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DETERMINATION OF RADIOACTIVITY LEVELS IN SOIL SAMPLES AT CHIKUN ENVIRONMENT OF KADUNA METROPOLIS USING GAMMA RAY SPECTROMETRY

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ABSTRACT
Fourteen (14) soil samples were collected in Chikun Local Government Area of Kaduna metropolis. The radio activity level in samples were detected and analysed by employing 76×76mm NaI (TI) detector crystal. The average activity of ^{226}Ra, ^{232}Th and ^{40}K were found to be 62.2827Bq/kg, 155.3592 Bq/kg and 459.5646 Bq/kg, respectively. The mean activity of ^{226}Rais lower than the world average of 400Bq/Kg, while those of^{232}Th and ^{40}K are higher than the world average of 50Bq/Kg and 420 Bq/kg, respectively.

Keywords: Radionuclides; soil samples, activity concentration, Chikun Area, Kaduna Metropolis.

INTRODUCTION

Exposure to ionizing radiation originates from two major sources, namely naturally occurring and man-made 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 (UNSCEAR, 2000).

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 30 nGyh^{-1} (UNSCEAR, 2000). External exposures to gamma radiation outdoors arise from terrestrial radio nuclides occurring at trace levels in all rock formations. Therefore, the natural environmental radiation mainly depends on geological and geographical condition (Nagaraja and Sathish, 2010). These have 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 radio nuclides (Ramachandra *et al*, 1995).

Ionizing radiation has always been part of the human environment. Along with natural radioactive sources present in the earth crust and cosmic radiation. Man's discovery of atomic and nuclear energy with advance in technology, the uses of man produced radiation, especially in the production of elastic power are being continuously multiplied, so are man's chances of exposure to harmful radiation (Agbalaje, 2010).

This research work determines the levels of the primordial radionuclides ^{40}K, ^{232}Th and ^{238}U in the selected sampling sites using a 76x76mm NaI (TI) detector crystal, primarily for monitoring rapid identification and assessment of radioactivity and absorbed dose rates in air.

MATERIALS AND METHOD

In this research Rados meter (RDS 120) was usedfor 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 makes it a perfectly suitable devicesor monitoring land detections 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 natural occurring radioactive materials(NORM) and technology enhance natural occurring radioactive materials(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 level were detected and analysed employing 76×76mm NaI (TI) detector crystal optically coupled to a photomultiplier tube (PMT), to determine each ofthe radioactivity level of these three radio nuclides, there is spectral energy window and energy calibration as presented in table 1 and 2.

Soil Sample Preparation

Each of the soil samples collected were dried and crushed to fine powder with the use of pulveriser. Packaging of the samples into radon-impermeable cylindrical plastic containers which were selected based on the space allocation of the detector vessel which measures76×76mm in dimension (geometry) was also carried out to prevent ^{222}Ra from escaping; the packaging in each case was triple sealed.

The sealing process include 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 then when the sample was sealed into the container it was then weighed again to know the weight of empty container and soil sample, then to get the weight of sample you are to subtract the weight of the empty container from the weight of container + sample. 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.

Gamma Ray Spectrometry

Gamma spectrometry offers a convenient, direct and non-destructive method for the measurement of the activity of different radionuclides in environmental samples.Gamma spectrometry techniques offer high efficiency (NaI (Tl) detectors) and high resolution semiconductors detection. The technique enables the use of large quantity of samples for counting.

Liquid nitrogen cooling is required only when the detectors are in use. Hence, gamma ray spectrometer was employed in the present work to determine the activity concentration of radio nuclides in environmental soil samples.

Evaluation of Radioactivity of Samples

The analysis was carried out using a 76×76mm NaI (Tl)detector crystal optically coupled to a photomultiplier tube (PMT). 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.

The data acquisition is maestro software by Canberra nuclear products. The samples were measured for a period of 29000 seconds (10hrs 50mins) for each sample. The peak area of each energy level in the spectrum was used to compute the activity concentrations in each sample by the use of equation (1) (Ibeanu, 1999).

$$c(Bq.kg^{-1}) = \frac{Cn}{C_{fk}} \qquad (1)$$

Where C is the activity concentration of the radio nuclides in the sample given in $Bqkg^{-1}$

C_{fk} is Calibration factor of the detecting system.

C_n is count rate (counts per second)

$$Counts\ per\ second\ (cps) = \frac{netcount}{livetime} \qquad (2)$$

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 the 66.16KeV of Cs- 137 and counted for 30 mins.

Backgrounds Readings

The background count rate was done for 29000 seconds (10hrs 50mins) and Table 1shows the spectral energy window used in the analysis.

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	*1460.000*	*1380 – 1550*

Table 2 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

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:

- The 295, 352, 607, 1120 and 1765 keV peaks in the Uranium series,
- The238, 510, 583, 911, 965 and 2615 keV peaks in the Thorium series, and
- 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.

Calculation of Activities 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 was reached between ^{238}U and their decay products. The ^{232}Th was determined from the average concentration of ^{204}Tl and ^{227}Ac, and that of ^{238}U was determined from the average concentration of ^{214}Pb and ^{208}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

$$A_{Ei} = \frac{N_{Ei}}{\epsilon_E \, t\gamma_d} \qquad (3)$$

Where N_{Ei} is the net peak area of a peak energy E
ϵ_E is the detection efficiency at energy N
t is the counting live-time
γ_d is the gamma ray yield per disintegration of specific nuclide for a transition of energy E

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 (Paramesh *et al*., 2003)

$$\sigma_{tot} = \sqrt{\sigma_{st}{}^2 + \frac{1}{3}\sum \sigma_{sys}{}^2} \qquad (4)$$

RESULTS AND DISCUSSION

The results of radio activity levels in Fourteen (14) soil samples within Chikun Local Government Area of Kaduna metropolis were determined. Samples contain a number of radio nuclides originating from both natural and man-made sources. Distribution of radio nuclides varies from place to place that is because of the different activities within the sample area. Activity concentration of some of the radio nuclides such as ^{226}Ra, ^{232}Th and ^{40}K were determined by direct counting methods employing the nuclear instruments.

The results obtained are presented in Table 3. From the tables there was no significant difference of the result from the survey meter and the result obtained from Centre for Energy Research and Training (CER) Zaria.

Fig.1 is the graphical representation of the result from background radiation readings and gamma spectrometry analysis obtained from soil samples in Chikun L.G.A.

Table 3: Result of Background Radiation Reading at Chikun L.G.A

S/N	EXAMPLE	RDS120 SERIAL NO 990100 (μSV/hr)			MEAN OF EXPOSURE RATE (μSV/hr)	RDS 120 SERIAL NO 210152 (μSV/hr)			MEAN OF EXPOSURE RATE (μSV/hr)	MEAN OF MEAN (μSV/hr)
	Code	1	2	3		1	2	3		
1	KP1	0.11	0.11	0.12	0.11±0.006	0.12	0.12	0.12	0.12±0.006	0.12±0.006
2	KP2	0.14	0.14	0.14	0.14±0.008	0.11	0.14	0.2	0.15±0.008	0.15±0.008
3	KP3	0.12	0.15	0.12	0.13±0.006	0.13	0.14	0.15	0.14 ±0.01	0.14±0.008
4	SBR1	0.11	0.12	0.13	0.12±0.008	0.11	0.11	0.13	0.12±0.013	0.12±0.011
5	SBR2	0.12	0.12	0.14	0.13±0.13	0.11	0.11	0.11	0.11±0.008	0.12±0.011
6	SBR3	0.11	0.11	0.11	0.11± 0.006	0.11	0.11	0.11	0.11±0.006	0.11±0.006
7	GN1	0.14	0.14	0.15	0.14± 0.016	0.13	0.13	0.15	0.14±0.008	0.14±0.012
8	GN2	0.11	0.12	0.12	0.12±0.010	0.14	0.14	0.14	0.14±0.013	0.14±0.012
9	GN3	0.13	0.14	0.14	0.14±0.008	0.12	0.12	0.14	0.13±0.008	0.14±0.008
10	RIDO1	0.12	0.13	0.15	0.13±0.010	0.15	0.16	0.18	0.16±0.013	0.15±0.012
11	RIDO2	0.11	0.11	0.11	0.11±0.006	0.1	0.11	0.11	0.11±0.006	0.11±0.006
12	BNH2	0.12	0.13	0.14	0.13 ±0.008	0.11	0.12	0.13	012±0.000	0.13±0.008
13	KRPC1	0.18	0.19	0.18	0.18±0.008	0.19	0.20	0.20	0.20±0.008	0.19±0.008
14	KRPC2	0.16	0.16	0.17	0.16±0.010	0.18	0.17	0.18	0.18±0.010	0.17±0.010

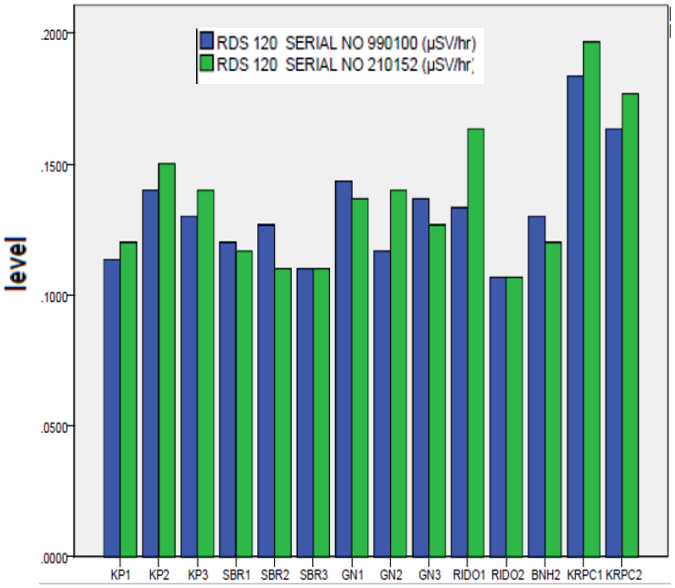

Fig 1: The Background Radiation reading at Chikun L.G.A.

Fig.1 is the chart for the background radiation reading obtained in Chikun L.G.A. It shows a minimum background reading at SBR_3 (0.110 ± 0.006) μSv/hr and Rido2 (0.11 ± 0.006) μSv/hr while $KRPC_1$ (0.190 ± 0.008) μSv/hr as the maximum background reading.

Table 4: The Result Obtain from Soil Samples in Chikun L.G.A

S/No	Sample ID	K-40 (CPS)	Error ± (CPS)	K-40 (Bq/Kg)	Error ± (Bq/Kg)	Ra-226 (CPS)	Error ± (CPS)	Ra-226 (Bq/Kg)	Error ± (Bq/Kg)	Th-232 (CPS)	Error ± (CPS)	Th-232 (Bq/Kg)	Error ± (Bq/Kg)
1	KP1	0.1468	0.0009	228.3048	1.3997	0.0243	0.0007	28.1576	0.8111	0.0253	0.0005	28.8483	0.5701
2	KP2	0.3328	0.0016	517.5739	2.4883	0.0169	0.0053	19.5829	6.1414	0.0445	0.0026	50.7412	2.9647
3	KP3	0.0786	0.0059	122.2395	9.1757	0.0772	0.0066	89.4554	7.6477	0.1294	0.0043	147.5485	4.9031
4	SBR1	0.0189	0.0025	29.3935	3.8880	0.0417	0.0025	48.3198	2.8969	0.0931	0.0003	106.1574	0.3421
5	SBR2	0.0981	0.0062	152.5661	9.6423	0.0285	0.0004	33.0243	0.4635	0.0922	0.0041	105.1311	4.6750
6	SBR3	0.2431	0.0042	378.0715	6.5319	0.0312	0.0036	36.1530	4.1715	0.0332	0.0024	37.8563	2.7366
7	GN1	0.0762	0.0051	118.5070	7.9316	0.0631	0.0028	73.1170	3.2445	0.0194	0.0016	22.1209	1.8244
8	GN2	0.1131	0.0007	175.8942	1.0886	0.0519	0.0018	60.1390	2.0857	0.0212	0.0021	24.1733	2.3945
9	GN3	0.0932	0.0021	144.9456	3.2659	0.0152	0.0016	17.6130	1.8540	0.0743	0.0005	84.7206	0.5701
10	RIDO1	0.0405	0.0001	62.9860	0.1555	0.0349	0.0034	40.4403	3.9397	0.0945	0.0031	107.7537	3.5348
11	RIDO2	0.0625	0.0003	97.2006	0.4666	0.0138	0.0011	15.9907	1.2746	0.022	0.0009	25.0855	1.0262
12	RIDO3	0.1357	0.0032	211.0420	4.9767	0.0304	0.0028	35.2260	3.2445	0.1173	0.0036	133.7514	4.1049
13	KRPC1	0.5721	0.0043	889.7356	6.6874	0.0937	0.0003	108.5747	0.3476	0.2132	0.0019	243.1015	2.1665
14	KRPC2	0.4921	0.0031	765.3188	4.8212	0.0895	0.0029	103.7080	3.3604	0.2531	0.0012	288.5975	1.3683

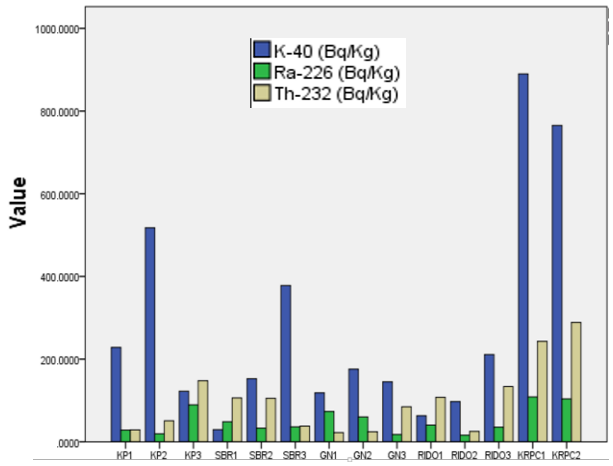

Fig 2: Chart showing Result for ^{40}k, ^{226}Ra and ^{232}Th.

Fig. 2 is the chart showing the result obtained from Gamma spectrometry done in Centre for Energy Research and Training (CERT) Zaria, on the soil samples. Thus, the result shows that K-40 is more abundant among the three radio nuclides and is peaked at $KRPC_1$(889.7356Bqkg^{-1}) and $KRPC_2$ (765.3188Bqkg^{-1})and is minimum at SBR_1.(29.3935Bqkg^{-1}) Th-232 is the second most, which is peaked at $KRPC_2$(288.5975Bqkg^{-1})and is minimum at GN_1.(22.1209Bqkg^{-1}) Ra-226 is the least among the three Radio nuclides and also the minimum is at $RIDO_2$ (15.9907Bqkg^{-1}).

CONCLUSION

When exposed to radiation the body absorbs some dose which causes chemical reactions to occur which may alter the normal function of the body. From the obtained result, the average exposure is 0.15mSv and average activity of ^{226}Ra, ^{232}Th and ^{40}K were found to be 62.2827Bqkg^{-1}, 155.3592 Bqkg^{-1} and 459.5646Bqkg^{-1}, respectively. Though the mean activity of ^{226}Ra (62.2827Bqkg^{-1}) is lower than the world average of 400Bqkg^{-1}, mean activity concentration of^{232}Th (155.3592Bqkg^{-1}) is higher than the world average of 50Bq/kg and the activity concentration of ^{40}K (459.5646 Bqkg^{-1}) is also higher than the world average of420 Bqkg^{-1}.

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