



Science

ESTIMATION ON THE RADIOLOGICAL LEVEL OF NATURAL OCCURRING RADIONUCLIDES PRESENT IN FLOODED SOIL SAMPLES IN KADUNA NORTH, NIGERIA

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Abstract

The background radiation of the areas was collected at random for each point using a rados survey meter. The detectors (two rados survey meter were used) were placed 1 meter above the ground with the operator positioned a few meters away. Three (3) readings were taking from each detector in other to reduce error or reach accuracy in obtaining the background readings from each randomly selected point where soil samples were later collected.

In the current study, the concentration levels of naturally occurring radioactive materials (NORMs) of 40K, 232Th, and 238U in the surface soil samples of selected areas in kigo road new extension Kaduna north, Kaduna in Nigeria were studied. The collected soil samples were analyzed by means of gamma-ray spectrometry. The mean activity concentrations of the natural occurring radionuclides of 226Ra, 232Th and 40K in the soil samples were estimated to be 62, 78.35, 227.17 Bq/kg respectively for kigo road new extension respectively. Radium equivalent activity, absorbed dose rate, annual effective dose equivalent were also calculated for assessment of radiological risk. External hazard value (Hex) is between 0.3163 and 0.9557 and Internal hazard value is between 0.4462 and 1.1618. The worldwide average activity concentrations of 226Ra, 232Th and 40K in soil samples from various studies around the world have values of 37, 30 and 400 Bq/kg respectively [UNSCEAR, 2000]. The values compared well with published data from UNSCEAR shows Ra-226, Th-232 from the location are well above the standards while K-40 below the risk value.

Keywords: Norms; Gamma-Ray Spectrometry; Sodium Iodide (Nai) Detector; Radium Equivalent Activity; Annual Effective Dose Equivalent; Radiation Hazard Indices.

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1. Introduction

Estimation of radiological level of flooded soil sample in Kaduna metropolitan, Kaduna state Nigeria.

Radioactivity is a natural occurring phenomenon in the environment. In radioactive processes, particles or electromagnetic radiation are emitted from the nucleus. The most common forms of radiation emitted have been traditionally classified as alpha (α), beta (β), and gamma (γ) radiations. Nuclear radiation occurs in other forms, including the emission of protons or neutrons or spontaneous fission of a massive nucleus.

Of the nuclei found on Earth, the vast majority is stable. This is so because almost all short-lived radioactive nuclei have decayed during the history of the Earth. There are approximately 270 stable isotopes and 50 naturally occurring *radioisotopes* (radioactive isotopes) (International Association of Oil and Gas, NORM Task Force -2016). Thousands of other radioisotopes have been made in the laboratory.

All minerals and raw materials contain radionuclides of natural origin. The most important for the purposes of radiation protection are the radionuclides in the U-238 and Th-232 decay series. For most human activities involving minerals and raw materials, the levels of exposure to these radionuclides are not significantly greater than normal background levels and are not of concern for radiation protection. However, certain work activities can give rise to significantly enhanced exposures that may need to be controlled by regulation. Material giving rise to these enhanced exposures has become known as naturally occurring radioactive material (NORM).

NORM is the acronym for Naturally Occurring Radioactive Material, which potentially includes all radioactive elements found in the environment. However, the term is used more specifically for all naturally occurring radioactive materials where human activities have increased the potential for exposure compared with the unaltered situation. Concentrations of actual radionuclides may or may not have been increased; if they have, the term Technologically-Enhanced (TENORM) may be used.

Long-lived radioactive elements such as uranium, thorium and potassium and any of their decay products, such as radium and radon are examples of NORM. These elements have always been present in the Earth's crust and atmosphere, and are concentrated in some places, such as uranium ore bodies which may be mined. The term NORM exists also to distinguish 'natural radioactive material' from anthropogenic sources of radioactive material, such as those produced by nuclear power and used in nuclear medicine, where incidentally the radioactive properties of a material maybe what make it useful. However from the perspective of radiation doses to people, such a distinction is completely arbitrary.

The most common terrestrial radio Elements that produce gamma-rays are uranium ^{238}U , thorium ^{232}Th , and potassium ^{40}K (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)).

All naturally encountered elements are present in the Earth's crust in varying Concentrations. However, only eight of these – oxygen (O), silicon (Si), aluminum (Al), iron (Fe), calcium (Ca), sodium (Na), magnesium (Mg) and potassium (K) – make Up 98.5%.

As their half-life ($t^{1/2}$) times are extremely long ($=1.405 \times 10^{10}$ years, ^{235}U : $t^{1/2} = 7.038 \times 10^8$ years and ^{238}U : $t^{1/2} = 4.468 \times 10^9$ years), these nuclides have been present ever since planet Earth was formed and are called primordial or terrestrial NORs. Potassium (K) is widely distributed in nature in plants, animals, and rocks with the current isotopic distribution.

In secular equilibrium, the daughter nuclide activity concentrations are identical to that of their respective parent NOR. (International Association of Oil and Gas, NORM Task Force -2016). The effects of these radionuclides are due to gamma exposure of the body and irradiation of the lung from inhalation of radon and its daughter's nuclide. Therefore, the measure of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of world population [IAEA, 2003]. These radioactive elements have always been present in various concentrations in every part of the Earth's mantle, and are also present in the tissues of every living being, including man. Such materials do have the potential to initiate cancers in persons who are exposed to them. Although the concentration of NORM in most natural substances is so low that the risk is generally regarded as negligible, higher concentrations may come up due to intervention of man on earth surfaces and or natural disasters such as flooding (washing away of top layer of soil particles exposing this radioactive materials or transporting them from a different location to a habituated environment).

1.1. Statement of Problem

In recent times, there is an increased awareness of the potential problems of NORM and this has resulted in most countries taking steps to implement regulations dedicated to natural sources of radiation in their national legislations. The potential hazard occurs when the operator of the practice or the regulatory authority is not aware of the problems associated with the enhanced levels of NORM in raw materials and no protective actions are put in place so that doses to the public do not exceed the prescribed dose limits. The relevant route of exposure of the public is internal, via inhalation of dust and aerosols and ingestion of water and food. Radionuclides, such as ^{226}Ra and ^{228}Ra are known to have high mobility in the environment due to their high comparative solubility in water. Most of these radionuclides are predominantly alpha emitters and alpha particles tend to cause more internal hazard. Farming is also an important activity within these areas.

The soil, water bodies, dust and crops could be potential sinks for these radionuclides. The ultimate substrate of these radionuclides is the human body, which is the main concern of this study.

It's essential to experimentally check for the availability of natural occurring radioactive materials (NORMs) around the specified environment of interest by testing flooded soil samples. Vicinity that have experienced floods over time, washing away top layers of the earth mantle thereby exposing such radioactive materials.

1.2. Research Aim

The central aim of this study (or research) is to estimate the radiological level of flooded soil sample in Kaduna metropolitan with a view to assessing the risk to the public living in the environment due to NORM as a result of flooding. The study focused on the determination of the levels and distribution of the naturally occurring radionuclides of the U/Th decay series and ⁴⁰K in the surrounding communities. As a result, background radiation readings is TAKING USING a rados survey meter and soil samples were collected at selected points for analysis by gamma spectrometry using a SODIUM IODIDE detector (NaI). There is general concern about the health risk of NORM and national regulatory authorities are establishing guidelines and criteria for radiation protection from NORM. The public, which is the focus of this study, has very little or no understanding of radiation and risks concepts. In general, perceptions about radiation derived from natural sources, including radon, and from artificial sources may be different. There is also lack of understanding of the biological effects from both sources [IAEA, 2005]. .

1.3. Significance of the Study

Norms in the nation are barely considered or regulated to ascertain level of radiological hazards, also there is NONE or little awareness to the general public on it. The availability of data from such studies is very vital to all stakeholders involved since it will add to the data required for the development of guidelines for the regulation of norm in Nigeria.

2. Materials and Methods

2.1. Sample Collections and Preparations

Soil samples were collected from Abubakar kigo road new extension communities. In order to ensure representative samples were taken from the area for the analysis, initial survey was carried out in the area to determine the sampling points. The selection of the sampling Locations was based on the accessibility to the public and proximity of the area to residential areas. In addition, the geological map of the area was used to identify the locations where samples will be taken. Based on these criteria, 15 locations in general were identified for the soil samples (KGRP location id given to point where samples were collected for kigo road new extension). In the communities, samples were taken in areas (farms) where crops were grown also.

The sampling strategy that was adopted for the soil samples was random [ASTM, 1983, 1986; IAEA, 2004]. At each identified location samples were arbitrary collected within defined boundaries of the area of concern. Each location was divided into 50 m x 50 m grids and samples taken at different points and mixed together to give a sample. Each sampling point was selected independent of the location of all other sampling points. By this approach all locations within the area of concern had equal chance of being selected. The soil samples were taken using a coring tool to a depth of 5-10 cm. At each sampling location, samples of soil were taken from at least five different sections of the area into labelled plastic bags. One kilogram (1 kg) of each sample was collected for analysis. The samples were transported to the laboratory for preparation and analysis.

The collected sample was sent to the center for energy research and training for analysis were each of the soil sample collected were dried and crushed to fine powder with the use of pulverizer. Packaging of the sample into radon – impermeable cylindrical plastic containers which was selected based on the space allocation of the detector vessel which measures 7.6 cm by 7.6 cm in dimension (geometry) was also carried out. To prevent radon -222 escaping, the packaging in each case was tripled sealed.

The sealing process include smearing of the inner rim of each container lid with Vaseline jelly, feeling the lid assembly gap with candle wax to block the gap between lid and container, and tight-sealing lid-container with masking adhesive tape. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days prior to gamma spectroscopy measurement.

3. Results and Discussions

In other to ascertain the quality and the reliability of measurements the NaI detector was calibrated with respect to energy and efficiency using standard radionuclides, the terrestrial gamma dose rates measured at 1 meter above the ground at the sampling points in the study area are shown in Table 1, 2. The estimated average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K soil samples are shown in Table 3. They also include the results of the estimated absorbed dose rate, annual effective dose, radium equivalent, external and internal hazard indices in table 4. Fig 1 shows the chart of the hazard external and internal indices in comparison with the safe threshold from UNSCEAR.

Table 1: Background reading using 210152 survey meter

LOCATION ID	1 ST READING	2 ND READING	3 RD READING	MEAN READING
KGRP1	0.13	0.13	0.14	0.13
KGRP2	0.11	0.11	0.12	0.11
KGRP3	0.11	0.13	0.12	0.12
KGRP4	0.11	0.11	0.12	0.11
KGRP5	0.11	0.12	0.14	0.12
KGRP6	0.11	0.12	0.12	0.11
KGRP7	0.11	0.12	0.12	0.11
KGRP8	0.11	0.13	0.11	0.11
KGRP9	0.11	0.11	0.11	0.11
KGRP10	0.11	0.11	0.14	0.12
KGRP11	0.11	0.11	0.14	0.12
KGRP12	0.11	0.11	0.12	0.11
KGRP13	0.12	0.15	0.16	0.14
KGRP14	0.11	0.11	0.11	0.11
KGRP15	0.11	0.11	0.11	0.11

Table 2: Background reading using 990100 survey meter

LOCATION ID	1 ST READING	2 ND READING	3 RD READING	MEAN READING
KGRP1	0.11	0.11	0.12	0.11
KGRP2	0.11	0.11	0.11	0.11
KGRP3	0.12	0.14	0.13	0.13
KGRP4	0.11	0.11	0.11	0.11
KGRP5	0.13	0.13	0.14	0.13
KGRP6	0.11	0.11	0.11	0.11
KGRP7	0.11	0.11	0.11	0.11
KGRP8	0.11	0.12	0.12	0.11
KGRP9	0.11	0.14	0.15	0.13
KGRP10	0.11	0.11	0.11	0.11
KGRP11	0.11	0.12	0.12	0.11
KGRP12	0.11	0.11	0.12	0.11
KGRP13	0.12	0.11	0.12	0.12
KGRP14	0.11	0.11	0.11	0.11
KGRP15	0.11	0.11	0.11	0.11

Table 3: The estimated average activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K soil at kigo road new extension area

S/No	Sample ID	Ra-226 (BqKg ⁻¹)	Th-232 (BqKg ⁻¹)	K-40 (BqKg ⁻¹)
1	KGRP1	48.0681	33.8143	268.3542
2	KGRP2	31.7257	99.2018	41.4544
3	KGRP3	85.5876	81.1937	293.5593
4	KGRP4	57.9374	58.3494	183.9974
5	KGRP5	45.7506	52.0583	316.7266
6	KGRP6	76.9569	54.8893	231.4581
7	KGRP7	58.2571	143.7502	239.9314
8	KGRP8	72.3618	57.1305	307.6098
9	KGRP9	66.1286	112.6096	286.8022
10	KGRP10	65.5292	44.5877	11.8518
11	KGRP11	102.5692	59.7649	135.6787
12	KGRP12	64.6502	61.1017	252.6948
13	KGRP13	27.2506	93.9724	243.0418
14	KGRP14	76.2377	175.2054	352.121
15	KGRP15	51.0249	47.6546	242.2374
	Range	102.6-27.3	175.2-33.8	316.7-11.9
	Mean	62.0024	78.3523	227.1679

Standard deviation	19.8309	39.9278	97.16033
Worldwide mean	37	30	400
Worldwide range	60-17	64-11	850-140

Table 3 shows the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the soil/rock samples. The mean value of the activity concentrations of ^{226}Ra is 62.0024 Bq/kg in a range of 102.6 - 27.3 Bq/kg. For ^{232}Th the mean activity concentration 78.3523 Bq/kg in range of 175.2-33.8 Bq/kg and that of ^{40}K is 227.1679 Bq/kg in a range of 316.7-11.9 Bq/kg. The worldwide average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples from various studies around the world have values of 37, 30 and 400 Bq/kg respectively [UNSCEAR, 2000]. The values compared well with published data from UNSCEAR shows Ra-226, Th-232 are well above the standards while K-40 is below the risk value

3.1. Calculation for Radiological Effect

The following equations is used in calculating Radium equivalent (Ra_{eq}), absorbed dose rate (D), annual effective dose equivalent (AEDE), external radiation hazard index (H_{ex}), and internal radiation hazard index (H_{int}) for assessment for naturally occurring radioactive materials (NORMs) ^{238}U , ^{232}Th , and ^{40}K . The template (which covers energy, percentage error, count, uncertainty, and Activity concentration, uncertainty in activity, Gamma probability, uncertainty in Gamma probability, Efficiency and uncertainty in Efficiency) will be used to determine the radionuclide concentration in each sample. The activity concentration A , in unit of $\text{Bq}\cdot\text{kg}^{-1}$, for a radionuclide with a detected photopeak at energy E , can be obtained from Equation given by Awudu *et al.* and Faanu *et al.*

$$A_i = \frac{NC_i}{\varepsilon * \gamma_i * M * T}$$

Where

A_i is the i th radionuclide concentration.

NC_i = net count of the i th radionuclide.

ε = detector efficiency at the energy of the i th radionuclide.

γ_i = the emission probability of the i th radionuclide.

M = Mass of the soil sample in kg.

T = counting time.

Radium Equivalent (Ra_{eq})

The exposure to the γ -rays of materials that contain ^{238}U , ^{232}Th , and ^{40}K is defined in terms of radium equivalent given by (UNSCEAR-2000-17, 19)

$$\text{Ra}_{\text{eq}} (\text{Bq kg}^{-1}) = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \leq 370$$

Where A_{Ra} , A_{Th} and A_{K} are specific activity concentrations of Radium, Thorium and Potassium respectively.

Absorbed Dose Rate (D)

Absorbed dose rate as defined by (UNSCEAR-1988,24)

$$D(\text{nGy h}^{-1}) = 0.427A_{\text{Ra}} + A_{\text{Th}} + 0.043A_{\text{K}}$$

The absorbed dose rate in air, outdoors at 1m above the ground surface due to specific activity concentrations of ^{238}U , ^{232}Th , and ^{40}K .

Annual Effective Dose Equivalent (AEDE)

Annual effective dose equivalent due to the activity in soil will be calculated by the equation:

$$\text{AEDE (mSv.y}^{-1}\text{)} = \text{Dose rate (nGy.h}^{-1}\text{)} \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv.Gy}^{-1} \times 10^{-6}$$

Where the values 0.7 SvGy^{-1} is the conversion coefficient from absorbed dose in the air to effective dose received by adults, 8760 is the time in hours in one years, 0.2 represents the outdoor occupancy factor (UNSCEAR-2000), and is the observed dose rate.

The maximum permissible limit of R_{aeq} activity is 370 Bqkg^{-1} corresponds to annual effective dose equivalent to 1 mSvy^{-1} for general public. For radiation workers, the five years average is 100 mSv (ICRP-60. 1990)

External (H_{ex}) and Internal (H_{in}) Radiation Hazard Indices

The external hazard index (H_{ex}) will be calculated from the equation (Beretka & Mathew, 1985)

$$H_{\text{ex}} = A_{\text{Ra}}/370 + A_{\text{Th}}/259 + A_{\text{K}}/4810 \leq 1$$

Whereas the internal hazard index (H_{in}) was calculated from the equation

$$H_{\text{in}} = A_{\text{Ra}}/185 + A_{\text{Th}}/259 + A_{\text{K}}/4810 \leq 1$$

Table 4: Different Radiological Effects of ^{238}U , ^{232}Th , ^{40}K

Sample ID	Raeq (Bq/Kg)	D(nGy h-1)	AEDE (mSv.y ⁻¹)	H _{in}	H _{ex}
KGRP1	117.0858	65.8786	0.0808	0.4462	0.3163
KGRP2	176.7763	114.5312	0.1405	0.5631	0.4774
KGRP3	224.2987	130.3627	0.1599	0.8372	0.6058
KGRP4	155.5448	91.0006	0.1116	0.5767	0.4201
KGRP5	144.5819	85.2131	0.1045	0.5141	0.3905
KGRP6	173.2709	97.7026	0.1198	0.6760	0.4680
KGRP7	282.2946	178.943	0.2195	0.9198	0.7624
KGRP8	177.7444	101.2562	0.1242	0.6757	0.4801
KGRP9	249.2441	153.179	0.1879	0.8519	0.6731
KGRP10	130.2022	73.0783	0.0896	0.5288	0.3517
KGRP11	198.4803	109.3961	0.1342	0.8134	0.5362
KGRP12	171.4831	99.5732	0.1221	0.6379	0.4632
KGRP13	180.3454	116.0592	0.1423	0.5607	0.4870
KGRP14	353.8947	222.9001	0.2734	1.1618	0.9557
KGRP15	137.8233	79.8584	0.0979	0.5102	0.3723
Study mean	191.538	114.595	0.14055	0.6849	0.51732

study range	355.895-117.086	222.900-65.8786	0.2734-0.0808	1.1618-0.4462	0.9557-0.3163
World mean	370	57	0.48	≤1	≤1
World range		93-18	0.6-0.3		

The radium equivalent value for the study are below the world expected value (370 Bq/Kg) given by (UNSCEAR-2000-17, 19), the mean of the absorbed dose rate is above the world given value. A sample point is above the estimated hazard internal indices in the study area.

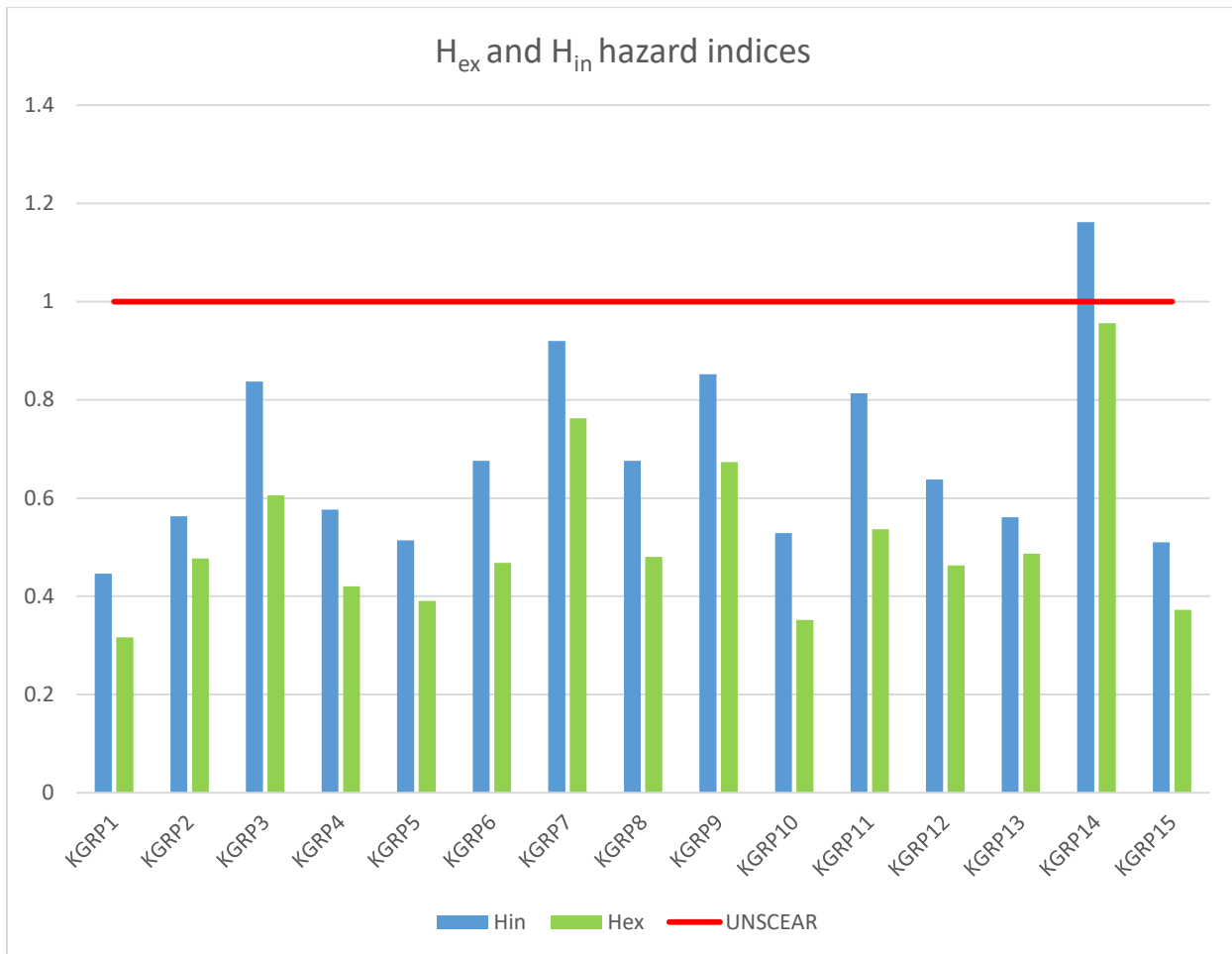


Figure 1: Showing the external and internal hazard indices of kigo road new extension in comparison with the world maximum permissible limit.

4. Conclusions & Recommendations

Radiation Exposure from NORM and Impact on the Public

The aim of this research work was to assess the risks to members of the public in the study area from exposure to natural sources of radiation from flooded soil samples. One (1) exposure

pathways considered for the study is; direct external gamma ray exposure from natural radioactivity concentrations in soil. The community covered during this study is kigo road new extension.

The study was motivated by the fact that the area is known to experience flooding annually. Prior to this study, no investigations have been conducted to obtain data on the activity concentration levels of the natural radionuclides ^{238}U , ^{232}Th and ^{40}K in the area. Consequently, the radiation doses and risks associated with these radionuclides have never been investigated. High levels of these elements could pose chemical and/or radiological hazards.

In this study, data on the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in different types of samples as well as radiation doses and risks have been established. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in different media for the potential pathways through which members of the public could be exposed were quantified using direct gamma spectroscopic analysis.

The mean activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the soil samples were estimated to be 62, 78.35, 227.17 Bq/kg respectively for kigo road new extension area. The results in this study compared well with other studies carried out in other countries and with the worldwide average activity concentrations (UNSCEAR, 2000).

The radiological hazards to the population in the study area were assessed based on the calculation of radium equivalent activity (Ra_{eq}), hazard indices (external and internal) for the soil/rock samples. The Ra_{eq} was found to be less than the recommended maximum value of 370 Bq/kg, and the external and internal hazard indices had values less than unity except at one point KGRP14 where the external hazard indices is above unity. It can be concluded that soil/rock materials that may be used for construction of buildings, farming etc. may not pose any significant radiological hazards.

The results obtained in this study show that the background radiation levels are within the natural limits. The data from this study can be used as baseline for future investigations.

The results from this study will serve as reference data for any future studies and also add up to data required to help develop guidelines for the regulation of NORM in Nigeria for radiation protection of workers and the public.

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