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Natural Radionuclides in Natural Spring Water Samples in Ikere – Ekiti Local Government Area, Ekiti State, Nigeria

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Abstract
The presence of the radionuclides in drinking water poses a number of health hazards. This study estimated the committed effective doses due the natural radiounuclides via consumption of spring water in the study area. 80 spring water samples were measured using gamma spectrometry. The annual committed effective doses in Ikere local government , Ekiti State, Nigeria varied from 0.16 mSv/y to 0.22 mSv/y with a mean value of 0.20 ± 0.03 mSv/y. The calculated annual committed effective dose is lower than WHO recommended limit of 1.0 mSv/y for public exposure. Due to the consumption of natural spring water there is no radiological health hazards to the public within the study area.
Keywords: Radionuclides, Activity concentration, Committed effective dose, Spring water, Drinking water

Introduction

Water is indispensable to human life and is therefore an important parameter of environmental science. The presence of radionuclides in drinking water poses a number of health hazards, especially when the radionuclides are deposited in the human body, through drinking the polluted water.

Water pollution is the contamination of water bodies such as lakes, rivers, ocean, underground water, spring and flowing water by human or natural activities which can be harmful to organisms and plants, which live or use the water. Waste product like industrial waste, mining wastes, agricultural wastes and toxic wastes often contain some amount of pollutants as a result of the materials used in generating the waste, thereby adding to the level of water pollution (Tajudeen, 2006).

Another form of water pollution is naturally occurring Radioactive materials (NORM) that emit alpha, beta and gamma radiation, these usually have elements in the Uranium and Thorium series whose radioactive gaseous daughter (radon and thoron) in particular cause an appreciable airborne particulate activity and contribute to the radioactivity of rain and ground water, this also affects drinking water. Furthermore, spring or flowing water passes through rocks that contains many radioactive materials, as a result, the flowing water that leaches and migrates affects the soil and plants on its way, and it could also be transported into wells, boreholes and tap water through burst pipes. The most important radionuclides in drinking water are tritium, potassium – 40, radium and radon, which are alpha, beta and gamma emitters (Tajudeen, 2006).

Radium and other natural radionuclides can enter ground water by dissolution of aquifer materials, by desorption from rock or sediment surfaces, and by ejection from minierals during radioactive decay. Radium in drinking water is known to increase cancer risk, primarily for bone and sinus cancers (USSG, 1998). As radium decays, the radiation that is emitted can strip electrons from the atoms with which it collides, causing the atoms to become charged or "ionized." The ionizing alpha and beta radiation emitted by radium consists of particles that move slowly and cannot penetrate skin. If radium is ingested, however, especially dissolved in water, the emitted alpha- and beta- particle radiation can ionize and damage cell tissue. Human bone tissue accumulates radium rather than allowing it to be removed from the body, thereby exposing the bones to tissue-damaging alpha or beta radiation. Damage from continuous exposure can potentially cause cancer. Radium is considered to pose a greater cancer risk than most other radioactive elements because it accumulated in the body, (USSG, 1998).

There is evidence from both human and animal studies that radiation exposure at low to moderate doses may increase the long term incidence of cancer and that the rate of genetic malformations may be increased by radiation exposure (Ottonm, 2015). It is therefore important to determine the amount of radioactivity in drinking water for every area where people live in, so as to guard against its health hazards (WHO, 2006). The radioactivity in Spring or flowing water comes mainly from radionuclides of the natural decay chains ^{238}U ^{40}K, and ^{232}Th in soil and bedrock. Some radionuclides can dissolve easily in water, depending on the mineralogical and geochemical composition of the soil and rock, redox conditions and the residence time of ground water in the soil and bedrock, as result of the reaction of the ground water with soil and bedrock (Vesterbacka, 2007). In rural area, spring or flowing water, without any purification processes are mostly consumed. The reason for evaluating the activity concentrations of the primordial radionuclides' activities is to ensure that the reference dose level (RDL) of committed effective dose of 0.1mSv y^{-1} consumption of drinking water is not exceeded. The RDL of 0.1mSv is equal to 10% of the dose limit for members of the public, recommended by the International Commission for Radiological Protection (ICRP, 1990) and the International Basic Safety Standard (IAEA, 1996).

Also, they are acceptable to most World Health Organization (WHO) member States.

The aim of this study is to determine the activity concentrations of the primodial radionuclides (^{40}K, ^{238}U and ^{232}Th) and to estimate the committed effective doses due the consumption of natural spring water in the rural areas of Ikere local government area in Ekiti – State, Nigeria.

Sample Collection and Preparation

Geologically, the study area lies within the south western Nigeria crystlline basement rock which is of Precambrian in age. The basement complex which consists of an assemblage of crustalline igneous and metamorphic rocks has been classified into three main groups namely: Migmatite Gneiss comples, schist belts and older granite (Fasae and Akinkuade, 2011). The spring water samples used were collected from springs in close proximity to the rural areas, mostly farm settlements in the study area, where there is no demand for treated water and only spring water are mostly consumed. The study was carried out in the dry season when water intake increases. The spring water samples were collected manually in the early hours of the day in the study area during the period where the weather conditions were fairly stable . The samples were collected through a distance of 600 m at 200 meters interval from the source of the spring. There was no rainfall during sample collection also, the containers were first rinsed three times with sample water before measurement in order to minimize contamination. The samples were collected in 2-litre plastic containers with about 1% air space left for thermal expansion and a few drops of hydrochloric acid were added to bring the pH to an appreciable level of 2 in order to prevent adherence of the radionuclides to the walls of the containers.

Radioelemental analysis using NaI(Tl) gamma spectroscopic system.

A NaI(Tl) detector of dimension, 7.62 cm × 7.62 cm housed in a 6 cm thick lead shield and lined with Cd and Cu sheets was used for the measurement. The IAEA supplied standard isotopic sources (^{60}Co, ^{137}Cs and ^{54}Mn) and reference standard sources RGTh-1, RGU-1 and RGK-1, for ^{232}Th, ^{226}Ra and ^{40}K, respectively (for bulk sample analysis) were used to calibrate for the estimation of concentration levels of the soil samples (Ibeanu, 1999). A computer-based MCA card system MAESTRO programme from Ortec was used for data acquisition and analysis of gamma spectra. In order to estimate the background contribution an empty container was counted using the same geometry as the samples.

The standards and samples were in turn counted for a period of 30,000 seconds per sample in reproducible sample-detector geometry and the count rate in counts per second (cps) obtained for each radionuclide. The gamma ray peak of 1460.0 keV was used in the assessment of ^{40}K, while 1764.0 keV and 2165.0 keV gamma lines of ^{214}Bi were used in the assessment of the activity concentrations of ^{226}Ra and ^{232}Th, respectively. According to published reports, the activity concentration (Bq/L) in the water samples is given by Faanu et. al. (Faanu et al., 2011):

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

where N_{sam} is the background corrected net counts of the radionuclide in the sample given in Bq /L, P_E is the gamma ray emission probability (gamma yield), ε is the detector efficiency of the detector system, T_c is the sample counting time , and M_s is the mass of the sample (kg) or volume (L).

The annual effective doses (mSv/y) from ingestion of radionuclides in water samples collected in the study area were calculated on the basis of the mean activity concentrations of the radionuclides. The daily water intake was considered to be 2 litres per day and the conversion factors or dose per unit intake by ingestion for naturally occuring radionuclides for adult members of the public was taken to be 6.2×10^{-6} mSv/Bq for ^{40}K, 4.5×10^{-6} mSv/Bq for ^{238}U and 2.3×10^{-5} mSv/Bq for ^{232}Th [10] were used.

The annual effective dose H was calculated using the expression below (ICRP, 1996):

$$H_{ing} = \sum_{j=1}^{3} D_{ing}(K, U, Th) . C_{sp} I \quad (2)$$

where, D_{ing} (K, U, Th) is the dose conversion coefficients of the radionuclides in Sv/Bq, C_{sp} is the specific activity concentrations of radionuclides in the water samples in Bq/L.

Results and Discussion

The results of the measured activity concentrations of ^{40}K, ^{238}U and ^{232}Th in spring water samples with respect to the locations and distances from the sources of the springs are presented in Tables 1 – 4 while Figs. 1 – 4 shows the variations across the distances. The mean activity concentrations of the primordial radionuclides ^{40}K, ^{238}U and ^{232}Th were 59.9 ± 2.0, 7.9 ± 2.6 and 8.8 ± 2.6 Bq/ L respectively in Odo Ilawe natural spring water. In spring water samples from Shasha area, the mean activity concentrations of the radionuclides in the water samples were 155 ± 3.6, 6.9 ± 1.4 and 6.6 ± 0.8 Bq/ L respectively. The mean activity concentrations of the

radionuclides were 53.3 ± 2.1, 7.0 ± 1.5 and 6.4 ± 1.4 Bq/ L respectively in Gbemiso natural spring water, while 65.8 ± 13.3, 12.4 ± 4.0 and 8.7 ± 3.0 Bq/ L for the radionuclides in Ori – Oga natural spring water in the study area.

One way ANOVA of Statistical package of social sciences (SPSS) was used to assess the relationship between the radionuclides of the spring water samples for the various locations. The results in Tables 1 – 4 indicate clearly that there is no significant differences between the radionuclides across the distances (0 , 200 m, 400 m and 600 m) with p – values of 0.996, 1.00, 0.998 and 0.941 for Odo Ilawe, Shasha, Gbemiso and Ori Oga lacations respectively. This implies that the activity concentrations of the radionuclides in the spring water samples are not significantly elevated as the water run off on the surface soil, this may further suggest that the primordial radionuclides are uniformly distributed across the surface soil layer along the path of the streams.

However, this is expected since the locations in the study area are not known for mining activities which may enhance the activity concentrations of uranium and thorium. Mining activities may enhance the activity concentrations of natural radionuclides as reported by Bahari et al. (2007).

The samples were collected during summer. The activity concentrations of the dissolved substances in water depend on the season therefore, the concentration increases in summer due to the high evaporation rates and the increase of the solubility of the salts due to the higher temperatures of the water (Kehagia, et al., 2007). Consequently there may be an increase in the concentrations of the radionuclides in the spring water samples in summer than samples collected during winter, Samples of spring water collected in winter will be analyzed in another study.

Using Hotelling T^2 statistical package for activity concentration values of the radionuclides obtained in the spring water samples as presented in Tables 1 – 4 for the different locations: Odo Ilawe, Shasha, Gbemiso and Ori oga. The p – value value of 0.0023 ($p < 0.05$) indicate that there is significant difference in the activity concentrations of the radionuclides across the locations in the study area. This may be attributed to the slight variations in the geological formation, discharge rate and the transport of radionuclides in the liquid phase in the area.

Table 1: Mean and Range of Activity concentrations (Bq/ kg) of ^{40}K, ^{238}U and ^{232}Th in Odo –Ilawe natural spring water (at different distances from the source) in Ikere local government area, Ekiti State, Nigeria

	No of Samples	Activity concentration of ^{40}K (Bq/ L)	Activity concentration of ^{238}U (Bq/ L)	Activity concentration of ^{232}Th (Bq/ L)	Ann. Effective Dose (mSv/y)
Source	5	57.9 ± 0.7 (56.8 – 58.5)	5.5 ± 0.4 (5.0 – 5.9)	6.3 ± 0.3 (6.0 – 6.8)	0.15
200m	5	58.9 ± 0.4 (58.5 – 59.4)	6.3 ± 0.4 (6.0 – 6.9)	7.4 ± 0.3 (7.0 – 7.8)	0.18
400m	5	60.4 ± 0.3 (60.1 – 60.8)	8.3 ± 0.2 (8.0 – 8.6)	9.0 ± 0.2 (8.8 – 9.2)	0.21
600m	5	62.5 ± 0.4 (62.0 – 63.0)	11.4 ± 0.6 (10.5 – 11.8)	12.3 ± 0.9 (11.5 – 13.8)	0.28
Mean/ Range	20	59.9 ± 2.0 (56.8 – 63.0)	7.9 ± 2.6 (5.0 – 11.8)	8.8 ± 2.6 (6.0 – 13.8)	0.21 (0.15 – 0.21)

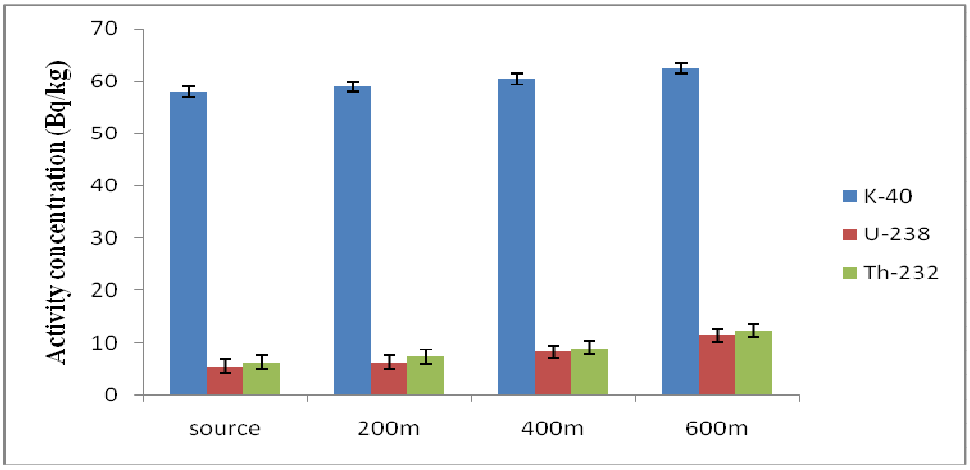

Fig. 1: The activity concentrations of the radionuclides in Odo Ilawe natural spring water against the different distances from the source

Table 2: Mean and Range of Activity concentrations (Bq/ kg) of ^{40}K, ^{238}U and ^{232}Th in Shasha natural spring water (at different distances from the source) in Ikere local government area, Ekiti State, Nigeria

	No of Samples	Activity concentration of ^{40}K (Bq/ L)	Activity concentration of ^{238}U (Bq/ L)	Activity concentration of ^{232}Th (Bq/ L)	Ann. Effective Dose (mSv/y)
Source	5	150.4 ± 0.3 (150.1 – 150.8)	5.2 ± 0.2 (5.0 – 5.5)	5.5 ± 0.3 (5.1 – 5.8)	0.18
200m	5	154.6 ± 0.4 (154.2 – 154.8)	6.4 ± 0.4 (5.9 – 6.8)	6.3 ± 0.3 (6.0 – 6.7)	0.20
400m	5	156.5 ± 0.2 (156.2 – 156.8)	7.5 ± 0.3 (7.1 – 7.8)	7.0 ± 0.2 (6.8 – 7.2)	0.22
600m	5	158.8 ± 0.7 (158.0 – 160)	8.5 ± 0.4 (8.0 – 9.0)	7.4 ± 0.3 (7.0 – 7.8)	0.23
Mean/ Range	20	155 ± 3.6 (150.1 – 160.0)	6.9 ± 1.4 (5.0 – 9.0)	6.6 ± 0.8 (5.1 – 7.8)	0.21 (0.18 – 0.23)

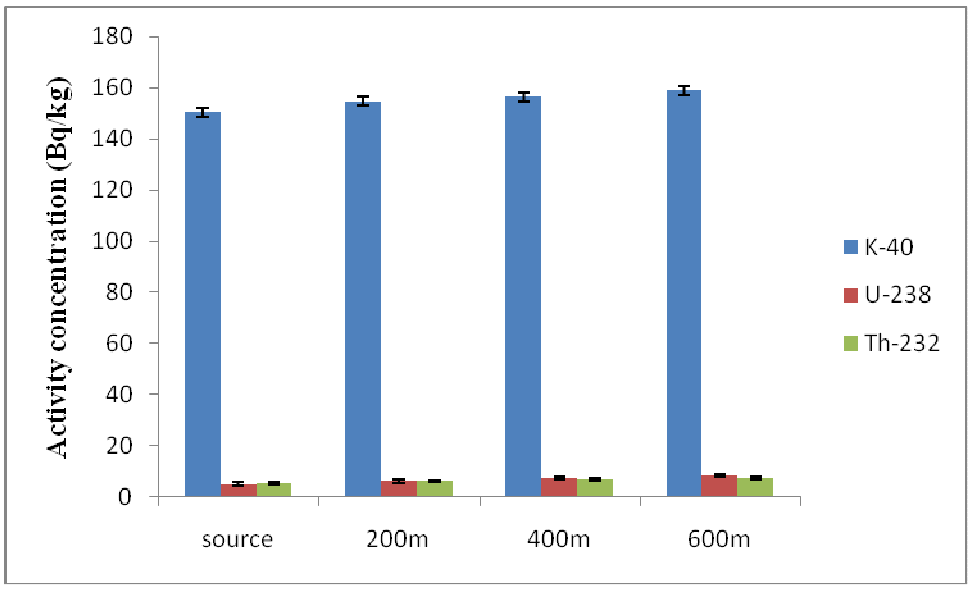

Fig. 2: The activity concentrations of the radionuclides in Shasha natural spring water against the different distances from the source

Table 3: Mean and Range of Activity concentrations (Bq/ kg) of ^{40}K, ^{238}U and ^{232}Th in Gbemiso natural spring water (at different distances from the source) in Ikere local government area, Ekiti State, Nigeria

	No of Samples	Activity concentration of ^{40}K (Bq/ L)	Activity concentration of ^{238}U (Bq/ L)	Activity concentration of ^{232}Th (Bq/ L)	Ann. Effective Dose (mSv/y)
Source	5	50.7 ± 0.5 (50.2 – 51.4)	5.3 ± 0.2 (5.0 – 5.6)	5.1 ± 0.2 (4.9 – 5.4)	0.13
200m	5	52.7 ± 0.3 (52.3 – 53.0)	6.4 ± 0.3 (6.0 – 6.8)	5.8 ± 0.2 (5.6 – 5.9)	0.15
400m	5	54.4 ± 0.4 (53.9 – 54.8)	7.4 ± 0.3 (7.0 – 7.8)	6.3 ± 0.3 (6.0 – 6.7)	0.16
600m	5	55.5 ± 0.4 (55.0 – 56.0)	8.8 ± 0.3 (8.3 – 9.2)	8.3 ± 0.3 (8.0 – 8.6)	0.20
Mean/Range	20	53.3 ± 2.1 (50.2 – 56.0)	7.0 ± 1.5 (5.0 – 9.2)	6.4 ± 1.4 (4.9 – 8.6)	0.16 (0.13 – 0.20)

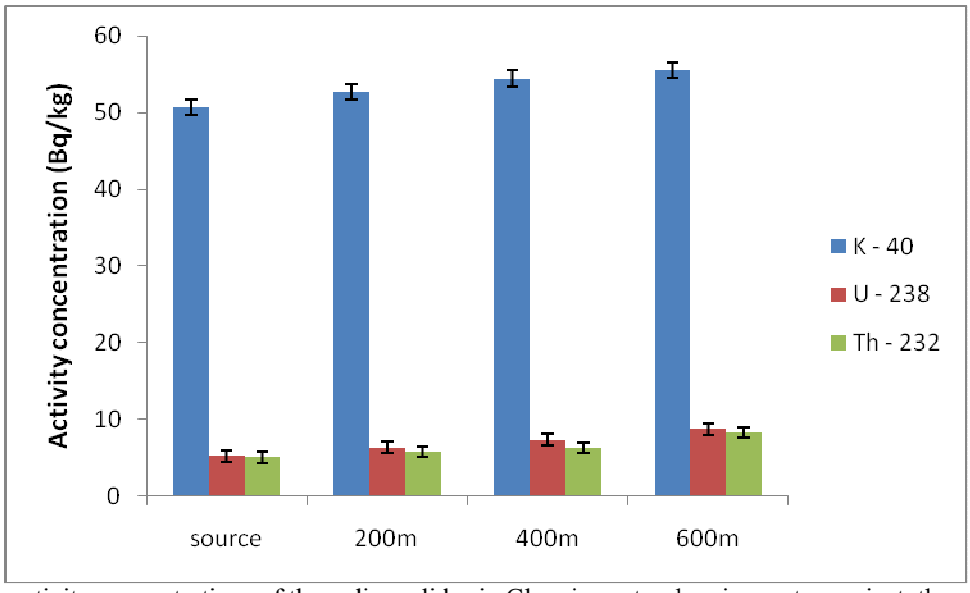

Fig. 3: The activity concentrations of the radionuclides in Gbemiso natural spring water against the different distances from the source

Table 4: Mean and Range of activity concentrations (Bq/ kg) of ^{40}K, ^{238}U and ^{232}Th in Ori –Oga natural spring water (at different distances from the source) in Ikere local government area, Ekiti State, Nigeria

	No of Samples	Activity concentration of ^{40}K (Bq/ L)	Activity concentration of ^{238}U (Bq/ L)	Activity concentration of ^{232}Th (Bq/ L)	Ann. Effective Dose (mSv/y)
Source	5	55.3 ± 0.3 (55.0 – 55.8)	8.0 ± 1.2 (7.0 – 9.5)	5.5 ± 0.4 (5.0 – 6.0)	0.15
200m	5	58.7 ± 0.4 (58.1 – 59.2)	11.0 ± 0.4 (10.4 – 11.5)	7.3 ± 0.4 (7.0 – 7.7)	0.19
400m	5	64.3 ± 3.2 (60.3 – 68.6)	13.2 ± 0.5 (12.5 – 13.8)	9.4 ± 0.3 (9.0 – 9.8)	0.24
600m	5	84.9 ± 3.6 (80.4 – 89.5)	17.5 ± 0.4 (17.0 – 17.9)	12.5 ± 0.8 (11.2 – 13.4)	0.31
Mean/Range	20	65.8 ± 13.3 (55.0 – 89.5)	12.4 ± 4.0 (7.0 – 17.9)	8.7 ± 0.3 (5.0 – 13.4)	0.22 (0.15 – 0.31)

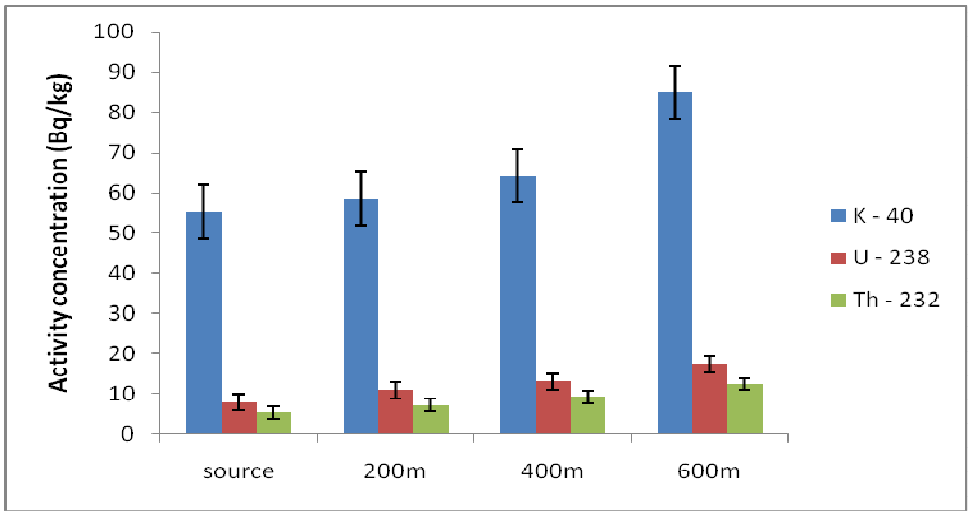

Fig. 4: The activity concentrations of the radionuclides in Ori – Oga natural spring water against the different distances from the source

The mean and range of activity concentrations of the natural radionuclides in the spring water samples across the locations in Ikere local government area considered in this study and the mean annual effective doses

are presented in Table 5. The mean activity concentration of ^{40}K in the local government area is 83.5 ± 48 Bq/L with the highest value of 155.0 ± 3.6 Bq/L from samples collected from Shasha, while the lowest value of 53.3 ± 2.1 Bq/L was recorded from Gbemiso location. The mean activity concentration of ^{238}U in the local government area is 8.6 ± 2.6 Bq/L with the highest value of 12.4 ± 4.0 Bq/L from samples collected from Ori – Oga, while the lowest value of 6.9 ± 1.4 Bq/L was obtained from Shasha location. The mean activity concentration of ^{232}Th in the local government area is 7.6 ± 1.3 Bq/L with the highest value of 8.8 ± 2.6 Bq/L from samples collected from Odo Ilawe, while the lowest value of 6.4 ± 1.4 Bq/L was recorded from Gbemiso location.

The results show low degree of variations in the activity concentrations of the radionuclides since the locations in the study area lie in the same basement complex that consists of an assemblage of crystalline igneous and metamorphic rocks. The mean activity concentrations of ^{232}Th recorded in the spring water from the study area are higher than the World Health Organization (WHO) maximum acceptable concentration of 0.6 Bq/L (WHO, 2004). The higher value may be due to the longer time of the run off on the bedrocks and the surface soil, during which minerals dissolve in the spring water. Moreso the mineralogical and geochemical composition of the soil and rock, chemical composition of water, degree of weathering of the rock and redox conditions also contribute to high activity concentrations [10].

The annual committed effective dose to an adult due to ingestion of the radionuclides in the spring water were calculated using equation 2. The annual committed effective doses in Ikere local government , Ekiti State, Nigeria varied from 0.16 mSv/y to 0.22 mSv/y with a mean value of 0.20 ± 0.03 mSv/y. The calculated annual committed effective dose is in line with World Health Organization (WHO) recommended limit of 1.0 mSv/y for public exposure [14]. The calculated mean annual committed effective dose is also lower than the ICRP recommended public dose limit of 1.0 mSv/y. Consumption of natural water from the streams in the study area does not pose any radiological burden to the village residents.

Table 5: Mean and Range of activity concentrations (Bq/ kg) of ^{40}K, ^{238}U and ^{232}Th natural spring water in Ikere local government area, Ekiti State, Nigeria

Location	No of Samples	Activity concentration of ^{40}K (Bq/ kg)	Activity concentration of ^{238}U (Bq/ kg)	Activity concentration of ^{232}Th (Bq/ kg)	Ann. Effective Dose (mSv/y)
Odo Ilawe	20	59.9 ± 2.0 (56.8 – 63.0)	7.9 ± 2.6 (5.0 – 11.8)	8.8 ± 2.6 (6.0 – 13.8)	0.21 (0.15 – 0.21)
Shasha	20	155 ± 3.6 (150.1 – 160.0)	6.9 ± 1.4 (5.0 – 9.0)	6.6 ± 0.8 (5.1 – 7.8)	0.21 (0.18 – 0.23)
Gbemiso	20	53.3 ± 2.1 (50.2 – 56.0)	7.0 ± 1.5 (5.0 – 9.2)	6.4 ± 1.4 (4.9 – 8.6)	0.16 (0.13 – 0.20)
Ori – Oga	20	65.8 ± 13.3 (55.0 – 89.5)	12.4 ± 4.0 (7.0 – 17.9)	8.7 ± 0.3 (5.0 – 13.4)	0.22 (0.15 – 0.31)
Mean/Range	80	83.5 ± 48.0 (59.9 – 155.0)	8.6 ± 2.6 (6.9 – 12.4)	7.6 ± 1.3 (6.4 – 8.8)	0.20 ± 0.03 (0.16 – 0.21)

Conclusion

Spring water has been the only source of drinking water for rural dwellers and mostly farmers. The determined activity concentrations of the primordial radionuclides are slightly higher than the values obtained in literatures [10]. The resulting effect on the annual commited effective dose of 0.20 ± 0.03 mSv/y due to intake of the radionuclides by an adult via spring water is lower than 1.0 mSv/y the recommended public dose by WHO and ICRP. The consumption of spring water may not pose any radiological health hazards to the public within the study area.

Further studies should be carried out to cover the entire Ekiti State, this should include epidemiological survey of incidences of radiation related health effects to obtain a baseline data for future reseach work. Results from these studies may be important for formulating local regulations for radioactivity levels in drinking water.

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