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Assessment of Concentrations and Distributions of Natural Radionuclides in Soils of Abandoned Mine Sites of Jos South and Barkin Ladi in Plateau State Nigeria

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Barkin Ladi and Jos South Local Government Area of Plateau State is situated in the central part of Northern Nigeria, on a rugged terrain of lowlands. It is the principal centre of tin and columbite mineralization, which forms the focal area of younger granites. The work was aimed at assessing the concentration and distribution of radionuclides in soils of abandoned tin mines and the soil's physicochemical properties. The samples were collected in July, from five different locations in both Barkin Ladi and Jos South where mining activities were predominant. The samples were analyzed using a gamma-ray spectrometer. The result obtained reveals a range of the concentrations of ^{40}K

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as 116.76 ± 0.21 to 250.15 ±0.,42 Bq/kg, ^{226}Ra from 24.37 ± 0.31 to 90.45 ± 0.86 Bq/kg, and ^{232}Th from 15.74 ± 0.71 to 88.43 ± 0.14 Bq/kg. The result of the physicochemical properties of the soil analyzed ranges from pH (3.12 to 6.81); organic carbon (0.01 to 1.62%); organic matter (0.22 to 3.01 %), cation exchange capacity (1.01 to 4,21Cmol/kg); clay (9.72 to 40.82 %); silt (8.32 to 40.32% and sand (32.42 to 75.38%) respectively. All the ^{40}K results analyzed in all the sampling sites were lower than the recommended limits of 412Bq/kg, whereas most of the results of ^{226}Ra and ^{232}Th were above the 33Bq/kg and 45Bq/kg recommended safe limits. Exposure to elevated levels of naturally occurring radionuclides in soil can pose potential health risks to nearby populations. The physicochemical properties of the soil varied across the sampling sites. Most of the sampling sites are polluted with radionuclides and migrations of the radionuclides from the ex-mining sites to other parts of the environment were noticed from the studies conducted.

Keywords: Mining point; pollution; radionuclides; soil; and tailings; physicochemical properties; radioactivity; global radiation.

1. INTRODUCTION

1.1 Background of the Study

Abandoned tin mines can potentially increase soil radioactivity due to the presence of naturally occurring radionuclides like thorium, uranium, and rare earth elements. The majority of environmental matrices contain radioactive elements including ^{232}Th and ^{238}U, as well as their decay products like ^{226}Ra. These elements can enter living things through a variety of paths, which can lead to the sources of human exposure [1].

Ever since life originally began in the universe, naturally occurring radioactive elements have exposed species including plants and animals to radiation [2]. There are more than 60 radionuclides in nature because of how radioactive our planet is [3]. The sources of radioactivity and the emitters of nuclear radiation that permeate our daily lives are these radionuclides. Everywhere there is radiation, humans are continually exposed to it, either directly or indirectly. Humans are exposed to radiation from a wide range of natural and artificial sources. Ionizing radiation comes from a variety of sources, including mining sites, water, air, soils, and construction materials [3]. We breathe in or absorb radionuclides from the food we eat, and in the water we drink.

Populations in communities may be exposed to radiation from both natural and man-made sources. Both terrestrial and extraterrestrial (space) sources can contribute to the natural radiation that surrounds life on Earth. Ionizing radiation from radionuclides in the earth, rocks, building materials, water, air, and mining sites is referred to as global radiation. Conversely, cosmic rays are high-energy radiation from space that reaches the Earth's atmosphere [4]. Radiation-sensitive elements like 238U, ^{232}Th, and the daughter products ^{226}Ra, According to [5], ^{222}Rn, ^{235}U, and 40K are significant naturally occurring radiation sources. Ionizing radiation, including α, β, and γ radiations, is released from many terrestrial materials. Among them, soil is a primary source of naturally occurring radioactivity and the primary pathway for radionuclide migration and transfer into the surrounding environment [6].

Numerous investigations have been conducted globally to ascertain the activity content of these radioactive elements, which are the primary causes of natural radiation in soil that humans have contaminated [7]. Geographical location and some human activities influence the extent of these natural exposures [8]. In addition to radiation from natural sources, radiation from man-made sources can also affect humans. Radiation applications may be found in many fields, including biology, agriculture, industry, medicine, and the production of electric power. Humans may come into contact with radiation from various radioactive sources during their uses, as well as radiation-related illnesses [9]. This study aims to assess the concentration and spatial distribution of radionuclides in soils of abandoned tin mines in Barkin Ladi and Jos South Local Government Area of Plateau State, Nigeria. Understanding the levels and distribution of these radionuclides is crucial for evaluating potential health risks to nearby communities."

2. MATERIALS AND METHODS

2.1 Materials

2.1.1 Reagent/ solvents

The reagents that were used for the research work are 1N Potassium dichromate ($K_2Cr_2O_7$), sodium fluoride, 0.5N ferrous ammonium sulfate $Fe(NH_4)_2(SO_4)_2.6H_2O$, Diphenylamine, Tetraoxosulphate (VI) acid 0.1M $BaCl_2.2H_2O$, and 0.1M $MgSO_4.7H_2O$.

2.1.2 Instruments/equipment

The materials that were used for the research work are Canberra Model 727/727R Lead Shield Gamma-ray Spectrometer with NaI(TI) detector, Oven Gallenham England, Hanna pH meter 209 Romania, Labtect digital conductivity meter USA, Beakers, Mortar and pestle, volumetric flask.

2.2 Area of Study

Barkin Ladi and Jos South Local Government Area of Plateau State are situated in the central part of Northern Nigeria, on a rugged terrain of lowlands. Five samples were collected each from Barkin Ladi and Jos South, Local Government Area. These areas are the principal centres of tin and columbite mineralization, which forms the focal area of younger granites. The mine tailings are associated with radioactive minerals as impurities such as monazite and zircon among others. These minerals are radioactive and can cause serious hazards to human health during mining and land cultivation for crop production.

2.3 Sample Collection

2.3.1 Soil sample

At each sampling point, about 0.50 kg of the soil sample was collected from a depth of between 0-20 cm from the surface of the soil, using a clean auger at a distance of 1m away from each other, and within an area of one square meter.

2.4 Sample Analysis

A gamma ray Spectrometer with NaI(TI) detector, was used to analyze the soil samples for the presence of ^{226}Ra, ^{232}Th, and ^{40}K. The Soil samples were collected into a very clean polythene bag and well-labeled to avoid mixed up of samples. The samples were transported to CERT, Ahmadu Bello University, Zaria. The samples were dried at ambient temperature until there was no detectable change in the mass of the sample. The dried samples were thoroughly crushed, grounded and pulverized to powder. The powder was passed through a 2mm sieve. Due to the limited space of the detector shield, only 200g - 300g of the samples (dry– weight) were used for analysis.

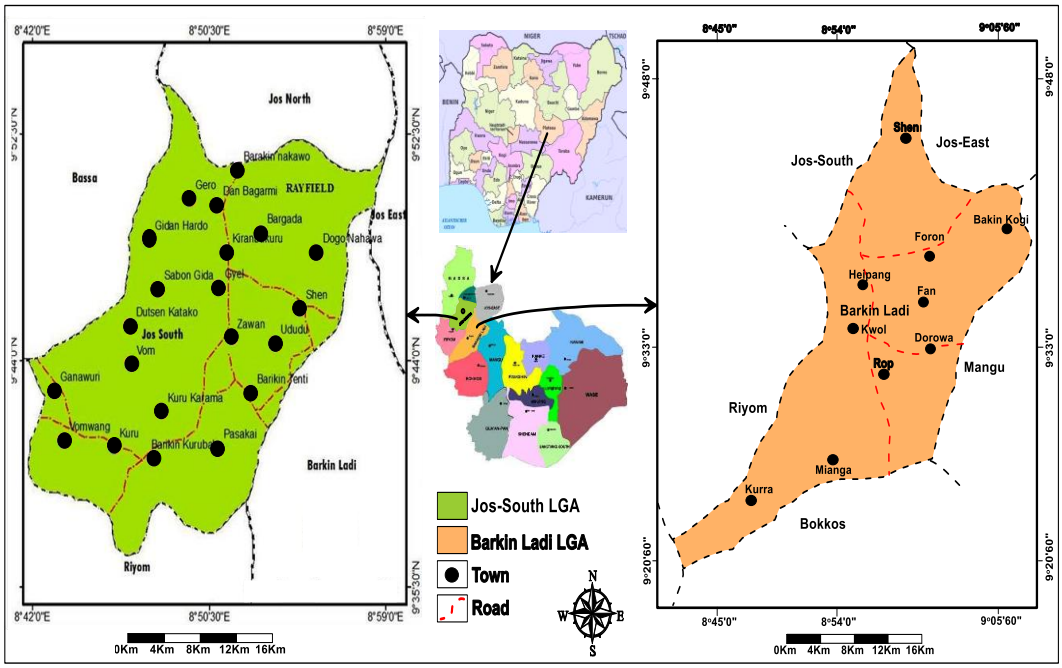

Fig. 1. Map of Barkin Ladi and Jos South Local Government Area

To prevent ^{222}Radon from escaping, the packaging in each case was triple sealed. The samples after weighing were transferred to radon-impermeable cylindrical plastic containers of uniform size (70mm height by 60mm diameter) and were sealed for about 30 days. This was done to allow Radon and its short-lived progenies to reach secular radioactive equilibrium prior to gamma spectroscopy. The reference soil was also transferred to a container of the same material and dimensions as were used for the plant samples. A lead-shielded 76 × 76 mm NaI(TI) detector crystal (Model No. 727 series, Canberra Inc.) that is coupled to a Canberra Series 10 plus Multichannel Analyzer (MCA) (Model No.1104) through a preamplifier was used for the radioactivity measurements. It has a resolution (FWHM) of about 8% at the energy of 662.0KeV (^{137}Cs) which is considered adequate to distinguish the gamma ray energies of interest in the present study. The choice of gamma-ray peaks of the radionuclides to be used for measurements was made because the NaI(Tl) detector used in this study had a modest energy resolution. This was to ensure that the photons emitted by the radionuclides would only be sufficiently discriminated if their emission probability and their energy were high enough, and the surrounding background continuum low enough. Therefore, the activity concentration of ^{214}Bi (determined from its 1760 KeV - ray peak) was chosen to provide an estimate of ^{226}Ra (^{238}U) in the samples, while that of the daughter radionuclide ^{208}Ti (determined from its 2615 KeV -ray peak) was chosen as an indicator of ^{228}Th (^{232}Th). Potassium-40 was determined by measuring the 1460 KeV -rays emitted during its decay.

The soil samples were placed symmetrically on top of the detector and measured for a period of 29000 seconds. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks.

2.4.1 Calibration and efficiency determinations

Calibration of the system for energy and efficiency was done with two calibration point sources, ^{137}Cs and ^{60}Co. These were done with the amplifier gain that gives 72% energy resolution for the 661.7Kev of Cs-137 and counted for 30 minutes.

Table of Spectral energy windows used in the Analysis

Isotope	Gamma Energy (Kev)	Energy Window(Kev)
R-226	1764.0	1620-1820
Th-232	2614.5	2480-2820
K- 40	1460.0	1380-1550

CEC was determined using the method described by Gillman and Sumpter (1986), OC was analysed using the method described by Walkley – Black revised in 2018 while the pH was determined using the methods described by Abubakar et al. (2015).

3. RESULTS AND DISCUSSION

3.1 Results

NB: 1 stands for samples collected directly from the mining spot, 2 stands for samples collected 200 m away from the mining spots, and 3 stands for samples collected 400 m away from the mining spots.

The activity concentration of ^{40}K, ^{226}Ra, and ^{232}Th expressed in Bq/Kg for the samples obtained from mining areas of Foron and Heipang district of Barkin Ladi Local Government Area and Kuru and Zawan district of Jos South Local Government area of Plateau State are presented in Table 1. The samples were taken during the second week of July 2021 at the various sites listed in Table 1. A total of thirty (30) samples were taken from various sampling points. From each of the settlements, three samples were taken. The samples were taken from the point source, 200 meters and 400 meters away, respectively. This was carried out to evaluate the distribution and fate of the natural radionuclides in the environment of the mining locations. The radioactive content in the samples from the various sites varied significantly. The findings show that the concentration of radionuclides near mining sites (point source) was higher than that at 200 meters away from the sites and those at 400 meters away respectively. There was however, a few exceptions where the opposite was true, these patterns were seen almost everywhere from the result of the analysis. At a 200-meter distance from the mining sites, KW_2 had a greater concentration of ^{40}K than KW_1 (i.e. point source). B_2 samples, which were taken 200 meters away from the point source, were likewise more abundant than B_1 samples collected from the mining locations (point source). SSK_3 was

found to be greater than SSk_2, as well. Given that ^{40}K is a nutrient for plants, the cause of this fluctuation may be related to the physicochemical characteristics of the soil and the surrounding vegetation. The overall results for ^{226}Ra and ^{232}Th exhibit a pattern, with the higher values (results) coming from samples taken directly from the mining sites, followed by samples taken from 200 and 400 meters distant from the sites, respectively. These findings show that radionuclides are typically released into the environment through a variety of activities, some of which may be connected to mining processing, transportation, water and wind erosion respectively. A significant additional factor that influences the distribution and fate of radionuclides in the soil is the properties of the particular radionuclide, its chemical form and reactivity, control of the nature of its retention in the soil, and the affinity to certain soil constituents [10]. The concentration of ^{226}Ra varies from 109.61 ± 0.04 to 62.54 ± 0.36 in mining sites, 99.91±1.11 to 43.78±0.14Bq/kg from 200m away from mining sites, 67.87±0.12 to 31.39±0.73Bq/kg from 400m away from mining sites respectively. These values are similar to those reported by [11], in Mazat Barkin Ladi. Almost all the results recorded are higher than the recommended safe limits of 33Bq/kg except for a few cases in KJ_3, $ZPSC_3$, and Z_3 which fall below the recommended limits of 33Bq/kg [12]. ^{232}Th ranges from 102.50±0.55 to 27.23±0.49 around the mining sites, 72.48±0.22 to 22.43±0.73 200m away from the mining sites and 64.23±0.10 to 6.99±0.23 Bq/kg 400 meters away respectively. K_2 and K_3 recorded a higher value than K_1. Soil physicochemical properties may also have been responsible for this abnormality. All the results of ^{232}Th around the mining sites are higher than 45 Bq/kg permissible limits, except for K_1 and $ZPSCs_1$ whose values are 31.94±0.51and 27.23±0.49.

The most significant property of soil is its pH level, it has effects on all other parameters of soil. Therefore, pH is considered while analyzing any kind of soil. If the pH is less than 6 then it is said to be an acidic soil. When pH ranges from 6 to 8.5, it is said to be normal soil, but greater than 8.5 then it is said to be alkaline soil. [13]. The pH of the soil samples from this investigation ranges from 3.12. to 6.8 which is lower than the 7.3 to 7.9 for raining season reported by [14], but similar to the report of [15]. A decrease in the pH values will lead to an increase in the mobility of the radionuclides [16]. Additionally, in this study, the organic carbon (OC) ranges from 0.51% to 0.02%, and Organic Matter ranges from 3.33% to 0.01%. Soil organic matter (OM) is of great importance due to its influences on the mobility, solubility, and complexation of the radionuclides in the soils [15]. Cation exchange capacity ranges from 4.91Cmol/Kg to 1.01Cmol/kg respectively. Other soil properties studied were clay content which ranges from 42.96 % to 10.02%, which is similar to the report of [15]. A decrease in clay content will increase the chances of the mobility of the radionuclides. Silt ranges from 40.39% to 8.32% and sand from 76.28 to 32.47% higher than the report of [15]. A decrease in sand content will lead to a decrease in the mobility of the radionuclides and vice versa [16].

Generally, the high concentration of radionuclides may be a result of the application of phosphate fertilizer in the farmland around the study area [17].

To draw the provenance of radionuclides and their relationship with the soil physiochemical properties, a Pearson correlation matrix was tabulated in Table 2. ^{40}K shows a strong positive correlation with ^{226}Ra ($r = 0.854$, $p \leq 0.05$), which signifies their common mineralogical affinity and/or similar sources, it also signifies that the absence of one will affect the other. Both ^{226}Ra and ^{40}k are released from parent minerals (e.g. clay minerals) as ions during weathering, and thereafter preferentially adsorbed by the clay [18], from the clay, Ra and K are transferred to the soil solution and are available for migration and uptake by plants/vegetables [19]. A moderate negative correlation was observed between ^{40}K and ^{232}Th ($r = -0.510$, $p \leq 0.05$), which indicates their different geochemical behaviour [18][19]. This negative correlation was similar to the report of Habib *et al*. (2018). ^{232}Th shows a very strong positive correlation with ^{226}Ra ($r = 0.701$, $p \leq 0.05$). pH of the soil shows a strong positive correlation with ^{226}Ra ($r = 0.918$, $p \leq 0.05$) which is an indication that the solubility and mobility of ^{226}Ra increase with increasing soil acidity [20]. ^{40}k was negatively correlated with OC and OM ($r = -0.910$, $p \leq 0.05$ and -0.894, $p \leq 0.05$), which reveals inorganic mineralogical (e.g. illite) affiliated with OM [19]. Clay has a strong correlation with ^{40}K and ^{226}Ra ($r = 0.935$ and 0.887), while a weak correlation was observed between Clay and ^{232}Th ($r = 0.348$). CEC has a strong positive correlation with Clay ($r = 0.923$). The reason may be a result of the fact that clay minerals exhibit the largest surface area, which is

Table 1. Summary of the result for the analysis of radionuclides ^{40}K, ^{226}Ra, and ^{232}Th sample collected in Barkin Ladi and Jos South LGA

SAMPLE ID	^{40}K	^{226}Ra	^{232}Th	pH	OC %	OM %	CEC cmol/kg	Clay %	Silt %	Sand %
KW_1	210.26±0.24	98.71±0.64	93.79±0.26	4.10	0.32	0.07	4.36	30.72	22.00	47.28
KW_2	276.08±0.08	97.32±0.40	39.08±2.40	4.11	0.24	0.09	4.52	40.12	12.42	47.46
KW_3	124.97±0.31	51.99±0.30	26.99±0.38	3.21	0.42	0.38	2.61	30.53	25.76	43.71
FJ_1	315.23±0.25	98.21±0.41	89.24±0.33	3.76	0.28	0.02	4.49	36.72	24.00	39.28
FJ_2	204.14±0.32	85.88±0.24	72.48±0.22	3.93	0.21	0.21	3.21	42.96	23.91	33.13
FJ_3	142.87±0.46	67.87±0.12	64.23±0.10	6.81	0.23	0.01	4,91	29,32	38.21	32.47
B_1	231.86±0.34	85.97±0.41	69.08±0.51	4.34	0.20	0.41	2.03	30.72	28.00	41.28
B_2	249.41±0.24	63.43±0.52	62.21±0.55	3.76	0.20	0.32	3.50	25.21	22.56	52.23
B_3	210.27±0.10	46.19±0.80	41.88±0.08	5.32	0.30	0.62	2.90	21.24	40.39	38.37
NK_1	301.91±0.24	83.41±0.30	76.20±0.37	5.16	0.16	2.97	1.29	20.72	24.00	55.28
NK_2	236.31±0.33	58.02±0.35	58.57±0.43	5.10	0.24	3.01	3.32	40.78	23.67	34.54
NK_3	139.92±0.24	43.10±0.62	35.73±0.51	4.21	0.41	1.41	2.90	10.02	35.32	54.66
N_1	208.11±0.34	105.42±0.1	86.65±1.73	3.96	0.16	2.96	3.61	34.61	22.41	42.98
N_2	157.12±0.48	99.91±1.11	67.39±0.94	4.50	0.08	3.12	1.01	20.50	38.67	38.83
N_3	167.32±0.15	57.02±0.22	61.20±0.22	6.32	0.19	2.74	4.90	36.00	12.19	51.81
K_1	259.34±0.44	62.54±0.36	31.94±0.51	4.61	0.24	0.45	1.16	9.72	13.00	76.28
K_2	172.15±0.12	53.32±0.18	47.61±0.41	5,28	0.04	1.84	3.76	31.87	19.38	48.75
K_3	161.39±0.18	44.32±0.32	36.92±0.54	6.71	0.31	2.59	2.41	30.97	22.43	56,60
SSK_1	305.49±0.31	79.13±0.81	98.40±0.52	4.06	0.40	0.10	1.34	10.72	14.00	75.38
SSK_2	201.64±0.37	62.51±0.37	47.38±0.42	4.32	0.24	1.93	3.21	29.67	36.41	33.92
SSK_3	209.74±0.22	35.31±0.33	37.18±0.57	3.70	0.50	2.03	3.82	31.90	21.87	46.23
Z_1	319.48±0.22	69.65±0.13	52.46±0.23	4.34	0.32	1.05	2.09	35.87	22.65	41.48
Z_2	211.94±0.14	43.78±0.14	23.87±0.40	3.12	0.12	3.32	3.52	25.12	26.57	48.31
Z_3	167.92±0.32	32.47±0.21	16.99±0.23	4.21	0.02	2.04	4.01	23.43	29.31	47.26
$ZPSC_1$	261.14±0.39	77.32±0.41	27.23±0.49	6.90	0.39	2.01	4.11	38.00	12.45	49.55
$ZPSC_2$	200.27±0.82	52.34±0.82	22.43±0.73	4.92	0.12	1.52	1.08	20.35	40.12	39.34
$ZPSC_3$	159.38±0.07	31.39±0.73	24.35±0.20	5.80	0.31	2.04	2.90	25.56	40.32	34.12
KJ_1	180.39±0.42	109.61±0.0	102.50±0.5	3.82	0.40	1.10	1.27	10.72	14.00	74.28
KJ_2	263.78±0.67	77.27±0.44	66.72±1.92	3.56	0.44	2.01	4.10	40.82	8.32	50.86
KJ_3	210.98±0.25	31.67±0.68	42.65±0.00	5.63	0.51	0.63	2.32	30.10	15.30	54.60
Permissible limits	420	33	45							

N=3

Key: kw= kwat, FJ= Foron junction, B=Ban, Nk=Nkan, N=Newo, K=Kuru, SSK=Science School Kuru, Z= Zawan, ZPSC=Zawan Police Staff College and KJ=Kuru Jenta

OC = Organic Carbon, OM = Organic Matter, CEC= Cation Exchange Capacity

Table 2. Mutual correlation matrix of radionuclides and soil properties of Jos South and Barkin Ladi Local Government Area

	^{40}k	^{226}Ra	^{323}Th	pH	OC	OM	CEC	Clay	Silt	Sand
^{40}k	1									
^{226}Ra	0.854	1								
^{232}Th	-0.510	0.701	1							
Ph	0.633	0.918	0.506	1						
OC	-0.910	-0.901	-0.350	-0.840	1					
OM	-0.894	-0.986	-0.767	-0.840	0.870	1				
CEC	0.884	0.996	0.650	0.911	-0.933	-0.982	1			
Clay	0.935	0.887	0.348	0.800	-0.997	-0.867	0.923	1		
Silt	-0.444	-0.514	0.236	-0.701	0.756	0.393	-0.559	-0.726	1	
Sand	-0.912	-0.771	-0.743	-0.457	0.687	0.864	-0.778	-0.722	0.049	1

important for soil chemistry and CEC, and also for water-holding capacity important for transporting nutrients and pollutants to soil organisms and plants [16]. Sand witnessed a strong negative correlation with CEC (r = -0.722), this is because of the inability of sand to retain some nutrients and some inorganic elements.

One-way analysis of variance (ANOVA) was performed to compare the radionuclide concentration and their fate and migratory patterns in the environment based on the different soil physiochemical properties. The one-way ANOVA revealed that there was a statistically significant difference in the mean

concentration of the radionuclides (F ratio values was 218.9890) p = 0.00001) at p = 0.05.

4. CONCLUSION

Natural radionuclide distribution into the environment and statistical analysis of the naturally occurring radioactive materials in soils of some abundant mining sites of Jos South and Barkin Ladi Local Government Area of Plateau State Nigeria were studied. It was found that the natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K are the main radiological constituents of the soil. The result of the analyses shows that the activity concentration of the natural radionuclide in some of the sampling sites is above the recommended limits whereas some are lower. The result for ^{40}k in all the soil samples is below the permissible limits. The result reveals that the activities of artisanal miners around the ex-mining site contribute to the higher concentrations of radionuclides in most of the areas, and also aid in the distribution of the radionuclides into the environment.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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