

Original Research Article

Measurement of radioactivity concentration in Pegmatite Rocks samples and dose assessment from Wamba in North Central Nigeria

ABSTRACT

Aims: To evaluate the radiological impact parameters on mine workers.

Study design: The research work was carried out by using a gamma – ray spectrometer with a NaI (TI) detector.

Place and Duration of Study: Centre for Energy and Research development (CERD), Obafemi Awolowo University, Ile- Ife Osun State, Nigeria between September 2016 and June 2017.

Methodology: The activity concentrations of ^{238}U , ^{232}Th and ^{40}K were determined in ten samples which were used to evaluate the absorbed dose rate, radium equivalent activity, external and internal hazard indices, representative level index and annual effective dose equivalent.

Results: The average activity concentrations of ^{238}U ($24068.11\text{Bq kg}^{-1}$), ^{232}Th (387.72Bq kg^{-1}) and ^{40}K (9509.24Bq kg^{-1}) **were higher than the recommended world values 35Bq kg^{-1} , 30Bq kg^{-1} and 400Bq kg^{-1} by UNSCEAR.** with the mean value of absorbed dose rate $11720.8\text{ nGy h}^{-1}$. **The mean values of radium equivalent activity, external hazard index (H_{ex}) and internal hazard index (H_{in}), representative level index and annual effective dose equivalent for the area under study were determined as $25346.82\text{ Bq kg}^{-1}$, 68.52 , 135.43 and 357.9 and $14374.36\text{ }\mu\text{Sv y}^{-1}$ respectively.** This study revealed that all the radiological parameters were higher than their respective recommended world average values.

Conclusion: With the high concentrations obtained in this study, it is therefore concluded that this may pose a serious health risk to the miners and the general public. It is therefore advice that necessary guidelines should be provided for exploration of minerals in this mining site.

Keywords: Mining; Radiological protection; spectrometer; pegmatite; Absorbed dose rate

1. INTRODUCTION

Human exposure to radiation can be from either natural or artificial radioactivity. Activities like mining of natural resources such as uranium and other minerals expose a lot of people to dangers which has generated a need for regulations [1, 2]. The natural occurring radionuclide materials (NORMs) has been in existence since human creation. This has been observed to have spread across the earth crust with presence of many radioactive elements such as Uranium (U) and Thorium (Th) which are referred to as parent's radioisotopes and Potassium (K) which is also seen in geological formations of soils, rocks, plants, water and air [3 - 5]. The level of radiation exposure can be determined by the type of rock that is mined. Higher radioactivity are observed to be mostly present in igneous rocks such as granite and pegmatite while lower levels of radioactivity are associated with sedimentary rocks with exceptions of Shale and Phosphate rocks **having relatively higher amount of radioisotopes** [6]. The level of radionuclides in mined rocks helps to know the extent of radioactive pollutant infused to the surroundings [7]. Therefore, the measurement of radioisotopes in rock samples before use is very important in order to prevent radiation exposure.

Several researches have been conducted to estimate natural radiation exposure and likely radiological hazards on people [8 -13]. Some of these studies have established that accumulation of ingested radionuclides through food, water and inhalation of tiny particles could have serious health repercussions like leukemia, bone cancer and high blood pressure [14, 15].

Other studies also revealed that the activity concentrations are not particular to a kind of rock alone but the release of these radionuclides overtime causes cancer and other related health hazards which can later damage some essential organs in the body [16 - 19]. According to [20], it was revealed that in an environment of uranium operational mining site, 68.33 % of the people living around this site do die before age 62.

Presently exploration of minerals in the area under study are done by small scale miners whose activities are dangerous and are not controlled by any regulated bodies. In that case, the illegal mining currently going on in the area reveals that the Wamba pegmatites may be of economic importance. It is therefore necessary to unveil more informations on the natural resources so that their economic mineral potential can be assessed.

This study focused on the extent of exposure of the miners to natural occurring radionuclides materials (NORMs) by evaluating the radiological parameters and compared with other literatures and with the United Nations Scientific Committee on Atomic Radiation Report [7] average values.

2. MATERIAL AND METHODS

2.1 Study area

Wamba town is located mid - northern part of Nasarawa State, Nigeria (**Figure 1**) between Longitudes $8^{\circ} 30'E$ and $8^{\circ} 40'E$ and Latitudes $9^{\circ} 00'N$ and $9^{\circ} 07' N$. It has an area of 1,156 km² and a population size of 72,894 people [21]. The pegmatite field of Wamba is one of the Pan- African rare - metal pegmatite broad belt which stretches from southwestern to central Nigeria [22].

It is found covering about 45 km² within the Pan-African reactivation zone. The exploration of minerals in this area had started as early as 1912 with tin - bearing pegmatites in the 40 km of Wamba area and ceased in the 1960s. The study area is characterized by hosting several hills with an average temperature of 31° C. Due to the peculiarity of this area mining activities is predominate. Therefore collection of pegmatite rock samples was done randomly to survey the natural radiation in the mining area. A total of ten samples were collected for radiation measurement and absorbed dose rate.

2.2 Sampling

The total of 10 pegmatite samples were collected randomly from the mentioned study area were left to dry for some days under a thorough laboratory conditions in order to remove any liquid present. These rock samples were pulverized in a Rocklab milling machine (Model C RC3E, Serial number 1288) situated at the Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile- Ife Osun State, Nigeria. The samples were oven dried at 100 °C for 24 hours to ensure that is free from humidity. The samples were sieved with a 2 mm mesh and thereafter, 150 g of each sample was packed into a cylindrical plastic container tightly closed, sealed and stored for 28 days to ensure secular equilibrium between the parent radionuclides ²²⁶Ra, ²³²Th and their respective daughters [23]. An identical empty plastic container was sealed to serve for background counting.

2.3 Instrumentations

The activity measurement was carried using a well calibrated **Sodium Iodide (NaI(Tl)) detector with its photomultiplier** at the Centre for Energy and Research development (CERD), Obafemi Awolowo University, Ile- Ife Osun State, Nigeria. The detector is shielded with a lead castle thickness of 5 cm and the resolution of about 8% at energy of 0.662 MeV (¹³⁷Cs). The preamplifier provided a voltage 750 V to the detector. The multichannel analyzer (MCA) connected through a preamplifier provides a stabilised spectrum which can be analyzed on an IBM personal computer. The energy calibration of the detector was done using a standard source which was shown in **Table 1**, with known gamma-ray energies prepared by the Isotope Products Laboratories, California, U.S.A.. The calibration was performed by determining the sources of known radionuclides with energy range **from 60 keV to 1836 keV**. This was achieved by measuring the **spectra point sources emitting known gamma-ray energies (¹³⁷Cs) 661.66 keV, (⁶⁰Co) 1173.24 and 1332.50 keV** with channel numbers of 738, 1504 and 1743 respectively. The standard sources have the same geometry with that of the sample. The gamma counting was done for 18000 s.

In order to quantify the radionuclides present in each sample, determination of the efficiency of the detector must be done. The efficiency measured the performance of the detector when the source was placed at 25 cm from the detector crystal. The efficiency of a detector measures number of pulses that occur for a given number of incident gamma rays, i.e., the fraction of all the photons that are emitted by the source or sample, which cause an event in the detector. This depends on the following: the detector features, counting geometry used, the sample shape, volume and distance of the sample to the detector.

As soon as the counting time started, the computer started recording the photo peaks and the same time storing them. The accumulation of counts at a specific energy range are due to emitted photons from the sample in addition to the one from background interference. However, if the background is constant, then a sample with high activity will accumulate relatively more counts in the peak compared to the background. Therefore, as the total area of the peak increases, the relative background size reduces as well as the proportional

background error. After the counting elapsed, the peaks were displayed. The radioactivity analysis was carried out by spectra- analysis program, SAMPO 90, which was used to analyze the samples on the computer. The spectrum of each sample was analyzed and the identification of radionuclides were determined from the energy of the peak centroids. Also as a sample activity increases, the estimated standard error decreases, and as the activity decreases, the relative error increases. An empty container of the same geometry was sealed and counted for gamma ray distribution count. This served as a background measurement which was done repeatedly at regular intervals for quality control. The spectrum obtained was recorded for analysis.

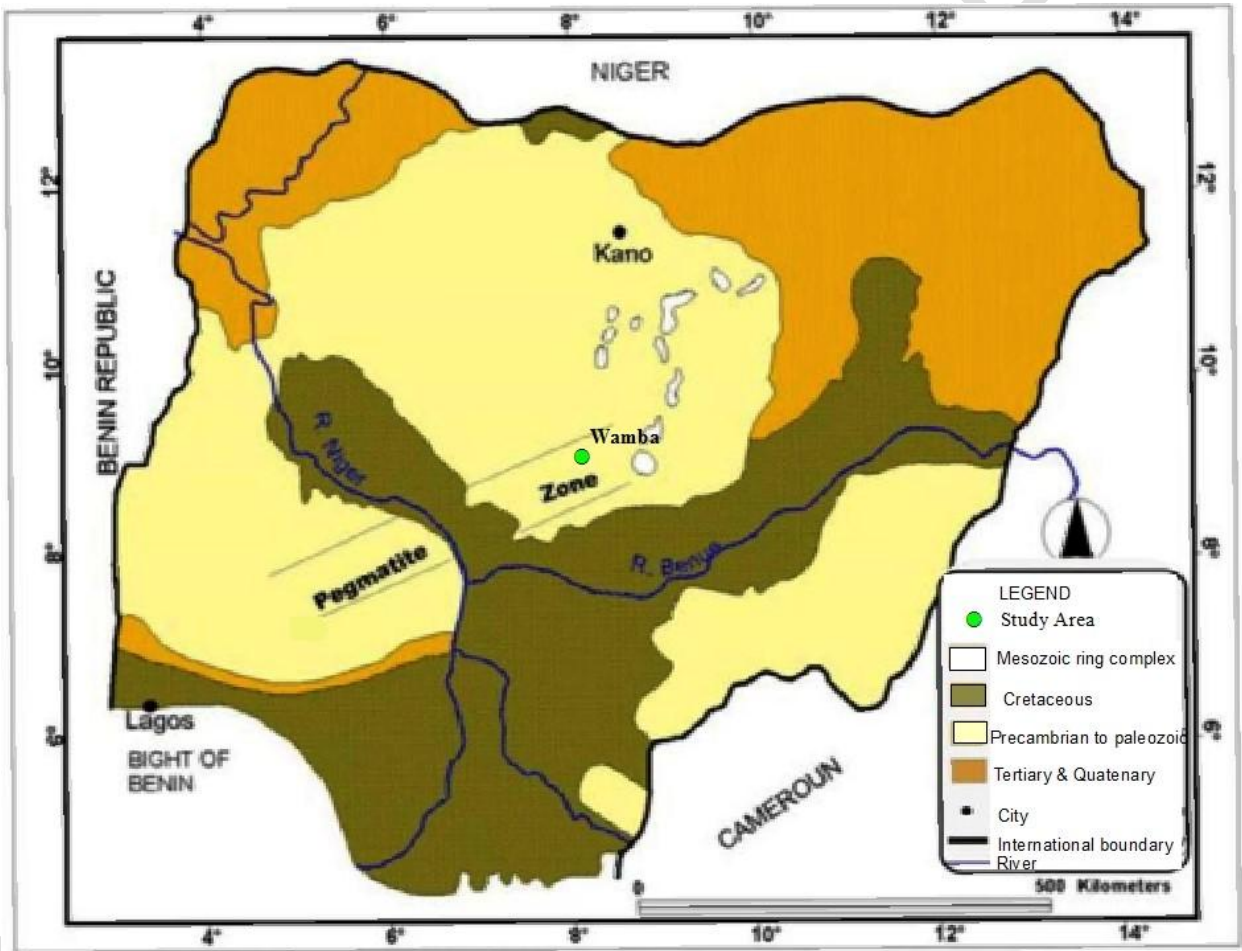


Figure 1: General Geology of Nigeria showing the Location of the Pegmatite Zone and the Study Area [24]

Table 1: Radionuclides used for Energy Calibration of NaI (TI)

Radionuclides	Energy (keV)	Channel number
¹³⁷ Cs	661.66	738
⁶⁰ Co	1173.24	1504
⁶⁰ Co	1332.50	1743

In **Table 2**, the activity concentration of the daughter radionuclide (²²⁶Ra) in the samples was obtained using the γ -ray peaks of 1120 keV from ²¹⁴Bi which is equivalent to the concentration of (²³⁸U) in the samples. The daughter radionuclide ²²⁸Ac was determined from its 911 keV γ -ray peak which was chosen as an indicator of ²³²Th. The concentration of ⁴⁰K was determined by measuring the photo peak of 1460 keV emitted during its decay.

Table 2: Spectral Energy Windows used in Analysis using NaI(Tl) Gamma Spectroscopy

Element analysed	Isotope Used	Energy (keV)
²³² Th	²²⁸ Ac	911
²³⁸ U	²¹⁴ Bi	1120
⁴⁰ K	⁴⁰ K	1460

The background radioactivity distribution in the environment around the detector was determined by counting the empty sealed container for 18000 s. The background measurements were repeated at regular intervals for quality control. The spectrum obtained was recorded for analysis. The specific activity concentration of the natural radionuclide **A** ($Bq\ kg^{-1}$) of each sample in this study was evaluated using Eq. 1 [25, 26].

$$A = \frac{C}{p_w t \varepsilon} \quad (1)$$

where **C** is the net count above the background, **p** is the absolute gamma ray emission probability of the radionuclide being measured, **w** is the net dry sample weight (kg), **t** is the sample measurement time (18000 s) and ε is the absolute efficiency of the detector.

The specific activity concentration (**C_s**) of the radionuclide was obtained by direct comparison with the same radionuclide in a given standard. The two activities, i.e. the sample and the standard are related by:

$$C_x = C_s \frac{m_s(A_x - A_b)}{m_x(A_s - A_b)} \quad (2)$$

These values obtained were compared in order to test for accuracy of the results obtained for specific activity concentration of the radioisotopes. The direct comparison method gives the same radionuclide in a given standard.

A_x , A_s and A_b are the areas of sample, standard and background respectively; m_x and m_s are the respective masses of sample and standard, while C_x and C_s are specific activity concentrations of sample and standard respectively.

The concentration of natural radionuclides to the absorbed dose rate in the air (D_R) depends on the concentration of the radionuclides present in the pegmatite rock samples. Absorbed dose radiation dose in an exposed person per unit mass at 1m above the ground level is define as the quantity of ionizing energy present in the body in **Gay/second....(Gy/s)**. This was determined using Equation 3.

$$D_R(nGy h^{-1}) = 0.462 C_U + 0.604 C_{Th} + 0.0417 C_K \quad 3$$

where C_U , C_{Th} and C_K , are the **activity concentrations** of ^{238}U , ^{232}Th and ^{40}K , in the rock samples ($BqKg^{-1}$) respectively.

2.4 Evaluation of Radiological Impact Parameters

2.4.1 Radium Equivalent Activity (Ra_{eq})

The radium equivalent activity ($Bq kg^{-1}$) represents a weighted sum of activities of ^{238}U , ^{232}Th and ^{40}K . Ra_{eq} is related to the external gamma dose and internal dose due to radon and its daughters. In this study Ra_{eq} was calculated using the equation 4 [27].

$$Ra_{eq} = C_U + 1.43 C_{Th} + 0.077 C_k \quad 4$$

where C_U , C_{Th} and C_K , are the **activity concentrations** of ^{238}U , ^{232}Th and ^{40}K , in the rock samples ($Bq kg^{-1}$) respectively.

2.4.2 External Hazard index (H_{ex})

The external hazard index (H_{ex}) connotes the level of health hazard incurred from gamma radiation that comes from the materials used in building walls of a dwelling. Equation 5 was used in calculating for external hazard index [7].

$$H_{ext} = \left(\frac{C_U}{370}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \quad 5$$

where, C_k , C_U and C_{Th} are the **activity concentrations** of ^{238}U , ^{232}Th and ^{40}K ($Bq kg^{-1}$) in respectively.

2.4.3 Internal Hazard index (H_{in})

The internal hazard index (H_{in}) also defined as the inhalation exposure of the respiratory organs to radon and its daughter nuclei inside a house was calculated using Equation 6 [7].

$$H_{int} = \left(\frac{C_U}{185}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \quad 6$$

C_U , C_{Th} and C_K are the **activity concentrations** in $Bq kg^{-1}$ of ^{238}U , ^{232}Th and ^{40}K respectively.

2.4.4 Representative level index (I_γ)

To ascertain whether a particular sample conform to the standard dose limit set for building materials [28], the representative level index or gamma index (I_γ) is used to estimate the level of γ – radiation hazard associated with the natural radionuclides in specific investigated samples. Equation 7 was used to evaluate the gamma index for samples in this study.

$$I_\gamma = 0.0067C_U + 0.01C_{Th} + 0.00067C_K \quad (7)$$

C_U , C_{Th} and C_K are the **activity concentrations** in $Bq\ kg^{-1}$ of ^{238}U , ^{232}Th and ^{40}K respectively. Therefore, (I_γ) must be ≤ 1 which also corresponds to an **annual effective dose of $\leq 1mS$**

2.4.5 Annual Effective Dose Rate (AEDR)

This is the dose that an individual receives after being exposed to radiation outside especially through air. The absorbed doses evaluated in $nGy\ h^{-1}$ were converted to annual effective dose in $\mu Sv\ y^{-1}$ by [7]. This was calculated using equation 8.

$$AED_{Routdoor}(\mu Sv\ y^{-1}) = D_R(nGy\ h^{-1}) \times 8760\ (h\ y^{-1}) \times 0.7\ (Sv\ Gy^{-1}) \times 0.2 \times 10^{-3} \quad (8)$$

where D_R is the absorbed dose rate in $nGy\ h^{-1}$, conversion factor of $0.7SvGy^{-1}$ is the dose conversion factor ($Sv\ Gy^{-1}$), 0.2 is the outdoor occupancy factor and 8760 is the time conversion factor ($h\ y^{-1}$).

3. RESULTS AND DISCUSSION

[The daughter lines for ^{238}U and ^{232}Th used in this study were ^{214}Bi and ^{228}Ac respectively. The results of the radioactivity concentrations were presented in **Table 3**. It was revealed that all the three radionuclides ^{238}U , ^{232}Th and ^{40}K investigated for in the pegmatite rock samples were observed to have the highest activity concentrations in sample 1 and lowest concentrations in samples 6. The mean activity concentrations of ^{238}U measured was $24068.11 \pm 368.03\ Bq\ Kg^{-1}$ which is higher than the world average value of $35\ Bq\ kg^{-1}$. The mean activity concentration of ^{232}Th estimated in this research was $387.72 \pm 18.50\ Bq\ kg^{-1}$, and thus higher than the world average value of $30\ Bq\ kg^{-1}$. The average activity concentration of ^{40}K was $9509.24 \pm 335.11\ Bq\ kg^{-1}$ which is higher than the world average value of $400\ Bq\ kg^{-1}$. **The high activity concentration values in the samples could be as a result of high concentration of uranium present in the pegmatite rock samples.** Both the ^{238}U and ^{40}K contributed to the increase in the activity concentration in the pegmatite rock samples.

Table 3: Specific activity of ^{238}U , ^{232}Th and ^{40}K in Pegmatite Rock Samples in $Bq\ kg^{-1}$

Samples	^{238}U	^{232}Th	^{40}K
K1	716.42±11.56	7.98±0.39	650.70±31.83
K2	27755.55±424.28	418.58±19.96	10473.55±12.27
K3	30028.46±459.02	842.49±40.18	16726.56±818.12
K4	5825.98±89.17	82.80±3.96	1627.15±79.60
K5	6616.20±101.24	92.80±4.44	3354.09±164.06
K6	65809.11±1005.94	962.84±45.91	22015.65±1076.81
K7	1950.28±30.10	25.36±1.22	1313.38±64.24
K8	18036.72±275.82	200.65±9.58	6943.71±339.63
K9	38854.63±593.93	550.27±26.24	14765.44±722.20

K10	45087.76±689.23	693.45±33.07	17222.13±842.36
Min.	716.42±11.56-65	7.98±0.39- 962.84	650.70±31.83
Max.	65809.11±101.24	962.84±45.91	22015.65±1076.81
Mean value	24068.11±368.03	387.72±18.50	9509.24±335.11

Table 4 revealed the estimated absorbed dose rate (D_R) which ranged from 362.49 to 31888.00 $nGy h^{-1}$ with mean concentration value of 11720.77 $nGy h^{-1}$. The mean value of the absorbed dose rate was found to be higher than the world average value of 59 $nGy h^{-1}$ [7]. **This could be as result of large quantity of uranium concentration and other radionuclides that are present in the rock samples.** The Radium activity equivalent (Ra_{eq}) values obtained varied from 777.87 to 68878.11 $Bq kg^{-1}$ with mean value of 25346.82 $Bq kg^{-1}$. The Ra_{eq} mean value exceeded the maximum limit of 370 $Bq kg^{-1}$ recommended by [29]. The evaluated values of the external hazard index (H_{ex}) and internal hazard index for the pegmatite rock samples in this study ranged between from 2.11 to 186.16 with mean value of 68.52. The calculated values of (H_{in}) for the rock samples ranged from 4.04 to 364.02 with mean value of 135.43 as shown in Table 4. Both external and internal indices mean values were higher than the world average value of 1 unity [7].

The representative level index I_γ was calculated using equation in order for the radiation hazard to be negligible it must be less than unity. The evaluated values varied from 14.84 to 1340.49 with the average value of 357.95. The average value obtained is much greater than the world recommended value of 1 unity. The annual effective dose rate (AEDR) ranged from 444.5 to 39107.4 $\mu Sv y^{-1}$ with mean concentration of 14374.36 $\mu Sv y^{-1}$. The AEDR average value is thus higher than the recommended safety limit value of 70 $\mu Sv y^{-1}$ for an individual [7].

In **Table 5**, comparative analysis of the mean values of ^{238}U , ^{232}Th and ^{40}K for pegmatite rock in this study with some selected studies across the world was given. It was observed that the mean values of both ^{238}U and ^{40}K obtained in this study were higher than the values reported by all the authors (in **Table 5**). The mean value of ^{232}Th was higher than the values reported by [32,12] (Nigeria), [33,34] (Nigeria), [35] in Turkey and [36] (India) except [31] in (Nigeria) that was 507.2 $Bq kg^{-1}$. **The differences in the activity concentrations of these radionuclides when compared with other studies may be as a result non-evenly spread of the radio-elements in the earth crust [12]. Therefore, their concentration depend mostly on the local geology.**

Table 4: The evaluated of radium equivalent Ra_{eq} , dose rate, External hazard index (H_{ex}), Internal hazard index (H_{in}) and gamma index of the samples under investigation.

Sample code	Ra_{eq}	D	H_{ex}	H_{in}	I_γ	AEDR
K1	777.87	362.49	2.11	4.04	14.84	444.56
K2	29159.18	13277.76	78.82	153.83	570.45	16283.84
K3	32518.68	15067.80	87.89	169.05	620.15	18479.15
K4	6002.83	2808.32	16.41	32.15	118.43	3444.12
K5	7006.78	3250.25	18.93	55.38	135.49	3986.11
K6	68878.11	31888.00	186.16	364.02	1340.49	39107.44
K7	2087.54	970.20	5.63	10.90	40.14	1189.85
K8	18857.49	8738.84	50.96	99.70	367.37	10717.31
K9	40776.53	18888.58	110.20	215.22	792.44	23164.95
K10	47403.18	21955.50	128.12	249.98	920.17	26926.23
Min.	777.87	362.49	2.11	4.04	14.84	444.56

Max.	68878.11	31888.00	186.16	364.02	1340.49	39107.44
Mean value	25346.82	11720.77	68.52	135.43	357.95	14374.36

Table 5. Comparison of the mean activity concentration with some selected studies.

Sample used	²³⁸ U	²³² Th	⁴⁰ K	Place	Country	References
Soil	3.00	33.30	122.10	Sango-Ota	Nigeria	[30]
Soil	67.40	507.19	8217.77	Abua/Odua	Nigeria	[31]
Soil	32.52	56.23	403.96	Nasarawa	Nigeria	[32]
Rock	11.51	15.42	4441.06	Asa	Nigeria	[12]
Soil	11.90	17.72	70.44	Maiganga	Nigeria	[33]
Soil	12.14	23.23	270.14	Ile-Ife	Nigeria	[34]
Soil	24.50	51.80	344.90	Anatolia	Turkey	[35]
Rock	12.25	45.17	639.24	Aravali	India	[36]
Rock	24068.11	387.72	9509.24	Wamba	Nigeria	Present study

Why ,big values!!!

4. CONCLUSION

The mean activity concentrations of ²³⁸U (24068.1±368.0), ²³²Th (387.7±18.5), ⁴⁰K (9509.2±335.1) Bq kg⁻¹ and absorbed dose rate of 11720.77 nGy h⁻¹ were higher when compared to the United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations Sources and Effects of Ionizing Radiation values of 30 Bq kg⁻¹, 35 Bq Kg⁻¹, 400 Bq kg⁻¹ and 60 nGy h⁻¹, respectively. This may pose a serious health risk to the miners and the general public. The result of this study can serve as a baseline data for distribution of radioactivity present in pegmatite rock in the area and also allow more research work to be carried out to investigate radioactive background level of the area. This will provide necessary guidelines for fruitful exploration of pegmatite so that their economic potential can be of use to the country at large. The illegal mining currently going on could be curb to the barest minimum so that the general populace will not be prone to radiation hazard.

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