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COMPARISON OF ACTIVITY CONCENTRATIONS OF NAT-URAL RADIONUCLIDES IN SOILS COLLECTED AT DIF-FERENT DEPTHS OF SELECTED HAND-DUG WELLS IN ABEOKUTA

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ABSTRACT

This study was aimed at measuring and comparing the activity concentration of soil samples collected from some selected hand – dug wells with their corresponding depths of collection in Abeokuta metropolis. Total of twenty (20) soil samples were collected from hand-dug wells in five sites (Obada, Adigbe, Kuto, Olorunsogo, and Obantoko) within Abeokuta with four (4) soil samples from each hand-dug well at the surface, (0.0m) through to 2.25m depth. Gamma ray spectroscopy with High Purity Germanium (HPGe) detector was used for the measurements. The average activity concentrations obtained for the three natural radionuclides 226 Ra, 232 Th and 40 K in Bq/Kg are 34.31 ± 2.01, 128.73 ± 4.41 and 152.31 ± 2.59 respectively at depth 0.00 m (surface), 23.00 ± 1.61, 68.39 ± 3.24 and 191.08 ± 3.11 respectively at depth 0.75 m, 31.52 ± 2.21, 145.37± 4.95 and 375.56 ± 5.50 respectively at 1.50 m and lastly 28.57±1.70, 95.61 ± 3.71 and 181.10 ± 3.94 respectively at 2.25 m depth. The world average activity concentrations for 226 Ra, 232 Th and 40 K are given to be 35 Bqkg⁻¹, 30 Bqkg⁻¹ and 400 Bqkg⁻¹ respectively (UNSCEAR 2000). 232 Th showed higher average values than the world's average while averages of 226 Ra and 40 K were lower but most of the activity concentration values obtained in some of the locations are higher than the world's average values, especially 226 Ra and 232 Th in the soil samples.

Keywords: Activity Concentrations, Depth, High Purity Germanium (HPGe) detector, Abeokuta

INTRODUCTION

Our natural environment is continuously bombarded with ionising radiations from both natural and man-made sources (UNSCEAR, 1999). With the knowledge of the negative effects of an unguided exposure to ionising radiation, there has been a tremendous effort to locate and control these sources.

Apart from the natural sources, there are other known technically enhanced sources: nuclear and electric generating plants, gas and oil production and solid mineral mining (Fasasi *et al.*, 1999).

Also known to contribute to naturally occurring radionuclides is rock phosphate. This is a basic raw material for all inorganic fertilizers, used in agriculture. The continuous application of fertilizers in a given field could enhance the radioactivity level of the field (Nwankwo, 2013; Hussein, 1994).

Water is vital and concurrently, one of the most important natural resources. About 70% of the Earth's surface is covered with water, (Ashton *et al.*, 2012). However, most of it is salty, and only around 2.5% of the global water resources consists of freshwater (Nwankwo, 2013).

Over 30% is stored as groundwater beneath the earth's surface which are useful for different domestic purposes through hand dug wells and the likes. However the quality of groundwater sources of many communities can be unfit for drinking as a result of contaminations (Nwankwo, 2013, Ibe and Njokwu, 1999).

The research motivation was based on the fact that it has been estimated that at least one-eighth of the mean annual effective dose due to natural sources of radiation is caused by the consumption of foodstuff and water (UNSCEAR 2000). Since Water plays a central role in human nutrition and existence, therefore studies of the dose from this radioactive sources and its effects on health are of primary importance from the point of view of environmental radiation protection. This motive is justified since the earth's crust contains naturally occurring radioactive materials (terrestrial radioactivity), which increases with depth (Kozinski, 1995) and these radioactive materials occur naturally and of most concern are the uranium series, thorium series and their progenies (radon and thoron). Because of this, drinking water from deep wells and boreholes is likely to contain a higher concentration of radioactive elements than surface water (Tajudeen, 2006). Spring or flow-

ing water, passes through rocks that contain many radioactive materials and this could be transported into wells and boreholes (Saidu and Ike 2013). Most of the sources of water supply in Nigeria are upland surface water or groundwater from boreholes and hand dug wells since portable tap water is not readily available to many communities. Many works have been done on radionuclides in water in Nigeria and around the world (Nwankwo, 2013, Ibe and Njokwu, 1999 and Saidu and Ike 2013. etc), but very few studies was carried out on radionuclides in soil samples from Hand dug wells. Researchers have also shown that Abeokuta, (the study area), is among the high background radiation areas in Nigeria (Jibiri et al., 2009, Farai and Jibiri, 2000). Which they also show that geology of an area enhances the level of radiation in the area. Hence the objectives of this study are to:

- Determine the activity concentrations of natural radionuclides in soil samples collected at different depths of some selected hand-dug wells from which water for different domestic purposes can be obtained in Abeokuta metropolis which is known to be a high background radiation area.
- Determine the relationship between activity concentrations of the natural radionuclides in the soils collected with depths
- Determine the level of radiological hazards that is associated with the use of these wells for drinking and domestic purposes.
- Disseminate the information obtained from this study to the communities involved.

MATERIALS AND METHODS

The materials and methods used to carry out this study and the study area are discussed below

The Study Area

The project work was carried out on five (5) sites within Abeokuta in Ogun State Nigeria (Table 1). Abeokuta means "Refuge Among Rocks" in the local language, this is because the caves of the outcrop rocks provided shelter and save havens for the early settlers from invaders (Britannica, 2012; Demeji, 2010). Abeokuta is the capital city of Ogun state, Southwestern Nigeria. It is situated between latitudes 3^o 20"and 3^o 54" and

longitude 7º 9" and 7º 39" on the east bank of Ogun river, around a group of rocky outcroppings that rise above the surrounding wooded savanna. The geology of Abeokuta comprises of sequence of rocks that starts with the Precambrian basement and consists of guartzites and biotite schist, hornblendebiotite, aranite and aneisses. The sedimentary rock sequences are from Cretaceous to Recent; the oldest of them, known as Abeokuta formation, consists of grey sand intercalated with brown to dark grey clay. It is overlain by the Ewekoro formation, which typically contains thick limestone layers at its base (Jibiri and Okeyode, 2012).

Table 1:	Locations	of the	hand	dug wells
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SN	SAMPLE POINTS	NORTH (Deg.)	EAST (Deg.)
1	ADIGBE	7.11717	3.318
2	KUTO	7.16069	3.34093
3	OBADA	7.07678	3.2935
4	OBANTOKO	7.17606	3.89022
5	OLORUNSOGO	7.17606	3.37086

Methodology

The Five (5) hand-dug well sites that were visited are in areas previously studied and had been known as high radiation areas in Abeokuta. (Adigbe, Kuto, Obada, Olo-runshogo and Obantoko. Four (4) soil samples from each well were collected at each depth of 0.0m, 0.75 m, 1.50 m and 2.25 m, making a total of twenty (20) soil samples in all. In order to quantify the natural radionuclides that maybe present in soil which could eventually filter/drain into the well water, the following processes were carried out.

Each soil sample was collected using handtrowel at each of the depths and the soil samples were labelled with their respective well locations and depths to avoid mix-up. Soil samples were air dried, pulverised/ crushed and made to pass through a 2 mm mesh sieve. A total of 150g of the dried, grounded and sieved sediment samples were put in plastic containers of uniform sizes. The containers were shielded hermetically and also shielded externally to ensure that all daughter products of uranium and thorium, in particular, radon isotope formed, do not escape. A time of 4 weeks was allowed after packing, which was a sufficient time required to attain a state of secular radioactive equilibrium after their progeny.

A computerized gamma ray spectrometry system with high purity germanium (HPGe) detector (model: GC 8023) was used (Fig 1). This is an instrument that changes the kinetic energy of an incident ionizing particle in a particular substance into a flash of light. The number of light photon emitted is proportional to the energy of the absorbed radiation which corresponds to the number of pulses displayed under a photo peak and is proportional to the intensity of the radiation reaching the detector from the samples. The activity concentration of a given natural radionuclide in a sample is proportional to the count per second obtained under a gamma photo peak due to the radionuclides present.

The equipment, (HPGe) detector (model: GC 8023) was calibrated using standard reference samples. It was ensured that the calibration standard reference samples used for the detector efficiency calibration represented the samples to be counted, i.e. calibration standard and samples to be counted are identical in size, shape, density, spatial distribution of active material, they have the same geometrical configurations (Alatise, 2007). The standard reference samples were from International Atomic Energy Agency (IAEA Agency's Laboratories) in Austria.

Having done with the equipment calibration, Computerized gamma ray spectrometry system with the mathematical relations of equations 1 and 2 (UNSCEAR 2000), were used to obtain the activity concentrations of the soil samples (Tables 2-5).

CaA	(1)
C = KA	(2)

Where C is the concentration in Bq/kg, A is the count rate i.e counts in 18,000 seconds (5 hours) under the corresponding peak and K is a constant of proportionality for each radionuclide at constant geometry.

Where C is the concentration in Bq/kg, A is the count rate i.e counts in 18,000 seconds (5 hours) under the corresponding peak and K is a constant of proportionality for each radionuclide at constant geometry.

The constant K is determined by the detector efficiency of the assembly due to the energy emitted by the standard source and it is

$$\frac{1}{(\varepsilon p_{\gamma} M_s)}$$

equal to γ is the detector efficiency at the specific gamma-ray energy,

 p_{γ} is the absolute transition probability of the specific gamma-ray and M_s is the mass



Figure 1. Schematic diagram of the detector, high purity germanium crystal

Absorbed Dose Rates

To know the amount of absorbed radiation dose for the samples (i.e the energy deposited per unit mass of medium, which may be measured as joules per kilogram and represented by the equivalent SI unit, gray (Gy)), equation 3 was used.

D (nGy h⁻¹) = 0.462 C_{Ra} + 0.621 C_{Th} + 0.417 C_K (3) Where C_{Ra}, C_{Th} and C_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K of the samples respectively and conversion factors of 0.462 nG4h⁻¹/Bqkg⁻¹, 0.621 nGyh⁻¹/Bqkg⁻¹ and 0.0417 nGyh⁻¹/Bqkg⁻¹ for ²²⁶Ra, ²³²Th and ⁴⁰K respectively (UNSCEAR 2000).

Outdoor Annual Effective Dose Rate

Annual effective dose rate was calculated

using a conversion factor of 0.7SvGy⁻¹ which was used to convert absorbed dose rate in air to human effective dose equivalent with an outdoor occupancy factor of 40%.. The annual effective dose was determined using equation (4) (UNSCEAR, 2000):

Outdoor Annual effective Dose Rate (mSv/ yr) = D x T x F (4)

Where; D is the calculated dose rate (nGyh⁻¹), T is the outdoor occupancy factor (0.4 x 24 x 365.25) hy⁻¹ and F is the conversion factor (0.7 x 10^{-6} SvGy⁻¹).

Radiological parameters

To estimate radiological hazards that could be associated with the use of the sampled wells for domestic purposes, external and internal hazard indices were calculated. The external hazard index H_{ex} can be calculated by using equation (5):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_{K}/4810 \le 1$$
(5)

where C_{Ra} , C_{Th} and C_{K} are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹, respectively from the soil samples. The value of this index is recommended to be less than unity in order to keep the radiation hazard insignificant.

Internal Hazard Index (H_{IN})

In addition to the external hazard index, Radon and its short – lives products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter product is quantified by the internal hazard index (H_{in}), which is given by equation (6) (Jibiri and Okeyode, 2012; UNSCEAR, 2000).

 $H_{IN} = (C_{Ra}/185) + (C_{Tn}/259) + (C_K/4810) \le 1$ (6)

where C_{Ra} , C_{Th} and C_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹, respectively from the soil samples. These values are also recommended to be less than unity in order to keep the radiation hazard insignificant.

DISCUSSION

Table 2 summarizes the results of measurements of natural radionuclide (226Ra, ²³²Th and ⁴⁰K) concentrations in the collected wells soil samples at the surface, Table 3 shows the results at depth 0.75m, Table 4, indicates the values at depth 1.5m and Table 5 shows the values for depth 2.25m. World average concentrations are 35 Bq kg⁻¹, 30 Bq kg⁻¹ and 400 Bq kq⁻¹ for ²²⁶Ra, ²³²Th and ⁴⁰K respectively (UNSCEAR, 2000). Comparing these values with the averages and ranges obtained in this study, it shows that in general, most of the activity concentration values are higher than the world's average values, especially ²²⁶Ra, and ²³²Th in the soil samples.

Table 2: Activity Concentration (Bq/kg) of 226Ra, 232Th and 40K in soil Samples at the Surface (0.0m)

Sample No	Location	236Ra	232Th	40K
1	Adigbe	22.52 ± 1.40	54.05 ± 2.70	11.85 ±0.84
2	Kuto	61.01 ± 3.39	257.07 ± 7.37	584.85 ± 7.95
3	Obada	14.57 ± 0.96	40.33 ± 2.19	31.55 ± 0.96
4 5	Obantoko Olorunsogo	27.71 ± 1.66 45.70 ± 2.63	64.85 ± 3.09 227.29 ± 6.69	18.67 ± 0.96 114.64 ± 2.22
Range	5	22.52 – 61.01	40.33 – 257.07	11.85 – 584.85
Average		34.31 ± 2.01	128.73 ± 4.41	152.31 ± 2.59

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at dep	oth 0.75m			
Samples No.	Location	226Ra	232Th	40K
1	Adigbe	29.50 ± 1.58	53.04 ± 2.70	19.53 ± 0.87
2	Kuto	18.36 ± 1.52	73.49 ± 3.79	500.97 ± 7.04
3	Obada	23.99 ± 1.13	46.06 ± 2.41	18.27 ± 0.94
4	Obantoko	35.61 ± 1.40	57.15 ± 2.99	12.95 ± 1.02
5	Olorunsogo	45.52 ± 2.42	112.21 ± 4.30	403.66 ± 5.68 12.95 –
Range		18.36 – 45.52	46.06 – 112.21	500.97
Average		23.00 ± 1.61	68.39 ± 3.24	191.08 ± 3.11

Table 3: Activity Concentration (Bq/kg) of 226Ra, 232Th and 40K in soil samples at depth 0.75m

Table 4: Activity Concentration	(Bq/kg) of 226Ra,	, 232Th and 40K	in soil samples
at depth 1.50m			

Samples No.	Location	236Ra	232Th	40K
1	Adigbe	29.45 ± 2.23	218.69 ± 6.57	507.93 ± 7.14
2	Kuto	31.18 ± 2.71	190.99 ± 6.12	485.42 ± 6.83
3	Obada	20.20 ± 1.29	43.96 ± 2.48	31.87 ± 1.12
4	Obantoko	44.74 ± 2.54	136.22 ± 4.77	411.93 ± 6.01
5	Olo- runsogo	32.01 ± 2.29	137.01 ± 4.83	439.73 ± 6.41
Range	-	20.20 - 44.74	43.96 – 218.69	31.87 – 507.93
Average		31.52 ± 2.21	145.37 ± 4.95	375.56 ± 5.50

Table 5: Activity Concentration (Bq/kg) of 226Ra, 232Th and 40K in soil samples at depth 2.25m

Sample No.	Location	226Ra	232Th	40K
1	Adigbe	26.46 ± 1.48	33.76 ± 2.30	8.75 ± 0.69
2	Kuto	30.03 ± 1.39	64.19 ± 3.41	31.87 ± 6.21
3	Obada	21.69 ± 1.53	64.59 ± 3.07	57.89 ± 1.46
4	Obantoko	42.29 ± 2.60	227.01 ± 6.76	504.49 ± 7.01
5	Olo- runsogo	22.40 ± 1.50	88.49 ± 3.01	302.49 ± 4.25
Range	C C	21.49 – 42.29	33.76 – 227.01	8.79 – 504.49
Average		28.57 ± 1.70	95.61 ± 3.71	181.10 ± 3.94

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At 0.0m, Kuto has the highest value in the three radionuclides. At 0.75m, Olorunsogo has the highest value of ²²⁶Ra and ²³²Th, while Kuto has the highest in ⁴⁰K. At 1.5m depth, Adigbe has the highest value of ²³²Th and ⁴⁰K, while Obantoko has highest value of ²²⁶Ra. At 2.25m, Obantoko has highest value of the three radionuclides. Farai and Vincent (2006) confirmed Obantoko to have high radiation level. From these results, it shows that the geology of the soil from the sampled wells are really enriched in Thorium and Radium. It implies that radon (daughter product of radium) would be emanating from these soils into the well water. The geology of Abeokuta is basement complex (Gbadebo, 2011; Jones and Hockey 1964) which accounts for the high values as this enhances level of radiation

(Gbadebo, 2011).

The calculated absorbed dose rates are plotted in Figure 2, while annual effective dose rates of the samples are plotted in Figure 3. The International Commission on Radiological Protection (ICRP) has recommended the annual effective dose equivalent limit of 1 mSv/yr for the individual members of the public and 20 mSv/yr for radiation workers (ICRP, 1993). The average absorbed dose rate (D) is nGyh-¹ for the five locations, Adigbe, Kuto, Obada, Obantoko, and Olorunsogo are 74.01, 123.90, 41.04, 102.59 and 117.34 respectively. These values are higher than 57 nGyh-1 reported by UNSCEAR 2000 except Obada with a value of 41.04n Gyh⁻¹.



Fig 2: Absorbed dose rate (D) nGyh-1 for the soil samples at various Locations and depths



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Figure 3. Outdoor Annual Effective Dose Rate in (mSvy-1) for the soil samples at various Locations and depths

The corresponding outdoor annual effective doses range from 0.08 to 0.42 mSv/yr from 0.0m through 2.25m depth for Adigbe, 0.14 to 0.52 mSv/yr for Kuto, 0.08 to 0.13 mSv/ yr for Obada, 0.13 to 0.45 mSv/yr for Obantoko and 0.19 to 0.41 mSv/yr for Olorunsogo respectively. The averages for each of the locations from depth 0.0m through 2.25m are 0.18 mSv/yr, 0.30 mSv/ yr, 0.10 mSv/yr, 0.25 mSv/yr and 0.29 mSv/yr respectively. The world's average annual effective dose is approximately given as 0.5 mSv, comparing with value in this study, all locations are below the world's average value and are less than the recom-

mended value of 1mSv/yr.

The calculated average values for External Hazard Index (H_{ex}) for Adigbe, Kuto, Obada, Obantoko, and Olorunsogo are 0.45, 0.75, 0.25, 0.62 and 0.71 respectively from depths 0.0m through 2.25m, Figure 4. The Internal Hazard Index (H_{in}) are 0.50, 0.84, 0.32, 0.71 and 0.81 for Adigbe, Kuto, Obada, Obantoko and Olorunsogo respectively Figure 5. Although, there are some locations that have high values greater than unity, the calculated averages are still less than unity in the two indices.



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Figure 4. External Hazard Index (Hex) for the soil samples at various Locations and depths



Figure 5: Internal Hazard Index (Hin) for the soil samples at various Locations and depths

CONCLUSION

The specific radioactivity values of 226 Ra, 232 Th and 40 K measured in the soil samples from the selected wells were determined by High Purity Germanium (HPGe) gamma ray spectrometer. For each sample in this study, the absorbed dose rate, annual effective dose rate, External Hazard Index (H_{ex}) and Internal Hazard Index (H_{in}) were determined to assess the radiological hazards that may be linked with the use of water from the wells from which the soil samples were obtained.

In this study, the order of the contributions of radionuclides to the activity concentrations is ${}^{40}K.>{}^{232}Th>{}^{226}Ra$. Although, these values are high but they still fall within the safe range of high radiation areas.

There was no relationship between the concentrations and the depths of ²²⁶Ra and ²³²Th, but for ⁴⁰K the mean activity concentrations increased from the surface with increasing depth to 1.50m and finally decreased at 2.25m. Therefore the activity concentrations of the radionuclides are neither permanently increasing nor decreasing with increasing depth.

Although the absorbed doses calculated are higher than the world's average value but the outdoor annual effective dose equivalent is lower than 1mSv/yr.

The hazard indices are higher than unity in some locations but the averages are all less than unity, showing that the soils from the wells do not pose any significant radiological hazard to the people using the wells for domestic purpose.

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