An Evaluation of ^{238}U, ^{40}K, and ^{232}Th Concentrations in the Top Soil of the University of Ibadan (UI), Southwestern Nigeria.

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ABSTRACT

The radioactivity concentration of the natural radionuclides (^{40}K, ^{238}U and ^{232}Th) in the top soils of University of Ibadan (UI), Ibadan, Nigeria have been determined by gamma ray spectrometry. The activity concentrations of ^{40}K, ^{238}U, and ^{232}Th varied from 22.80 to 923.79, 5.59 to 92.30, and 5.00 to 154.25 Bq kg^{-1}, respectively, with mean values of 261.37 ± 192.17,50.01 ± 29.00, 84.66 ± 37.88 Bq kg^{-1}.

The mean absorbed dose rate recorded for ^{40}K, ^{238}U and ^{232}Th were 10.95 nGy h^{-1} for ^{40}K, 21.49 nGy h^{-1} for ^{238}U and 56.38 nGy h^{-1} for ^{232}Th. The mean total absorbed dose rate was found to be 88.92 nGy h^{-1} .

The outdoor annual effective dose equivalent ranged from 11.21 to 178.45 μSv y^{-1} with mean of 109.05 μSv y^{-1}. This is considered to be low when compared to natural external radiation of about 2000 μSv y^{-1} to which no harmful effect is expected directly.

(Keywords: naturally occurring radionuclides, gama ray spectrometry, potassium, uranium, thorium, natural radiation dose, dosage)

INTRODUCTION

Radioactivity is part of the human physical environment. The largest contribution (about 98%) to the radiation field is of natural origin and this is due to cosmic rays from outer space and radioactive elements in the Earth. Artificial man-made radioactivity is emitted by nuclear power plants, industrial plants, and research facilities. The emissions due to artificial radioactivity are very small in normal operations; however a large quantity of radioactivity can be released to the environment through accidents and dumping of radioactive waste in a particular environment. At times contribution of long-lived decay and activation products from nuclear weapon tests and/or nuclear facilities (mainly ^{137}Cs) add to the natural contribution. The artificial radiation at the surface varies with time due to decay and downward migration of cesium. Natural terrestrial radiation comes from the natural occurring radionuclide headed by ^{238}U and ^{232}Th, while others are in non-series type called ^{40}K. There have always been some seventy naturally radioactive isotopes such as radium and polonium in human skin and skeletons, radioactive potassium and carbon in muscles, as well as tritium and radioactive noble gases in human lungs (Olomo, 2006). It is estimated that in the US, the most important single source of the dose received by an average man is the radioactive gas radon (Rn-222) a decay product of uranium. However uranium is found in many common rocks, notably granite.

The high geochemical mobility of radionuclides in the environments allow them to move easily and to contaminate mainly the environment with which human come in contact. U-238, in particular is easily mobilized in ground water and surface water. As a result, uranium and its decay product enter the food chain through irrigation water, and enter the water supply through ground water, well and surface water streams and rivers (Otton, 1994). The ubiquitous gaseous ^{222}Rn formed from the decay of uranium enters the atmosphere through emanation from the soil and building materials (Okeyode, 2007). The problem of ^{222}Rn occurs when houses are built in uranium-rich regions, since it is impossible to prevent ^{222}Rn from entering such houses from the ground underneath such houses (Beizer, 1995). As a cause of lung cancer, ^{222}Rn is a second only to

cigarette smoking. Other minor contributions to radiation exposure come from non-series primordial radionuclides, primarily ^{87}Rb (NCRP,1987) and cosmogenic radionuclides produced in the atmospheric Ar, O_2 and N_2. These latter radionuclides (of cosmic origin) reach the earth through precipitation, scavenging, atmospheric mixing and gravitational setting.

Exposures from these results primarily from ingestion, but they are relatively constant throughout the world. The public concern for environmental quality necessitates the radiation monitoring program to identify all sources of radiation exposure within an environment. This enables assessment to be made of the radiation exposure of the members of the public living in the geographical location. Such assessment permits timely detection of changes in radiation parameter which may lead to increased exposures and to produce sufficient information on the causes and necessary steps to safeguard the health of the inhabitant of such location (NOHSC, 1995).

STUDY LOCATION

The University of Ibadan (popularly known as UI) was the first university in Nigeria established in 1948 and affiliated with the University of London at that time called University College London. The University has an estimated area of 10.47sq km. The university is located in Ibadan (the third largest cosmopolitan city in Africa and the capital city of Oyo state). UI has a mean elevation of 200meters above sea level and lies between Latitude $N7^0\ 26^1$ - $7^0\ 27^1$ and longitude East $3^0\ 53^1$-$3^0\ 54^1$. This university (UI) is located in the south-western part of Nigeria and Northern part of Ibadan along Oyo road.

The city of Ibadan itself is said to fall within the basement complex of the geological setting of south western Nigeria characterized mainly by the metamorphic rock types of Precambrian age, but with few intrusion granite and porphyries of Jurassic age (Jibiri, 2006). Generally 75% of the rocks in and around Ibadan are banded gneisses while granite gneisses and quartzite, share the remaining 25% of the area in almost equal halves (Oyewole, 1970). Rock quarrying in Ibadan is a growing economic activity in the last decade, this is attributable to growing urbanization, demand for housing and infrastructure, road construction and maintenance and availability of rocks in the surrounding.

In specific terms the main type of rock found within UI are gneiss, quartzites and magnetite which are of igneous and sedimentary origins. The basement rocks are covered by superficial deposits which vary in thickness with locations (Oresanya, 1984). The deposits are capped with laterite and clay topsoil. Igneous rock like granite has high concentration of uranium. Also the solubility of ^{222}Th in natural water such as river is detected in high concentration in sediments and deposits (Arogunjo, 1994). Moreover, ^{222}Th and ^{238}U are more abundant in sedimentary rocks than in igneous and metamorphosed sediments. Adequate knowledge of the soil in this location is useful in terms of formation of natural soil deposit and describing the source of the soil.

Presently, the focus of UI is manpower development in terms of postgraduate studies. The ratio of admission of undergraduates to postgraduates is about 2:3. There are several research projects going on in different departments. Among the research institutes that uses ionizing radiation is Federal Radiation Protection Service (FRPS) located in the Department of Physics (DOP) established in 1964 after a nuclear weapon was reportedly tested in the Sahara Desert which is close to the northern boundary of Nigeria (Agu, 1994). However, only the air medium was monitored on the roof of FRPS building (Olomo, 2006).

This (FRPS) metamorphosed into Nigerian Nuclear Regulatory Authority (NNRA) established by Decree 19 of 1995. Other research activities are carried out in the Departments of Chemistry, Pharmacy, Agriculture and Engineering. Movement of quarried rocks and granite into the campus has been on for more than fifty years due to continuous infrastructure development of UI.

The aim of this study is to assess the activity concentration of top soil within UI and to ascertain whether the activity level is within the acceptable level. This study stems from the fact that UI is fast turning to postgraduate research centre (especially area around the Physics, Chemistry, Pharmacy and Agriculture departments) and the likelihood of some of the activities leading to contamination of the environment cannot be downplayed.
The result of this study will provide a baseline of value of activity concentration for assessing

radioactive pollution of the environment in any case of future accidental radioactive release in UI.

MATERIALS AND METHODS

Sample Collection and Preparation: Soil samples were collected from thirty different locations at about 400m apart within University of Ibadan. This distribution is to allow even representation of the different positions of the UI campus. The samples were collected at a depth of about 10cm deep (topsoil) and packed in polythene bag of non radioactive material sealed and labeled L1-L30 for easy identification and to prevent mix-up. The samples were sun dried and kept sealed in a polythene bag and kept for some days to ensure that parent and daughter nuclei would be in a state of equilibrium before the assessment of the concentration was carried out.

The Counting Assembly and Counting Process: The Counting system used for this analysis of the natural radiological content of the soil samples consisted of scintillation detector or photomultiplier tube and a Canberra multichannel analyzer (MCA). The scintillation detector is connected to the multichannel analyzer (MCA). The MCA is a microprocessor used in spectroscopic analysis. The scintillation detector is a 76cm X 76cm NAI (TL) manufactured by Bicron. This is connected to the MCA through coaxial cables. The MCA consisted of an analog-to-digital converter (ADC), control logic (CL) with input and output devices, memory, display and analysis logic (DAC) unit with an on-screen Display (SD) unit. The MCA has an inbuilt high voltage power supply (HVPS), and for convenience, it has rechargeable batteries of Nickel Cadmium (for that can run for at least 6 hours, if there is power outage and counting will not be hindered) or it can be powered from a 12V automobile battery.

Initial Measurement and Calibration of Equipment: Each height of a given pulse and its corresponding channel number is directly proportional to the gamma energy producing it. This forms the basis of our measurement and calculation. The system was tested for its linearity and calibrated with standard source samples potassium, Thorium and Uranium (K_2SO_4 IAEA/RGK-1, Th-ore IAEA/RGTH-1). These were Geological certified Reference material for Radiometric Measurement from International Atomic Energy (IAEA), Vienna. Equal quantity of these standard sources of known energy were mixed together, place inside the gamma-ray spectroscopy and counted for 7200sec (2hrs).

The calibration of known energy of the source used to determine the linear equation relating the gamma energy, E, to the channel number as shown in Figure 1. The linearity (R^2= 0.9928) of the system used for this work was observed by plotting the gap energy of the known source agent from different sources for calibration used to get a straight line for the energy channel calibration which can be fitted with Equation 1. This can be fitted with the Equation 1:

$$E = K n + \xi \qquad \textbf{(1)}$$

where ξ and K are constants (intercept and slope of the graph. Using the system, the curved fitting was carried out and the equation obtain is given below (Equation 2)

$$E(MeV) = 0.0084n - 0.0655 \qquad \textbf{(2)}$$

where E is the energy and n the channel number, the Equation 2 was stored in the memory of the operational system and it remained as the operational setting of the system through out the experimental observation. Three regions of interest were created for the purpose of this work. Region one was photo peak corresponding to gamma energy 1.465Mev of ^{40}K, region two was photo peak of energy 1.765Mev for ^{238}U, while region three was photopeak with channel number 310 of energy 2.615Mev for ^{232}Th.The energy peaks were obtained from region of interest (ROI) created in the neighborhood of observed energies.

Background Shielding and Counting Process: For effective and reliability of the measurement in gamma spectroscopy, effective shielding against the natural radiation background has been accomplished in the study by building a lead shield castle around the source–detector combination (Farai, 1988).The sample being measured also provide additional shielding from the residual radiation emanating from the lead castle itself. The lead castle has dimension of 5cm x 10 cm x 20 cm lead block arranged such that the thickness is 5 cm.

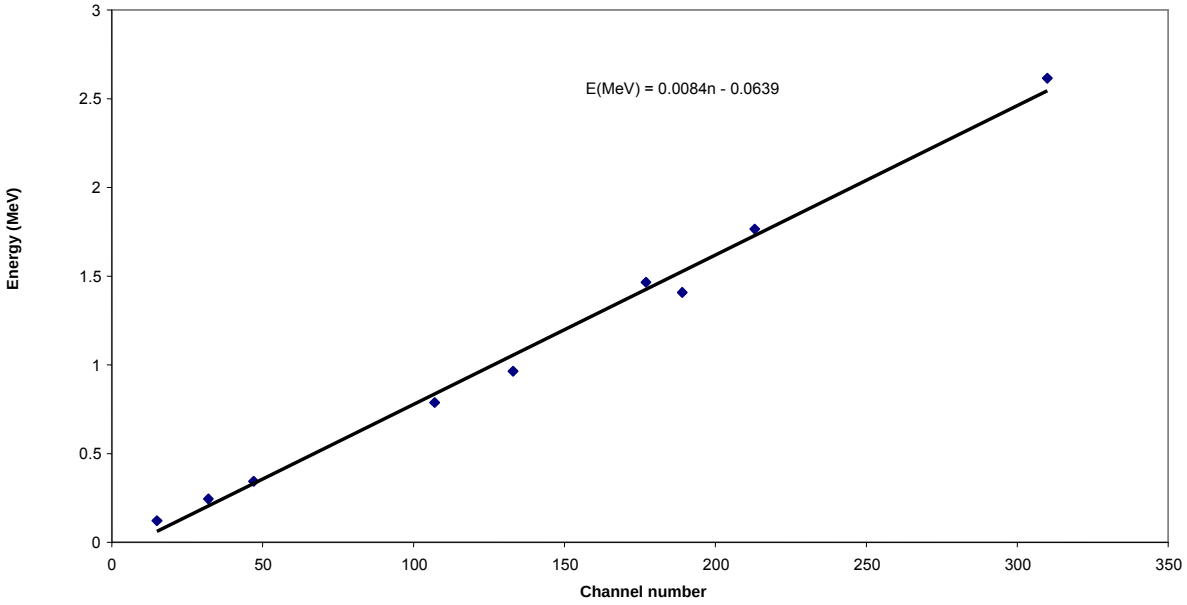

Figure1: Energy (MeV) against Channel Number.

The counting assembly described earlier was employed for the counting procedure. Necessary adjustment were carried out on the MCA, for example the high voltage (1000 V) settings, gain and memory settings, time settings (7200 seconds). The region of interest (ROI) for each radionuclide observed was defined with the aid of corresponding channel number in which the peak occurs. After 7200 seconds of counting the area under each photopeak under the region of interest computed and presented by MCA for each of the 30 soil sample.

RESULTS

Table 1 is the area under the photopeak due to the background radiation. The area under the photopeak due to standard sample used for the calibration is displayed in Table 2.

Table 3 is the result of the area under the photopeak due to standard sample plus the background radiation. And Table 4 is the result of the activity concentration (Bq kg^{-1}) of the standard source. The mean and coefficient of variation of the activity concentration (Bq kg^{-1}) of the radionuclides present in the soil at different sample location and the gamma absorbed dose rate are presented in Table 5.

Specific Activity Concentration and Absorbed Dose Rate Determination: The area under the photopeak counted for each radionuclide by the MCA is a measure of the activity concentration of the radionuclide in the soil. In this study, a fixed geometry was maintained for all the samples measured and since the sensitivity of the Canberra system used was constant, the activity concentration is proportional to the net area under the photopeak (Okeyode and Farai, 2007),

Therefore: $C = kA$ **(3)**

$$k = \frac{C}{A} \quad \textbf{(4)}$$

where *C* is the concentration and *A* is the net area, *k* is multiplicative factor which is constant for each radionuclide at constant geometry; *k* is determined by the detector efficiency of the assembly. The calibration factors for the three radionuclides were calculated using a standard soil sample (Geological Certified Reference Material for Radiometric Measurement) obtained from International Atomic Energy Agency, Vienna using equation (3).

Table 1: Area Under the Photopeak due to Background Radiation.

^{40}K	^{238}U	^{232}Th
987± 30.04	86 ±204.48	537±25.11

Table 2: Area Under the Photopeak due to the Standard Sample.

^{40}K	^{238}U	^{232}Th
1992± 355.71	24414 ±647.18	203±81.20

Table 3: Area Under the Photopeak of the Standard Sample plus the Background Radiation.

^{40}K	^{238}U	^{232}Th
2979± 15.96	2500 ± 13.10	776±26.5

Table 4: Activity Concentration (Bq/kg) of the Standard Source.

^{40}K	^{238}U	^{232}Th
510	631	232

Table 5: Mean and Coefficient of Variation of the Activity Concentration of the Radionuclides and Gamma Absorbed Dose Rate.

Sample location	Mean activity concentration (Bq/kg)	Coefficient of variation	Mean absorbed dose rate (nGy/h)	Coefficient of variation
1	19.23	0.41	3.05	0.94
2	219.38	0.02	27.00	0.01
3	178.62	0.02	38.19	0.08
4	232.25	0.01	46.04	0.02
5	178.62	0.02	26.73	0.01
6	356.00	0.01	42.17	0.02
7	61.58	0.06	23.26	0.03
8	185.58	0.02	48.50	0.002
9	80.15	0.05	22.24	0.02
10	67.01	0.10	3.58	0.57
11	102.25	0.03	17.19	0.01
12	109.99	0.03	40.86	0.01
13	87.76	0.14	28.86	0.02
14	115.99	0.02	27.49	0.01
15	100.13	0.03	39.36	0.01
16	223.13	0.01	29.60	0.01
17	235.05	0.01	46.81	0.001
18	141.29	0.01	41.89	0.01
19	129.15	0.02	29.49	0.01
20	129.59	0.03	26.67	0.01
21	46.94	0.09	22.64	0.18
22	152.74	0.02	44.98	0.01
23	134.61	0.08	24.86	0.01
24	122.96	0.04	25.21	0.03
25	140.08	0.02	22.56	0.001
26	84.88	0.14	16.52	0.04
27	74.42	0.05	24.75	0.02
28	54.75	0.21	19.72	0.03
29	106.05	0.03	37.55	0.01
30	90.62	0.04	35.15	0.01

The calibration factors were found to be:

k (^{40}K) = 0.278 Bq kg^{-1}/cps
k (^{238}U) = 0.266 Bq kg^{-1}/cps
k (^{232}Th) = 0.250 Bq kg^{-1}/cps

The calibration factor; k (^{40}K), k (^{238}U), k (^{232}Th) were multiplied by the area under the photopeak to get the corresponding activity concentration of ^{40}K, ^{238}U, ^{232}Th for the different sample location. The result obtained is given in Table 5.

Moreover, the absorbed dose rates were calculated using equation (4) according to Beck, *et al*, (1972) and Okeyode and Farai, (2007).

$$\check{D}_{air}=0.042S_k+0.42S_u+0.666S_{Th} \quad \textbf{(5)}$$

where D_{air} is the total absorbed dose rate in air in nGy h^{-1} , and S_k, S_u, S_{Th}, are the activity concentration of potassium, uranium, and thorium. The result of $\check{D}_{air}$ obtained for each sample location using Equation (4) is given in table 5.Additionally, the total absorbed dose rate were converted into outdoor annual effective dose equivalent, E_{air} using 0.2 as the outdoor occupancy factor, Equation (5) (Jibiri and Okorie, 2006) was used to determine the outdoor human effective dose equivalent to the population.

$$E_{air}=TQ\check{D}_{air}\varepsilon \quad \textbf{(6)}$$

where E_{air} is the annual effective dose rate (μSvy^{1}), T is the time being 8760 hy^{-1}, Q is the quotient of the effective dose rate and absorbed dose rate in air (0.7SvGy^{-1}), ε is the conversion factor. The result obtained and the total absorbed dose rate is displayed in Table 6.

DISCUSSION

The radionuclide analysis of soil sample carried out within the University of Ibadan (UI) campus revealed that the basic composition of the radioactive element of soils in UI include: ^{40}K, ^{238}U and ^{232}Th .

Figure 2 shows the absorbed dose rate for the three radionuclides found in the soil of UI. The absorbed dose rate of ^{40}K for various sample locations were relatively low, it ranged between 0.96±5.65 and 38.80 ± 1.44 nGy h^{-1} while that of ^{238}U is in the range of 3.88 ± 8.32 and 44.96 ± 1.05 nGy h^{-1}. However, Figure 4 shows that the absorbed dose rate of ^{232}Th was relatively high with mean value of 56.38 ± 25.23 nGy h^{-1}. The highest absorbed dose rate was found in location 8(department of forestry fish farm river bank) with a value of 102.73 ±5.58 nGy h^{-1}.

Table 6: Annual Effective Dose Equivalent, E_{air} (μSv/y)

Sample location	Annual effective dose equivalent (μSv/y)
1	11.21
2	99.35
3	140.50
4	169.40
5	98.34
6	155.15
7	85.58
8	178.45
9	81.82
10	37.69
11	63.26
12	150.35
13	106.19
14	101.16
15	144.81
16	108.92
17	172.21
18	154.14
19	108.49
20	98.13
21	81.73
22	165.51
23	91.46
24	92.74
25	83.04
26	60.78
27	91.05
28	72.54
29	138.16
30	129.32

The result here shows uneven distribution of specific activity concentration of the radionuclides as indicated in Figure 3. The distribution of radionuclide could be attributed to the nature of the parent rock from which the soil was formed. Other factors that affect the distribution of radionuclide include soil utilization pattern and precipitation. Figure 3 revealed that ^{40}K has the highest specific activity concentration with mean

value of 261.34 (22.80-923.79) $Bqkg^{-1}$.

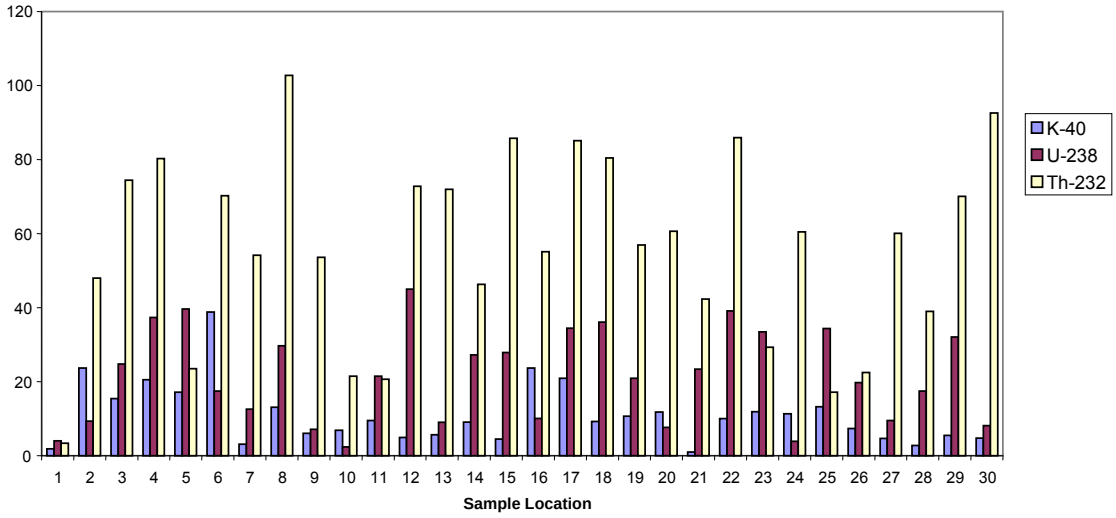

Figure 2: Absorbed Dose Rate Against Sample Location.

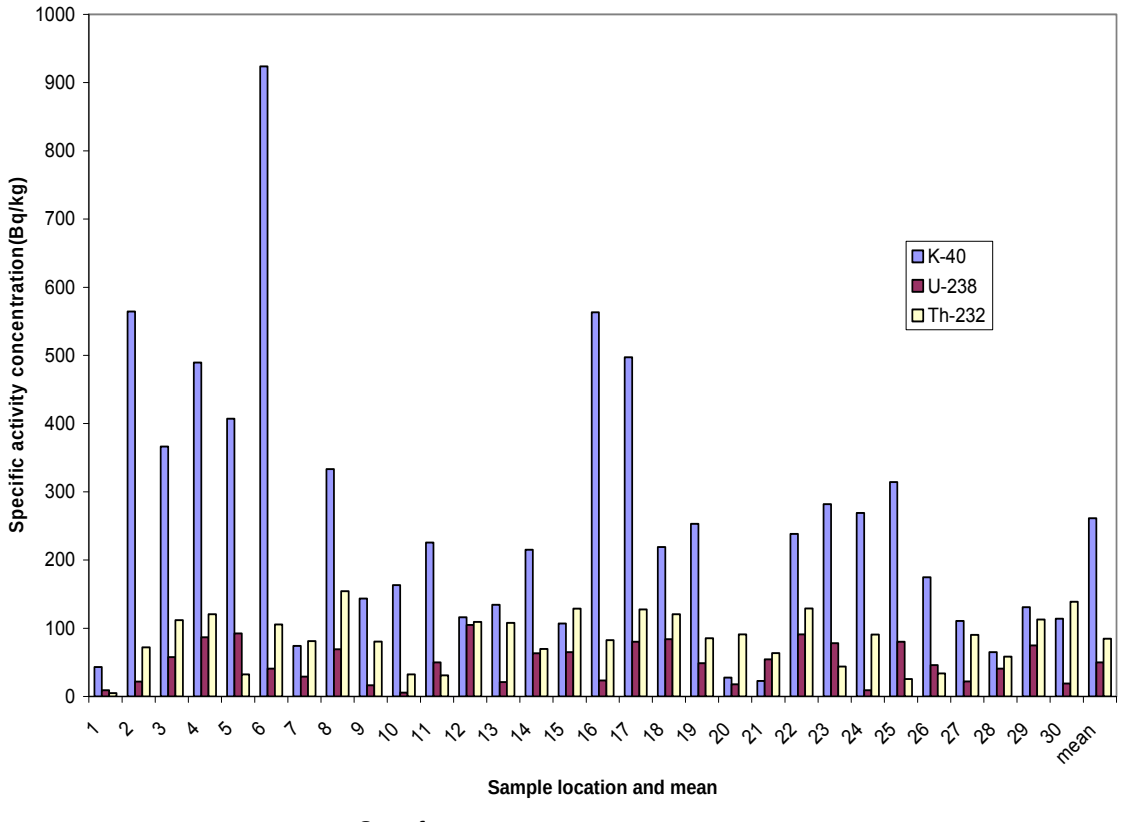

Figure 3: Specific Activity Concentration Against Sample Location.

The range factors (ratio of maximum individual specific activity concentration to the minimum specific activity for the same sample location) as indicated in Figure 5 for each of the radionuclide show that there were distinct variations in the distribution of the radionuclides in the soil.

It is worthy of note to state that the highest value of ^{238}U activity concentration was found to be 104.80 ± 1.04 Bqkg^{-1} at sample location 12 beside the Federal Environmental Protection Agency (FEPA UI linkage centre) within the premises of Chemistry Department. This location is not far from the Nigeria Nuclear Regulatory Agency (NNRA) .

In addition the highest total activity concentration recorded in this study was found to be 1069. 99 Bq kg$^{-1.}$ This was found in sample location 6 (the main entrance to the university). This is the busiest part of UI, here commercial activities are carried out. However, the lowest total activity concentration of 57.58 Bq kg^{-1} was found in location 1 (Department of forestry and fish farm) . The soils around this area are moist most part of the year. The study indicate that 12 sample locations have activity concentration above the mean value while 18 locations were below the mean value of 396.11 Bq kg^{-1}.

The range factor (displayed in Figure 5) indicates the variation in the distribution of the radionuclides in the soil. The variation in the distribution of the radionuclides is attributable to the varied rock types found at various sample locations within UI. The rock types with sedimentary origin are prone to high activity concentration as seen in this study. This stems from the fact that quartzites are made up of fine grains of silky clay and have natural potassium as its essential constituent (Arogunjo, 1994). Therefore there is possibility of having high activity concentration at a particular location and low values at another location.

Table 5 shows the mean and coefficient of variation of activity concentration and the mean absorbed dose rate. The range of mean values of activity concentration and absorbed dose rate obtained in this study for the 30 locations is 19.23-256.10 Bq kg^{-1} and 3.05-48.50 nGy h^{-1}, respectively.

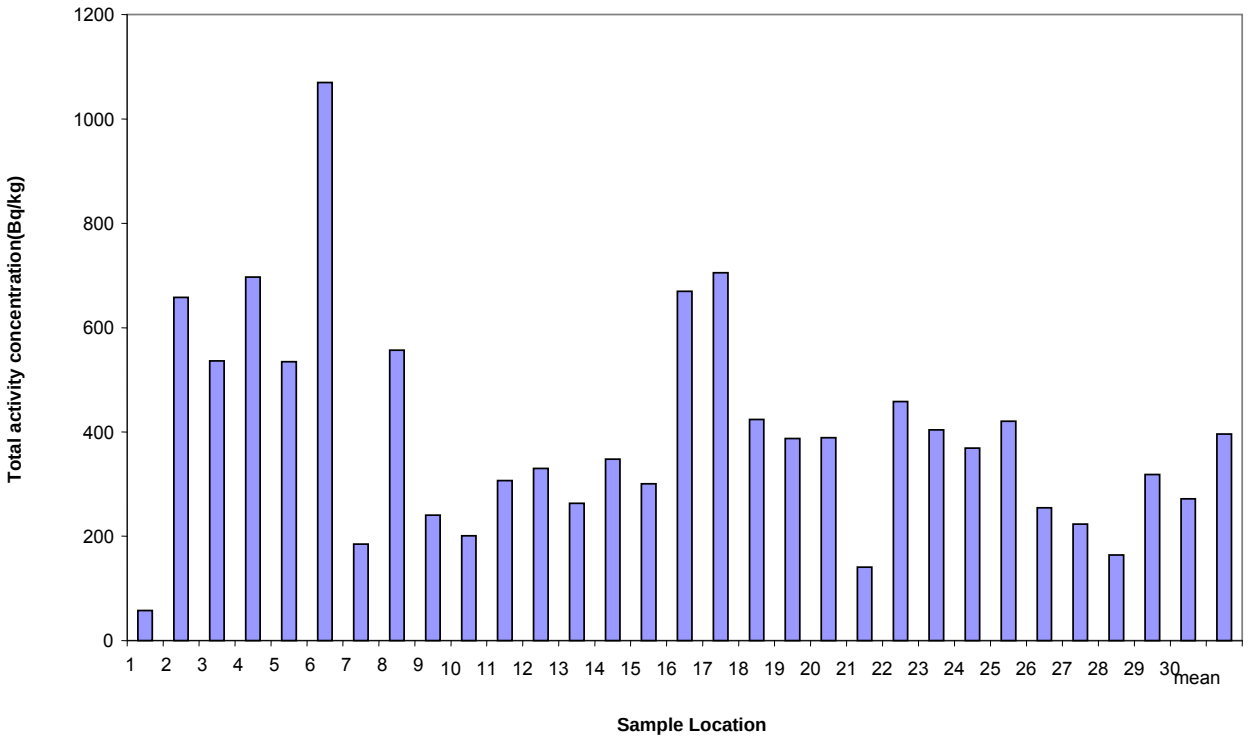

Figure 4: Total Activity Concentration Against Sample Location.

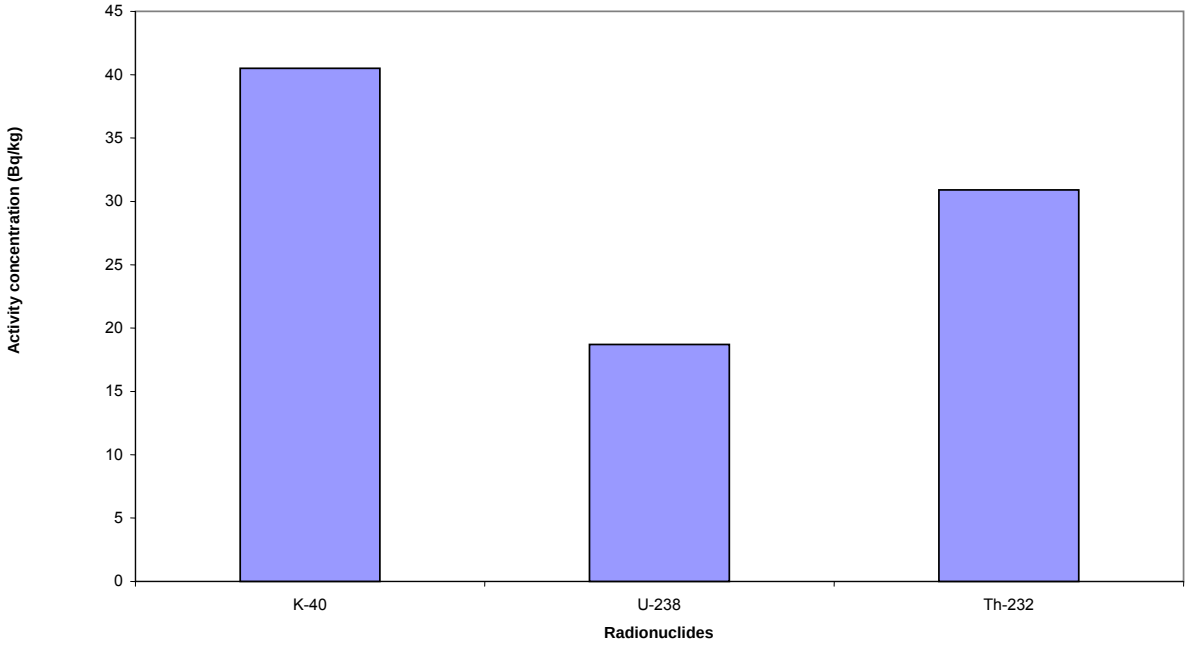

Figure 5: Range Factor of Activity Concentration of Radionuclides.

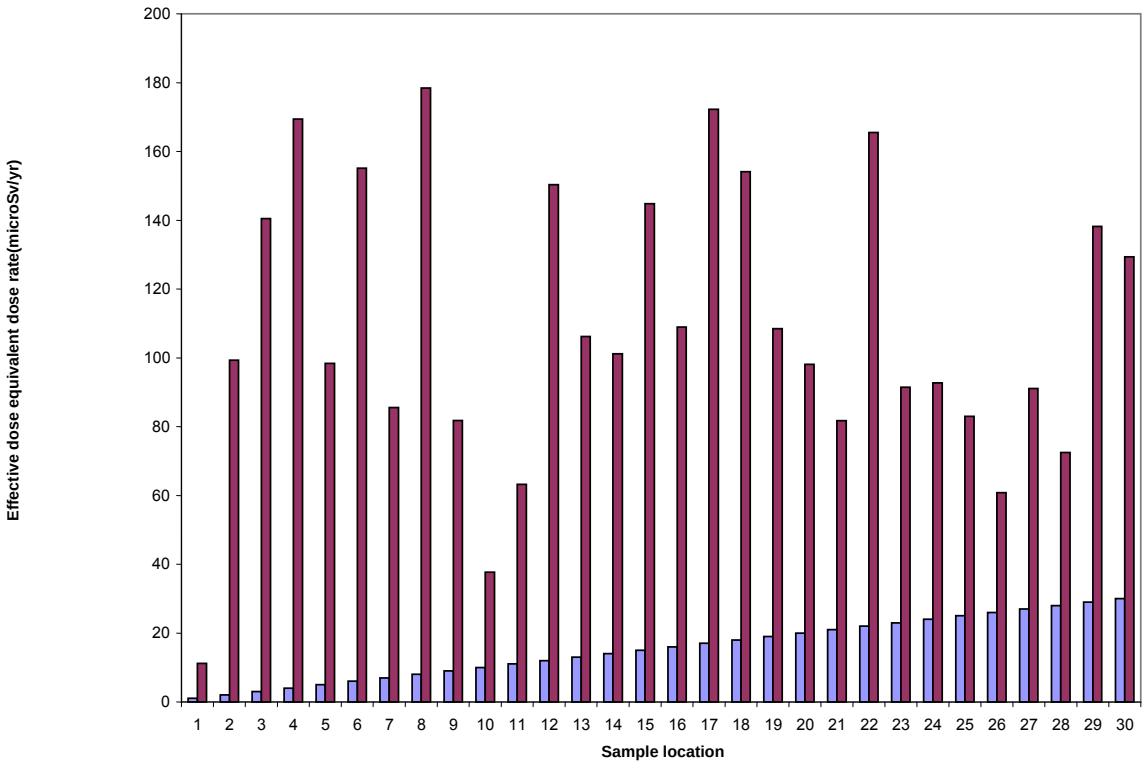

Figure 6: Effective Equivalent Dose Rate Against Sample Location.

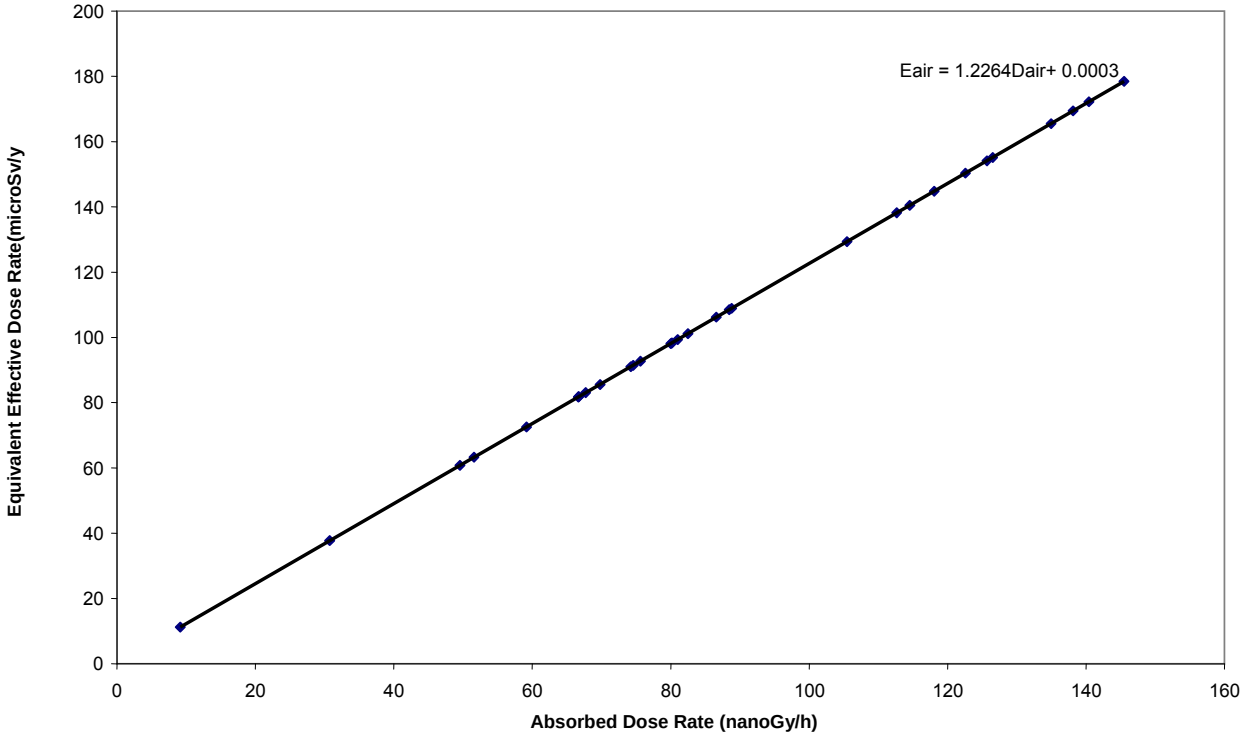

Figure 7: Equivalent Effective Dose Rate Against Absorbed Dose Rate.

Table 6 is the annual effective dose rate,E_{air} .This ranged between 11.21 and 178.45 μSv y^{-1} and the mean of the 30 sample locations was 109.05 μSv y^{-1}. The percentage of sample location with E_{air} above the mean value was 36.7%, while 63.3% was below the baseline value. The outdoor annual effective dose rates in UI are shown in Figure 6 where the highest value was found in sample location 8.

The highest value of 178.45 μSv y^{-1} and the baseline value obtained in UI were both low when compared to the natural external radiation of about 2000 μSv y^{-1} to which no harmful effect will occur directly (Wollenberg and Smith 1990, UNSCEAR, 2000) The result of this study represents reference information on radiation level at UI to which future measurement may be compared. Finally the plot of calculated annual effective dose rate against the absorbed dose rate is shown in Figure 7. The effective annual dose rate is describable by a linear function given by:

$$E_{air}=I\check{D}_{air}+m \qquad (7)$$

where E_{ai} is the outdoor annual effective dose rate and $\check{D}_{air;}$ the constants I =1.2264 and m =3.0x10^{-4}. Using Equation (7) one can easily convert the absorbed dose rate (outdoor) to the outdoor effective dose rate.

CONCLUSION

The radionuclide analysis of top soil of the University of Ibadan (UI) has been carried out using NaI (Tl) as detector. The result obtained indicate that the three types of radionuclides which include ^{40}K, ^{238}U, and ^{232}Th were present and were unevenly distributed due to the slight difference in geological setting of different locations where the samples were collected within UI. Moreover, the result of the investigation revealed that the baseline and the highest annual effective dose rate equivalent were lower than the recommended value of 2000 μSv y^{-1} . These findings are indications that the radionuclides present in the topsoil of UI do not constitute any hazard to the University community. However, the result of this study represents reference

information on radiation dose level at UI against which future measurement may be compared.

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