

RADIOLOGICAL ASSESSMENT OF DAM WATER AND SEDIMENTS FOR NATURAL RADIOACTIVITY AND ITS OVERALL HEALTH DETRIMENTS

*Ibikunle S. B., Ajayi, O.S.; Arogunjo, A.M. and Salami, A.A.

Department of Physics, Federal University of Technology, P.M.B 704, Akure, Nigeria.

(Corresponding Author: tuncom7084@yahoo.co.uk ; Tel. No.+2348037218023)

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ABSTRACT

The activity concentrations of natural radionuclides were measured in surface water and sediment samples. The samples were collected from water dam in Abeokuta area in the southwestern Nigeria. The activity concentration was determined using a low- γ -spectroscopy. The activity concentration of ^{40}K , ^{238}U and ^{232}Th ranged between 86.55 - 36.81 - 122.74 - 57.85 Bq kg⁻¹, 6.78 - 2.42 - 10.97 - 3.75 Bq kg⁻¹ and 5.27 - 2.65 - 8.19 - 3.17 Bq kg⁻¹ with a mean of 102.15 ± 60.99 Bq kg⁻¹, 9.00 ± 3.34 Bq kg⁻¹ and 7.13 ± 2.63 Bq kg⁻¹ respectively for the water samples from the dam, and 118.37 - 28.73 - 217.91 - 49.47 Bq kg⁻¹, 7.98 - 4.02 - 13.51 - 5.68 Bq kg⁻¹ and 7.86 - 2.65 - 11.28 - 4.24 Bq kg⁻¹ with a mean of 174.69 ± 39.36 Bq kg⁻¹, 9.94 ± 4.31 Bq kg⁻¹ and 9.12 ± 3.25 Bq kg⁻¹ respectively for sediment samples from the dam. The variability of the values showed the wide disparity in the measured activity concentrations. No artificial gamma emitting radionuclide was detected in the samples. The projected collective health detriment for infants (0-1y), children (7-12y) and adults (>17y) from the ingestion of water from the dam were 28, 38 and 145 persons respectively. However, the chances of radiological hazard to the health of human from radioactivity in the soil were generally low.

Keywords: Radionuclides, Activity Concentration, Water, Sediments, Health Detriment, Radiological Hazard.

INTRODUCTION

The issue of radionuclides and its concentration in environmental matrices has been on an increasing wave of interest in recent time. The carcinogenic nature and the overall effect on human make it paramount in the heart of everyone. The earth has been radioactive ever since its formation into a solid mass over 4½ billion years ago. However, we have only known about radiation and radioactivity for just over one hundred years. The early discovery of naturally occurring radiation and environmental radioactivity has led to an extensive survey in many countries all over the world (UNSCEAR, 2000). Such investigations can be useful for both the assessment of public dose rates and the performance of epidemiological studies, as well as to keep reference-data records, in order to ascertain possible changes in the environment radioactivity due to nuclear, industrial, and other human activities. Natural radioactivity arises mainly from primordial radionuclides, such as ^{40}K and the nuclides from the ^{232}Th and ^{238}U series and their decay products, which occur at trace levels in all ground formations. Primordial radionuclides are formed by the process of nucleo-synthesis in stars and are characterized by half-lives comparable to the age of the earth (Tzortzis,

2003).

Researchers, both at the developed and developing nations of the world have worked extensively in determining the activity concentration of radionuclides in various sources of water and different types of soil, as it affects community health (Strain, 1979). This has been of help in calculating the annual collective effective dose due to exposure through ingestion or inhalation. Olomo *et al.* (1994) investigated the concentration of naturally occurring radionuclides in the community water supply within the Obafemi Awolowo University Campus. They reported low mean specific activity for ^{238}U and ^{232}Th without any trace amount of ^{40}K . Tahir *et al.* (2005) measured the activity concentration of naturally occurring radionuclides in soil samples from Punjab province of Pakistan. They found out that potentially hazardous radionuclides from ^{232}Th , ^{226}Ra and ^{40}K were present in the soil samples at a moderate mean activity concentration. Since these estimated radiological impact assessment factors are lower than the recommended values, they concluded that no potentially radiological health hazard associated with the soil in Punjab-Pakistan. (Oyedele, 2006) estimated the concentration of naturally occurring radionuclides in the soils of

Windhoek city, Namibia, Southern African. His findings showed that ^{238}U , ^{232}Th and ^{40}K were present in trace amount and the mean effective dose is insignificant as it is less than the maximum permissible dose of 1 mSv^{-1} recommended by ICRP. Misdaq *et al.* (2006) investigated the activity concentration of ^{238}U and ^{232}Th in potable waters in Morocco. They reported the medium range activity concentration of ^{238}U and ^{232}Th in the range 0.37 ± 0.02 to $13.60 \pm 0.97\text{ BqL}^{-1}$ and 0.33 ± 0.02 to $7.10 \pm 0.49\text{ BqL}^{-1}$ respectively. Ajayi *et al.* (2007) detected the presence of ^{226}Ra , ^{228}Ra , ^{238}U , ^{232}Th and ^{40}K in private dug wells in Akure south western Nigeria in amounts higher than the limits recommended by both WHO and ICRP. Also Ajayi *et al.* (2008) made an assessment of natural radioactivity of soils and its external radiological impact in south western part of Nigeria. The result showed the presence of ^{238}U and ^{232}Th decay series, ^{40}K , ^{137}Cs and ^{235}U in the soils. They also measured their activity concentration. Some areas of high background radiations were noticed. (Akinloye, 2008) assessed the radioactivity level in water supplies across Ladoke Akintola University Campus, in which ^{226}Ra , ^{228}Ra and ^{40}K were found to be present in trace amount, as the data obtained is still below the maximum permissible dose recommended by ICRP (1991).

Researchers in epidemiological studies have attempted to relate health effects as a result of exposure to elevated radiation from natural sources, as well as exposure from air, food, industry, soil and community water (Roberto and Willibald, 1991). Hence, this work was carried out to undertake the measurement of the activity concentrations of natural radionuclides (^{40}K , ^{226}Ra , ^{228}Ra), in water and sediment from Oyan river dam in Abeokuta, Ogun State. To assess the concentration and effects of these radionuclides, environmental samples were collected for analyses to obtain a valid data for activity concentration of natural radionuclides in water and sediments from the dam under study. The data were used to estimate the absorbed effective and ingested doses that the occupant of the environment were exposed to and determined the overall health risk on human in the study area.

Description of the Study Area

The Oyan River Dam is a popular dam located in Abeokuta North local government area of Ogun State in the Southwestern part of Nigeria, about 20 km North West of the state capital Abeokuta. The dam was constructed across the Oyan River, a tributary of the Ogun River. It was designed to supply raw water to Lagos and Abeokuta, and to support the 3,000 hectare Lower Ogun Irrigation Project. Three turbines of 3 megawatts each were installed in 1983 but as of 2007 had not been used (Ikenweibe *et al.*, 2007).

The dam was commissioned in 1983 and is operated by the Ogun-Osun River Basin Development Authority. The lake is in the savannah region, with sparse trees and grasses and low soil fertility. It covers 4,000 hectares and has a catchment area of $9,000\text{ km}^2$. The dam has a crest length of 1044 m, height 30.4 m and gross storage capacity of 270 million m^3 (Ofoezie, 1997).

MATERIALS AND METHODS

Sampling Collection and Processing

Samples of water were taken from the dam reservoir at different locations. The water samples were each collected into a 100 cl bottle and acidified with 11M HCL solution at the rate of 10 ml per litre of sample. This was done to avoid absorption of radionuclide into the walls of the plastic container while it also releases any radionuclide already absorbed back into the samples. In the laboratory, water samples were turned into marinelli beakers. Each marinelli beaker with a volume of 1 litre was first rinsed with dilute Sulphuric Acid (H_2SO_4) and then the samples were sealed and left for about 1 month to achieve a state of secular equilibrium between the parent and daughter nuclides within each sample. After this equilibrium was achieved, the water samples were analyzed and the activity of the radionuclide present in each sample determined.

Samples of soil sediments were collected and processed according to the recommended procedure by the IAEA (1989). The soil samples were air-dried for between 2–4 days, pulverized and homogenized by grinding it into powdery form. The powdered sample was then sieved using a 2mm sized mesh screen to obtain a fine texture of soil samples. The pulverized samples were then

oven-dried at 110°C until they attained constant weight. Three hundred grams (300 g) of each dried sample was sealed in radon impermeable, cylindrical plastic geometry containers (8 cm height and 7.5 cm diameter). The samples were then stored for upward of 1 month before gamma activity counting, so as to ensure radioactive equilibrium between the long-lived radionuclides and their short-lived decay products. The storage of the samples for this period was essential, because the activities of ^{238}U and ^{232}Th were measured by estimating ^{214}Bi and ^{208}Tl respectively (Singh *et al.* 2010).

Counting and Analysis of Samples

The gamma analysis was performed on a 76 mm x 76 mm Sodium Iodide (Thallium doped) NaI(Tl) scintillation detector. The output of the detector was connected to a Canberra Series 10 plus Portable Multichannel Analyzer (MCA) which recorded the gamma spectra of the soil samples as well as background radiation. Measurements of radionuclide contents of the soil sample in this study was carried out at the Centre for Energy Research and Development, Obafemi Awolowo University, Ile-Ife. The meshed soil sample was weighed out and transferred to a 0.573 litre capacity container for gamma analysis. Cross contamination was avoided by maintaining utmost cleanliness while preparing each sample. The counting time for background radiation in the soil sample was 2 hours (7,200 s).

For calibration, a 500 cc sand standard radionuclide source was prepared using 0.07721 g measured gravimetrically from a master radionuclide solution source which was calibrated using the NaI(Tl) gamma spectrometer system. The gamma spectroscopy analysis was based on a computer program which matched gamma energy at various energy levels to a library of possible isotopes. For the purpose of this study, three regions of interest were defined for ^{40}K , ^{238}U and ^{232}Th . ^{232}Th concentration in soil was determined by the 911 keV gamma lines of ^{228}Ac while the ^{238}U concentrations were determined by the 609 keV gamma lines of ^{214}Bi . The activity concentration of ^{40}K was determined from the peak areas at 1460 keV.

Standard Radiological Indices

Standard radiation hazard indices were used to evaluate the effects of radiation doses on the health of humans that are exposed to natural environmental radiation through ingestion and inhalation. The indices to be evaluated are discussed below.

The annual Effective Dose (Sv/y) due to ingestion of radionuclides from water samples was calculated using the relation by Alam *et al.* (1999)

$$E_d = A_c A_i C_f \quad 1$$

Where E_d is the annual effective dose, A_c is the activity concentration in Bq/l, A_i is the annual consumption rate of water in l/y, and C_f is the ingested dose conversion factor (Sv/Bq).

Committed effective dose (C_d) to an individual over an average life span of 50 yrs was estimated from the equation:

$$C_d = 50 \times E_d \quad 2$$

The collective effective dose equivalent to a population, is a measure of the collective detrimental effects and the percentage of people at risk of incurring radiation-induced diseases; which is calculated using the expression ICRP (1991) defined as:

$$S_E = \sum N_i H_{Ei} \quad 3$$

Where S_E = collective effective dose equivalent (person-Sv)

N_i = the numbers of individual exposed to radiation and H_{Ei} is the mean outdoor effective dose equivalent (μSv^{-1}).

The collective health detriment (G) (person), due to exposure to gamma radiation in an environment, can be calculated using the relation

$$G = R_T S_E \quad 4$$

Where R_T = Total risk factor the body organs are exposed to and according to ICRP (1991). It has a value of $16.50 \times 10^{-3} \text{Sv}^{-1}$, S_E = Collective effective

dose equivalent (person – Sv).

Radiological Indices for Soil Sediment Sample

The absorbed dose rate in air, D (nGyh^{-1}) due to a partial evaluation of the radiological hazard posed by the exposure to these estimated radioactivity concentrations at 1 m above the ground containing the naturally occurring radionuclide is calculated from UNSCEAR, (2000)

$$D = 0.042A_k + 0.462A_{Ra} + 0.604A_{Th} \quad 5$$

Where A_k , A_U , and A_{Th} are the specific activity concentration in Bqkg^{-1} of K, U and Th respectively in the soil sample and 0.042, 0.462 and 0.604 (nGyh^{-1} per Bqkg^{-1}) are the concentration-to-dose conversion factors.

Using an outdoor occupancy factor (OF) of 0.20 and the Dose Conversion Factor (DCF) of 0.70 SvGy^{-1} (UNSCEAR, 1988), the Annual Effective Dose Equivalent (AEDE) from the calculated outdoor terrestrial gamma radiation at 1 m above the ground were calculated using the relation:

$$AEDE = D \times DCF \times OF \times T \quad 6$$

where T is 8760 h.

Radionuclides of ^{238}U , ^{232}Th and ^{40}K are not homogeneously distributed in soil. The inhomogeneous distribution from these naturally occurring radionuclides is due to disequilibrium between ^{238}U and its decay products. For uniformity in exposure estimates, the radionuclide concentrations have been defined in terms of radium equivalent activity (Ra_{eq}) in Bqkg^{-1} . This allows comparison of the specific activity of materials containing different amounts of ^{238}U ,

^{232}Th and ^{40}K according to Beretka and Mathew (1985) as follows:

$$(\text{Ra}_{eq} = A_U + 1.43A_{Th} + 0.077A_K) \quad 7$$

The external hazard index (H_{ex}) was calculated from the equation:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad 8$$

The internal hazard index (H_{in}) was calculated from the equation:

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad 9$$

RESULTS AND DISCUSSION

Activity Concentration

The measured activity concentration of natural radionuclides such as ^{40}K , ^{232}Th and ^{226}Ra were in the range of 86.55 ± 36.81 to $122.74 \pm 57.85 \text{ Bq/l}$, 6.78 ± 2.42 to $10.97 \pm 3.75 \text{ Bq/l}$ and 5.27 ± 2.65 to $8.19 \pm 3.17 \text{ Bq/l}$ respectively in the water samples, and 118.3 ± 28.73 to $217.91 \pm 49.47 \text{ Bq/kg}$, 7.98 ± 4.02 to $13.51 \pm 5.68 \text{ Bq/kg}$, and 7.86 ± 2.86 to $11.28 \pm 4.24 \text{ Bq/kg}$ respectively in sediment samples. These values are presented in Table 1 and Figure 1.

^{40}K displayed the highest activity. This is expected because ^{40}K is a naturally occurring radionuclide which abounds in the earth crust. The activity concentration of natural radionuclide in the study area is in agreement with the global trend on the distribution of natural radionuclide in the soil.

Figure 2 shows the correlation between the activity concentrations of Ra-226 and Th-232 in water and sediment samples.

Table 1: Activity Concentration in Collected Samples

Collected Samples	Activity Concentration		
	K-40	Ra-226	Th-232
Water (Bq/L)			
Sample 1	97.15 ± 27.32	6.78 ± 2.42	5.27 ± 2.65
Sample 2	86.55 ± 36.81	10.97 ± 3.75	8.19 ± 3.17
Sample 3	122.74 ± 57.85	9.26 ± 3.86	7.93 ± 2.08
Range	86.55 ± 36.81 - 122.74 ± 57.85	6.78 ± 2.42 - 10.97 ± 3.75	5.27 ± 2.65 - 8.19 ± 3.17
Mean	102.15 ± 40.66	9.00 ± 3.34	7.13 ± 2.63
Sediments (Bq/kg)			
Sample 1	118.37 ± 28.73	7.98 ± 4.02	8.22 ± 2.86
Sample 2	187.79 ± 39.87	8.34 ± 3.24	7.86 ± 2.65
Sample 3	217.91 ± 49.47	13.51 ± 5.68	11.28 ± 4.24
Range	118.37 ± 28.73 - 217.91 ± 49.47	7.98 ± 4.02 - 13.51 ± 5.68	7.86 ± 2.65 - 11.28 ± 4.24
Mean	174.69 ± 39.36	9.94 ± 4.31	9.12 ± 3.25

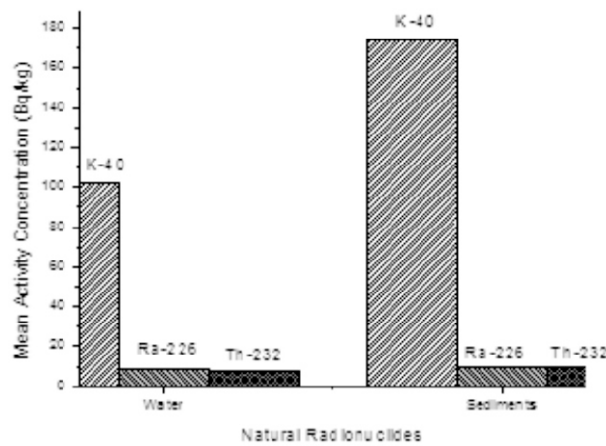


Fig 1: Mean Activity Concentration of Natural Radionuclides in Dam Water and Sediments

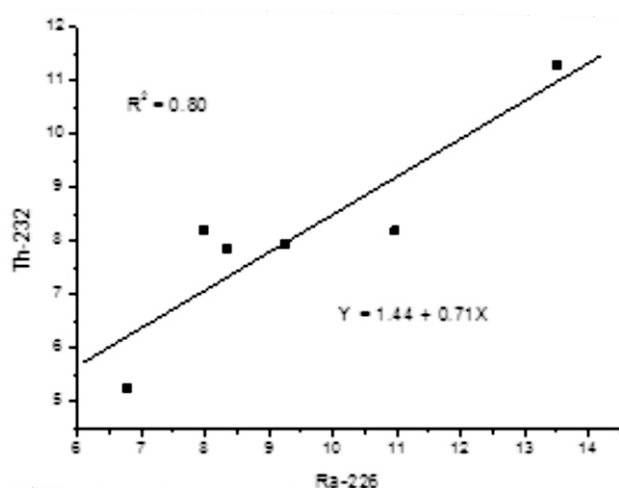


Fig 2: Correlation between Ra-226 and Th-232 in Water and Sediment Samples

Total Annual Effective Dose and Committed Effective Dose

The total annual effective dose E_d (Sv/y) estimation to an individual due to the ingestion of the natural radionuclides present in water samples was carried out using the equation 1

Only three age groups were used for the purpose of this study;

Infant : 0-1y; Children: 7-12y; Adult: >17y

The variation of the different age groups with the Ingested dose conversion factor was provided by IAEA (1996) and shown in Table 2 while it's

variation with the annual consumption rate of water in a year was supplied by WHO (2003) and shown in Table 3. Figure 3 and Table 4 show the mean total annual effective dose for adults (>17y), children (7-12y), and infants (0-1y) as 3.50 1.31, 3.71 1.39 and 16.29 6.06 mSv⁻¹ respectively. These values, most especially for infants, were extremely higher than the 1 mSv⁻¹ recommended by NCRP, 1993.

Committed Effective Dose to an individual (evaluated here only for the >17 age group) over an average life span of 50 yrs was estimated at 175.00 65.50 mSv using equation 2.

Table 2 Radionuclides and Their Ingested Dose Conversion Factor

Radionuclide	Ingested Dose Conversion Factor Per Age Group					
	(0-1)yr Sv/Bq	(1-2)yr Sv/Bq	(2-7)yr Sv/Bq	(7-12)yr Sv/Bq	(12-17)yr Sv/Bq	>17yrs Sv/Bq
⁴⁰ K	6.2E-8	4.2E-8	2.1E-8	1.3E-8	7.6E-9	6.2E-9
²¹⁴ Bi	1.4E-9	7.4E-10	3.6E-10	2.1E-10	1.4E-10	1.0E-10
²¹⁴ Pb	2.7E-9	1.0E-9	5.2E-10	3.1E-10	2.0E-10	1.4E-10
²²⁴ Ra	2.7E-6	6.6E-7	3.5E-7	2.6E-7	2.0E-7	6.5E-8
²²⁶ Ra	4.7E-6	9.6E-7	6.2E-7	8.0E-7	1.5E-6	2.8E-7
²²⁸ Ac	7.4E-9	2.8E-9	1.4E-9	8.7E-10	5.3E-10	4.3E-10
²³² Th	4.6E-6	4.5E-7	3.5E-7	2.9E-7	2.5E-7	2.3E-7

Source: LAEA 1996

Table 3 ANNUAL WATER CONSUMPTION PER AGE GROUP

AGE GROUP (y)	ANNUAL WATER CONSUMPTION (ly ⁻¹)
0-1	2.0E2
1-2	2.6E2
2-7	3.0E2
7-12	3.5E2
12-17	6.0E2
>17	7.3E2

Source: WHO 2006

Table 4 Total Annual Effective Dose and Collective Effective Dose Equivalent for Different Age Groups

Age Group	Total Annual Effective Dose (mSv ⁻¹)	Collective Effective Dose Equivalent (Person-Sv)
Infants (0 – 1 y)	16.29 ± 6.06	1,696.75
Children (7- 12 y)	3.71 ± 1.39	2,324.57
Adults (> 17 y)	3.50 ± 1.31	8,816.62

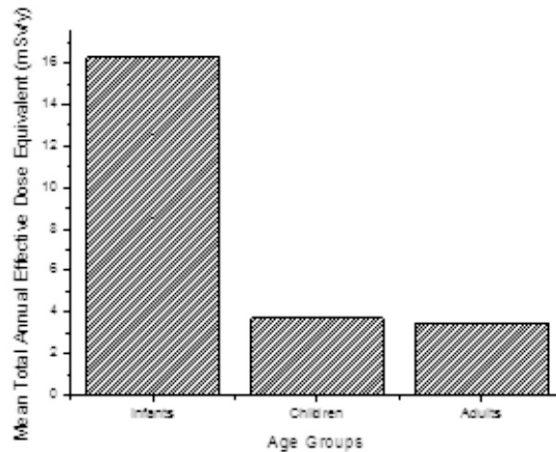


Fig 3: Mean Total Annual Effective Dose Equivalent for Different Age Group through Radionuclide Ingestion From Dam Water

Collective Effective Dose Equivalent and Health Detriment

The collective effective dose equivalent for the different age groups was estimated using equation 3 and presented in Table 5 while the population of the different age group were presented in Table 6. From the 140,431,790 people living in Nigeria according to NPC (2006), and projected

population of 183,390,767 (Worldometers, 2015). 3,728,098 according to NPC (2006) and projected population of 4,868,547 (Worldometers, 2015) people are living in Ogun State across which the dam supplies water. The number of people within the age range of 0-1, 7-12 and >17 years in Ogun State are 104,159; 626,568; and 2,519,033 respectively. It was discovered that though the

infant group has the highest estimated mean total effective dose, the adults because of their large population has more people susceptible to a radiation induced disease. However, the greatest

fraction of health detriment comes from the infant group. This could be linked to the fact that infants' body system is more radiosensitive than others.

Table 5 Collective Health Detriment for Different Age Groups

Age Group	Collective Health Detriment in (Person)	% Health Detriment in (Person)
Infants (0 – 1 y)	28	0.0269 (269 from every 1,000,000)
Children (7- 12 y)	38	0.0061 (61 from every 1,000,000)
Adults (> 17 y)	145	0.0058 (58 from every 1,000,000)

Table 6 Ogun State, Nigeria Population for Different Age Groups

Age Range (Years)	Population (N _i) (2006) <i>Source: NPC (2006)</i>	Population (N _i) (2015 Projection) (Worldometers, 2015)
0-1	79,760	104,159
7-12	479,796	626,569
>17	1,928,954	2,519,033

Radiological Indices for Soil Sediment Sample

The specific activities measured in the collected sediment sample were presented in Table 1. The absorbed dose rate in air from measured activity concentrations in the sediment sample was estimated using equation 5 and the values are presented in Table 7 alongside with radium equivalent and the hazard indices. The result shows a lesser value than the world average value of 55 nGyh⁻¹ UNSCEAR (2000). This hazard indices, both internal and external, show values less than unity which means that the radiation hazard through the soil is insignificant, that is, the

area is safe for human.

The mean annual effective dose equivalent estimated from equation 6 is given as 21.39 6.89 μSv/y. This is about 30.56% of the world average which is 70 μSv/y, and lower than 1.0 mSv/y recommend by the International Commission on Radiological Protection ICRP (1992) as the maximum permissible AEDE for members of the public. The mean Ra_{eq} was estimated at 36.44 11.99 Bq/kg, which is less than 10 % of the recommended maximum of 370 Bq/Kg UNSCEAR (2000).

Table 7 Absorbed Dose Rate, Radium Equivalent, External and Internal Hazard Indices of Radionuclides in Sediment Sample

Samples	Absorbed Dose Rate in Air (nGyh ⁻¹)	Radium Equivalent (Bq/Kg)	External Hazard Index (Bq/Kg)	Internal Hazard Index (Bq/Kg)
Sample 1	13.62 ± 4.79	28.85 ± 10.32	0.078 ± 0.028	0.099 ± 0.039
Sample 2	16.49 ± 4.77	34.04 ± 10.10	0.092 ± 0.027	0.114 ± 0.036
Sample 3	22.21 ± 7.26	46.42 ± 15.55	0.125 ± 0.042	0.162 ± 0.057
Mean	17.44 ± 5.61	36.44 ± 11.99	0.098 ± 0.032	0.125 ± 0.044

CONCLUSION

Natural radioactivity in the sediments and water of dam has been determined using gamma-ray spectrometer. The activity concentrations of ^{40}K , ^{238}U and ^{232}Th obtained in this study are in agreement with the global trend on the distribution of natural radionuclide in the soil. Although the soil samples show lower activity than the world average and recommended standard, the water samples show relatively higher activity. This implies that the activity level of surface water may have been determined by the activity of the sediments and other sources which include the country rocks and anthropogenic activities such as disposal into the water body of domestic, agricultural and industrial wastes (Manyama and Elis, 1993 and Akinloye, 2008). Based on estimates of the collective health detriment for different age groups, the dam water is not considered potable, without treatment.

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