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Radiometric investigation of naturally occurring radionuclides in soils from Igbokoda, a coastal area in Ondo State Nigeria

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Abstract

The activity concentration of naturally occurring radionuclides was evaluated in soil samples collected from Igbokoda, a coastal area of Ondo state in Southwest Nigeria, using a NaI(TI) detector. According to the results, the average levels of ^{238}U, ^{232}Th, and ^{40}K present in the soils are 37.63 ± 3.82, 23.20 ± 2.55, and 657.17 ± 45.15 Bq·kg^{-1}, respectively. The radiological results from Igbokoda, Nigeria, offer a varied comparison with other coastal regions. While ^{238}U and ^{232}Th levels in Igbokoda are within global averages and generally lower than in some areas, the ^{40}K levels are significantly higher, leading to an elevated radium equivalent activity (Raeq). Despite this, Raeq remains below the global safety threshold. Following the computation of the mean radiological risks, the reported values are absorbed gamma dose rate: 57·684 nGy·h^{-1}; annually effective dose rate: 70.744 μSv·y^{-1}; representative level index: 0.921; and radium equivalent: 121.413 Bq·kg^{-1}, respectively. The study results demonstrate that the population's radiation exposure resulting from the reported concentration of radionuclides in the soil of the study area is less than the levels recommended by global organizations. Therefore, the soil in the study region will not endanger the public. Nonetheless, more research is required to estimate the radionuclide concentration in the agricultural produce cultivated in the study area.

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Keywords: Igbokoda, NaI(TI), Natural radionuclide, Radiological risk, Soil

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1. Introduction

Naturally occurring radionuclides, particularly those from the uranium, thorium, and potassium decay series, are ubiquitous in the environment and can be found in varying soil concentrations [1, 2]. The assessment of these radionuclides is critical due to their potential impact on human health and the environment [3]. In Nigeria, several studies have focused on the radiometric analysis of soils, exploring the distribution, concentration, and health implications of these radionuclides across different regions [4–7]. However, a gap still needs to be in understanding the specific radiometric characteristics of soils in coastal areas, especially those affected by industrial activities such as crude oil exploration and spillage, such as Igbokoda.
Previous research conducted in various parts of Nigeria has highlighted elevated radionuclide levels in areas subjected to industrial activities [8–12]. Studies have shown that regions with significant oil exploration activities, such as the Niger Delta, tend to exhibit higher concentrations of radionuclides in soils

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[13–17]. These findings suggest a possible link between crude oil exploration, environmental contamination, and the accumulation of radionuclides in soils. However, these studies often focus on inland areas or specific regions, with limited attention given to coastal environments where the dynamics of radionuclide distribution may differ due to factors such as tidal influences, sediment transport, and the unique geochemistry of coastal soils.

Igbokoda, a coastal area in Ondo State, Nigeria, is of significant interest due to its proximity to crude oil exploration sites and its exposure to potential environmental contaminants, including oil spillages. The transportation of crude oil through pipelines and oil spills in coastal areas can lead to the contamination of soils with hydrocarbons and associated radionuclides [18, 19]. The interaction between oil spillages and the geochemical properties of coastal soils can enhance the mobility and concentration of radionuclides, posing a potential risk to the environment and the local population [20–22]. Despite the environmental significance of these interactions, more studies need to be conducted explicitly investigating the radiometric characteristics of soils in coastal areas like Igbokoda, which are vulnerable to oil-related pollution.

Estimating the concentration of radionuclides in soil is essential for determining the natural radiation levels in various environmental matrices, including plants, water, and buildings [9, 23]. Prolonged exposure to these soil radiation levels poses numerous radiological risks to humans, including cancer [24]. Therefore, assessing the degree of radioactivity in environmental samples remains one of the most important methods for estimating the potential risks of radionuclides in the environment.

This study focuses on the assessment of natural radionuclides (^{238}U, ^{232}Th, and ^{40}K) in the soil of Igbokoda, a coastal area in Ondo State, southwest Nigeria. The radiological risks related to the radionuclides in the research area were evaluated using well-established mathematical models. The activity concentration values and associated radiological risk assessments reported for various regions of the world will be compared with the results of this study.

2. Materials and techniques used for measuring radionuclides in soil

2.1. The study area

The study area, Igbokoda, is situated on the coastline of Ondo State, Nigeria, and serves as the administrative center of the Ilaje local government area. Based on estimates from Nigeria's 2006 population census, the population of Ondo state is estimated to be 170,123,740, with 2,509 and 290,615 people living in the Igbokoda area and Ilaje LGA, respectively [25]. Igbokoda is a region that spans the coastal sand bars that run from Okitipupa to the Atlantic Ocean in a northwest-to-southeast orientation. It is between longitudes of 4′3′′ E and 4′53′′ E and latitudes of 6′10′′ N and 6′25′′ N. Igbokoda's land areas are 8–10 meters above sea level; it experiences 27°C average annual temperatures and 2030 mm of precipitation on average. Its population density is 52,257 people per km², occupying a land area of 48,012 m² [26]. For the people of Igbokoda, fishing is the primary source of income, supported by related sea-based industries such as boat building, net production and repair, and trading.

Additionally, crude oil exploration is a significant revenue source for Nigeria's federal government. The lithological units in Igbokoda consist primarily of sedimentary formations influenced by nearby crystalline basement rocks. These formations significantly affect the distribution of naturally occurring radionuclides like ^{238}U, ^{232}Th, and ^{40}K. Alluvial deposits of sands, silts, and clays, typically low in radionuclides, can have increased levels due to their proximity to granitic and basement rocks, which are rich in U, Th, and K [27]. The weathering and erosion of these rocks contribute to the radionuclide content in the coastal plains [28]. Additionally, crude oil exploration and spillage further enhance radionuclide concentrations in the area. A geographic positioning system was employed to mark the sampling locations, which were then used to create the study area map, as illustrated in Figure 1.

2.2. Samples collection

At a depth of 5–15 cm, a clean hand trowel was used to take twelve (12) soil samples in this study. Four (4) grab samples, each weighing about one kilogram, were collected for each sample to create a composite sample. This means four soil samples weighing approximately one kilogram were collected from each sampling location. These samples were then mixed to form a single composite sample, representing the overall characteristics of that specific location. When collecting these samples, the precise boundaries of the sampling locations were noted, and sampling points were recorded. According to International Atomic Energy Agency Report No. 295, sampling complied with the guidelines [29, 30]. At the collection site, each soil sample was appropriately labeled with the sample code using paper tape and put in a plastic bag for identification.

2.3. Preparation of samples

The composite soil samples were suitably blended after removing extraneous materials like roots, dirt, and gravel. Extraneous materials were removed to access the soil aggregates. A different piece of paper was used to spread out the samples and allowed them to air dry for five days. After air-drying, larger debris masses were broken into smaller pieces by gently hammering the dried soil sample. At 105°C in an electric oven, soil samples were dried until they had a consistent dry weight [31, 32]. After that, a 2 mm sieve was used to filter the dried soil samples to achieve homogeneity [33]. Each prepared soil sample was weighed in cylindrical plastic containers that were identically sized and shaped to ensure the maximum level of counting accuracy and efficiency. The sample containers' geometric dimensions were comparable to those used for the reference standard sources to provide precise calibration requirements [34]. Then, the containers were sealed tightly with vinyl tape and stored for at least four weeks. This is done to ensure that, before measurement, the radium and its daughter nuclei have attained secular equilibrium [34, 35].

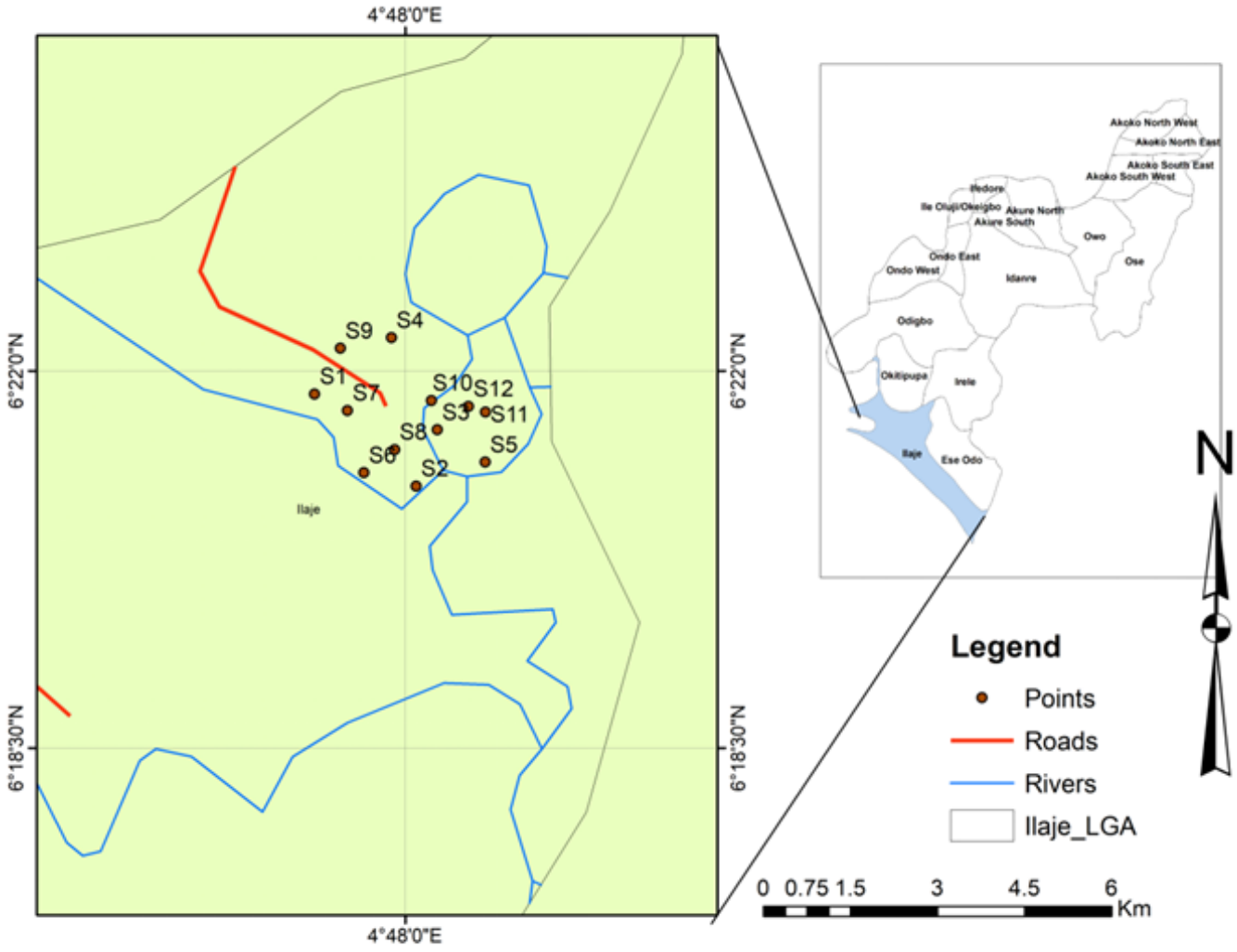

Figure 1. Ondo state's map with the study's sampling points highlighted.

2.4. *Measuring the activity levels of ^{238}U, ^{232}Th, and ^{40}K in the samples.*

The measurement used a NaI(TI)-doped gamma-ray spectrometry detector at the Radiation Physics Research Laboratory of Ladoke Akintola University, Nigeria. The system comprises an enclosed detector and a multichannel computer-resident quantum analyzer (MCA2100R). Figure 2 displays a typical NaI(TI) detector schematic representation.

The NaI(Tl) detector system detects gamma rays using a crystal that produces light when exposed to radiation. This light is converted into an electrical signal by a photomultiplier tube, amplified by a pre-amplifier and main amplifier. The signal is processed by a Multichannel Analyzer (MCA), which categorizes it based on energy levels to create a gamma radiation spectrum. A high-voltage power supply supports the system, and the base connects the detector to the photomultiplier tube, ensuring the entire system functions appropriately to measure gamma radiation.

The spectrum analysis was done using gamma analysis software (Palmtop Multichannel Analyzer) installed on a computer. The AQCS (USA) provided the reference standard sources to calibrate the detector efficiency and verify the activity of the specified radionuclides. Calibration with the standard sources was done to measure the detector's efficiency, and the samples being counted had the same geometry as the standard references. Gamma lines of ^{214}Pb at 351.92 keV and ^{214}Bi at 609.32 keV were used to measure the activity of ^{238}U. To determine ^{232}Th, gamma-energies of ^{228}Ac at 911.16 keV and ^{208}Tl at 2614 keV were utilized. The γ-ray at 1460.8 keV provided the basis for directly deciding the ^{40}K activity concentration. The sample mass was weighed using an electronic scale, and each sample took 3600 seconds to count when it was placed in the detector. Equation (1) was utilized to determine the activity concentration based on the detector's efficiency curve [37]:

$$A_E = \frac{C_E}{C_{eff}.\gamma_p.m.t} \quad (1)$$

A_E: activity concentration in Bq·kg^{-1} of the radionuclides of in-

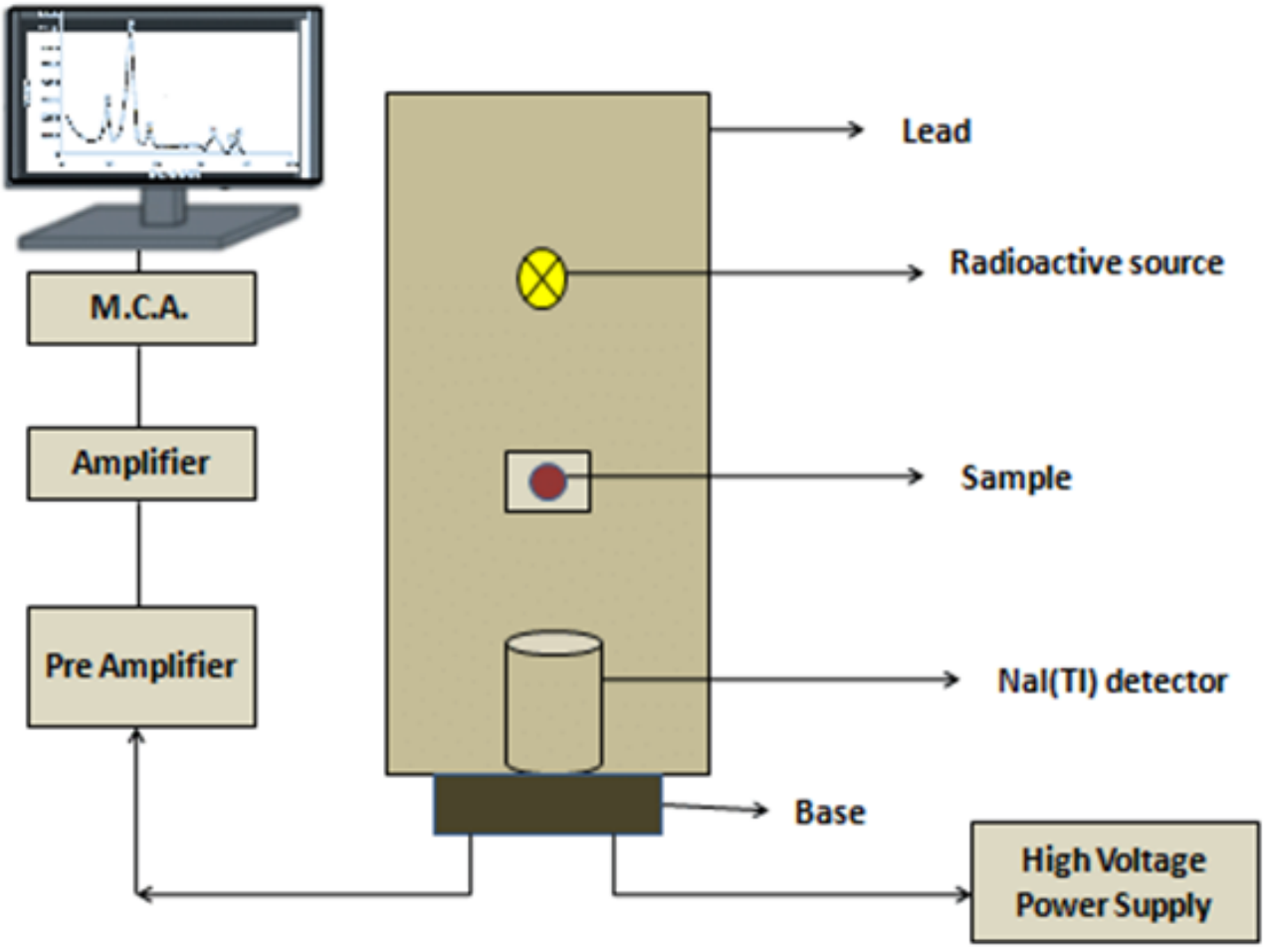

Figure 2. A schematic depiction of the NaI(Tl) detector (modified after [36]).

terest; C_E: net gamma counting rate in count per second; C_{eff}: detector's efficiency at energy E (keV); m: sample's dry mass in kg; t: sample counting time in seconds (s); and γ_p: gamma emission probability for a transition energy E (keV).

The gamma-ray detector system's ability to operate independently of the sample is indicated by its minimal detection limit (MDL) [38]. Equation (2) calculated the minimum detection limit required for each sample's precise radionuclide measurement [37]:

$$DL\ (Bqkg^{-1}) = \frac{F\sqrt{B}}{E.I_{\gamma}.T.m} \quad (2)$$

Where E: counting photo-peak efficiency ($cps{\cdot}Bq^{-1}$); I_{γ}: probability of emitting gamma rays; T: counting time in seconds (s); m: mass of the sample in kilograms (kg); F: conversion factor between counts per second (cps) and Bq which is 1.96 at a 95% confidence level; B: region of interest's net background count rate for the specific radionuclides. The present study shows that ^{238}U, ^{232}Th, and ^{40}K have minimum detection limits of 3.42, 3.80, and 14.08 $Bq \cdot kg^{-1}$, respectively.

3. Result and discussions

3.1. Activity concentrations of radionuclides in the soil samples

Table 1 presents the activity concentration values for radionuclides ^{238}U, ^{232}Th, and ^{40}K in the soil samples of the study area.

The soil samples from Igbokoda showed varying activity concentrations: ^{238}U ranged from 4.98 ± 1.95 to 59.48 ± 6.74 Bq·kg^{-1}; ^{232}Th ranged from 3.35 ± 1.60 to 55.12 ± 6.49 Bq·kg^{-1}; and ^{40}K ranged from 75.79 ± 24.06 to 895.26 ± 82.68 Bq·kg^{-1}. The average concentrations were 37.63 ± 3.82 Bq·kg^{-1} for ^{238}U, 23.20 ± 2.55 Bq·kg^{-1} for ^{232}Th, and 657.17 ± 45.15 Bq·kg^{-1} for ^{40}K.

The recommended limit of 33 Bq·kg^{-1} for ^{238}U, as given by UNSCEAR [2], is exceeded in soil samples S3 (34.62 ± 5.14 Bq·kg^{-1}), S5 (35.14 ± 5.18 Bq·kg^{-1}), S6 (45.08 ± 5.87 Bq·kg^{-1}), S7 (59.48 ± 6.74 Bq·kg^{-1}), and S9 (39.43 ± 5.49 Bq·kg^{-1}). These elevated levels suggest localized areas of higher uranium concentration, possibly due to geological or environmental factors specific to those sampling points. Similarly, the recommended limit of 45 Bq·kg^{-1} for ^{232}Th is ex-

Table 1. Concentrations of ^{238}U, ^{232}Th, and ^{40}K in soil samples from the study area.

Samples codes	^{238}U (Bq·kg^{-1})	^{232}Th (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})
S1	18.59 ± 3.77	13.25 ± 3.18	132.52 ± 31.81
S2	19.50 ± 3.86	12.90 ± 3.14	346.97± 51.47
S3	34.62 ± 5.14	8.52 ± 2.55	895.26 ± 82.68
S4	31.98 ± 4.94	18.34 ± 3.74	549.86 ± 64.80
S5	35.14 ± 5.18	19.04 ± 3.81	688.24 ± 72.49
S6	45.08 ± 5.87	48.65 ± 6.09	808.97 ± 78.59
S7	59.48 ± 6.74	55.12 ± 6.49	871.67 ± 81.58
S8	4.98 ± 1.95	3.35 ± 1.60	75.79 ± 24.06
S9	39.43 ± 5.49	21.06 ± 4.01	687.14± 72.43
S10	32.84 ± 5.01	11.12 ± 2.91	708.94± 73.57
S11	25.64 ± 2.05	9.15 ± 2.11	421.45 ± 33.56
S12	29.05 ± 4.91	11.48 ± 1.85	384.93 ± 41.05
Mean	37.63 ± 3.82	23.20 ± 2.55	657.17 ± 45.15
Min.	4.98 ± 1.95	3.35 ± 1.60	75.79 ± 24.06
Max.	59.48 ± 6.74	55.12 ± 6.49	895.26 ± 82.68
World Average (UNSCEAR, 2000)	33	45	450

ceeded in samples S6 (48.65 ± 6.09 Bq·kg^{-1}) and S7 (55.12 ± 6.49 Bq·kg^{-1}). The exceedance of thorium levels in these specific samples may indicate the presence of thorium-rich minerals or sediments in these areas. Most of the ^{40}K values exceed the recommended maximum limit of 450 Bq·kg^{-1} [2], including samples S3 (895.26 ± 82.68 Bq·kg^{-1}), S4 (549.86 ± 64.80 Bq·kg^{-1}), S5 (688.24 ± 72.49 Bq·kg^{-1}), S6 (808.97 ± 78.59 Bq·kg^{-1}), S7 (871.67 ± 81.58 Bq·kg^{-1}), S9 (687.14 ± 72.43 Bq·kg^{-1}), and S10 (708.94 ± 73.57 Bq·kg^{-1}). The consistently high levels of ^{40}K across multiple samples suggest a widespread presence of potassium-bearing minerals, which could be naturally elevated in this coastal region. The consistently high levels of radionuclides (^{238}U, ^{232}Th, and ^{40}K) across multiple samples suggest that the coastal area has a natural abundance of radionuclide-bearing minerals, possibly influenced by the presence of crude oil, as supported by studies from Agbalagba *et al.* [5], Iwetan *et al.* [8], Jibiri and Emelue [13], and Ilori *et al.* [41]. These elevated levels could be attributed to the region's unique geological formations and proximity to granitic and basement rocks, which contribute to higher radionuclide concentrations, consistent with the findings of Isinkaye *et al.* [12]. The coastal area's geological history and mineral distribution patterns may naturally result in the elevated radionuclide levels observed. Hence, radionuclides may be present in high concentrations in soils from Igbokoda due to the exploratory activities associated with crude oil. These align with studies demonstrating how the presence of crude oil in an environment raises the concentration of radionuclides in the surrounding media [40, 41]. Figure 3 illustrates the radioactivity distribution in soil samples.

3.2. Radiological risk assessments

Table 2 indicates the radiological risk assessments based on radioactivity measurements in soil samples from the research area:

An indicator of radium activity known as the "radium equivalent" (Raeq) offers a practical basis for securely monitoring radioactivity in an environment [42]. Equation (3) was used to calculate the radium equivalent activity resulting from the concentration of radionuclides in the soil [2]:

$$Ra_{eq}(Bq \cdot kg^{-1}) = A_u + 1.43A_{Th} + 0.077A_K \quad (3)$$

A_u, A_{Th}, and A_K depict the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in $Bqkg^{-1}$ in the soil sample.

Table 2 shows the calculated values of radium equivalent activity, which ranged from 15.606 to 205.420 $Bq \cdot kg^{-1}$, with 121.413 $Bq \cdot kg^{-1}$ as the mean value. The reported values for Ra_{eq} were below 370 $Bq \cdot kg^{-1}$, the recommended global average [2].

The absorbed gamma dose rate (D) at one meter above ground was calculated (equation (4)) using the values of ^{238}U, ^{232}Th, and ^{40}K in $Bq \cdot kg^{-1}$ [43]:

$$D(nGy \cdot h^{-1}) = 0.427A_u + 0.604A_{Th} + 0.042A_K, \quad (4)$$

where A_u, $A_{Th,}$ and A_K are the values of ^{238}U, ^{232}Th, and ^{40}K, and D is the absorbed gamma dose rate in $nGy \cdot h^{-1}$.

The absorbed gamma dose rate in the air varied from 7.333 to 95.301 $nGyh^{-1}$, with a mean value of 57.684 $nGy \cdot h^{-1}$ (Table 2). The majority of recorded absorbed gamma radiation rates fell below UNSCEAR's recommended safe limit of 60 $nGy \cdot h^{-1}$ [2], except for values found in soil samples S6 (82.629 $nGy \cdot h^{-1}$) and S7 (95.301 $nGy \cdot h^{-1}$).

The annual effective dose rate (AEDR) in$\mu Sv \cdot y^{-1}$ is the equivalent dose exposed to each body organ [44]. This was calculated using a conversion coefficient of 0.7 $Sv \cdot Gy^{-1}$, and the annual percentage of time spent outdoors by humans was set at 0.2 (20 percent). We computed the annual effective dosage rate [2] using equation (5):

$$AEDR(\mu Sv \cdot y^{-1}) =$$

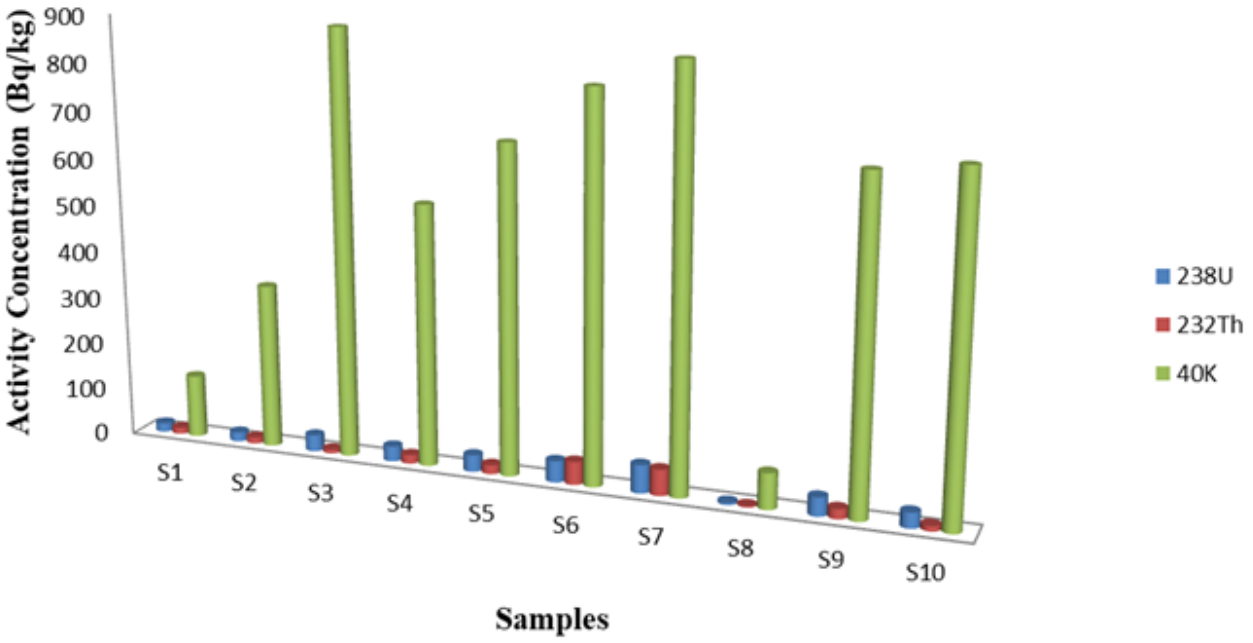

Figure 3. ^{238}U, ^{232}Th, and ^{40}K distribution in the study area's soil samples.

Table 2. Radiological hazard indices due to radioactive contamination in soil samples from the study area.

Samples codes	Ra_{eq} $(Bq \cdot kg^{-1})$	D$(nGy \cdot h^{-1})$	AEDR $(\mu Sv \cdot y^{-1})$	Iyr
S1	47.742	21.507	26.376	0.345
S2	64.664	30.691	37.639	0.490
S3	115.739	57.530	70.554	0.913
S4	100.545	47.827	58.655	0.763
S5	115.362	55.411	67.956	0.883
S6	176.983	82.629	101.336	1.327
S7	205.420	95.301	116.877	1.529
S8	15.606	7.333	8.993	0.117
S9	122.456	58.417	71.642	0.932
S10	103.330	50.515	61.951	0.803
S11	71.176	34.176	41.913	0.543
S12	75.106	35.505	43.544	0.565
Mean	121.413	57.684	70.744	0.921
Min.	15.606	7.333	8.993	0.117
Max.	205.420	95.301	116.877	1.529
World Average [2]	370	60	70	1

$$D(nGy \cdot h^{-1}) \times 8760(h \cdot y^{-1}) \times 0.2 \times 0.7(Sv \cdot Gy^{-1}) \quad (5)$$

With a mean value of 70.744 $\mu Sv \cdot y^{-1}$, the yearly effective dosage rate assessed from the research area's soil samples ranged from 8.993 to 116.877 $\mu Sv \cdot y^{-1}$. With a few exceptions, the majority of the AEDR values in soil samples S3 (70.554 $\mu Sv \cdot y^{-1}$), S6 (101.336 $\mu Sv \cdot y^{-1}$), S7 (116.877 $\mu Sv \cdot y^{-1}$), and S9 (71.642 $\mu Sv \cdot y^{-1}$) were below the 70 $\mu Sv \cdot y^{-1}$ recommended by UNSCEAR [2].

Using the representative level index ($I_{\gamma r}$), an area's soil can be assessed for radioactive contamination with ^{238}U, ^{232}Th, and ^{40}K. It determines the risk of radiation from radionuclides in a specific area [29]. Equation (6) established the representative level index for soil samples collected from Igbokoda [45]:

$$I_{\gamma r} = (A_U/150Bq \cdot kg^{-1}) + (A_{Th}/100Bq \cdot kg^{-1}) + (A_K/1500Bq \cdot kg^{-1}) \quad (6)$$

where A_U, A_{Th}, and A_K are the activity concentrations in Bq$\cdot kg^{-1}$ for ^{238}U, ^{232}Th, and ^{40}K.

The representative level index values for soil samples taken from the study area were 0.117 to 1.529, respectively, with a mean value of 0.921. Except for soil samples S6 (1.327) and S7 (1.529), all of the values in the study area fell under the recommended safe limit of 1 [2]. The radium equivalent, absorbed

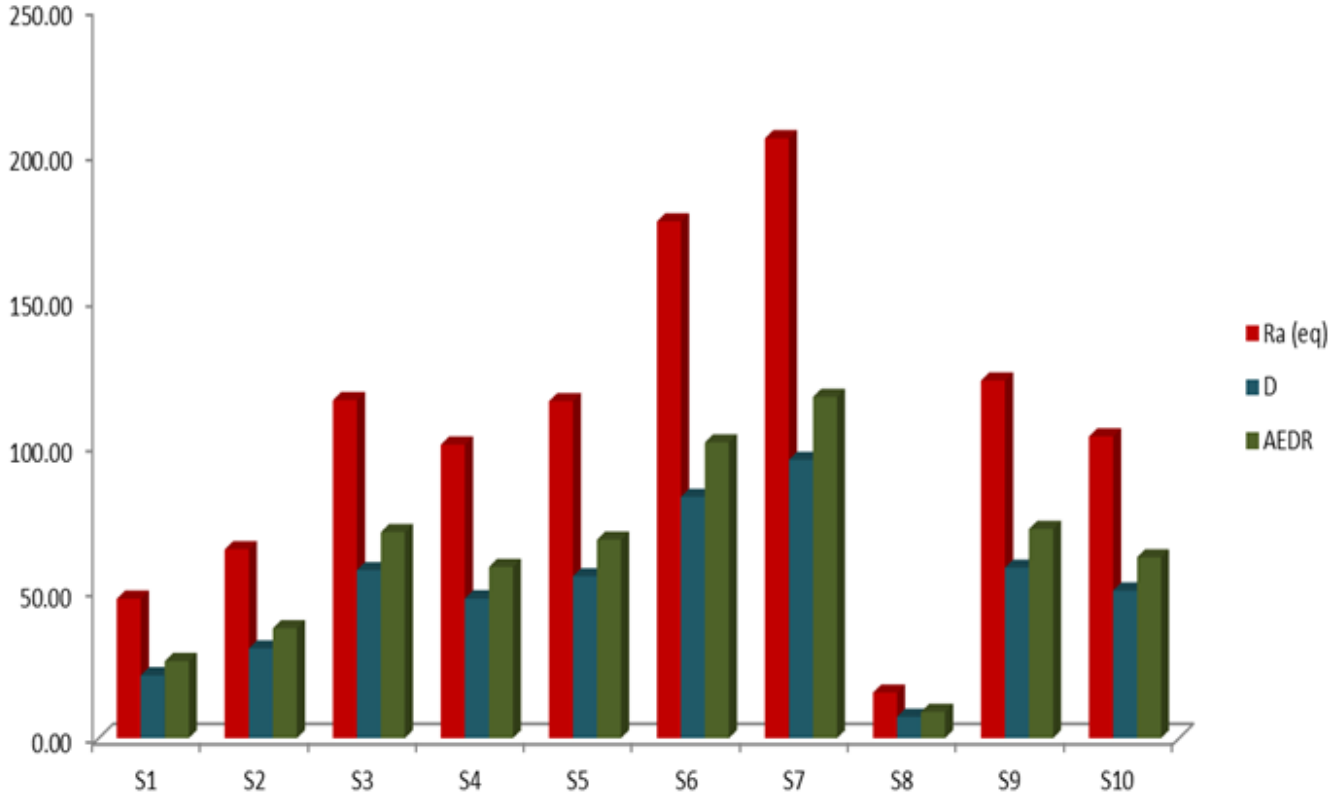

Figure 4. A graphic representation of the absorbed gamma dose rate, the annual effective dose rate, and the radium equivalent from soil samples collected at the study area.

Table 3. The ^{238}U, ^{232}Th, and ^{40}K concentrations in soil samples from the studied areas were compared with those of coastal regions worldwide.

Location	Detector	^{238}U	^{232}Th	^{40}K	Ra_{eq}	D	AEDR	Iyr	References
Ota, Nigeria	HPGe	40.44	94.44	134.25	185.82	81.32	-	-	[9]
Southwest China	HPGe	37.5	37.5	551	146	87.8	-	-	[46]
Western Cameroon	BEGe 6530	99	157	671	416.9	188.2	-	-	[47]
Coast of Gulf, Thailand	ICP-MS	5 - 49	4 - 108	3 - 714	11.1–257	5.0–117	6.1–143	-	[48]
Kaiga, India	HPGe	31.3	27.5	159.9	-	-	-	-	[49]
Northern Jordan	HPGe	49.9	26.7	291.1	103.1	51.5	63.2	-	[50]
Yanbu, Saudi Arabia	NaI(Tl)	40.65	42.89	513.16	140.8	65.8	-	-	[51]
Yobe, Nigeria	NaI(TI)	23	36	395	106	50	-	1.073	[52]
Metekel, Ethiopia	HPGe	64	70	330	133 - 229	172	85.0	0.48 - 0.81	[53]
Igbokoda, Nigeria	NaI(TI)	37.63	23.20	657.17	121.413	57.684	70.744	0.921	Present Study
World Average		33[a]	45[a]	450[a]	370[b]	60[b]	70[b]	1.00[a]	[a][30] [b][2]

gamma dose rate, and annual effective dose rate from soil samples collected within the study area are shown graphically in Figure 4. Table 3 compares the radiation concentration levels found in soil samples from the study area and published values from coastlines worldwide.

The radiological parameters from the study have significant implications for the environment and human health. Variations in gamma and annual effective dose rates (AEDR) reveal areas within the study region that may experience higher environmental radiation levels. Elevated radiation levels in these locations

could negatively impact local ecosystems. For instance, plants and animals in these areas might encounter increased radiation exposure, potentially disrupting growth, reproductive patterns, and other ecological functions.

From a health perspective, although most soil samples fall within the recommended radiological limits, the higher dose rates and AEDR observed in certain areas are concerning. Prolonged exposure to elevated radiation levels could pose serious health risks to residents, including an increased risk of cancer. These higher values highlight the need for continuous monitoring and possible corrective actions to safeguard public health.

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4. Conclusion

A thallium-doped sodium iodide (NaI(TI)) gamma-ray spectrometry detector was used to assess the specific activity of natural radionuclides in soils from Igbokoda, a coastal area in Ondo State, Nigeria. The findings reveal that while most radionuclide concentrations in the soil fall within safe limits, certain levels of ^{238}U, ^{232}Th, and ^{40}K exceed global safety recommendations. These elevated concentrations pose potential long-term health risks for the local population and underscore the need for ongoing monitoring and intervention. The region's geological characteristics, potentially influenced by crude oil and related activities, may contribute to the higher levels of certain radionuclides. Comparing the study's results with global benchmarks and data from other coastal regions is essential to ensure compliance with international safety standards. It is crucial to address areas where radiological parameters exceed recommended limits to mitigate potential environmental and population risks. The study concludes that proactive measures and strict regulatory adherence are vital to maintaining safety and preventing adverse health and environmental consequences.

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