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Research article

Radioactivity levels and transfer factor for granite mining field in Asa, North-central Nigeria

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ABSTRACT

Natural radioactivity measurement and dose assessment are important aspects of radiation protection. The goal of this study is to validate the previous results obtained from the in-situ measurements in the study area in order to ascertain the level of radiation hazards to the populaces living around the mining site.A3 × 3-inch lead-shielded NaI(Tl) detector was used to measure the activity concentrations of ^{40}K, ^{238}U and ^{232}Thin soil, water and guinea corn grain samples collected from a granite mining field in Asa, Kwara State, North-central Nigeria. The overall mean activity concentrations of ^{40}K, ^{238}U and ^{232}Th are 441.06, 11.51 and 15.42 Bqkg^{-1}for the soil samples, 20.67, 0.66, and 0.88 BqL^{-1} for the water samples and 214.31, 5.25 and 8.86 Bqkg^{-},^{1}respectively for the grain samples. The bioaccumulation/transfer factors are 0.49, 0.46 and 0.58 for ^{40}K,^{238}U and ^{232}Th respectively. The mean values of all the radiological hazard parameters are within the permissible limit recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation. Consequently, the risk of indoor and outdoor gamma radiation exposure is comparatively less for these Granite soils. Hence, the results in this study will reference future studies in terms of basic radiological data.

1. Introduction

Natural radionuclides such as ^{238}U, ^{232}Th, their progenies and the non-series ^{40}K are generally spread in the earth. Considerable amounts of these radionuclides exist in many mineral rocks including granites. So, granites may possess significant amount of natural radionuclides like ^{238}U, ^{232}Th, their progenies and the non-series ^{40}K (Orosun et al., 2019; USEPA, 2018; Usikalu et al., 2016). The concentrations of these radionuclides are not evenly spread within a particular brick of granite. These radionuclides decay to release dangerous ionizing radiations that are known to cause cancer and other radiation health effects, damaging critical organs of the body (Ajayi and Ajayi, 1999; Orosun et al., 2016; Orosun et al., 2017; Akinyose et al., 2018; USEPA, 2018). Radionuclides in mineral soil like granite find their way into waterways (drinking water) and possibly taken up by plants, thereby becoming available for further redistribution within food chains. They can therefore, eventually be passed on to human beings through food chains, and so may present an environmental threat to the health of local populations. So, information about theconcentrations of these radionuclides inthe environment is fundamental for estimating the level of public exposure to ionizing radiations.

Studies on the levels of these natural radionuclides and their respective progenies have been carried out in different parts of Nigeria (Farai and Ademola, 2001; Ademola, 2005; Obed et al., 2005; Ademola et al., 2008; Jibiri and Esen, 2011; Ajayi et al., 2012; Usikalu et al., 2017, 2018; Isinkaye et al., 2015; Orosun et al., 2016; Adagunodo et al., 2018; Omeje et al., 2018). An in-situ measurement of these radionuclides was carried out on this granite mining field using handheld RS125 gamma-spec by an earlier work by Orosun et al. (2019), which reveals that the activity concentrations of ^{40}K, ^{238}U and ^{232}Th are higher than their respective recommended limits. This call for further investigation into waterways and food chain using higher resolution '3×3' lead shielded NaI (Tl) detector. This is important because in-situ measurements may not sufficiently provide the quantitative activity concentrations of radionuclides. Therefore, the goal of this research is to validate the results obtained from the in-situ measurements in the study area in order to ascertain the level of radiation hazards to the populaces living around the mining site. Also, this study will serve as baseline radiological

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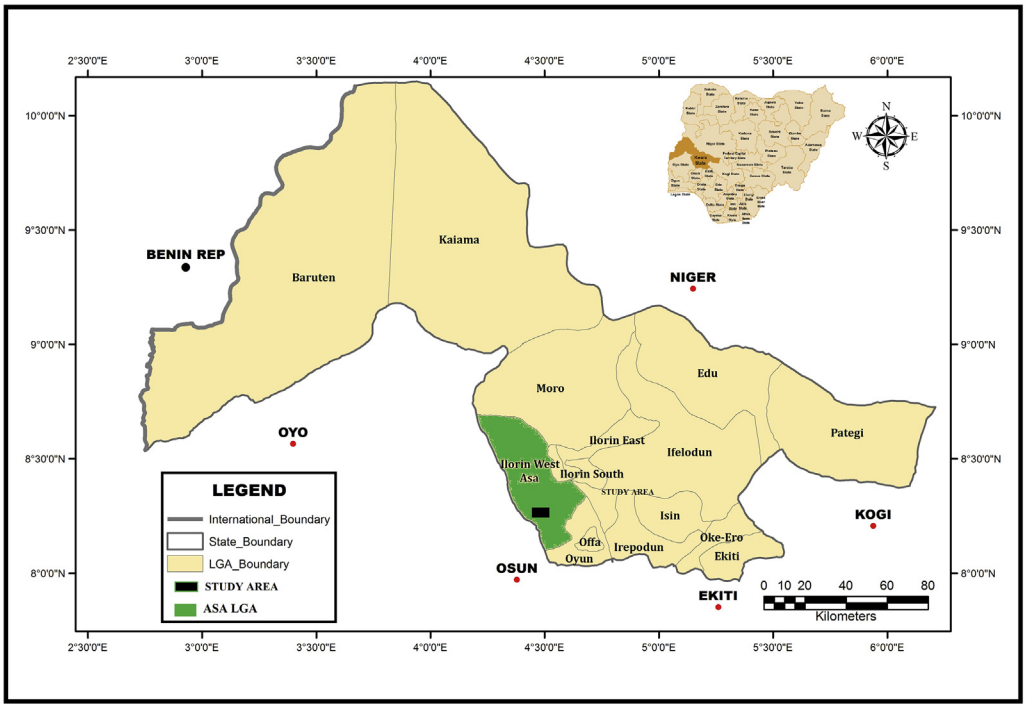

Figure 1. Map of Nigeria showing the survey area.

risk assessment for this granite mining field in Asa LGA, Kwara State, North-central Nigeria.

2. Material and methods

2.1. Study area

Asa is a Local Government Area in Kwara State, Nigeria. It has an area of 1,286 km^2 and a population of 168,300 (City Population, 2016). The study area lies between latitudes 4°12′N and 4°29′N and longitudes 8°07′E and 8°42′E (Figure 1). The study area is underlain by basement complex rock. The soils are formed from metamorphic and igneous rocks which are about 95%. The metamorphic rocks consist of biotitegnesiss, banded gnesiss, quartzite augitegnesissand granitic gnesiss. The intrusive rock comprises of pegmatite and vein quartz (Oyegun, 1985; Ibiremo et al., 2010; Ajadi et al., 2016; Usikalu et al., 2019). Detail geology of the study area can be found in (Oyegun, 1985; Ibiremo et al., 2010; Megwara and Udensi, 2014; Kayode et al., 2015; Ajadi et al., 2016; Orosun et al., 2020).

2.2. Sample preparation

Twenty-four (24) samples of granite bricks were collected randomly from the mining sites under study. These samples were sent to the laboratory where macroscopic traces of glass, rubber, hair, animal and plant matter were removed to ensure that the materials to be analysed are free from such contaminants. The samples were grinded using agate mortar and sieved through a 1 mm sieve mesh and stored in well labelled plastic containers (Marinelli cylindrical beakers) sealed using adhesive tape to prevent the escape of Rn gas and kept for 40 days to ensure secular radioactive equilibrium before the Gamma-Ray spectrometry. A total of 12 samples of water and 12 samples of guinea corn were also collected randomly from the mining site under study. The guinea corn was grinded into powder form using electric blender. The water and the grinded guinea corn samples were collected in a fit rubber test containers (Marinelli cylindrical beakers). Each Marinelli beaker was washed thoroughly with liquid detergents, dried in an oven, wiped with acetone and then dried again in an oven (Faanu et al., 2011a). All the samples were stored in marinelli cylindrical beakers sealed using adhesive tape to prevent the escape of Rn gas and kept for 40 days to ensure secular radioactive equilibrium before the gamma-ray spectrometry.

2.3. Gamma-ray spectrometry

The detector that was used for the radioactivity measurements is a 3 × 3 inch lead-shielded NaI(Tl) detector produced by Princeton Gamma Tech. USA. The NaI(Tl) detector is coupled to gamma spectacular (GS-2000-Pro) multichannel analyzer (MCA) through a pre-amplifier.In order to derive a qualitative and quantitative relationship between the peak position in the spectrum and the corresponding gamma-ray energy, the NaI(Tl) spectrometry system was calibrated. Energy calibration of the detector was carried out using the RSS8 gamma source set (from Spectrum Techniques LLC, USA). It was accomplished by measuring the spectra of point sources emitting gamma-rays of precisely known energies (^{137}Cs and ^{60}Co) and obtaining the measured peak positions for 18000s.

The efficiency calibration of the detector was also carried out using IAEA-RGU1 and a reference source consisting of known radionuclide activities: ^{40}K (578.4 Bqkg^{-1}), ^{238}U (20.9 Bqkg^{-1}) and ^{232}Th (10.47 Bqkg^{-1}). The standard sources are designed for the determination of natural radionuclides in environmental matrices. The source was prepared in a container that has the same geometry as the sample and counted for a period of 18000 s. The full energy peak efficiency was employed as it relates the peak area in the spectrum to the amount of radioactivity present. It is denoted by ε and expressed by

$$\varepsilon = \frac{C_{net}}{A \times P_{\gamma} \times T} \quad (1)$$

where C_{net} is the net peak count for each radionuclide present in the source, A is the activity concentration of the radionuclide present in the source, Pγ is the absolute gamma ray emission probability of the radionuclide being measured and T is the acquisition time.

The uncertainty of the activity concentration measurements deduced according to DKD-3 of Germany, established on the standard uncertainty multiplied by a coverage factor of k = 2 at a confidence level of 95% is

Table 1. Statistical summary of the measured activity concentrations of ^{40}K, ^{238}U, ^{232}Th in the selected soil samples collected from mining sites using 3 × 3 inch NaI(Tl).

Sample location	Sample stat	^{40}K (Bqkg^{-1})	^{238}U (Bqkg^{-1})	^{232}Th (Bqkg^{-1})
Asa L.G.A (Granite) Soil	Min-Max	132.76–643.61	4.48–17.92	6.85–22.93
	Skew	-1.49	-0.41	-0.58
	Kurt	0.19	-0.08	0.15
	Mean ± SD	441.06 ± 271.35	11.51 ± 6.78	15.42 ± 8.09
	Global Limits	**420.00**	**32.00**	**45.00**

3.0% (Faanu et al., 2011a). Prior to the sample measurement, an empty container was counted for 18000 *s* so as to determine the background gamma-ray distribution count. The sealed samples after attaining a state of secular equilibrium were each placed on the detector one after the other for analysis. Each sample was then counted for the same period of time as that of the empty container. The activity concentration of^{214}Bi (determined from its 609.31 and 1764.5 keV γ-ray peak) and ^{214}Pb (295.21 and 351.92 keV) were selected to provide an estimate of ^{238}U in the samples, while 2614.7 keV of ^{208}Tl and 911.21 keV of ^{228}Ac were used as an indicators of ^{232}Th. ^{40}K was determined by measuring the 1460 keV γ-rays emitted during its decay.In the background spectrum, the photopeaks of ^{40}K and ^{137}Cs, which occurred at 1460.83 and 662 keV respectively, were used to determine their LLD and MDA (Cember, 1996).

The activity concentration A (Bqkg^{-1} or Bql^{-1}) of each identified radionuclide in the sample was calculated using:

$$A = \frac{Cnet}{\gamma \times \varepsilon(E\gamma) \times T \times Ms} \quad (2)$$

where, *Cnet* is the net count rate under the corresponding photopeak, γ is the absolute gammaintensity, ε(*E*γ) is the detector efficiency at the specific gamma ray energy (*E*γ), *t* is the totalcounting time (18000 *s*), and *Ms* is the sample mass in *kg*.

2.4. External absorbed dose rate

The absorbed dose rate (nGyhr^{-1}) in air due to the mean specific activities of ^{40}K, ^{238}U, and ^{232}Th (Bqkg^{-1}) in the collected samples was calculated at 1m above the ground surface. It can be calculated using Eq. (3) (UNSCEAR, 2000)

$$D\ (nGyh^{-1}) = 0.0417C_K + 0.462C_U + 0.623C_{Th} \quad (3)$$

where $DCF_K = 0.0417$, $DCF_U = 0.462$ and $DCF_{Th} = 0.623$ are the dose conversion factors (UNSCEAR, 2000) and C_K, C_U, and C_{Th} are the activity concentrations of ^{40}K, ^{238}U, and ^{232}Th in the soil samples respectively.

2.5. Annual effective dose for external exposures ($AED_{external\ exposure}$)

The annual effective dose received outdoor by a member of the public was calculated from the absorbed dose rate by applying dose conversion factor of 0.7 SvGy^{-1} and occupancy factor for outdoor was 0.2 (UNSCEAR, 2000). $AED_{outdoor}$ was determined using Eq. (4) (Issa et al., 2013; UNSCEAR, 2000).

$$AED_{outdoor}(\mu Svy^{-1}) = D(nGyh^{-1}) \times 8760h \times 0.7\ (SvGy^{-1}) \times 0.2 \times 10^{-3} \quad (4)$$

The $AED_{outdoor}$ involves a consideration of the absorbed dose emitted from radionuclide in the environment such as ^{238}U, ^{232}Th and ^{40}K.

2.6. Representative gamma index ($I\gamma$)

This is used to estimate the gamma radiation hazard associated with the natural radionuclide in specific investigated samples. The representative gamma index was estimated as shown in Eq. (5) (UNSCEAR, 2000)

$$I_\gamma = \frac{C_u}{150} + \frac{C_{Th}}{100} + \frac{C_k}{1500} \leq 1 \quad (5)$$

where, C_U, C_{Th}, and C_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K in the granite sample.

2.7. 7Annual effective dose for ingested radionuclide ($AED_{Internal\ exposure}$)

The annual effective dose rate for all the ingested radionuclides from food (guinea corn) and water was calculated using Eq. (6)

$$AED_{Internal\ Exposure} = 365 \sum_i I_i \times D_i \quad (6)$$

where I_i is the daily intakes of radionuclide (Bqd^{-1})= (concentration of radionuclide in food or water in Bqkg^{-1} or Bql^{-1}) × (consumption rate of food or water in kgd^{-1} or ld^{-1})and $D_i$is the ingestion dose coefficient for adults. D_i for ^{238}U, ^{234}Th and ^{40}K are:4.5 × 10^{-8}, 2.3 × 10^{-7} and 6.2 × 10^{-9} SvBq^{-1}, respectively (Orosun et al., 2018a & b; UNSCEAR, 2000; ICRP, 2012). The daily intake of water per person is 2 ld^{-1} for adults. While anaverage Nigerian consumes 24.8 kg of guinea corn grains per year (Bamidele et al., 2010; FAO, 2004; IRRI, 2001).

2.8. Excess lifetime cancer risk (ELCR)

The Excess Lifetime Cancer Risk (ELCR) was calculated using Eq. (7) (UNSCEAR, 2000)

$$ELCR = AED \times DL \times RF \quad (7)$$

where, AED is the annual equivalent dose, DL is the average duration of life (estimated to 70 years) and RF is the risk factor (Sv^{-1}), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for public (UNSCEAR, 2000).

2.9. Bioaccumulation/transfer factor (BAF)

The bioaccumulation factor (BAF) is defined as the ratio of the activity concentration of the radionuclides in the grain to that in the soil. The BAF was calculatedusing the following equation (Karatas et al., 2007);

$$BAF = \frac{A_{guinea-corn}}{A_{soil}} \quad (8)$$

where $A_{guinea-corn}$ and A_{soil} represent the activity concentrations of the radionuclides in the guinea corn grain and the soil, respectively. Soil-to-plant transfer is one of the key processes of human exposure to radionuclides through the food chain. When BAF <1 or BAF = 1, it signifies that the plant only absorbs the radio elements but does not accumulate and when BAF >1, it follows that the plant accumulates the radio elements.

3. Results and discussion

The summary of the results of the gamma spectrometry measurements using the 3 × 3 inch lead-shielded NaI(Tl) detector and their estimated standard deviation (SD), Skew and Kurt for the soil, water, and

Table 2. Statistical summary of the measured activity concentrations of ^{40}K, ^{238}U, ^{232}Th in the selected water samples collected from mining sites using 3 × 3 inch NaI (Tl).

Sample Location	Sample Stat	^{40}K (Bql^{-1})	^{238}U (Bql^{-1})	^{232}Th (Bql^{-1})
Water	Min	8.80	0.33	0.49
	Max	30.77	1.04	1.36
	Mean ± SD	20.67 ± 11.09	0.66 ± 0.36	0.88 ± 0.44
	Global limits	10.00	1.00	1.00

Table 3. Statistical summary of the measured activity concentrations of ^{40}K, ^{238}U, ^{232}Th in the selected guinea corn samples collected from mining sites using 3 × 3 inch NaI (Tl).

Sample Location	Sample Stat	^{40}K (Bqkg^{-1})	^{238}U (Bqkg^{-1})	^{232}Th (Bqkg^{-1})
Guinea corn	Min	105.15	2.14	5.48
	Max	418.89	10.30	15.48
	Mean ± SD	214.31 ± 77.30	5.25 ± 2.41	8.86 ± 2.73
Transfer factor		0.49	0.46	0.58

Table 4. Comparison of the mean activity concentration with some selected studies.

Case Study	*U-238* (Bqkg^{-1})	*Th-232* (Bqkg^{-1})	*K-40* (Bqkg^{-1})	Country	References
Soil	19.16	48.56	1146.88	India	Chandrasekaran et al. (2014)
Kaolin (soil)	82.00	94.80	463.60	Turkey	Turhan (2009)
Clay (soil)	39.30	49.60	569.50	Turkey	Turhan (2009)
Floor ceramic	101.22	87.53	304.57	Iraq	Amana (2017)
Wall ceramic	102.12	70.90	328.60	Iraq	Amana (2017)
Kaolin (soil)	964.70	251.60	58.90	Eqypt	El-Dine et al. (2004)
Phosphogypsum	206.80	99.10	15.10	Brazil	Mazzilli and Saueia (1999)
Kaolin (soil)	38.20	65.10	93.90	Nigeria (Dahomey Basin)	Adagunodo et al., 2018
Building materials	51.50	48.10	114.70	Australia	Berekta and Mathew (1985)
Sands (soil)	78.00	33.00	337.00	Egypt	El-Afifi et al. (2006)
Soil Samples	55.30	26.40	505.10	Nigeria (Itagunmodi)	Ademola et al. (2014)
Soil and Rock	13.60	24.20	162.10	Ghana	Faanu et al. (2011b)
Laterite (soil)	30.00	41.00	65.00	Nigeria (Obajana)	Ajayi et al. (2012)
Granite (In-situ)	18.15	42.86	570.91	Nigeria (Asa In-situ)	Orosun et al. (2019)
Granite (Soil)	**11.51**	**15.42**	**441.06**	**Nigeria (Asa)**	**Present Study**
Soil and Rock	32.00	45.00	420.00	Global Limit	UNSCEAR, 2000

the guinea corn are presented in Tables 1, 2, and 3 respectively. The radionuclide observed with reliable regularity belonged to the decay series chain headed by ^{238}U and ^{232}Th as well as the non-series ^{40}K. The ^{40}K activity concentration dominated over the ^{238}U and ^{232}Th elemental activities in all the locations as expected. The measured values for all the parameters (i.e. ^{238}U, ^{232}Th and ^{40}K) were moderately skewed (the distribution is approximately or moderately symmetric) since most of the measure of the asymmetry of their probability distribution about their means is in the range of -2 and +2 (Normality Testing, 2019).

As expected, the mean activity concentration of ^{40}K in the soil samples is higher than the ^{238}U and ^{232}Th mean activities.^{40}K has highest activity concentration of 643.61 Bqkg^{-1} and lowest value of 132.76 Bqkg^{-1}. The highest and lowest activity concentrations of ^{238}U and ^{232}Th were found to be 17.92; 4.48 Bqkg^{-1} and 22.93; 6.85 Bqkg^{-1} respectively. The overall mean of the activity concentrations of the measured radionuclides in the soil samples was calculated and found to be 441.06, 11.51 and 15.42 Bqkg^{-1} for ^{40}K, ^{238}U and ^{232}Th respectively. The estimated mean value for ^{40}K is slightly higher than the global average of 420.00 Bqkg^{-1} for normal background radiation levels given by UNSCEAR. Surprisingly, unlike the results of the in-situ measurements whose values are higher than their global limits (Orosun et al., 2019), the mean activity concentrations of ^{238}U and ^{232}Th are lower than their corresponding global average of 32.00 Bqkg^{-1} and 45.00 Bqkg^{-1} respectively provided by ICRP (1991), IAEA (1996) and UNSCEAR (2000) report. This follows that all the measured activity concentrations of ^{40}K, ^{238}U and ^{232}Th in all the locations are lower than their respective *in situ* measurements reported by Orosun et al. (2019). This could be due to the contribution of earth materials to the gamma ray detection for the *in situ* measurements.

In general, comparative analysis of these mean values of ^{40}K, ^{238}U, ^{232}Th for the Granite mine field under study with some selected studies from literatures across the world is given in Table 4. . It was observed that the mean values of ^{238}U and ^{232}Th obtained in this study are only lower than the values reported by all the authors (see Table 4). The values of ^{40}K though higher than the values reported by Amana (2017) (in Iraq), El-Dine et al. (2004) (in Egypt), Mazzilli and Saueia (1999) (in Brazil), Adagunodo et al. (2018) (in Nigerai) and Ajayi et al. (2012) (in Nigeria), it is certainly less than the in-situ measurements carried out earlier by Orosun et al. (2019). The elevated values recorded during the in-situ measurements may be due to contribution of the earthly materials composition of the study area (Orosun et al., 2019). The variation observed in the activity concentrations of these radionuclides when compared with other studies was also believed to be because these radio-elements are not evenly spread in the earth crust. So their concentration level depends mostly on the local geology.

Table 5. Summary of the estimated mean values of radiological impact parameters for the soil samples.

Location	D ($nGyh^{-1}$)	$AED_{outdoor}$ ($mSvy^{-1}$)	RLI	$ELCR$ ($\times 10^{-3}$)
Asa LGA (Soil)	32.72	0.04	0.53	1.08
Global Limits	**59.00**	**0.07**	**≤1**	**3.75**

Table 6. Summary of the estimated mean values of radiological impact parameters for the water and guinea corn samples.

Locations	$AEDing$ ($mSvy^{-1}$)	$ELCR$ ($\times 10^{-3}$)
Water	0.26	0.92
Guinea corn	0.09	0.31
Global limits	**1.00**	**3.75**

The overall mean of the activity concentrations of the measured radionuclides (^{40}K, ^{238}U and ^{232}Th) was calculated and found to be 20.67, 0.66 and 0.88 Bql^{-1} respectively for water and 214.31, 5.25 and 8.86 $Bqkg^{-1}$ respectively for the guinea corn.The mean activity concentration of ^{40}K in the water and guinea corn samples is higher than the ^{238}U and ^{232}Th mean activities. While the mean values of ^{238}U and ^{232}Th for the water samples are within the permissible limits of 1 Bql^{-1}, their values in the guinea corn samples exceeds this universal limit of 1 Bql^{-1}. The mean activity of ^{40}K for both the water and the guinea corn is higher than global limit of 10 Bql^{-1} provided by UNSCEAR.The activity concentrations of these measured radionuclides are much higher in the grains than the water.

The bioaccumulation/transfer factor was obtained which is the ratio of the mean activity concentration of a given radionuclide in soil to its mean activity concentration in crop plant. The bioaccumulation factors estimated are 0.49, 0.46 and 0.58 respectively for ^{40}K, ^{238}U and ^{232}Th. These values are comparable with the findings of Oluyide et al. (2018). The presence of the radionuclides in high concentration in the grains is a call for serious concern because their high activity concentration can bring about internal exposure to ionizing radiation which is very detrimental to human health. The level of damage though depends on the amount or rate of consumption of the food crop.

The results of the activity concentrations of these radionuclides were used to estimate the corresponding radiation impact parameters. This is in order to assess the level of radiological hazards associated with theuse of the soil, and the consumption of water and guinea corn from the mining site.

The mean value of dose rate (D) and $AED_{outdoor}$for the soil samples are less than their corresponding recommended limits given by UNSCEAR (Table 5). This showed that the risk of indoor and outdoor gamma radiation exposure is comparatively less for these granite soils. However, the populaces may not be safe from exposure to ionizing radiation since no amount of radiation is safe for stochastic effects. The mean values of $AED_{internal}$ for ingested radionuclides in the water and the grains are lower than the recommended value of 1 $mSvy^{-1}$ (Table 6).

The estimated mean representative gamma index for the soil samples (a radiological hazard parameter used as screening tool for identifying building materials that may possibly be of radiological concern to be used as construction materials) is within the recommended value of 1. This follows that from a radiation protection point of view, the granite bricks qualify to be used as building materials. The estimated values for the $ELCR$ for the soil, water and grain samples were also below the recommended limits of 3.75×10^{-3}.

4. Conclusion

A well calibrated 3 × 3 inch lead-shielded NaI(Tl) detector was used to measure the activity concentrations of ^{40}K, ^{238}U, and ^{232}Th in soil, water and guinea corn grains cultivated around a granite mining field in Asa, Kwara State, North-central Nigeria. The results of the activity concentrations obtained were used to estimate the corresponding radiation impact parameters in order to assess the level of radiological hazards to the populace in the study environment. The results of the activity concentrations showed that the mine field is more loaded with ^{40}K compared with ^{238}U and ^{232}Th. Also, all the measured activity concentrations of ^{40}K, ^{238}U and ^{232}Th are lower than their respective *in situ* measurements reported by Orosun et al. (2019). This was believed to be due to the contribution of earth materials to the gamma ray detection for the *in situ* measurements. The estimated mean values of the entire radiation hazard index are within the recommended limits. Hence, the danger of exposure to ionizing radiation is less. It is recommended that the Nigerian Environmental Protection Agency (NEPA) and other regulatory bodies in Nigeria should enforce statutory requirements of mining activities in the State and the country at large in accordance with international regulations.

Declarations

Author contribution statement

Muyiwa Michael Orosun: Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Mojisola Rachael Usikalu: Contributed reagents, materials, analysis tools or data.

Kayode John Oyewumi: Conceived and designed the experiments.

Justina Ada Achuka: Analyzed and interpreted the data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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