



Assessment of naturally occurring ⁴⁰K, ²³²Th and ²³⁸U and their associated radiological hazard indices in soils used for building in Ondo West Local Government area, Southwestern, Nigeria

Lasun T. Ogundele^{1*}, Patrick O. Ayeku², Samuel O. Inuyomi³, Oluwakunle M. Ogunsakin⁴, Olubusayo F. Oladejo⁵, Isaiah A. Adejoro⁶

- 1 Department of Physics, University of Medical Sciences, Ondo, Nigeria
- 2 Department of Biological Sciences, Wesley University, Ondo, Nigeria
- 3 Department of Physics and Engineering Physics, Obafemi Awolowo University, Ile-Ife
- 4 Department of Physical Sciences, Wesley University, Ondo, Nigeria
- 5 Department of Physics, Osun State University, Osogbo, Nigeria.
- 6 Department of Chemistry, University of Ibadan, Nigeria.

* Corresponding author e-mail: logundele@unimed.edu.ng

ARTICLE INFO

Received 20/6/2019; received in revised form 5/9/2019; accepted 8/10/2019. DOI: <u>10.6092/issn.2281-4485/9473</u> © 2019 The Authors.

Abstract

The naturally occurring ⁴⁰K, ²³²Th and ²³⁸U radionuclide contents in the soil used as building materials in Ondo West Local Government, Southwest Nigeria were determined using gamma spectrometric technique. The radiological hazards were estimated by employing several indices consisting of radium equivalent (Raeq), representative level index (RLI), activity utilization index (AUI), absorbed dose (D), annual effective dose, external hazard index (H_{ex}), internal hazard index (H_{in}) and gamma index (I_y). The average activity concentrations of ²³⁸U, ⁴⁰K, ²³²Th and Raeq were 171.8, 146.2, 19.8 and 211. 3 Bqkg⁻¹ while the average values of H_{in}, H_{ex}, RLI, D, AED, ELCR, AUI and I_y were, 1.0, 0.6, 0.2, 97.4, 0.5, 3.9 x 10⁻³, 1.8 and 0.7, respectively. The multivariate statistical analysis was employed to identify the relationships between the radionuclides and the estimated radiological hazard parameters. The estimated radiological indices were within the internationally acceptable limits confirming the safe use of these soil for building construction for human dwelling without any radiological implications.

Keywords

natural radionuclide, soil, exposure, radiological indices

Introduction

Soil is a major source of human exposure to radiation and medium of migration for the transfer of radionuclides within the environment and to human body (Ahmad et al., 2015; Senthilkumar and Narayanaswamy, 2016; Alajeeli, et al., 2019). Apart from serving as a medium for plant growth and hydrological buffer, it is used as one of the major materials in construction of building and other physical infractstructures. Most of the building materials such as cement, sand, limestone, granite, marble and soil had been reported to contain varying quantity of naturally occurring radionuclides (Mavi and Akkurt, 2010; Ademola et al., 2014; Raghu et al., 2017). The presence of radionuclide in building materials most especially soil can be source of internal and external human exposure to gamma ray from both short and long life radioactive decay activity. Natural radionuclides are formed through the interaction of the high energy rays from the sun with the atmospheric

particles (cosmogenics) or at the creation of the earth (primordial) (Alajeeli et al., 2019). The primordial radionuclides had been reported to be the most significant sources of over 60 radioactive elements most especially ²³⁸U and ²³²Th decay chains and ⁴⁰K (Akozcan, 2014) and they remain the largest contributor (85 %) to external dose among the exposed population (Mavi and Akkurt, 2010). It contributes approximately 85 % of the total global average annual radiation (UNSCEAR, 2001) while the remaining 15 % might be from cosmic rays and nuclear process (Ademola et al., 2014). A large percentage of the total global average annual radiation might be from the building materials, most especially soil. Several authors had reported soil as the basic indicator of radiological contamination in the environment (Karahan and Bayulken, 2000; Xinwei et al., 2006; Ravisankar et al., 2014; Feroz and Sajad, 2015; Senthilkumar and Narayanaswamy, 2016; Sivakunar, 2014; Raghu et al., 2017). The knowledge of the level of the radionuclides content in the soil is very crucial in evaluation of the absorbed dose that can produce radiological hazard, leading to severe health problems among the exposed population.

Soil is most frequently and quantitatively utilize in large quantities in building construction and the inhabitants of such building spend several years in the buildings most likely to be exposed to the radiation from naturally occurring radionuclides present in the soil. As a result, there is a need to evaluate the radiological hazard in the population living in such houses (Ravisankar et al., 2014). The gamma radiation from the soil used in building is a source of external radiation exposure route while inhalation of radon gas (a potentially dangerous radioactive element) is the principal route of internal exposure to humans (Ademola et al., 2014). Therefore in some cases, reference is often made to ²²⁶Ra being a short live decay product of ²³⁸U (Ravisankar et al., 2012). In an indoor micro-environment, building materials and indoor-outdoor gaseous exchange influence the activity concentration as well as the decay products of radionuclides. The epidemiological studies on external and internal radiation exposure from natural radionuclides had indicated severe implications and detrimental impacts on human health (Xinwei et al., 2006; Gao et al., 2014). These effects include potentially harmful genetic damage, cancer incidence and other health outcomes such as benign tumors, cataract, diarrhea, leukemia, loss of appetite, malaise, vomiting, scarring, skin redness and diarrhea. Depending on the dose of radiation received or the absorbed dose rate, the health implications such as cardiovascular disrupt, gastrointestinal breakdown, hematopoietic and central nervous system dysfunction can also occur (USEPA, 2017). The radiological hazard vary from an absorbed dose depends on the type of radiation and the sensitivity of different tissues and organs. Pregnant women and children are especially vulnerable to radiological hazards. The cells in children and fetuses divide rapidly, providing more opportunity for radiation damaging consequences and cause cellular dysfunction. The assessment of activity concentration of naturally occurring radionucles in the soil used for building had drawn the attention of researches worldwide (Sroor et al., 2001; Darko et al., 2005; Xinwei et al., 2006; Tufail et al., 2007; Mavi and Akkurt, 2010; Ravisankar et al., 2012; Hassan and Khoo, 2014; Usikalu et al., 2014; Raghu et al., 2017). In most of these studies showed a large variation of radioactivity concentrations from one country to another

The detailed assessment of the radioactivity and radiological hazard of 238U, 232Th and 40K in the soil of the studied area is lacking in the literature. The present study therefore determines the radioactivity level of ²³⁸U, ²³²Th and ⁴⁰K in the soil samples obtained from Ondo, Southwestern Nigeria. This was with the aim of establishing baseline information on radioactivity level in the soil commonly used as building materials in the study area and to estimate the potential radiological hazards by employing the various indices such as Radium equivalent activity (Ra_{ed}), representative level index (RLI), activity utilization index (AUI), absorbed dose (D), external hazard index (H_a), internal hazard index (H_{in}), annual effective dose (AED), excess life cancer risk (ELCR) and gamma index (I_y) . The obtained results were compared to the world average and corresponding values of 238 U, 232 Th, 40 K and Ra_{eq} soil from different countries. The activity concentrations of ²³⁸U, ²³²Th, ⁴⁰K and the estimated radiological indices were subjected to multivariate statistical analysis in order to establish the relationship among them.

Materials and Method

Sample collection and preparation

A total of thirty soil samples were collected at the functional excavating pits in Ondo West Local Government area, Ondo, Nigeria where soils used for building materials are obtained at 0 - 20 cm depth. For each sample at each site, 5 representative samples were taken, pooled and homogenized to form a representative sample. The soil samples were packed in polythene

bags, coded serially (S1 to S30) for easy identification and they were brought to the laboratory for further preparation. In the laboratory, the samples were dried at ambient temperature for about two weeks. The organic materials, plant debris, coarse stones, gravel lumps and pebbles were removed from each sample. Afterward, they were crushed and grounded using agate mortar and then sieved through a 150-mm mesh. In order to completely remove the moisture content of the samples and obtain a constant weight, the powdered samples were oven dried at 110°C. Prior to gamma spectrometric measurement, two hundred grams (200 g) of each of the processed soil samples were weighed and packed into cylindrical plastic (polyvinylchloride) containers (6 cm diameter and 7 cm height) and uniform mass, hermetically sealed with adhesive masking tape in a radon impermeable airtight condition. The samples were then stored for a period of 40 days so as to ensure secular equilibrium between ²²²Ra and its short-lived progenies with ²²⁶Ra (Ravisankar et al., 2012; Senthilkumar and Narayanaswamy, 2016).

Gamma spectroscopy

The activity concentration of natural radioactivity in the soil samples were measured using a Cs(Ti) detector and a multi-channel analyzer (model URSA II) was used to record the gamma spectra. A custom made lead shielding array was employed to reduce the background radiation by a factor of about 95 %. The qualitative and quantitative analysis of the radionuclides present in each sample was conducted by energy calibration using standard sources of known gamma ray energies and activities. The activity concentrations of radionuclides were determined in Bqkg-1 using the count spectra obtained from each of the samples. The gamma ray photo peaks corresponding to energy of 1460 keV for 40K, 352 keV (²¹⁴Pb) for ²⁸³U and 583 kev (²⁰⁸Tl) for ²³²Th were considered to determine the activity of ⁴⁰K, ²³²Th and ²³⁸U in each soil samples. The counting time for each sample was 36,000 s (10 hours). Each sample was counted twice and the average was then taken. The gamma spectrometric measurement was conducted at the Biological Trace Element Laboratory, Department of Physics and Engineering Physics, Obafemi Awolowo University, Ile Ife, Nigeria.

Determination of Radium equivalent and radiation hazards indices

Radium equivalent. Radium equivalent (Ra_{eq}) is common radiological index use to estimate the actual activity level of radionuclides in the samples and

their associated radiation hazards (Mavi and Akkurt, 2010). The ⁴⁰K, ²³²Th and ²³⁸U radionuclides in the soil samples under investigation are not uniformly distributed. Therefore, Ra_{eq} is a universally accepted single index which represent the weighted sum of the activity concentration of primordial radionuclides of ⁴⁰K, ²³²Th and ²³⁸U in the soil samples based on the fact that 10 Bq kg⁻¹ of ²³⁸U, 7 Bqkg⁻¹ of ²³²Th and 130 Bq kg⁻¹ ⁴⁰K produce an equal gamma ray dose (Laili et al., 2012). In order to estimate the Ra_{eq} in the soil used as building materials in the study area, equation [1] was used (Gbenu et al. 2016; Kaliprasad et al. 2017).

$$Ra_{eq} = 1.43A_{Th} + A_{U} + 0.077A_{K}$$
 [1]

where

 A_{Tb} , A_U and A_K are the activity concentrations of ²³²Th, ²³⁸U and ⁴⁰K in Bqkg⁻¹, respectively.

Internal bazard index. Inhalation of gaseous radionuclide and its short-lived progeny had been reported to be hazardous to respiratory organs from the inhalation exposure route (Ademola et al., 2015; Gbenu et al., 2016; Kaliprasad et al., 2017). The hazardous impact of short-lived products was quantified by the internal hazard index (H_{in}) following the equation [2] developed by (Krieger, 1981):

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1$$
[2]

where

 A_{Th}, A_U and A_K are the activity concentrations of ⁴⁰K, ²³²Th and ²³⁸U in Bqkg⁻¹, respectively. For a negligible internal radiation hazard and a safe use of the soil material for in the construction of building, the estimated H_{in} should also be less than unity (Senthilkumar et al., 2014).

External bazard index. The external hazard index (H*ex*) is the excess gamma radiation emanating from the soil samples and it was assessed using the widely employed hazard index equation introduced by Bereka and Mathew (1985). In order to ensure the safe limit as well as the suitability of soil as building materials and to ascertain that the radiation hazard is kept insignificant, the H_{cr} must be less than unity.

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1$$
[3]

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

Representative level index. The representative level index (RLI) is primarily denotes the level of gamma radioactivity associated with different concentrations of some specified radionuclide measured in the sample matrix and it is estimated using equation [4]:

$$RLI = \frac{A_U}{150BqKg^{-1}} + \frac{A_{Th}}{100BqKg^{-1}} + \frac{A_K}{1500BqKg^{-1}}$$
[4]

where A_U , A_T and and A_K are the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K, respectively The maximum permissible limit for RLI is 1 (Alam et al., 1999).

Absorbed dose rate. To quantify the outdoor absorbed dose rate (nGyh⁻¹) in air from the terrestrial sources of gamma radiation in the soil samples at 1 m above the ground surface for uniform distribution of the studied naturally occurring radionuclide (Gbenu et al., 2016), the activity concentration of A_U , A_T and A_K were used. This is based on the assumption that contribution of other naturally occurring radionuclide like ²³⁵U, ¹³⁸La, ¹⁴⁷Sm, ⁸⁷Rb and ¹⁷⁸Lu to actual dose rates are less important (Senthilkumar et al., 2014). The formula developed by the United Nations Scientific Committee on Effect of Atomic Radiation (UNSCEAR, 2010) and the European Commission (EC, 1999) were adopted to determine the gamma ray emission from the studied soil samples. The conversion coefficient of 0.462, 0.0417 and 0.604 nGyh $^{\scriptscriptstyle 1}$ for $^{\scriptscriptstyle 238}\text{U},~^{\scriptscriptstyle 232}\text{Th}$ and $^{\scriptscriptstyle 40}\text{K},$ respectively were used in the computation of absorbed dose rate from the activity concentration by employing equation [5]:

 $D = 0.64A_U + 0.4462A_{Th} + 0.0417A_{K}$ [5]

where A_U , A_T and and A_K are the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K, respectively.

Annual effective dose. The indoor exposure to gamma rays due to naturally occurring ²³⁸U, ²³²Th and ⁴⁰K in the measured soil samples was determined due to the fact that indoor exposure inherently higher than that of the outdoor provided the earth materials are used for construction (UNSCEAR, 2000). A factor 0.8 was used as the indoor occupancy factors (meaning that on the average around the world, 80 % of time is spent indoors) while and 0.7 nGyh⁻¹ was the conversion coefficient from absorbed dose (D) in air to effective dose received by adults. The indoor component of the annual effective dose, AED (mSvy⁻¹) was estimated as follows equation [6]:

 $AED = D * 24hours * 365 days * 0.8 * 0.7 * 10^{-6}$ [6]

Excess Lifetime Cancer Risk. Low doses of ionizing radiation can increased the risk of cancer can be increased in ionizing radiation even at low dose. The risk of cancer increases as the dose of radiation increases. Exposure to one Sievert of radiation spread out over time is estimated to increase the lifetime risk of fatal cancer in an average adult by around 4 % and a 0.8 % chance of hereditary defect in future offspring. The excess lifetime cancer risk (ELCR) due to external gamma dose exposure in the soil used for building was calculated as follows equation [7] (Jaillad, 2016; Mohammed and Ahmed, 2017):

ELCR = AED * ALE * RF[7]

where AED, ALE and RF are annual effective dose, average life expectancy of the exposed population (70 year) and RF is the fatal risk factor (0.005 Sv⁻¹), respectively. For stochastic effects due to low dose background radiation, RF value of 0.057 was used in this study as suggested by International Commission on Radiological Protection (ICRP-103) for public exposure (ICRP, 2007).

Activity utilization index. The activity utilization index (AUI) is the dose rates in air obtained from the combination of ⁴⁰K, ²³²Th and ²³⁸U(Bq kg⁻¹) in soil samples. Some suitable conversion factors are applied in calculating AUI. The AUI values of the soil samples were calculated using equation [8] (El-Gamal et al., 2007).

$$4UI = \frac{A_U}{50} f_U + \frac{A_{Th}}{50} f_{Th} + \frac{A_K}{500} f_K \quad [8]$$

The exception criterion and upper limit dose 0.3 and 1 mSvy⁻¹ were introduced by the European Commission for gamma dose of the building materials. The upper limit is mostly applied by most countries. The value of AU*I* should be below 0.5 for materials used in bulk. If the upper level of unity is, then the values of AUI should be below 1 for such materials.

Multivariate statistical analysis (Correlation and Factor analysis)

Correlation analysis statistical tool used to measure the strengths of association between two variables. In statistics, correlation coefficient is used as a measure of relationship between two variables. The correlation coefficient varies between ±1. Spearman's correlation coefficient is a statistical measure of the strength of a

monotonic relationship between paired data. In this study, Spearman rank correlation was used. It is an alternative to Pearson correlation coefficient when nonparametric statistics is involved (Gbenu et al., 2016). The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K and the evaluated variables Ra_{ea}, H_{in}, H_{ex}, RLI, D, AED, ELCR, AUI and I_{γ} were subjected to Spearman's correlation analysis in order to determine the mutual relations and the degree of association between pairs of variables. Factor analysis is a multivariate statistical method that is conducted to describe the underlying dimensions of the variables. Factor analysis provides a unique solutions and the interpretation of the factor output is straight forward. In factor analysis approach, only the factors with eigenvalues greater than 1 are extracted and factor loading above 0.5 (50 % association) are interpreted (Raghu et al., 2017). The fundamental equation, basic principle and procedure of factor analysis can be found elsewhere (Marcazzan et al., 2003; Pekey et al., 2005; Kothai et al., 2008; Ogundele et al., 2018).

Results and discussion

Activity concentrations and Radium equivalent results

The measured average values together with their respective standard deviations (±SD) of activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th are presented in Table 1. The activity concentration of ⁴⁰K varied from 94.1 to 267.9 Bqkg⁻¹ with the average of 146.2 Bqkg⁻¹. The activity values of ⁴⁰K in all the samples were less than the set limit 370 Bqkg⁻¹. The activity concentrations of ²³⁸U varied from 98.9 to 303.9 Bqkg⁻¹ and it has a mean of 184.8 Bqkg⁻¹. All the samples were 3 to 6 times the criteria 35 Bqkg⁻¹ for ²³⁸U. In case of ²³²Th, the activities concentration ranged from 12.6 to 32.3 Bqkg⁻¹ with the average of 19.7 Bqkg⁻¹. The variation in activity concentration could be related to differences of geological structures of the area. The average ratio of ⁴⁰K/²³⁸U is 0.8, signifying high activity concentration of ²³⁸U compared to ⁴⁰K. The higher activity concentration of ²³⁸U compared to ⁴⁰K is consistent with IAEA report (IAEA, 2007). The high activity concentrations of ²³⁸U might be traced to geochemistry of the soil of the study studied area (Mir and Rather, 2015). The average value of ⁴⁰K in this study is similar to the values obtained for Nigeria soil samples but several order of magnitude higher than the values reported by Tufail et al. (2007) for Pakistan soil samples. The ²³⁸U, ²³²Th and ⁴⁰K were greater than the values stipulated by United Nations Scientific Committee on the Effects of Atomic

Radiation (UNSCEAR, 2010) stipulated the world mean values of 226 Ra, 232 Th and 40 K as 50, 50 and 500 Bqkg⁻¹.

The radionuclide that could probably contribute to radiological hazard from the soil

Code	⁴⁰ K	²³⁸ U	²³² Th	Ra _{eq}
S1	105.8	144.0	15.8	174.8
S2	243.3	126.4	21.5	175.9
S3	102.3	145.0	16.7	176.8
S4	104.5	149.1	15.4	179.2
S5	168.2	25.9	23.2	72.0
S6	131.4	18.9	17.7	54.4
S7	124.0	178.3	17.6	213.0
S8	134.0	208.7	20.4	248.1
S9	117.5	164.5	15.8	196.1
S10	191.7	270.2	25.0	320.6
S11	112.3	171.4	16.3	203.3
S12	201.6	293.1	26.7	346.9
S13	104.1	177.1	19.7	213.3
S14	137.5	158.8	16.4	192.9
S15	129.7	216.2	21.6	257.1
S16	177.8	224.7	21.0	268.5
S17	102.8	130.2	12.6	156.2
S18	175.6	303.9	23.1	350.5
S19	94.1	134.0	13.6	160.7
S20	133.1	211.5	22.4	253.8
S21	134.9	180.0	16.4	213.8
S22	186.5	98.9	19.6	141.2
S23	111.9	167.6	17.2	200.8
S24	196.9	125.3	23.2	173.7
S25	108.0	165.6	18.9	200.9
S26	119.7	131.9	14.8	162.2
S27	109.3	161.5	16.7	193.9
S28	205.1	301.6	29.5	359.6
S29	268.0	143.1	32.3	209.9
S30	154.4	225.4	22.3	269.2
Mean	146.2	171.8	19.8	211.3

Table 1. Activity concentration and Radium equivalentresults (Bqkg⁻¹).

Country	⁴⁰ K	²³⁸ U	²³² Th	Ra	References
Australia	62.9	162.8	403.3	326.7	Bereka and Mathew, (1985)
China	44.0	47.0	593.1	156.8	Ziqiang et al. (1988)
Turkey	317.0	210.0	342.0	-	Karahan and Bayulken, (2000)
Egypt	13.0	6.0	433.0	-	Sroor et al., (2001)
China	40.7	21.5	302.6	96.4	Xinwei et al., (2006)
Pakistan	46.5	60.8	698.6	187.2	Tufail et al. (2007)
India	6.5	19.0	314.9	57.9	Ravisankar et al., (2014)
Nigeria	19.7	18.1	167.2	58.5	Nwaka et al., (2001)
Malaysia	16.8	20.1	38.5	48.5	Hassan and Khoo, (2014)
Malaysia	60.7	70.2	229.1	185.8	Solehah et al., (2016)
India	116.1	43.5	300.1	201.5	Raghu et al. (2017)
Nigeria	146.2	171.9	19.7	211.3	This study

Table 2. Comparison of 238 U, 232 Th, 40 K activity concentrations and Ra_{eq} with some other studies around the world.

of the studied areas is ²³⁸U considering its high activity concentration compared to that ⁴⁰K and ²³²Th. The calculated radium equivalent activity (Ra_{eq}) values ranges from 54.4 to 359.6 Bqkg⁻¹ with a mean value of 211.3 Bqkg⁻¹. In all the soil samples, the estimated Ra_{eq} values were lower than the 370.0 Bqkg⁻¹ stipulated as the safe limit for soil as building materials. The average value of *Ra_{eq}* obtained in this study were found to be higher than values reported by Hassan and Khoo (2014) (48.5 Bqkg⁻¹), Nwaka et al. (2001) (58.5 Bqkg⁻¹), Ravisankar et al. (2014) (57.9 Bqkg⁻¹), Xinwei et al. (2006) (96.4 Bqkg⁻¹) and Tufail et al. (2007) (187.2 Bqkg⁻¹) in other countries. However, the Ra_{eq} values were lower than 326.8 Bqkg⁻¹ reported by Bereka and Mathew (1985).

Radiological indices results

Table 3 shows the results of the estimated values of the radiological indices (H_{in}, H_{ex}, RLI, D, AED, ELCR, AUI and I_y). The values of H_{in} in soil samples were closed to maximum permissible value of unity except in some few samples (S5, S10, S12, S18 and S28). The measured H_{er} ranges from 0.4 to 0.9 with the mean of 0.6. All the H_{ex} values were below the criterion (≤ 1). For the RLI values, it was observed that most of the sample had the RLI values close to 1 except some few samples S5, S8, S10, S12, S15, S16, S18, S28 and S30. The value of absorbed dose rate vary from 24.9 to 165.7 nGyh⁻¹.. The average value of the absorbed dose was 97.4 nGyh⁻¹ which was about 3.5 times the recommended world average value of 60 nGyh⁻¹ (UNSCEAR, 2000). The differences could arise from the geological setting of the studied area, soil physical and chemical properties which are common phenomenon in assessment of radiation in the environment (Usikalu et al., 2014). The annual effective dose ranges from 0.12 to 0.80 mSv⁻¹. The mean value of annual effective dose is 0.47 mSvy⁻¹ which is quite less than 1.0 mSv stipulated by UNSCEAR (2000) as the maximum tolerable dose from building materials. Mavi and Akkurt (2010) found that dose rates higher than 1 mSvy⁻¹ should be permitted only in some very exceptional cases where materials are used locally. The estimated average of excess lifetime cancer risks as a result of the level of radionuclides in the soil samples is 3.9×10^{-3} . This value is lower than the world average value of 2.9×10^{-3} recommended by (UNSCEAR, 2000) due to low level radiation exposure from the soil. About two-third of the total soil samples had the AUI to be less than 2, signifying the safety use of soil in the studied area for building construction. The I_y values vary from 0.70 to 1.22. The average value I is 0.72, which was less than the recommended value (≤ 1). This implies that the gamma activity in soil samples does not exceed the established criterion level, hence pose no significant radiological hazards when used in the construction of dwelling.

Correlation analysis results

Table 4 shows the summary of the correlation coefficient matrix of all the radiological variables considered for the soil samples in Ondo, Southwest, Nigeria. ⁴⁰K is weakly correlated with all the measured variables but strongly correlated with ²³²Th (0.80). Similarly, ²³⁸U is strongly correlated with other radiological parameters except ²³²Th. There is a weak correlation between ⁴⁰K and ²³⁸U,

	obtained	a by (00		linar allo	u i talay		, 2010).	
Code	$\mathbf{H}_{_{\mathrm{in}}}$	H _{ex}	RLI	D	AED	ELCR (* 10 ⁻³)	AUI	I_{γ}
S1	0.86	0.47	1.19	80.5	0.39	3.27	1.53	0.59
S2	0.82	0.48	0.12	81.5	0.39	3.31	1.45	0.61
S3	0.87	0.48	0.12	81.4	0.39	3.31	1.55	0.60
S4	0.89	0.48	0.12	82.5	0.40	3.35	1.57	0.61
S5	0.26	0.19	0.05	33.0	0.16	1.34	0.53	0.26
S6	0.20	0.15	0.04	24.9	0.12	1.01	0.40	0.20
S7	1.06	0.58	0.14	98.2	0.48	3.99	1.87	0.72
S8	1.23	0.67	0.17	114.3	0.55	4.65	2.19	0.84
S9	0.97	0.53	0.13	90.4	0.44	3.68	1.72	0.67
S10	1.60	0.87	0.22	147.9	0.72	6.01	2.81	1.09
S11	1.01	0.55	0.14	93.7	0.45	3.81	1.79	0.69
S12	1.73	0.94	0.24	160.0	0.77	6.50	3.05	1.18
S13	1.06	0.58	0.14	98.1	0.47	3.99	1.88	0.72
S14	0.95	0.52	0.13	89.0	0.43	3.62	1.68	0.66
S15	1.28	0.69	0.17	118.3	0.57	4.81	2.27	0.87
S16	1.33	0.73	0.18	123.9	0.60	5.04	2.35	0.91
S17	0.77	0.42	0.11	72.1	0.35	2.93	1.36	0.53
S18	1.77	0.95	0.24	161.7	0.78	6.57	3.10	1.19
S19	0.80	0.43	0.11	74.1	0.36	3.01	1.41	0.55
S20	1.26	0.69	0.17	116.8	0.57	4.75	2.24	0.86
S21	1.06	0.58	0.15	98.7	0.48	4.01	1.87	0.73
S22	0.65	0.38	0.10	65.3	0.32	2.65	1.17	0.49
S23	1.00	0.54	0.14	92.5	0.45	3.76	1.77	0.68
S24	0.81	0.47	0.12	80.1	0.39	3.26	1.45	0.60
S25	0.99	0.54	0.14	92.4	0.45	3.76	1.77	0.68
S26	0.79	0.44	0.11	74.8	0.36	3.04	1.41	0.55
S27	0.96	0.52	0.13	89.3	0.43	3.63	1.70	0.66
S28	1.79	0.97	0.24	165.7	0.80	6.74	3.16	1.22
S29	0.95	0.57	0.15	96.8	0.47	3.93	1.74	0.73
S30	1.34	0.73	0.18	124.0	0.60	5.04	2.37	0.91
Mean	1.04	0.57	0.18	97.4	0.47	3.96	1.84	0.72
Min	0.20	0.15	0.04	24.9	0.12	1.01	0.40	0.20
Max	1.79	0.97	1.19	165.7	0.80	6.74	3.16	1.22

which is similar to the results obtained by (Senthilkumar and Narayanaswamy, 2016).

Table 3. Radiological indices results.

	⁴⁰ K	²³⁸ U	²³² Th	Ra _{ea}	H	H _{ex}	RLI	D	AED	ELCR	AUI	Ι _γ
⁴⁰ K	1			-4								
²³⁸ U	0.201	1										
²³² Th	0.790	0.384	1									
Ra _{eq}	0.359	0.969	0.538	1								
H _{in}	0.293	0.989	0.482	0.990	1							
H _{ex}	0.376	0.965	0.550	0.999	0.989	1						
RLI	0.305	0.858	0.462	0.878	0.873	0.873	1					
D	0.376	0.968	0.539	0.999	0.990	0.999	0.878	1				
AED	0.380	0.965	0.549	0.997	0.988	0.997	0.878	0.998	1			
ELCR	0.376	0.968	0.539	0.999	0.990	0.999	0.884	1.000	0.998	1		
AUI	0.310	0.982	0.512	0.996	0.997	0.994	0.881	0.994	0.993	0.994	1	
Ι _γ	0.416	0.950	0.573	0.996	0.980	0.996	0.872	0.997	0.996	0.997	0.987	1

Table 4. Spearman correlation analysis results.

A strong positive correlation co-efficient exists between most of the measured radiological parameters, implying a strong association among the pairs. A correlation coefficient value of 1 also exists between ELCR and D indicating a perfect monotonic relationship and a reflection of mutual dependence among them.

Multivariate factor analysis results

The results of the rotated factor loading of ⁴⁰K, ²³⁸U and ²³²Th and measured radiological indices are presented in table 5.

V7 · 11		Factors	
Variables -	1	2	3
К	0.114	0.963	-0.047
U	0.994	0.088	0.066
Th	0.351	0.896	-0.015
Raeq	0.975	0.213	0.058
Hin	0.986	0.153	0.062
Hex	0.975	0.213	0.058
RLI	0.105	-0.055	0.993
D	0.975	0.215	0.058
AED	0.975	0.215	0.058
ELCR	0.975	0.214	0.058
AUI	0.984	0.168	0.061
I_{γ}	0.972	0.230	0.057
Variance (%)	78.44	12.79	7.66

Table 5. Rotated factor loading of the radionuclides and measured radiological indices.

Three factors were extracted and explained about 99 % of the total variance of the data set. Factor 1 is loaded with concentration of ²³⁸U and the estimated radiological health hazards (except RLI) and explained 78.44 % of the total variance of the data. Factor 2 exhibits high loading of concentrations of ²³²Th and ⁴⁰K with 12.79 % as the percentage explained variance. The presences of ²³²Th and ⁴⁰K in factor 2 is similar to the result of the correlation analysis earlier obtained in this study, indicating their common association the soil of the study area. Factor 3 is singly loaded with RLI, which explains 7.66 % of the total variance.

Conclusions

The activity concentration of the naturally occurring ⁴⁰K, ²³⁸U and ²³²Th in the soil samples from Ondo, Southwest, Nigeria have been studied using the convectional gamma ray spectrometry. The results showed that the activity concentration ⁴⁰K, ²³⁸U and ²³²Th in the soil samples of the study area are within tolerable levels. The average activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th are 146.20, 184.83 and 19.70 Bqkg⁻¹, respectively. The averages values of the radiological indices consisting of Ra_{eq}, H_{in}, H_{ex}, RLI, D, AED, ELCR, AUI and I_Y were 224.47, 1.01, 0.61, 1.53, 203.60, 0.47, 1.65 x 10⁻³,

0.96 and 15.30, respectively. The values obtained in this study are within the recommended and tolerable safety limits, showing that these soil samples do not pose any significant radiological hazard. The results of this study are of great relevance in the environmental radiological protection and safety because soil is used as building material on a large scale in the studied area. It can be used as a reference data for future environmental radioactivity monitoring in the study area.

Acknowledgement

The authors are grateful to the technical staff of the Department of Physics, University of Medical Sciences, Ondo for the assistance rendered during the sampling.

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-forprofit sectors.

Conflict of interest

The authors declare that no conflict of interest exists.

References

ADEMOLA A. K., BELLO A. K., ADEJUMO A. C. (2014) Determination of natural radioactivity and hazard in soil samples in and around gold mining area in Itagumodi south western, Nigeria. J. Rad Res. Appl. Sci., 7:249–255. Doi: <u>10.1016ijrras.2014.06.1001</u>.

ADEMOLA, A. K., OLAOYE, M. A., ABODUNRIN, P. O., 2015. Radiological safety assessment and determination of heavy metals in soil samples from some waste dumpsites in Lagos and Ogun state, south-western, Nigeria. J. Rad. Res. Appl. Sci., 8:148–153. Doi: <u>10.1016/j.jrras.2014.12.010</u>.

AHMAD N., MOHAMAD SUHAIMI JAAFAR M. S., BAKHASH M., RAHIM M. (2015) An overview on measurements of natural radioactivity in Malaysia. J. Rad. Res. Appl. Sci., 8:136–141. Doi: <u>0.1016/j.jrras.2014.12.008</u>.

AKOZCAN S. (2014) Natural and artificial radioactivity levels and hazards of soils in the Kucuk menders basin, Turkey. Environ Earth Sci. 71: 4611-4614. Doi: <u>10.1007/</u><u>s12665-013-2861-6</u>.

ALAJEELI A., ELMAHROUG, Y., MOHAMMED, S., TRABELSI, A., (2019). Determination of natural radioactivity and radiological hazards in soil samples: Alhabba and Abuscabkh agricultural projects in Libya. Environ Earth Sci, 78:194-102. Doi: <u>10.1007/s12665-019-8213-4</u>.

ALAM, M.N., MIAH, N.M.H., CHOWDHURY, M.I., KAMAL, M., GHOSE, S., ISLAM, M.N., (1999). Radiation dose estimation from the radioactivity analysis of lime and cement used in Bangaladesh. J. Environ. Rad., 42(1):77–85. Doi: 10.1016/S0265-931X(98)00027-7.

BEREKA, J. MATHEW, P.J., (1985). Natural radioactivity of Australian building materials, wastes and by-products. Health Phys., 48:87–95. Doi: <u>10.1097/00004032-198501000-00007</u>.

CHANDRASEKARAN, A., RAVISANKAR, R., SENTHILKUMAR, G., THILLAIVELAVAN, K., DHINAKARAN, B., VIJAYAGOPAL, P., (2014). Spatial distribution and lifetime cancer risk due to gamma radioactivity in Yelagiri hills, Tamilnadu, India. Egyptian J. Basic Appl. Sci., 1(1):38–48. Doi: <u>10.1016/j.ejbas.2014.02.001</u>.

DARKO, E.O., TETTEH, G.K., AKAHO, E.H.K., (2005). Occupational radiation exposure to norms in a gold mine. Rad. Prot. Dos., 114(4):538–545. Doi: <u>10.1093/rpd/nci300</u>.

EC (1999). European Commission on radiological protection, principles concerning the natural radioactivity of building materials. directorate-general environment, nuclear safety and civil protection.

EL-GAMAL A., NASR S., EL-TAHER A., (2007) Study of the spatial distribution of natural radioactivity in upper Egypt Nile River sediment. Rad. Measure., 42:475–465. Doi: 10.1016/j.radmeas.2007.02.054.

FEROZ A.M., SAJAD A.R. (2015) Measurement of radioactive nuclides present in soil samples of district ganderbal of Kashmir province for radiation safety purposes. J. Rad. Res. Appl. Sci., 8:155–159. Doi: <u>10.1016/</u>j.jrras.2014.03.006.

GAO J., CAO C., LUO Z., ZHANG X. (2014) Inhalation exposure to particulate matter in rooms with under floor air distribution. Indoor Built Environ., 23:236–245. Doi: 10.1177/1420326X13482478.

GBENU S.T., OLADEJO O.F., ALAYANDE O., OLUKOTUN S.F., FASASI M.K. (2016) Assessment of radiological hazards of quarry products from southwest Nigeria. J. Rad. Res. Appl. Sci., 9, 20–25. Doi: <u>10.1016/j.jrras.2015.08.004</u>.

HASSAN N.N., KHOO K.S. (2014) Measurement of natural radioactivity and assessment of radiation hazard indices in soil samples at Pengerang, AIP Conference Proceedings 1584: 190:190–195. Doi: <u>10.1063/1.4866130</u>.

ICRP - International Commission on Radiological Protection (2007) Recommendations of the ICRP. Annals of the ICRP, 7, 2–4. Technical Report.

JAILLAD K.N. (2016) Radiation hazard indices and excess lifetime cancer risk in sand from the northern and eastern regions of Kuwait. Environ Earth Sci. 75:156. Doi: <u>10.1007/</u><u>s12665-015-5028-9</u>.

KALIPRASAD C.S., VINUTHA P.R., NARAYANA Y. (2017) Natural radionuclides and radon exhalation rate in the soils of cauvery river basin. Air, Soil, Water Res., 10:1–7. Doi: 10.1177/1178622117746948.

KARAHAN G., BAYULKEN A. (2000) Assessment of gamma dose rates around Istanbul, Turkey. J. Environm. Rad, 47:221–237. Doi: <u>10.1016/S0265-931X(99)00034-X</u>.

KOTA T.J. (2006) AIP Conference Proceed., 190:1584. IAEA, Database of prompt gamma rays from slow neutron capture for elemental analysis. Vienna. Technical report. Doi: 10.1063/1.4866130.

KOTHAI I.V.S., PRATHIBHA P., HOPKE P. K., PANDIT V.D.P. (2008) Source apportionment of coarse and fine particulate matter at Navi Mumbai, India. Aero Air Qual Res., 8:423–436. Doi: <u>10.4209/aaqr.2008.07.0027</u>.

KRIEGER R. (1981) Radioactivity of construction materials. Betonwerk Fertigteil-Tech, 47:468–473.

LAILI Z., IBRAHIM M.Z., MAHMUD A.N., OMAR M. (2012) Natural radioactivity content and radionuclides lechability of bricks containing industrial waste, seminar Nuclear Malaysia 2012 (rnd12), dewan tun dr. ismail, Agensi Nuklear Malaysia, 26-28 September 2012.

MARCAZZAN G.M., CERIANI M., VALLI G., VECCHI R. (2003) Source Apportionment of PM_{10} and $PM_{2:5}$ in Millan (Italy) using receptor modeling. Sci Total Environ, 317:137-147. Doi: <u>10.1016/S0048-9697(03)00368-1</u>.

MAVI B., AKKURT I. (2010) Natural radioactivity and radiation hazards in some building materials used in Isparta, Turkey. Rad. Phys. Chem., 79:933–937. Doi: <u>10.1016/j.</u> radphyschem.2010.03.019.

MOHAMMED R.S., HAMMED R.S. (2017) Estimation of excess lifetime cancer risk and radiation hazard indices in southern Iraq. Environ Earth Sci 76:303. Doi: <u>10.1007/s12665-017-6616-7</u>.

NWAKA B.U., EMELUE H.U., NWOKOCHA C. (2001) Natural radiation levels and health hazard indices of soil in Owerri Nigeria. The Intern. J. Eng. Sci., 3(12):5–9. ISSN (e):2319–1813.

OGUNDELE L.T., OLASINDE R.T., OWOADE O.K., OLISE F.S. (2018) Composition and Source Identification of Chemical Species in Dust from Selected Indoor Environments in Ile-Ife, Nigeria. Earth Syst. Environ., 2:323–330. Doi: 10.1007/s41748-018-0052-z.

PEKEY H., BAKOGLU M., PEKEY B. (2005) Sources of heavy metals in the Western Bay of Izmit surface sediments, Int. J. Environ. Anal. Chem., 85(14):1025–1036.

QURESHI A.A., TARIQ S., DIN K.U.U., CALLIGARIS C., WAHEED A. (2014) Evaluation of excessive lifetime cancer risk due to natural radioactivity in the rivers sediments of Northern Pakistan. J. Radiat. Res. Appl. Sci., 7:438–447. Doi: 10.1016/j.jrras.2014.07.008.

RAGHU Y., RAVISSANKAR R., CHANDRASEKRAN A.P., VENKATRAMAN V. (2017) Assessment of natural radioactivity and radiological hazards in building materials used in the Tiruvannmalali District, Tamilnadu, India, using a statistical approach. J. Taibah Uni. Sci., 11:523–533. Doi: 10.1016/j.jtusci.2015.08.004.

RAVISANKAR R., VANASUNDARI K., CHANDRASEKARANA., RAJALAKSHMIA., SUGANYA M., VIJAYYAGOPAL P., MEENAKSHISUNDARAM V. (2012) Measurement of natural radioactivity in building materials of namakkal, Tamilnadu, India using gamma ray spectrometry. Appl. Rad. Isot., 70:699–704. Doi: <u>10.1016/j.</u> apradiso.2011.12.001.

RAVISANKAR R., VANASUNDARI K., SUGANYA M., RAGHU R., RAJALAKSHMI, A., CHANDRASEKARAN A., SIVAKUNAR S., CHANDRAMO-HA S., VIJAYAGOPALP, VENKATRAMAN B. (2014) Multivariate statistical analysis of radiological data of building materials used in Tiruvan-namalai, Tamilnadu, India. Appl. Rad. Isot., 85:114–127. Doi: 10.1016/j.apradiso.2013.12.005.

SENTHILKUMAR G., RAGHU Y., SIVAKUMAR S., CHANDRASEKARAN A., ANAND D. P., RAVISANKAR R. (2014) Natural radioactivity measurement and evaluation of radiological hazards in some commercial flooring materials used in Thiruvannamalai, Tamilnadu, India. J. Rad. Res. Appl. Sci., 7:116–122. Doi: <u>10.1016/j.jrras.2013.12.009</u>.

SENTHILKUMAR R.D., NARAYANASWAMY R. (2016) Assessment of radiological hazards in the industrial effluent disposed soil with statistical analyses. J. Rad. Res. Appl. Sci., 9:449–456. Doi: <u>10.1016/j.jrras.2016.07.002</u>.

SIVAKUMAR R. (2014) An assessment of natural radioactivity levels and radiation hazards in the soil of Conoor, South India. Environ earth Sci, 72:5063–5071. Doi: 10.1016/j.jtusci.2014.03.004.

SOLEHAH A.R., YASIR M.S., SAMAT S.B. (2016) Activity concentration, transfer factors and resultant radiological risk of ²²⁶Ra, ²³²Th, and ⁴⁰K in soil and some vegetables consumed in Selangor, Malaysia. AIP Conference Proceedings, Doi: 10.1063/1.4966802.

SROOR S.M.E., AHMAD F., ABDUL-HALIM A.S. (2001) Natural radioactivity and radon exhalation rate of soil in southern Egypt. Appl. Rad. Isot., 55:873–879. Doi: 10.1016/S0969-8043(01)00123-3.

TUFAIL M., AKHHTAR N., JAVIED S., HAMID T. (2007) Natural Radioactivity hazard of building bricks fabricated from saline soil of two district of Pakistan. J. Rad. Prot., 27:481-492. Doi: 10.1088/0952-4746/27/4/009.

UNSCEAR (2000) United Nations Scientific Committee on the Effects of Atomic Radiation. Sources Effects and Risks of Ionizing Radiation. <u>https://inis.iaea.org/search/search.</u> <u>aspxorig.</u> RN:20037090. Accessed on September 2018.

UNSCEAR (2001) Sources and effects of ionizing Radiation. United Nations Scientific committee on Effects of Atomic Radiation (UNSCER), 2001. Report to the General Assembly. Accessed on July, 2018. UNSCEAR (2010) Sources and effects of ionizing radiation. united nations scientific committee on effects of atomic radiation (UNSCEAR), 2010. report to the general assembly with scientific annexes, vol. 1 New York, United Nation.

USEPA (2017) United States Environmental protection Agency, Radiation Health Effects. Technical report.

USIKALU M.R., AKINYEMI M.L., ACHUKA J. (2014) Investigation of radiation levels in soil samples collected from selected locations in Ogun State, Nigeria. IERI Procedia, 9:156–161. XINWEI L., LINGQUIG W., XIAODAN J., LEIPENG Y., GELIAN D. (2006) Specific activity and hazards of archeozoiccambrian rock samples collected from the Weibei area of Xhaanxi, China. Rad. Prot. Dos., 118(3):352–359. Doi: 10.1093/rpd/nci339.

ZIQIANG P., YIN Y., MINGQIANG G. (1988) Natural radiation and radioactivity in China. Rad. Prot. Dos., 24:88–99. Doi: <u>10.1093/oxfordjournals.rpd.a080236</u>.