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ORIGINAL PAPER

Distribution dynamics and descriptive statistical analysis of radionuclides in the farmland soils near mining areas in Southwestern Nigeria

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Abstract Human exposure to ionizing radiation in the environment is mainly due to naturally occurring radionuclides in the soils, building materials and rocks, but the level may vary depending on the anthropogenic activities prevalent in each location. Presently, in Nigeria, there are concerns due to environmental health implications of all sorts of mineral mining and processing spreading across the southwestern states of the country. This work determines the activity concentrations of naturally occurring radionuclide materials (NORMs) in the farmland soil with the aim of evaluating the radiation hazards. A total of 200 composite soil samples were taken from five states in the southwest of Nigeria, close to active mining sites at the root (0.2 m) and at deep planting zones (0.5 m) for analysis by gamma-ray spectrometry using NaI(Tl) detector. The activity concentrations of natural radionuclides in the composite soil samples were determined to vary in the order of ^{40} K > ^{232}Th > ^{226}Ra/^{238}U for all locations. In contrast to the other locations, Olode and Igbokoda had average radium equivalent activities (Ra_{eq}) to be 1.6 and 1.8 times, respectively, higher than the reference limit of 370 Bqkg^{-1}. The estimated excess life cancer risk values were lower than the 0.29×10^{-3} global average value for soil by United Nations on Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and International Commission on Radiological Protection (ICRP). Negative and low skewness values of 0.61 and 1.20 were obtained for ^{40} K in Itagunmodi, and also 0.47 and 0.66 for ^{232}Th were obtained in Sagamu. The kurtosis analysis of the activity concentrations was low and negative for soil at Itagunmodi for ^{40} K and ^{226}Ra/^{238}U; Olode for ^{40} K and ^{232}Th; and Igbokoda for ^{226}Ra/^{238}U and ^{232}Th where mining activities are commonly practiced. The variation in the results has been attributed to different agriculture practices and artisanal mining operations in each location.

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Introduction

Environmental matrices such as crude oil, solid minerals and rocks can be huge sources of naturally occurring radioactive materials (NORMs). Enhancement in the levels of these materials is unavoidable where technological and human activities are involved during mineral extraction or mining processes, making NORMs to be referred to as Technologically Enhanced Natural Occurring Radioactive Materials (TENORMs) (Ali et al., 2019). Under natural conditions, the presence of radioactive elements in the soil does not disturb the ecosystem's dynamic equilibrium or the food chain (Niamatullah, 2021). However, high concentrations of radioactive elements might change the biosphere's natural ecosystem, offering serious risks to those who are exposed. The two main methods of exposure to radionuclides are external exposure from radionuclides in soils and internal exposure from eating plants grown in farmland soil and/or inadvertent ingestion during cultivation of the farmland soil (UNSCEAR, 2008; Kakati et al., 2013; Kpeglo et al., 2011; Özdiş et al., 2017; Olatunji et al., 2022). The safety of lives against potential radiological hazards ensuing from exposure to the radionuclides requires proper understanding of their concentrations, migration within the soil and influential knowledge of factors responsible for the entire dynamics within the soil (Adebiyi et al. 2021; Geras'kin, 2016).

Uranium, thorium, and their progenies are the most common natural radionuclides and also the main sources of radiation exposure for humans. The disintegration of the atomic nuclides occurs with the emission of alpha or beta particles followed by gamma radiation until a final and most stable nuclide such as ^{206}Pb, ^{207}Pb or ^{208}Pb is attained. The gamma radiation that originates from ^{226}Ra, ^{232}Th and their daughter radionuclides and ^{40} K is responsible for external radiation exposure while the short-lived progenies of radon (^{222}Rn) after the decay of ^{226}Ra are the main cause of internal radiation exposure.

The earth's crust is stratified into several layers of which soil is the uppermost layer. Among the layers of the earth's crust, soil plays major roles in the ecological balance, hydrological buffer, mining operations and sustainable development for human survival (Ademola et al., 2014). Apart from mining activities being carried out on the soil, agricultural processes especially cultivation of crops for the production of foods for human consumption are practiced on the soil. Farming is one of the most crucial means of human survival because humans depend totally on plants for feeding (Olufunmbi et al., 2016). However, anthropogenic activities such as cultivating near mining environments may serve as a major sink of environmental contaminants.

Both external and internal radiation exposure routes have detrimental implications on human health. The inhalation of radon gas and its short-lived daughters from soil material in the farmland may expose the population to radiation risks. Also, once radon and its progenies enter the human body, they are deposited in the respiratory tract and can cause damage to the sensitive tissues of the lungs. Radon gas could also penetrate the bloodstream from the lungs and irradiate the whole human body through an internal gaseous exchange, leading to a permanent source of internal radiation exposure (Ogundele et al., 2021; Ononugbo et al., 2016; Özdiş et al., 2017). The translocation and uptake of natural radionuclides in crops obtained from farmlands are dependent on the farmland-soil composition; arising from agricultural practices (such as deposition of mineral resources like fertilizers in the subsurface layers of soil), atmospheric deposition, plant soil characteristics, and other environmental contamination (Gbenu et al., 2016; Olatunji et al., 2022). Accidental or incidental exposure to ionizing radiations over a short period (acute exposure) or a long period (chronic exposure) can result in skin burns, cell damage, cancer, cardiovascular disease, and even death. Low levels of environmental exposure do not immediately have a negative impact on health, but like most toxic environmental pollutants, prolonged exposure increases the risk of cancer (Ndontchueng et al., 2014; Njinga & Tshivhase, 2016). The biological effects of ionizing radiation exposure have been precisely described (Njinga & Tshivhase, 2016; Qureshi et al., 2014; Taskin et al., 2009).

The mining of solid minerals had increased tremendously over the last few decades in Nigeria with a consequential impact on land degradation most especially

on agricultural farmlands. The United State Environmental Protection Agency (USEPA, 2007) reported that in nature, mining generates a lot of waste that can contaminate soils over a large area, which has a detrimental effect on the environment and human health. Several minerals are mined and processed in close proximity to farmlands and these affect soil geochemistry. Some of the potential adverse impacts of mining operations include ecological disturbance and radiation hazards to the farmers that cultivate the farmland and the entire population that consumes the farm products.

In Nigeria, studies have been carried out on the environmental radioactivity in the soil around the farmland areas where active artisanal mining is going on (Ogundele et al., 2021). The existence of various radionuclides with varying activity concentrations and radiological threats associated with exposure to the measured activity concentrations of radionuclides have also been reported. The consequence of radiation exposure is always cumulative from frequent and continuous exposure, leading to radiation build-up which could be devastating for human health (Olatunji et al., 2022). In this study, the radionuclides' concentrations of soil samples collected from farmlands around active mining activities in part of Nigeria were determined. The mining operations are carried out haphazardly by inexperienced and unlicensed artisanal miners without land reclamation. In most cases, such disturbed soil enhanced the spread of the natural radionuclides in the immediate environment of the mining. This study is therefore aimed at determining the activity concentrations of NORMs in the farmland soils near and far away from mining sites and estimating the probable radiological hazards and risks due to the exposure from radiation in the farmland soil. This will also involve the dynamics computation of NORMs redistribution in farmland soils due to vertical migration from surface to deep soil layers. The quantum GIS software is used to map the radioactive levels of the radionuclides in the root zone to the deep soil layers within the five states in the Southwest region of Nigeria.

Methodology

Geological setting and study location

The study areas lie between latitudes 4° and 9°N and longitudes 30° and 7°E right in the equatorial rain forest region of Africa and fall within the Precambrian Basement Complex of Southwestern Nigeria. As described by other authors (De Swardt, 1953; Hockey et al., 1986; Elueze, 1988), the geology of the Basement Complex of Nigeria consists of six major rock groups. These are migmatite-gneiss complexes, metasedimentary and metaigneous rocks that are slightly migmatised to non-migmatised, charnockitic, older granite, metamorphosed to non-metamorphosed calc-alkaline volcanics and hypabyssal rocks, and non-metamorphosed dolerite dykes, basic dykes, and syenite dykes. The Basement Complex of Southwestern Nigeria hosts important deposits of gold and chromite in the Ife-Ilesha Schists Belt. Local geological mapping showed that the rock groups in the Olode area in Oyo state are porphyritic granite, porphyroblastic gneiss and undifferentiated gneiss complex. Prominent locations in the study area are known to harbour precious rocks and gemstones, in which the important host rocks are pegmatites. Most of the mineralized pegmatites in this area occur as minor intrusions within the gneissic rocks as evidenced by the elevated level of mining activities in the study area. The geological settings of Ogun State with other southwestern states including Oyo and Lagos States are that of a sedimentary terrain located within the eastern Dahomey basin. This basin extends into western Nigeria as far as the Okitipupa Ridge or Ilesha Spur and as far west as the Volta Delta complex in Ghana. The rocks within the Dahomey are extensive wedges of Cretaceous, Paleocene, and Neocene sediments which thicken markedly from the onshore margin of the basin. The Cretaceous sediments rest unconformably on the basement complex and west of the Okitipupa high, consisting mainly of coarse-grained classic sediments known as the Abeokuta Formation in western Nigeria and the "Maestrichtian Sableux" in Benin (Dahomey). From oldest to youngest, the Abeokuta, Ewekoro, Akinbo, Oshosun, Ilaro, and Benin Formations were identified within the stratigraphy of the eastern Dahomey basin. Some workers have described the Cretaceous Abeokuta Formation as a group consisting of Ise, Afowo, and Araromi Formations. Further description of the geological mapping of southwestern Nigeria has been detailed elsewhere (Maxwell et al. 2014; Rahaman, 1988).

Meanwhile, only four of the five states involved in the study have notable records of mining activities near farmlands. Each of these states has its

own solid minerals commonly mined, except Lagos State which does not have any known record of solid mineral mining but known for regular application of fertilizers in growing crops. Even though Lagos State is the economic hub of Nigeria due to the presence of heavy industrial activities (excluding mining or extraction of minerals), there are no known industrial processes involving manufacturing or processing of raw materials in Epe area of Lagos State where samples were collected for this study. In short, the location of sample collection in Epe is known only for farming and boast of vast farmlands where Lagos State Government trains the youths in the state for agro-entrepreneurial empowerment (AGRIC-YES). However, it is important to mention also that a private refinery project (Dangote Refinery) is nearing completion in the neighbouring Lekki Ibeju which is a town next to Epe. Hence, it is envisaged that the data from this study will not only serve as control for other states but also as reference radiation data for monitoring the effect of the activities of this refinery when in operation.

Table 1 shows the geographical location in each state where the samples were collected and their prevalent minerals/mining activities. The climates of the southern states of Nigeria are characterized by tropical rainforests, although there is minute variation in the meteorological variables such as annual rainfall, temperature and relative humidity due to their distances from the Atlantic Ocean. It is important to note that the mining activities carried out by the people in the study locations are illegal and mostly carried out by untrained artisanal miners without license. Figure 1 shows the map of Nigeria and the five southwestern states where samples were collected and Fig. 2 is a typical of artisanal gold mining site which is a scenario for other mining sites in the study locations.

Sample collection and preparation

Two farmlands were identified close to the mining sites in each state, making a total of ten (10) farmlands involved in this study. At each farmland location, a plot of 1,000 m^2 was marked out and subdivided into 10 m by 10 m (100 m^2) making a total of ten (10) subdivisions. Thirteen (13) points were located for sample collection in each subdivision at the root zones layer of 0.2 m and deep soil layer of 0.5 m in the farmland (at approximately 25 m apart) to active mining. The reason for the sample collection at two depths is so as to investigate if there is a vertical relationship between top soil and deep soil layers. The thirteen soil samples in each area were collected as follows: four samples were collected at the extreme corners of the demarcated areas; eight samples were collected in the midway to the centre of the square and one sample at the centre. The collected thirteen samples each at top soil and deep soil layers from each sub-demarcated area were then mixed thoroughly to form composite soil sample, making a total of two (2) composite soil samples. Hence, within the 1,000 m^2 area of demarcation, twenty (20) composite soil samples (10 at top soil layer and 10 at deep soil layer) were collected for each farmland. The collection procedures were uniform in all five states. A total of 40 samples were collected from two farmlands from each area and 200 samples altogether for the five States. It is important to mention that the collected samples are all from existing farmlands at an area extent of 100 m^2, leading to partial distortion and impact on the farmlands. However, the farmers were adequately compensated, and the food crops and vegetables grown on the farmlands were bought from the farmers for further research.

The geographical coordinate of each location was determined using hand-held Global Positioning System (GARMIN-GPS map 78 s). The samples

Table 1 Description of the study locations

S/N	State	Location	Activities
1	Oyo	Olode	Mining of gemstone and precious minerals
2	Ondo	Igbokoda	Mining of Bitumen
3	Osun	Itagunmodi	Gold mining
4	Ogun	Sagamu	Mining of limestone
5	Lagos	Epe	Intensive agricultural activities with fertilizer application

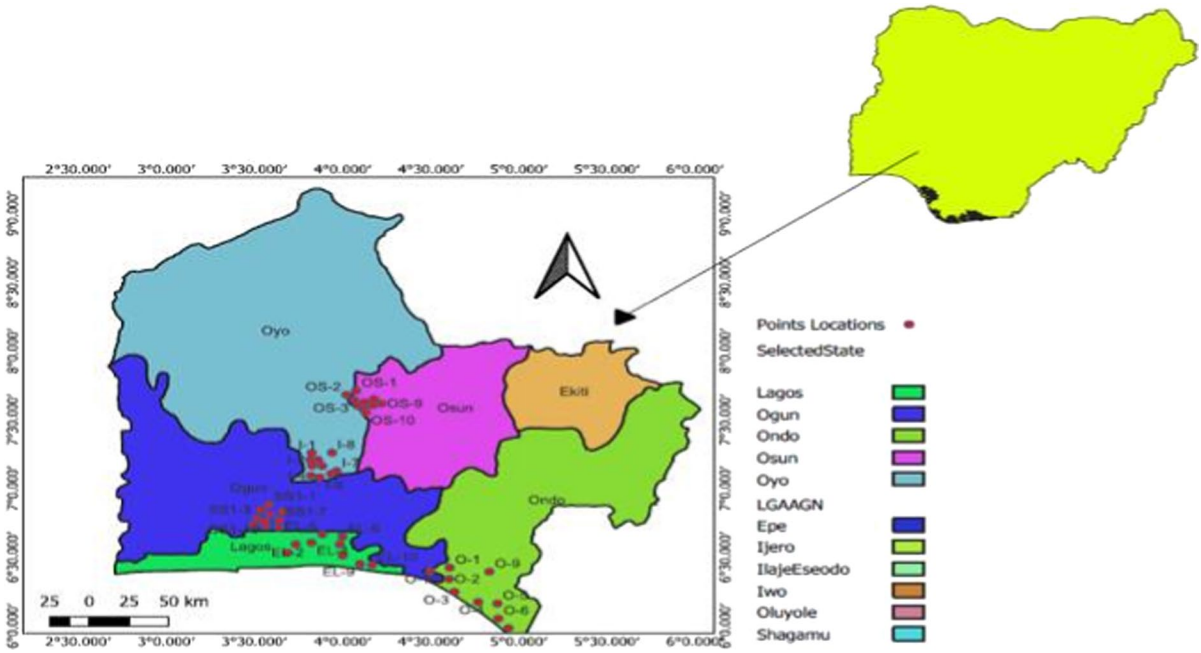


Fig. 1 The map of the study location

Fig. 2 A typical artisanal gold mining site (https://punchng.com/tackle-illegal-miners-devastating-osun-ekiti/)

collected were kept in properly labeled polyethylene bags to avoid cross-contamination and easy identification. These soil samples were transported to the laboratory for further preparation prior to gamma spectrometric analysis. The samples were air dried for two weeks in the laboratory and the gravel lumps, pebbles, and coarse stones, were handpicked from each sample. The samples were then oven dried at a temperature of 110 to remove the moisture and attain constant mass. Afterward, the samples were crushed and grounded with an agate mortar, then sieved with 2 mm mesh size sieve. Two hundred (200 g) of each fine sample was weighed using an electric weighing balance (METLR MT-501) and sealed hermetically in a 250-ml plastic bottle and kept for 30 days to allow for radon and its short-lived progenies to reach secular radioactive equilibrium prior to gamma-ray spectroscopy.

Gamma-ray spectrometric analysis

Measurement of the activity concentrations of the gamma-emitting radionuclides in the samples was conducted by gamma-ray spectrometry using NaI(Tl) detector. The gamma-ray detector is an energy conversion medium for the incident photons to have a high interaction probability to produce fast electrons. The amplitudes of the pulse (gamma ray photons) are analysed after amplification and digitization. The output of the gamma spectrometer represents the energy spectrum of the detected NORMs which provides complete information useful for the identification of unknown NORMs available in the sample (Avelar et al., 2013; Worsfold et al., 2019). In this study, the thallium-doped sodium iodide (NaI(Tl)) detector

was used along with some standard sources ordered from Isotope Products Laboratories, Valencia, California, USA consisting of ^{241}Am, ^{57}Co, ^{137}Cs and ^{60}Co. The detector was connected to a 16 k MCA for data acquisition and the gamma-rays emitted from the sample were analysed by the Gamma Vision 5.0 software (EG&G ORTEC). The detector is shielded with 15-cm-thick lead on all sides, including the top, to reduce the background contribution from the surroundings. The inner sides of the lead shielding are lined with 2-mm-thick cadmium and 1-mm-thick copper to attenuate lead X-rays and cadmium x-rays, respectively. The measured detection efficiencies were fitted by a polynomial fitting function and the fitted efficiency was used to calculate the activity concentration of the regular samples. These samples were counted for a sufficiently long time (10 h) to reduce counting errors and the background counts for the same counting time were deducted to obtain the net counts. The gamma lines of 351.93 keV (35.6%) from ^{214}Pb, 609.32 keV (45.49%) from ^{214}Bi, and 1120.294 keV (14.92%) from ^{214}Bi were used to determine the activity concentration of ^{226}Ra; the gamma lines of 238.632 keV (43.6%) from ^{212}Pb, 583.19 keV (85.0%) from ^{208}Tl to 911.204 keV (25.8%) from ^{228}Ac were used to determine the activity concentration of ^{232}Th; while the single gamma line 1460.822 keV (10.66%) was used to determine the activity concentration of ^{40} K. The spectrometric measurement was carried out at the Natural Radioactivity Laboratory, Department of Earth Sciences, University of Coimbra, Portugal. The statistical modeling was conducted using GNU Octave software and the coding for linear regression is presented as a supplementary material I. Equation (1) is the usual model used to obtain the activity concentration of ^{226}Ra, ^{232}Th and ^{40} K radionuclides in each sample (Olatunji et al., 2014):

$$A = \frac{N \times 1000}{\varepsilon_\gamma I_\gamma m t} \left(Bqkg^{-1}\right) \tag{1}$$

where, A is the activity concentration of the radionuclide in the sample, N is the net counts or counting area under a photopeak, ε_γ is the efficiency of the detector for particular γ–ray energy, m is the mass of each counted sample, I_γ is the intensity of the emitted gamma–ray, and t is the counting time.

Associated radiological health risk parameters

To determine the radiation risks, radium equivalent (Ra_{eq}), external health (H_{ex}), and internal health (H_{in}) risks were employed while absorbed dose (D), annual effective dose equivalent (AEDE) and excess lifetime cancer risks were utilized to estimate the radiation dose due to the exposure to radionuclides in the farmland soil samples near the mining environments. The activity concentrations of naturally occurring radionuclides are not uniformly distributed. Therefore, the Ra_{eq} is introduced as a weighted sum of activity concentrations of ^{226}Ra, ^{232}Th and ^{40} K being a factor that described the gamma output from different mixtures of activity concentrations. Equation (2) was used to estimate radium equivalent activity as follows (Njinga et al., 2016):

$$R_{a_{eq}} = A_{Ra} + 1.43\,Th_{Ra} + 0.077\,A_K \tag{2}$$

where $A_{Ra}, A_{Th}, and A_K$ are specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40} K, respectively.

The external hazard index (H_{ex}) is a widely useful index for safety standard regulation and radiation protection from the emission of gamma radiation by various radionuclides. The H_{ex} was evaluated using Eq. (3) (Stevanović et al., 2018):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{3}$$

Inhalation of gaseous radionuclide and its short-lived progeny are hazardous to respiratory organs and other internal living cells from radiation exposure route (Ademola et al., 2014; Gbenu et al., 2016). The hazardous effect of short-lived products was quantified by the H_{in} parameter as shown in Eq. (4) (Stevanović et al., (2018)):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{4}$$

For a homogeneous distribution of naturally occurring radionuclides in the soil samples, the outdoor absorbed dose rate in air from the terrestrial sources of gamma radiation in the soil samples at 1 m above the ground surface is determined using Eq. (5) (Olatunji et al., 2022):

$$D(nGy/h) = 0.042A_K + 0.462A_{Ra} + 0.604A_{Th} \tag{5}$$

where $A_K, A_{Ra} and A_{Th}$ are the specific activity in Bq kg^{-1} for ^{40} K, ^{226}Ra(^{238}U), and ^{232}Th, respectively. The concentration-to-dose conversion factors of 0.042, 0.462 and 0.604 $Gyh^{-1}/Bqkg^{-1}$ are used for ^{40} K, ^{226}Ra and ^{232}Th, respectively.

The United Nations Scientific Committee on Effect of Atomic Radiation (UNSCEAR, 2000) report specified 0.2 as the outdoor occupancy factor (OF) and 0.70 Sv Gy^{-1} as the dose convention factor (DCF) for external irradiation (which converts the external outdoor absorbed dose rate to outdoor effective dose rate) (Olatunji et al., 2022). In this regard, the outdoor annual effective dose equivalent (AEDE) was calculated using Eq. (6) (UNSCEAR, 2000):

$$AEDE(\mu Sv/y) = D(nGy/h) \times DCF \times OF \times T \quad (6)$$

where T = annual exposure time (8760 hy^{-1}).

The probability of developing cancer in a lifetime as a result of external gamma dose exposure from naturally occurring radiation source defined as excess lifetime cancer risk (ELCR) was determined from the annual effective dose using Eq. (7) (Isinkaye, 2018; Taskin et al., 2009):

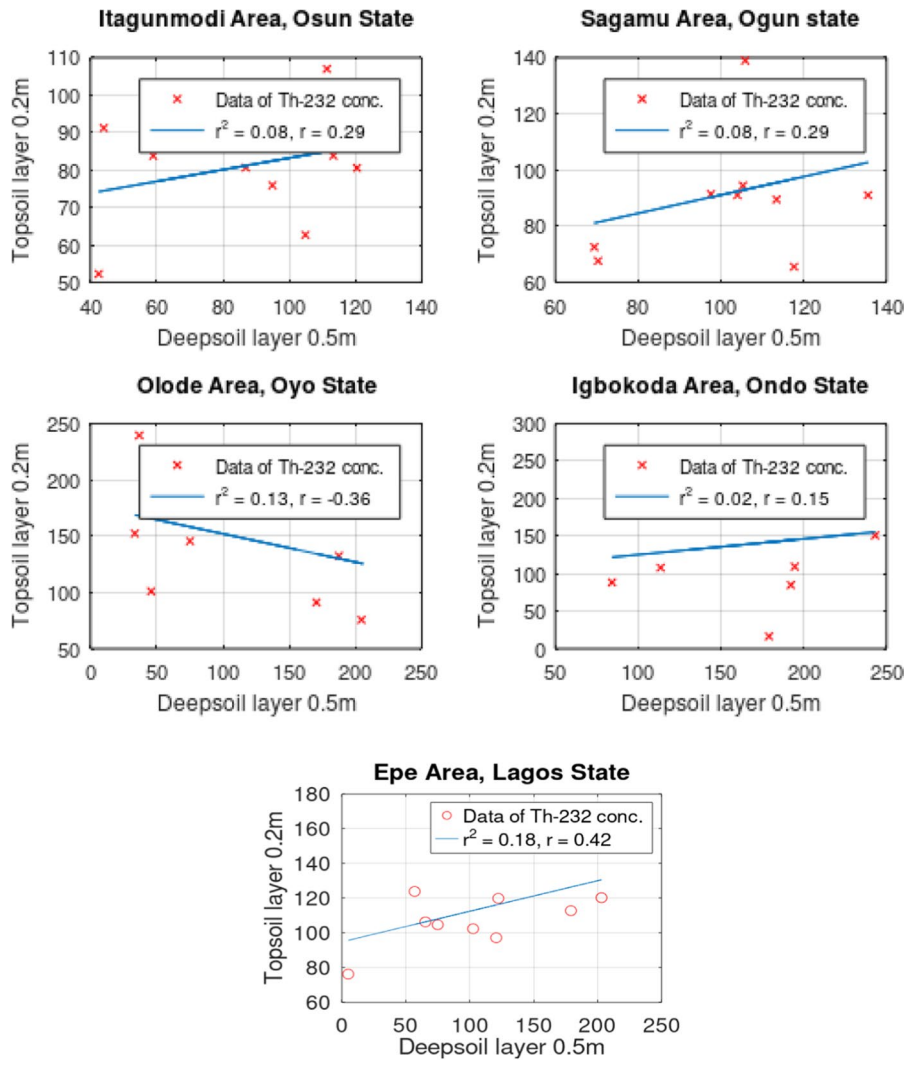
$$ELCR = AEDE \times DL \times RF \times 10^{-3} \quad (7)$$

where *DL* is the average duration of life and was estimated to be 45.5 years in Nigeria, *RF* is the detriment-adjusted nominal risk coefficient (Sv^{-1}), and ICRP uses *RF* as 5.5×10^{-2} for the public (ICRP, 2007; Olatunji et al., 2022).

Result and discussion

Statistical analysis of activity concentration in the farmland soil samples

The results of the correlation and regression analysis of the top and deep layers soil for ^{232}Th, ^{226}Ra, and ^{40} K (Farm 1 & 2) are presented in Figs. 3 a, b, and c, respectively. The measurement of the linear relationship in the variability of each of the measured radionuclides in the top and deep layer zone of the soil profile was indicated by the coefficient of determination (r^2) and correlation coefficient (r). For ^{232}Th, considering Farm 1 & 2, the correlation coefficient (r) for Itagunmodi LGA (Osun State), Sagamu LGA (Ogun State), Olode LGA (Oyo State), Igbokoda LGA (Ondo State), and Epe LGA (Lagos State) were 0.29, 0.29, − 0.36, 0.15 and 0.42 for Farm 1 and Farm 2, were 0.30, 0.43, 0.47, − 0.59 and 0.48, respectively. The dataset "top soil layers, radionuclides distributions, and deep soil layers" contains the radionuclides pollutant levels and the number of sampling points (10 at top soil layer and 10 at deep soil layer). These variables reflect the level of pollutant distributions in the two farmlands per LGA in each of the State under study. It is reasonable to assume that there is some correlation between the top and deep soil layers. From the dataset in Farm 1, ^{232}Th have a positive moderate correlation of 0.42 for Epe LGA (Lagos State) and the r^2 value of 0.18 (the square of the correlation coefficient), indicating that 18% of the variation in the top soil layer is influenced by the deep soil layers. The remaining LGA showed weak correlation in the distribution of ^{232}Th. In Farm 2 (^{232}Th), negative moderate correlation of r = − 0.59 and r^2 value of 0.36 was observed. This shows that 36% of the variation in the top soil layer is reversely related to the deep soil layer. However, a positive moderate correlation (*r*) of 0.48 ($r^2 = 0.23$) for Epe LGA, 0.43 ($r^2 = 0.18$) for Itagunmodi LGA and 0.47 ($r^2 = 0.23$) for Igbokoda LGA were observed. This revealed that 18% for Itagunmodi LGA, 23% for Epe LGA & Igbokoda LGA distribution of ^{232}Th, the variation in the top soil layers is directly related to the deep soil layers in each of the LGA, respectively. The *r*-values of ^{226}Ra for Farm 1 (F1) and Farm 2 (F2) at Itagunmodi, Sagamu, Olode, Igbokoda and Epe were − 0.63 F1 & − 0.39 F2, 0.22 F1 & 0.24 F2, 0.94 F1 & 0.76 F2, 0.42 F1 & 0.95 F2 and 0.63 F1 & 0.48 F2, respectively. The r^2-values were 0.88 F1 and 0.57 F2 in Olode LGA, 0.89 F2 in Igbokoda LGA indicating a perfect positive correlation coefficient of 88%, 57% and 89% of ^{226}Ra distribution variation of the deep soil layers being influenced by the top soil layers. In the case of the remaining LGAs, weak r^2 values were observed. The r–values of ^{40} K distributions were − 0.63 F1 & − 0.39 F2, 0.22 F1 & 0.24 F2, 0.10 F1 & − 0.34 F2, 0.73 F1 & 0.20 F2 and 0.15 F1 & 0.48 F2, respectively, and the r^2 values of 0.23 F2 (Epe LGA), 0.39 F1 (Itagunmodi LGA) showing moderate relations of the correlation coefficient variation of the top and deep soil layer for ^{40} K. All the remaining LGAs revealed weak



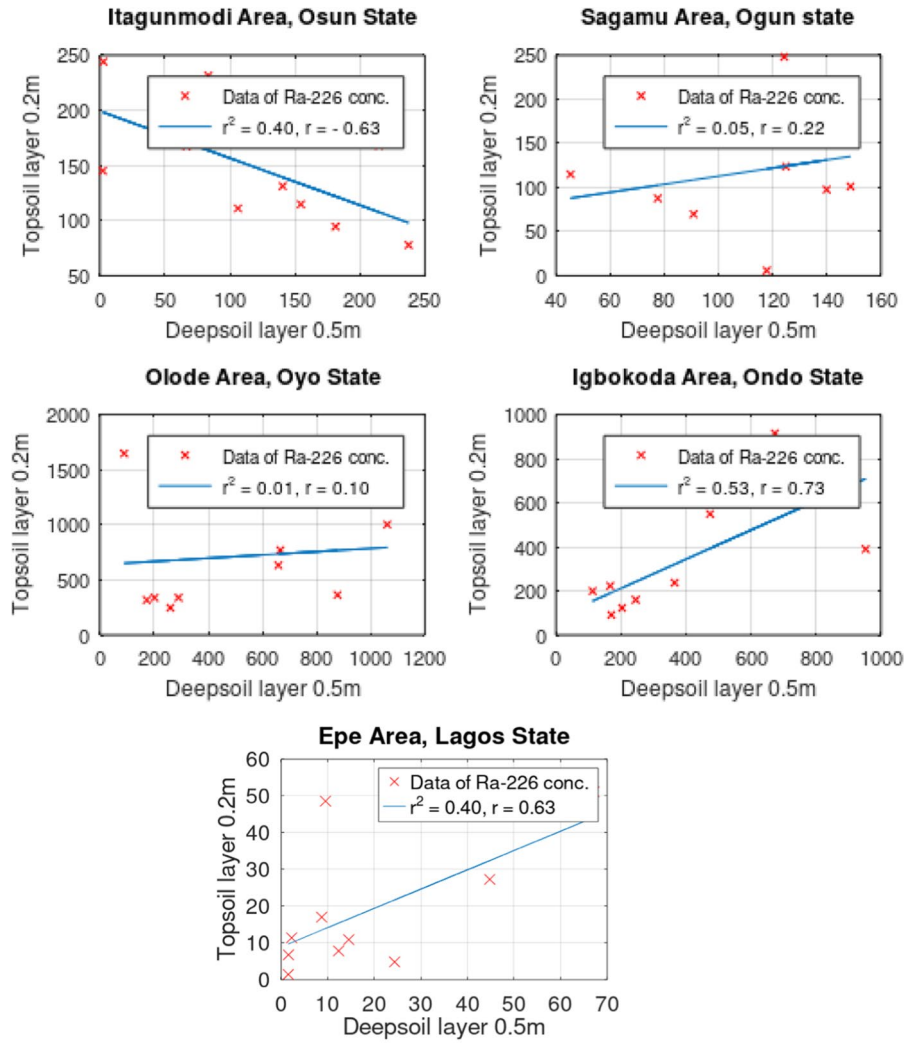
(a)

Fig. 3 (**a**) Results of the Bivariate Statistical Analysis of ^{232}Th in the topsoil and deep layer. (**b**) Results of the Bivariate Statistical Analysis of ^{226}Ra in the topsoil and deep layer. (**c**) Results of the Bivariate Statistical Analysis of ^{40} K in the topsoil and deep layer

relationships between the top and the deep soil layers. In general, the long-term behaviour of radionuclides in the environments (farmlands) is most susceptible to their vertical distribution and migration in the soils. Each *r*-value explained the linear relation in the activity concentrations. It indicates the gradual increase and, in some cases, decrease in the activity concentration of the measured radionuclides in the farmland soil samples. The observed differences in the trend might be related to the differences in physical–chemical properties of the soil (such as mineral composition, air, water, organic matter content and decaying remains of once-living creatures) which have many contributions to the vertical distributions of radionuclides (Nenadović et al., 2012; Olatunji et al., 2022).

The descriptive statistics of the activity concentrations of ^{40} K, ^{232}Th and ^{226}Ra (^{238}U) in the farmland soil samples at the root zone (layer) and the deep layer zone are presented in Table 2 and Table 3, respectively. A basic feature of statistical analysis is to characterize the variability of the data set in a simplified form for easy interpretation and deductions. The average values, standard deviation (±SD) minimum, maximum, skewness, and kurtosis of the measured activities described the central tendency and the variation around the mean as well as the distribution

Fig. 3 (continued)

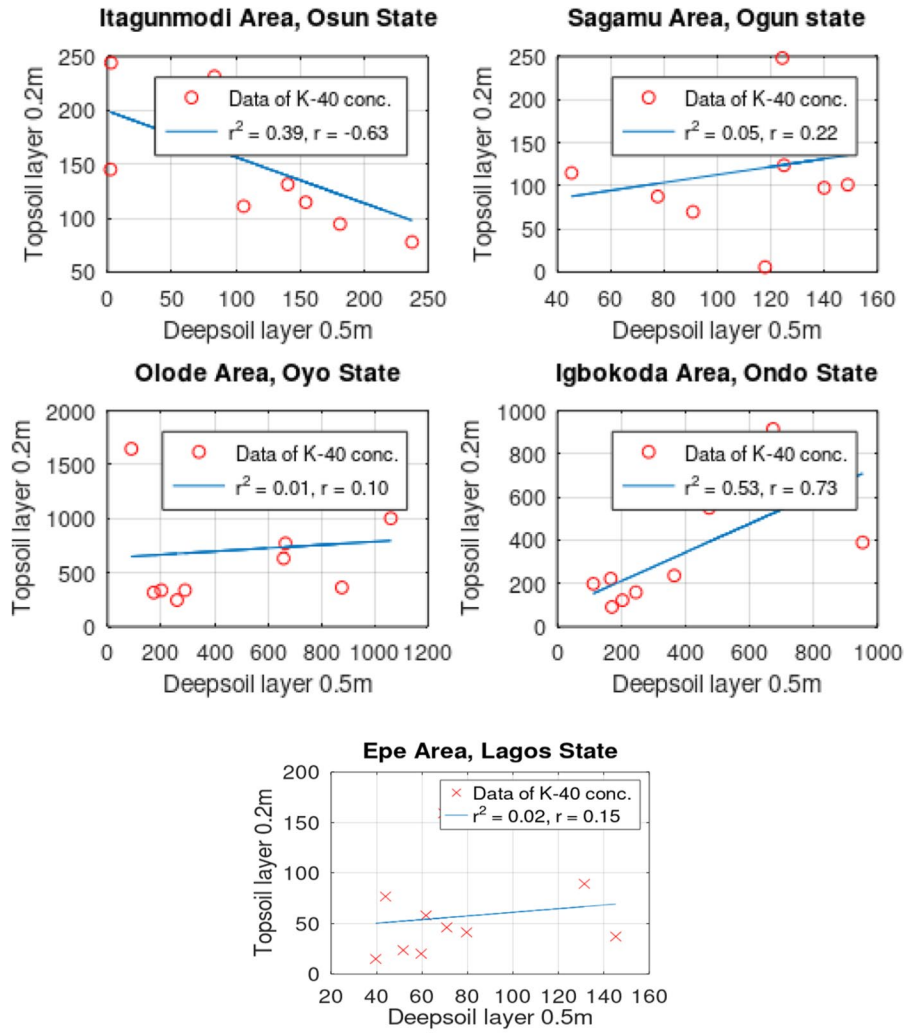


(b)

of the activity concentrations across each sampling location.

The ^{40} K and ^{232}Th radionuclides have negative and low skewness values (0.61–1.20) and (0.36–0.66) in the root zone layer in Itagunmodi (close to area where artisanal mining of gold is practiced), in Igbokoda (near bitumen mining activities are practiced) and in Sagamu (near where the mining of limestone is actively carried out), indicating a distribution with an asymmetric tail extending towards values that are more negative and general normal distribution. Also, the positive skewness values of the activity concentrations of the radionuclides in all the locations implied that the distributions are asymmetric. Similarly, the results of the kurtosis analysis of the activity concentrations at the surface and deep layer zone were low and negative values at Itagunmodi for ^{40} K, and ^{226}Ra; Olode for ^{40} K and ^{232}Th; and Igbokoda for ^{226}Ra and ^{232}Th where mining activities are actively practiced. This implies a relatively flat distribution and lack of extreme deviation in the activity concentrations (Adam & Mohamed, 2012; Ravisankar et al., 2014). In this study, the trend of the average of the activity concentrations in the root zone layer in Itagunmodi (near mining area), Sagamu, Igbokoda, Epe (far from mining area) and Olode is ^{40} K > ^{232}Th > ^{226}Ra, ^{40} K > ^{232}Th > ^{226}Ra, ^{40} K > ^{232}Th > ^{226}Ra, ^{232}Th > ^{40} K > ^{226}Ra and ^{40} K > ^{232}Th > ^{226}Ra, respectively. The variation in activity concentration may be linked to the artisanal

Fig. 3 (continued)



(c)

mining activities and the intensive agricultural activities with fertilizer application in the study locations.

Among the measured radionuclides, ^{40}K has the highest average activity concentrations which might be related to the high potassium contents (K-rich feldspar) in the mineral compositions of parent rocks and the most abundant terrestrial radionuclide in the crustal materials (Alajeeli et al., 2019; Ogundele et al., 2020). Minerals make up the majority of the potassium in soils. This potassium is arranged in the lattice of silicate minerals like feldspar and mica to a degree of 95–99 percent (fixed potassium). These minerals release potassium ions during weathering, which are then transported to soil fluids and adsorbed on clay cation-exchange sites (Nenadović et al. 2012). Apart from the geological composition of the soil, the high activity concentrations of ^{40}K can also be attributed to large scale utilization of predominantly potassium-bearing fertilizers in agricultural activities (Olatunji et al., 2014, 2022).

For the deep layer zone (0.5 m), the trend of the average of the activity concentrations in the Itagunmodi, Sagamu, Olode, Igbokoda and Epe is ^{40}K > ^{232}Th > ^{226}Ra, ^{40}K > ^{232}Th > ^{226}Ra, ^{40}K > ^{232}Th > ^{226}Ra, ^{40}K > ^{232}Th > ^{226}Ra and ^{232}Th > ^{40}K > ^{226}Ra, respectively. As shown, the lower concentration is observed at Epe in Lagos State which is far from mining area and therefore serves as a control. The average activity concentrations (Fig. 4) at different sites were similar except in a few cases.

Table 2 Descriptive statistics of the activity concentrations (Bq Kg^{-1}) at root zone layer (0.2 m)

Locations	Statistical parameters	^{40}K	^{226}Ra(^{238}U)	^{232}Th
Itagunmodi	Min	7.26	8.61	42.49
	Max	218.82	53.16	120.45
	Mean ± SD	122.17 ± 63.99	23.96 ± 14.29	87.01 ± 29.75
	Kurtosis	− 0.31	0.47	− 1.09
	Skewness	− 0.61	0.95	− 0.66
Sagamu	Min	45.40	13.98	69.44
	Max	148.78	105.76	135.55
	Mean ± SD	113.53 ± 32.35	52.81 ± 24.97	102.38 ± 20.05
	Kurtosis	− 0.81	1.69	0.40
	Skewness	− 1.20	0.83	− 0.47
Igbokoda	Min	112.81	7.99	84.30
	Max	953.08	111.71	243.14
	Mean ± SD	419.65 ± 302.04	42.84 ± 30.03	171.39 ± 49.79
	Kurtosis	− 0.84	2.32	− 0.40
	Skewness	0.82	1.37	− 0.36
Olode	Min	89.39	29.59	33.46
	Max	1060.71	97.08	205.96
	Mean ± SD	487.45 ± 330.61	58.86 ± 24.19	118.66 ± 72.40
	Kurtosis	− 1.07	− 0.72	− 2.13
	Skewness	0.47	0.54	0.03
Epe	Min	39.63	1.52	4.92
	Max	145.26	67.16	203.05
	Mean ± SD	75.35 ± 35.2	18.68 ± 21.50	105.56 ± 58.57
	Kurtosis	0.70	2.05	− 0.04
	Skewness	1.31	1.61	0.08

SD standard deviation

In all the locations ^{40}K had the highest activity concentrations except in Epe where the activity concentration of ^{232}Th was higher than ^{40}K. The respective maximum values of ^{40}K at Igbokoda and Olode were 2 and 3 times the value of 500 Bq kg^{-1} stipulated by the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR, 2014). The higher concentration of ^{232}Th in soils may be associated mainly to the active mining activities which are uncontrolled (Jeelani et al., 2022).

Table 4 presents the average soil activity from this study compared with the published data from different regions of the world. Significant discrepancies were observed in the values presented in Table 4. It was observed that the activity of ^{226}Ra in the farmland soils of the study area was higher compared to those reported for Nigeria at Ile-Ife (Oluyide et al., 2019), Mangoro-Agege (Ilori & Alausa, 2019), Bajoga (Kolo et al., 2019) and other parts of the world such as Laghouat in Algeria (Saadi et al., 2020) and Tarkwa in Ghana (Faanu et al., 2011). However, the highest average activity concentration of ^{226}Ra was reported at Jos, Plateau State in Nigeria (Adesiji & Ademola, 2019). This state is well known for age-long mining of tin and columbite and in many cases, the elevated level of activity concentrations of radionuclides has been linked to these anthropogenic activities. In the case of ^{232}Th, activity concentrations in the farmland soils of the study areas are higher than at all others regions, except in Abeokuta (Ekhaguere et al., 2019), Ile-Ife (Oluyide et al., 2019), Akwa Ibom (Akpan et al., 2020), Cairo (El-gamal et al., 2013), Northern Cyprus (Abbasi & Mirekhtiary, 2019), Laghouat in Algeria (Saadi et al., 2020), Tarkwa in Ghana (Faanu et al., 2011), Assam in India (Rangaswamy and Sannappa, (2021), Athina in Greek (Anagnostakis et al., 1996), and Sao Paulo in Brazil (Dantas et al., 2019). It is worth noting that Owo, Jos and some parts of Ogun State have been reported as high radionuclide background regions. The lowest

Table 3 Descriptive statistics of the activity concentrations (Bq kg^{-1}) at deep zone layer (0.5 m)

Locations	Statistical parameters	^{40}K	^{226}Ra(^{238}U)	^{232}Th
Itagunmodi	Min	63.87	2.12	52.21
	Max	229.93	52.82	106.98
	Mean ± SD	138.44 ± 50.30	27.86 ± 14.77	81.15 ± 15.47
	Kurtosis	− 0.29	0.68	− 0.62
	Skewness	0.31	0.14	− 0.39
Sagamu	Min	5.74	10.63	65.51
	Max	248.16	114.24	138.96
	Mean ± SD	119.06 ± 65.96	46.61 ± 37.37	91.84 ± 22.36
	Kurtosis	1.02	0.50	1.13
	Skewness	0.39	1.29	0.99
Olode	Min	246.53	17.20	75.64
	Max	1646.68	106.89	239.59
	Mean ± SD	709.71 ± 502.69	44.15 ± 30.22	147.23 ± 49.95
	Kurtosis	− 0.28	0.71	− 0.17
	Skewness	1.02	1.35	0.30
Igbokoda	Min	91.12	6.45	16.36
	Max	914.99	77.69	253.63
	Mean ± SD	365.57 ± 273.95	37.39 ± 24.29	140.08 ± 71.30
	Kurtosis	0.32	− 0.75	− 0.52
	Skewness	1.13	0.58	− 0.09
Epe	Min	15.09	1.45	76.36
	Max	159.24	51.14	170.70
	Mean ± SD	56.74 ± 43.29	18.73 ± 17.90	113.53 ± 24.40
	Kurtosis	2.99	0.06	3.48
	Skewness	1.63	1.20	1.25

SD standard deviation

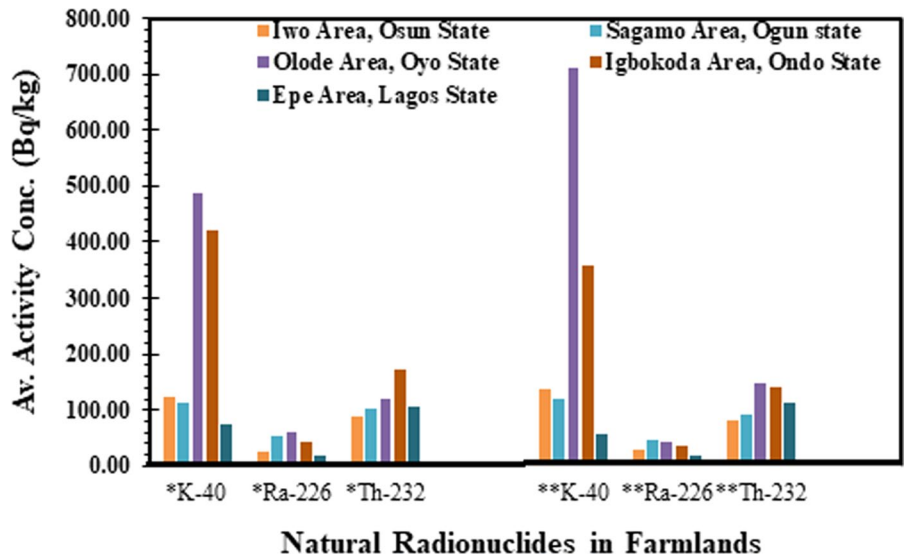


Fig. 4 Average activity concentration (Bq kg^{-1}) at 0.2 m and 0.5 m of the natural radionuclides in the farmlands

Table 4 Comparison of activity concentrations in the different areas of the world

Area & states in Nig	Radionuclide concentration ($BqKg^{-1}$)			Reference
	Ra-226	Th-232	K-40	
*Osun state (Iwo Area)	23.96	87.01	122.17	This study
**Osun state (Iwo Area)	27.86	81.15	138.44	
* Ogun state (Sagamo Area)	52.81	102.38	113.53	
** Ogun state (Sagamo Area)	46.61	91.84	119.06	
*Oyo state (Olode Area)	58.86	118.66	487.45	
**Oyo state (Olode Area)	44.15	147.23	709.71	
*Ondo state (Igbokoda Area)	42.84	171.39	419.65	
**Ondo state (Igbokoda Area)	37.39	140.08	356.57	
*Lagos state (Epe)	18.68	105.56	75.35	
** Lagos state (Epe)	18.73	113.52	56.74	
Abeokuta, Ogun state	30.87	47.10	261.29	Ekhaguere et al., 2019
Delta state	54.43	561.67	413.64	Ononugbo et al., 2019
Ile-Ife, Osun state	12.14	23.23	270.14	Oluyide et al., 2019
Coastal area, Akwa Ibom state	23.00	36.00	145.00	Akpan et al., 2020
Mangoro-Agege, Lagos state	11.47	10.44	403.07	Ilori & Alausa, 2019
Asa, Kwara state	42.86	18.15	570.91	Oluyide et al., 2019
Elere, Oyo state	36.55	29.05	537.3	Ilori & Alausa, 2019
Jos, Plateau state	242.13	1776.08	374.01	Adesiji & Ademola, 2019
Bituminous sand deposit area, Ogun state	42.60	113.00	461.00	Gbadamosi et al., 2018a
Onikitinbi, Ogun state	30.51	103.19	350.75	Gbadamosi et al., 2018b
Agbara, Ogun state	40.30	26.00	103.00	Gbadamosi et al., 2017
Egbeda, Oyo state	30.50	50.80	200.00	Owoade et al., 2019
Agbaaru, Oyo state	25.30	26.20	381.80	Ademola, 2019
Bajoga, Gombe state	7.41	16.27	196.11	Kolo et al., 2019
Zone A, Benue state	39.10	29.44	113.02	Kungur et al., 2020
Ajaokuta, Kogi state	31.00	36.00	712.00	Usikalu et al., 2018
Owo, Ondo state	64.64	110.18	1190.10	Aladeniyi et al., 2019
Rayfield-Du, Jos, Plateau state	168.83	436.08	346.10	Atipo et al., 2020
Ondo, Ondo state	171.80	19.80	146.20	Ogundele et al., 2020
Sao Paulo, Brazil	38.80	43.40	555.10	Dantas et al., 2019
Laghouat, Algeria	16.05	17.44	260.31	Saadi et al., 2020
Athina, Greek	25.00	21.00	355.00	Anagnostakis et al., 1996
Istanbul, Turkey	43.00	129.00	43.00	Karakelle et al., 2002
Assam, India	36.93	51.60	567.00	Rangaswamy & Sannappa, 2021
Izmir, Turkey	43.00	352.00	128.00	Nguyen et al., 2019
Cairo, Egypt	46.15	30.57	553.14	El-gamal et al., 2013
Northern Cyprus	83.70	53.60	593.90	Abbasi & Mirekhtiary, 2019
Tarkwa, Ghana	13.60	24.20	162.71	Faanu et al., 2011
World	33	45	420	UNSCEAR, 2000

*Represent 0.2 m (top soil layer), **Represent 0.5 m (deep soil layer)

activity concentration of ^{40}K in the farmland soils of the study areas was higher than that reported in Istanbul, Turkey (Karakelle et al., 2002), while the highest level of ^{40}K was reported in Owo, Ondo State (Aladeniyi et al., 2019).

Health risk parameter measurement in the farmlands

The results of the radiation hazard indices are presented in Table 5. The average values of the radium equivalent (Ra_{eq}) for Olode, Igbokoda, Sagamu, Itagunmodi, and Epe are 613.68, 697.10, 268.57, 255.99 and 232.11 Bqkg^{-1}, respectively. The Ra_{eq} values were less than the reference value of 370 Bqkg^{-1}, except in Olode and Igbokoda where the average values were 1.6 and 1.8, respectively, higher than the 370 Bqkg^{-1} reference value by the ICRP (ICRP, 2007).

The computed average values of internal hazard (H_{in}) and external hazard (H_{ex}) were both below one. For radiation hazard to be insignificant, the estimated values of H_{ex} and H_{in} must be less than one (Raghu et al., 2017). Since the values obtained in this study were less than one, the criterion condition (≤ 1) was satisfied, inferring that the risk due to internal exposure from radon gas and external exposure from terrestrial radiation is insignificant in the soil samples of the studied area. Hence, the implication is that the use of the soils in the studied areas for construction or building purposes does not warrant any health worries from the radiation physics point of view. However, periodic assessments of radioactivity levels of the soils are encouraged to forestall possible radiological hazards in the future due to changes in the radionuclide levels.

For excess lifetime cancer risks, naturally occurring radionuclides in environmental matrices, most especially soil, are known to produce carcinogenic effects by the accumulation of radon gas and its progeny in the air. The tendency of developing lung cancer due to exposure to gaseous radionuclides (radon gas) incurred over the lifetime of an individual defines the excess lifetime cancer risk (ELCR) (Kolo et al., 2015; Qureshi et al., 2014). Based on this, the calculated average values of the ELCR for Olode, Igbokoda, Sagamu, Epe, and Itagunmodi are 3.69×10^{-4}, 3.59×10^{-4}, 2.46×10^{-4}, 2.23×10^{-4} and 1.91×10^{-4}, respectively (as shown in Table 5). The respective ELCR values were 1.27, 1.24, 0.85, 0.76 and 0.65 times lower than the world average value of 0.29×10^{-3} for soil recommended by UNSCEAR and ICRP (Kolo et al., 2015; Qureshi et al., 2014). Progressive exposure to radon gas in the studied areas might enhance the risk of radiation carcinogenesis.

As it is shown in Table 5, the estimated dose indices, the average values of the total outdoor absorbed dose rate in the air for Olode, Igbokoda, Sagamu, Epe and Itagunmodi are 132.18, 128. 80, 88.05, 80.07 and 68.36 nGyh^{-1}, respectively. For such study location, the mean absorbed gamma dose rate obtained for the studied soil samples was higher than the worldwide population-weighted mean of 66 nGyh^{-1} as a stipulated reference value for safety purposes (UNSCEAR, 2008).

The estimated outdoor annual effective dose equivalent (AEDE) ranges from 83.84 μSvy^{-1} (Itagumodi) to 162.80 μSvy^{-1} (Olode) which were higher than the reference worldwide average value of 70.0 μSv y^{-1} recommended by the International Commission on Radiological Protection for soil (Adedokun et al., 2020). The location with high values implies high radiological risk which might occur, though gradually over the entire lifetime of the exposed population. Such risk involves radiation-induced cancer or development of heritable effects at some time in life (ICRP, 2007; Olatunji et al., 2022). Generally, the estimated values of the radiation risks and dose obtained in this study were lower than the permissive values recommended by the international bodies (ICRP and

Table 5 Estimated radiation dose and risk indices

Location	H_{ex}	H_{in}	Radiation hazard indices		Dose estimation indices	
			Ra_{eq} (Bq kg^{-1})	ELCR$\times10^{-4}$	D(nGy h^{-1})	AEDE (μSvy^{-1})
Itagunmodi	0.42	0.49	255.99	1.91	68.36	83.84
Sagamu	0.53	0.67	268.67	2.46	88.08	108.03
Olode	0.78	0.92	613.68	3.69	132.18	162.10
Igbokoda	0.79	0.90	697.10	3.59	128.80	157.96
Epe	0.49	0.54	232.11	2.23	80.07	98.20

UNSCEAR) saddled with the radiological safety from radiation exposure assessment. Also, the risks from the low-level NORMs in the farmland soil of the study region should not be neglected due to gradual exposure and bioaccumulation among the populace and the high possibility of transfer into the food crops being cultivated on the farmland.

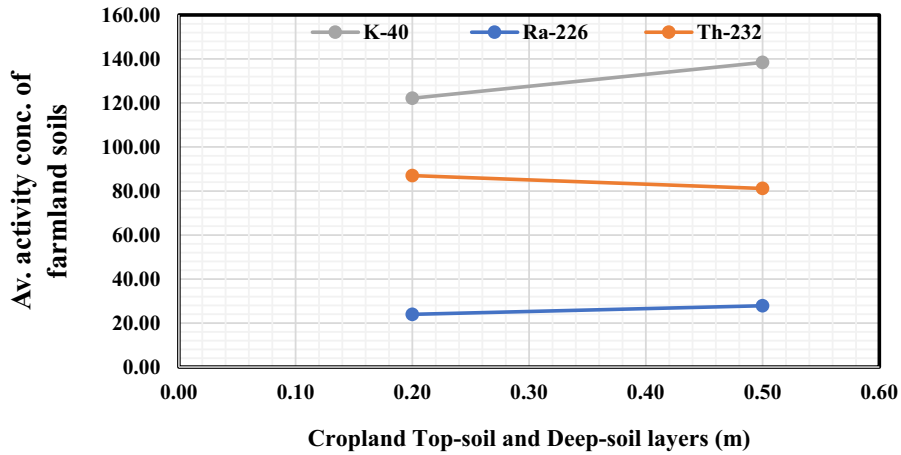
Distribution of NORMs in Soils from farmlands

The results of the vertical distribution of the mean activity concentrations of natural radionuclides from Itagunmodi, Sagamu, Olode, Igbokoda and Epe are presented in Figs. 5 (a, b, c, d, e). The vertical distributions of ^{232}Th, ^{226}Ra, and ^{40} K in all the studied locations show that the total content of radionuclides depends mainly on the parent material. It also revealed the fairly even distribution throughout the profile. Although in some cases, the accumulation of ^{232}Th, ^{226}Ra and ^{40} K occurs in the 0.5 m depth. A significant concentration of ^{232}Th, ^{226}Ra and ^{40} K might migrate and leach downward during physical processes such as percolation within the soil. This factor is greatly influenced by the physical and chemical soil properties. The vertical variability of the activity concentrations of the measured radionuclides in the soil samples could also be influenced by surface erosion activities, soil humic nature and soil chemical stability through the formation of stable aggregates. In addition, ^{40} K is part of mineral components of clay rather than organic matter, and its mobility in the soil is greatly controlled by its solubility (Bajoga et al., 2017).

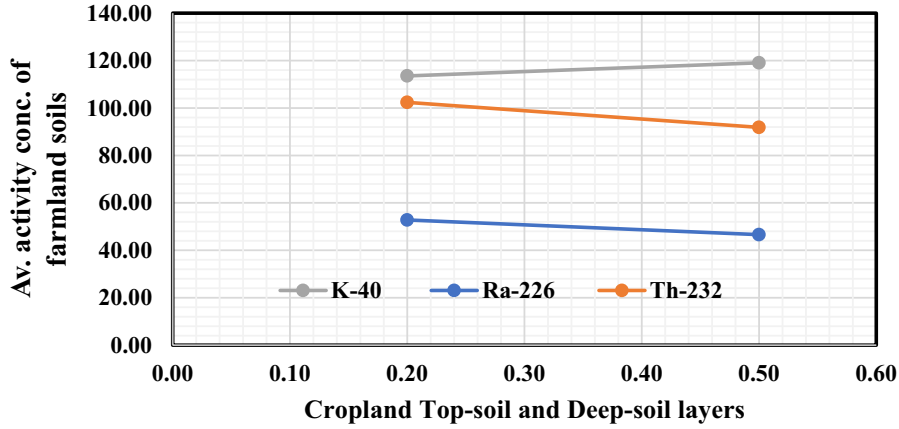
The pattern of the distribution was relatively constant except in few cases. This is due to the fact that the mobility of uranium in soil and its vertical transport (leaching) depend on properties of the soil (Bibler & Marson, 1992). The retention of uranium by the soil may be due to physical or chemical sorption (Allard et al., 1981) and the surface layers of the soil dominate the process between uranium and humic substances (Schultz et al., 1998). This is consistent with the presented results. In addition to ^{232}Th and ^{238}U/^{226}Ra, potassium isotope ^{40} K also contributes to the natural radioactivity especially those areas in close proximity to the active mining areas. The vertical distribution of ^{40} K with soil depth in farmland profiles shows a trend of gradual increase in concentration followed by a gradual decrease as observed in Igbokoda and Epe. Meanwhile, in Itagumodi, Sagamu and Olode, a trend of gradual decrease in concentration followed by a gradual increase of ^{40} K was observed. These differences in the trend might be due to the variation in physicochemical properties of the soil as a result of the mining activities which may have contributions to the vertical distributions of radionuclides.

Conclusion

This study has measured the activity concentration, estimated the radiological indices, and established the vertical distribution of the naturally occurring ^{40} K, ^{232}Th and ^{226}Ra/^{238}U in the farmland soil samples of southwestern Nigeria. The statistical model was performed using GNU Octave software to simulate the relationship between radionuclides in the root zone and the deep planting zone. Among the measured radionuclides, ^{40} K has the highest activity concentration in all the sampling locations except in a few cases. Negative and low skewness values of 0.16 and 1.20 for ^{40} K and ^{232}Th were obtained in the root zone layer in Olode and Sagamu, the two locations are close to an area where artisanal mining activities of gemstone and precious minerals, and limestone are practiced. The kurtosis analysis of the activity concentrations at the surface and deep layer zone were low and negative at Itagunmodi for ^{40} K and ^{226}Ra/^{238}U; at Olode for ^{40} K and ^{232}Th; and at Igbokoda for ^{226}Ra, ^{232}Th and ^{226}Ra/^{238}U where mining activities are actively practiced. The trend of the average of the activity concentrations in the root zone layer and deep layer zone across all states show that Epe in Lagos State where there is no record of mining activities has the lowest activity concentration values for all radionuclides compared to the other states known for mining activities. The average values of the radium equivalent (Ra_{eq}) for all study locations were also less than the reference value of 370 Bqkg^{-1}except in Olode and Igbokoda where average values were 1.6 and 1.8 times higher than the limit value. All other radiation indices estimated have also revealed some variations in values compared to the reference values recommended by standard organizations such as UNSCEAR and ICRP. These variations may



(a)



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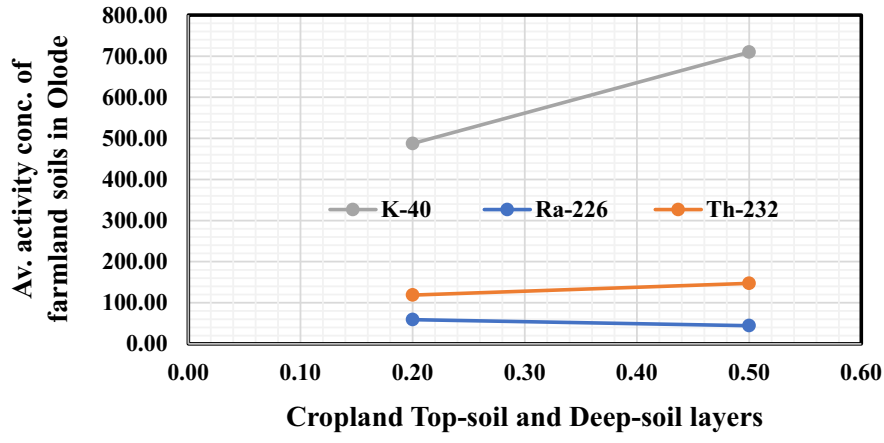
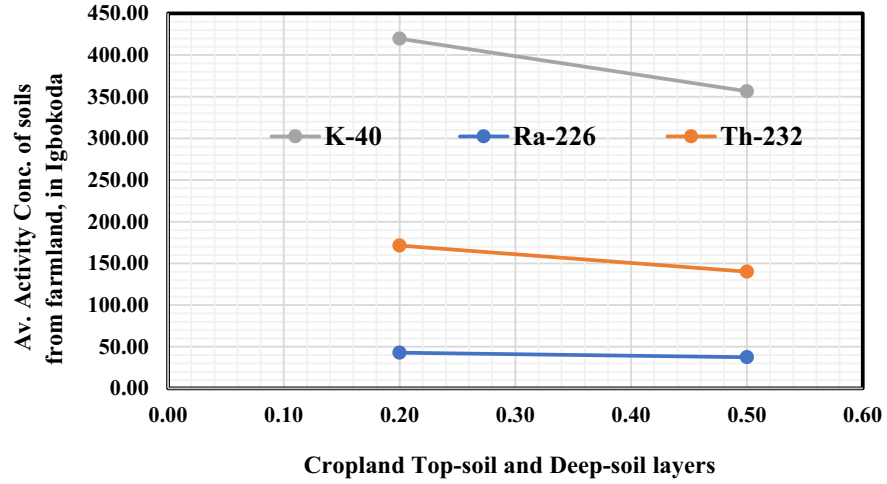


(c)

Fig. 5 (**a**) Vertical distribution of the mean activity concentrations of NORMs from Itagunmodi in Osun state (**b**) Vertical distribution of the mean activity concentrations of NORMs from Sagamu in Ogun state (**c**) Vertical distribution of the mean activity concentrations of NORMs from Olode in Oyo state. (**d**) Vertical distribution of the mean activity concentrations of NORMs from Igbokoda, Ondo state. (**e**) Vertical distribution of the mean activity concentrations of NORMs from Epe, Lagos state

Fig. 5 (continued)



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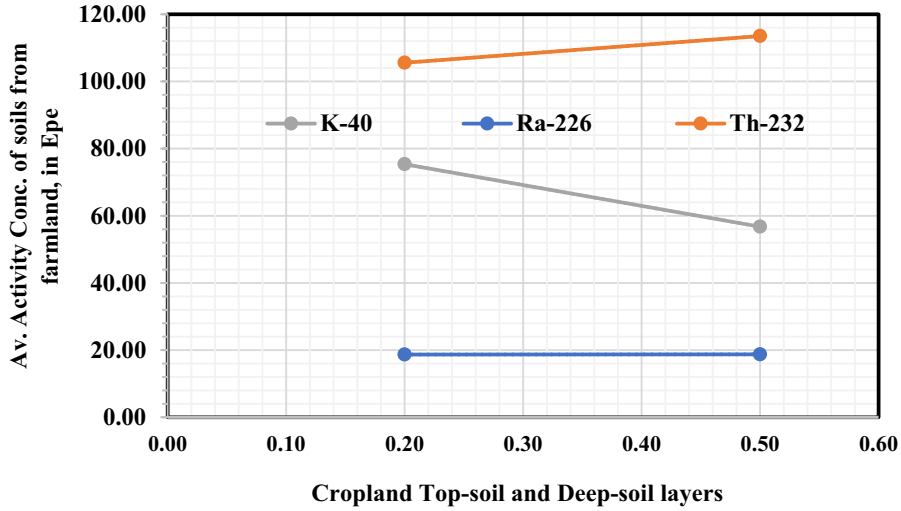

(e)

be linked to the artisanal mining activities and different agricultural activities in the study locations.

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Declarations

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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