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Assessment of geogenic natural radionuclide contents of soil samples collected from Ogun State, South western, Nigeria

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ABSTRACT

Background: Natural radionuclides are always present in the environment. Human exposure to the background radiation is inevitable. It is therefore important to assess health risk associated with these radionuclides. ***Materials and Methods***: The distribution of natural radionuclides ^{238}U, ^{232}Th and ^{40}K in soil samples collected from all the twenty (20) local Governments headquarter areas in Ogun state, Nigeria were determined by gamma spectroscopy using a high-purity germanium detector. The measured concentrations were used in estimating the radiological risk pose to people living in these communities. ***Results:*** The activity concentrations measured ranged between 3 ± 1 $Bqkg^{-1}$(Otta) to 27 ± 6 $Bqkg^{-1}$(odeda) for ^{238}U, 10 ± 1 $Bqkg^{-1}$(Otta) to 126 ± 6 $Bqkg^{-1}$(Ijebu Igbo) for ^{232}Th and 7 ± 6 $Bqkg^{-1}$(Aiyetoro) to 497 ± 1 $Bqkg^{-1}$(Odeda) for ^{40}K. The mean radium equivalent calculated was 77.6 $Bqkg^{-1}$ which is lower than 370 $Bqkg^{-1}$ of the world average and the hazard indices calculated were lower than unity. ***Conclusion:*** The absorbed rate and annual effective dose estimated for most of the locations are in good agreement with world average values except for Akomoje, Ake-Abeokuta, Ogere, Ijebu-Igbo and Odeda.

Keywords: *Natural radioactivity, absorbed dose rate, radium equivalent, hazard indices, Ogun State.*

INTRODUCTION

The environment is filled with radionuclides which can be dangerous if they are present at elevated concentrations. These radionuclides can be divided into two groups based on their sources, naturally occurring radionuclides such as ^{238}U, ^{232}Th and ^{40}K and artificial radionuclides such as ^{137}Cs. The naturally occurring radionuclides are present in the rock, soil and are easily transported into the environment through plants and water [1, 2].

The radiological impact from the above nuclides is due to radiation exposure of the body by the gamma rays and irradiation of the lung tissues from inhalation of Radon and its progeny [3]. From the natural risk point of view, it is necessary to know the dose limits of public exposures and to measure the natural environmental radiation level provided by ground, air, water, foods, building interiors, etc., for the estimation of the exposures to natural radiation sources. In assessing the radiation exposure, it is pertinent to determine the distribution of the radionuclide. The terrestrial component of the background is often due to various radioactive nuclides that are present in the air, soil, water and building materials whose abundances vary significantly depending on the geological and geographical features of a region. Nonetheless, the level of background radiation in a region is also considerably affected from

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man-made sources including those from nuclear activities and accidents [4]. The environment and health are interrelated. Health risks related to natural radioactivity are of great concern and require assessment in order to estimate the risks. Thus, the aim of this study is to measure the activity concentrations and estimate the radiological hazard indices in soil samples collected from all local government areas in Ogun State, Nigeria. The results obtained in this study will provide information on natural occurring radionuclides in soil in Ogun State, Nigeria and add to the existing data on radioactivity in soils in UNSCEAR data bank.

Study area

Ogun state is situated in the sub-humid tropical region of Southwest Nigeria with a tropical climate with distinct wet and dry season periods of about 130 days. It has twenty local government headquarters with a population of 3,751,140 people [5]. The mean annual rainfall and temperature are about 1,270 mm and 28°C respectively while the estimated mean annual potential evaporation is 1,100 mm. The geology of the study area has been extensively discussed by several authors [6, 7]. The Location is within the Dahomey basin and the stratigraphy of the basin has been grouped into six lithostratigraphic formations namely from oldest to youngest Abeokuta, Ewekoro, Akinbo, Oshosun, Ilaro and Benin formations (figure 1). Aizebeokhai and Oyeyemi (2014) [8] described the cretaceous Abeokuta formations as a group consisting Ise, Afowo and Araromi formations. The occurrence of phosphate rocks from Lower Eocene sedimentary phosphatic sediments have been known from southern Nigeria since 1921 [9, 10]. Phosphatic sediments occur between the Ifo junction and Oshosun in Southwest Nigeria, approximately 43 km and 48 km north of Lagos [10].

MATERIALS AND METHODS

Sample collection and preparation

Samples were collected by digging the ground to at least 3cm so as to take samples free from debris and vegetation. Five (5) soil samples were taken from different points at each location for better sampling. They were kept in ziplock bags and labelled accordingly making a total of one hundred (100) soil samples from all the locations. The samples were oven-dried at 110 ºC to ensure complete removal of moisture and passed through a 2mm sieve. 100g of each sample were placed in plastic radon-tight of 9 cm diameter vessels with a total capacity of 300 cm^3. The vessels were weighted and sealed for 30 days to allow secular equilibrium in the ^{238}U and ^{232}Th with their respective progeny [11].

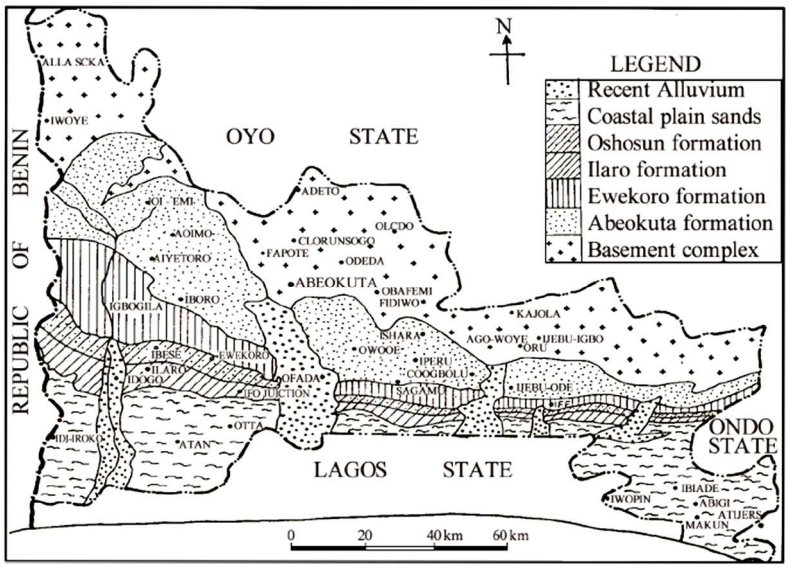

Figure 1. The map of Ogun State showing the geological formations at each sampling locations.

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Activity measurements

A 45 % efficiency p-type high purity germanium (HPGe) detector (Canberra model GC4520) coupled to a multichannel analyzer (MCA) was used for radioactivity measurements. The HPGe detector had an energy resolution of 2.2 keV (FWHM) for the 1332.5 keV gamma-ray transition of ^{60}Co source. The detector was situated in a well consisting of 5cm thick lead, to shield the measuring station from external background radiation [12]. The spectral for ^{238}U, ^{232}Th and ^{40}K were obtained using IAEA reference materials RGU-1 (4940 Bqkg^{-1} uranium ore); RGTh-1 (3250 Bqkg^{-1} thorium ore) and potassium chloride (16259 Bqkg^{-1} ^{40}K) from Merck company with 99.5 % purity. Each sample was placed on the detector and counted for 28,800 s with the same geometry as the reference materials. Background spectra were also collected for the same period of time. The net sample count rate at each energy peak was obtained after subtraction of the corresponding background rate. The absolute detection efficiency of the HPGe detector for gamma ray energy was calculated using equation (1)

$$\varepsilon = \frac{n}{tP_\gamma(E)N_o e^{-\lambda t_d}} \qquad (1)$$

where n is the net area under the full energy peak E is the gamma energy, t is the counting time , P_γ (E) is the gamma emission probability at energy E, N_o is the activity of the source (Becquerel), λ is the decay constant and t_d is the decay time. The activity concentration were calculated using the photo peaks corresponding to ^{226}Ra (186 keV), ^{214}Bi (1238 keV and 1378 keV), ^{214}Pb (295 keV and 351 keV) for ^{238}U; ^{208}Tl (860 keV), ^{228}Ac (338 keV, 911 keV, 969 keV) for ^{232}Th and 1460 keV for ^{40}K with palmtop MCA software according to equation 2:

$$\text{Activity (Bq kg}^{-1}) = \frac{\text{net area - B.G}}{t\varepsilon P_\gamma M} \qquad (2)$$

where Net Area is the net area under energy peak (count), B.G is the number of counts for the background spectrum, ε is the absolute efficiency of the detector, and M is the mass of the dried sample (kg).

RESULTS AND DISCUSSION

The average activity concentrations (Mean ± S.D) of ^{238}U, ^{232}Th, and ^{40}K for soil samples from the twenty [20] Locations are presented in table 1. The image maps of the radionuclides measurements in the area localising each soil sample with their geographical coordinates is presented in figures 2 and 3 using Surfer 11 software. Odeda has the highest level of ^{238}U and ^{40}K with values 27 ± 6 Bq kg^{-1} and 497 ± 6 Bq kg^{-1}, respectively while Ijebu Igbo (figure 2) has the highest concentration (126 ± 6 Bq kg^{-1}) of ^{232}Th. Otta has the activity concentration levels of ^{238}U and ^{232}Th with values 3 ± 1 Bq kg^{-1} and 10 ± 1 Bq kg^{-1} respectively while Ayetoro has the lowest concentration (7 ± 2 Bq kg^{-1}) of ^{40}K. The activity levels of ^{232}Th at Akomoje (109 ± 6 Bq kg^{-1}), Ake-Abeokuta (97 ± 5 Bq kg^{-1}), Ogere (56 ± 6 Bq kg^{-1}), Ijebu-Igbo (126 ± 6 Bq kg^{-1}) and Odeda 56 ± 5 Bq kg^{-1}), were higher than the global average (45 Bq kg^{-1}) for ^{232}Th in soil. The concentration of ^{40}K at Odeda (497 ± 6 Bq kg^{-1}) was higher than the global average (420 Bq kg^{-1}) for ^{40}K in soil [4]. ^{232}Th has the highest contribution to the environmental radioactivity in all the locations. The Th/U ratio in the samples is greater than unity in all the locations. This shows that thorium has high affinity of association to solid state mater and low geochemical mobility. All the locations with the high concentrations (Abeokuta and environs) are located within the basement complex. Basement complex is known to contain granites which have high concentrations of uranium, thorium and potassium [13, 14]. The uranium and thorium are incorporated into the rocks during crystallization of the last magmas and residual solutions as a result of their large ionic radii which hinder them from crystallizing in the early silicates. High activity concentration level of ^{40}K in Odeda may not have perhaps been from the denudation of the phosphatic rocks reported by [15] but probably from inorganic fertilizer used in agricultural activities. The continuous application of fertilizers in farms within this region enhances the radioactive levels which in turn increases the radionuclide concentration [16].

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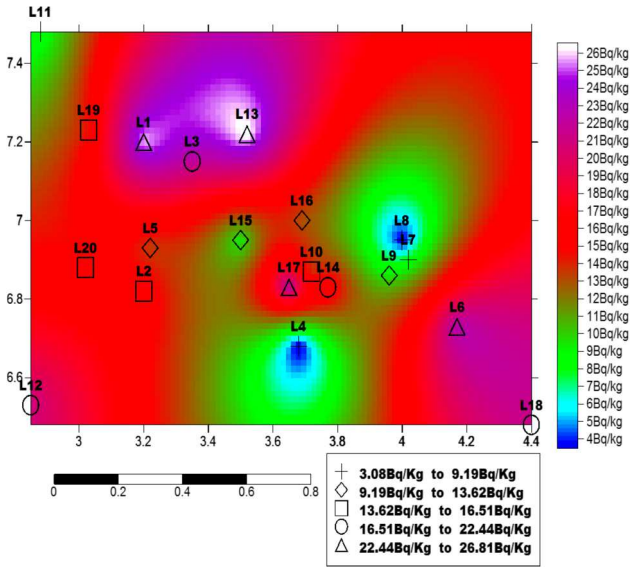

Figure 2. The image map of ^{238}U activity concentration for the study area.

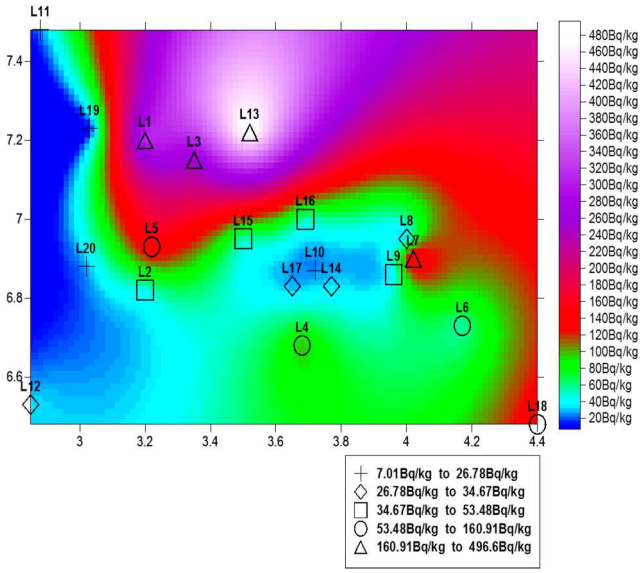

Figure 3. The image map of ^{232}Th activity concentrations for the study area.

Table 1. The activity concentration from twenty (20) locations in Ogun state, Southwest Nigeria.

Sample Codes	Location	Northing/ Easting	^{238}U(Bq kg^{-1})	^{232}Th(Bq kg^{-1})	^{40}K(Bq kg^{-1})
L1	Akomoje	7°12'N 3º12'E	26 ± 5	107 ± 6	312 ± 6
L2	Ifo	6°49'N 3º12'E	16 ± 4	23 ± 3	42 ± 3
L3	Ake-Abeokuta	7°09'N 3º21'E	22 ± 5	97 ± 5	252 ± 5
L4	Otta	6°41'N 3º41'E	3 ± 1	10 ± 1	97 ± 5
L5	Itori	6°56'N 3º13'E	13 ± 1	32 ± 3	151 ± 5
L6	Ogbere	6°44'N 4º10'E	23 ± 1	56 ± 6	53 ± 6
L7	Atan	6°54'N 4º01'E	8 ± 1	12 ± 3	161 ± 7
L8	Ijebu-Igbo	6°57'N 4º01'E	3± 1	126 ± 6	29 ± 5
L9	Ijebu-Ode	6°49'15"N 3º55'15"E	9 ± 1	13 ± 1	35 ± 5
L10	Ikenne	6°52'N 3º43'E	14 ± 2	13 ± 4	23 ± 5
L11	Imeko	7°29'N 2º53'E	8 ± 1	13 ± 3	21 ± 5
L12	Ipokia	6°32'N 2º51'E	21 ± 4	37 ± 6	32 ± 6
L13	Odeda	7°13'N 3º31'E	27 ± 6	56 ± 5	497 ± 6
L14	Odogbolu	6°50'N 3º46'E	17 ± 2	17 ± 5	33 ± 5
L15	Owode	6°57'N 3º30'E	9 ± 2	17 ± 5	51 ± 5
L16	Isara	7°00'N 3º41'E	13 ± 2	18 ± 5	50 ± 5
L17	Shagamu	6°50'N 3º39'E	22 ± 9	38 ± 7	27 ± 7
L18	Abigi	6°29'N 4º24'E	21 ± 4	28 ± 5	142 ± 5
L19	Aiyetoro	7°14'N 3º02'E	15 ± 3	20 ± 2	7 ± 2
L20	Ilaro	6°53'N 3º01'E	15 ± 1	23 ± 1	26 ± 6
Mean			**15.3**	**37.8**	**102.0**
Range			**3 - 27**	**10 -127**	**7 – 497**

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Estimation of radiological hazard indices

The radiological hazard index values estimated for the locations are shown in table 2 (column 2) and Radium equivalent activity (Ra_{eq}) [17] which is based on the estimation that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4180 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rate [19] expressed as equation (3)

$$Ra_{eq} = 370\ Bq\ kg^{-1} H_{ex} \quad (3)$$

H_{ex} is the external hazard index which is calculated by using Equation [7].

The Ra_{eq} estimated in this work ranges from 26 Bq kg^{-1} to 203 Bq kg^{-1} obtained from soils collected from Otta and Akomoje respectively. The values of Ra_{eq} calculated are lower than the world average maximum admissible value of 370Bq kg^{-1} [18].

In order to estimate the absorbed dose rates in air at 1m above the ground level equation [4] was used. The measured activity of ^{238}U, ^{232}Th and ^{40}K were converted into dose (nGyh^{-1}Bq^{-1}kg^{-1}) by applying the factors 0.462, 0.604, and 0.0417 for uranium, thorium and potassium respectively [19].

$$\text{Absorbed dose (nGyh}^{-1}) = 0.462C_U + 0.604C_{Th} + 0.0417C_K \quad (4)$$

C_u, C_{Th}, and C_k are the measured activity (Bq kg^{-1}) of uranium, thorium and potassium in the soil samples. Figure 4 represents the correlation of absorbed doses with the radium equivalent activity within the study areas. Regression analysis technique was used in drawing a trend line between the points. The regression result was high, positive and linear. The average absorbed dose rates calculated vary between 11.22 nGyh^{-1} (Otta) and 89.46 nGyh^{-1} (Akomoje). The mean absorbed dose rates obtained for Akomoje, Ake-Abeokuta, Ijebu-Igbo and Odeda exceeded the world average of 60 nGyh^{-1} [5] which may be attributed to the fact that the soils originated from igneous rock.

The annual outdoor and indoor effective dose equivalent H_E received by a member is calculated using equations 5 and 6 with a conversion factor of 0.7 SvGy^{-1}, applied to convert the absorbed rate to annual effective dose with an outdoor occupancy of 20% and 80% for indoors [20].

The indoor annual effective doses (table 2 column 4) were found to vary from 0.06 mSv y^{-1} to 0.44 mSv y^{-1} and the outdoor effective doses (table 2 column 5) vary from 0.01 mSv y^{-1} to 0.11 mSv y^{-1}. Akomoje, Ake-Abeokuta and Odeda exceeded the world average (0.07 mSv y^{-1}) for outdoor effective dose while only Akomoje slightly exceeded the world average (0.41 mSv y^{-1}) indoor effective dose recommended for general public [4] in the soil samples.

Indoor annual effective dose (mSv) = (Absorbed dose) nGyh^{-1}×8760h×0.8×0.7SvGy^{-1} (5)

Outdoor annual effective dose (mSv) = (Absorbed dose) nGyh^{-1}×8760h×0.2×0.7SvGy^{-1} (6)

$$H_\alpha = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (7)$$

$$I_\gamma = \sum_x \frac{C_x}{A_x} \quad (8)$$

Cx(Bq kg^{-1}) is the measured activity of each nuclide in the building material, A_χ(Bq kg^{-1}) is the activity concentration of each nuclide in the material and it is assumed to produce the same gamma dose rate, i.e. 300, 200, and 3000 Bq kg^{-1} for U^{238}, Th^{232}, and K^{40}, respectively. The estimation of external indices includes the determination of the external hazard index (H_{ex}), and gamma index ($I\gamma$) using equation 7 and 8. These indices are obtained in order to examine how suitable the soil materials are for construction. The (H_{ex}) values obtained in this study ranged between 0.07 obtainable at Otta and 0.55 (Akomoje) since the values are lower than unity (18, 20). Table 2 (column 7) showed that calculated $I\gamma$ is less than unity in the soil samples for all the locations.

The Internal index (I_α) which is a quantity used to determine the exposure level due to radon inhalation originating from materials [21] was obtained using equation 9.

$$I_\alpha = \frac{C_U}{200\ Bqkg^{-1}} \quad (9)$$

The alpha indices in the samples did not exceed the recommended limit given by International Commission on Radiological Protection, 1994 [22] (100 and 200 Bqkg^{-1}) as the

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concentration of ^{238}U (table 1) were below 200 Bq kg^{-1} in all the locations. This suggests that the soils are considered safe for construction for human accommodation purposes.

CONCLUSION

The natural radioactivity (^{238}U, ^{232}Th and ^{40}K) from soil samples collected from all the local Government areas of Ogun state, Nigeria were determined using high resolution HPGe gamma ray spectrometer. The highest measured activity levels for ^{232}Th, and ^{40}K of 126 ± 6 and 497 ± 6 Bq kg^{-1} were higher than the global average for soil were higher than the average world values for soil [4]. A strong correlation coefficient (0.998) is observed between the absorbed dose and the radium equivalent activity. The mean absorbed and annual equivalent dose rates obtained for Akomoje, Ake-Abeokuta, Ijebu-Igbo and Odeda exceeded

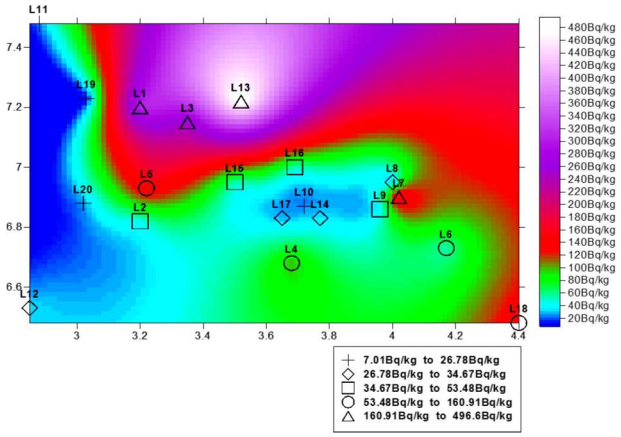

Figure 4. The image map of ^{40}K activity concentration for the study area.

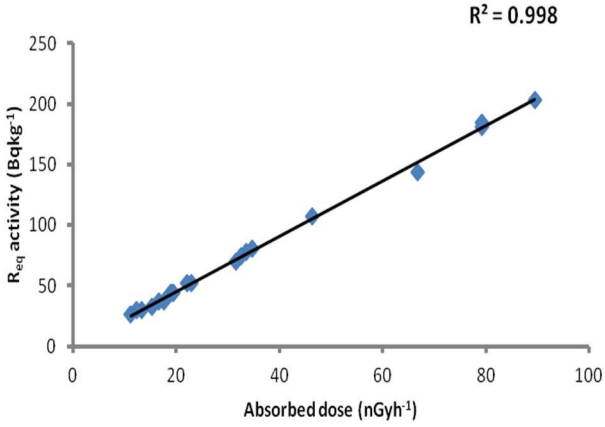

Figure 5. Variation of absorbed dose with Ra_{eq} activity.

Table 2. Radium equivalent, absorbed dose, dose equivalents and external-internal indices for soil samples.

Sample Codes	R_{eq} (Bq/kg)	Absorbed dose (n Gyh^{-1})	Indoor H_E (mSv)	Outdoor H_E (mSv)	External index (H_{ex})	Gamma index (I_γ)	Internal Index (I_α)
L1	203	89.46	0.44	0.11	0.55	0.72	0.13
L2	52	22.95	0.11	0.03	0.14	0.18	0.08
L3	181	79.26	0.39	0.09	0.49	0.64	0.11
L4	26	11.22	0.06	0.01	0.07	0.09	0.02
L5	70	31.63	0.16	0.04	0.19	0.25	0.07
L6	107	46.38	0.23	0.06	0.29	0.37	0.11
L7	37	17.68	0.09	0.02	0.1	0.14	0.04
L8	185	79.13	0.39	0.1	0.5	0.65	0.02
L9	30	13.34	0.07	0.01	0.08	0.1	0.05
L10	33	15.35	0.08	0.02	0.09	0.12	0.07
L11	30	12.4	0.06	0.02	0.08	0.1	0.04
L12	78	33.59	0.16	0.04	0.21	0.27	0.11
L13	144	66.7	0.32	0.08	0.39	0.53	0.13
L14	44	19.5	0.1	0.02	0.12	0.15	0.08
L15	37	16.65	0.08	0.02	0.1	0.13	0.05
L16	44	19	0.09	0.02	0.12	0.15	0.06
L17	81	34.7	0.17	0.04	0.22	0.28	0.11
L18	74	32.51	0.16	0.04	0.2	0.26	0.11
L19	44	19.27	0.09	0.02	0.12	0.15	0.07
L20	52	22.06	0.11	0.03	0.14	0.18	0.07
Mean	77.6	34.139	0.168	0.041	0.21	0.273	0.077
Range	26-203	11.22-89.46	0.06-0.44	0.01-0.11	0.07-0.55	0.09-0.72	0.02-0.13

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the world average of 60 nGyh^{-1} [4]. However, the radium equivalent R_{eq} values for the samples were found to be less than the upper recommended value of 370 Bq kg^{-1}. Thus, the study revealed that the health risk from natural background radiation from soil is low except for Akomoje, Ake-Abeokuta, Ijebu-Igbo and Odeda.

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Conflicts of interest: *none to declare.*

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