

Baseline radioactivity and associated radiological hazards in soils around a proposed nuclear power plant facility, South-South Nigeria

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ABSTRACT

Increased concerns about human population exposure to increased ionizing radiations emanating from anthropogenic sources of radiation, as well as the associated environmental risk, have necessitated radioactivity baseline studies in many regions. This work based on standard procedures designed suite nuclear power plant (NPP) radiological monitoring plan was conducted to assess the radiological baseline of Itu, Nigeria (5°10'0 N, 7°59'0 E). A total of 44 soil samples were collected and analyzed using High Purity Germanium detector. The average activity concentration values for soil samples were $47.98 \pm 8 \text{ Bqkg}^{-1}$ for Ra, $15.15 \pm 1 \text{ Bqkg}^{-1}$ for ^{238}U , $38.65 \pm 6 \text{ Bqkg}^{-1}$ for ^{232}Th , and $41.55 \pm 6 \text{ Bqkg}^{-1}$ for ^4K . These mean values were lower than the UN Scientific Committee on the effects of atomic radiation's world mean values of 33, 45, and 420 Bqkg^{-1} for $^{238}\text{U}/^{226}\text{Ra}$, ^{232}Th , and ^4K , respectively. The hazard indices calculated using the measured activity concentrations indicated 32.04 nGy/h, 0.042 mSv/yr, and 0.17 mSv/yr for mean GDR, outdoor, and indoor AEDE, respectively. In addition, the average radium equivalent resulting from terrestrial gamma of activity concentration was 113.94 Bqkg^{-1} , with a mean ELCR of 0.15×10^{-3} . These baseline data show that there is no substantial radiological concern for human health.

1. Introduction

In recent years, there has been a great deal of research on low-level exposure of human beings to ionizing radiation from naturally occurring radionuclides, specifically ^{232}Th , ^{238}U , their decay species, and ^4K in soils. However, depending on environmental conditions such as local climate and geological characteristics, the concentrations of these radionuclides in soils, as well as the resultant rate of human exposures, vary greatly (Luiz do Carmo Leal et al., 2020; Sirin, 2020; Ekong et al., 2019; Manigandan and Chandar Shekar (2014); UNSCEAR, 2008). Measurements of natural and artificial radionuclides in soil are critical radiological tool for the evaluation of human and non-human biota exposures to ionizing radiation and identification of areas of elevated environmental radioactivity levels (Movsisyan et al., 2021; Baltas et al., 2019; Barnekow et al., 2019; Cinelli et al., 2018; El Samad et al., 2013).

The significant proportion of the natural background radiation in the environment is caused by Naturally Occurring Radioactive Materials (NORM), which are classified as radioactive materials containing

negligible amount of radionuclides (Missimer et al., 2019; IAEA, 2007a, b; Kathren, 1998). Natural radiation exposure is divided into three types: planned, emergency, and existing. The planned exposure scenario is when an ionizing radiation source is introduced on purpose; the emergency radiological exposure is when a planned introduction of an ionizing radiation source goes out of control; and the existing exposures are those radiological cases triggered by natural background radiations from the earth crust. The sources of radiation are classified as primordial, cosmic and anthropogenic radionuclides. Primordial or terrestrial radionuclides are radioactive materials that emanate from the Earth's crust and mantle, and are predominantly composed of uranium-238 (^{238}U), thorium (^{232}Th) and potassium (^4K) (IAEA, 2015; Clark and Veil, 2009).

Natural alpha emitters ^{232}Th , ^{238}U , ^{226}Ra , ^{210}Po , ^{234}U , beta emitter ^4K , ^{210}Pb , ^{228}Ra , and artificial radionuclides ^{241}Am , ^{90}Sr , ^{137}Cs are present in varying concentrations in the environment and are mostly dependent on the geological formation of a region (Njinga et al., 2015; UNSCEAR, 2000a). Cosmogenic NORM comes from neutron interactions with

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elements like carbon-14 (^{14}C) hydrogen-3 (^3He) under high temperatures in stars (IAEA, 2015). Anthropogenic radionuclides are formed by radiological exposure to materials used in human-mediated activity such as medical applications, oil and gas exploration and exploitation, energy production (coal), mineral mining and milling, and nuclear reaction operations (Giwa et al., 2018; Njinga et al., 2015; Sohrabi, 1998).

This study is an important measure toward meeting the radiological requirements for selecting sites for nuclear power plant (NPP) construction in Nigeria, as stated in the media by NPP promoters (Yudewei, 2017; Anuforo and Onyedika (2016); Onwuemenyi, 2010). The aims of this study are to measure and provide a data set of natural radionuclides in soil of the area, and to calculate the radiological hazard indices for the detected radionuclides in order to provide baseline data that can be used to evaluate the potential radiation levels from the area.

2. Materials and method

2.1. Instrumentation

The major instruments used in this work included GARMIN etrex 10 (GPS Finders – Serial number 3964), and ArcGIS. The High Purity Germanium (HPGe) radiation detector (8023 Model: Gc with Serial Number: 9744 and Pre-Amplifier Model: 2002csl) at the Environmental Monitoring Laboratory of National Institute of Radiation Protection and Research (NIRPR), University of Ibadan was employed gamma spectrometric analysis. The NIRPR has participated in inter-comparison analysis with other environmental analytical laboratories organized by the International Atomic Energy Agency (IAEA), Vienna, Austria. The HPGe detector is a Canberra coaxial type with 50% relative efficiency, having a resolution of 2.4 keV at 1.33 MeV of Co-60. The HPGe detector is well shielded by a lead shield to protect it from external radiation interference during measurement. The calibration of the HPGe detector was performed using IAEA calibration Multi-Gamma Ray Standard (MGS6M315), to acquire spectrum peaks of radionuclides spanning through energy lines of ^{241}Am at 59.5 keV to ^{208}Tl at 2614 keV, with

which all other unknown radionuclides were fully detected and identified using Genie 2K software (Lilley, 2001; Knoll, 2010). The general quality control of the radiochemical procedures, as well as the efficiency calibration of the gamma-counting devices, were performed using standard reference material from the IAEA for varying energies of interest in the specified sample geometry.

2.2. Study area

Itu ($5^{\circ}10'0''\text{N}$, $7^{\circ}59'0''\text{E}$) is a Local Government Area in Akwa Ibom State, Nigeria. It has a landmass that covers approximately 606.10 km^2 , and is located about 27 km from Uyo, the capital city of Akwa Ibom State (Fig. 1). The Itu people's major occupations are farming, fishing, and trading, with a projected population of 161,572 in 2013 (AKS, 2014). The study area is bounded to the north by Eki/Odukpani in Cross River State, to the south by Uyo, to the east by Anakpa/Uruan in Akwa Ibom State, and to the west by Oko Ita/Ibiono Ibom in Akwa Ibom State. The climate is tropical, with rainy and dry seasons. The rainy season has an average annual precipitation of 2409 mm, with temperatures varying from 25.5 to 28.3°C (Ayoade, 1998). The area is drained by the Cross River on the east, which branches and flows to form part of the Itu River, the Imo River on the southwest, and the Qua Iboe River on the south-central parts; all of these rivers flow from the State's northern highlands and drain into the Atlantic Ocean in the south. The elevation ranges from 66 to 131 m above sea level (Beka and Udom, 2014).

2.3. Sampling zones and samples collection

To ensure optimum coverage and representativeness, the entire study area was divided into four (4) zones in accordance with the NPP radiological monitoring plan (MoEF, 2010). The zones are as follows: (1) the Exclusion Zone (EZ) of approximately 1.5 km; (2) the Sterilized Zone (SZ) of approximately 5 km; (3) the Emergency Planning Zone (EPZ) of approximately 16 km; and (4) the Impact Assessment Zone (IAZ) of approximately 30 km radius. The research design also included a 5 km

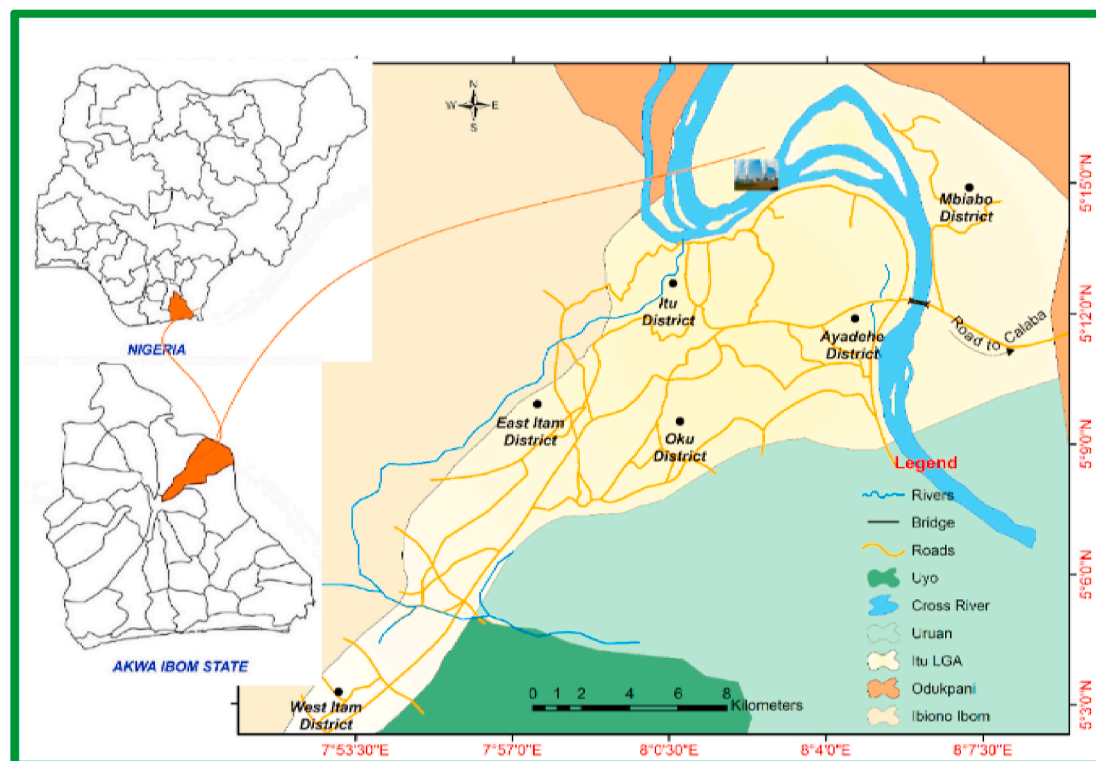


Fig. 1. Map of Itu showing districts and major river (Insert: Nigeria and Akwa Ibom State).

radius (5) Radiological Assessment Zone (RAZ) for collecting control samples. The NPP radiological monitoring demarcated arrangements for the purpose of this study is presented in Fig. 2.

Soil samples were obtained by removing the top soil to a depth of 15 cm after clearing the top vegetation of grass and other debris. In accordance with the NPP radiological monitoring plan, a systematic random sampling approach was used for each zone considered to have the same homogeneity. It is important to note that the sampling device was thoroughly cleaned after each collection point to prevent cross contamination of samples. Each sample was appropriately labeled with codes for proper identification before being carefully packed in a polyethylene bag, sealed, and transported for analysis.

2.4. Sample preparation and analysis

A total of 44 soil samples were collected from the villages in Itu covering the entire four radiological monitoring zones and the 5 km villages as control. The samples were air-dried to remove excess moisture and then oven-dried at 140 °C to obtain constant weight. Hard or stony samples were crushed and pulverized into powdery form and sieved with a mesh of 500 μm , then packaged and sealed in Marinelli beakers and kept for 28 days to attain secular equilibrium between parent and progeny for ^{238}U and ^{232}Th . The samples were then counted for 18,000 s with HPGe detector. The ^{238}U decay chain were considered by identifying γ -ray photo-peaks corresponding to ^{226}Ra at 186 keV, ^{214}Pb at (242.0 keV, 295.2 keV & 351.9 keV), ^{214}Bi (609.3 keV, 768.4 keV, 806.19 keV, 1120.3 keV, 1377.669 keV and 1401.516 keV respectively) and ^{234}Pa (1001.03 keV). Also, the ^{232}Th day chain in the samples were considered by identifying γ -ray photo-peaks of ^{228}Ac at (209.3 keV, 338.3 keV, 409.5 keV and 911.1 keV), ^{208}Tl at (277.4 keV, 583.2 keV, 860.6 keV and 2614 keV), ^{212}Bi at (727.33 keV and 785.37 keV), ^{212}Pb (238.6 keV and 300.1 keV and ^{224}Ra (240.986 keV), etc. Lastly, the ^4K and ^{137}Cs non decay chain radionuclides were estimated from emitted gamma ray with energies of 1460.8 keV and 661 keV, respectively. Upon attainment of secular equilibrium, analyzed photo peaks counts of progenies with greater intensity or those with higher energy were collated under respective parents of ^{238}U and ^{232}Th for evaluation using equation (2).

In the process of photo peak selection, often times there are overlapping contributing influence of radionuclides with close proximities thereby increasing the activity concentration. To circumvent this effect, the use of HPGe detector provided better energy resolution by analyzes complex spectra or distinguish any energy lines of close proximity like ^{235}U at 185.72 keV and ^{226}Ra at 186 keV was employed in this study.

The efficiency of the HPGe detector was estimated using the standard IAEA source to calibrate the detector prior to sample analysis. The absolute efficiency (ϵ_γ) of a HPGe detector at specific gamma energy is given by Equation (1) (ASTM, 2005).

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

where C_{net} is the source net count, A is the activity in Bq of gamma ray sources used in calibration and I_γ is absolute gamma decay intensity of specific energy peak (is the probability of emission per transformation for a photopeak specific energy) and T is the counting time (18000 s). Activity concentration (A_c) was calculated from analyzed using the equation (ASTM, 2005): as:

$$A_c = \frac{C_{net}}{\epsilon_\gamma \times I_\gamma \times m \times t} \quad (2)$$

where m is the mass of the sample. The unit of activity concentration of a soil sample is given as Bqkg^{-1} . The activity concentration was used to estimate radiological hazards indices such as Gamma dose rate (GDR), Outdoor and indoor annual effective dose rate (AEDE), External hazard index, and Excess life cancer risk (ELCR).

Gamma dose rate (D) - This is estimated as absorbed dose in the air at 1 m above the ground, determined by postulation that all the progenies of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their predecessors. This is given (UNSCEAR, 2008; Rani and Singh, 2005) as

$$D = 0.461A_{Ra} + 0.623A_{Th} + 0.0414A_K \quad (3)$$

Annual effective dose rate (AEDE) - The factors used are from D obtained from the with a conversion factor of 0.7 Sv/Gy of absorbed dose in the air. The occupancy factor (O_f) is the fraction of time spent indoors and outdoors and were estimated as 0.8 and 0.2, respectively (Al-Sulaiti, 2009; UNSCEAR, 2008):

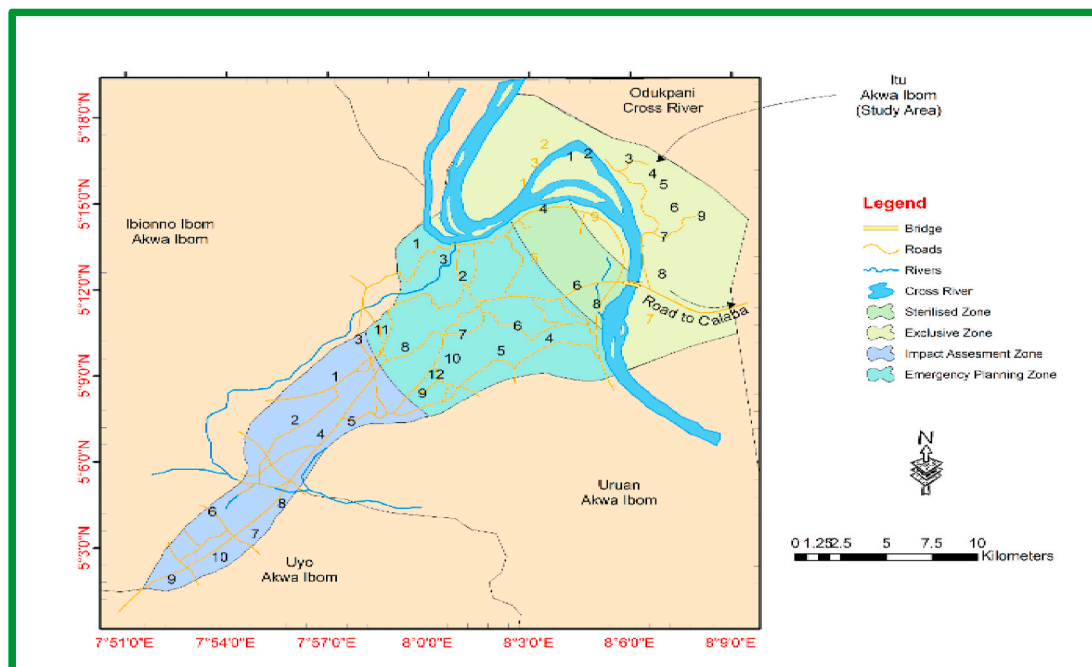


Fig. 2. Demarcated zones of NPP radiological monitoring zones and sampling points.

$$AEDE_{\mu Sv} = D_{nGy/h} \times 8760_{h/y} \times Of \times 0.7_{Sv/Gy} \times 10^{-3} \tag{4}$$

Excess life cancer risk (ELCR) is an estimation of values obtained from the product of determined AEDE with Duration of Life (DL, 70 years for children and 50 years for an adult), using (Taskin et al., 2009; Usikalu et al., 2011):

$$ELCR_{mSv/y} = AEDE_{mSv/yr} \times RF \times DL \tag{5}$$

Radium equivalent activity (Ra_{eq}) was calculated for relative purposes, assessing the hazard associated with material containing different concentrations of ^{226}Ra , ^{232}Th and 4K , Radium equivalent activity was estimated using the expression (Usikalu et al., 2011; Mantazul, 1979; Beretka and Mathew, 1985).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.007A_K \tag{6}$$

where A_{Ra} , A_{Th} and A_K are the specific activities of ^{226}Ra , ^{232}Th and 4K (in Bq/kg). In defining Ra_{eq} activity the assumption is made that 370 Bq/kg for ^{226}Ra , 259 Bq/kg for ^{232}Th and 4810 Bq/kg for 4K yields the same

gamma dose rate (UNSCEAR, 1982).

External hazards index (H_{ex}) was evaluated from natural gamma radiation and the prime objective is to limit the activity concentration (A) of ^{226}Ra , ^{232}Th and ^{40}K to ensure that a permissible dose rate of 1 mSv/y and is not exceeded by the expression (Beretka and Mathew, 1985; UNSCEAR, 1982; Al-Sulaiti, 2009).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1. (\text{without doors and windows}) \tag{7}$$

For the hazard to be considered as negligible, the H_{ex} value must be less than unity. However, the denominator values for A_{Ra} changes to 740 and A_{Th} changes to 540 with doors and windows (Hewamanna et al., 2001), also the denominator value for A_{Ra} changes to 185 if internal hazards are taken into account (Beretka and Mathew, 1985; Xinwei, 2005).

Table 1
Activity concentrations in soil samples from monitoring zones in the study area.

Code Name	Activity concentration (Bqkg ⁻¹)				
	²²⁶ Ra	²³⁸ U	²³² Th	⁴ K	¹³⁷ Cs
Exclusive Zone (EZ)					
EZ01	68.12 ± 12	23.61 ± 2	47.39 ± 5	38.72 ± 4	-
EZ02	83.35 ± 13	23.49 ± 2	51.41 ± 5	35.57 ± 4	-
EZ03	110.72 ± 14	41.95 ± 3	68.44 ± 6	84.20 ± 7	-
EZ04	42.19 ± 8	14.62 ± 1	31.67 ± 4	29.27 ± 4	-
EZ05	58.65 ± 12	22.46 ± 2	42.36 ± 4	36.04 ± 3	-
EZ06	56.52 ± 9	18.73 ± 2	34.59 ± 4	38.45 ± 4	-
EZ07	23.21 ± 7	9.03 ± 1	13.92 ± 2	28.46 ± 3	-
EZ09	26.46 ± 7	10.47 ± 1	17.81 ± 3	21.85 ± 3	-
Sterilized Zone (SZ)					
SZ01	75.80 ± 12	23.25 ± 2	40.15 ± 4	36.76 ± 4	-
SZ02	83.35 ± 13	23.49 ± 2	51 ± 4	35.59 ± 4	-
SZ03	41.79 ± 9	9.54 ± 1	15.19 ± 3	350 ± 20	-
SZ04	ND	3.83 ± 1	6.54 ± 2	120.40 ± 8	-
SZ05	85.82 ± 12	23.32 ± 2	53.11 ± 5	78.38 ± 6	-
SZ06	80.76 ± 14	31.15 ± 3	67.50 ± 6	40.98 ± 5	-
SZ07	125.60 ± 15	40.27 ± 3	62.92 ± 6	86.61 ± 7	-
SZ08	48.16 ± 9	20.48 ± 2	38.80 ± 4	36.42 ± 3	-
SZ09	75.46 ± 15	31.50 ± 3	56.60 ± 7	42.97 ± 5	-
Emergency Planning Zone (EP)					
EPZ 01	112.37 ± 15	45.99 ± 3	77.03 ± 7	128.11 ± 9	0.51 ± 0.2
EPZ 03	64.42 ± 11	21.42 ± 2	46.03 ± 5	34.62 ± 4	-
EPZ 04	63.18 ± 10	23.75 ± 2	44.66 ± 5	39.50 ± 4	1.02 ± 0.2
EPZ 05	66.30 ± 11	24.00 ± 2	48.93 ± 5	37.34 ± 4	-
EPZ 06	84.21 ± 9	25.46 ± 2	51.05 ± 6	45.89 ± 5	-
EPZ 07	54.68 ± 9	13.98 ± 1	26.79 ± 3	27.39 ± 3	0.37 ± 0.2
EPZ08	ND	5.83 ± 1	14.96 ± 2	22.26 ± 3	-
EPZ 09	52.07 ± 11	21.47 ± 2	44.94 ± 5	28.17 ± 4	1.65 ± 0.4
EPZ 10	54.16 ± 9	15.23 ± 1	27.10 ± 3	26.71 ± 3	0.79 ± 0.3
EPZ 11	63.86 ± 11	20.98 ± 2	37.95 ± 4	37.64 ± 3	-
EPZ 12	28.93 ± 7	11.14 ± 1	17.57 ± 3	20.64 ± 2	-
Impact Assessment Zone (IAZ)					
IAZ01	26.14 ± 8	10.78 ± 1	19.77 ± 3	30.69 ± 3	-
IAZ 02	44.10 ± 9	13.18 ± 1	26.26 ± 3	19.29 ± 3	-
IAZ 03	44.98 ± 10	12.72 ± 1	24.38 ± 3	28.33 ± 3	-
IAZ 04	45.34 ± 10	12.80 ± 1	20.51 ± 3	21.99 ± 3	-
IAZ 05	62.77 ± 12	24.77 ± 2	43.46 ± 5	39.35 ± 4	-
IAZ 06	137.31 ± 18	51.25 ± 4	109.27 ± 9	98.40 ± 8	-
IAZ 07	56.80 ± 9	22.08 ± 2	37.84 ± 4	44.54 ± 4	0.80 ± 0.2
IAZ 08	32.11 ± 9	16.40 ± 1	27.44 ± 4	32.58 ± 3	1.77 ± 0.2
IAZ 09	72.03 ± 11	18.35 ± 2	36.73 ± 4	38.87 ± 4	-
IAZ 10	53.30 ± 9	17.35 ± 2	29.33 ± 3	42.29 ± 4	0.87 ± 0.2
5 km Control					
Eki	56.04 ± 12	15.54 ± 1	28.80 ± 3.44	52.45 ± 5	-
Okolta	27.34 ± 8	15.76 ± 1	21.70 ± 2.91	19.97 ± 3	-
Anakpa	60.68 ± 12	15.53 ± 1	33.56 ± 3.56	34.94 ± 4	-
Uyo	38.63 ± 8	14.28 ± 1	18.39 ± 2.81	28.60 ± 3	-
Range	23.21 ± 7–110.72 ± 14	9.03±1–41.95 ± 3	13.92±2–68.44 ± 6	21.85 ± 3–84.20 ± 4	0.32 ± 0.1–2.97 ± 0.4
Mean	47.98 ± 8	15.15 ± 1	38.65 ± 6	41.55 ± 6	1.07 ± 0.3
World Mean	33	33	45	420	100

3. Results and discussions

3.1. Soil activity concentrations

The activity concentration (Bqkg⁻¹) for soil samples from the zones evaluated in Equation (2) are presented in Table 1 while mean activity concentrations for ²³⁸U radionuclides from the various radiological monitoring zones are presented in Fig. 3.

The activity concentration values for ²²⁶Ra ranged between 23.21 ± 7 Bqkg⁻¹ - 110.72 ± 14 Bqkg⁻¹ with average value of 47.98 ± 8 Bqkg⁻¹; the values for ²³⁸U ranged between 9.03±1–41.95 ± 3 Bqkg⁻¹ with average values of 15.15 ± 1 Bqkg⁻¹, ²³²Th range between 13.92 ± 2 Bqkg⁻¹- 68.44 ± 6 Bq/kg with average values of 38.65 ± 6 Bqkg⁻¹, and the values for ⁴K ranged between 21.85 ± 3 Bqkg⁻¹ - 84.20 ± 38 Bqkg⁻¹ with average values 41.55 ± 6 Bqkg⁻¹. These mean activity concentration values were lower than the admissible limits (UNSCEAR, 2000a). The low activity concentrations of ²²⁶Ra, ²³⁸U, ²³²Th, ⁴K implies that there are no human TENORM generated activities that have impacted on land, and may have altered the concentrations in the soil. However, the present concentration might be attributed to geological formations of the area (Njinga et al., 2015). Fig. 3 is a specific 3-Dimensional Arc-GIS map presentation of ²³⁸U activity concentration data obtained from Table 1 and was placed in NPP radiological monitoring zones of Fig. 2.

According to geological data, the study area is underlain by Imo formations and is located on the southeastern edge of the Anambra Basin. The analyses of minerals show that deposits of kaolinite, smectites, palygorskite, and other minerals can be found in rocks ranging in colour from medium grey to dark grey, such as clay/shale and sandstone (Okunlola and Egbulem, 2015). Furthermore, the Nigerian coastal plain is thought to cross through the Niger Delta sedimentary basins, a pro-grading depositional complex within the shape of the Abalaliki Trough in Eastern Nigeria to the Benin Flank in Western Nigeria, which opens up to the Atlantic Ocean. The sedimentary source in the area consists of mainly crystalline rock of Guinea high-lands basement complex along with cretaceous and tertiary sediments derived from Cameroun volcanic zone (Allen, 2011). These activity concentration findings are a reflection of what emanates as background ionizing radiation measurement of this study area, and results were within the

admissible limits (Ekong et al., 2019).

The ¹³⁷Cs radionuclide concentrations found in few locations in the area ranged between 0.32 ± 0.1–2.97 ± 0.4 Bqkg⁻¹ with mean 1.07 ± 0.3 Bqkg⁻¹ below admissible limits of 100 Bqkg⁻¹. However, its insignificant presence may either be from scrap metal recycling facility and/or may have been spread into the stratosphere and troposphere, then gradually return to the earth as radiological fallout after a nuclear test (IAEA, 2011; ICRP, 2002; IAEA, 2014a, 2014b; IAEA, 2005a; Martin et al., 2012; Ogundare and Nwankwo, 2015; Ekong et al., 2016). The mean activity concentration for all radionuclides under consideration of soil sample from all the radiological monitoring zones as well as the results of 5 km impact from 4 villages/LGAs surrounding Itu, Nigeria serving as control, was also found to be of low concentration (Fig. 4).

3.2. Radiological hazards evaluation

Table 2 presents evaluated radiological hazards indices considerations arising from analyzed activity concentration results were evaluated using Equations (3)–(8), which are but not limited to Gamma Dose Rate (GDR), Outdoor and Indoor Annual Effective Dose Rate (AEDR), Radium equivalents, External Hazard index and Excess Life Cancer Risk (ELCR).

The range of GDR from terrestrial gamma activity concentration for the entire four zones were 10.82–95.78 nGy/hr with a mean of 32.04 nGy/hr. The range of AEDE outdoor was 13.28–117.46 µSv/yr with a mean concentration of 42.98 µSv/yr. The range of AEDE indoor was 53.10–469.83 µSv/yr with mean of 171.91 µSv/yr; The Radium Equivalent arising from terrestrial gamma of activity concentration for entire four zones ranged 10.20–294.25 Bq/kg with a mean of 113.94 Bq/kg and ELCR ranged (0.05–0.41) × 10⁻³ with the mean of 0.15 × 10⁻³. These values were lower than the respective admissible limits as presented in Table 2 (Al-Sulaiti, 2009; UNSCEAR, 2000b; ICRP, 1990; UNSCEAR, 2000b). These low estimated values of radiological hazards indices considerations as regards to Gamma Dose Rate (D), Outdoor and Indoor Annual Effective Dose Rate (AEDR), Radium equivalent, External Hazard index and Excess Life Cancer Risk (ELCR) suggest that the radioactivity level in the area does not pose a significant threat to the health of the populace.

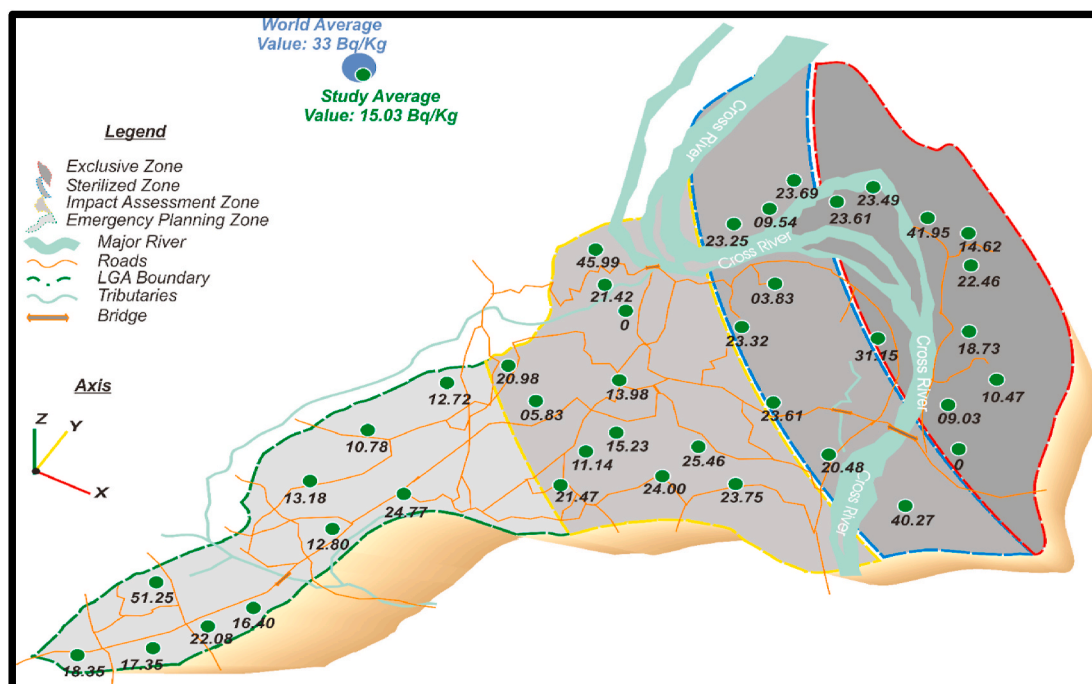


Fig. 3. A 3-Dimensional Arc-GIS map for ²³⁸U.

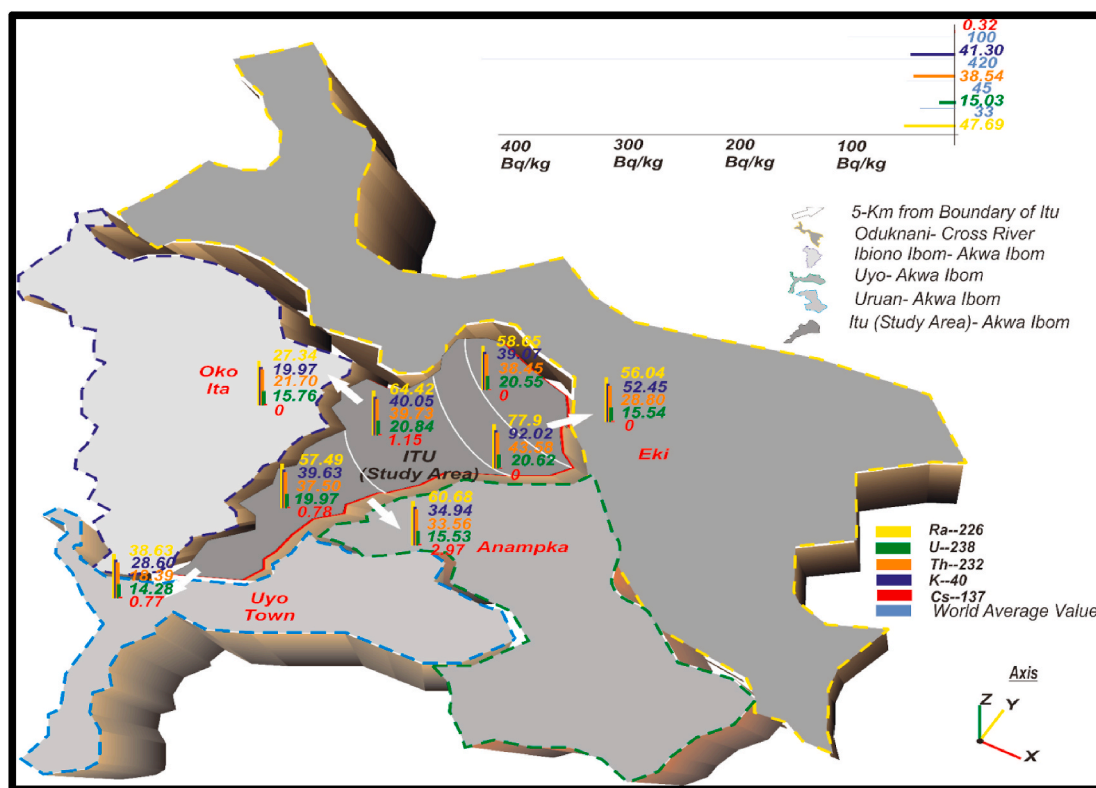


Fig. 4. A 3-Dimensional Arc-GIS map of mean activity concentration for radionuclides. in soil sample of the monitoring zones.

Table 2

Hazard Indices from activity concentrations at various zones.

ZONES	GDR (nGy/hr)	R _{req} (Bg/Kg)	H _{ex} (mSv/yr)	AEDR(O) (mSv/yr)	AEDR(I) (mSv/yr)	ELCR (10 ⁻³)
Exclusive Zone	35.04	113.91	0.32	0.042	0.17	0.15
Sterilized Zone	40.46	131.49	0.37	0.049	0.20	0.17
Emergency Planning	34.21	111.39	0.31	0.041	0.17	0.15
Impact Assessment	32.20	104.88	0.29	0.039	0.16	0.14
5 km control	24.41	82.54	0.23	0.042	0.12	0.10
Study Range	10.82–95.78	10.20–294.25	0.05–0.81	0.013–0.12	0.05–0.47	0.05–0.41
Study Average	35.04	113.94	0.32	0.043	0.17	0.15
World Average	59	370	1	0.46	0.46	0.29

Table 3 presents other radioactivity studies conducted in Nigeria and elsewhere. A study whose results are very close to this study was carried out randomly from some quarry site soil samples collected at different locations in Ayadehe, Oku Iboku, Odiok Itam and Ntak Inyang villages in Itu, Nigeria. The radionuclide activity concentrations measured in Bqkg⁻¹ for ⁴K, ²³⁸U and ²³²Th from quarry samples using gamma spectroscopy method with NaI(Tl) detector were found to be 143.54 ± 8, 2.47 ± 0.3, 3.70 ± 0.2, respectively for Ayadehe, 73.69 ± 4, 2.04 ± 0.2, 2.47 ± 0.3 respectively for Oku Iboku, 33.96 ± 2, 8.84 ± 0.9, 3.01 ± 0.2, respectively for Odiok Itam and 63.77 ± 3, 7.81 ± 0.8, 2.31 ± 0.1, respectively Ntak Inyang (Essien and Akpan (2016)).

These values were found to be low when compared to world mean values, which may be associated with geological formations, and the absence of substantial anthropogenic activities to increase the radionuclide concentration in the zone. The measured levels of radiological hazard exposures from the study areas were relatively low, posing no significant health risk; therefore, quarry products can be used as building materials (Essien and Akpan (2016)). The radionuclide activity concentrations and radiological hazards assessment associated with samples from various streams of metal recycling facilities have been identified in Delta State (Ogundare and Nwankwo, 2015). The activity

concentrations were found to be 12.93 ± 2, 19.00 ± 3 and 54.44 ± 3 for Bqkg⁻¹ of ²³⁸U, ²³²Th and ⁴K, respectively, which is lower than the world mean. These results are in agreement with the current study. Furthermore, the activity concentration of this present study is less than an evaluation conducted from farmland soil samples of ex-tin mining locations Jos Plateau Nigeria. Activity concentration of Bitsichi area is 163 ± 92, 451 ± 358 and, 455 ± 221 for ²²⁶Ra, ²³²Th and, ⁴K; Bukuru area 109 ± 28, 154 ± 56 and, 961 ± 263 for ²²⁶Ra, ²³²Th and, ⁴K; Ropp area 129 ± 65, 147 ± 75 and, 1052 ± 199 for ²²⁶Ra, ²³²Th and, ⁴K. From the evaluated results, of all the three farmlands were above the world means, which were attributed to mining activities (Jibiri et al., 2011).

Radiological evaluation was conducted in some soil samples from five states of South-western, Nigeria. The mean concentration of ²³⁸U in soil was 28.81 ± 4 Bqkg⁻¹; that of ²³²Th was calculated to be 5.93 ± 0.6 Bqkg⁻¹ while and ⁴K appeared undetected. The activity concentrations of the radionuclides in soil agreed with that of the present study and found to be lower than the world mean (Giwa et al., 2018). The estimation of the radioactivity of tin tailing samples collected from a mining site in Jos, Nigeria to determine their activity concentrations using a gamma-ray spectrometer with a HPGe detector has also been reported (Ademola, 2008). The ⁴K was below detectable limits in the studied

Table 3
Comparison of primordial radionuclides concentrations.

Locations	Evaluation Results (Bq/kg)			References
	²³⁸ U	²³² Th	⁴ K	
Assessment in South West, Nigeria	28.81 ± 4	5.93 ± 0.6		Giwa et al. (2018)
Metal recycling facilities, Niger Delta	12.93 ± 2	19.00 ± 3	54.44 ± 3	Ogundare and Nwankwo (2015)
Quarry site samples from three locations in Itu, Nigeria i.	2.47 ± 0.3	3.70 ± 0.2	143.54 ± 8	Essien and Akpan (2016)
Ayadehe	7.81 ± 0.8	2.31 ± 0.1	0.3	
ii. Oku Iboku,			33.96 ± 2	
iii. Odiok Itam			63.77 ± 3	
iv. Ntak Inyang				
Mining sites in Jos, Plateau Nigeria	72.2 × 10 ² (17.1 × 10 ² –16.6 × 10 ³)	16.8 × 10 ³ (52.9 × 10 ² –47.5 × 10 ³)		Ademola (2008)
Ex-tin mining locations in Jos plateau Nigeria from farmland i.	163 ± 92	451 ± 358	455 ± 221	Jibiri et al. (2011)
Bitsichi area	109 ± 28	154 ± 56	961 ± 263	
ii. Bukuru Area	129 ± 65	147 ± 75	1052 ± 199	
iii. Ropp area				
Itu, Nigeria	15.15 ± 1	38.65 ± 6	41.55 ± 6	Present Study
World mean	33	45	420	UNSCEAR (2000a)

samples. The activity concentrations of ²³⁸U ranged between (17.1 × 10²–16.6 × 10³) Bqkg⁻¹ with means of 72.2 × 10² Bqkg⁻¹ and ²³²Th ranged between (52.9 × 10² to 47.5 × 10³) Bqkg⁻¹ with means of 16.8 × 10³ Bqkg⁻¹. These comparison studies show that activity concentrations in areas where there are no radiological anthropogenic activities are found to be lower or within the world average concentration, whereas areas where radiological anthropogenic activities are found tend to indicate higher concentrations.

4. Conclusion

The natural activity concentrations of radionuclides (²³⁸U, ²³²Th, and ⁴K) measured by the HPGe detector were assessed in soil samples collected from designated zones in Itu, South-south Nigeria. The radionuclides' activity concentration indicated marked variability based on studied zones. A fission product (¹³⁷Cs) was also detected. The radiological hazard indices were calculated using the measured activity concentrations with mean GDR, outdoor, and indoor AEDE computed as 32.04 nGy/hr, 0.042 mSv/yr, and 0.17 mSv/yr, respectively. Most of these results suggest values that are less than the permissible limits. The low values observed may be attributable to the area's geology and geographical location, rather than to any anthropogenic activity. The values presented in this paper can be used as baseline radiological data in future analyses, as well as epidemiological studies of local population groups. Furthermore, if radiological or nuclear activities begin in the proposed region, periodic monitoring and assessment are recommended.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- Ademola, J.A., 2008. Exposure to high background radiation level in the tin mining area of Jos Plateau, Nigeria. *J. Radiol. Prot.* 28 (1), 93–99.
- Akwa Ibom State (AKS), 2014. Projected Population 2007 – 2015, vol. 1. Ministry of Economic Development Uyo, pp. 1–11.
- Allen, J.R.L., 2011. The Nigerian continental margin, bottom sediments, submarine morphology and geological evolution. *J. Mar. Geol.* 1 (4), 289–332.
- Al-Sulaiti, H., 2009. Determination of Natural Radioactivity Levels of the State of Qatar Using High-Resolution Gamma-Ray Spectrometry. MPhil to PhD Transfer Report. Department of Physics (University of Surrey).
- Anuforo, E., Onyedika, U.N., 2016. Nigeria Russia okay pact to build nuclear plants in Kogi, Akwa Ibom. *Guardian Newspaper online*. <https://guardian.ng/news/nigeria-> (Accessed 21 November 2018).
- American Society for Testing and Materials (ASTM), 2005. Standard guide for high-resolution gamma-ray spectrometry of soil samples. In: *Annual Book of ASTM Standard International, Standard Worldwide*, 12, pp. 1402–1404, 1.
- Ayoade, B.O., 1998. Adaptation to Climate Change in Agriculture, Forestry and Fisheries, Perspective Framework and Priorities. Food and Agriculture Organization of the United Nations Rome, pp. 149–155.
- Baltas, H., Yesilkanat, C.M., Kiris, E., Sirin, M., 2019. A study of the radiological baseline conditions around the planned Sinop (Turkey) nuclear power plant using the mapping method. *Environ. Monit. Assess.* 191 (11), 1–14.
- Barnekow, U., Fesenko, S., Kashparov, V., Kis-Benedek, G., Matisoff, G., Onda, Yu, Sanzharova, N., Tarjan, S., Tyler, A., Varg, B., 2019. Guidelines on Soil and Vegetation Sampling for Radiological Monitoring. International Atomic Energy Agency (IAEA). Technical Reports Series No. 486.
- Beka, J.E., Udom, G.J., 2014. Quality status of groundwater in Akwa Ibom State, Nigeria. *Int. J. Sci. Invent. Today* 3 (5), 436–449.
- Beretka, J., Mathew, P.J., 1985. Natural radioactivity of Australian building materials, industrial wastes and by-products. *Health Phys.* 48, 87–95.
- Cinelli, G., Tondeur, F., Dehandschutter, B., 2018. Mapping potassium and thorium concentrations in Belgian soils. *J. Environ. Radioact.* 184–185, 127–139. <https://doi.org/10.1016/j.jenvrad.2018.01.025>.
- Clark, C.E., Veil, J.A., 2009. Produced Water Volumes and Management Practices in the United States, ANL/EVS/R-09/1. Science Division, Argonne National Laboratory for the U.S. Department of Energy. Prepared by the Environmental.
- Ekong, G.B., Sambo, I., Sayaidi, S., 2016. Determination of radionuclides surface concentration and radiation level in fukushima prefecture, Japan. *Modern Environ. Sci. Eng.* 2, 757–764.
- Ekong, G., Akpa, T., Umaru, I., Lumbi, W., Akpanowo, M., Benson, N., 2019. Assessment of radiological hazard indices from exposures to background ionizing radiation measurements in South-south Nigeria. *Int. J. Environ. Monit. Anal.* 7 (2), 40–47.
- El Samad, O., Baydoun, R., Nsouli, B., Darwish, T., 2013. Determination of natural and artificial radioactivity in soil at North Lebanon province. *J. Environ. Radioact.* 125, 36–39. <https://doi.org/10.1016/j.jenvrad.2013.02.010>.
- Essien, I.E., Akpan, E.N., 2016. Evaluation of radiological hazard indices due to radioactivity in quarry sites in Itu, Akwa Ibom state, Nigeria. *Int. J. Sci. Res. Environ. Sci.* 4 (3), 71–77.
- Giwa, K.W., Osahon, O.D., Amodu, F.R., Tahiru, T.I., Ogunsanwo, F.O., 2018. Radiometric analysis and spatial distribution of radionuclides within the terrestrial environment of Southwestern Nigeria using ERICA tool. *Environ. Nanotechnol. Monitor. Manag.* 10, 419–426.
- Hewamanna, R., Sumithrarachchi, C.S., Mahawatte, P., Nanayakkara, H.L.C., 2001. Natural radioactivity and gamma dose from Sri Lankan clay bricks used in building construction. *Appl. Radiat. Isot.* 54 (2), 365–369.
- International Atomic Energy Agency (IAEA), 2005. *Radiological Conditions At the Former French Nuclear Test Sites In Algeria: Preliminary Assessment And Recommendations*, Radiological Assessment Report Series. IAEA, Vienna, Austria, pp. p1–59.
- International Atomic Energy Agency IAEA, 2007a. Terminology Used in Nuclear Safety and Radiation Protection. In: *IAEA Safety Glossary*. IAEA, Vienna, Austria.
- International Atomic Energy Agency IAEA, 2007b. Update of X-Ray and Gamma-Ray Decay Data Standards for Detector Calibration and Other Applications, vol. 1. IAEA, Vienna, Austria, pp. 4–47.
- International Atomic Energy Agency (IAEA), 2014a. *Managing Environmental Impact Assessment For Construction And Operation In New Nuclear Power Programme*, Nuclear Energy Series No. NG-T-3.11. IAEA, Vienna, Austria, pp. 1–30.
- International Atomic Energy Agency (IAEA), 2014b. *Radiation Protection and Safety Of Radioactive Source*, International Basic Safety Standard, vol. 3. General Safety Requirements Part, Vienna, Austria, pp. 1–110. IAEA.
- International Atomic Energy Agency (IAEA), 2015. Naturally occurring radioactive material (NORM VII). In: *Proceedings of Seventh International Symposium*. STI/PUB/1664, Beijing, China.
- International Commission on Radiological Protection (ICRP), 1990. *The Recommendations of the International Commission on Radiological Protection*, vol. 21. Publication 60, pp. 1–3. Annals.
- International Commission on Radiological Protection (ICRP), 2002. *Guide for the Practical Application of the ICRP Human Respiratory Tract Model*. Supporting Guidance 3, vol. 90. ICRP Publication, pp. 1–7, 32.

- Jibiri, N.N., Alausa, S.K., Owofolaju, A.E., Adeniran, A.A., 2011. Terrestrial gamma dose rate and physical – chemical properties of farm soil from ex-tin mining locations in Jos Plateau Nigeria. *Afr. J. Environ. Sci.* 5 (11), 1039–1049.
- Kathren, R.L., 1998. NORM sources and their origins. *Appl. Radiat. Isot.* 49 (3), 149–168.
- Knoll, G.F., 2010. *Radiation Detection and Measurement*, fourth ed. John Wiley and Sons, New York, pp. 123–506.
- Lilley, J., 2001. *Nuclear Physics-Principle and Applications*, second ed. Wiley, Chichester.
- Luiz do Carmo Leal, A., da Costa Lauria, D., Ribeiro, F.C.A., Viglio, E.P., Franzen, M., de Albuquerque Medeiros Lima, E., 2020. Spatial distributions of natural radionuclides in soils of the state of Pernambuco, Brazil: influence of bedrocks, soils types and climates. *J. Environ. Radioact.* 211 <https://doi.org/10.1016/j.jenvrad.2019.106046>, 106046.
- Manigandan, P.K., Chandar Shekar, B., 2014. Evaluation of radionuclides in the terrestrial environment of Western Ghats. *J. Rad. Res. Appl. Sci.* 7, 310–316. <https://doi.org/10.1016/j.jrras.2014.04.001>.
- Mantazul, I.C., 1979. Concentration of radionuclides in building and ceramic materials of Bangladesh and evaluation of radiation hazard. *J. Radioanal. Nucl. Chem.* 231 (2), 122.
- Martin, A., Harbison, S., Beach, K., Cole, P., 2012. *An Introduction Radiation Protection*, fifth ed. Hodder Arnold, an imprint of Hodder Education, Hachette UK, pp. p6–209.
- Missimer, T.M., Teaf, C., Maliva, R.G., Danley-Thomson, A., Covert, D., Hegy, M., 2019. Natural radiation in the rocks, soils, and groundwater of southern Florida with a discussion on potential health impacts. *Int. J. Environ. Res. Publ. Health* 16 (10), 1793. <https://doi.org/10.3390/ijerph16101793>.
- MoEF Ministry of Environment and Forests, 2010. *Environmental Impact Assessment Guidance Manual – for Nuclear Power Plants, Nuclear Fuel Reprocessing Plants and Nuclear Waste Management Plants*. Government of India, New Delhi, India, pp. 7–8.
- Movsisyan, N., Demirtchyan, D., Pyuskyulyan, K., Belyaeva, O., 2021. Identification of radionuclides' altitudinal distribution in soil and mosses in highlands of Armenia. *J. Environ. Radioact.* 231 <https://doi.org/10.1016/j.jenvrad.2021.106550>, 106550.
- Njinga, R.L., Ibrahim, Y.V., Ishoriyi, I.J., 2015. Radioactivity Analysis in underground drinking water sources in Niger State University of Nigeria. *Pollution* 1 (3), 315–324.
- Ogundare, F.O., Nwankwo, C.U., 2015. Radionuclide content of, and radiological hazards associated with samples from different streams of metal recycling facilities. *Radioprotection* 50 (1), 55–58.
- Okunlola, O., Egbulem, C., 2015. Geological setting, compositional and economic appraisal of shale occurrence in itu-mbonuso/iwere area, south-eastern Nigeria. *J. Geogr. Geol.* 7 (1), 85–96.
- Onwuemenyi, O., 2010. Agency Picks Four Sites for Nuclear Power Plants. Vanguard Newspaper online.
- Rani, A., Singh, S., 2005. Natural radioactivity levels in soil samples from some areas of Himachal Pradesh, India using γ -ray spectrometry. *Atmos. Environ.* 39 (34), 6306–6314.
- Sirin, M., 2020. Investigation of accumulation of radionuclides in different tissues of Whiting fish (*Merlangius merlangus euxinus* Nordmann, 1840) caught on the coasts of Rize in the eastern Black Sea region of Turkey. *Microchem. J.* 152 <https://doi.org/10.1016/j.microc.2019.104349>, 104349.
- Sohrabi, M., 1998. State-of-the-Art on worldwide studies in some environments with elevated natural occurring radioactive material. *Appl. Radiat. Isot.* 49 (3), 169–188.
- Taskin, H., Karavus, M., Topuzoghi, P. Ay, Hindiroglu, S., Karaha, G., 2009. Radionuclide concentrations in soil and lifetime cancer risk due to the gamma radioactivity in Kizilirmaci. *J. Environ. Radioact.* 100, 49–53.
- UNSCEAR, 1982. *Ionizing Radiation, Sources and Biological Effect* United Nations Scientific Committee on the Effect of Atomic Radiation (New York).
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000a. *Exposures from Natural Radiation Sources*. Annex B, New York.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000b. *Effects and Risks of Ionizing Radiations*. United Nations, New York.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), New York United Nations, 2008. *Sources and Effects of Ionizing Radiation*. United Nations, New York, ISBN 9211422426.
- Usikalu, M.R., Anoka, O.C., Balogun, F.A., 2011. Radioactivity measurement was of the tin mine of the Jos Plateau northern Nigeria. *Arch. Phys. Res.* 2, 80–86.
- Xinwei, L., 2005. Natural radioactivity in some building materials of Xi'an, China. *Radiat. Meas.* 40 (1), 94–97.
- Youdeowei, T., 2017. Nigeria signs pact with Russia on nuclear energy. Vanguard Newspaper Online. <https://www.vanguardngr.com/2017/11/nigeria-signs-pact-russia-nuclear-energy/>. (Accessed 21 November 2018).