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NATURAL OCCURRING RADIONUCLIDE VARIATION WITH A SOIL DEPTH PROFILE OF UDI AND EZEAGU LOCAL GOVERNMENT AREAS OF ENUGU STATE, NIGERIA

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Abstract. *The assessment of the natural radionuclide concentration with a depth profile of two drilled boreholes in Enugu State, Nigeria was carried out using a gamma spectroscopy detector. Soil samples were collected at 20m depth intervals down the boreholes (wells) with a depth of about 200m and 140m for Well 1 (Amagu-Umuene) and well 2 (Ogulogu-Olo) respectively. The average values of ^{40}K, ^{226}Ra and ^{232}Th obtained is 57.17Bqkg^{-1}, 13.71Bqkg^{-1} and 10.49Bqkg^{-1} respectively for Well 1 and 59.77Bqkg^{-1}, 11.49Bqkg^{-1} and 8.83Bqkg^{-1} respectively for Well 2.The computed absorbed dose and equivalent dose rate has a mean value of 15.20ηGyhr^{-1} and 0.13mSvyr^{-1}for Well 1 and 13.30ηGyhr^{-1} and 0.12mSvyr^{-1} for Well 2, respectively. These values obtained are well within the world average value and may not result in any significant health effects on human and environmental hazard. The variation of the radionuclides for both wells is irregular down the depth of the boreholes profile. The irregular distribution pattern down the aquifer is attributed to the mineralogy and the geology of the study area.*

Key words: *Natural radioactivity variation, soil depth profile, Enugu, Nigeria*

1. INTRODUCTION

The environment is a complex ecological system and adverse impact on one part can ultimately affect other parts (Eze, 2008). The environment, when exposed to a deleterious substance, can become polluted, thus having a harmful effect on man and other biotic organisms in the environment. Radionuclides are one of the sources of pollution in the environment, if allowed to build up due to natural occurrence or anthropogenic activities. Radionuclides which emit radiation are found naturally in air, water and soil. Natural radioactivity is common in the rocks and soil that make up our planet, in waters and oceans, and in building materials. The monitoring of radioactive materials are therefore of primary importance to man and environmental protection (El-Bahi, 2004).

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Studies on radionuclide concentration variation with soil type and depth profiles are relatively new in the field of radiation and radioactivity concentration studies in the environment, especially in Nigeria. Studies have shown that in soil and water, terrestrial radiations and radioactivity are highly dependent on the soil type and mineral content of the environment (Nevas et al., 2002a); however, reports on activity variation with a soil depth profile are scarse. Nevas et al., (2002b) determined the distribution of natural gamma-emitting radionuclides in three soil profiles developed on tertiary sedimentary materials in the mountain landscapes of the Central Spanish Pyrenees and reported a depletion of activity with depth. They concluded that the variation in radionuclide activities of the soil may be due to the differences in carbonate content, organic matter and/or grain size. Nevaset al. (2002a) report that soil properties differently affected the mobilization of natural radionuclides and the association of some radiologic properties with soil layers, suggesting a relationship between soil processes and radionuclide distribution.

Vukašinović et al., (2009) measured the radionuclide variation from a series of soil cores between 13 and 40 cm of depth collected in different lithology and reported that radionuclides show variations in the depth profile as well as in the different morphoedaphic environments studied. Furthermore, they reported that variability in some radionuclides seems to be related to mineralogy derived from parent materials as well as to cryogenic and soil processes affecting the depth distribution of the granulometric fractions and the organic matter. Avwiri et al. (2010) measured radionuclide concentration with depth in the lithology of Port Harcourt, Nigeria and revealed that radionuclide concentration reduced with depth in a significant manner down the aquifer (0-40 meters in depth). This present study is aimed at assessing the variability of the natural radionuclides with a depth profile up to 200m in depth. The results obtained from the study will form a baseline data of radionuclide soil profiles of the study area, it will reveal the variation behavior of the radionuclides at further depth profiles from previous studies and will add to the world database of radionuclide variation with depth profiles. It will be useful for geologist and authorities in charge of the implementation of radiation protection standards for the public. During borehole drilling, drillers are literally bathed in the mud drilled out of the reservoir and other sediments associated with the process. The need to ascertain the radiological health hazard content also created a basis for this work.

1.1. Study area

Drilling of water borehole in Nigeria, especially in the urban areas, has become a regular activity. Water exploration through the drilling of boreholes in the Enugu State is a hectic task because of the lithology and structure of the environment. Depths of about 450ft to 600ft are required to get to water table and this can take several weeks to achieve. The area is also noted for the exploration of solid minerals, especially coal.

The study areas are Ogulogu-Olo, E007^{o}08'12.2" N06^{o}24'44.5" in the Ezeagu Local Government Area and Amagu-Umuene, E007^{0}25'15.4" N06^{o}34'03.7" in the Udi Local Government Area, both in the Enugu State. The Enugu State is located in the South-East region of Nigeria with its capital of Enugu, and has coal as its predominant natural resource. The entire area occupies a relatively hilly terrain though there are a few isolated low-lying areas. The area is part of the Guinea Savanna vegetation zone and dominated

by the rainy and dry seasons; drainage in the area is essentially due to dendrites with low infiltration as a result of the dominant underlying shale lithology (Ezeh, 2008).

2. MATERIALS AND METHODS

2.1. Sampling and sample preparation

Soil/sediment sampling was carried out in two deep boreholes during the process of well drilling at the location of Ogulogu-Olo in Ezeagu L.G.A and Amagu-Umuene, IbeteOpkatu, Udi Local Government Areas of Enugu State, in the period between January and April 2012. The soil samples were collected at the depth interval of 20m during the drilling process. The wells were of various depth with Well 1 having a depth of 200m (650ft) and Well 2 with a depth of 140m (460ft) to the aquifer.

The samples were collected using a steel hand geological auger, which was cleaned with acid, detergent and rinsed with tap water. Samples of about 450g wet-weight of soil were collected in new aluminum foil, labeled and placed in polythene bags. The water content in the soil sample was separated from the sample by decantation. The samples were then oven dried at a temperature of 60^0 to 80^0C for about 24 hours. The dried samples were grounded with a mortar and pestle and then allowed to pass through a 100-mesh sieve. Sample of 250g dry-weight were packed in an air tight cylindrical plastic container which is of detector geometry, and stored for a period of four weeks before counting, so that the secular equilibrium could be attained between ^{226}Ra and its short lived progeny (Zarie and Al Mugren, 2010).

2.2. Sample analysis

The samples were analyzed using a thallium-activated Canberra vertical high purity 3"×3" Sodium iodide [NaI(TI)] detector connected to an ORTEC 456 amplifier. The detector was connected to the MAESTRO computer program window that matched the gamma energies to a library of possible isotopes. The cylindrical plastic containers holding the samples were left to sit on a high geometry 7.6cm × 7.6cm NaI(TI) detector. High level shielding against the environmental background radiation was achieved by counting in the Canberra 100mm thick lead castle. Spectra were accumulated for background for 29000 s at 900 volts to produce strong peaks at gamma emitting energies of 1461keV for ^{40}K; 609keV of ^{214}Bi and 911keV of ^{228}Ac, which were used to estimate the concentration of ^{238}U and ^{232}Th, respectively. The energy resolution of the detector using Cs-137 from the International Atomic Energy agency (IAEA) is 8% at the 662keV Cs-137 line, while the activity of the standard at the time of calibration is 25.37KBq. The background spectra, measured under the same conditions for both the standard and sample measurements, were used to correct the calculated sample activities concentration in accordance with Arogunjo et.al. (2005). The activity concentration (C) in Bqkg^{-1} of the radionuclides in the samples was calculated after decay correction, using the expression:

$$C_s\,(\mathrm{Bqkg^{-1}}) = \frac{C_a}{\varepsilon_\gamma \times M_s \times t_c \times P_\gamma} \tag{1}$$

Where C_s = Sample concentration, C_a = net peak area of a peak at energy, ε_y = Efficiency of the detector for a γ-energy of interest, M_s=Sample mass, t_c= total counting time, P_γ = Emission probability of radionuclide of interest.

2.3. Radiation hazard indices calculation

Different known radiation health hazard indices analysis was used in radiation studies to arrive at a better and safer conclusion on the health status of a radiated or irradiated person and environment in recent studies (Diab et al., 2008; Kabir et al., 2009; Zarie and Al Mugren, 2010; Senthilkumar et al., 2010; Agbalagba and Onoja, 2011). To assess the radiation hazards associated with the study soil samples, the absorbed dose rate and Annual dose rate were estimated.

a) Absorbed dose rate (D):

The absorbed dose rates outdoors (D) due to gamma radiation in the air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on the guidelines provided by UNSCEAR (2000).The conversion factors used to compute absorbed the γ–dose rate (D) in air per unit activity concentration in Bqkg^{-1} (dry weight) corresponds to 0.462ηGyhr^{-1} for ^{226}Ra (of U- series), 0.621 ηGyhr^{-1} for ^{232}Th and 0.0417 ηGyhr^{-1} for ^{40}K (UNSCEAR, 2000; Ashraf et al., 2010)

$$D\ (\eta Gyhr^{-1}) = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_{K} \quad (2)$$

b) Annual effective dose rate:

To estimate the annual effective dose rates outdoors, one has to take into account the conversion coefficient from the absorbed dose in the air to the effective dose (0.7Sv.Gy^{-1}) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) are used. Therefore, the annual effective dose rate (mSv.yr^{-1}) was calculated by the formula (UNSCEAR, 2000):

$$\text{Effective dose rate } (mSvyr^{-1}) = D(\eta Gy.hr^{-1}) \times 8760h.yr^{-1} \times 0.7 \times (10^{3}mSv/10^{9})\ \eta Gy \times 0.2$$

$$E_{ff}\ Dose = D \times 1.2264 \times 10^{-3} \quad (3)$$

The worldwide annual effective dose from the natural sources of radiation in areas of normal background is estimated to be 1mSvyr^{-1} by UNSCEAR (1993)

RESULTS

Table 1 Activity Concentration of Well 1 (Amagu-Umuene, IbiteOkpatu) (Bqkg^{-1})
Site Location: N06^{o}34'03.7" E007^{0}25'15.4"

Depth (M)	Soil type	Activity concentration (Bqkg^{-1}) ^{40}K	^{226}Ra	^{232}Th	Absorbed dose rate (ηGyhr^{-1})	Equivalent dose rate (mSvyr^{-1})
20	Laterite	68.18±19.53	12.32±9.21	14.97±5.65	18.03	0.16
40	Laterite	54.98±18.75	14.32±7.65	12.65±4.69	16.80	0.15
60	Clay	46.73±73.00	17.17±6.98	8.56±2.87	14.98	0.13
80	Clay/silt	49.56±16.08	9.64±4.45	9.17±2.53	12.27	0.11
100	Silt	62.79±17.28	13.36±6.76	9.87±3.78	14.88	0.13
120	Silty sand	84.54±23.73	15.09±5.76	15.47±5.35	20.23	0.18
140	Sand stone	44.75±13.48	18.64±6.98	11.42±3.65	17.41	0.15
160	Sand stone	65.16±14.24	16.38±6.98	5.06±1.82	13.10	0.11
180	Sharp Sand	46.60±15.43	6.45±2.78	7.34±2.98	9.57	0.08
200	Sharp sand	48.41±19.13	13.71±5.19	10.36±3.83	14.75	0.13
Average		57.17±13.06	13.71±6.27	10.45±3.72	15.20±5.74	0.13±0.02
Worldwide Background Soil Standard		30 (10-50)	35 (10-50)	400 (100-700)	{60 (18- 93) }	1.0

Table 2 Radionuclide Concentration of well 2 (Ogulogu-Olo, Ezeagu) (Bqkg^{-1})
Site Location: N06°24'44.5" E007°08'12.2"

Depth (M)	Sample code	Activity concentration (Bqkg^{-1})			Absorbed dose rate (ηGyhr^{-1})	Equivalent dose rate (mSvyr^{-1})
		^{40}K	^{226}Ra	^{232}Th		
20	Laterite	79.12±19.46	16.75±5.21	11.17±2.25	17.88	0.16
40	Laterite/shale	39.68±14.39	13.39±5.76	7.49±2.73	12.35	0.11
60	Shale	56.15±17.51	9.54±4.64	11.04±3.14	13.73	0.12
80	Shale	42.67±16.02	8.56±2.74	9.07±4.01	11.46	0.13
100	Shale	67.17±14.92	12.79±4.30	9.64±1.79	14.67	0.13
120	Shale/sand	62.74±12.85	11.75±5.02	7.21±3.54	12.41	0.11
140	Sand	70.86±21.54	7.65±3.22	6.20±3.02	10.45	0.09
Average		59.77±16.67	11.49±4.43	8.83±2.93	13.30±3.87	0.12±0.03
Worldwide Background Soil Standard		30 (10-50)	35 (10-50)	400 (100-700)	{60 (18- 93) }	1.0

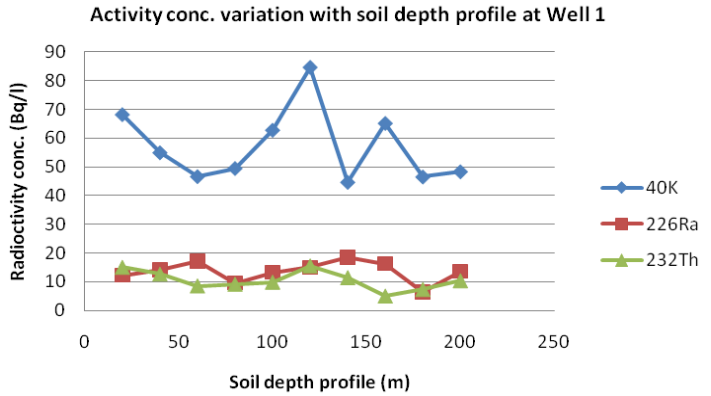

Fig. 1 Radioactivity concentration variation with soil profile depth of Well1

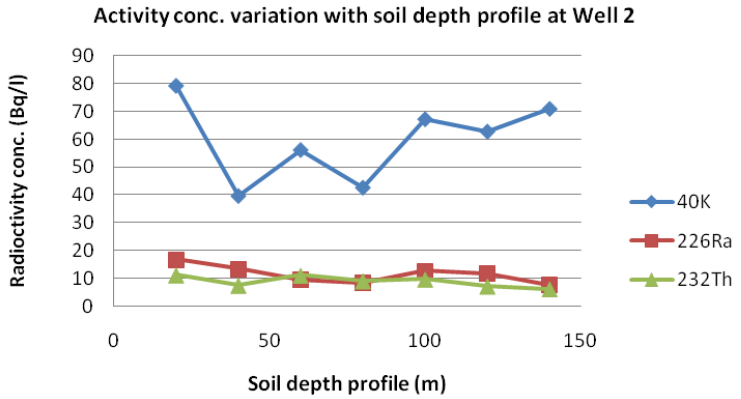

Fig. 2 Radioactivity concentration variation with soil profile depth of Well2

DISCUSSION

Tables 1 and 2 present the three (^{40}K, ^{226}Ra and ^{232}Th) natural radionuclide isotopes present in the two study boreholes. The average values of ^{40}K, ^{226}Ra and ^{232}Th is 57.17Bqkg^{-1}, 13.71Bqkg^{-1} and 10.49Bqkg^{-1} respectively for Well 1 (Amagu-Umuene) and 59.77Bqkg^{-1}, 11.49Bqkg^{-1} and 8.83Bqkg^{-1} respectively for Well 2 (Ogulogu-olo).These radioactivity concentration values obtained in these two boreholes are below the world average value of 400Bqkg^{-1} for ^{40}K, 35Bqkg^{-1} for ^{226}Ra and 30Bqkg^{-1} for ^{232}Th (UNSCEAR, 2000). The relatively high values of ^{40}K are comparable with the values reported by Jankovic et al., (2011) and maybe as a result of its abundance in the earth crust (Tchokossa et al., 1999). The average concentrations of ^{226}Ra in the boreholes are slightly higher compared to that of ^{232}Th. This may be attributed to the fact that ^{226}Ra is moderately soluble in water and is found more abundant than ^{232}Th in the atmosphere (Ashraf et al., 2001).

The computed absorbed dose and equivalent dose rate has a mean value of 15.20ηGyhr^{-1} and 0.13mSvyr^{-1} for Well 1 (Amagu-Umuene) and 13.30ηGyhr^{-1} and 0.12mSvyr for Well 2 (Ogulogu-Olo),respectively. The obtained values are lower compared to those obtained by Avwiri et al., (2010) in the lithology of Port Harcourt, Nigeria. These values are well within the world average value of 60ηGyhr^{-1} for absorbed dose and 1.0mSvyr^{-1} equivalent dose rate(ICRP, 1991 and UNSCEAR 2000). It was observed from Figure 1 and 2 that the variation of the radioisotopes down the aquifer depth was not in a definite order. The distribution pattern of the radionuclide for both Well 1 and Well 2 is irregular (neither ascending nor descending), but has a pattern which could divide the soil profile of the study areas into segmented layers. The change in the characteristic pattern was observed at a periodic depth interval of about 60meters for Well 1 and 25meters for Well 2.

The analysis of the results pattern showed that for Well 1, the process of infiltration (layers acting as filters and buffers) was observed down the laterite zone until a sudden and sharp increase was observed at the clay/silt layer. It is known that clay/slit contains high concentrations of minerals and radionuclide (Nevas et al., 2002b). At the sandstone region, the infiltration process continued down to the sand region where the concentration of the radionuclide increased because of the sandstone. For Well 2, there is also a reduction of radionuclide concentration down the well at the laterite region. This may also be the result of the infiltration process down the soil profile. But at the shale zone, the radionuclide concentration was retained and it fluctuated at an irregular order. This indicated that shale soil has the ability to retain and conserve minerals and radionuclides. At the sand zone, there was a decrease in the radionuclide concentration which further shows the process of infiltration down the well. Following from these observations, one may infer that the radionuclide concentration down the well of an aquifer is affected by the process of infiltration (i.e. it reduces down the well) except at some soil layers such as shale, clay and silt where the concentration is either retained or increased due to the mineral content of the layer. These results compared and agreed satisfactorily with those presented by Nevas et al., (2002a) where the ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K exhibited a very irregular pattern down the aquifer depth. Variation in the radionuclides seems to be related to mineralogy derived from parent materials and soil processes.

CONCLUSION

The study on the natural radionuclides present in the soil profile of two borehole aquifers in Enugu, Nigeria was carried out. The radionuclides were observed to be distributed at an irregular pattern down the aquifer, which could be attributed to the mineralogy, carbonate content, soil texture and organic matter. The average radionuclide levels of the aquifer are lower than the world standards for such an environment and as such exposure to the drilling mud of the drillers and other workers will pose no significant health threat to human lives and the environment is said to be safe radiologically. Therefore, the irregularities in the distribution pattern observed in this study may be attributed to the irregular distribution of the minerals in Enugu State.

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PRIRODNI POJAVNI OBLICI VARIJACIJA RADIONUKLEIDA U SASTAVU ZEMLJIŠTA NA RAZLIČITIM DUBINAMA NA UDI I EZEAGU OBLASTIMA ENUGU STATE U NIGERIJI

Procena koncentracije radionukleida koji se prirodno javljaju u sastavu zemljišta na raznim dubinama u okviru dve bušotine u Enugu State u Nigeriji vršena je uz pomoć gama spektroskopa. Uzorci zemlje vađeni su na intervalima od 20 metara dubine u bušotinama (bunarima), gde je dubina bunara 1 (Amagu-Umuene) bila 200 a dubina bunara 2 (Ogulogu-Olo) bila 140m. Prosečne vrednosti ^{40}K, ^{226}Ra i ^{232}Th dobijene su 57.17Bqkg^{-1}, 13.71Bqkg^{-1} i 10.49Bqkg^{-1} tim redosledom iz bunara 1 i 59.77Bqkg^{-1}, 11.49Bqkg^{-1} i 8.83Bqkg^{-1} tim redosledom iz bunara 2. Izračunate absorbovane doze i odgovarajuće stope doziranje imaju srednj vrednost od 15.20ηGyhr^{-1} i 0.13mSvyr^{-1} za bunar 1 i 13.30ηGyhr^{-1} i 0.12mSvyr^{-1} za bunar 2 tim redosledom. Ove vrednosti se nalaze u okviru granica preporučenih u svetu i može se reći da ne predstavljaju neki značajniji negativan uticaj na zdravlje ljudi ili životnu okolinu. Varijacije radionukleida u oba bunara su nepravilne duž samih bušotina. Nepravilna distribucija pripisuje se mineralnom i geološkom sastavu istraživane oblasti.

Ključne reči: *Varijacije radioaktivnih materija u prirodi, sastav zemljišta na različitim dubinama, Enugu, Nigeria*