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Radioactivity Concentrations in Soil Samples from Kandam, Gyalgyal, Burmawan masaka, Dinbisu and Giyawa Mines in Wurno LGA, Sokoto State, Nigeria

Y. M. Ahijjo[1*], A. N. Baba-Kutigi[2], M. Momoh[1], A. M. Bayawa[3] and M. Hussain[4]

[1]Department of Physics, Usmanu Danfodiyo University, P.M.B. 2346, Sokoto, Sokoto State, Nigeria.
[2]Department of Physics, Federal University, Dutsin-Ma, Katsina State, Nigeria.
[3]Department of Pure/Applied Chemistry, Usmanu Danfodiyo University, P.M.B. 2346, Sokoto State, Nigeria.
[4]Department of Physics, GC University, Lahore, Pakistan.

Authors' contributions

This study was successful through the collaboration between all authors. Author YMA did the field work, participated in the laboratory activities, performed the statistical analysis and drafted the blue-print of the manuscript. Authors ANBK, MM and AMB recast the expressions, phrases, clauses, paragraphs and managed the literature in analyzing the literature and results. Author MH provides adequate consumables; laboratory and logistics for all the experimentations. All authors read and approved the final manuscript.

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ABSTRACT

Aims: This study was conducted in a bid to ascertain the concentration of NORMS in the soil samples dugout of some mining sites in Kandam, Gyalggal, Burmawan masaka, Dinbisu and Giyawa town in Wurno LGA, Sokoto State Nigeria. The concerns about NORMS radioactivity in the environment are frequently linked to natural radioactivity which is more of primordial radionuclides.
Study Design: A total number of nine soil samples were randomly collected for this study. The randomness was in order to represent over 90% of the sample population of different mines in Wurno LGA, Sokoto state for the analysis of gamma-ray spectrometry of High Purity Germanium (HPGe) detector coupled to a Multi-Channel Analyzer (MCA).

**Corresponding author: E-mail: yusuf.musa@udusok.edu.ng;*

Place and Duration of Study: Department of Physics, Usmanu Danfodiyo University, Sokoto, Nigeria, between August 2015 and June 2016.
Methodology: The concentrations of the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were ascertained through a high-resolution gamma-ray spectrometry of HPGe detector coupled with a Multi-Channel Analyzer, Centre for Advanced Studies in Physics (CASP), Government College University (GCU), Lahore, Pakistan. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples were obtained and used to calculate the radiological parameters (Ra_{eq}, H_{ex}, H_{in}, D_{Abs} and D_{Eff}) in order to assess the radiological impacts on the inhabitants.
Results: The mean activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K in the soil samples were found to be 58.34, 53.76 and 679.70 $Bqkg^{-1}$ while the radiological hazards parameters revealed mean values of 187.55 $Bqkg^{-1}$, 0.51, 0.66, 87.77 $nGy.h^{-1}$ and107.64 $\mu Svyr^{-1}$ for Ra_{eq}, H_{ex}, H_{in}, D_{Abs} and D_{Eff} respectively. These were compared with the results of similar studies conducted elsewhere and around the world. It was noticed that ^{40}K contributes the highest level of radioactivity in this study which could enhance the amount of background radiation received by the population in the study area. The activity concentration unveiled from the soil samples were considerably lower for ^{226}Ra and ^{232}Th when compared to the activity of ^{40}K. The mean value of the dose calculations shows that an elevated exposure to radioactivity by the inhabitants around this mine is eminent.
Conclusion: Cancer and various forms of ailment due to radioactivity exposure are further anticipated for the workers in the pits and the dwelling communities.

Keywords: Radioactivity monitoring; Wurno; Sokoto; mines; soil; cancer.

1. INTRODUCTION

The basic premise of this research is on the philosophy of ICRP, UNSCEAR, USEPA, WHO and IAEA that strengthen the effort of radiological protection. It was as well conceptualised in preventing the deterministic effects of exposure to environmental radioactivity, and also reducing the occurrence of the stochastic effect of cancer and other relative ailments. Evidence has been established that the major sources of human exposure to ionising radiations are the natural sources [1].

This means that there is a quest for individual protection by determining exposure limits or regulation on the risk of eminent exposure to radiological impacts in the environment [2].

The recent challenges in the global economy have raised the dependence on mineral extraction, exploration, mining etc. to an alarming level [3].

Materials like sand, gravel, rock, talc, asbestos, limestone etc. are dramatically harnessed from mining for aggregates, concretes, roofing slates, cement, ceramics etc. These are enjoyed with relatively high exposure to impurities inform of toxics to health status and pollution to the environment [4]. The exposure to radioactivity in the environment was found to account for roughly 85% of annual exposure dose received by the populations around the world [5].

Radioactivity exposures arise in the mining sites and their neighbourhood through three significant pathways which includes external gamma radiation from ores, inhalation of dust containing long-lived alpha-emitting radionuclides and inhalation of the short-lived decay progenies of some radionuclides [6].

Various studies concerning environmental radioactivity have provided information on the sources and levels of background radiation in some observed changes on radioactivity levels and particular areas, but little evidence have unveiled and assessed the activities in the mining communities that could lead to elevated exposure of commonly occurring radionuclides of ^{40}K, ^{238}U, ^{226}Ra, ^{232}Th, ^{137}Cs with an interest on the maladies to human health [7].

Hence, this paper attempts to unveil the concentration of radioactivity in the soil samples collected from the mines. The radiological tendencies of the inhabitants around these mines will be also assessed through theoretical calculations. The results of this study may also signal some dangers attributable to indiscriminate mining of raw materials like Kaolin,

Gypsum, Limestone etc., from the study area in Sokoto state.

The outcome of this finding could be added to the obtainable data of similar studies in other locations. Similar studies were conducted by [8,9,10,11,12,13,1] in a bid to unveil the environmental radioactivity and elemental analysis from environmental samples of farmland or mining sites.

2. MATERIALS AND METHODS

The sampling was considered based on proportions, representation of the sample, homogeneity, sample points code and size (taking the weight) respectively. Soil samples were randomly collected from five mines in Wurno LGA, Sokoto State in a fashion that satisfies the outlines according to [14,15,16,17,18]. The soil samples were collected with the aid of a cleaned hand auger from 0 *cm* depth where hips of excavated soil from the mines are dumped in a distribution of 5 different points at about 25 *km* apart. Three separately collected samples were homogenised within 50 *cm* parts to achieve a 95 to 98% representation of the soil for a particular location. The weight of the collected samples was measured so as to achieve a 2.5 kg gross weight of the sample from the field. They were then parked into a 300 ml plastic container, labelled with a distinct code and then stored in their sealed form before shipping to a laboratory for further experimental exercise. They were the measurement by gamma-ray spectrometry of HPGe detector coupled with an MCA. The samples were dried for seven days under the ambient temperature of the air and manually sorted by removing stones, leaves and other organic particles. They were gently dried at $75^{o}C$ for 24 hours in an oven for a careful removal of potential residual moisture and to avoid loss of volatile radionuclides to enable equilibration [19].

They were crushed, pulverised and homogenised with the aid of a mortar and pestle and then sieved through a standard mesh of 2.5 mm sizes using a vibratory sieve shaker to enable uniform distribution of radionuclides [20].

The samples were hermetically sealed with a PVC tape to prevent gaseous nuclide flux in a standard 250 ml Marinelli beakers for 4 weeks to enable equilibration of the radionuclide progenies and to minimise partial accumulations of the radionuclides after preparation. To attain radioactive secular equilibrium, 4 weeks maximum duration was set between ^{222}Rn (radioactive noble gas), its decay products (^{214}Pb and ^{214}Bi) and radium (^{226}Ra), from the ^{238}U decay series. For the ^{232}Th decay series, the radon isotope (^{220}Rn) poses no serious problem in equilibration due to its short half-life of 55 seconds and in the ^{40}K decay series, no equilibrium is accounted for [20].

The techniques of gamma-ray spectrometry of HPGe detector was used to count all the soil samples in this study as outlined by Umar et al. [21].

2.1 Gamma-ray Spectrometry

The measurement of the activity concentrations of the radionuclides was performed using a P-type coaxial HPGe detector in a low-background configuration. The HPGe was made of concentric cylinders that were closed at the end. The Peak-to-Compton ratio of the HPGe detector was 1332.5 keV at the photopeak of ^{60}Co. The detector was housed in a 12 cm of lead shielding to reduce the interference of background radiation from the surrounding with a cooling system of liquid nitrogen in a vertical cryostat Dewar (50 litres) to suppress the leakage current [22].

The detector cooling was incorporated for the system to enhance operation on low temperature for optimal performance of the energy and detection efficiency during operation that requires cooling below 110^{o}C. The detector system was fully automated with a multi-channel analyser that was installed in a Personal computer for data acquisition. A fixed geometry was maintained for the standard source and the samples by choosing a 250 ml Marinelli beaker for the coaxial HPGe detector. The signals processing in the gamma-ray spectrometry set-up used the main amplifier which was incorporated with an efficient pile-up ejector in the multi-channel buffer that consists of an analogue-to-digital converter (ADC).

For a linear amplification, the pulse height appears proportional to the gamma-ray energies absorbed. The pulses from the amplifier were then sorted by their pulse height and then converted to a digital number referred to as channel by the analogue-to-digital converter

(ADC) in the multichannel analyser (MCA). The identification of individual radionuclides was then achieved with the aid of gamma-ray analysis software, *GENIE 2000* of Canberra Company. It was used to acquiring spectrum signal for analysis in the MCAs. It was also used to perform Multiple Channel Analysis control, spectral display, basic spectrum analysis for the gamma-ray spectroscopy and quality control [23].

The spectroscopic windows in the MCA were used for energy and efficiency calibration options and peak to total calibration which were maintained and stored for use. The spectrum results were obtained from the MCA spectra of the gamma-ray photopeaks of the typical radionuclide in the sample as well as all the interactions ranging from scattering events to photo peaks that were initiated by photoelectric effects. The two well-known ^{60}Co peaks (1173.2 and 1332.5 keV) were identified by their direct gamma-ray emissions. Other peaks of ^{241}Am (59.54 keV), ^{109}Cd (88.03 keV), ^{57}Co (122.06 keV), ^{139}Ce (165.86 keV), ^{203}Hg (279.20 keV), ^{113}Sn (391.69 keV), ^{85}Sr (514.01 keV), and ^{88}Y (898.04 keV and 1836.1 keV) were obtained from the standard source. This spectrum was found with additional peaks at 60.0, 88.0, 122.1 and 661.7 keV for ^{241}Am, ^{109}Cd, ^{57}Co, and ^{137}Cs respectively.

The efficiency of the HPGe detector was 60% with a resolution of 1.85 keV in its full-width at half maximum (FWHM) of the gamma-ray energy line of ^{60}Co at 1332.5 keV. A unified preamplifier was connected to the terminal for varying transmissions of the signals. The energy calibration of the detector was performed with the calibration standard source containing the following radionuclides: ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}S, ^{137}Cs, ^{60}Co, and ^{88}Y by inserting the standard source inside the detector setting for 86,400s counting time.

The soil samples were then counted on the HPGe detector for 86,400s. The activity concentration of the radionuclides in the samples was determined on a dry weight basis in $Bqkg^{-1}$. The selected peaks were then manually calibrated by carefully entering and matching the known energies to a corresponding ROI centroid. The standard source channels range were defined by setting the energy window for detecting ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs. Although, ^{137}Cs and ^{40}K are known to decays directly in gamma-ray by their common emission lines of 661 keV and 1460 keV. A linear fit was then used to obtain the energy calibration parameters. The amplifier gain was set such that 1 spectrum channel corresponds to 0.5 keV. The linear relationship of gamma-ray energies (0 - 2000) and channel numbers (0 - 6000) was set within the ROI obtained from the linear equation 1 by the computer [24,25].

$$E = A_1 + A_2 ChannelNumber \quad (1)$$

where: E is the energy in keV, A_1 and A_2 are calibration constant for a given geometry.

Hence, a linear relationship was obtained for energy in keV against respective channel number.

The activity concentration of a radionuclide in the soil samples is given by equation (2):

$$A_c = \frac{N_E}{\varepsilon_\gamma p_\gamma t_c m} \quad (2)$$

Where A_c is the activity concentration of a particular nuclide in of $Bqkg^{-1}$. N_E the net count at the interest peak energy as the corrected background counts of the corresponding full-energy peak, ε_γ is the absolute full-energy peak detection efficiency, p_γ is the gamma-ray emission probability, t_c is the counting time in 86,400 seconds, and m is the mass of the samples in kilogram. The minimum detectable activity (MDA) for each radioelement was obtained by equation 3 according to Gilmore and Hemingway [23,26].

$$MDA = \frac{DL}{\varepsilon_\gamma p_\gamma t_c m} \quad (3)$$

Where DL is the detection limit under the background counts B such that:

$$DL = 2.71 + 465\sqrt{B} \quad (4)$$

The errors in the measurements were expressed in terms of standard deviation (±σ), where σ is expressed as in equation 5 [23,26].

$$\sigma = \left[\frac{N_S}{T_S^2} + \frac{N_b}{T_b^2} \right]^{1/2} \quad (5)$$

Where, N_S is the sample counts measured in time T_S and N_b is the background counts measured in time T_b. The standard deviation $\pm\sigma$ in cps was converted into activity concentration in $Bqkg^{-1}$ according to equation 4 [23,26].

2.2. Radiation Hazards

2.2.1 Radium equivalent activity

Based on the detected values of the measured ^{226}Ra, ^{232}Th and ^{40}K for all the samples, the radium equivalent activity was calculated from equation 6;

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (6)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$. Due to the assumption that radiologically contaminated materials from around mine are usually locally used for the construction of homes in most sampled localities, hence the external hazard index (H_{ex}) and internal hazard index (H_{in}) could be calculated from equations 7 and 8, [27].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (7)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (8)$$

where: A_{Ra}, A_{Th} and A_K are the activities concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$

The assessment of radiological hazard to the inhabitants around these mines due to exposure to ionising radiation from the commonly occurring radionuclides in the environment was based on the guidelines of [6].

The mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K ($Bqkg^{-1}$ and Bql^{-1}) in the samples were used to calculate the absorbed dose rate deposited on an individual by equation 9;

$$D_{Abs.}(nGy.h^{-1}) = (0.462A_{R_a} + 0.604A_{Th} + 0.0417A_K). \quad (9)$$

where $D_{Abs.}$ is the absorbed dose rate in $nGyh^{-1}$, A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The dose coefficients in units of $nGyh^{-1}$ per $Bqkg^{-1}$.

To account for the annual effective dose rates, the conversion coefficient from absorbed dose in air ($D_{Abs.}$) to effective dose (0.7 $SvGy^{-1}$) and outdoor occupancy factor (0.2 $SvGy^{-1}$). The effective dose rate in units of $\mu Svyr^{-1}$ was calculated from equation 10 [23,26].

$$D_{Eff}(\mu Svyr^{-1}) = D_{Abs.}(nGyh^{-1}) \times 8760h \times 0.2 \times 0.7SvGy^{-1} \times 10^{-3} \quad (10)$$

where $0.7SvGy^{-1} \times 10^{-3}$ is the conversion factor for absorbed to effective dose rate, 0.2 is the outdoor occupancy factors for 20% time spent by an individual, $8760h$ is the number of hours around the year.

3. RESULTS AND DISCUSSION

3.1 Activity Concentration

The spectrometric results revealed a wide range of spectra from the counting of the samples. The spectral signals were used for the identification of different radionuclides from the soil samples. The results of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K from the soil samples are presented on Table1 while Table 2 presents the radiological hazards parameters.

Table 2 presents the result of radiological hazards parameters in this study. It could be observed from the table that the calculated values of Ra_{eq} for the entire soil samples were in lower levels from 127.33 to 278.41 $Bqkg^{-1}$ with a mean of 187.55 $Bqkg^{-1}$ respectively. Also, from these results, Ra_{eq} values were comparably lower than 370.00 $Bqkg^{-1}$ which is the international threshold set by [6]. Similarly, the calculated mean values of H_{ex} and H_{in} from the activity concentrations of the radionuclides were

Table 1. Measured values of activity concentration of ^{226}Ra, ^{232}Th soil samples, Wurno LGA, Sokoto State

S/n		Activity concentration ($Bqkg^{-1}$)		
		^{226}Ra	^{232}Th	^{40}K
1.	Kandam	59.8±8.2	31.4±0.3	589.15±23.8
2.	Gyalggal	56.4±3.1	52.3±2.4	547.18±24.9
3.	Burmawan masaka	69.6±9.3	75.8±3.1	398.05±19.2
4.	Dinbisu	89.4±8.1	67.5±2.6	1201.07±19.3
5.	Giyawa	16.5±9.1	41.8±0.8	663.04±18.5
Minimum		16.50	31.40	398.05
Maximum		89.40	75.80	1201.07
Mean		58.34	53.76	679.70
World Mean		35.02	45.03	420.12

found to be 0.51 and 0.66 respectively. These values were found fairly close unity threshold for international exposure for both inhabitants and occupational safety. Also, to assess the radiological impacts from the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K on the inhabitants of these communities, the absorbed dose and the annual effective dose rates were calculated using equations 9 and 10. The mean absorbed dose in the ambient air due to terrestrial gamma-ray was found to be 87.77 $nGy.h^{-1}$ and 107.64 $\mu Svyr^{-1}$ for D_{Abs} and D_{Eff} respectively. For the annual effective dose rates, a conversion factor of 0.7 $SvGy^{-1}$ was utilised to obtain the effective dose from the absorbed dose with a special emphasis on 0.2 as the outdoor occupancy factor.

The activity concentrations of the five samples were obtained in a fashion of Concentration Level ± Standard Error of the mean for ^{226}Ra, ^{232}Th and ^{40}K and they were presented in ranges. The results graphically illustrated in Figure 1. ^{40}K was detected from its direct decay in gamma at 1461 keV while other radionuclides of ^{238}U and ^{232}Th series with numerous decay daughters were identified from the spectra of their progenies. The activity concentrations of ^{226}Ra and ^{232}Th were calculated from the spectra generated using equation 2.

The activity concentration from the soil of these mines in Kandam, Gyalgyal, Burmawan Masakka, Dinbisu and Giyawa in Wurno LGA, Sokoto state is presented in the chart of Figure 1. The mean values of the activity concentration measured were found to be 48.34±7.0, 80.76±1.8 and 463.52±21.1 $Bqkg^{-1}$. The activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K did not show any definite pattern in distribution with respect to any particular soil sample from the mines, but the activity concentration of ^{40}K appears to be highly significant when compared to those of ^{226}Ra and ^{232}Th respectively. The lowest activity concentrations of the three radionuclides were found to be 48.34±7.0 $Bqkg^{-1}$ for ^{226}Ra which may be due to a higher occurrence of ^{232}Th and ^{40}K in the most soil of the study area. The samples from Dinbisu was found with the highest level of activity concentrations for ^{226}Ra and ^{40}K by 89.4±8.1 and 1201.07±19.3 $Bqkg^{-1}$ while ^{232}Th had its highest value of 75.8±3.1 $Bqkg^{-1}$ at Burmawan Masakka. These values of radioactivity were found in significant levels above the world mean threshold of 35.02, 45.03 and 420.12 $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K according to UNSCEAR report [6]. The concerns on this hike in the values of radioactivity could be prominent due to the prevalence of younger granite rocks, Feldspars and Phosphate rocks in the Iullemeden basin where Wurno LGA form apart [28].

Most precious minerals that are mined in parts of Sokoto basin are also from quartz-mica rocks, granites rocks and Monazites which are known to be responsible for the higher level of radioactive exposure to inhabitants from their embedded primordial sources [12,11].

The results of this study were found in good agreement with the results obtained in the work of other researchers in Nigeria and around the world. The results are presented in comparison with other results in Table 3.

Table 2. Radiological hazards parameters for the soil samples collected from Wurno LGA

S/n sample point	Radiological parameters				
	Ra_{eq}	H_{ex}	H_{in}	$D_{Abs}(nGy.h^{-1})$	$D_{Eff}(\mu Svyr^{-1})$
1. Kandam	150.07	0.41	0.57	71.16	87.27
2. Gyalggal	173.32	0.47	0.62	80.46	98.68
3. Burmawan masaka	208.64	0.56	0.75	94.54	115.94
4. Dinbisu	278.41	0.75	0.99	132.16	162.08
5. Giyawa	127.33	0.34	0.39	60.52	74.22
Minimum	127.33	0.34	0.39	60.52	74.22
Maximum	278.41	0.75	0.99	132.16	162.08
Mean	187.55	0.51	0.66	87.77	107.64
World Mean	370.00	≤1	≤1	57.00	100.00

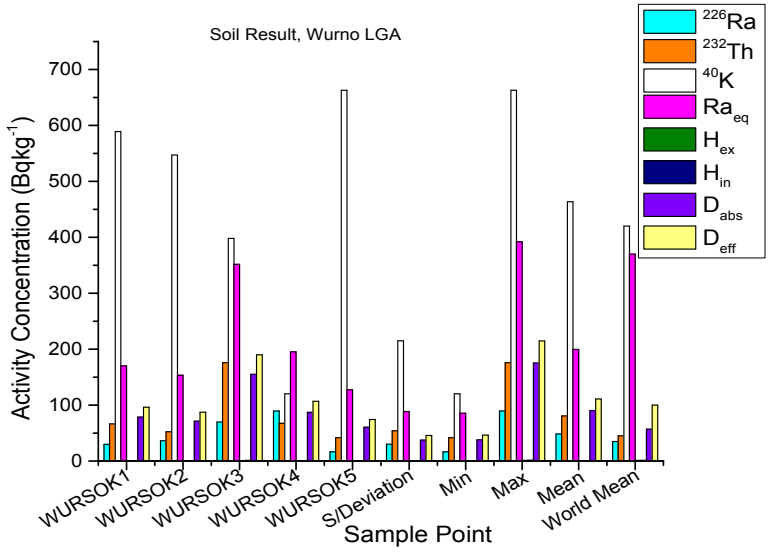

Fig. 1. A combined graph of radionuclides activities and radiological parameters for soil samples from Wurno LGA, Sokoto state, Nigeria.

Table 3. Comparison of the results obtained in this study with previous studies

Sn	Country	Activity concentration ($Bqkg^{-1}$)			Reference
		^{226}Ra	^{232}Th	^{40}K	
1.	Nigeria (Wurno, Sokoto)	58.34	53.76	679.70	This study
2.	Nigeria (Zaria, Kaduna)	-	215.18±8.70	476.04±28.07	[29]
3.	Nigeria (Sokoto)	720±4.2	33.5±1.4	315.3±6.7	[12]
4.	Saudi Arabia (Riyadh)	14.5±3.9	11.2±3.9	225±63	[30]

4. CONCLUSION

In this paper, the method of gamma-ray spectrometry has been used to ascertain the concentration of radioactivity from some mines in Wurno LGA, Sokoto state, Nigeria. The activity concentration of ^{40}K was found topping the chart of the results of natural radioactivity of the commonly occurring radionuclides from the mines. ^{226}Ra, ^{232}Th and ^{40}K were found significantly higher in a concentration above the world mean values which entails an enhanced background radiation levels in the study area. There are also significant levels of some radiological parameters unveiled in this study. Hence, there are potential dangers of exposure

to radioactivity to the workers in these mines and the general public dwelling in these communities. The soil from this mine must be carefully handled in case of any domestic or industrial needs to mimic radiological hazards.

Lastly, due to lack of sufficient accounts on the level of radioactivity in the soil of mines from these localities, the results obtained from this study stands a good chance of baseline data for research on the radioactivity measurement in the environment and mining sites of this region.

However, effective radiation monitoring measures are recommended to enable the frequent exercise of determining radioactivity levels so as to checkmate any potential radio-toxic burden on the general public.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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