

Determination of Radioactivity Levels of ^{222}Ra , ^{40}K and ^{232}Th using Gamma Ray Spectrometry in Kaduna South Local Government Area of Kaduna State, Nigeria

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ABSTRACT: A total of twelve (12) soil samples were collected from four different locations in Kaduna South Local Government Area of Kaduna state, Nigeria; the radioactivity level in these samples were detected and analysed by means of a gamma-ray spectrometer with $76 \times 76\text{mmNaI}$ (TI) detector crystal. The average activity of ^{226}Ra , ^{232}Th and ^{40}K were found to be 38.0263Bq/Kg , 82.3071Bq/Kg and 163.5174Bq/Kg respectively. The mean activity of ^{226}Ra (38.0263Bq/Kg) was found to be lower than the world average of 400Bq/Kg , while the mean activity concentration of ^{232}Th (82.3071Bq/Kg) and ^{40}K (163.5174Bq/Kg) were found to be higher than the world average of 50Bq/Kg and lower than the world average of 420Bq/Kg respectively.

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

I. INTRODUCTION

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. The latter is known as Technologically Enhanced Natural Radiation (TENR). Natural radiation is the largest contributor to the collective world radiation dose rate. The major contribution of high dose from natural radiation in normal background regions arise due to inhalation of radon and its progeny [1].

Gamma radiation from radionuclides, such as ^{40}K and ^{232}Th series, and their decay products, represents the main external source of irradiation to the human body. The absorbed dose rate in air from cosmic radiation outdoor at sea level is about 30nGy h^{-1} [1]. External exposures to gamma radiation outdoors arise from terrestrial radionuclides occurring at trace levels in all rock formations. Therefore, the natural environmental radiation mainly depends on geological and geographical conditions [2]. These have a direct effect by modifying the soil composition and natural radioactivity concentration levels; and hence the level of γ –absorbed dose received at a locality. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shale and phosphate rocks have relatively high content of radionuclides [1].

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

II. MATERIALS AND METHODS

In this research, Rados meter was used (RDS 120) 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 the perfectly suitable devices for monitoring land detections of radiological hazards.

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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 naturally occurring radioactive materials(NORM) and technology enhance natural occurring radioactive materials(TENORM) have some suitable gamma rays, allowing qualitative and quantitative determination of the radionuclides by high-resolution gamma spectrometry. Measurements of radiation levels and the concentrations of radionuclides in the environment are accomplished employing appropriate nuclear instruments. The radioactivity level was detected and analysed employing $76 \times 76\text{mmNaI}$ (TI) detector crystal optically coupled to a photomultiplier tube (PMT) to determine the radioactivity level of each of the three radionuclides having spectral energy window and energy calibration.

A. Sample collection

A total of twelve soil samples were collected from four different towns namely, Kudendan, Nassarawa, Kakuri and Barnawa within Kaduna South local Government area, Kaduna state, Nigeria, with three points selected from each town.

Kaduna South local government area occupies the southern part of the state capital and lies within latitudes $10^{\circ}26'N$ and $10^{\circ}32'N$ of the equator and on longitudes $7^{\circ}24'E$ and $7^{\circ}28'E$ of the Greenwich meridian. With an area of 59km^2 and a population of 402,390 as at the 2006 census, it falls into the tropical continental climate with seasonal rainfall pattern of high intensities, with distinct wet and dry seasons. Mean annual temperature is about 24°C and mean annual rainfall is about 1,185mm [3]. Kaduna South local government area is the industrial hub of Kaduna state and boasts of industries like Defence Industries Corporation of Nigeria (DICN), Peugeot Automobile company of Nigeria (PAN), Kaduna Refinery and Petro-Chemical Company of Nigeria (KRPCN), United Nigeria Textile Limited, Nigerian Brewery Limited, Nigerian Bottling Company Ltd, NOCACO Ltd, Arewa Pharmaceuticals Ltd and Dino Agricultural and Dairies Ltd to mention but a few [4].



Figure 1 Google map of the study areas

Figure 1 depicts the Google map of the study area showing Kudendan, Nassarawa, Kakuri and Barnawa within Kaduna South local Government area, Kaduna state, Nigeria where the twelve (12) soil samples were collected.

B. Soil Sample Preparation

Each of the soil samples collected was dried and crushed to a fine powder with the use of pulveriser. 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 $76 \times 76\text{mm}$ in dimension (geometry) 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 container 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 weight of empty container and soil sample, then the weight of empty container and sample was subtracted from the weight of empty container 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.

C. Evaluation of Radioactivity of Samples

The majornuclear technique employed in the analysis of samples for background activityis the Gamma Spectrometry using a 76 × 76mmNal (TI)detector[5]. The assembly has a preamble incorporated into it and a 1kilovolt 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.

D. Dose calculation

To assess any radiological hazard, the absorbed dose rate ($nGyh^{-1}$) in air at 1m above the ground surface due to the mean specific activities of ^{40}K , ^{226}Ra , and $^{232}Th(Bqkg^{-1})$ in the collected samples, can be computed using,

$$D(nGyh^{-1}) = DCF_k C_k + DCF_{Ra} C_{Ra} + DCF_{Th} C_{Th} \tag{1}$$

Where D is the air absorbed dose rate, $DCF_k(0.0417)$, $DCF_{Ra}(0.462)$ and $DCF_{Th}(0.604)$ are the dose rate conversation factors and C_k , C_{Ra} and C_{Th} are the concentration of ^{40}K , ^{226}Ra , and ^{232}Th in the respective materials[5].

E. Calibration and Efficiency Determinations

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

F. Background measurement

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 29000 seconds (10hrs 50mins). Table I shows the spectral energy window used in the analysis in the subsequent section, while the energy calibration for quantitative spectral analysis is given in Table II.

TABLE I. 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	1460.000	1380 – 1550

TABLE II. Energy Calibration for quantitative Spectral Analysis.

Isotope	Calibration Factors		Conversion Factors(BqKg ⁻¹)	Detection Limits	
	10 ⁻³ (cps/ppm)	10 ⁻⁴ (cps/ppm)	Conversion Factor (BqKg ⁻¹)	ppm	Bq/Kg
⁴⁰ K	0.026	6.431	0.032	454.54	14.54
²²⁶ Ra	10.500	8.632	12.200	0.32	3.84
²³² Th	3.612	8.768	4.120	2.27	9.08

G. Spectra Analysis

The computer based multi-channel analyser 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 γ -ray emission probability were considered.

H. 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 ^{232}Th , ^{238}U and their decay products. The concentration of ^{238}Th was determined from the average concentration of ^{208}Tl and ^{238}Ac and that of ^{238}U was determined from the average concentration ^{214}Pb and ^{214}Bi decay products. Thus, accurate radionuclide concentration of ^{232}Th and ^{238}U were determined, whereas a true measurement of ^{40}K concentration was made.

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

$$A_{Ei} = \frac{N_{Ei}}{\epsilon_E t \gamma_d M_s} \tag{2}$$

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. For more than one peak in the energy range analysis, the peak activities are averaged and the result is weighted average nuclide activity.

The total uncertainty (σ_{tot}) and weighted systematic activity value is composed of counting statistic (σ_{st}) and weighted systematic errors (σ_{sys}) calculated by the following formula

$$\sigma_{tot} = \sqrt{\sigma_{st}^2} + \frac{1}{3} \sum \sigma_{sys}^2 \tag{3}$$

[6]

III. RESULTS AND DISCUSSIONS

The result of radioactivity level in twelve (12) soil samples from Kudendan, Nassarawa, Kakuri and Barnawa within Kaduna South local government area of Kaduna metropolis was measured by means of gamma-ray spectrometry with NaI (TI) detector to establish a data for activity concentration ^{40}K , ^{226}Ra and ^{232}Th . Table III presents results of background radiation reading at the twelve (12) different sites.

TABLE III. Background Radiation Reading

S/N	Location	RDS120 Serial NO. 990100 ($\mu SV/hr$)			Mean of Exposure Rate ($\mu SV/hr$)	Mean of Exposure Rate ($\mu SV/hr$)			RDS 120 Serial NO. 210152 ($\mu SV/hr$)	Mean of Mean ($\mu SV/hr$)
1	KDD1	0.12	0.13	0.13	0.13 ± 0.013	0.12	0.14	0.14	0.14 ± 0.006	0.14 ± 0.010
2	KDD2	0.12	0.12	0.13	0.12 ± 0.006	0.11	0.12	0.14	0.12 ± 0.001	0.12 ± 0.008
3	KDD3	0.11	0.11	0.12	0.11 ± 0.013	0.12	0.13	0.13	0.13 ± 0.008	0.12 ± 0.011
4	NAS1	0.11	0.11	0.11	0.11 ± 0.006	0.11	0.11	0.11	0.11 ± 0.010	0.11 ± 0.008
5	NAS2	0.13	0.13	0.14	0.13 ± 0.010	0.11	0.11	0.11	0.11 ± 0.010	0.12 ± 0.010
6	NAS3	0.11	0.11	0.12	0.11 ± 0.006	0.12	0.12	0.12	0.11 ± 0.008	0.11 ± 0.006
7	KR1	0.13	0.13	0.14	0.13 ± 0.008	0.11	0.12	0.12	0.12 ± 0.008	0.13 ± 0.008
8	KR2	0.11	0.11	0.11	0.11 ± 0.016	0.11	0.11	0.12	0.11 ± 0.013	0.11 ± 0.015
9	KR3	0.13	0.12	0.15	0.13 ± 0.013	0.13	0.13	0.13	0.13 ± 0.006	0.13 ± 0.010
10	BN1	0.11	0.11	0.11	0.11 ± 0.010	0.12	0.12	0.13	0.12 ± 0.006	0.12 ± 0.008
11	BN2	0.11	0.11	0.12	0.11 ± 0.026	0.12	0.12	0.13	0.12 ± 0.006	0.12 ± 0.016
12	BN3	0.14	0.14	0.15	0.14 ± 0.016	0.12	0.12	0.13	0.12 ± 0.013	0.13 ± 0.015

The highest activity concentration of ^{40}K , ^{226}Ra and ^{232}Th were found in soil collected from location NAS3 (380.7154 Bq/kg), NAS1 (61.2978 Bq/kg) and BN1 (205.9293 Bq/kg) respectively, where the mean activity of ^{40}K (163.5174 Bq/kg) is lower than the world average of 420 Bq/kg and the mean activity concentrations of ^{232}Th (82.3071 Bq/kg) is above the world average of 50 Bq/kg, ^{226}Ra have the lowest mean activity concentration of 38.0263 Bq/kg, which is lower than the world average of 370 Bq/kg.

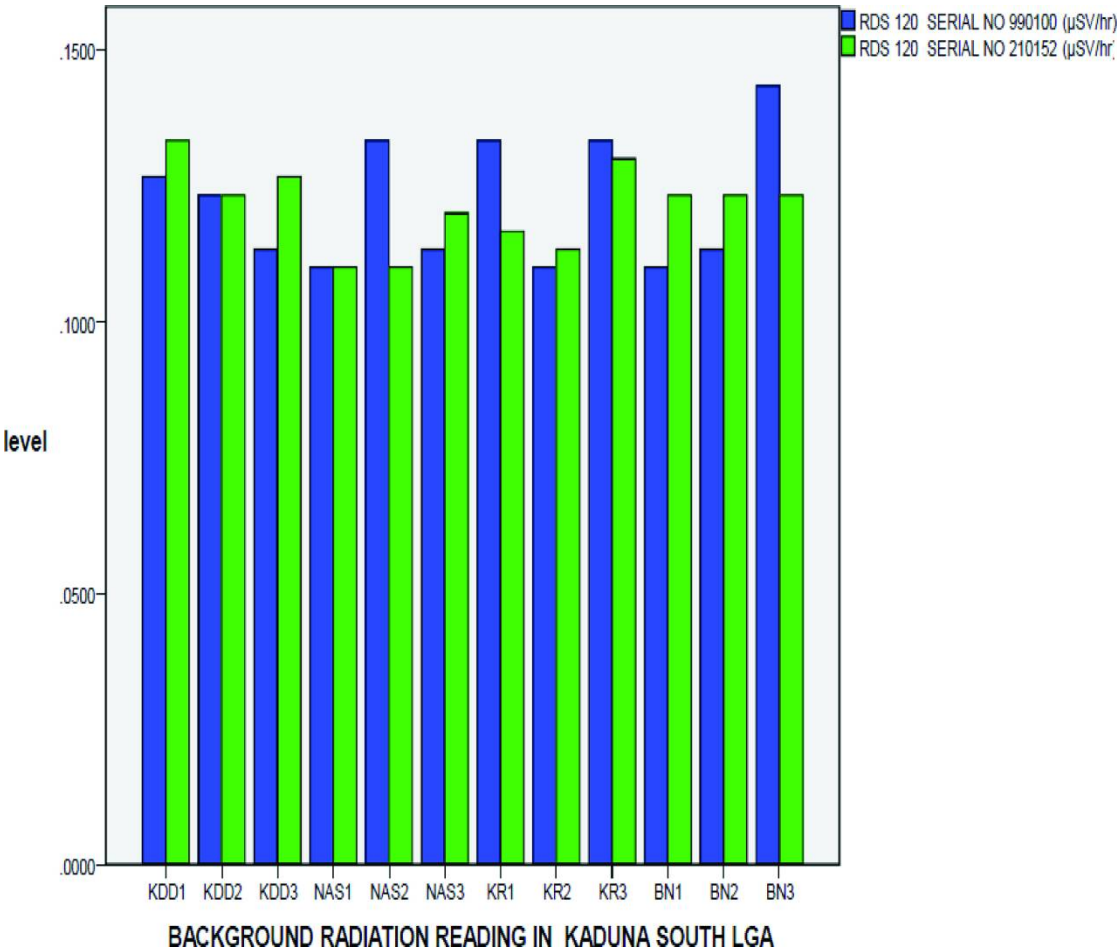


Figure 1The Background Radiation reading at Kaduna South L.G.A.

Figure 2 is the graphical representation of the result from background radiation readings and gamma spectrometry analysis obtained from soil samples in Kaduna South L.G.A. Here KDD (1-3), NAS (1-3), KR (1-3) and BN (1-3) represents Kudendan, Nassarawa, Kakuri and Barnawa respectively, while the numbers 1, 2 and 3 are the three locations where the soil samples were taken for each of the towns.

The background radiation reading obtained in Kaduna South L.G.A as presented in Fig.2 shows a minimum background reading at NAS₁ (0.11 ± 0.008) µSv/hr while BN₃ (0.13 ± 0.015) µSv/hr has the maximum Background reading.

TABLE IV. Gamma Ray Spectrometry Reading

S/N0	Sample ID	K-40	Error ±	K-40	Error ±	Ra-226	Error ±	Ra-226	Error ±	Th-232	Error ±	Th-232	Error ±
		(CPS)	(CPS)	Bq/kg	Bq/kg	(CPS)	(CPS)	Bq/kg	Bq/kg	(CPS)	(CPS)	Bq/kg	Bq/kg
1	KDD1	0.0631	0.0036	98.1337	5.5988	0.0346	0.0047	40.0927	5.4461	0.0594	0.0068	67.7309	7.7537
2	KDD2	0.0534	0.0029	83.0482	4.5101	0.0269	0.0035	31.1703	4.0556	0.0449	0.006	51.1973	6.8415
3	KDD3	0.0903	0.0003	140.435	0.4666	0.0223	0.0039	25.8401	4.5191	0.0817	0.0025	93.1585	2.8506
4	NAS1	0.1206	0.0016	187.5583	2.4883	0.0529	0.005	61.2978	5.7937	0.0813	0.0023	92.7024	2.6226
5	NAS2	0.2118	0.0003	329.3935	0.466	0.0413	0.0029	47.8563	3.3604	0.0982	0.0028	111.9726	3.1927
6	NAS3	0.2448	0.0028	380.7154	4.3546	0.0457	0.0038	52.9548	4.4032	0.1072	0.0031	122.2349	3.5348
7	KR1	0.771	0.0012	119.9067	1.8663	0.0025	0.0007	2.8969	0.8111	0.0109	0.0025	12.4287	2.8506
8	KR2	0.351	0.0004	54.5879	0.6221	0.0348	0.0019	40.3244	2.2016	0.0413	0.0012	47.0924	1.3683
9	KR3	0.1373	0.0004	213.5303	0.6221	0.0288	0.0025	33.3720	2.8969	0.0651	0.002	74.2303	2.2805
10	BN1	0.0436	0.0018	67.8072	2.7994	0.0526	0.0042	60.9502	4.8667	0.1806	0.0047	205.9293	5.3592
11	BN2	0.0712	0.0027	110.7309	4.1991	0.0482	0.0035	55.8517	4.0556	0.0731	0.0008	83.3523	0.9122
12	BN3	0.1134	0.0032	176.3608	4.9767	0.0032	0.0062	3.7080	7.1842	0.0225	0.0013	25.6556	1.4823

Table IV presents the result of the Gamma Ray Spectrometry measurements carried out on the twelve (12) soil samples from four (4) different cities within Kaduna South L.G.A., which is used in the plot given by Figure 3.

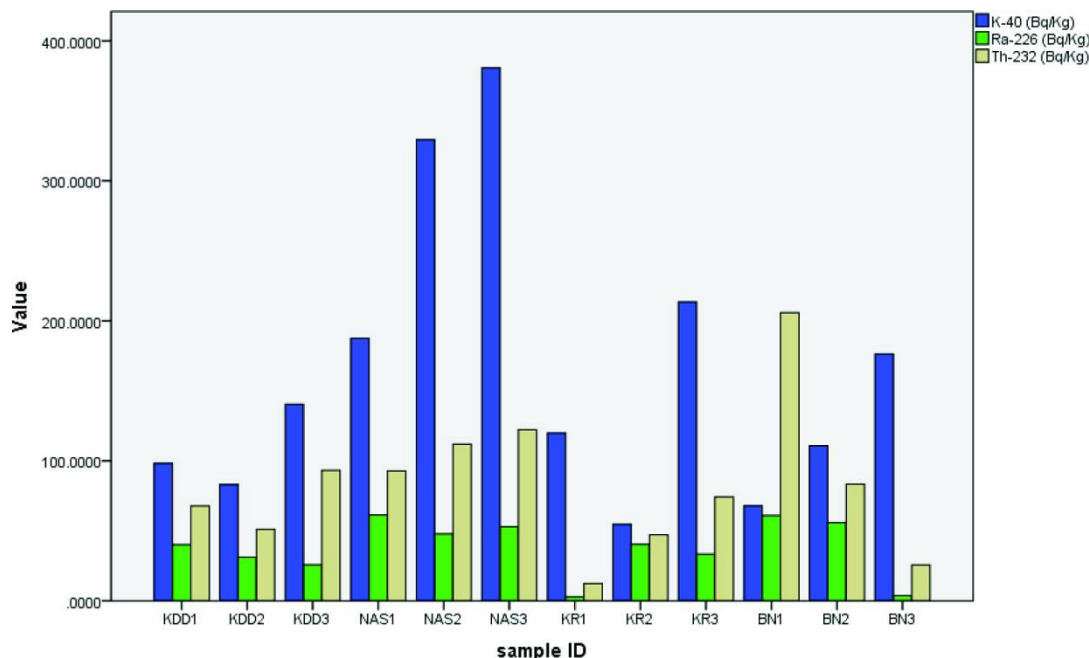


Figure 2Result of Gamma Ray Spectrometryfor⁴⁰K,²²⁶Ra, and ²³²Th done on twelve (12) soil Samples within Kaduna South L.G.A

Here, Figure 3is a representation of the result obtained from Gamma spectrometry done in Centre for Energy Research and Training (CERT) Zaria, on the twelve soil samples obtained from four different locations in Kaduna South local Government area. From the results obtained, the activity concentration of ⁴⁰Kis higher than the activity concentration of theother two radionuclides and is peaked at $NAS_3(380.7154BqKg^{-1})$ and minimum at $KR_2(54.5879BqKg^{-1})$. ²³²This is the second with its peak at $BN_1(205.9293 BqKg^{-1})$ and it is minimum at $KR_1(12.4287BqKg - 1)$, while²²⁶Ra is the least among the three radionuclides with its peak at $NAS_1(61.2978 BqKg^{-1})$ and minimum at $KR_1(2.8969BqKg^{-1})$.

IV. CONCLUSION

Radioactivity levels of twelve samples taken from four different towns within Kaduna South local Government area of Kaduna state, Nigeria has been measured by means of gamma-ray spectrometry with *NaI* (TI) detector to determine activity concentration ⁴⁰K, ²²⁶Ra and ²³²Th.

From the obtained results, the average exposure is 0.15mSv/yr and average activity of ²²⁶Ra, ²³²Thand⁴⁰K were found to be 38.0263Bq/Kg, 82.3071 Bq/Kg and 163.5174Bq/Kg respectively. The mean activity of ²²⁶Ra (38.0263Bq/Kg) isestablished to be lower than the world average of400Bq/Kg, while the mean activity concentration of (82.3071Bq/Kg) is seen to be higher than the world average of 50Bq/Kg and the activity concentration of ⁴⁰K (163.5174Bq/Kg) lower than the world averageof420 Bq/Kg.

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