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Radioactivity Concentration Level of ^{238}U, ^{232}Th, and ^{40}K in Soil Samples of some Abandon Mining Sites in Nasarawa, Nasarawa State, Nigeria

Idris M. Mustapha[1*], *Sidi, M. Aliyu*[1], *Obiri G. Okara*[1], *Ibrahim A. Mohammad*[2], *Umar Adamu*[1], *Abdullahi A. Aisha*[1], *Abdullahi S. Halimatu*[3],

[1]Department of Physics, Nasarawa State University, Keffi, Nigeria.

[2]Department of Science Laboratory Technology, Nasarawa State University, Keffi, Nigeria.

[3]Department of Biochemistry and Molecular Biology, Nasarawa State University, Keffi, Nigeria.

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Abstract: A comprehensive study was conducted to determine the radioactivity concentration level in soil samples of some abandon mining sites in Nasarawa, Nasarawa State using sodium iodide-doped thallium gamma spectrometer. A total of ten soil samples were collected each from different locations in the study area. The soil samples were air-dried, sieved, and sealed in a plastic container. The samples were then analyzed using NaI(Tl) detector for γ-ray spectral measurements. The mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in the samples are 5.66 ± 1.291 $BqKg^{-1}$, 0.904 ±0.09$BqKg^{-1}$ and 277.62±21.6$BqKg^{-1}$ respectively. The concentration of ^{232}U and ^{232}Th in all the samples is lower than the word average of 30 Bq/kg and 35 Bq/kg respectively as recommended by regulatory bodies. The concentration of ^{40}K in S6, S8, and S9 is higher than the average standard of 400 Bq/kg as recommended by regulatory bodies. The absorbed dose rate, radium equivalent, external hazard indices, annual effective dose equivalent, are in the range (mean) of 1.070-29.37 nGy/h (14.74 nGy/h), 2.251- 55.63 Bq/kg (28.33 Bq/kg), 0.006- 0.0765 mSv/yr (0.1502 mSv/yr), 0.005-0.144 mSv/yr (0.072 mSv/yr), and 0.0018-0.504 µSv/yr (0.253 µSv/yr) respectively. The absorbed dose rate for the entire locations are lower than the average standard of 55 Bq/kg set by regulatory bodies. The radium equivalent activity for all locations is lower than the average standard of 370 Bq/kg as recommended by regulatory bodies. Similarly, the calculated external hazard index for all the sample is lower than the average standard of 1 Bq/kg recommended. The calculated annual effective dose equivalent for all the sample is lower than the recommended standard of 1 mSv/yr for public exposure. The calculated excess lifetime cancer risk in S6, S7, S8, S9, and S10 samples were higher than the acceptable safe limit of 0.29 x 10^{-3} recommended by regulatory bodies.

Keywords: Radioactivity concentration level, Abandon mining sites, NaI(Tl) detector, Annual effective dose equivaent and Excess lifetime cancer risk.

1 Introduction

Gamma radiation emitted from naturally occurring radioisotopes, such as ^{40}K and the radionuclides from the ^{232}Th and ^{238}U series and their decay products (also called terrestrial background radiation), which exist at trace levels in all ground formations, represents the main external source of irradiation to the human body [1, 2]. More specifically, natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [3- 7]. The specific levels of terrestrial environmental radiation are related to the geological composition of each lithologically separated area, and to the content in thorium (Th), uranium (U) and potassium (K) of the rock from which the soils originate in each area. In terms of natural radioactivity, it is well known, for instance, that igneous rocks of granitic composition are strongly enriched in Th and U (on an average 15 µg/g of Th and 5 µg/g of U), compared to rocks of basaltic or ultramafic composition (< 1 µg/g of U) [4, 5]. For that reason, higher radiation levels are associated with igneous rocks and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of those radionuclides [3, 6- 11].

From a geological point of view, the Nasarawa, which is located in the north central Nigeria (within the middle

*Corresponding author e-mail: idrismustapham@nsuk.edu.ng

Benue Trough), as the name infers the middle portion of the Nigerian Benue Trough. The Benue Trough itself is a rift basin in Central West Africa that extends NNE–SSW for about 800km in length and 150km in width. The Trough contains up to 6000M of Cretaceous Tertiary Sediment of which those pre–dating the mid–Santonian Compressional deformed [6]. The soil cover of the central area is highly alkaline, while the soils on the slopes, lower down, are covered by neutral sheeted diabase. The weathering of the sedimentary rocks (chalks, marls, etc.) in the foothills that fringe Troodos, gave rise to alkaline, calcium-rich soils. Most of these rock types belongs to the category of silica-oversaturated, which usually is associated with high Th and U elemental concentrations [7].

Rilwan et al. [7] carried out an assessment on gamma radiation from ^{232}Th, ^{226}Ra and ^{40}K in Nassarawa, Nigeria. Their research reported that the radiation dose of the study area is minimal and seems to have low exposure for the inhabitants in and around the contaminated areas. Rilwan et al. also in 2020 carried out the evaluation of the radiation hazard indices from some selected mining sites in Nasarawa West, using Sodium Iodide Thallium Gamma Spectrometry. The research also concluded that, there may not be serious radiological effects of these radionuclides (K, Th and U) to the populace.

Since few systematic data on environmental radioactivity in Nasarawa west were available, this work is piloted with the objective to systematically measure the terrestrial gamma radiation in the area, and determine its contribution to the annual effective dose equivalent to the population. The main feature that make this study particularly important and interesting to radiometric studies is that, it provides information on the analysis of selective elemental abundance (^{238}U, ^{232}Th, and ^{40}K).

2 Materials and Method

2.1 Study Area

The "Nasarawa Local Government", located in the West Senatorial Zone of Nasarawa State has over 200,000 workers grouped under approximately 12,000 independent people involved in Vehicle Repair & Maintenance, Metal working, Sale of engineering materials and automobile spare- parts. The major occupation of the people are farming and artisanal mining.

Sampling Technique

This study was conducted in March, 2022. A systematic sampling technique was used for the sample collection. This is a probability sampling method in which sample members from a larger population are selected according to a random starting point but with a fixed, periodic interval. This interval, called the sampling interval, is calculated by dividing the population size by the desired sample size.

Sample Collection

Ten sample locations which consisted of abandoned mining sites were visited to conduct a radiometry study. The samples were collected at a depth of 0.5 m from the surface of the soil at each abandoned mining sites visited. The samples were sealed in a well labelled polythene bags and enclose into one sack for easy transportation. Global Positioning System (GPS) was used to record the elevation and altitude of the area.

Sample Preparation

The samples collected were taken to the laboratory and left open for at least 24 hours to dry under ambient temperature. After drying, the samples were pulverized using a ceramic mortar and pestle, and were passed through a 5 mm-mesh sieve to remove the larger particles and collect the fine powder. The 300 g of the prepared samples were packed in a well-sealed cylindrical plastic container to prevent the escape of radon and were stored for at least 24 days to allow radium to attain equilibrium with the daughters. The samples were sent to the National Institute of Radiation Protection and Research, Ibadan Nigeria were Gamma spectrometry analysis of the samples was performed.

Method of Data Analysis

Evaluation of radiological hazard effects depending upon the activity concentration of primordial radioactive elements, various radiological hazards delivered to the surrounding living biota are calculated based on the following hazard parameters;

i. **Absorbed Dose Rate (D):** The total absorbed dose rate (D) in nGy/h is calculated using the following formula [12, 13]:

$$D\ (nGy/h) = 0.462\ A_U + 0.604\ A_{Th} + 0.0417\ A_K \qquad 1$$

where, A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Bqkg^{-1}.

ii. **Radium Equivalent Activity (Ra_{eq}) :** The Radium Equivalent Activity (Ra_{eq}) was calculated using [13];

$$Ra_{eq}\ (Bq/kg) = A_{Ra} + 1.43A_{Th} + 0.077A_K \qquad 2$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/kg).

iii. **External Hazard Indices (HI_{ex}):** The gamma ray radiation hazards due to the specified radioactive elements in soil samples are assessed by calculating the following two hazard indices using the relationship [14]:

$$HI_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \quad 3$$

where, A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Bq kg^{-1}. The recommended value by UNSCEAR [3] report for the hazard indices is less than unity.

iv. **Annual Effective Dose Equivalent (AEDE):** The annual effective dose equivalent (AEDE) in outdoor air is determined following UNSCEAR [3] as:

AEDE (mSv/y) = D (nGy/h) x 8760h x 0.2 x 0.7 Sv/Gy x 10^{-6} 4

where 8760 is the time in hours for one year, and 10^{-6} is the factor converting from nano to milli.

v. **Excess Lifetime Cancer Risk (ELCR):** Excess lifetime cancer risk (ELCR) is calculated using the formula [15-17]:

ELCR = AEDE x DL x RF 5

where, AEDE, DL, and RF are annual effective dose equivalent, duration of life (70 years) and risk factor (0.05 Sv^{-1}), respectively.

vi. **Annual Gonadal Dose Equivalent (AGDE):** Annual gonadal dose equivalent (AGDE) due to the specific activities of 238U, 232Th, and 40K is calculated using the formula [18, 19]:

AGDE (µSv/y) = 3.09 A_U +4.18A_{Th} + 0.314 A_K 6

3 Results and Discussion

Table 1 shows the experimental results obtained from the spectra of ten soil samples under investigation. For the effective computation of the experimental data, the absorbed dose rate, annual effective dose rate, external hazard index and internal hazard index, and excess lifetime cancer risk were calculated using equation 1 to 6 and the results were presented in Table 2. Chats have been plotted to compare the activity concentrations of A_U, A_{Th} and A_K as well as the radiological parameters with literature. In the charts, S1 –S10 represent the sampling location in the study area.

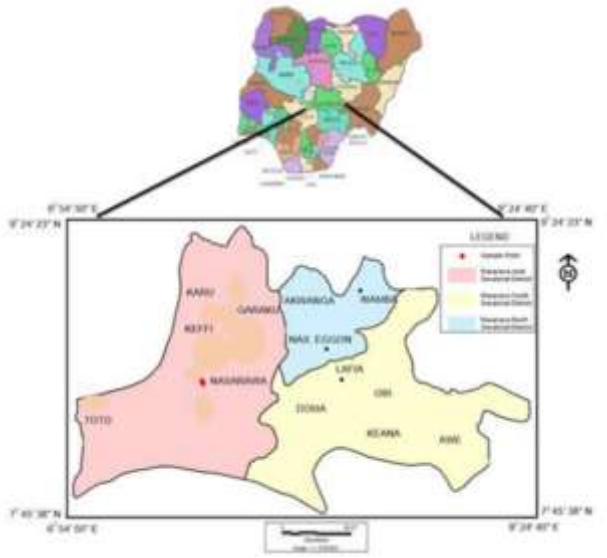

Fig.1: Map showing the location of the study area in Nigeria.

The raw result of activity concentration level of ^{238}U, ^{232}Th, and ^{40}K are shown Table 1 and Figure 3. The mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in the sediment sample are 5.66 ± 1.291 BqKg^{-1}, 0.904 ±0.09BqKg^{-1} and 277.62±21.6BqKg^{-1} respectively; shown in Table 1. The concentration of ^{40}K in S6, S8 and S9 is higher than the average standard of 400 Bq/kg as recommended by regulatory bodies. Figure 4 that the concentration of ^{232}U for all locations is lower than the average standard of 30 Bq/kg as recommended by regulatory bodies. It was observed that the concentrations of ^{232}Th in the samples are lower than the average standard of 35 Bq/kg as recommended by regulatory bodies.

The absorbed dose rate, radium equivalent, external hazard indices, annual effective dose equivalent, are in the range (mean) as 1.070-29.37 nGy/h (14.74 nGy/h), 2.251- 55.63 Bq/kg (28.33 Bq/kg), 0.006- 0.0765 mSv/yr (0.1502 mSv/yr), 0.005-0.144 mSv/yr (0.072 mSv/yr), and 0.0018-0.504 µSv/yr (0.253 µSv/yr) respectively. The absorbed dose rate for the entire locations is lower than the average standard of 55 Bq/kg set by regulatory bodies. The radium equivalent activity for all locations is lower than the average standard of 370 Bq/kg as recommended by regulatory bodies. Similarly, the calculated external hazard index for all the sample is lower than the average standard of 1 Bq/kg recommended. The calculated annual effective dose equivalent for all the sample is lower than the recommended standard of 1 mSv/yr for public exposure.

The calculated excess lifetime cancer risk in S6, S7, S8, S9, and S10 samples were higher than the acceptable safe limit of 0.29 x 10^{-3} recommended by regulatory bodies [20-22].

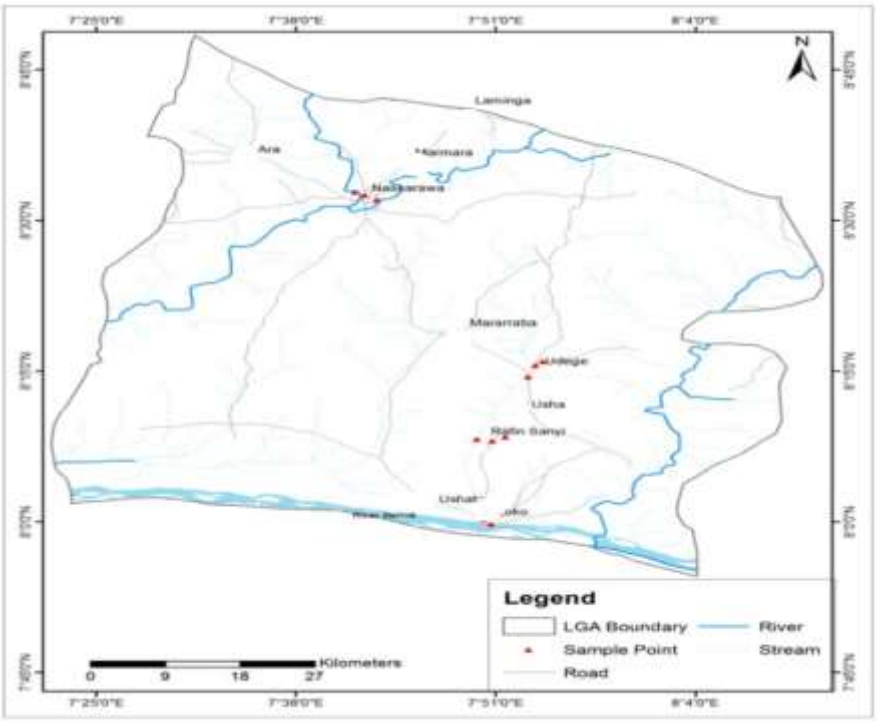

Fig. 2: Map of study locations.

Table 1: Raw result of radioactivity concentration levels in the samples.

Sample Code	Geopoint			Activity concentration (mSv/yr)		
	Longitude	Latitude		U-238	Th-232	K-40
S1	8°40’24.30’’ N	7°48’37.48’’E		0.35 ±0.08	1.30 ±0.120	48.92±3.740
S2	8°39’31.78’’ N	7°48’10.27’’E		2.26 ± 0.62	2.27 ± 0.24	69.60±6.500
S3	8°37’52.67’’ N	7°47’13.21’’E		8.40 ± 2.41	1.13 ± 0.120	126.07±12.0
S4	8°37’27.56’’ N	7°47’02.28’’E		1.26 ± 0.28	0.22 ± 0.02	90.74±6.910
S5	8°35’26.87’’ N	7°45’27.97’’E		0.39 ± 0.08	0.69 ± 0.06	11.35±0.890
S6	8°21’12.03’’ N	7°42’31.62’’E		9.38 ± 2.09	0.22 ± 0.020	574.24±43.2
S7	8°31’34.67’’ N	7°42’36.22’’E		9.51 ± 2.10	0.04 ± 0.00	332.3±26.11
S8	8°29’53.30’’ N	7°42’54.05’’E		3.88 ± 1.03	0.49 ± 0.05	553.61±42.5
S9	8°32’08.55’’ N	7°42’47.36’’E		5.80 ± 1.04	1.72 ± 0.18	615.22±46.4
S10	8°32’28.05’’ N	7°41’49.82’’E		15.41±3.18	0.96 ± 0.100	354.17±27.8
Mean				**5.66±1.291**	**0.904 ±0.09**	**277.62±21.6**
Min				**0.35 ± 0.08**	**0.04±0.000**	**11.35±0.890**
Max				**15.41±3.18**	**2.27± 0.240**	**615.22±46.4**

Table 2. Evaluated results for the radiological hazard parameters.

Sample Code	D (nGy/h)	Ra_{eq} (Bq/kg)	H_{ex} (mSv/yr)	AEDE (mSv/yr)	ELCR (µSv/yr)
S1	2.987	5.976	0.0161	0.015	0.051
S2	5.318	10.87	0.0293	0.026	0.091
S3	9.820	19.72	0.0533	0.048	0.169
S4	4.499	8.562	0.0231	0.022	0.077`
S5	1.070	2.251	0.0061	0.005	0.018
S6	28.41	53.91	0.1456	0.139	0.488
S7	18.27	35.15	0.0949	0.090	0.314
S8	25.17	47.21	0.1275	0.123	0.432
S9	29.37	55.63	0.1502	0.144	0.504
S10	22.47	44.05	0.1190	0.110	0.386
Mean	**14.74**	**28.33**	**0.0765**	**0.072**	**0.253**
Min	**1.070**	**2.251**	**0.0060**	**0.005**	**0.018**
Max	**29.37**	**55.63**	**0.1502**	**0.144**	**0.504**

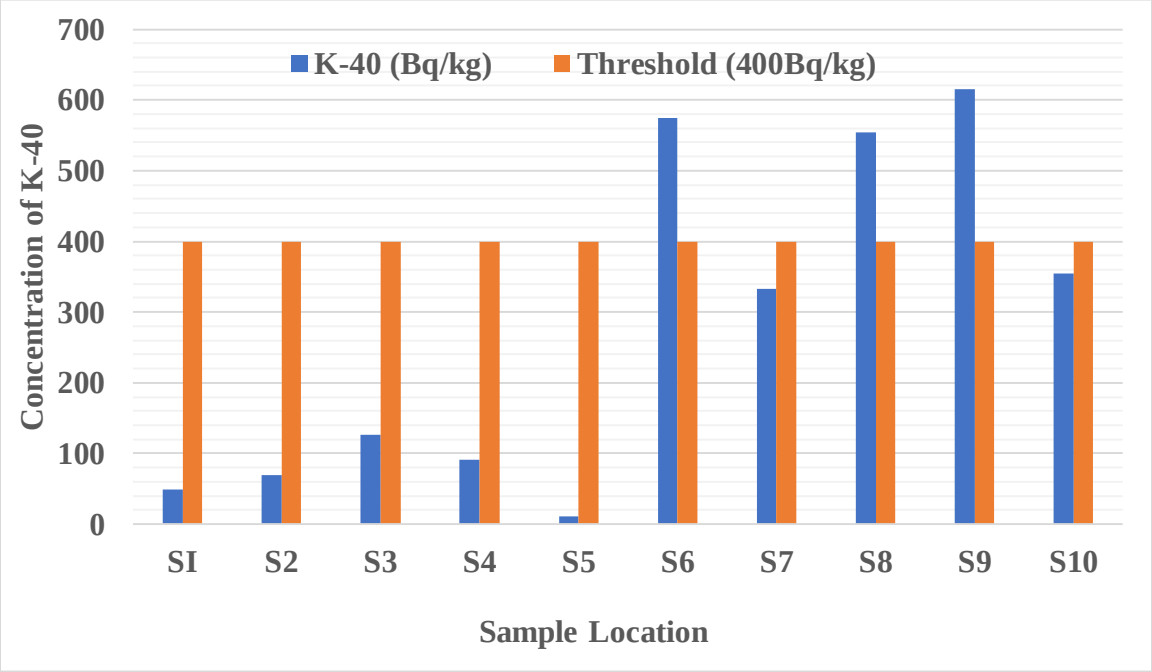

Fig. 3: Comparison of the activity concentration of K-40 with Threshold.

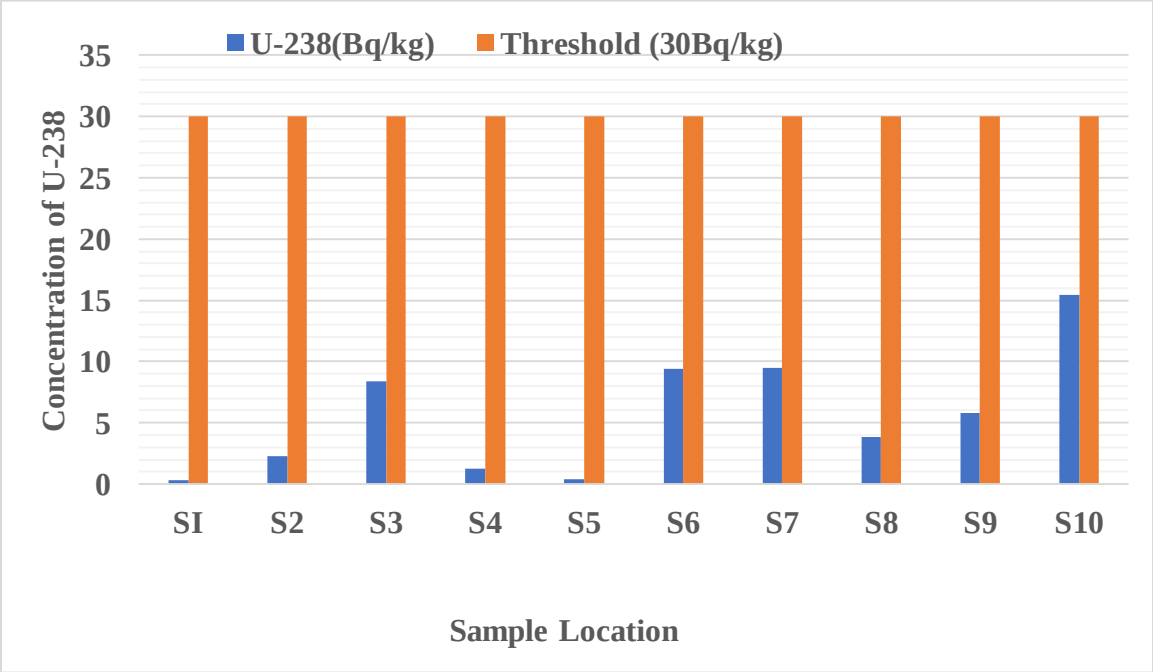

Fig. 4: Comparison of the activity concentration of U-238 with Threshold.

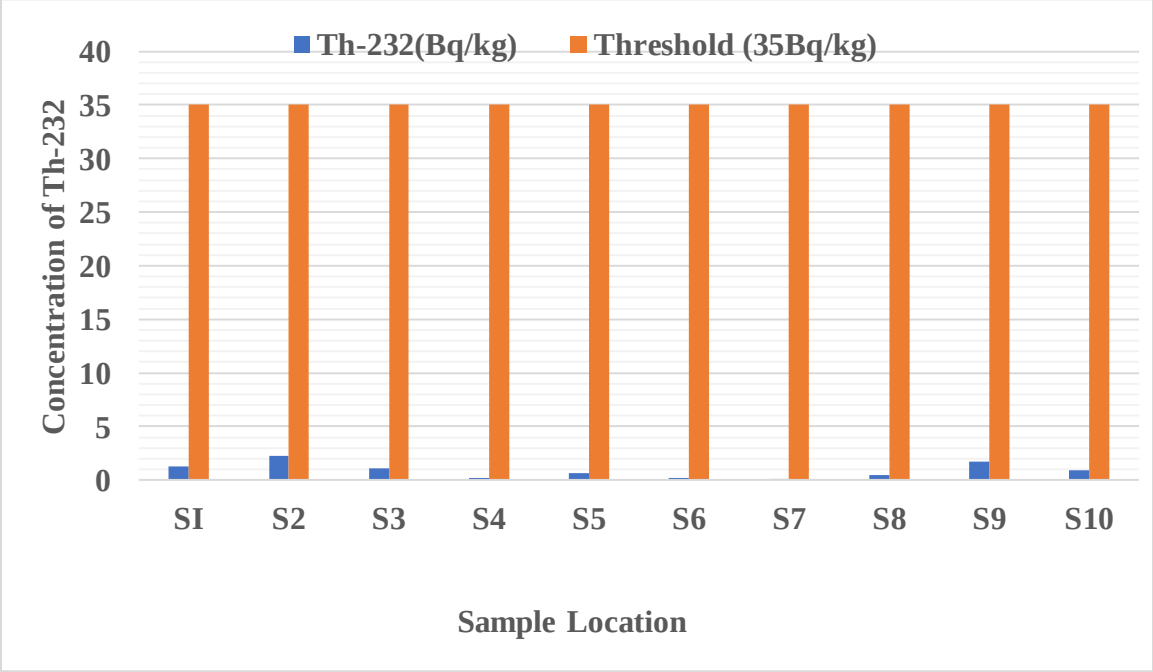

Fig. 5: Comparison of the activity concentration of Th-232 with Threshold.

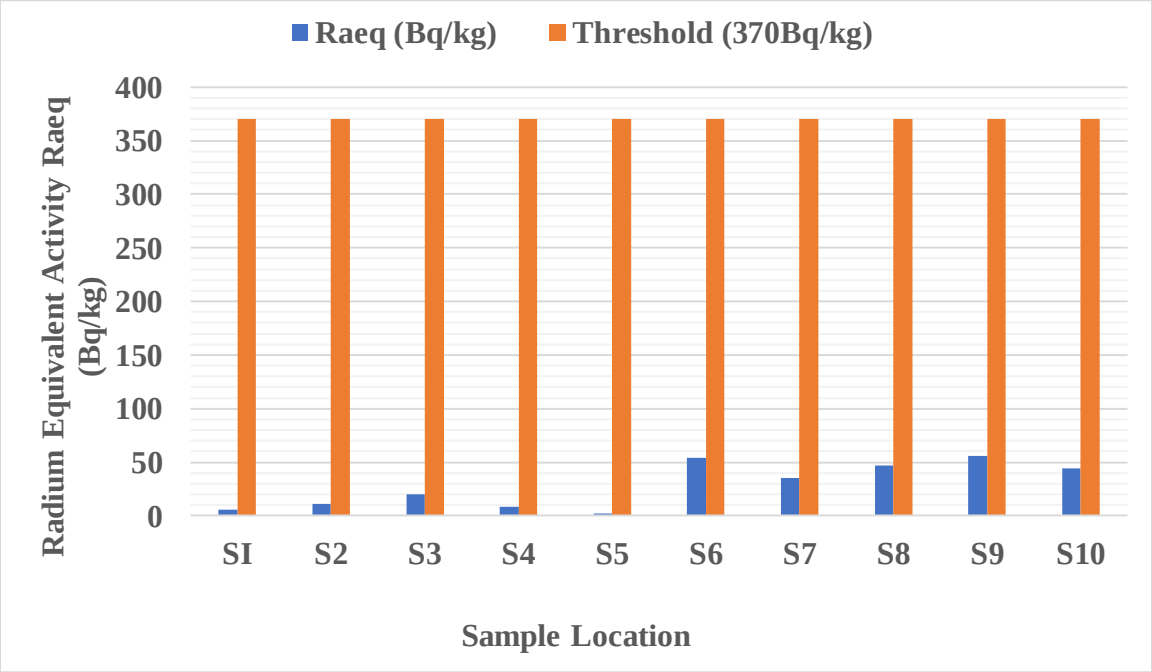

Fig. 6: Comparison of the Radium Equivalent Activity (Ra_{eq}) with Threshold.

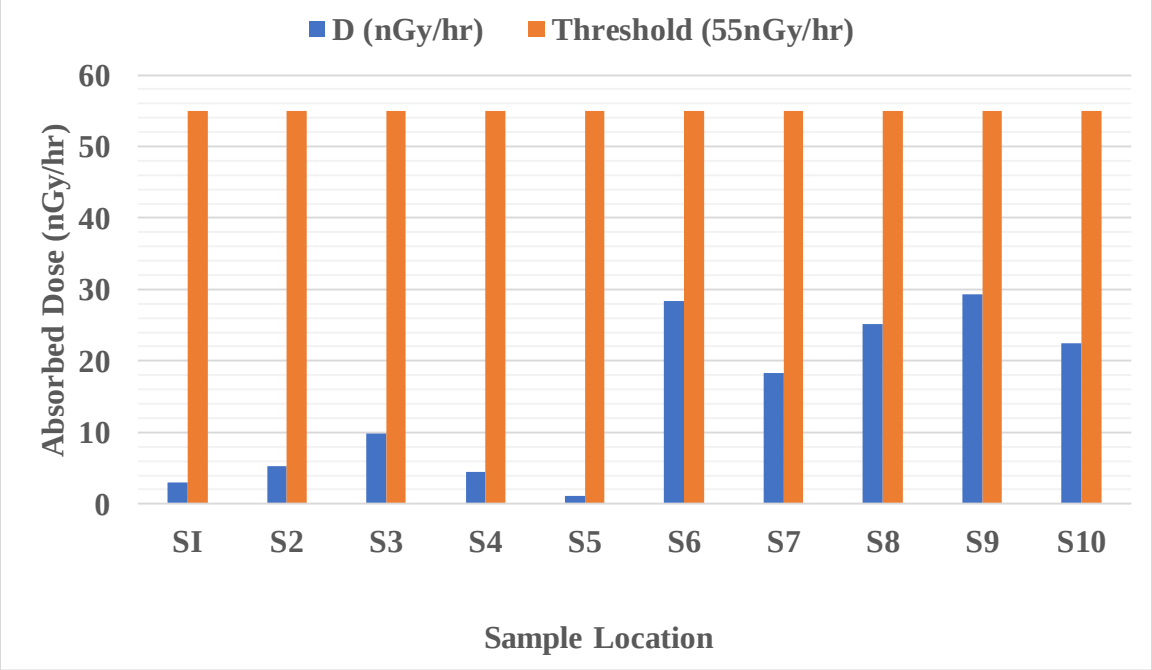

Fig. 7: Comparison of the Absorbed Dose Rate (D) with Threshold.

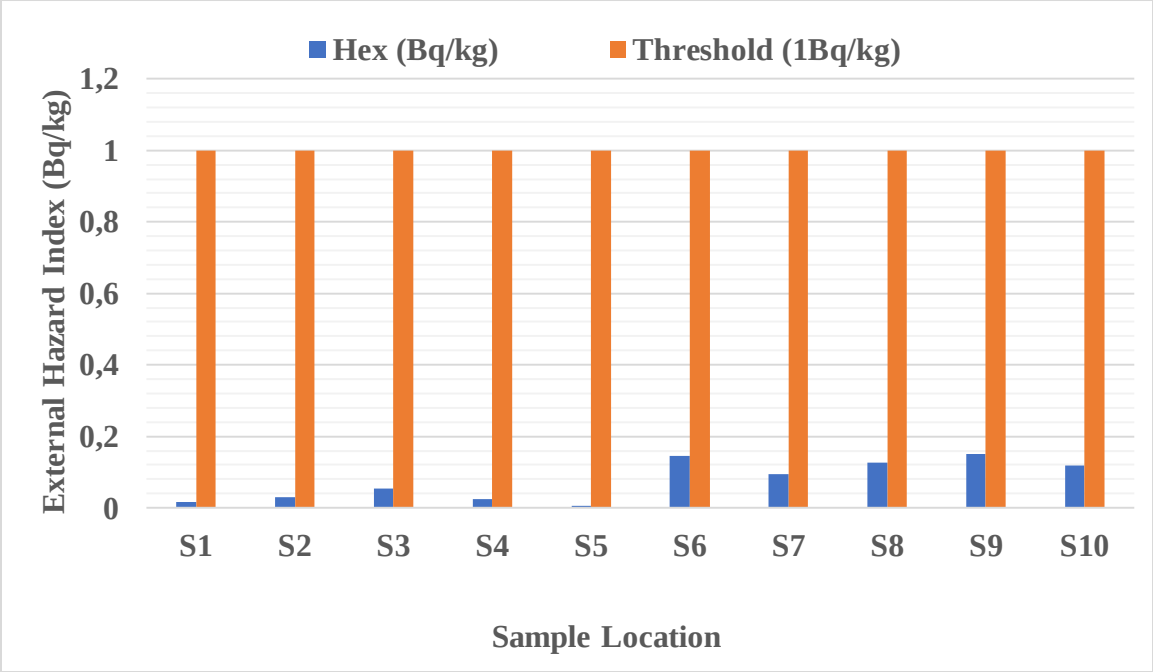

Fig. 8: Comparison of the External Hazard Index (Hex) with Threshold.

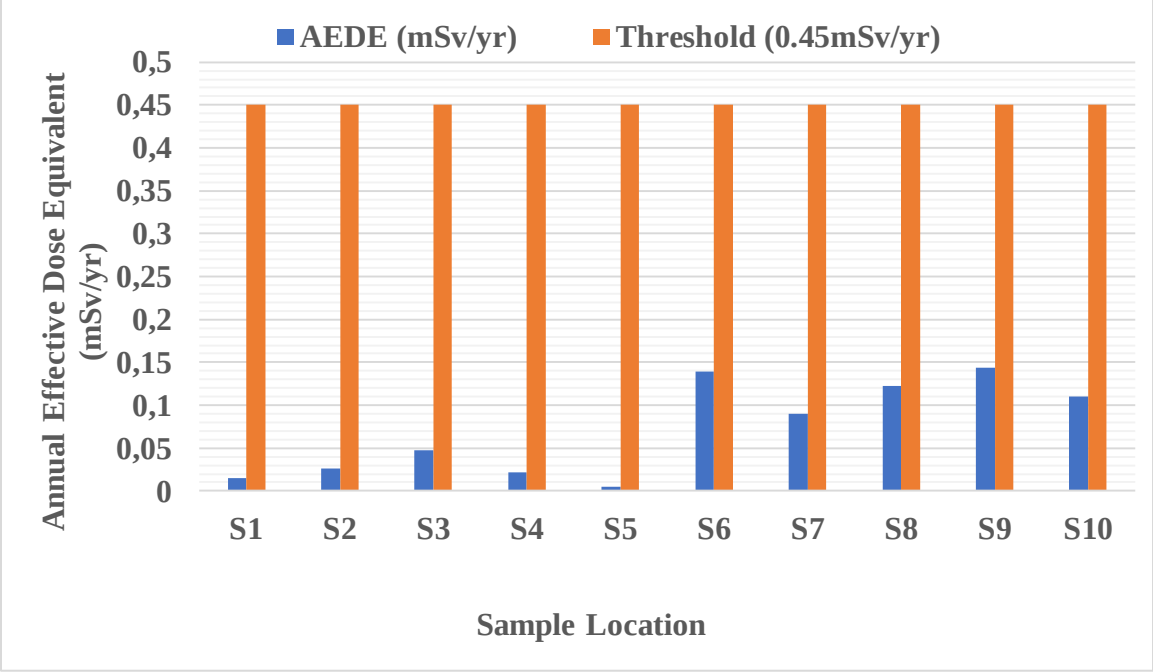

Fig. 9: Comparison of the Annual Effective Dose Equivalent with Threshold.

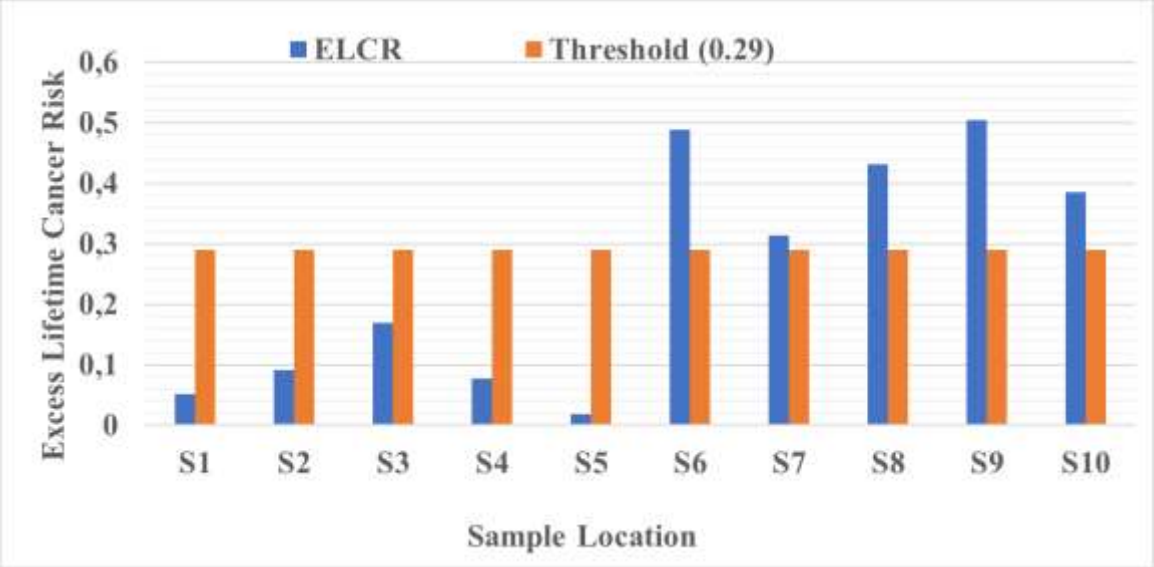

Fig.10: Comparison of the Excess Lifetime Cancer Risk with Threshold.

4 Conclusions

In the course of this radiometric study, it was discovered that some of the abandon mining site show high activity concentration of ^{40}K and excess lifetime cancer risk. These sites with high level of radiation need regulatory control. The level of radiation in those areas is sufficiently high and can cause radiological hazard to the member of the public. Thus, further investigation is needed to safeguard the people residing around the abandon mining sites.

Conflict of Interest

The authors declared that there was no conflict of interest.

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