Radioactivity level of drilled well water across selected cities in Ondo and Ekiti states, Southwestern Nigeria and its radiological implications

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► ***Original article***

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Revised: May 2019
Accepted: August 2019

Int. J. Radiat. Res., April 2020; 18(2): 351-358

DOI: 10.18869/acadpub.ijrr.18.2.351

ABSTRACT

Background: The increasing health effects of nuclear radiation occasioned by enhanced human activities in the environment necessitated the need for constant evaluation and assessment of radiological impact on the general populace within a confined developmental area. Hence, Ten (10) drilled well water samples were collected from various cities distributed across Ondo and Ekiti states and analyzed for gamma-emitting radiations. **Materials and *Methods:*** The collected water samples were analyzed using n-type co-axial HPGe detector (Canberra Inc, USA), to determine the activity concentrations of the gamma emitting radiations, which was used with dose conversion factors to calculate the age dependent total annual effective dose equivalents to six different age groups and the committed effective dose for the age group >17y. ***Results:*** Activity concentrations ranged from 2.25±0.39 to 35.61±6.22 Bq l^{-1} ^{232}Th, 7.08±1.71 to 56.68±12.50 Bq l^{-1} ^{226}Ra, 45.42±2.98 to 467.61±31.69 Bq l^{-1} ^{40}K and 1.66±0.46 to 25.55±5.76 Bq l^{-1} ^{232}Th, 4.90±1.08 to 54.18±13.34 Bq l^{-1} ^{226}Ra, 41.50±2.89 to 558.82±31.69 Bq l^{-1} ^{40}K, respectively for Ondo and Ekiti States . Furthermore, the mean total annual effective dose equivalent for the age groups was found to be within the range of (2.81 ± 0.46 – 26.91 ± 5.11) mSv/y and (4.71 ± 0.92 – 23.58 ± 5.12) mSv/y respectively for Ondo and Ekiti states. ***Conclusion:*** This is above the 1.0 mSv y^{-1} and 0.1mSv y^{-1} respectively set by ICRP and WHO. Hence, the drilled wells are recommended for water screening to remove radionuclides.

Keywords: *Drilled well, HPGe, Total Annual effective dose, committed effective dose.*

INTRODUCTION

The indispensability of water for domestic, commercial, industrial and agricultural uses is apparent and cannot be over-emphasised. This places a demand on adequate water sources for their appropriate applications. It may, however, be well said that nature has prepared for this high demand as water constitutes over 70% of the earth. Of all the different streams of water use, its drinking function seems to have received the most attention, particularly with respect to the quest for good quality water supply and this has recurrently been a challenge for many public and private water suppliers in spite of advances in science and technology. Notable sources of water used for drinking include: dug-wells, boreholes, rivers, streams etc. The choice and adoption of one or more sources would basically depend on source availability, standard of living, processing technique, source purity etc.

Water purity is defined by the level of contaminants which could be: physical, chemical, biological etc. [1]. Toxicity, diseases, and health hazards associated with water are also linked to the quantity of these contaminants. Packaged water in bottles and sachets are usually labelled in terms of its chemical composition with no account of the level of its biological and physical contaminants to the public. Radiological constituents such as ^{232}Th, ^{226}Ra, and the non-decay series ^{40}K which can pose serious health risk for humans are even more shielded from public knowledge. ^{226}Ra and ^{228}Ra, which are decay products of the Uranium series, are known to be very hazardous (2). Radium share similar human in vivo attachment with calcium and its accumulation over a long time can result into bone or sinus cancer (3). Higher concentrations of radioactivity in environmental media are connected with risk of health challenges to humans such as kidney damage, mutagenicity, leukemia as well as cancer of the bladder, kidney, testis and lung (4). In view of the radiological health hazard potentials there is a need to assess the level of radioactivity present in various water sources and to ascertain whether this is above or below the minimum permissible dose due to exposure. While many developed nations of the world have set up radiological regulatory agencies for their water supplies, it is not certain that developing nations have closely toed this line (5). The World Health Organization (WHO) and the United states Environmental Protection Agency (EPA) have issued regulations and guidelines on the quality of drinking water (5). Generally, water supplies from drilled or dug wells are sourced from the soil, which is the product of weathering from the parent rock. The distribution of radionuclides in any water supply is a function of the local geology of the parent rock or soil that represents the sources of primordial radionuclides (6, 7).

Some researchers have asserted that some sachet drinking water in Northern Nigeria were not suitable for drinking because the beta activity concentration of radionuclides in them was above the recommended levels by international standard organisations (8). Also an elevated activity concentrations of radionuclides was reported in dug well water sampled across Ondo and Ekiti states, with the mean total annual effective dose equivalents of (11.49 ± 1.92, 3.02 ± 0.48, 2.52 ± 0.41) mSv y^{-1} and (26.52 ± 5.33, 6.63 ± 1.32, 5.77 ± 0.76) mSv y^{-1} for three different age groups respectively (9). Additionally, the physical parameters and the total radioactivity concentrations in some borehole water were investigated in Zaria, Northwestern Nigeria (10). They noted that some of the samples met the requirements of good water supply, while some had the alpha and beta radioactive concentrations above the set values recommended by the World Health Organization and the US Environmental Protection Agency (EPA). Furthermore, the radionuclides concentrations in some public water were assessed in Markudi Metropolis of Benue state, Nigeria using a Geiger Muller Counter. It was reported that there existed the highest radiation concentration in borehole water ranging of 2.86 × 10^{-1} Bq to 3.69 × 10^{-1} Bq and the least in bottled water was in the range 0.55 × 10^{-1} – 0.77 × 10^{-1} Bq. It is evident that physical contaminant in drinking water should not be under estimated as it poses a great risk to human health (11). Evaluation of the influence of the target tissue mass and activity concentrations of ^{238}U, ^{232}Th, ^{222}Rn and ^{220}Rn on the annual committed equivalent doses in the compartment of the human body was also carried out. It was concluded that for all groups of European consumers considered, annual committed equivalent doses due to ^{238}U and ^{232}Th increase with increase in corresponding activity integrals and decrease in the mass of the target tissue (12).

Drilled and dug well are the major sources of water available for human use in town and cities of Nigeria. Hence, this work is intended to assess the level of physical contaminants (i.e. radionuclides) present in ten drilled well water distributed across selected cities in Ondo (50 48'N, 40 45'E) and Ekiti states (80 15'N, 60 05'É), South western Nigeria using high resolution gamma ray spectrometry and to estimate the age dependent total annual effective doses accrued to individuals due to ingestion.

MATERIALS AND METHODS

Collection and preparation of samples

Ten (10) Water samples were collected from the selected drilled wells, five from each state using electric pumping system. In this case, the tap was first fully turned on for sometimes to flush the pipes and to ensure access to only fresh water. Then the tap was turned low to reduce water flow rate and radon loss before the samples were collected. The collected water was then emptied into a one and half litres keg and labelled based on the alphabet ascribed to each city. Figures 1a and 1b are the maps of Ondo and Ekiti states showing the samples location. Water samples collected from the various drilled wells across the two states using a one and half litres plastic keg were acidified with 11M of HCl at the rate of 10ml per liter of samples as soon as possible, to avoid absorption of radionuclide onto the wall of container (13). Marinelli beaker of 1L volume capacity previously washed, rinsed with a dilute sulphuric acid (H_2SO_4) and dried to avoid contamination was filled with sample from the container used for sampling. This was later sealed for at least four weeks to ensure that there was no loss of radon and to achieve secular equilibrium between the daughters and the parents' nuclides.

Sample analysis

The activity concentrations of the drilled well water samples were measured using an N-type coaxialHPGe gamma-ray detector at the laboratory of Ghana Atomic Energy Commission Accra with ORTEC Multichannel Analyzer (MCA; Canberra Inc, USA) and MAESTRO-32 evaluation software for spectrum acquisition and processing. The relative efficiency of the detector was 28.5 % with energy resolution of 1.8 keV at gamma ray energy of 1332 keV of ^{60}Co. The gamma lines 609.31 and 1764.49 keV of ^{214}Bi were used to determine ^{226}Ra. The gamma lines 583.19 of ^{208}Tl were used to determine ^{232}Th and that of ^{40}K was determined from the gamma line of 1460.83 keV. The samples were counted for 18,000 seconds (5 hours). The energy and efficiency calibrations were performed using mixed radionuclide calibration standard in the form of solid water, serial number NW 146 A with approximate volume 1000 mL and density 1.0 gcm^{-3} in a 1.0 L Marinelli beaker. The standard was supplied by Deutscher Kalibrierdienst (DKD-3), QSA Global GmBH, and Germany. Background measurements were made for the same period. Density corrections were also done where appropriate.

The specific activity concentrations (Asp) of ^{232}Th, ^{226}Ra and ^{40}K were determined in Bq $l^{-1}$for the water samples using the equation (1) (14-16) after decay correction.

$$A_{sp} = \frac{N_{sam}}{P_E.\varepsilon.T_c.M} \quad (1)$$

where;

N_{sam} - net counts of the radionuclide in the sample

P_E - gamma ray emission probability (gamma yield)

E - total counting efficiency of the detector system

Tc - sample counting time

M - mass or weight of the sample

The specific activity obtained using equation (1) coupled with appropriate dose conversion factors form the basis for the evaluation of the radiological health hazards posed by the analysed samples from the study area. Excel software was used in computation and statistical analysis with cognisance to the method of error propagation described by (17).

Calculation of annual effective dose and committed effective dose

Estimation of annual effective dose E_d (Sv/y) to an individual due to the ingestion of the natural radionuclides present in the drilled well water samples was carried out using the equation (2) (18).

$$E_d = A_cA_iC_f \quad (2)$$

where A_c is the activity concentration of the radionuclide in the drilled well (Bq l^{-1}), A_i is the consumption per annum or annual intake of drinking water and C_f is the ingested dose conversion factor for radionuclides (Sv/Bq). The ingested dose conversion factor and the consumption per annum vary with both radionuclides and the age of the individual ingesting the radionuclides. The ingested dose

[DOI: 10.18869/acadpub.ijrr.18.2.351]

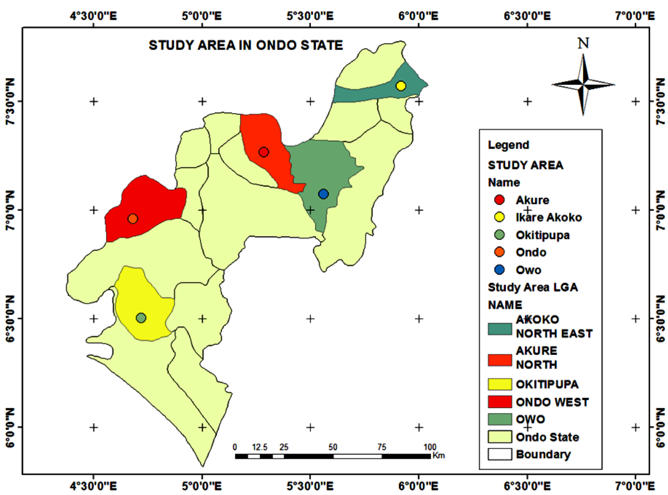
conversion factor C_f used for ^{232}Th, ^{226}Ra and ^{40}K, in this work and the corresponding consumption per annum in l y^{-1} are presented in tables 2 and 3 respectively for the six different age groups [19, 20]. The total effective dose equivalent D (Sv/y) to an individual was established by summing contributions from all radionuclides present in the water samples, shown as equation (3).

$$D = \Sigma A_c A_i C_f \tag{3}$$

The annual effective dose values were calculated for six different age groups The committed effective dose C_d, which is a measure of the total effective dose received over an average life time of 50 years was calculated for the age >17 years due to the ingestion of radionuclides using equation (4):

$$C_d = 50 \times D \tag{4}$$

Where D is the total effective dose to an individual.



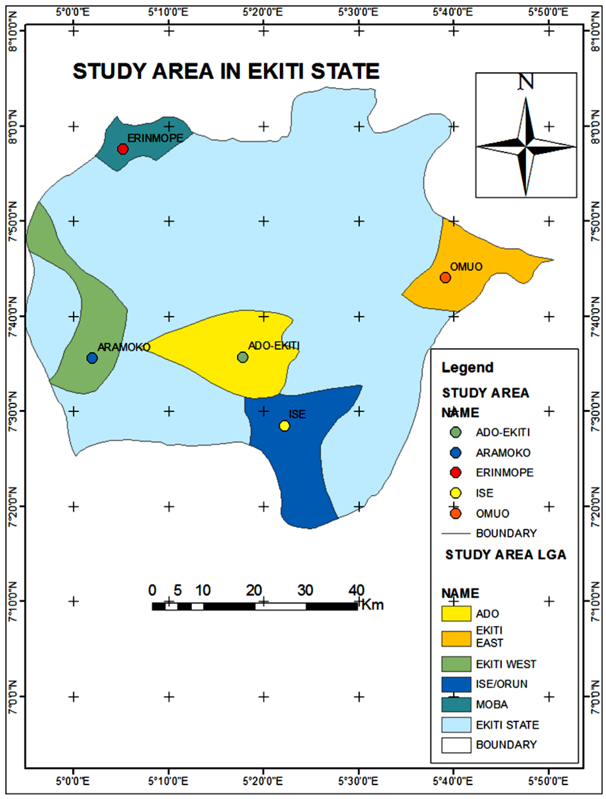


Figure 1. Ondo state map showing the sample locations **(a)**. Ekiti State map showing the sample location **(b)**.

RESULTS

The measured activity concentrations of ^{232}Th, ^{226}Ra, ^{40}K in the drilled well water samples of the two states is presented in table 1. ^{137}Cs were not detected in all the samples. The activity concentrations were found to be within the ranges of 2.25±0.39 to 35.61±6.22 Bq l^{-1} ^{232}Th, 7.08±1.71 to 56.68±12.50 Bq l^{-1} ^{226}Ra and 45.42±2.98 to 467.61±31.69 Bq l^{-1} ^{40}K, for Ondo state drilled well water samples, while Ekiti State drilled well water samples laid within the ranges of 1.66±0.46 to 25.55±5.76 Bq l^{-1} ^{232}Th, 4.90±1.08 to 54.18±13.34 Bq l^{-1} ^{226}Ra, and 41.50±2.89 to 558.82±31.69 Bq l^{-1} ^{40}K. ^{40}K had the highest activity concentration, followed by ^{226}Ra and the least activity was found in ^{232}Th for the two states. The highest activity concentration of the three radionuclides (^{232}Th, ^{226}Ra, and ^{40}K) was recorded in the sample taken from Erinmope-Ekiti and the least was found in the sample taken from Ise-Ekiti with the exception of Ado-Ekiti that had the least activity concentration for ^{40}K. Ondo state had its highest activity concentration in the sample taken from Ikare-Akoko and the least activity concentrations, for ^{232}Th, ^{226}Ra and ^{40}K in the

[DOI: 10.18869/acadpub.ijrr.18.2.351]

sample taken from Owo, Okitipupa and Ondo.
The age dependent annual effective dose equivalent was calculated using equation (2) and the total annual effective dose equivalent using equation (3) for six different age groups (i.e. 0-1 y, 1-2 y, 2-7 y, 7-12 y, 12-17 y, and >17 y). The ingested dose conversion factor C_f for ^{232}Th, ^{226}Ra and ^{40}K, and the corresponding consumption of water per annum in l y^{-1} for different age groups are presented in tables 2 and 3 respectively. Then the calculated total annual effective dose equivalents are presented in tables 4 and 5 for Ekiti and Ondo states respectively. The total annual effective dose equivalents for the six different age groups ranged between (6.65±1.48 – 81.40±18.20, 1.88±0.36 – 22.60±4.40, 1.35±0.27 – 16.30±3.29, 1.73±0.36 – 20.30±4.46, 4.85±1.05 – 55.10±13.00 and 1.47±0.31 – 17.90±3.84) mSv y^{-1} respectively for the drilled well water samples from Ekiti state. For Ondo state drilled well water samples, the total annual effective dose equivalents for the six different age groups ranged between (10.50±1.77 – 91.80±17.90, 2.34±0.42 – 3.03±0.45, 2.04±0.55 – 17.20±3.18, 2.68±0.44 – 21.60±4.28, 7.24±1.67 – 58.50±12.30 and 2.36±0.38 – 19.70±3.74) mSv y^{-1} respectively. The graphical illustrations of these variations are shown in figures 2 and 3 for Ondo and Ekiti states respectively, while the variation of the mean total annual effective dose equivalent for the two states against the age groups is presented in figure 4.

Table 1. Activity concentrations (Bq l^{-1}) of radionuclides in Ondo & Ekiti borehole water samples.

		Samples	^{232}Th	^{226}Ra	^{40}K	^{137}Cs
Ondo state	1.	H - BH	2.77 ± 0.21	8.08 ± 1.59	53.95 ± 3.43	BDL
Borehole	2.	E - BH	2.88 ± 0.53	7.78 ± 1.33	45.42 ± 2.98	BDL
Water	3.	K - BH	35.61 ± 6.22	56.68 ± 12.50	467.61 ± 31.69	BDL
Samples	4.	A - BH	3.55 ± 0.74	7.08 ± 1.71	73.13 ± 4.22	BDL
	5.	I- BH	2.25 ± 0.39	8.25 ± 1.50	64.81 ± 3.07	BDL
Ekiti state	1.	T - BH	2.43 ± 0.59	6.09 ± 1.23	41.50 ± 2.89	BDL
Borehole	2.	S - BH	25.55 ± 5.76	54.18 ± 13.34	558.82 ± 31.69	BDL
Water	3.	Q - BH	2.53 ± 0.90	5.50 ± 1.33	61.64 ± 3.74	BDL
Samples	4.	M - BH	1.66 ± 0.46	4.90 ± 1.08	41.96 ± 3.07	BDL
	5.	O - BH	4.10 ± 0.76	8.92 ± 1.42	78.93 ± 3.84	BDL

BDL= Below Detection limit, BH = Borehole, H = Akure, E = Ondo, I = Owo, K = Ikare Akoko, A = Okitipupa, T = Ado-Ekiti, S = Erinmope-Ekiti, O = Omuo-Ekiti, Q = Aramoko-Ekiti, M = Ise-Ekiti.

Table 2. Radionuclides and their ingested dose conversion factors per age groups[19].

Radionuclide	Ingested Dose Conversion Factor Per Age Group					
	(0-1)y	(1-2)y	(2-7)y	(7-12)y	(12-17)y	>17y
^{40}K	6.20E-08	4.20E-08	2.10E-08	1.30E-08	7.60E-09	6.20E-09
^{226}Ra	4.70E-06	9.60E-07	6.20E-07	8.00E-10	1.50E-06	2.80E0-7
^{232}Th	4.60E-06	4.50E-07	3.50E-07	2.90E-07	2.50E0-7	2.30E-07

Table 3. Annual water consumption per age groups[21].

Age Group (y)	Annual Water Consumption (l y^{-1})
0 – 1	2.00E+02
1 – 2	2.60E+02
2 – 7	3.00E+02
7 – 12	3.50E+02
12 – 17	6.00E+02
> 17	7.30E+02

[DOI: 10.18869/acadpub.ijrr.18.2.351]

Table 4. Total annual effective dose equivalent for six different age groups and committed effective dose resulting from Ekiti drilled well water samples.

Samples	Total annual Eff dose (0-1y) mSv/y	Total annual Eff dose (1-2y) mSv/y	Total annual Eff dose (2-7y) mSv/y	Total annual Eff dose (7-12y) mSv/y	Total annual Eff dose (12-17y) mSv/y	Total annual Eff dose >17y mSv/y	Commited Eff dose >17y mSv/y
T – BH	8.47 ± 1.73	2.26 ± 0.41	1.65 ± 0.31	2.14 ± 0.42	6.03 ± 1.21	1.84 ± 0.36	92.00 ± 18.20
S – BH	81.40 ± 18.20	22.60 ± 4.40	16.30 ± 3.29	20.30 ± 4.46	55.10 ± 13.00	17.90 ± 3.84	895.00 ± 192.00
Q – BH	8.27 ± 2.12	2.34 ± 0.48	1.68 ± 0.37	2.08 ± 0.48	5.61 ± 1.35	1.83 ± 0.44	91.40 ± 22.00
M – BH	6.65 ± 1.48	1.88 ± 0.36	1.35 ± 0.27	1.73 ± 0.36	4.85 ± 1.05	1.47 ± 0.31	73.40 ± 15.60
O – BH	13.10 ± 2.08	3.57 ± 0.49	2.59 ± 0.37	3.27 ± 0.50	9.00 ± 1.41	2.87 ± 0.43	143.00 ± 21.70
Mean	23.58 ± 5.12	6.53 ± 1.23	4.71 ± 0.92	5.90 ± 1.24	16.12 ± 3.60	5.18 ± 1.08	

Table 5. Total annual effective dose equivalent for six different age groups and committed effective dose resulting from Ondo drilled well water samples

Samples	Total annual Eff dose (0-1y) mSv/y	Total annual Eff dose (1-2y) mSv/y	Total annual Eff dose (2-7y) mSv/y	Total annual Eff dose (7-12y) mSv/y	Total annual Eff dose (12-17y) mSv/y	Total annual Eff dose >17y mSv/y	Commited Eff dose >17y mSv/y
H – BH	10.80 ± 1.73	2.93 ± 0.46	2.13 ± 0.34	2.79 ± 0.48	7.93 ± 1.48	2.36 ± 0.38	118.00 ± 18.80
E – BH	10.50 ± 1.77	2.77 ± 0.43	2.04 ± 0.55	2.68 ± 0.44	7.64 ± 1.29	2.28 ± 0.37	114.00 ± 18.70
K – BH	91. 80 ± 17.90	2.34 ± 0.42	17.20 ± 3.18	21.60 ± 4.28	58.50 ± 12.30	19.70 ± 3.74	984.00 ± 187.00
A – BH	10.86 ± 2.34	2.98 ± 0.56	2.15 ± 0.42	2.69 ± 0.58	7.24 ± 1.67	6.09 ± 1.58	304.00 ± 79.00
I – BH	10.60 ± 1.81	3.03 ± 0.45	2.18 ± 0.34	2.83 ± 0.47	8.06 ± 1.42	2.36 ± 0.39	118.00 ± 19.30
Mean	26.91 ± 5.11	2.81 ± 0.46	5.14 ± 0.97	6.52 ± 1.25	17.87 ± 3.63	6.56 ± 1.29	

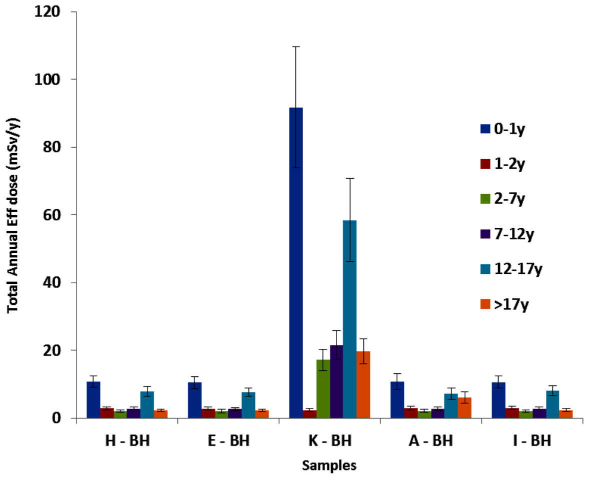


Figure 2. Total annual effective dose (mSv y-1) in the drilled well water samples of Ondo state.

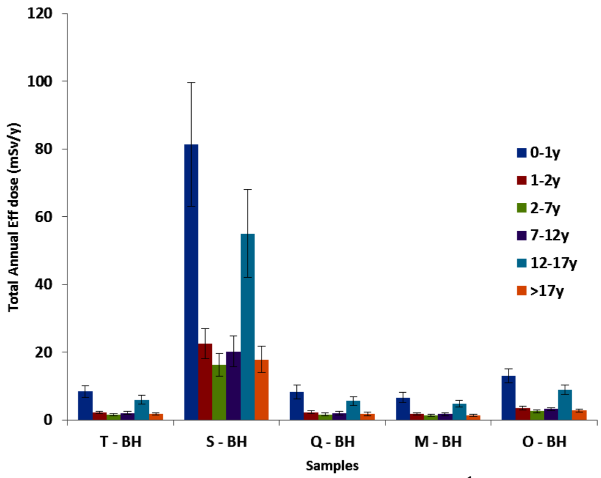


Figure 3. Total annual effective dose D (mSv y^{-1}) variation in the drilled well water samples of Ekiti state.

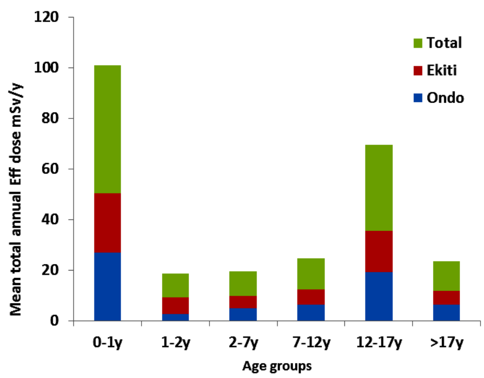


Figure 4. Variation of mean total annual effective dose equivalent against age groups.

[DOI: 10.18869/acadpub.ijrr.18.2.351]

DISCUSSION

From the result of the measured activity concentrations of the three radionuclides presented in table 1, the mean activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K for both Ondo and Ekiti drilled well water samples are 8.43 ± 1.47 Bq l^{-1}, 15.83 ± 3.39 Bq l^{-1}, 129.67 ± 8.27 Bq l^{-1} and 7.25 ± 1.69 Bq l^{-1}, 15.92 ± 3.68 Bq l^{-1}, 156.57 ± 9.05 Bq l^{-1} respectively. The result obtained in both cases for ^{226}Ra exceeds the recommended limits of 1.00 Bq l^{-1} set by the International Atomic Energy Agency [13] and 0.185 Bq l^{-1} set by the United state Environmental Protection Agency [21]. This result corroborates the 7.15 ± 6.94 Bq l^{-1} reported for ^{226}Ra by [22] in private drilled well in Akure Southwestern Nigeria. The mean measured activity concentrations of ^{232}Th and ^{40}K for Ondo state (8.43 ± 1.47Bq/l and 129.67 ± 8.27 Bq l^{-1}) and Ekiti state (7.25 ± 1.69 Bq l^{-1} and 156.57 ± 9.05 Bq l^{-1}) were above the values reported in other part of the world [6, 22].

Similarly, from tables 4.0 and 5.0, the mean total annual effective dose equivalent for each of the age groups (0-1 y, 1-2 y, 2-7 y, 7-12 y, 12-17 y, and >17 y) resulting from the radionuclides in the drilled well water samples of both Ondo and Ekiti states is within the ranges of (2.81 ± 0.46 – 26.91 ± 5.11) mSv y^{-1} and (4.71 ± 0.92 – 23.58 ± 5.12) mSv y^{-1} respectively. This is above the ICRP and WHO recommended limits of 1 mSv y^{-1} and 0.1 mSv y^{-1} respectively [23, 20]. For Ondo state, the samples taken from Ikare-Akoko had the highest total annual effective dose equivalent for almost all the age groups with the exception of the (1-2 y) age groups. The least total annual effective dose equivalent was recorded for all the age groups in the sample collected from Ondo town, with the exception of (1-2 y) and (12 – 17 y) age groups. This is evident from figure 2. The high total annual effective dose equivalent recorded in Ikare-Akoko might be as a result of local geology coupled with emerging industries.

Ekiti state had the highest total annual effective dose equivalent for all the age groups in the sample collected from Erinmope-Ekiti and the least total annual effective dose equivalent in the sample collected from Ise-Ekiti for all the age groups considered. This is a reflection of the activity concentrations of the radionuclides recorded in Ekiti state drilled well water samples.

This is also evident from the graphical illustration presented in figure 3 for Ekiti state. The high total annual effective dose equivalent recorded in the sample collected from Erinmope can be attributed to its closeness to Guinea Savannah where the soil needs huge application of Potash fertilizers to grow crops and the presence of granite rock in the terrain. Therefore the drilled wells, most especially the one from Ikare-Akoko in Ondo state and Erinmope-Ekiti in Ekiti state should be screened for radionuclides. The variation of mean total annual effective dose equivalent for the two states against all the age groups presented in figure 4, affirms that, (0-1 y) is most susceptible to radiation for both states and the least susceptible is (1-2 y) and (2-7 y) for Ondo and Ekiti states respectively. This result is in tandem with the following findings [22, 24, 25]. The committed effective dose which is a measure of the radiation received over a lifetime of 50 y is presented for the >17 y age group.

CONCLUSION

The calculated age dependent mean total annual effective dose equivalent due to the ingestion of these radionuclides from the drilled well of Ondo and Ekiti states was found to be above the recommended limits of 1.0 mSv y^{-1} and 0.1 mSv y^{-1}set by ICRP and WHO respectively, most especially the samples collected from Erinmope Ekiti and Ikare-Akoko. Hence the drilled wells are recommended for radionuclide screening to prevent the outbreak of radiation induced diseases.

Overall, the samples considered across the two states (0-1 y) were most susceptible to radiation and the least were (1-2 y) and (2-7 y) age groups for Ondo and Ekiti states respectively.

Therefore nursing mothers are advised not to feed their lactating babies (0-1 y) with water of unverified quality.

[DOI: 10.18869/acadpub.ijrr.18.2.351]

ACKNOWLEDGEMENTS

The authors wish to acknowledge the support of Mr Nicholas Opata of the Ghana Atomic Energy Commission in analyzing the samples using gamma ray spectrometry.

Conflicts of interest: *Declared none.*

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