Journal of Radiation and Nuclear Applications
An International Journal

http://dx.doi.org/10.18576/jrna/080304

Examination of Natural Radioactivity Concentration and Radiological Exposure of Soil Within Mining Site in Umuahia South Abia State Nigeria, Using High Purity Germanium (HPGe) Gamma Ray Spectrometry

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Received: 02 Jul 2023, Revised: 04 Jul 2023, Accepted: 24 Aug. 2023.
Published online: 1 Sep 2023.

Abstract: Mining activity is one of the major sources of exposure to radiation. The main goal of this study was to determine the natural radioactivity level and its radiological exposure risk in mining site Umuahia South Abia State, Nigeria, using High Purity Germanium (HPGe) gamma ray spectrometry. The activity concentration of ^{40}K, ^{226}Ra and ^{232}Th in the soil ranges from 31±2 to 367±27 Bq kg^{-1}, with mean value of 142±11 Bq kg^{-1}; from 26±2 to 65±5 Bq kg^{-1}, with mean value of 49±4; and from 53±9 to 109±17 Bq kg^{-1}, with mean value of 77±16 Bq kg^{-1}, respectively. The activity concentrations of ^{226}Ra and ^{232}Th are above the global values of 32 and 45 Bq kg^{-1} while ^{40}K is depleted in relation to the global mean of 412 Bq kg^{-1}. Radiological parameters are generally in agreement with the values observed for other Nigerian and other countries soil reported in literature. The main responsible for the values observed for the radiological parameters is ^{232}Th followed by ^{226}Ra. The use of this soil for dwelling construction is unlikely to pose any radiological harm for the residents.

Keywords: Natural Radioactivity, Radiological Parameters, Exposure Indices, Soil, Mining, High Purity Germanium (HPGe) Gamma Ray Spectrometry.

1 Introduction

Soils are the actual store house of radionuclide naturally occurring on the Earth's crust responsible for transferring these elements into biological systems [1]. Radioactivity is an observable fact that is connected with energetic atomic nuclei that are automatically decomposed releasing beta, alpha, and neutron particles, or electromagnetic radiation in the form of gamma rays [2]. Soil radioactivity is gaining great scholar attention widely as it one of the main factors in public doses and may help predict changes in the environment [3,4]. Soil characteristics, geological formations and human practices are important factors for potentially elevating the concentrations of background natural radiation [5]. The level of natural radioactivity in an area depends on the soil types, rocks types and its geology [4]. Moreover, the distribution and variation of radionuclides in surface soil relies on the composition and distribution of radioelements in the bedrock, their physical properties and mechanical properties such as porosity, permeability [5]. Soil, rocks and minerals commonly

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contains naturally occurring radioactive materials (NORM), with half-lives as longer as the Earth's age, known as primordial radionuclides [2]. The main radio nuclides of this category are ^{40}K, and the generators of decay series ^{232}Th and ^{238}U.

The radioactivity in the environment needs attention because of the potential harmful effects of the ionizing radiations on tissues of living organisms, though their occurrence is uncommon. Hence, scholars examined natural environmental radioactivity in soil to carry out background study and detect environmental radioactivity levels [6].

Urbanization, mining activities, waste disposal systems, and several human activities are contributors to the radioactivity of the environment [3, 4], when soil, rock and dust material that contains these NORM are spread form the mine areas, during mining and processing, to the surrounding area where they can be amassed in the food chain [6].

Of interest for this study, mining activities and the processing of natural resources have impacted strongly on human being and the environment [5] because NORM is largely responsible for the external or internal irradiation dose received by workers and neighbor population of the mining sites [6]. Internal exposure takes place via the intake of terrestrial radionuclides through inhalation or ingestion [7-8].

In Nigeria, mining activities for coal, lignite, lithium, bitumen, iron ore, and clay, among others, goe on mostly by a domestic and underdeveloped mining industry. In the country there are about 44 different types of minerals that have been identified in over 500 locations [9]. Among the minerals, clay holds significant contributions due to its high demand for construction, cement building, and block industry [10]. In Umuahia South, Abia State, South East, Nigeria, present study area, where clay mining goes on, people are involved in mining activities as their source of income and subjected to the unavoidable NORM emission which aids the constant release of radionuclides in the environment. The main goal of this paper is to study the natural radioactivity in the soil on the Umuahia mining sites and to ascertain radiological parameter and the hazard indices, therefore comparing with international organisation recommendations and global average.

2 Materials and Methods

2.1 Study Area

The study area is Umuahia South, South East, Nigeria, where clay mining is going on. Umuahia South is one of the local government areas of Abia State and it has a geographic coordinate of 5° 31' 0" North, 7° 26' 0" mean area of 140 km² and population of 138,570 [11]. The mining site is about 10 km from Umuahia main town and about 45 km to Aba, Enyimba City of Abia state. The average annual temperature is 26.4 °C. About 2333 mm of precipitation falls annually. The vegetation is ordinarily considered part of tropical rainforest which is the dominant natural vegetation in most parts of southern Nigeria [12].

2.2 Sampling

All of the 15 soil samples were collected using a soil auger sampler at a depth of 5-15 cm, for every sampling point about 1 kg to 1.5 kg were collected. The soil samples were lowered into labelled plastic bags and every bag was properly tied [13].

2.3 Samples Preparation

After the collection of samples from the field, they were kept in natural air–dried in a room at ambient temperature for about one month. Moreover, at the laboratory, the samples were air dried in trays for 7 days and then oven dried at a temperature of 105 ^{0}C for between 3-4 hours until the samples were well dried with a constant weight [14], at the central lab of Federal University Wukari, for preparation. Soil samples were pounded using mortar and pestle. A sieve of 2mm pore size mesh was used to obtain uniform particle sizes, as recommended by IAEA [15]. The sieved prepared soil samples were fled to Nuclear and Energy Research Institute (IPEN) or Instituto de Pesquisas Energéticas e Nucleares (IPEN), Sao Paulo Brazil for analysis.

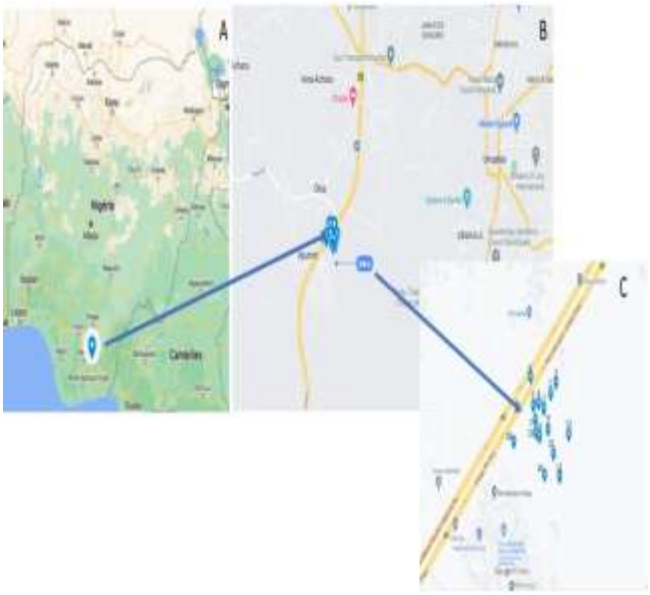

Fig.1: Location of the studied area. A) Nigeria; B) Umuahia region; C) Mine sampling points.

2.4 Analysis of Samples

In this work, the radionuclides ^{40}K, ^{228}Ra, and ^{232}Th were determined using p-type High Purity Germanium Detectors (HPGe), Canberra (USA) with relative efficiency $\geq$ 20%, and resolution $\leq$ 2.00 keV at 1.33MeV. The activity concentration of the radionuclides was determined using

the following energies: ^{40}K was determined using the 1460 keV gamma-ray peak; ^{226}Ra was determined using the 351 keV (^{214}Pb) and 609 keV (^{214}Bi) energies, and ^{232}Th was determined using 727 keV (^{212}Pb) and 238 keV (^{212}Bi). The samples were counted for 86,400 seconds, background was counted for 172,800 seconds while the certified reference materials (CRM) IAEA-RGU-1 uranium ore , IAEA-RGTh-1, thorium ore and IAEA-RGK-1, potassium sulfate, were counted for 7,200 seconds. Activity concentrations were determined via a punctual calibration comparing the intensity of peak in samples to the respective peaks of the standard reference material. For certainty and to ensure the quality assurance of this work, the IAEA-327 reference material (Radionuclides in Soil) from IAEA was used [16]. The obtained values were 626 $\pm$ 43 Bq kg^{-1} for ^{40}K, 36 $\pm$ 3 Bq kg^{-1} for ^{226}Ra and, 38 $\pm$ 7 Bq kg^{-1} for ^{232}Th. All the obtained result agrees with the 95% confidence interval of the reference material: 612 – 630, 32.7 – 35.5 and 37.2 – 39.2 Bq kg^{-1}, for ^{40}K, ^{226}Ra and ^{232}Th, respectively.

2.5 Activity Concentration

The activity concentration of the radionuclides has been calculated using the expression Equation 1 [17].

$$A = \frac{Ne^{\lambda t_0}}{\varepsilon F_c I_\gamma M t_c} \quad (1)$$

Where, A is the activity concentration of the radionuclide; N is number of counts at a given energy, subtracted from the BG; λ is the decay constant; t_c is the time of counting; t_0 is the time difference between sampling and start of count; M is the sample mass; ε is the efficiency; I_γ is the emission probability and F_c is the correction factor.

2.6 Radiological Parameters (RP) and Exposure Indices (EIs)

Radiological parameters of exposure indices are commonly used to assess the radiation exposure originating from the environment and building material [1, 18, 19, 20]. The indices calculated in this work are listed and defined below, denoting with Ra_{At}, Th_{At} and K_{At} the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively, obtained as described in Equation 2 to Equation 7.

Radium Equivalent Activiy: $Ra_{eq} = 0.077 K_{At} + Ra_{At} + 1.43 Th_{At}$ (2)

Absorbed Dose Rate in Air: $AD = 0.0417 K_{At} + 0.462 Ra_{At} + 0.604 Th_{At}$ (3)

Representative Level Index: $RLI = \frac{K_{At}}{1500} + \frac{Ra_{At}}{150} + \frac{Th_{At}}{100}$ (4)

Annual Effective Dose Equivalent: AEDE = D x OF_t x CF_t (5)

$$AEDE_{OUT} = D \times 0.7 \times 0.2 \times 8760 \times 10^{-6} \quad (5a)$$

$$AEDE_{OUT} = D \times 0.7 \times 0.8 \times 8760 \times 10^{-6} \quad (5b)$$

Annual Gonadal Equivalent Dose: $AGDE = 0.314 K_{At} + 3.09 Ra_{At} + 4.18 Th_{At}$ (6)

Excess Lifetime Cancer Risk: $ELCR = AEDE \times MLE \times RF$ (7)

Radium Equivalent Activiy (Ra_{eq}) represents a weighted sum of the activities of ^{226}Ra, ^{232}Th, and ^{40}K in the building materials samples, and is defined as the activity concentration of a radionuclide equivalent to 370 Bq kg^{-1} of ^{226}Ra that gives outdoors an external effective dose rate of 1 mSv y^{-1}. So, the Ra_{eq} index should not exceed 370 Bq kg^{-1} for building materials [21, 22]. The Absorbed Dose Rate in Air (AD) is employed in measuring the exposure that enables ascertaining the quantity of radiation received by the human body due to the concentrations of radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in soil [21].

The Representative Level Index (RLI) expresses the extent the soil materials from a site of interest is convenient for using as construction material [23]. It is also applied to estimate the rate of γ-radiation risks in connection with natural radionuclide in investigated samples (3, 6).

The Annual Effective Dose Equivalent (AEDE) computes the annual effective dose for human beings exposed of a certain dose rate [21, 22]. The AEDE can be obtained by converting the total absorbed dose and the product of occupancy factor (0.2 for outdoor and 0.8 for indoor exposure), of a year, expressed in seconds.

Annual Gonadal Equivalent Dose (AGDE) is used to evaluate the potential effects of exposure to radiation on certain important organs, such as gonads, the activity bone marrow and the bone surface cells since those are the most sensitive parts of the human body [23-24].

Excess Lifetime Cancer Risk (ELCR) is an index related to the risk of developing cancer as a result of amassed exposure to a toxic substance liable to the existence time of human being for a period of lifetime [19].

3 Results and Discussion

3.1 Activity Concentration of Soil the Mining Site

In Tables 1 and 2 the activity concentration of ^{40}K, ^{226}Ra and ^{232}Th determined in the soil samples of the present study and comparison of activity concentration of radionuclides of soil (Bq kg^{-1}) determined in the literature, respectively, are presented. The activity concentration of ^{40}K, ^{226}Ra and ^{232}Th in this study ranges from 31 ± 2 to 367 ± 27 Bqkg^{-1} with mean value of 142 ± 11 Bq kg^{-1}; 26 ± 2 to 65 ± 5 Bq kg^{-1} with mean value of 49 ± 4 and 53 ± 9 to 109 ± 17 Bq kg^{-1} with mean value of 77 ± 16 Bq kg^{-1},

respectively.

The activity concentrations ratio $^{226}Ra/^{40}K$, $^{232}Th/^{40}K$ and $^{232}Th/^{226}Ra$ are shown in Figure 2. The point 16, in the figure, were calculated using the mean values for these radionuclides in soil, according to [25]. It can be seen that ^{40}K is depleted in relation to the reference, while ^{232}Th and ^{226}Ra ratio present the same ratio value (around 1.5) of the global mean. This can indicate that ^{232}Th and ^{226}Ra have the same distribution patter in these soil samples and that the ^{226}Ra activity concentration must be approximately equal to that of ^{238}U considering the crustal Th/U activity ratio with an average of about 1.1 [26]. The ^{40}K depletion must be related to the soil type and the predominant weathering occurring in the region forming the soil known as ferruginous tropical [27, 28].

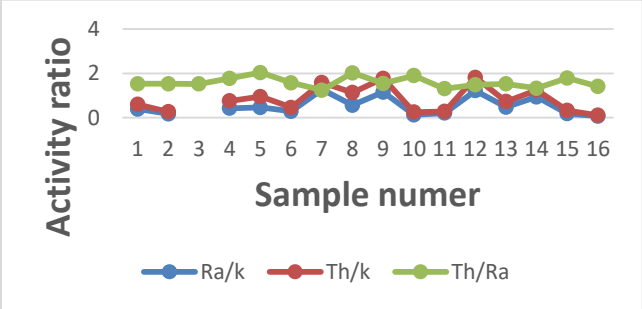

Fig. 2: Activity ratios of the determined radionuclides in soil samples. Point 16 was calculated using the UNSCEAR [25] global average values.

Figure 3 shows the correlation coefficients between ^{40}K, ^{226}Ra and ^{232}Th. It can be seen that ^{40}K do not show significant correlation with ^{226}Ra and ^{232}Th, indicating they probably are linked to different mineral phases or granulometric fractions. Confirming the results showed in Figure 2, the correlation coefficient (Figure 3) between ^{226}Ra and ^{232}Th indicates a strong correlation (r = 0.74).
The strong positive correlation between ^{226}Ra and ^{232}Th in the samples indicates that their rock content must be mostly influenced and controlled by the similar source origin [29], and the fact that radium (from the ^{238}U-series) and thorium decay series occur combined together in nature [30].

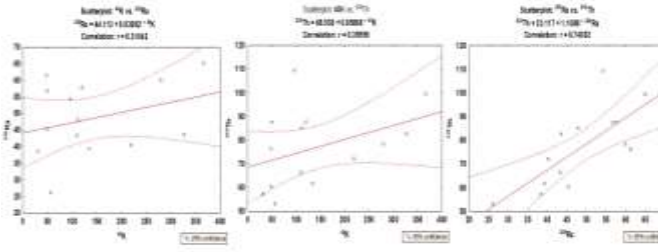

Fig.3: Linear regression and correlation coefficient ^{40}K x ^{226}Ra, ^{40}K x ^{232}Th, and ^{226}Ra x ^{232}Th

The observed values of ^{226}Ra and ^{232}Th in these samples were higher than global average while the ^{40}K activity concentration had lower value (3). The increasing order of the activity concentrations for the analysed radionuclides is $^{226}Ra < ^{232}Th < ^{40}K$. Comparing the present study with local and multinational related reports, as depicted in Table 2, the mean activity concentration of ^{40}K of similar research is higher than the present study except that of [31] also located in the South-East Enugu region of Nigeria. The ^{226}Ra activity concentrations measured in Nigerian research reports indicate a variation ranging from 12 to 66 Bk kq^{-1}, which is in agreement with the values here reported. The same is also observed in the comparing with international research studies whose variation ranges from 22 to 86 Bk kq^{-1}.

3.2 Radiological Parameters (RPs) and Exposure Indices (EIs)

Table 3 depicts the RPs and EIs. The AD values ranged from 70 ± 8 to 105 ± 12 ($nGyh^{-1}$), with almost all the samples presenting higher values than the global average with mean values ranging from 50 to 59 nGy h^{-1} [25]. According to the same previous reference, 90% of the world population are exposed to about 20 to over 100 nGy h^{-1}, indicating that the analyzed soil is in well agreement with the absorbed dose registered in most of the world. The annual outdoor and indoor AEDE ranged from 0.06 ± 0.01 to 0.13 ± 0.01 $mSvy^{-1}$ and 0.23 ± 0.03 to 0.52 ± 0.06 mSv y^{-1}, respectively, and are in good agreement with the global average of 0.08 and 0.42 mSv y^{-1}. Radium equivalent presented a variation from 106 to 236 Bq kg^{-1} and all the samples presented values lower than the recommended value of 370 Bq kg^{-1} indicating that this soil used as construction material will not pose unnecessary exposures for the population that can result in late deterministic effects [32]. The indoor ELCR for the soil samples in this study ranged from 0.17 x 10^{-3} to 0.39 x 10^{-3} which is very close to the world average of 0.29 x 10^{-3}. The RLI in the soils analyzed in this study varied from 0.74 to 1.67 and is above the reference value of 0.46 mSv y^{-1} [33].
The model used for EGDE calculation, for a given material be used as construction material, consider a typical cavity type house with infinitely thick walls, which makes it possible to compare the AGED of a house containing concentration of ^{226}Ra, ^{232}Th, and ^{40}K equal with the world average in soil with those obtained using that given material [34]. The value of AGDE considering the global mean of the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K is 416 µSv y^{-1}. The calculated AGDE for samples from 320 to 732 µSv y^{-1}, slightly higher than the recommend value

[35] Principal component and classification analysis was applied to the results of radionuclides determined activity concentrations and calculated radiological parameters. The result is shown in Figure 4 where the projection of the cases as a function of factors 1 and 2 are presented. The samples in the right side, with positive loading factor, in the factor 1, are those presenting the lowest ^{226}Ra and ^{232}Th activity concentrations and were collected in places near the border of the mine. Samples located in the left side of the Figure 4, have negative loading factors and present the higher activity concentration for ^{226}Ra and ^{232}Th.

The projection of the variables, determined activity concentrations and calculated radiological parameters, as a function of factors 1 and 2 are presented in Figure 5 showing that 40K is negatively correlated with ^{226}Ra and ^{232}Th and that all the calculated radiological indices are dependent of the activity concentration of the last two nuclides.

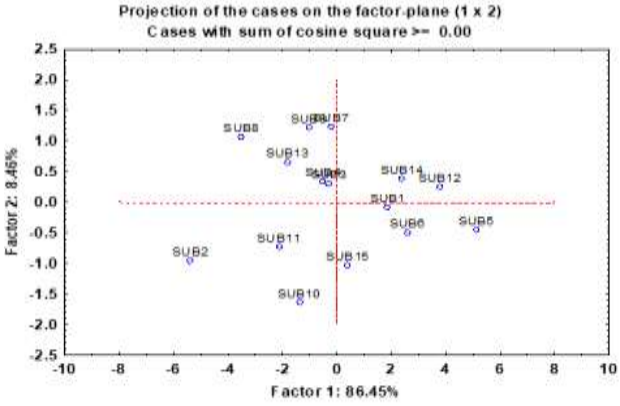

Fig. 4: Factors obtained in component analysis showing the loading factor for Factor 1 x Factor 2

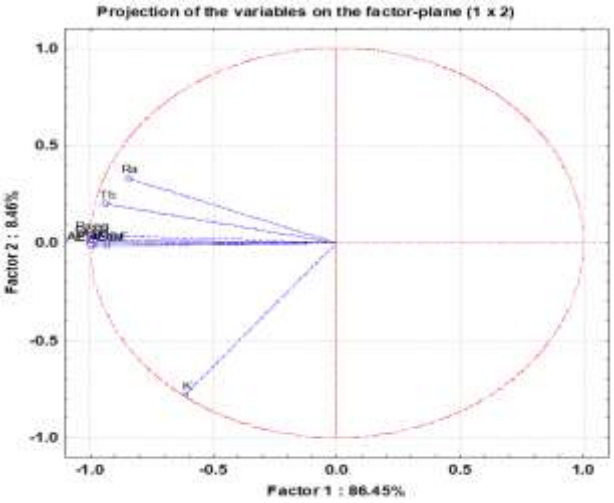

Fig. 5: Correlation between activity concentrations ^{226}Ra, ^{232}Th, and ^{40}K and the radiological indices

Table 4 presents radiological indices values of some studies from Nigeria and other world locations. For AD the present study was higher than reported values except in Tanzania and in Nigeria (Anka). The RLI value of the present study was higher than those reported values, except for the value in Al-Khor, Qatar. ELCR value found for the samples of this study is in the same order of others. AGED in present study was lower than the value registered in Tamilnadu, India, and higher than the value reported for Ibadan Oya, in Nigeria. Hence, the mining activities in the local government may influence the radiological characteristics of the Umuahia region soil and it is recommended that constant monitoring of the mining activities and workers be adopted. If used as construction material for brick production, it is unlikely that any radiological harm may be posed by these samples.

Table 1. Activity of soil Umuahia, SUB (BqKg^{-1}).

SID	^{40}K	^{226}Ra	^{232}Th
SUB1	109.72±8.48	43.31±3.35	66.25±8.62
SUB2	366.59±27.03	65.01±4.89	99.49±13.71
SUB3	BDL	54.73±4.47	83.37±10.93
SUB4	111.61±9.49	47.93±4.32	84.99±11.77
SUB5	55.81±4.47	26.06±2.04	53.05±8.62
SUB6	133.82±10.17	39.25±2.95	61.65±8.61
SUB7	47.93±5.02	61.26±5.38	76.15±10.61
SUB8	96.4±8.16	54.27±4.37	109.43±16.89
SUB9	49.21±4.89	56.78±5.06	87.48±87.48
SUB10	326.37±25.58	43.56±3.28	82.64±11.95
SUB11	279.04±22.13	59.97±5.21	78.33±10.33
SUB12	31.45±2.34	38.4±3.08	57.09±7.45

SUB13	119.92±9.18	57.69±4.36	87.63±15.47
SUB14	48.35±5.04	45.28±4.08	60.23±8.93
SUB15	218.71±15.53	40.31±3.54	72.18±11.09
MEAN	142.49±11.25	48.92±4.02	77.33±16.16
GLOBAL MEAN	412	32	45

Table 2. Comparison of soil ($BqKg^{-1}$) with other scholar's work.

LOCATIONS	^{40}K	^{226}Ra	^{232}Th	REFERENCE
Nigeria (Abia, Umuahia South)	142.49±11.25	48.92±4.02	77.33±16.16	Present Study
Brazil (State of Parana) Soil1	237 ± 15	50 ± 9	48 ± 3	[36]
Brazil (State of Paraná) Soil 2	258 ± 17	23 ± 4	23 ± 1	
India (Punjab and Himachal Pradesh)	143.04	56.74	87.42	[37]
Iraq	335.8	21.7	9.4	[19]
Turkey	771.57	85.75	51.08	[38]
Nigeria (Itagunmodi, south-western)	505.5	55.3	26.4	[4]
Nigeria (Zamfara)	426.51	12.12	60.117	[39]
Nigeria (Kaduna)	459.56	62.28	155.36	[20]
Nigeria (Niger Delta)	210	18	22	[40]
Nigeria (Alade, Southwestern)	383.77	20.82	16.32	[41]
Nigeria (Ijero Town)	635.41	42.02	43.27	[23]
Nigeria, South-East (Enugu)	100.65	33.16	77.68	[31]
Nigeria, North West (Anka)	380.34	41.60	151.15	[42]

Table 3. Radiological parameters for soil Umuahia.

Sample	AD ($nGyh^{-1}$)	Raeq ($BqKg^{-1}$)	RLI	$AEDE_{out}$ (mSv y^1)	$AEDE_{in}$ (mSv y^1)	ELCRx10^{-3} (mSv y^1)	AGDE ($\mu Sv\ y^{-1}$)
SUB1	65±7	146±16	1.02±0.11	0.08±0.01	0.32±0.03	0.24±0.03	445±49
SUB2	105±12	236±27	1.67±0.19	0.13±0.01	0.52±0.06	0.39±0.04	732±81
SUB3	76±9	174±20	1.2±0.14	0.09±0.01	0.37±0.04	0.28±0.03	518±59
SUB4	78±9	178±22	1.24±0.15	0.1±0.01	0.38±0.05	0.29±0.04	538±66
SUB5	46±6	106±15	0.74±0.1	0.06±0.01	0.23±0.03	0.17±0.02	320±44
SUB6	61±7	138±16	0.97±0.11	0.07±0.01	0.3±0.03	0.23±0.03	421±48
SUB7	76±9	174±21	1.2±0.15	0.09±0.01	0.37±0.04	0.29±0.03	523±62
SUB8	95±13	218±29	1.52±0.2	0.12±0.02	0.47±0.06	0.36±0.05	655±87
SUB9	81±55	186±130	1.29±0.91	0.1±0.07	0.4±0.27	0.30±0.21	557±383
SUB10	83±10	187±22	1.33±0.16	0.1±0.01	0.41±0.05	0.31±0.04	583±68
SUB11	86±9	193±22	1.37±0.15	0.11±0.01	0.42±0.05	0.32±0.04	600±66
SUB12	54±6	122±14	0.85±0.1	0.07±0.01	0.26±0.03	0.20±0.02	367±41
SUB13	85±12	192±27	1.34±0.19	0.1±0.01	0.41±0.06	0.32±0.04	655±81
SUB14	59±7	135±17	0.94±0.12	0.07±0.01	0.29±0.04	0.22±0.03	556±51
SUB15	71±9	160±21	1.14±0.14	0.09±0.01	0.35±0.04	0.27±0.03	583±62
Mean	75±12	137±28	1.19±0.2	0.09±0.01	0.37±0.06	0.28±0.05	530±83

Global Mean	59	370	1	0.080	0.42	0.29	416

Table 4. Comparison of radiological parameter of soils of present study and other works.

Location	AD (nG $yh^{-1)}$	Ra_{eq} (Bq kg^{-1})	RLI (Bq kg^{-1})	AEDE (mSv y^{1})	ELCR	AGED	Reference
Present work	75±12	137±28	1.19±0.2	0.09±0.01	0.23±0.05	417±83	Present Study
Egypt (Suez Canal)	18.88	39.24	0.30	0.020			[43]
Iraq	23.27	46.82	0.359	0.142			[19]
India (Tamilnadu)	60.73	133.14	0.96	0.075	0.261	744.7	[44]
Tanzania	106.8	230.6	0.831	1.07			[6]
Iraq (Karbala)	29.7	61.11	0.463	0.03	0.12		[19]
Turkey	45	96		0.056			[45]
Qatar (Al-Khor)	22	46		0.026	0.941		[46]
Nigeria (Ibadan Oya)	42.0	90.97	0.62	0.36		284.94	[2]
Nigeria (Anka)	121.8	273.1		0.149			[42]

5 Conclusions

The NORM radionuclides ^{40}K, ^{226}Ra and ^{232}Th, resulting from clay mining activities, in the soil mining site in Umuahia SouthAbia State Nigeria were determined. Compared to the global average, the activity concentrations of ^{226}Ra and ^{232}Th are above and ^{40}K are below the mean values. Potassium is depleted in this soil samples, probably as a result of the bad rock weathering that occurs in the region. The activity ratio $^{232}Th/^{226}Ra$ indicates that these radioactive elements must have the same provenance in the soil formation and link to the same mineral phases. On the other hand, ^{40}K did not present correlation with ^{226}Ra and ^{232}Th. Comparing with published values for other Nigerian sites and other countries, ^{226}Ra and ^{232}Th was found in agreement with the range of values reported in literature and the low values measured for ^{40}K is also observed in reports for the same region, i.e, Umuahia South, South East, Nigeria. Samples with the highest activity concentrations tend to be concentrated more in the centre of the mining area.

The radiological parameters calculated indicated values slightly above global means for absorbed dose and annual gonadal equivalent dose with ^{232}Th responding for the higher contribution, followed by ^{226}Ra. It was observed that ^{40}K do not present correlation with the radiological indices. The results indicate that use of this soil samples for brick production to be used as construction material is unlikely to cause any radiological harm for the individuals.

Acknowledgement

The authors mmensely appreciate the staff of Instituto de Pesquisas Energe´ticas e Nucleares (IPEN), Centro do reator de pesquisa (CRPq), USP, Brazil and Tertiary Education Trust Fund (TETFUND for their support.

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