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Applying *in-Situ* and Laboratory-based Gamma Spectrometry to Determine Activity Concentration and Distribution of ^{40}K , ^{226}Ra and ^{232}Th in Abeokuta, Southwest Nigeria

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Abstract: The applications of radionuclides are potential sources of health risk and also a concern in the area of nuclear security. It is therefore imperative to determine the presence of the different radionuclides present in the environment at all times, because it is necessary to control and assess the risk level in the environment. The present study compares activity concentrations of the primordial radionuclides ^{40}K , ^{226}Ra and ^{232}Th obtained from laboratory gamma spectrometry measurements with the activity concentrations of the radionuclides as obtained from *in-situ* measurements. Soil samples were randomly collected from nineteen different points within Abeokuta city ensuring good coverage of the city area. A mobile gamma spectrometry system was used to collect gamma spectra measurements in the field. The obtained values are presented. The ranges of activity concentrations for ^{40}K , ^{226}Ra and ^{232}Th have been found to be 113 – 1975, 5 – 128 and 181 – 3284 Bqkg⁻¹, respectively for laboratory gamma spectrometry and 104 – 1312, 31 – 121 and 104 – 2578 Bqkg⁻¹, respectively for *in-situ* gamma spectrometry measurements. This study showed that the average activity concentrations of the primordial radionuclides in Abeokuta were much higher than worldwide averages of 400, 35 and 30 Bqkg⁻¹ for ^{40}K , ^{226}Ra and ^{232}Th , respectively. From both methods, ^{232}Th is seen to be the major contributor to the environmental radioactivity of Abeokuta. Good correlations also were deduced between the activity concentration results obtained from laboratory and *in-situ* gamma spectrometry, which therefore implies a significant relationship between the two methods used in the study.

Keywords: In-situ gamma, Gamma spectrometry, Activity concentration, Radionuclides.

Introduction

The presence of radionuclides in the environment is both of interest as well as of concern. It is of interest because radionuclides have been widely applied for various purposes, such as medicine, agriculture, electric power and education [1]. Examples of such radionuclides used are ^{131}I , ^{95}Zr , ^{241}Am , ^{60}Co , ^{137}Cs , ^{192}Ir , ^{232}U , ^{14}C , among others. Their presence is a potential source of health risk as well as a concern in the area of nuclear security. It is therefore

imperative to be able to determine the presence of different radionuclides present in the environment at all times so as to control and assess the risk level in the environment. Gamma-ray spectrometry measurements give practical ways to assess dispersed radionuclides in soils to determine possible changes in the radioactivity of the environment [2]. Both laboratory and *in-situ* gamma spectrometry are usually used for the purpose of detection, monitoring and assessment

of levels of radioactivity in the environment and radiation dose rates as a result of natural and artificial sources [3].

The traditional method of evaluating the distribution of radioactivity in the environment is laboratory gamma spectrometry measurements. It is well established and widely used. Samples are extracted from the site, packaged, transported to the laboratory and prepared for analysis. The method has the advantage of being fairly straight forward and the samples can be used for other types of analysis, like elemental analysis. Sampling, however, is labour-intensive with regard to the collection of samples and concerning laboratory measurements. This method suffers from the disadvantages arising from the representative nature of the samples, difficulty in accessing samples, coverage of sampling area, time required for laboratory analyses and delays in obtaining results [4], because all samples will have to be measured in the laboratory before any results are produced. *In-situ* gamma ray spectrometry entails the collection of a spectrum of the ambient gamma ray flux at a given site for analysis, principally to identify and quantify the radionuclides present at the site. Since its development in the late sixties and at the beginning of the seventies, *in-situ* gamma spectrometry has gone through a lot of modifications. It has become a useful method for rapid assessment of radionuclide concentrations of gamma emitters present in the environment [5]. The main advantages of this method include its modest time consumption, its portability and the reasonable cost of the detector compared to laboratory-based gamma spectrometry [6]. *In-situ* gamma-ray spectrometry is however complex and requires a more rigorous calibration procedure [7]. The present study compares activity concentrations of radionuclides calculated from laboratory gamma spectrometry and activity concentrations of radionuclides obtained from *in-situ* measurements.

Materials and Methods

Abeokuta, the capital of Ogun State, is located in the southwestern part of Nigeria. It is situated between latitudes 7°9'N and 7°39'N and longitudes 3°20'E and 3°54'E [8]. It is made up mainly of two local government areas, which are Abeokuta North and Abeokuta South. It covers a geographical area of 1,256 km². Abeokuta is found in a sub-humid tropical region within

southwestern Nigeria [9]. The city is drained mainly by River Ogun, which passes through it. The drainage pattern is generally dendritic. Geologically, the city is characterized by underlying basement complex rocks which are basically hornblende-biotite gneiss, porphyroblastic gneiss, granite, porphyritic granite and pegmatitic intrusions [8]. These rocks are noted for high content of radionuclides and poor water-bearing properties [10]. Characterizing the city is the Abeokuta formation which is sedimentary and comprises mainly sand with sandstone, siltstone, silt, clay, mudstone and shale interbeds [11]; [8]. The sands are coarse grained, clayey, micaceous and poorly sorted and indicative of short distances of transport and possible derivation from the granite rocks [12].

Calibration of *in-Situ* Gamma Spectrometer

The calibration of the detector was done in two stages. The first was to convert channel numbers into gamma energy in keV. The calibration procedure involved selecting the calibration option of the maestro digiBASE software and inserting the gamma energies of each of the peaks of interest against their channel numbers. A relation of the gamma energy *versus* channel number was established by the software, thereby calibrating the detector. The second was to convert the count rates (cps) under a photopeak into soil activity concentration (Bqkg⁻¹) of the radionuclide. In this stage of the calibration process, factors that related the count rate under a photopeak to soil radioactivity concentration and dose rate in air were determined.

According to [2], the number of counts per second, N_f , obtained under a photopeak due to a particular gamma energy, E , is related to the soil radioactivity concentration, A , of the radionuclide producing the peak by Eq. 1 [13].

$$\frac{N_f}{A} = \frac{N_f N_o}{N_o \phi A} \quad (1)$$

where $\frac{N_f}{A}$ is the photopeak count rate at the gamma energy per unit activity concentration of the radionuclide in the soil ($\frac{cps}{Bqkg^{-1}}$), $\frac{N_f}{N_o}$ is called the angular correction factor and accounts for the non-uniformity of the detector response to gamma rays incident at varying angles. This detector-specific quantity is dimensionless and dependent on the source distribution, gamma

energy, soil density and soil composition, $\frac{N_0}{\phi}$ is the on-axis response of the detector (normal to detector face) given in count rate per uncollided flux of parallel gamma rays ($\frac{\text{cps}}{\gamma\text{m}^{-2}\text{s}^{-1}}$), $\frac{\phi}{A}$ represents the total un-collided flux per unit source activity concentration ($\frac{\gamma\text{m}^{-2}\text{s}^{-1}}{\text{Bqkg}^{-1}}$) and ϕ is the gamma ray un-scattered flux on the detector ($\gamma\text{m}^{-2}\text{s}^{-1}$).

***In-Situ* Gamma Spectrometry**

The mobile gamma spectrometry system was used to collect gamma spectra measurements in the field. Concerted efforts were made to make sure that the sites where measurements were carried out were well spread across Abeokuta, ensuring also that at each location, the measurement represented the true terrestrial gamma radiation. The NaI(Tl) detector was mounted on the iron stand with the face of the detector pointing downward to receive gamma radiation emanating from the soil below, as shown in Fig. 1. The shield with a thickness of 4 cm and a depth of 17.5 cm was expected to

attenuate radiation from other sources or from outcrops and man-made constructions. Although the shield was not able to eliminate the effect of incident radiation completely, a larger shield would not have been easily movable, so a compromise had to be made between practicability and accuracy of the spectroscopy measurement. The detector itself was accurately folded with shock-resistive material in order to prevent it from possible damage in the field. The central axis of the detector was kept perpendicular to the extended plane of the soil surface while it faced the soil so that it could measure gamma radiation. The distance between the detector and the ground was kept at 140 mm and counting was for a preset time of 600 s. The photo-peaks used were 1460, 1760 and 2614 keV for ^{40}K , ^{226}Ra and ^{232}Th , respectively. Regions of interest (ROIs) were carefully determined around these photo-peaks in order to obtain total count rate, N_f , due to each radionuclide under its reference peak. N_f was converted into soil radioactivity concentration of the radionuclides using the conversion factors.

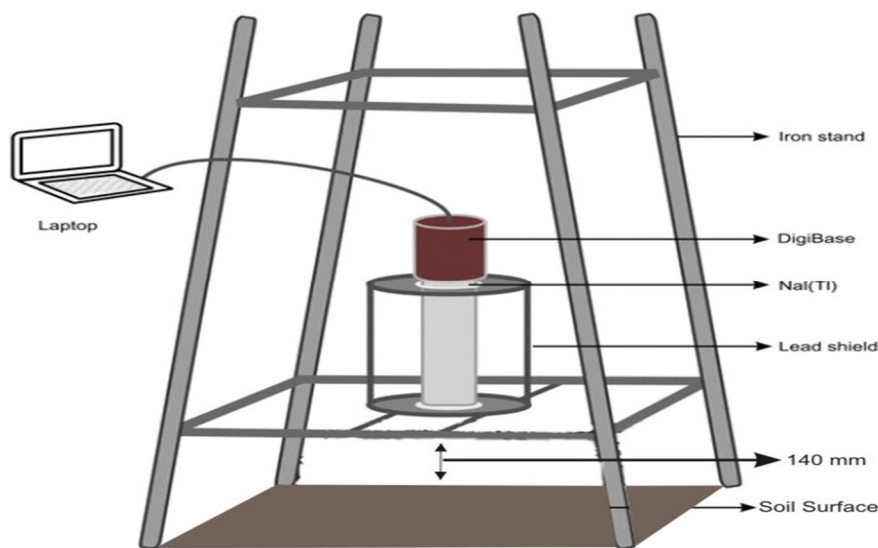


FIG. 1. Schematic diagram showing the arrangement of the detector during *in-situ* measurement.

Soil Sampling

Soil samples were collected from the same *in-situ* measurement points. The soil samples were taken from topsoil within the thickness of 0 – 50 mm from the surface. The collected soil samples were then packed in labeled cellophane bags and taken to the Radiation and Health Physics Laboratory of the Department of Physics, Federal University of Agriculture, Abeokuta (FUNAAB), where they were air-dried at room

temperature [14], pulverized into fine grain sizes and sieved with a 1 mm mesh sieve. The prepared samples were then packed in 0.2 kg of mass in plastic containers of about 80 mm in diameter so that the sample could sit with high geometry on the NaI(Tl) detector, as shown in Fig. 2. The plastic containers were then sealed with adhesive paper tape and kept for about 30 days which was a sufficient time to allow for ^{226}Ra and its decay products to attain a state of secular radioactive equilibrium.

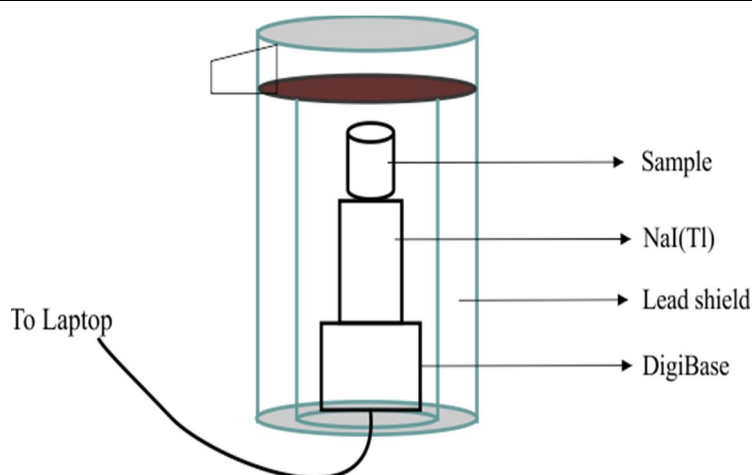


FIG. 2. Schematic diagram showing the soil sample on the NaI(Tl).

Calibration of Laboratory-based Gamma Spectrometer

Standard material used for energy calibration of the laboratory-based gamma spectrometer consists of RGU-1, RGTh-1 and RGK-1. These are standard materials that were produced under the auspices of the International Atomic Energy Agency (IAEA) and were distributed through its Analytical Control Services (AQCS) program. According to the manufacturers, RGU-1 was prepared from a dilution of Uranium ore (BL-5) with silica sand, RGTh-1 was prepared from a dilution of Britholite material (OKA-2) with silica sand and RGK-1 was prepared from potassium sulphate. The standard materials were put into the sample containers and were kept for about one month to achieve equilibrium between radium and its decay products. The standard materials were counted using the NaI(Tl) detector. Counting was for about 1800 s, which was long enough to capture spectra with well-resolved peaks. Three suitable energy peaks of 352, 609 and 1764 keV were selected. The calibration procedure continued by selecting the calculation option of the maestro digiBase software, calibration option was further selected and the gamma energies of each of the peaks of interest were inserted against their channel numbers. A relation of the gamma energy *versus* channel number was established by the software, then the channel numbers and the corresponding energy values were matched. The activity of each of the three primordial radionuclides was determined by relating the area under each photo-peak in the soil samples to the same photo-peak in the standard materials after counting. The photopeaks used were 1460 keV,

1764 keV and 2614 keV for ^{40}K , ^{226}Ra and ^{232}Th , respectively.

Laboratory-based Gamma Spectrometry

Gamma-ray spectrometry analysis of the prepared soil samples was carried out using the NaI(Tl) gamma ray spectrometer in the Radiation and Health Laboratory, Department of Physics, FUNAAB. The counting time was set at 10800 seconds. Regions of interest were carefully determined from the downloaded spectra and were used to obtain total counts under the three major peaks of 1460 keV ^{40}K , 1764 keV ^{214}Bi and 2614 keV ^{208}Tl . The peaks had a range of 160 - 190, 200 - 235 and 295 - 335 keV, respectively. The background count was also determined by counting another container, similar to the one used for the soil samples, filled with distilled water which was sealed and kept for about 30 days. The net area, after background counting was taken away, and was related to the radioactivity concentration of each of the three primordial radionuclides. Activity concentrations of the primordial radionuclides in the soil samples was calculated using Eq. 2 [15].

$$\frac{A_{\text{sample}}}{A_{\text{std}}} = \frac{CR_{\text{sample}} - CR_{\text{Bgd}}}{CR_{\text{std}} - CR_{\text{Bgd}}} \quad (2)$$

where A_{sample} is activity concentration of the sample (Bqkg^{-1}), A_{std} is activity concentration of the standard (Bqkg^{-1}), CR_{sample} is count rate of sample (counts sec^{-1}), CR_{Bgd} is count rate of background (counts sec^{-1}), CR_{std} is background of standard (counts sec^{-1}), CR_{Bgd} is count rate of background (counts sec^{-1}).

Fig. 3 shows the area of the study where both *in-situ* gamma spectrometry as well as the soil sampling were done.

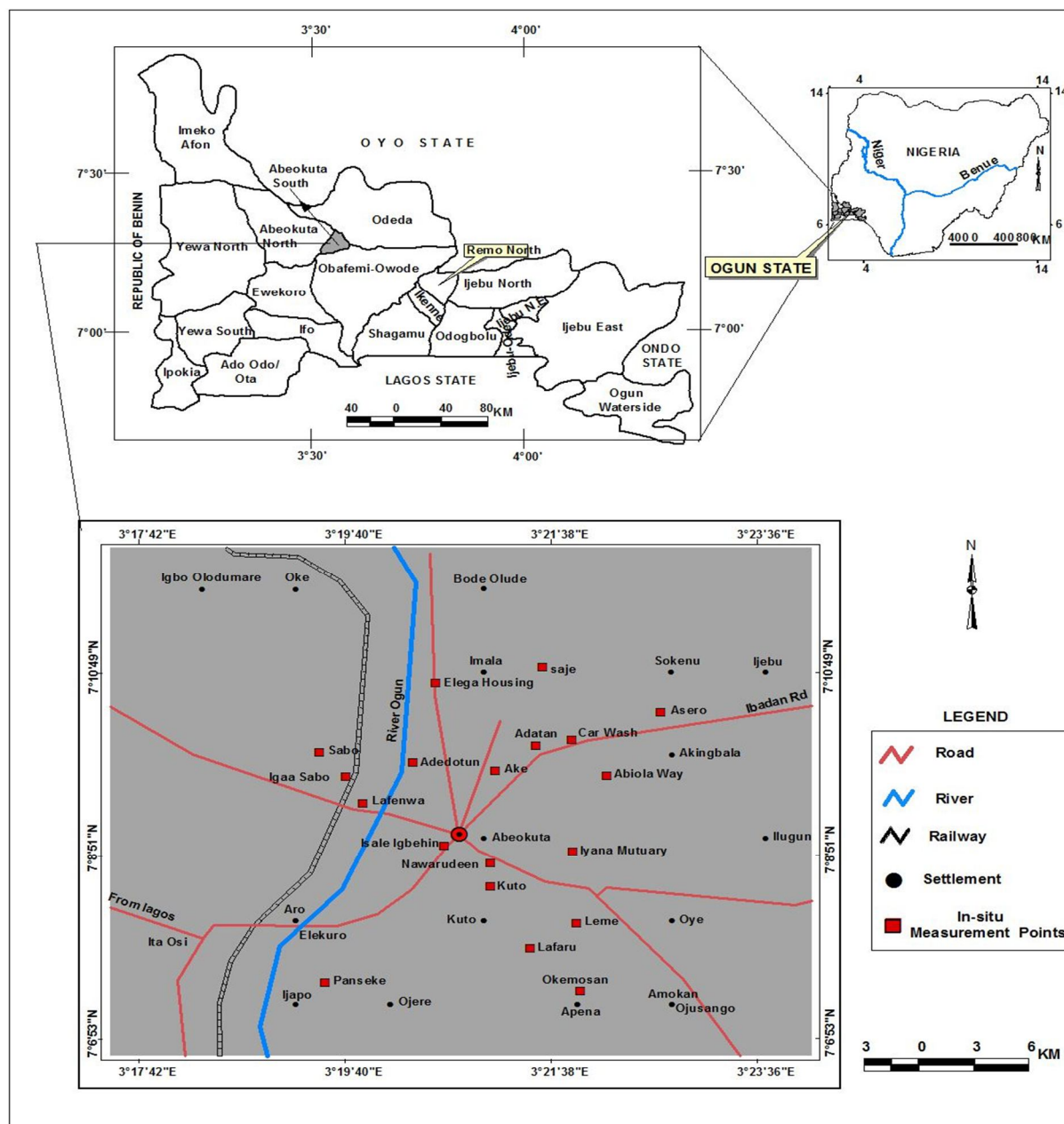


FIG. 3. Map of the study area showing Abeokuta South and Abeokuta North.

Results and Discussion

The three primordial radionuclides, ^{40}K , ^{226}Ra and ^{232}Th have been detected and measured. Activity concentrations of the radionuclides from the soil samples as determined in the laboratory and from *in-situ* gamma spectrometry are presented in Table 1. From the laboratory-based gamma spectrometry, of special interest are concentrations of ^{232}Th at Kuto and Lafaru at 3284 and 2857 Bqkg^{-1} , respectively which are both over three times the average of the study area and concentrations of ^{40}K at Kuto and Lafaru at 1924 and 1816 Bqkg^{-1} , respectively

which are both over two times the average of the study area. From the *in-situ* gamma spectrometry, concentrations of interest are ^{232}Th at Lafaru and Kuto at 2578 and 2362 Bqkg^{-1} , respectively which are both over three times the average of the study area and concentrations of ^{40}K at Leme and Adedotun at 1312 and 1147 Bqkg^{-1} , respectively which are both over two times the average of the study area. These anomalously high concentrations at some points account for the relatively high activity concentration level of the city. This confirms the general report in different studies carried out by different authors from Nigeria, that Abeokuta

town has a high radiation dose [10]; [16]. Frequency distributions of activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th from both methods are presented in Figs. 4 to 9. The distributions of the concentrations of the three radionuclides in the study area show significant variations in both laboratory-based spectrometry

and the *in-situ* spectrometry. This shows non-homogenous distributions of ^{40}K , ^{226}Ra and ^{232}Th . The frequency distributions of the values of the activity concentrations obtained from laboratory and *in-situ* gamma spectrometry show same pattern, where ^{40}K is near normal, ^{226}Ra is normal and ^{232}Th is skewed to the left.

TABLE 1. Results of activity concentrations of naturally occurring radionuclides from both laboratory and *in-situ* gamma spectrometry.

S/N	Location	lab	<i>in-situ</i> $^{40}\text{K}(\text{Bqkg}^{-1})$	lab	<i>in-situ</i> $^{226}\text{Ra}(\text{Bqkg}^{-1})$	lab	<i>in-situ</i> $^{232}\text{Th}(\text{Bqkg}^{-1})$
1	Ake	1796	233	57	68	1432	234
2	Abiola Way	113	219	30	64	181	419
3	Adatan	2036	598	97	79	1826	725
4	Adedotun	1887	1147	35	64	1554	634
5	Asero	482	346	43	54	645	353
6	Car Wash	1082	402	70.3	47	824	222
7	Elega Housing	643	202	64	82	735	626
8	Igaasabo	848	188	34	35	540	185
9	Iyanamutary	768	188	51	42	229	185
10	Isaleigbehin	778	281	5	31	491	1030
11	Kuto	1924	937	68	100	3284	236
12	Lafaru	1816	896	66	90	2857	2578
13	Lafenwa	996	169	10	36	608	104
14	Leme	1975	1312	66	121	1513	1321
15	Nawarudeen	1616	293	128	74	1893	467
16	Okemosan	501	104	31	56	365	200
17	Panseke	680	315	27	65	237	153
18	Sabo	1831	291	26	45	686	191
19	Saje	1240	794	46	85	877	756
MEAN		1211	469	50	65	1094	559
MIN.		113	104	5	31	181	104
MAX.		1975	1312	128	121	3284	2578

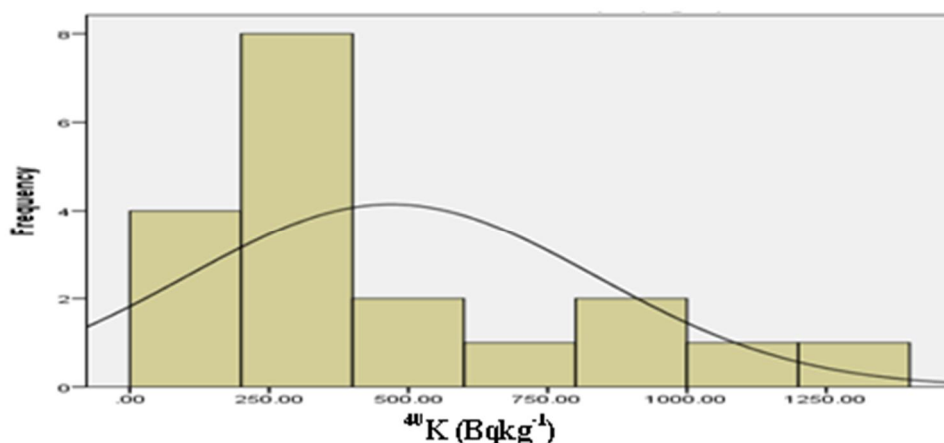


FIG. 4. Frequency distribution of activity concentration of ^{40}K of *in-situ* gamma spectrometry in Abeokuta.

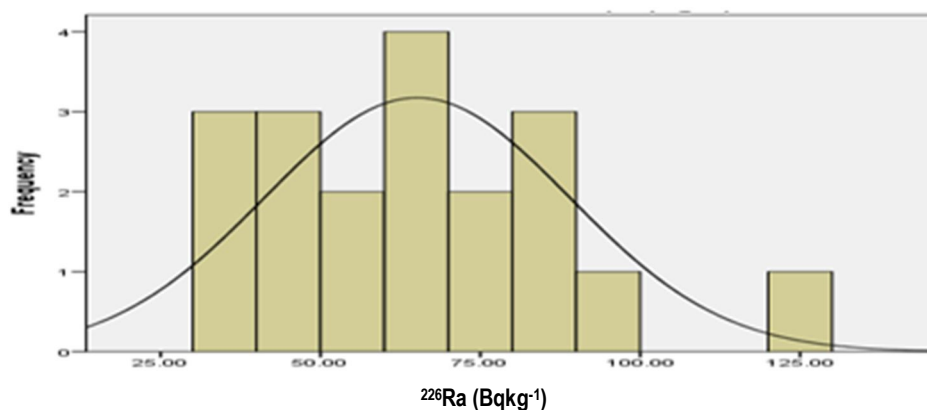


FIG. 5. Frequency distribution of activity concentration of ^{226}Ra of *in-situ* gamma spectrometry in Abeokuta.

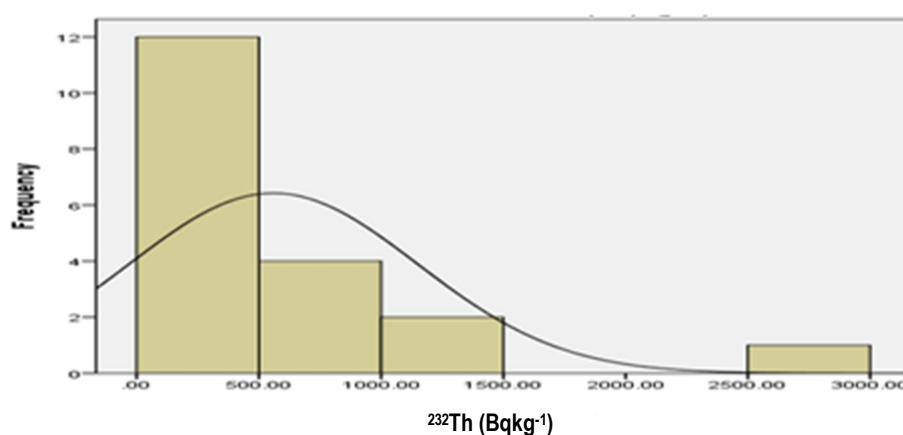


FIG. 6. Frequency distribution of activity concentration of ^{232}Th of *in-situ* gamma spectrometry in Abeokuta.

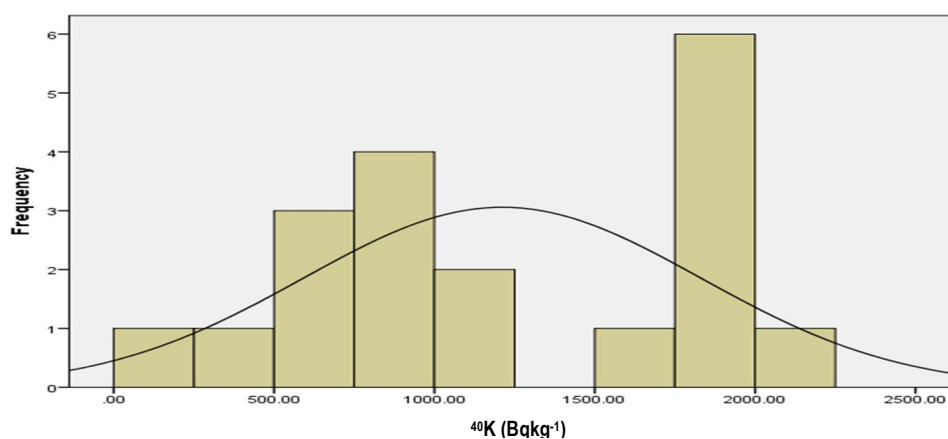


FIG. 7. Frequency distribution of activity concentration of ^{40}K of laboratory-based gamma spectrometry in Abeokuta.

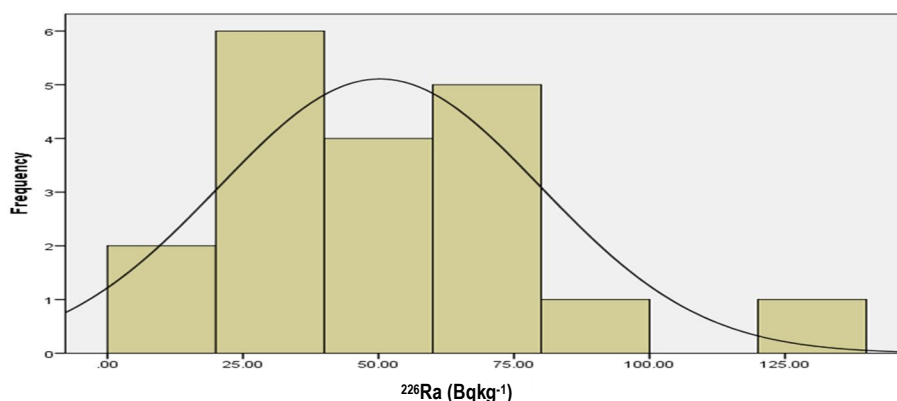


FIG. 8. Frequency distribution of activity concentration of ^{226}Ra of laboratory-based gamma spectrometry in Abeokuta.

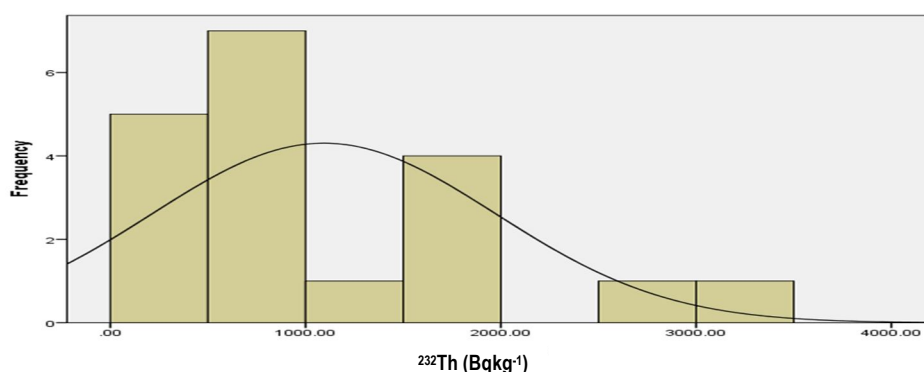


FIG. 9. Frequency distribution of activity concentration of ^{232}Th of laboratory-based gamma spectrometry in Abeokuta.

Conclusion

The activity concentrations of primordial radionuclides have been measured using both the laboratory-based and *in-situ* gamma spectrometry methods. The ranges of the activity concentrations for ^{40}K , ^{226}Ra and ^{232}Th have been found to be 113 – 1975, 5 – 128 and 181 – 3284 Bqkg⁻¹, respectively for laboratory-based gamma spectrometry and 104 – 1312, 31 – 121 and 104 – 2578 Bqkg⁻¹, respectively for *in-situ* gamma spectrometry. This study determined that the average activity concentrations of the primordial radionuclides in Abeokuta were much higher than worldwide averages of 400, 35 and 30 Bqkg⁻¹ for ^{40}K , ^{226}Ra and ^{232}Th , respectively.

From both methods, thorium is seen to be the major contributor to environmental radioactivity of the city Abeokuta. This could be associated to the geological setting of the city. The two methods used in this work have proven to be able to allow for scientific hypothesis to be tested with sufficient statistical power. Good correlations were deduced between the activity concentration results obtained from laboratory-based and *in-situ* gamma spectrometry, which implies a significant relationship between the two methods used in the study area. The *in-situ* gamma spectrometry therefore, is not inferior to the laboratory-based gamma spectrometry.

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