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Assessment of Radiological Hazard of Natural Radioactivity in Drinking Water in Ondo, Nigeria

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

The paper determines the natural activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in water samples from various ponds and rivers in some parts of Ondo State, Nigeria. The measurement was done by gamma-ray spectrometry system with high-purity germanium detector. The activity concentrations in ponds for ^{226}Ra, ^{232}Th and ^{40}K varied from (0.15±0.01 to 0.82±0.04 Bq/l), (0.52±0.10 to 1.64±0.20 Bq/l) and (0.25±0.05 to 3.60±0.21 Bq/l) respectively and the activity concentration in rivers varied from (0.17±0.01 to 0.23±0.04 Bq/l), (0.56±0.02 to 1.38±0.20 Bq/l) and (0.46±0.02 to 5.38±0.10 Bq/l) respectively. The results of the calculated annual effective doses for different age groups due to ingestion and the health implication with respect to the hazard limit set by the International Commission on Radiological Protection (ICRP) were considered.

Keywords: Groundwater; natural radioactivity; HPGe detector; cancer risk; severe hereditary effect.

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1. INTRODUCTION

Naturally occurring radionuclides can present very harmful biological effect to human cells and tissues as a result of continuous ingestion. Exposure to ionizing radiation causes damage to living tissue, and can result in mutation, cancer, and death. Of particular concern are naturally-occurring uranium and radium, which can accumulate to harmful levels in drinking water. As radionuclides decay, they emit radioactive particles such as alpha particles, beta particles and gamma rays. Each type of particle produces different effects on humans. Radionuclides usually enter drinking water through natural erosion and chemical weathering of mineral deposits [1]. The maximum permissible dose for the general public is set as 1 mSv/y by the International Commission on Radiological Protection (ICRP).

The earth is radioactive and humans are continuously irradiated by sources outside and inside their bodies. Outside sources include space and terrestrial radiations while inside sources are from the radionuclides that enter our bodies in the food and water people ingest and the air they breathe. The human body cannot sense exposure to radiation directly except at levels that are invariably lethal, and at these levels the human body cannot provide defence against it [2]. Due to the severity of this problem, the acceptable levels of radiation exposure and consequently radiation doses (maximum permissible dose) have been set by various bodies, based on research findings in this field.

Various research works have been carried out by scientists on the measurement of radionuclide concentration in both surface and underground waters of different areas of the world [3]. The radionuclide concentration levels of ^{40}K, ^{238}U and ^{232}Th in soil and water samples around three cement manufacturing companies in Port Harcourt, South-South Nigeria were measured [4]. The mean activity concentration of ^{40}K, ^{238}U and ^{232}Th measured in Bq/kg for the soil samples were found to be 473.95±165.27, 49.90±17.34 and 5.51±1.72 respectively, while the mean activity concentration for the water samples, measured in Bq/l were found to be 0.362±0.19, 48.29±12.07 and 0.038±0.030 respectively. The radionuclides concentration in underground and drinking waters in some areas in Upper Egypt were measured using gamma ray spectrometry with Hyper-pure Germanium detector [5]. The analyzed waters differ in radioactivity concentration, depending on their origin and places. In drinking water in Qena, Upper Egypt, the mean value of ^{226}Ra concentrations was 1.32 ± 0.70 pCi/l, while in ground water in Safaga and Qusier in the Red Sea region where there are phosphate mines, ^{226}Ra and ^{232}Th mean values were 3.05 ± 0.90 and 1.39 ± 0.60 pCi/l respectively. The mean annual effective dose taken into the body by the populace drinking the tap water was found to be 0.008 mSv, which is lower than the limit recommended by the World Health Organisation [6,7]. Nwankwo measured natural radioactivity in groundwater in Tanke-Ilorin, Nigeria [8]. The activity concentration values ranged from 0.81±0.08 to 7.40±2.20 Bq/l for ^{226}Ra, 1.80±0.30 to 5.60±2.60 Bq/l for ^{228}Ra respectively. The mean contribution of both ^{226}Ra and ^{228}Ra activities to the committed effective dose from a year's consumption of drinking water in the study area was, therefore, higher than the tolerable level of 1 mSv/y to the general public for prolonged exposure as recommended by International commission on Radiological Protection (ICRP). The levels of radionuclide activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in various sources of drinking water in some parts of Ondo State, Southwestern, Nigeria was measured by [9]. The activity concentrations obtained for ^{226}Ra, ^{232}Th and ^{40}K varied from 0.20±0.01 to 1.12±0.10 Bq l^{-1}, 0.60±0.04 to 1.45±0.06 Bq l^{-1}, 0.72±0.04 to 8.02±0.35 Bq l^{-1}, 0.18±0.01 to 1.06±0.14 Bq l^{-1}, 0.52±0.04 to 1.04±0.05 Bq l^{-1}, 0.94±0.06 to 5.90±0.15 Bq l^{-1}, 0.15±0.01 to 0.34±0.03 Bq l^{-1}, 0.52±0.04 to 0.90±0.04 Bq l^{-1}, 0.37±0.09 to 4.95±0.12 Bq l^{-1} in dug-well, borehole and stream respectively. These values when compared with the ICPR standard were found to be very low.

This present work therefore, intends to study comprehensively and measure the radionuclide concentrations in drinking water in some parts of Ondo State, Southwestern, Nigeria. The major source of drinking water in these areas is from ponds and rivers. The annual effective doses resulting from the ingestion and the radioactivity level of the water are considered within tolerable limits in order to check health hazards among the populace.

2. MATERIALS AND METHODOLOGY

2.1 Sample Collection and Preparation

Twelve (12) samples of drinking water from ponds and rivers were collected from selected Local Government Areas in Ondo State which

include: Akure-South, Akure-North, Idanre and Ifedore Local Government Areas respectively. A1 to A7 represent water samples from pond and B1 to B5 represent water samples from river. The geographical locations are as shown in Fig. 1 for pond and river samples. The water samples are collected using 1-litre plastic containers after they had been thoroughly rinsed with distilled water. Two drops of 1M hydrochloric acid was immediately added to the water samples after collection to prevent adherence of the radionuclides on the wall of the container. For activity concentration measurement, the water samples were prepared into 1 litre Marinelli beakers and sealed hermetically. The samples are kept for about 30 days to establish secular equilibrium between the radionuclides and their daughter products [10].

2.2 Instrumentation

The activity concentrations of radionuclides in the water samples were determined by a non-destructive analysis using a computerized gamma ray spectrometry system with high-purity germanium (HPGe) detector. The relative efficiency of the detector system was 25%, and resolution was 1.8 keV at 1.33MeV of ^{60}Co. The gamma spectrometer was coupled to conventional electronics connected to a multichannel analyzer card (MCA) installed in a desk top computer. A software program called MAESTRO- 32 was used to accumulate and analyze the data and to calculate the natural radioactivity concentrations in the samples.

2.3 Activity Concentration and Analysis

The detector was located inside a cylindrical lead shield of 5 cm thickness with internal diameter of 24 cm and height of 60 cm. A counting time of 36,000 seconds (10 h) was used to acquire spectra data for each sample. The energy and efficiency calibrations of the spectrometer were carried out using standard water sources emitting gamma rays in the energy range 200-1500KeV, covering all gamma energies of radionuclides of interest. The activity concentrations of the uranium-series were determined using γ -ray emission of ^{214}Pb at 351.9 keV (35.8%) and ^{214}Bi at 609.3 keV (44.8%) for ^{226}Ra, and for the ^{232}Th-series, the emission of ^{228}Ac at 911 keV (30.1%) were used as showed in Figs. 2 and 3. The ^{40}K activity concentration was determined directly from its emission line at 1460.8 keV.

2.4 Calculation of Activity Concentration

The specific activity concentrations (A) of the radionuclides in the water samples were determined using the following expression [10].

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

where N_{sam} is the net counts of the radionuclides in the samples, P_E is the Gamma ray emission probability (gamma yield), $\boldsymbol{\varepsilon}$ is the total counting efficiency of the detector system, T_c is the sample counting time and M is the Volume (L).

2.5 Calculation of Annual Effective Dose

Estimation of Annual Effective Dose (E_d) to different age groups due to the consumption of radionuclide present in water samples from different sources was done using the following relations:

$$E_d = A_c A_i C_f \quad (2)$$

Where A_c is the activity concentration of the radionuclide in the water samples (Bq l^{-1}), A_i is annual intake of drinking water sample (l y^{-1}), C_f = ingested dose conversion factor for radionuclides (Sv Bq^{-1}) by [10]. The total effective dose D (Sv Bq^{-1}) to an individual was established by summing contributions from all radionuclides present in the water samples i.e.

$$D = \sum A_c A_i C_f \quad (3)$$

The annual effective dose was calculated for the ICRP age groups with annual average water intake of 200, 260, 300, 350, 600 and 730 litres respectively. The conversion factors used in the estimations were taken from [9,11].

2.6 Calculation of Cancer Risk and Hereditary Effects

The risk to population was estimated using the annual effective dose and the 2007 recommended risk coefficients in ICRP report and assumed 70 years lifetime of continuous exposure of population to low level radiation [12]. According to ICRP methodology [13]:

Cancer Risk = Total Annual Effective Dose (mSv/y) x Cancer Risk Factor (4)

Hereditary Effects = Total Annual Effective Dose (mSv/y) x Hereditary Effect Factor (5)

For fatal cancer risk, the coefficient in 2007 Recommendations of the ICRP for members of the public 5.5 x 10^{-2} was used in equation (4). For hereditary effect, the detriment adjusted risk coefficient for the whole population as stated in [12] for stochastic effects after exposure at low dose rate 0.2 x 10^{-2} was used in equation (5).

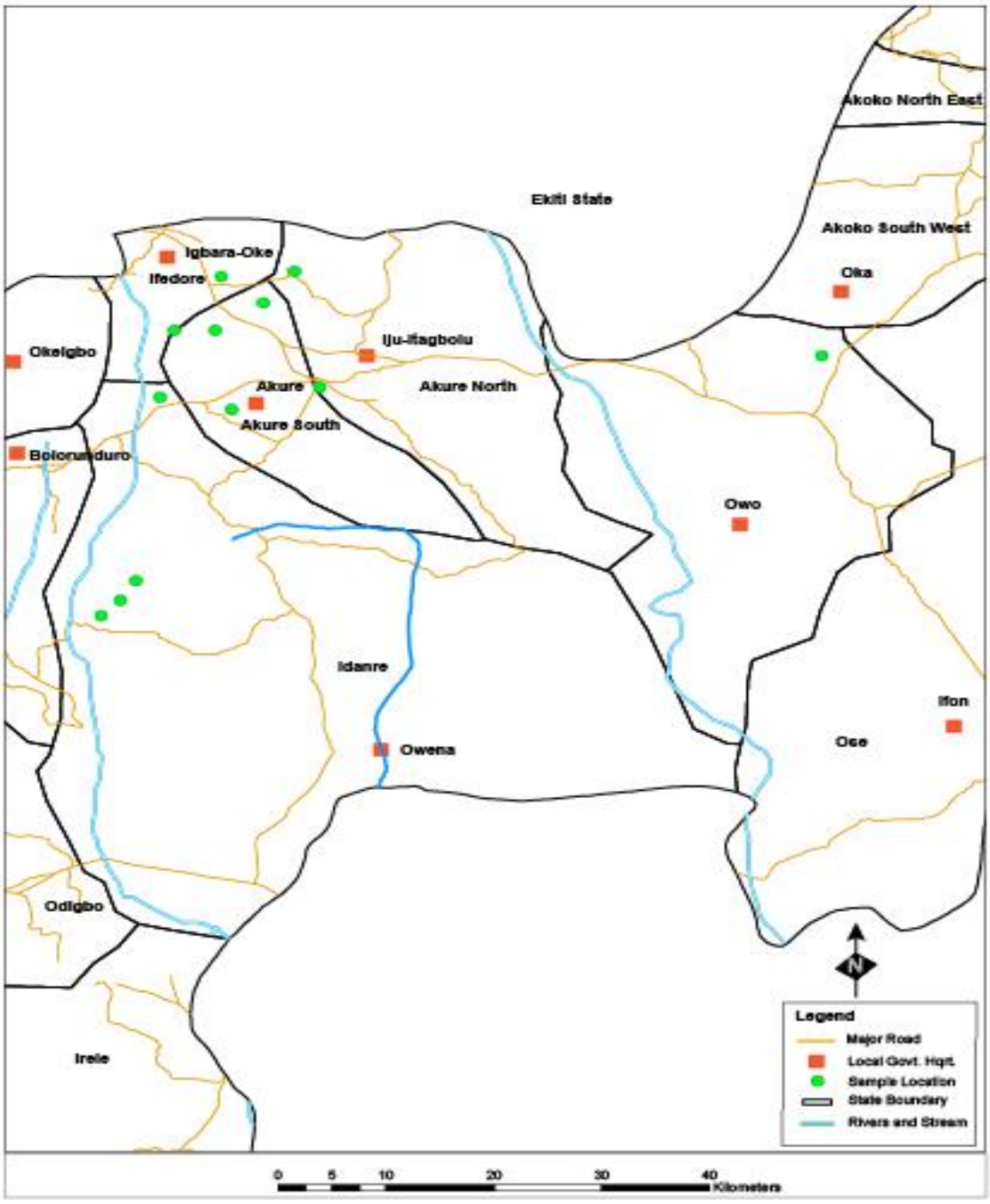

Fig. 1. Geological Map of Study Area showing sample locations

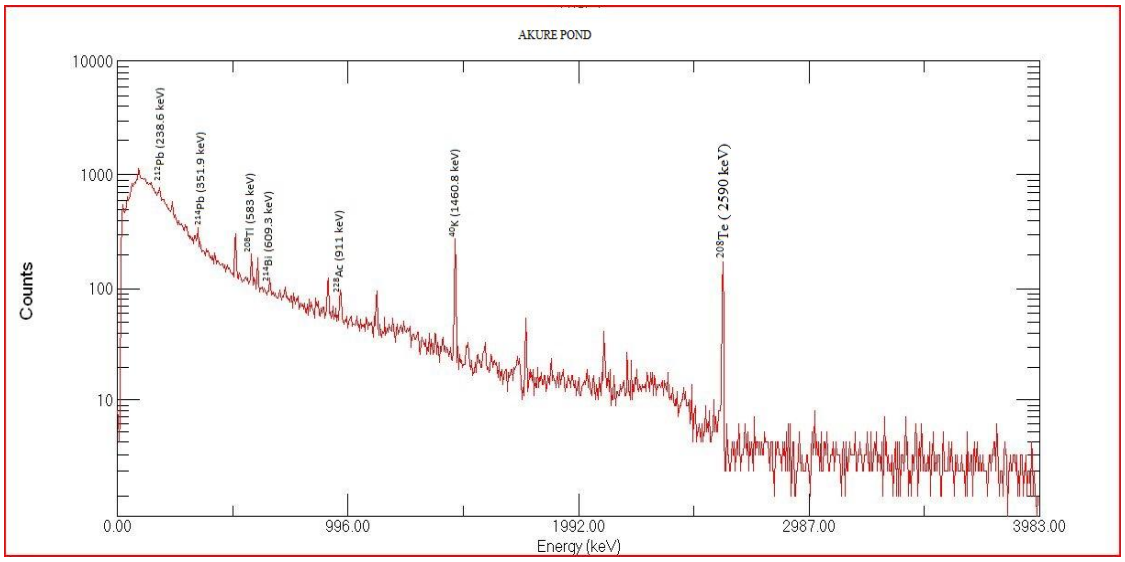

Fig. 2. Gamma ray spectrum of drinking water sample (pond)

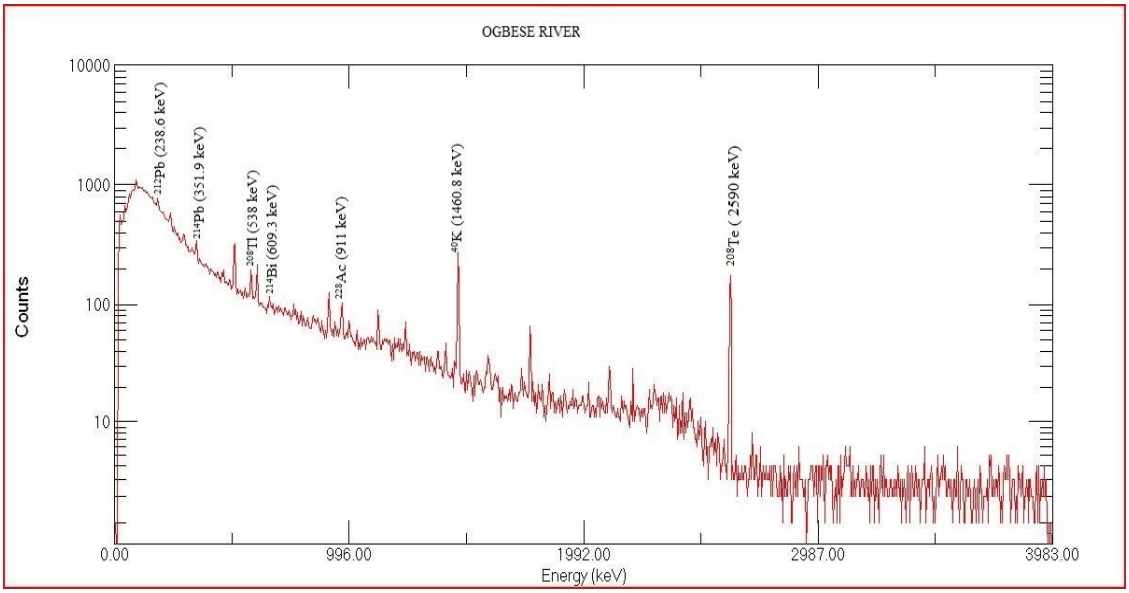

Fig. 3. Gamma ray spectrum of drinking water sample (river)

3. RESULTS AND DISCUSSION

The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in water from pond and river sources in some parts of Ondo State, Southwestern, Nigeria are presented in Tables 1 and 2. The annual effective doses (mSv/y) for different age categories (for pond and water samples) are presented in Tables 3 and 4. The estimated cancer risk and hereditary effects for adults are presented in Tables 5 to 6. Fig. 4 shows the bar graph of the total annual effective doses to the six age groups from pond water consumption. Fig. 5 shows the contribution of each pond water sample to the total annual effective doses. Fig. 6 shows the bar graph of the total annual effective doses to the six age groups from river water consumption. Fig. 7 shows the contribution of each river water sample to the total annual effective doses.

As seen in Pond, the activity value of ^{40}K concentration varied from 0.25±0.05 to 3.60±0.21 Bq/l with an average value of 2.04 Bq/l. The specific activity concentration of ^{226}Ra ranged from 0.15±0.01 to 0.82±0.04 Bq/l with an average value of 0.34 Bq/l. The activity concentration of ^{232}Th varied from 0.52±0.10 to 1.64±0.20 Bq/l with an average value of 0.92 Bq/l. In River, the activity values of ^{40}K concentration varied from 0.46±0.02 to 5.38±0.10 Bq/l. The specific activity concentration of ^{226}Ra ranged from 0.17±0.01 to 0.23±0.04 Bq/l with an average value of 0.20 Bq/l. The activity concentration of ^{232}Th varied between 0.56±0.02 to 1.38±0.20 Bq/l with an average value of 0.92 Bq/l.

This variation in activity concentration of ^{40}K, ^{226}Ra and ^{232}Th observed in these samples indicates that the origins of these waters are not the same and that they come from different depths and pass through different geological layers [13]. However, these irregular distributions of activity concentrations of the selected nuclides in this water from different sources depend on the geological formation of the area where the waters are located. Tables 1 and 2 show the activity concentrations of the radionuclides detected in water samples from the pond and river respectively in which the concentration of ^{40}K was the highest follow by ^{232}Th, and ^{226}Ra was the least. Thus ^{40}K contributed the largest activity concentration while ^{226}Ra contributed least activity in all the water samples from different sources. The ± values associated with the mean values represented the variability (standard deviation) in the activity concentration values of the radionuclides [14].

When comparing these data, the activity concentration of ^{226}Ra in the water samples varies in agreement with reports by many authors. The activity concentration of ^{226}Ra in water from different sources in Nigeria also varies. This wide range of ^{226}Ra concentration may be due to the geological structure of the areas.

The activity concentrations of ^{226}Ra in all the samples do not exceed the limit of 1.00 Bq/l set by the International Atomic Energy Agency [15].

The total annual effective doses to the six age groups from the pond considered in this study are presented in Table 3 and pictorially in Fig. 4. The figure shows that babies (0-1 y old) are most exposed internally to radium in the water samples followed by the adult (12-17 y old) and the highest exposure is from Ibule-Soro pond while the least is from Igoba as shown in Fig. 5. The total annual effective doses to the six age groups from the river considered in this study are also presented in Table 4 and pictorially in Fig. 6.

The figure shows that babies (0-1 y old) are most exposed internally to radium in the water samples followed by the adult (12-17 y old) and the highest exposure is from Aponmu while the least is from Ipogun river as shown in Fig. 7. Fig. 8 compares the total annual effective dose from different drinking water sources. The pond water sample has higher value than the river. Radium is highly radiotoxic and it builds up in the growing bones of babies and children where it can cause bone cancer.

The calculated radiation doses for different age groups due to consumption of water from the pond were ranged from 0.42 to 1.26 mSv/y with the average value of 0.70 mSv/y for 0-1 y, 0.12 to 0.34 mSv/y with the average value of 0.21 mSv/y for 1-2 y, 0.10 to 0.27 mSv/y with the average value of 0.16 mSv/y for 2-7 y, 0.12 to 0.33 mSv/y with an average value of 0.19 mSv/y for 7-12 y, 0.27 to 0.89 mSv/y with the average value of 0.46 mSv/y for 12-17 y, 0.14 to 0.39 mSv/y with the average 0.23 mSv/y for >17 y, respectively. The calculated radiation dose for different age groups due to consumption of water from river were ranged from 0.46 to 0.67 mSv/y with the average value of 0.55 mSv/y for 0-1 y, 0.16 to 0.24 mSv/y with the average value of 0.19 mSv/y for 1-2 y, 0.12 to 0.19 mSv/y with the average value of 0.14 mSv/y for 2-7 y, 0.13 to 0.21 mSv/y with an average value of 0.16 mSv/y for 7-12 y, 0.27 to 0.38 mSv/y with the average value of 0.33 mSv/y for 12-17 y, 0.15 to 0.28 mSv/y with the average 0.20 mSv/y for >17 y, respectively. It can be seen that radiation doses received by 0-1 y (babies) are relatively higher than that received by the other age groups. From the analysis, the age group with the highest exposure dose is 0-1 y (babies) followed by the 12-17 y age group. Following the ICRP recommendation, the recommended reference levels of the effective dose corresponding to one year consumption of drinking water is 1.0 mSv/y.

The dose obtained in the present study is lower than the recommended reference level and from radiation protection point of view; life-long consumption of these investigated sources of water may not cause any significant radiological health risk.

In order to evaluate the radiation risk due to ingestion of the selected radionuclides, the ICRP methodology was adopted in this study and the results are shown in Tables 5 and 6 for pond and river waters respectively. The results of the cancer and non-cancer risk components were evaluated from the estimated annual effective dose of the water from the different sources.

The results of the evaluated fatal cancer risk to adult (>17) per year in each of the water sample from the ponds ranged from 0.77×10^{-5} to 2.15×10^{-5} with the associated lifetime fatality cancer risk of 0.54×10^{-3} to 1.50×10^{-3} per year. The results of the evaluated fatal cancer risk to adult (>17) per year in each of the water sample from the rivers ranged from 0.83×10^{-5} to 1.54×10^{-5} with the associated lifetime fatality cancer risk of 0.58×10^{-3} to 1.08×10^{-3} per year. The evaluated severe hereditary effect to adult (>17) per year in each of the water sample from the river varied from 3.0×10^{-7} to 5.60×10^{-7} with the associated lifetime hereditary effect in adult of 2.10×10^{-5} to 3.92×10^{-5} per year.

Furthermore, in terms of lifetime fatality cancer risk to adult, about 1 out of 1000 may suffer from some form of cancer fatality risk and for the lifetime hereditary effects; about 5 out of 100,000 may suffer some hereditary effects by drinking water from the pond in the study area and about 3 out of 100,000 may suffer some hereditary effects by drinking water from the river.

The negligible cancer fatality risk value recommended by United State environmental Protection Agency [3] is in the range of 1.0×10^{-6} to 1.0×10^{-4} [13] meaning, one person out of one million or one person out of ten thousand. Comparing the estimated results of the lifetime fatality risk in the present study with the acceptable risk factor, it can be concluded that, all estimated results of the life time fatality risk in adult member of some parts of Ondo State population due to ingestion of radionuclides in water from different sources are above the range of acceptable risk values recommended by USEPA.

Table 1. Activity concentrations (Bq/l) of ^{40}K, ^{226}Ra and ^{232}Th in pond water samples

Sample ID	Pond Sample	Sample location		^{226}Ra	^{232}Th	^{40}K
		Lat. (N)	Long. (E)			
A1	Oda	7.23°	5.12°	0.54 ± 0.09	1.64 ± 0.20	0.25 ± 0.05
A2	Akure	7.25°	5.19°	0.15 ± 0.01	0.97± 0.12	3.60 ± 0.21
A3	Ilara	7.35°	5.11°	0.20 ± 0.04	0.85 ± 0.02	0.46 ± 0.02
A4	Ijare	7.36°	5.17°	0.28 ± 0.04	0.83 ± 0.05	2.18 ± 0.08
A5	Igoba	7.01°	5.01°	0.20 ± 0.02	0.52 ± 0.10	1.74 ± 0.05
A6	Ibule- Soro	7.31°	5.10°	0.82 ± 0.04	0.95 ± 0.03	2.55 ± 0.02
A7	Ipinsa	7.33°	5.15°	0.22 ± 0.03	0.65 ± 0.03	3.47 ± 0.10
Average				0.34	0.92	2.04
Standard deviation				0.25	0.36	1.33
Range				0.15-0.82	0.52-1.64	0.25-3.60

Table 2. Activity concentrations (Bq/l) of ^{40}K, ^{226}Ra and ^{232}Th in River water samples

Sample ID	River Sample	Sample location		^{226}Ra	^{232}Th	^{40}K
		Lat. (N)	Long. (E)			
B1	Ogbese	7.02°	5.02°	0.22 ± 0.05	0.94 ± 0.08	1.95 ± 0.12
B2	Aponmu	7.24°	5.06°	0.23 ± 0.04	0.85 ± 0.02	0.46 ± 0.02
B3	Ipogun	7.31°	5.08°	0.17 ± 0.01	1.38 ± 0.20	3.65 ± 0.08
B4	Owene	7.03°	5.03°	0.19 ± 0.03	0.88 ± 0.06	1.55 ± 0.04
B5	Omifunfun	7.27°	5.61°	0.19 ± 0.02	0.56 ± 0.02	5.38 ± 0.10
Average				0.20	0.92	2.59
Standard deviation				0.02	0.29	1.93
Range				0.17-0.23	0.56-1.38	0.46-5.38

Table 3. Total annual effective doses (mSv/y) for different age categories (pond water samples)

Sample ID	0-1 y	1-2 y	2-7 y	7-12 y	12-17 y	>17 y
A1	1.14	0.32	0.27	0.32	0.74	0.39
A2	0.51	0.19	0.15	0.15	0.31	0.20
A3	0.50	0.15	0.13	0.14	0.31	0.18
A4	0.60	0.18	0.14	0.17	0.38	0.21
A5	0.42	0.12	0.10	0.12	0.27	0.14
A6	1.26	0.34	0.26	0.33	0.89	0.34
A7	0.50	0.16	0.10	0.15	0.29	0.17
Average	0.70	0.21	0.16	0.19	0.46	0.23

Table 4. Total annual effective doses (mSv/y) for different age categories (river water samples)

Sample ID	0-1 y	1-2 y	2-7 y	7-12 y	12-17 y	>17 y
B1	0.57	0.18	0.14	0.16	0.34	0.20
B2	0.53	0.16	0.12	0.14	0.34	0.19
B3	0.67	0.24	0.19	0.21	0.38	0.28
B4	0.52	0.17	0.14	0.15	0.31	0.2
B5	0.46	0.18	0.13	0.13	0.27	0.15
Average	0.55	0.19	0.14	0.16	0.33	0.20

Table 5. Estimated cancer risk and hereditary effects for adult from pond water samples

Sample ID	Fatality Cancer Risk to Adult per year (10^{-5})	Lifetime Fatality Cancer Risk to Adult (10^{-3})	Severe Hereditary effect in Adult per year (10^{-7})	Estimated Lifetime Hereditary effect in Adult (10^{-5})
A1	2.15	1.50	7.8	5.46
A2	1.10	0.77	4.0	2.80
A3	0.99	0.69	3.6	2.52
A4	1.16	0.81	4.2	2.94
A5	0.77	0.54	2.8	1.96
A6	1.87	1.31	6.8	4.76
A7	0.94	0.65	3.4	2.38

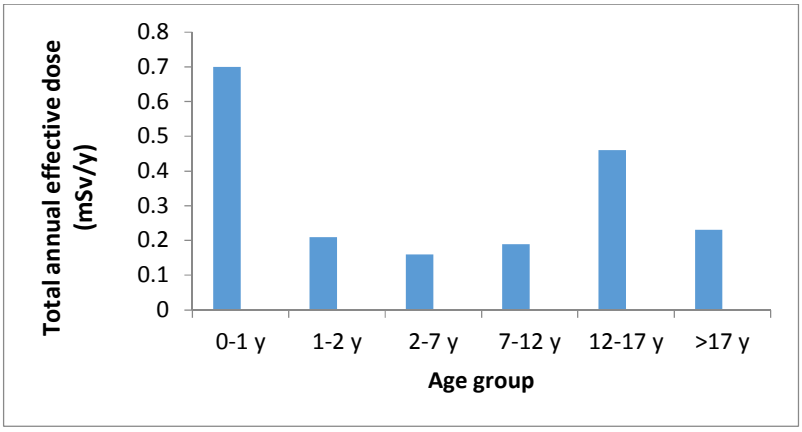

Fig. 4. Total annual effective dose (mSv/y) to the six age groups from Pond

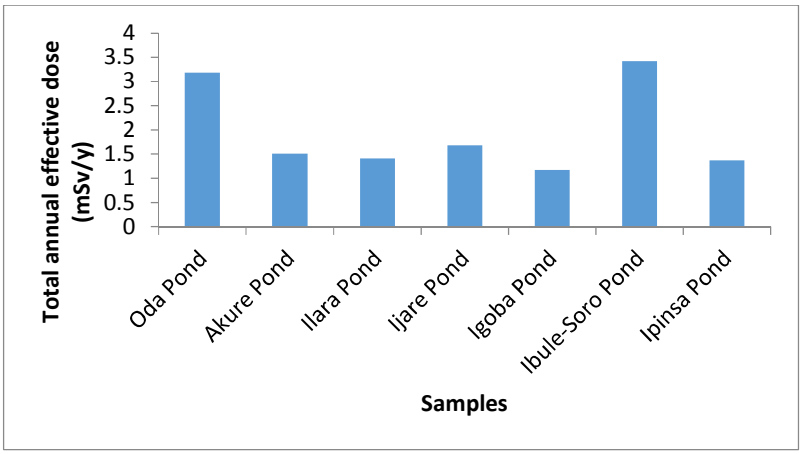

Fig. 5. Comparison of total annual effective dose (mSv/y) from different samples (Pond)

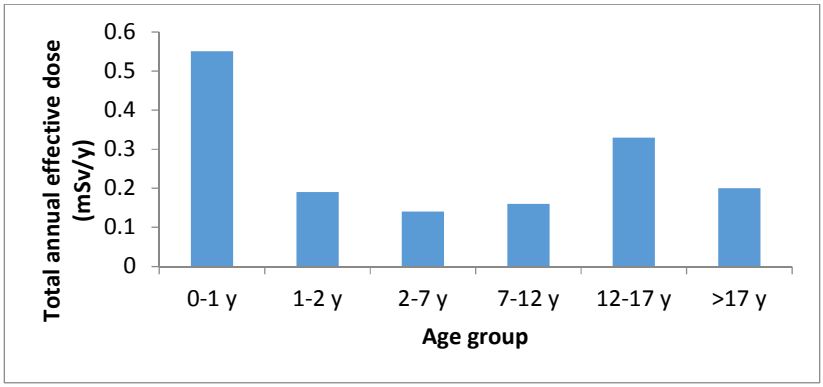

Fig. 6. Total annual effective dose (mSv/y) to the six age groups from Rivers

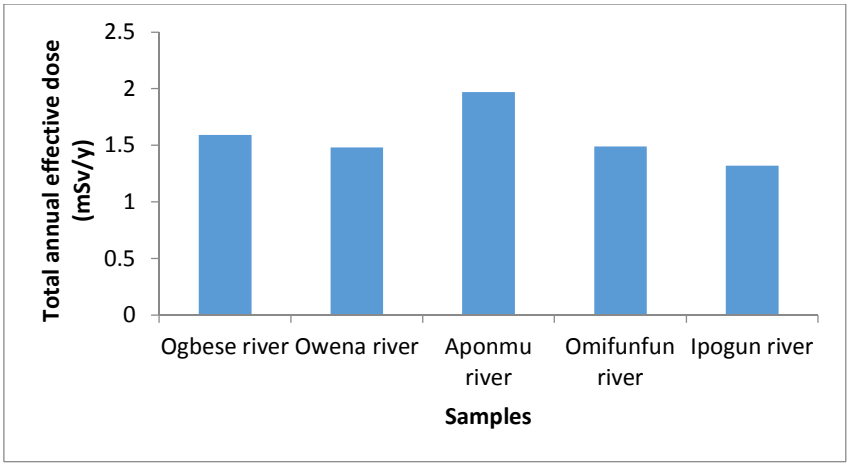

Fig. 7. Comparison of total annual effective dose (mSv/y) from different samples (river)

Table 6. Estimated cancer risk and hereditary effects for adult from river water samples

Sample ID	Fatality Cancer Risk to Adult per year (10^{-5})	Lifetime Fatality Cancer Risk to Adult (10^{-3})	Severe Hereditary effect in Adult per year (10^{-7})	Estimated Lifetime Hereditary effect in Adult (10^{-5})
B1	1.10	0.77	4.0	2.80
B2	1.05	0.73	3.8	2.66
B3	1.54	1.08	5.6	3.92
B4	1.10	0.77	4.0	2.80
B5	0.83	0.58	3.0	2.10

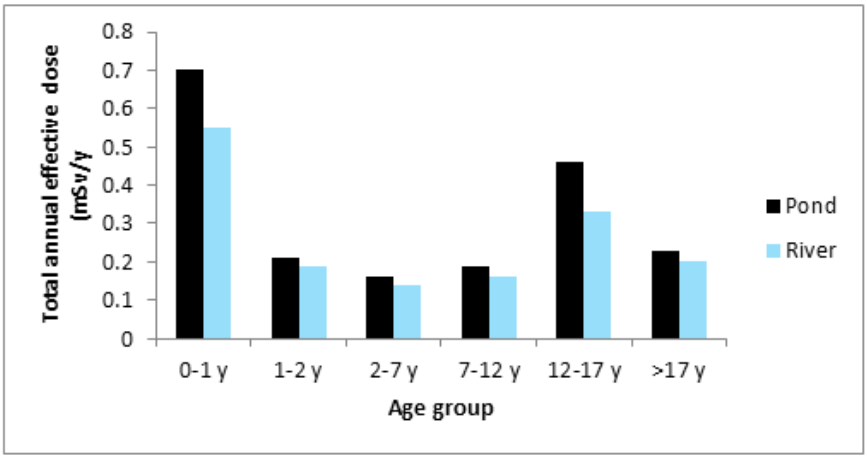

Fig. 8. Total annual effective dose (mSv/y) to the six age groups in water from pond and river water

4. CONCLUSION

This work presents detailed natural radioactivity measurements in drinking water from pond and river in some parts of Ondo State, Southwestern, Nigeria. The activity profiles of the radionuclides estimated from the pond and river have clearly shown low activity concentrations. This may be attributed to the low level of natural radioactivity in bedrocks in the areas where the water is coming from. The exposure doses from drinking water from the sources are within the reference limit of ICRP (1.0 mSv/y). The estimated lifetime fatality risk in adult member of some parts of Ondo State, Southwestern Nigeria due to ingestion of radionuclides in water from the sources are above the range of acceptable risk values recommended by USEPA. Therefore, based on the results obtained from this study, we can conclude that the annual effective doses estimated from the pond and river drinking water samples show that the waters are suitable for human consumption in the study area.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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