe is also illustrated from the study. Moderate pollution with respect to Cu in the soil of the area is also depicted from the study. The study holds importance owing to the extensive industrial and mining activities in the region.

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Recommendations

- i. Regular monitoring of soil samples in the environs of the mining areas is recommended so as to know the post mining effect with respect to radionuclide and metals.
- ii. The content of radionuclides and metals in the agricultural produces should be done since there is a strong probability for the pollutants to get into the food chain through the soil
- iii. Human health risk assessment is suggested to be carried out owing to the exposure related to radionuclides and metals in the soil through the ingestion, dermal and inhalation pathways.

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