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DETERMINATION OF ACTIVITY CONCENTRATION LEVEL OF ²²⁶RA, ⁴⁰K AND ²³²TH IN SOIL WITHIN IGABI LOCAL GOVERNMENT AREA OF KADUNA STATE, NIGERIA

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ABSTRACT

This paper is aimed at determining the levels of primordial radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in selected sampling sites within Igabi local government area of Kaduna state, Nigeria using Gamma Ray Spectrometry. Radioactivity levels of fifteen samples taken from seven different locations within Igabi local Government area of Kaduna state, Nigeria were measured by means of gamma-ray spectrometry with NaI(Tl) detector to determine activity concentration of 40K, 226Ra and 232Th. Results obtained indicate highest activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th from soil samples collected from NOU (502.4883 $Bqkg^{-1}$), BP1 $(759.0962 \, Bqkg^{-1})$ and EC2 $(127.5941 Bqkg^{-1})$. The mean activity of 40 K (237.4184 $Bqkg^{-1}$) was found to be lower than the world average of $420 Bqkg^{-1}$; that of ²³²Th $(86.2182 \, Bqkg^{-1})$ was found to be above the world average of $50~Bqkg^{-1}$; and that of 226 Ra $(128.3507~Bqkg^{-1})$ was found to be lower than the world average of $370 Bqkg^{-1}$...

Keywords: Radionuclides, soil samples, radioactivity level, activity concentration, Gamma Ray spectrometry

INTRODUCTION

The earth and all living things on it are continuously bombarded by radiation from space which is akin to a steady light rain. Furthermore, the earth crust has always contained radionuclides with long half-lives, such as \$^{40}\$K, \$^{238}\$U, and \$^{232}\$Th, and as a result of their activity, these radionuclides cause natural radiation. The human environment is composed largely of soil, water and gases. Man uses soil or land for various purposes ranging from citing of industries, agriculture and erecting permanent structures for dwelling purposes. Soil is a product of weathering and contains fossils, organic and inorganic matter, gases and physical contaminants called radionuclides or radioisotopes. Radionuclides occur naturally in the soil in the form of the Uranium and Thorium decay series (\$^{226}\$Ra and \$^{232}\$Th) and natural potassium \$^{40}\$K (UNSCEAR, 1993).

Soil has been one of the major sources of radiation exposure to humans and animals alike, through the transfer of radionuclides into the environment, consequently determining the activity levels of these radionuclides in soil, waters, local vegetation and air of an area can help to determine the natural radioactivity in that area (Ahmad *et al.*, 2015).

Natural environmental radioactivity and the associated external exposure, as a result of gamma radiation, depend primarily on the geological and geographical conditions and appear at different levels in the soil samples of any region of the world. The specific

levels of terrestrial environmental radiation are related to the composition of each lithological-separated area, not to mention to the content of the rock from which the soil has originated. There are many types of soils, depending on their physical and chemical composition (UNSCEAR, 2000).

All living organisms including man are constantly exposed to varying degrees of ionizing radiation from naturally occurring radioactive materials (NORMs) in the environment and radionuclides generated by human activities which is called technologically enhanced naturally occurring radioactive materials (TENORM). Many artificial sources of radiation have been introduced since the discovery of X-rays and radioactivity at the end of the nineteenth century, and particularly since the exploitation of the process of nuclear fission in the middle of the twentieth century (Alan et al., 2012).

Exposure to ionizing radiation originates from two major sources namely naturally occurring and manmade sources. Naturally occurring radioactivity present on the earth's crust can be further classified into two distinct categories such as virgin and modified natural sources. Virgin sources of radiation are of cosmogenic or primordial (terrestrial) origin and have existed on the earth since primordial times. Modified natural sources are mainly from activities like mining, usage of fossil fuel, production of fertilizers or usage of natural materials for building constructions. Natural radiation is the largest contributor to the collective world radiation dose rate (Ramachandran, 2011; Alan et al., 2012). The major contribution of high dose from natural radiation in normal background regions arise due to inhalation of radon, a naturally occurring gas which emanates from rock and soil and its progeny (UNSCEAR, 2000). The activity concentrations of these Primordial radionuclides vary from one location to another and the distribution has been found to be largely dependent on geological and geographical conditions, and appear at different levels in the soils of each region of the world (UNSCEAR, 1993). Numerous artificial sources of radiation have been introduced since the discovery of X-rays and radioactivity at the end of the nineteenth century, and particularly since the exploitation of the process of nuclear fission in the middle of the twentieth century (Alan et al., 2012). Furthermore, radionuclides enter the environment during nuclear weapon testing, nuclear accidents, medical and industrial radiation applications. These artificial sources now add a significant contribution to the total radiation exposure of the population (Avwiri et al., 2014).

Several studies have been carried out to assess the activity concentrations of radionuclides in soils/ sediments within and outside Nigeria. These include natural radioactivity concentrations

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and dose assessment in shore sediments along the coast of greater Accra, Ghana (Amekudzie et al., 2011), measurement of natural radioactivity levels in soil along the bank of river Kaduna, Nigeria (Abdullahi et al., 2013), natural radioactivity levels of Australian building materials, industrial wastes and by-products (Beretka and Mathew, 1985), natural radioactivity levels in environmental samples in North Western Desert of Egypt (El-Daly and Hussein, 2008), measurement of activity concentration of soil samples collected from various areas of Riyadh city (Alaamer, 2008), determination of the activity concentrations of naturally occurring radioactivity of soil samples collected from selected cities across Ondo and Ekiti States (Ayodele et al., 2017); determination of radioactivity levels of naturally occurring radionuclides within Chikun local government area, Kaduna State, Nigeria (Gyuk et al., 2017), determination of radioactivity levels of naturally occurring radionuclides within Kaduna South Local Government Area of Kaduna state, Nigeria (Sarki et al., 2017) and determination of activity concentration within Kaduna North Local Government Area of Kaduna State, Nigeria (Sarki et al., 2019a; Sarki et al., 2019b).

This research work is aimed at determining the levels of primordial radionuclides $^{226}\text{Ra},~^{232}\text{Th},~\text{and}~^{40}\text{K}$ in selected sampling sites within Igabi local government area of Kaduna state, Nigeria using a $76\times76~mm~NaI$ detector crystal device used primarily for monitoring, rapid identification and assessment of radioactivity and absorbed dose rates.

MATERIALS AND METHODS

In this research Rados meter (RDS 120) was used for background mapping. The Rados is a versatile gamma radiation detector designed for wide range of applications involving the detection of abnormal or elevated radiation levels. It could also be used to determine background radiation of a place. Its performance and its user-friendly interface make it a perfectly suitable device for monitoring and detection of radiological hazards.

Monitoring of any release of radioactivity to the environment is important for environmental protection against ionizing radiation. Rapid and accurate methods for the measurements are essential. Many important isotopes in the NORM and TENORM have some suitable gamma rays, allowing qualitative and quantitative determination of the radio nuclides by high-resolution gamma spectrometry. Measurements of radiation levels and the concentrations of radio nuclides in the environment are accomplished employing appropriate nuclear instruments. Then radioactivity levels were detected and analyzed employing 76×76mm NaI (TI) detector crystal optically coupled to a photomultiplier tube (PMT). To determine each of the radioactivity level of these three radio nuclides, there is spectral energy window and energy calibration as presented in tables 1 and 2.

Study area

Igabi Local Government Area (LGA) of Kaduna State, Nigeria, is one of 774 local government areas (LGAs) in Nigeria, and one of the four that constitute Kaduna metropolitan city. It was created in 1989 out of Zaria local government, with headquarters in Turunku. The local government is made up of three districts namely: Igabi, Rigachikun and Rigasa (Igabi, 2020a). According to the 2006 National Census figures, Igabi has a population of 430,753 people, (NPC, 2006). The local government is dominated by farmers who produce crops on commercial level, with yams,

maize, guinea corn, beans and sugar cane being the major crops. Igabi Local Government is located on latitude 10° 47' 0"N and longitude 7° 46' 0"E (Igabi, 2020b). Annual rainfall is between 250 mm - 1000 mm and usually begins early May and ends in October, with the dry season from October to April (Ishola and Olukotun, 2019).

Sample collection

A total of fifteen (15) soil samples were collected from seven different locations namely: Bye Pass (BP1-3), Eye Center (EC1-3), Mando (MD1-3), Mando Road (MR1-3), National Open University (NOU), National Teachers Institute (NTI) and Rigasa (RGC) respectively. At each of the designated locations, soil samples were collected from a depth of $10,30,60\ cm$ using a hand trowel. In each study region, $1\ mm^2$ sampling site was covered, with each site located $1\ m$ straight from the other, except for NOU, NTI and RGC where each has only one sample site. About $150\ g$ of soil samples were collected from each location, packaged in cellophane bag and labeled for proper identification. Figure 1, presents a Google map of the sample locations within Igabi Local Government Area, Kaduna State, Nigeria.



Fig.1. Google map of the sample locations within Igabi Local Government Area, Kaduna State, Nigeria

Soil Sample Preparation

Each of the soil samples collected was dried and crushed to a fine powder with the use of pulveriser at Centre for Energy Research and Training, Zaria. The packaging of the samples into radon-impermeable cylindrical plastic containers which were selected based on the space allocation of the detector vessel which measures $76 \times 76 \ mm$ in dimension was also carried out to prevent ^{222}Ra from escaping; the packaging in each case was triple sealed.

The sealing process includes smearing of the inner rim of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container, and tight-sealing lid-container with masking adhesive tape. After the samples were prepared, the empty containers were weighed to know the weight of the empty container and when the sample was sealed into the container it was then weighed again to know the

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weight of empty container and soil sample, then the weight of empty container was subtracted from the weight of empty container and sample to get the weight of soil samples. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days at ambient temperature prior to gamma spectroscopy measurements.

Sample analysis

The major nuclear technique employed in the analysis of samples for background activity is the Gamma Spectrometry using a $76 \times 76 \ mm \ NaI(TI)$ detector (Ibrahim *et al.*, 2013). The assembly has a preamble incorporated into it and a 1 kilovolt external source. The detector is enclosed in a 6cm lead shield with cadmium and copper sheets. This arrangement is aimed at minimizing the effects of background and scattered radiation.

Calibration and Efficiency Determinations

Calibration of the system for energy and efficiency were done using two calibration point source; $\mathit{Cs}-137$ and $\mathit{Co}-60$. These were done with the amplifier gain that gives 72% energy resolution for $66.16\,keV$ of $\mathit{Cs}-137$ and counted for 30 minutes.

The standards used to check for the calibration are the IAEA gamma spectrometric reference materials RGK-1 for K-40, RGU-1 for Ra-226 (bi-214 peak) and RTG -1 for Th-232 (Ti-208).

Background measurement

Background radiation is a measure of the level of ionizing radiation present in the environment at a particular location which is not due to deliberate introduction of radiation sources. Generally, the background count rate is not constant but keeps fluctuating. This is because radioactive decay is a random process. The background count rate was done for 29,000 seconds. Table I shows the spectral energy window used in the analysis, while the energy calibration for quantitative spectral analysis is given in Table 2.

Table 1. Spectral energy window used in the analysis

Isotope	Gamma Energy (KeV)	Energy Window (KeV)
Ra – 226	1764.000	1620 – 1820
Th – 232	2614.500	2480 - 2820
K – 40	1460	1380 - 1550

Table 2. Energy calibration for quantitative spectral analysis

Isotope	Calibration	Factors	Conversion Factors (<i>Bqkg</i> -1)	Detection Limits		
	ppm	ppm	ppm	ppm	Bqkg ⁻¹	
K – 40	0.026	6.431	0.032	454.54	14.54	
Ra - 226	10.500	8.632	12.200	0.32	3.84	
Th – 232	3.612	8.768	4.120	2.27	9.08	

Spectra Analysis

The computer based multi-channel analyzer system with emulsion software (MAESTRO-32) was used for spectra acquisition. Based on two-point energy calibration as set for the operation, the prominent peaks were identified in a bench-mark spectrum and the appropriate regions of interest were set up. These peaks which are characteristic of typical environment spectral are:

- (a) The 295,352,607,1120 and 1765 *keV* peaks in the Uranium series.
- (b) The 238, 510, 583, 911, 965 and 2615 keV peaks in the Thorium series,
- (c) The 1460 keV peak of potassium.

The set energy bands define the peaks of where the left and right channel markers are representative of the Compton continuum. Detector's specific calibration factors (efficiency) were applied to convert from net count rate to concentration. Only peaks with reasonable $\gamma - ray$ emission probability were considered.

Calculation of Activity Concentration

Following spectrum analysis, calculation of count rates for each detected photo peaks and radiological concentration (activities per unit mass or specific activities) of detected radio nuclides depends on the establishment of secular equilibrium reached between ²³²Th, ²³⁸U and their decay products. The concentration of ²³²Th was determined from the average concentration of ²⁰⁸TI and ²³⁸Ac and that of ²³⁸U was determined from the average concentration ²¹⁴Pb and ²¹⁴BI decay products. Thus, accurate radionuclide concentrations of ²³²Th and ²³⁸U were determined, whereas a true measurement of ⁴⁰K concentration was made.

The specific activity $(Bqkg^{-1})A_{Ei}$ of a nuclide and for a peak at energy E, is given by

$$A_{Ei} = \frac{N_{Ei}}{\epsilon_E t \gamma_d M_S} \tag{1}$$

Where N_{Ei} is the net peak area of a peak energy E, ϵ_E is the detection efficiency at energy E, t is the counting live-time, γ_d is the gamma ray yield per disintegration of specific nuclide for a transition of energy E and M_S the mass in kg of the measured sample (Gyuk et al., 2017).

RESULTS AND DISCUSSION

The radioactivity level in twelve (15) soil samples from, Bye Pass (BP1-3), Eye Center (EC1-3), Mando (MD1-3), Mando Road (MDR1-3), National Open University (NOU), National Teachers Institute (NTI) and Rigasa (RGC) within Igabi local government area of Kaduna metropolis were measured by means of gammaray spectrometry with $NaI\ (TI)$ detector to establish a data for activity concentration $^{226}{\rm Ra},~^{232}{\rm Th},~{\rm and}~^{40}{\rm K}.$ Table 3 presents results of background radiation reading at the fifteen (15) sites.

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Table 3. Background radiation reading of Igabi local government area

S/N0	Sample ID	RDS 120 SERIAL NO. 990100 (μSV/hr)			MEAN OF EXPOSURE RATE	RDS 120 SERIAL NO. 210152 (μSV/hr)			MEAN OF EXPOSURE RATE	MEAN OF EXPOSURE RATE	
		1	2	3	(μSV/hr)	1	2	3	(μSV/hr)	(μSV/hr)	
1	BP1	0.11	0.11	0.11	0.11 ± 0.006	0.11	0.11	0.11	0.11 ± 0.011	0.11 ± 0.004	
2	BP2	0.12	0.13	0.13	0.13 ± 0.13	0.12	0.12	0.13	0.12 ± 0.008	0.12 ± 0.011	
3	BP3	0.11	0.12	0.12	0.12 ± 0.006	0.12	0.12	0.13	0.12 ± 0.016	0.12 ± 0.011	
4	EC1	0.12	0.12	0.13	0.12 ± 0.008	0.12	0.12	0.12	0.12 ± 0.008	0.13 ± 0.008	
5	EC2	0.11	0.11	0.12	0.11 ± 0.016	0.12	0.11	0.12	0.12 ± 0.008	0.12 ± 0.011	
6	EC3	0.11	0.11	0.11	0.11 ± 0.013	0.11	0.11	0.12	0.11 ± 0.006	0.11 ± 0.010	
7	MD1	0.12	0.12	0.13	0.12 ± 0.013	0.12	0.12	0.13	0.12 ± 0.006	0.12 ± 0.010	
8	MD2	0.11	0.11	0.11	0.11 ± 0.013	0.11	0.11	0.11	0.11 ± 0.006	0.11 ± 0.010	
9	MD3	0.11	0.12	0.12	0.12 ± 0.013	0.11	0.11	0.12	0.11 ± 0.006	0.11 ± 0.010	
10	MR1	0.11	0.11	0.11	0.11 ± 0.008	0.11	0.11	0.12	0.11 ± 0.013	0.11 ± 0.011	
11	MR2	0.12	0.12	0.13	0.12 ± 0.006	0.12	0.12	0.12	0.12 ± 0.016	0.12 ± 0.011	
12	MR3	0.12	0.12	0.13	0.12 ± 0.016	0.11	0.12	0.12	0.12 ± 0.008	0.12 ± 0.024	
13	NOU	0.12	0.12	0.12	0.12 ± 0.010	0.11	0.11	0.11	0.11 ± 0.013	0.12 ± 0.012	
14	NTI	0.12	0.12	0.12	0.12 ± 0.008	0.11	0.11	0.12	0.11 ± 0.013	0.12 ± 0.011	
15	RGC	0.11	0.11	0.11	0.11 ± 0.010	0.11	0.11	0.11	0.11 ± 0.010	0.11 ± 0.010	

The highest activity concentration of 40 K, 226 Ra and 232 Th were found in soil collected from NOU (502.4883 $Bqkg^{-1}$), BP1 (759.0962 $Bqkg^{-1}$) and EC2 (127.5941 $Bqkg^{-1}$). The mean activity of 40 K (237.4184 $Bqkg^{-1}$) was found to be lower than the world average of 420 $Bqkg^{-1}$. Similarly, the

mean activity concentration of 232 Th $(86.2182 \, Bqkg^{-1})$ was found to be above the world average of $50 \, Bqkg^{-1}$, while the mean activity concentration of 226 Ra $(128.3507 \, Bqkg^{-1})$ was also found to be lower than the world average of $370 \, Bqkg^{-1}$

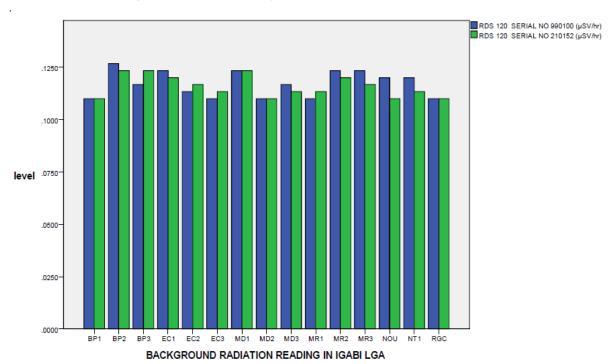


Figure 1. Background radiation reading for Igabi Local Government Area

Figure 1 is the graphical representation of the result from background radiation readings and gamma spectrometry analysis obtained from soil samples in Igabi Local Government Area. Here BP (1-3), EC (1-3), MD (1-3), MR (1-3), NOU, NTI and RGC

represents Bye Pass, Eye Center, Mando, Mando Road, National Open University, National Teachers Institute and Rigasa respectively, while the numbers 1, 2 and 3 are the three locations where the soil samples were taken for each of the towns.

Table 4. Gamma ray spectrometry reading

S/N0	Sample	K-40	Error ±	K-40	Error ±	Ra-226	Error ±	Ra-226	Error ±	Th-232	Error ±	Th-232	Error ±
	ID	(CPS)	(CPS)	(Bqkg ⁻¹)	(Bqkg ⁻¹)	(CPS)	(CPS)	(Bqkg ⁻¹)	(Bqkg ⁻¹)	(CPS)	(CPS)	(Bqkg ⁻¹)	(Bqkg ⁻¹)
1	BP1	0.5721	0.0043	426.6421	6.4768	0.0931	0.0002	759.0962	5.1024	0.0212	0.0018	16.2214	1.2431
2	BP2	0.4936	0.0040	228.7593	5.6812	0.0423	0.0003	27.6625	1.2312	0.0289	0.0012	106.8821	2.1764
3	BP3	0.5024	0.0031	363.4173	5.7041	0.0331	0.0001	690.5169	3.3674	0.0236	0.0015	58.6723	3.1642
4	EC1	0.0475	0.0006	73.8725	0.9331	0.0252	0.0026	29.2005	3.0127	0.0826	0.0031	94.1847	3.5348
5	EC2	0.0614	0.0012	95.4899	1.8663	0.0078	0.0042	9.0382	4.8667	0.1119	0.0038	127.5941	4.3330
6	EC3	0.0421	0.0007	65.4743	1.0886	0.0091	0.0050	10.5446	5.7937	0.0942	0.0025	107.4116	2.8506
7	MD1	0.2123	0.0018	330.1711	2.7994	0.0471	0.0027	54.5771	3.1286	0.0853	0.0031	97.2634	3.5348
8	MD2	0.1424	0.0046	221.4619	7.1540	0.0329	0.0042	38.1228	4.8667	0.1011	0.0030	115.2794	3.4208
9	MD3	0.1541	0.0025	239.6579	3.8880	0.0329	0.0029	38.1228	3.3604	0.0931	0.0007	106.1574	0.7982
10	MR1	0.1404	0.0007	218.3515	1.0886	0.0207	0.0012	23.9861	1.3905	0.0581	0.0027	66.2292	3.0787
11	MR2	0.1338	0.0055	208.0871	8.5537	0.0232	0.0052	26.8830	6.0255	0.0765	0.0076	87.2292	8.6659
12	MR3	0.2131	0.0028	331.4152	4.3546	0.0643	0.0036	74.5075	4.1715	0.1109	0.0043	126.4538	4.9031
13	NOU	0.3231	0.0062	502.4883	9.6423	0.0721	0.0003	83.5458	0.3476	0.0922	0.0031	105.1311	3.5348
14	NTI	0.1285	0.0038	199.8445	5.9098	0.0471	0.0029	54.5771	3.3604	0.0487	0.0030	55.5302	3.4208
15	RG	0.0361	0.0004	56.1431	0.6221	0.0080	0.0010	9.2700	1.1587	0.0012	0.0012	23.0331	1.3683

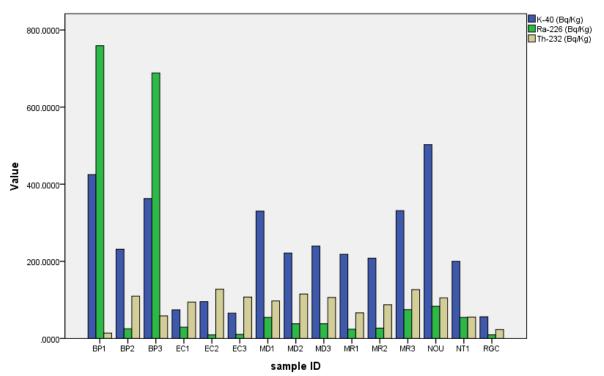


Figure 2. Results of gamma ray spectrometry for 226ra, 232th, and 40k done on fifteen (15) soil samples within Igabi L.G.A

Figure 2 shows the chart result obtained from Gamma spectrometry done in Centre for Energy Research and Training, Zaria, on the soil samples taken from Igabi L.G.A. The results show that \$^{226}\$Ra is more abundant among the three radio-nuclides and is peaked at BP1 and minimum at EC2, EC3 and RGC. \$^{40}\$K is the second with its peak at NOU, and it is minimum at RGC and EC3, while \$^{232}\$Th is the least among the three Radio nuclides and it peaked at EC2 with a minimum at BP1. Thus, Figures 1 and 2

show that BP1 and BP2 have the maximum background readings and it also shows that among the three radio nuclides $^{\rm 226}{\rm Ra}\,$ is maximum.

Conclusion

Radioactivity levels of fifteen samples taken from seven different locations within Igabi local Government area of Kaduna state, Nigeria have been measured by means of gamma-ray

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spectrometry with NaI(Tl) detector to determine activity concentration of 40 K. 226 Ra and 232 Th.

Measured results indicate highest activity concentration of $^{40}\mathrm{K}, ^{226}\mathrm{Ra}$ and $^{232}\mathrm{Th}$ in soil collected from NOU (502.4883 $Bqkg^{-1})$, BP1 (759.0962 $Bqkg^{-1})$ and EC2 (127.5941 $Bqkg^{-1})$, respectively. The mean activity concentration of $^{40}\mathrm{K}$ (237.4184 $Bqkg^{-1})$ was found to be lower than the world average of 420 $Bqkg^{-1}$. The mean activity concentration of $^{232}\mathrm{Th}$ (86.2182 $Bqkg^{-1})$ was found to be above the world average of 50 $Bqkg^{-1}$, and that of $^{226}\mathrm{Ra}$ (128.3507 $Bqkg^{-1})$ was found to be lower than the world average of 370 $Bqkg^{-1}$.

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