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Assessment of Radioactivity and Soil Dose Rate in Some Selected Areas of Wurno Mines-Nigeria

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ABSTRACT

Radiation hazards pose significant risks to individuals working in or residing near mining sites. This study undertakes a comprehensive assessment of radiation hazards emanating from soil in selected mining sites using gamma spectroscopy. In this research, Soil samples were obtained from various mining and residential areas, and radiometric analysis was conducted using a High-Purity Germanium (HPGe) gamma-ray spectrometer. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured as 186.6±9.7 Bq/kg, 52.2±2.8 Bq/kg, and 111.7±5.9 Bq/kg, respectively. Furthermore, radiation parameters were evaluated, with the absorbed dose rate, annual effective dose, external hazard index, internal hazard index, annual gonadal dose equivalent (AGDE), and excess lifetime cancer risk (ELCR) yielding average values of 122±6.2 nGy/h, 0.15±0.01 mSv/y, 0.73±0.04, 1.23±0.06, 830.4±41.94 µSv/y, and 0.526×10^{-3}, respectively. Many of these values exceeded recommended limits, indicating potential radiological risks to both the environment and public health. The findings of this research demonstrate the urgent need for radiation monitoring and mitigation strategies in these mining sites to protect the health and safety of the population.

Keywords: Gamma-ray Spectroscopy, Radionuclides, Soil, Dose Rates, Wurno.

INTRODUCTION

Radiation is the emission or transmission of energy in the form of electromagnetic waves through space or a medium (Khan, 2009; Park and Kang, 2011). Radiation applications play substantial role in several areas of science and technology such as therapy, agriculture, elemental analysis and so on. Despite its advantages, several drawbacks were attributed to ionizing radiations such as gamma rays. Environmental radiation monitoring involves the organized gathering and examination of specific environmental materials, including air, milk, and water, to assess the levels of radioactivity (Dankawu et al., 2022). The Earth's atmosphere, particularly its human population, is subject to exposure from both non-ionizing and ionizing radiation originating from various sources, both natural and artificial. Key natural sources include primordial radionuclides like ^{238}U, ^{232}Th, ^{40}K and the products. In contrast, artificial sources comprise anthropogenic radionuclides such as ^{137}Cs and ^{90}Sr (Oyeyinka et al., 2012; El-Arabi et al., 2007).

Radiation interactions damage cells. Ionizing radiation transmits sufficient energy to eject closely bound electrons from atoms or molecules, creating ions. Gamma, Alpa, Beta and x-rays gives ionizing radiation due to high energy sufficient to liberate electrons from their atoms (WHO, 2012). Ionizing radiation damages the DNA and causes chromosomal abnormalities in cells. It yields to cell transformation; one of the stages in cancer development. If the radiation didn't kill the cells, it leads to their modification which is then transmitted to the daughter cells eventually leading to cancer in the tissue or organ of the exposed individual. Health problems like abnormal cell growth can be caused due to high level exposure to radiation. Naturally occurring radionuclide

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materials (NORMs) like ^{238}U, ^{232}Th and ^{40}K can be found in soil and rocks surrounding the human population. These materials depending on the concentration of their activities, provide doses of radiation that at a certain limit becomes harmful to the inhabitants.

Several studies employing various sampling methods, sample locations and objectives have been conducted to assess radiological contamination levels globally. In Nigeria, Adabanija *et al,* (2020) the level and radiogenic heat potential of crystalline basement rocks in Okene, Nigeria. They employ gamma spectroscopy in determining the NORMs activity concentration (^{232} Th, ^{238} U and ^{40} K) in 19 rock samples collected from different lithologies An average value of 63.3nGy/h was determined for the gamma absorbed dose and a value of 3.11mSv/y as the annual effective dose equivalent were found to above the recommended safe values of 60nGy/h and 2.4mSv/y respectively. The natural radioactivity level of ^{40}K, ^{238}U and ^{232}Th in soils and mine tailings of Awo and Ede areas of Osun state Nigeria were assessed by Adewale *et al*, (2015) using gamma-ray spectroscopy of 30 samples as well as an on-site measurement of the area's radioactivity levels with a portable survey meter. Their findings indicated that the variations activity concentrations of the radionuclides from the samples might be attributed to differing geographical settings. Geology of Wurno, located in Sokoto State, Nigeria, is primarily characterized by sedimentary rock formations situated within the Sokoto Basin, one of Nigeria's significant sedimentary basins (Obaje, 2009). These basins have accumulated sedimentary rock layers over millions of years, containing valuable mineral resources such as gypsum and limestone (Ojo, 2005; Musari et al., 2012). The geological formations in the Wurno area include sandstones, shales, and mudstones, with variations in age and composition (Ojo, 2005). Wurno is an area known for its phosphate deposits. The presence of phosphate deposits in the soil can serve as a reservoir for uranium and radium due to the chemical similarities between phosphate ions and these radionuclides.

Isah et al. (2022) also studied the mean cancer mortality and morbidity risks due to ^{238}U and ^{226}Ra, the lifetime average daily dose (LADD) due to ^{238}U and ^{226}Ra as well as their respective hazard quotients from forty-five (45) well and borehole water samples from Wurno L.G.A of Sokoto State. There results revealed that carcinogenic risks were also above the recommended standard while the non-carcinogenic risks due to LADD for both uranium and radium were below the set limit. Their study concluded that that health risks associated with the intake of ground water of the study area are due to radiological factor rather than chemical harmfulness thus recommending further studies to ensure safety in these regions. Given the significance of these findings, it is crucial to further investigate soil samples using different sampling methods, techniques and locations to provide a more comprehensive understanding of radiological contamination from various mining and living areas of Wurno L. G. A.

The aim of this research is to use gamma spectroscopy system with a high purity germanium detector to analyse the Naturally Occurring Radionuclide Materials (NORMs) present of some selected mining areas in Wurno LGA of Sokoto State. Studying the concentrations of these radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in soil is vital for evaluating potential risks to human health. Findings from this study can inform policymaking and regulatory decisions related to environmental protection, land use, and radiation safety standards. In cases of soil contamination with radionuclides, this research can guide remediation efforts to mitigate environmental and health risks.

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Gamma Radiation Interaction with Matter

Gamma radiation is an electromagnetic form of packets of energy (photon) emitted. Gamma- rays may be generated by nuclear processes like annihilation, beta decay processes and nuclear reactions (Knoll, 2010). Three major types of interaction are photoelectric absorption, Compton scattering, and pair production (Knoll, 2010).

In the first mechanism, a photon interacts with the atom given by equation (1)

$$E_b = h\nu_0 + E_b \quad (1)$$

E_b epitomises binding energy of the photoelectron in original shell while $h\nu_0$ is incident photon energy.

In the process of Compton scattering incoming photon interacts with the electron in the absorbing material and then it is deflected over an angle Θ in respect to original direction as depicted in equation (2) (Knoll, 2010). Parts of the photon energy is transferred to the electron which was assumed to be at rest initially thereby been a recoil electron.

$$h\nu' = \frac{h\nu}{[1+\frac{h\nu}{m_o c^2}(1-\cos\theta)]} \quad (2)$$

The term $m_o c^2$ represents the rest-mass energy of an electron (0.511 MeV), while $h\nu'$ and $h\nu$ denote the energy of the outgoing and incoming photons, respectively.

Pair production occurs within the Coulomb field of a nucleus when the gamma-ray energy surpasses twice the electron's rest-mass energy (1.02 MeV). During this process, the gamma-ray photon is annihilated, giving rise to the formation of an electron-positron pair.

Gamma-Ray Spectroscopy

This procedure of identifying radioactive elements present in a sample using the spectrum of gamma ray energies produced by the elements. It is a non-destructive analysis used in carrying out different research works. In this case of study, the type of radiation detector utilized was the semiconductor detector. The interaction of radiation and semiconductors generates electron-hole pairs which can be collected and measured as an electrical signal (Das and Ferbel, 2003; Lutz, 2007). Semiconductor detectors offer high sensitivity and resolution, making them suitable for applications such as X-ray spectroscopy, gamma-ray detection, and particle tracking. Example is the High Purity Germanium (HpGe) detector.

Collection and Preparation of Samples

The soil samples were collected from three (3) mining areas in the Wurno local government area of Sokoto State. For each mining site, samples were collected from four (4) different angles which were then homogenized as one sample to obtain representative sample of the area. A sample was also collected from a neighbouring settlement to each mining area. Six (6) soil samples are collected at 20cm depth below the surface level with a soil auger before taken to the lab for preparation. Fine mesh of (~0.4 mm) was used in sieving the sample after crushing The soil samples were crushed into smaller fragments and passed through a fine mesh (~0.4 mm). The sieved fragments were subsequently dried at 100°C to eliminate moisture. 1000g of each sample was then sealed in plastic polythene bags and taken to the lab for the analysis. To prevent the escape of radium and for equilibrium to be reached between ^{226}Ra and its progeny (Mustapha et al., 1997), the soil samples were kept airtight in standard 500 ml Marinelli beakers for about four weeks at the lab. Names, elevation, latitude, and longitude of the different sample points are tabulated in Table 1 below

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Table 1: Names, Elevation, Latitude and Longitude of the different sample points.

Sample point No	Sample Location name	Elevation	Latitude (°)	Longitude (°)
1	Dabagin-Rama (The settlement)	341	N13.217	E005.500
2	Dabagin-Rama (The mining site)	328	N13.233	E005.492
3	Lugu (The settlement)	277	N13.282	E005.468
4	Lugu (The mining site)	316	N13.277	E005.449
5	Kaurare (The settlement)	289	N13.24	E005.33
6	Kaurare (The mining site)	275	N13.259	E005.265

Figure 1 presents the sampling site map, indicating the sample points within the local government area.

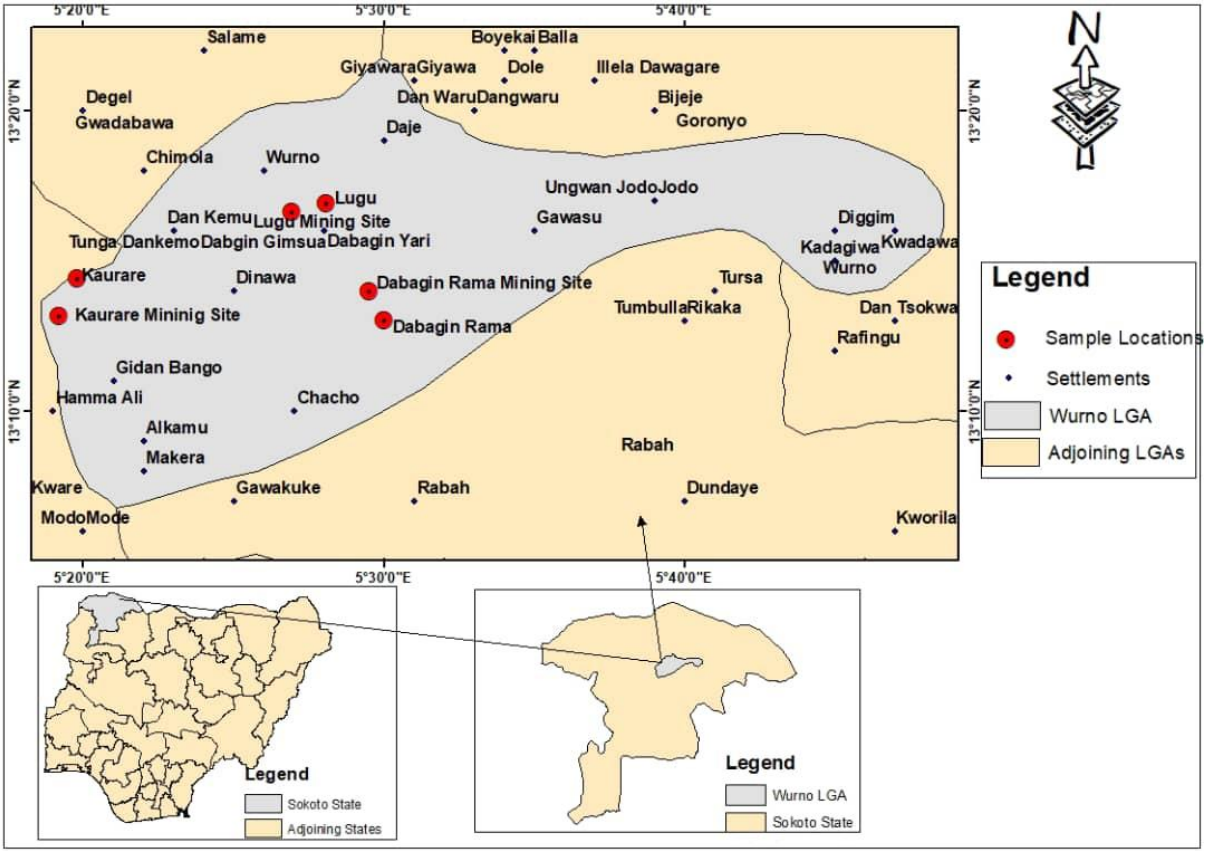

Figure 1: Map of Wurno showing the sampling points.

HpGe Gamma- Ray Spectrometer

A coaxial p-type high-purity germanium (HpGe) detector with 80% relative efficiency from Canberra (Model: GC 8023; Serial Number: 9744) was used to analyze the samples. This was coupled to an analogue-to-digital converter (ADC) and a multichannel analyzer, which interfaced with a personal computer-based data acquisition system running Genie 2000 (VI.3) software from Canberra. The analysis was conducted at the National Institute of Radiation Protection and Research (NNRA) located at the University of Ibadan, Nigeria.

System Calibration

Energy calibration was performed to align channel numbers with the corresponding gamma-ray peak energies in keV. Efficiency

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calibration was conducted to evaluate the detector's efficiency across a spectrum of different energies. The energy calibration utilized gamma-emitting sources including isotopes of ^{241}Am, ^{137}Cs and ^{60}Co at 59.54 keV, 661.66 keV and 1173.24/1332.49 keV respectively, with spectra acquired using the HPGe detector over a 1000-second period. A linear curve was derived by analyzing channel numbers against gamma-ray energies. Data storage was managed through the multichannel analyzer memory. Employing a blended 0.65kg Canberra reference soil in a Marinelli beaker comprising ^{125}Sb, ^{155}Eu, ^{54}Mn, ^{65}Zn and ^{40}K, the efficiency calibration was performed with energy ranging from 35.5keV to 1460.8 keV.

Determination of Background Activity

The background intensity for each spectrum was determined using the relationship below (Righi et al., 2009);

$$I_g - I_b = I_n \quad (3)$$

Here, I_g represents the gross spectral measurement, I_b is the background radiation measurement, and I_n is the net spectral measurement for the analyzed samples.

Radioactivity Assessment

The activity concentration of ^{226}Ra was determined using the energy lines at 295.2–351.9 keV from ^{214}Pb and 609.3–1238.1 keV from ^{214}Bi. For ^{232}Th, the activity was calculated using gamma lines at 300.1 keV from ^{212}Pb, 277.4–860.6 keV from ^{208}Ti, 209.3–911.1 keV from ^{228}Ac, and 723.3–785.3 keV from ^{212}Bi. The activity concentration of ^{40}K was directly assessed from its 1460.8 keV gamma line. The radionuclide activity concentration was calculated using the formula from Mustapha et al. (1999):

$$\frac{A_R M_R}{I_R} = \frac{A_S M_S}{I_S} \quad (4)$$

Where A_R, M_R, and I_R respectively denote the activity concentration, mass, and net spectral measurement of the radionuclide contained in the standard sample. A_S, M_S, and I_S respectively refer to the activity concentration, mass, and net spectral measurement of the radionuclide contained in the analysed sample.

RESULTS AND DISCUSSION

The activity concentration of radionuclides ^{226}Ra, ^{232}Th and ^{40}K in soil samples were measured using the HPGe detector were tabulated in Table 2. The activity concentrations for ^{226}Ra, and ^{232}Th were found to exceed the global average of 35 Bqkg^{-1} and 45Bqkg^{-1} respectively. However, they remained within the global average of 420 Bqkg^{-1} for ^{40}K, as stated in UNSCEAR (2016).

Table 2: Measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in this work.

Sample point	Activity Concentration (Bq/kg)			
	Ra-226	Th- 232	k-40	Ra_{eq}
1	176.32 ± 9.13	40.22 ± 2.11	55.04 ± 2.93	238.073±12.37
2	209.15± 10.81	60.07 ± 3.17	120.33 ± 6.37	304.316±11.97
3	155.19 ± 8.03	34.05 ± 1.80	47.92 ± 2.55	207.571±10.8
4	264.15 ±13.65	77.94 ± 4.10	206.60±10.93	391.512±20.35
5	130.87 ± 6.79	25.36 ± 1.35	34.02 ± 1.83	169.754±8.86
6	183.41 ± 9.50	75.79 ± 3.99	206.43±10.92	307.685±16.04
Average	186.515±9.65	52.238±2.75	111.723±5.93	270.033±13.62
Max	264.15±13.65	77.94±4.1	206.6±10.93	411.867
Min	130.87±6.79	25.36±1.35	34.02±1.83	160.893

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The present work has revealed a noteworthy phenomenon of high radium (^{226}Ra) activity concentration in the study areas of Wurno, Sokoto State. The elevated radium level can be primarily attributed to the presence of phosphate rocks in the geological composition of the local government area. (Isah et al; 2022) Phosphate rocks often contain higher concentrations of uranium and its decay products, including radium (IAEA, 2014). Furthermore, the activity concentration for ^{40}K is higher than that of ^{232}Th but lower than that of ^{226}Ra in all the samples from nearby settlements except for sample point 5. The high activity concentrations of ^{226}Ra and ^{40}K clearly explain that the major sources of gamma radiation in the location are ^{226}Ra and ^{40}K

Table 3: Absorbed dose rate (D), Annual effective dose (E), Hazard indices (H_{ex} & H_{in}), Annual gonadal dose equivalent (AGDE) and Excess lifetime cancer risk (ELCR) measured in this work.

Sample point	Radiation Hazard Parameters					
	D (nGy/h)	E (mSv/y)	H_{ex}	H_{in}	AGDE(μSv/y)	ELCR
1	108.048±5.62	0.133±0.01	0.643±0.034	1.12±0.058	730.231±37.95	0.464
2	137.927±5.54	0.169±0.007	0.822±0.032	1.387±0.061	935.15±37.37	0.592
3	94.262±4.90	0.116±0.006	0.561±0.029	0.980±0.051	636.913±33.14	0.405
4	177.728±9.24	0.218±0.011	1.058±0.055	1.772±0.092	1206.885±62.75	0.763
5	77.198±4.03	0.095±0.005	0.459±0.024	0.812±0.042	521.075±27.20	0.331
6	139.121±7.23	0.171±0.009	0.831±0.043	1.327±0.069	948.358±49.45	0.597
Average	122.472±6.19	0.150±0.007	0.73±0.037	1.234±0.063	830.397±41.94	0.526
Max	186.967	0.229	1.113	1.864	1269.634	0.803
Min	73.169	0.090	0.435	0.770	493.877	0.314

Radium Activity Equivalent (Ra_{eq}) Evaluation

The formula provided by Mohamed et al. (2016) was used to calculate the radium equivalent activity Ra_{eq} as:

$$Ra_{eq}\ (Bq/kg) = A_{Ra\text{-}226} + \frac{10}{7}A_{Th\text{-}232} + \frac{10}{130}A_{K\text{-}40} \quad (5)$$

Here, $A_{Ra\text{-}226}$, $A_{Th\text{-}232}$ in and $A_{K\text{-}40}$ represent the concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides respectively. The Ra_{eq} values ranged from 160.893 Bq/kg to 411.867 Bq/kg, with an average of 270.033±13.62 Bq/kg, exceeding the global average of 89 $Bqkg^{-1}$ but remaining below the UNSCEAR suggested limit of 370 $Bqkg^{-1}$ (UNSCEAR, 2016).

Absorbed gamma radiation dose rate

The absorbed gamma dose rate in air, measured at 1 meter above ground level, ranged from 73.169 nGy/h to 186.967 nGy/h, with an average of 122.472±6.19 nGy/h. The calculation followed the equation from Mohamed et al. (2016):

$$D\ (nGyh^{-1}) = 0.462A_{Ra} + 0.6044A_{TH} + 0.0417A_{K} \quad (6)$$

Where $A_{Ra,}$ A_{Th} and A_K represent the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The average value exceeds the global average of 60 nGy/h (UNSCEAR, 2016), indicating that the radiation dose in the study area is higher than in most regions worldwide.

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Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent (AEDE) for outdoor exposure ranged from 0.090 mSv/y to 0.229 mSv/y, with a mean value of 0.150±0.007 mSv/y. This was calculated using the (7) equation from UNSCEAR (2016):

$$E\ (msvy^{-1}) = T.Q.D \times 10^{-6} \quad (7)$$

Where D is the absorbed dose rate, Q is the conversion factor (0.7×0.2 Sv/Gy), 0.20 is the outdoor occupancy factor, and T represents the total outdoor occupancy time (8760 hours/y). The average value is below the global average of 1 mSv/y (Mohamed et al., 2016).

External Hazard Index

The external hazard index (H_{ex}) evaluates the radiological risk posed by natural radionuclides ^{40}K, ^{226}Ra, and ^{232}Th in the environment. It was calculated using the formula from Mohamed et al. (2016):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{TH}}{259} + \frac{A_K}{4810} \quad (8)$$

The results ranged from 0.435 to 1.113, with an average of 0.73±0.037, remaining below the UNSCEAR recommended value of 1 (UNSCEAR, 2016).

Internal Hazard Index

To assess the internal exposure risk from radon and its progeny, the internal hazard index (H_{in}) was calculated using the formula from Mohamed et al. (2016):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{TH}}{259} + \frac{A_K}{4810} \quad (9)$$

The values ranged from 0.770 to 1.864, with an average of 1.234±0.063, exceeding the recommended value of 1.

Annual Gonadal Dose Equivalent

The annual gonadal dose equivalent (AGDE), which quantifies the genetic impact of radiation on reproductive organs, was estimated using the UNSCEAR (2000) formula:

$$AGDE\ (\mu Svy^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (10)$$

The average AGDE was 830.397±41.94 µSv/y, significantly exceeding the permissible limit of 300 µSv/y, indicating a notable radiological risk.

Excess Lifetime Cancer Risk (ELCR)

The excess lifetime cancer risk (ELCR) was calculated to estimate the likelihood of cancer development from prolonged gamma-ray exposure, using the equation from Taskin et al. (2009):

$$ELCR = AEDE \times DL \times RF \quad (11)$$

Where AEDE is the annual effective dose equivalent, DL is the average lifespan (70 years), and RF is the risk factor (0.05 per Sievert). The ELCR values ranged from 0.314×10^{-3} to 0.803×10^{-3}, with a mean value of 0.526×10^{-3}, exceeding the international benchmark of 0.29×10^{-3}.

CONCLUSION

The sampling sites registered a mean activity concentration above the allowable limit for Radium and thorium and a mean absorbed gamma dose rate that was twice above the allowable limit of 60nGy/h. The average external radiation hazard index (H_{ex}) was determined to be 0.73±0.037, while the average internal radiation hazard index (H_{in}) was 1.234±0.063, exceeding the value of unity. Mean value for excess lifetime cancer risk is about twice the allowable limit of 0.29×10^{-3}. The samples describe that the radiological contamination of the study location may pose a significant radiological threat to the miners and the population. The observed high radium activity concentration in the selected places of Wurno, Sokoto State can be attributed to the presence of phosphate rocks in the region. Public awareness regarding the potential health risks associated with elevated radionuclide

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concentrations, particularly radium should be raised. This will help to promote understanding and adoption of appropriate radiation protection measures especially the miners, such as minimizing the amount of time spent in areas with elevated radiation levels, use of protective clothing and so on.

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