Determination of Radiological Health Risk Due to Gamma Exposure from River Water around Oil Bunking Centre in Rivers State, Nigeria

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Authors' contributions

This work was carried out in collaboration between both authors. Author PCO designed the study, performed the statistical analysis, wrote the protocol, wrote the first draft of the manuscript and managed the analyses of the study. Author ACO handled the field work and managed the literature searches. Both authors read and approved the final manuscript.

ABSTRACT

**Aim:** The aim of this study is to assess radiation health risk due to gamma exposure from river water around oil bunkering centers in Rivers state, Nigeria.

**Study Design:** This study was purely an experimental work.

**Place and Duration of Study:** Sampling started from the meeting point of Otamiri tributary and Imo River at the Abia /Rivers boundary to over seven kilometers along the Imo River; between July 2016 and January, 2017.

**Methodology:** 20 samples of river water were collected along coastal shore of Imo River with pre-washed 1.5 ml Polypropylene bottles. The bottles were rinsed with the water before collection and acidified immediately after collection with few drops of nitric acid. The bottles were sealed tightly with vinyl tapes and kept in the laboratory for 4 weeks for secular equilibrium of the radionuclides. The activity concentration of the radionuclides was measured using well calibrated Sodium Iodide detector.
Results: The mean activity concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ were 2.02±0.02, 3.59±0.21 and 10.43±1.13 BqL$^{-1}$. The mean annual effective dose estimated for infants, children and adult citizens that ingest river water sampled were 58.64, 0.19 and 0.24 mSv$y^{-1}$ respectively. The values of annual effective dose for infants and adults exceeded the reference levels of 0.26, 0.2 and 0.10 mSv$y^{-1}$ respectively while that for children is within the safe reference level. The estimated fatal cancer risk to adult citizens and the lifetime hereditary effects show that 53 out of 10,000 citizens may suffer some form of cancer fatality and 596 out of 1000,000 citizens may suffer some form of hereditary effect since the values exceeded the USEPA recommended range.

Conclusion: The result of this study show that the river water under study has been radiologically impacted by oil bunking activities and may cause significant health risk. Hence few recommendations were made in this work which will help to reduce radiation exposure and possible health impact.

Keywords: Radionuclide; lifetime cancer risk; committed dose; hereditary effect; Imo River.

1. INTRODUCTION

River water does not exist in a pure form for any appreciable length of time in nature. Even while water falls as rain, it picks up small amount of contaminants from the atmosphere and moves as it filters through the ground [1]. Those contaminants may be natural or anthropogenic including biological, chemical, physical and radiological impurities such as industrial and commercial solvents, heavy metals, acid salts, and radioactive materials. The natural radionuclides in water result from weathering and recycling of terrestrial minerals and rocks that give rise to $^{40}\text{K}$, $^{232}\text{Th}$, $^{235}\text{U}$ and $^{238}\text{U}$. The later three decay naturally to produce other important radioactive isotopes which include radium (Ra), radon (Rn), polonium (Po) and lead (Pb) [2,3]. Water can also become contaminated as it picks up radioactive materials from the surrounding rocks, soils or cracked cement as it flows past.

In a closed system the progeny of thorium (Th) and uranium (U) are present in concentrations determined by the concentration of parent uranium and thorium isotopes and the time since the system became closed to nuclide migration. In nature closed systems rarely exist and predictions regarding nuclide concentrations in water bodies invariably include large uncertainties. These nuclides and their decay products are found in ground and spring waters in specific concentrations dependent on complex hydrogeologic processes and conditions (dissolution, transport and ion-exchange processes as well as redox potentials and pH-conditions of the aqueous system). These hydrogeological processes result in non-equilibrium conditions between parent nuclides and their progeny. However, characteristic behaviour in the natural environment can provide a basis for assumptions regarding probable behaviour of nuclides used in the radioactivity screening assessment [4].

In the oxidised zone of the earth’s near-surface environment $^{232}\text{Th}$ and $^{238}\text{U}$ may both be mobilised, but in different ways. The former has an extremely low solubility in natural waters. There is a close correlation of thorium concentration and detrital content of water. This nuclide is almost entirely transported in particulate matter and is bound in insoluble resistant minerals or is adsorbed on the surface of clay minerals. The radioactive decay of $^{234}\text{U}$ it rapidly hydrolyses and adsorbs on to the nearest solid surface. Products of radioactive decay in the U and Th series include radon (Rn) gas of which three isotopes exist. Of these $^{222}\text{Rn}$ is abundant will cause disequilibrium between members of a decay chain. $^{222}\text{Rn}$ has an appreciable solubility in water and is often found in concentrations far in excess of the parent nuclide radium ($^{226}\text{Ra}$). A $^{222}\text{Rn}$/$^{226}\text{Ra}$ activity ratio of 450 has been observed in ground waters from central England [5]. Aeration of water and short half-lives make the contribution of radon negligible in ingestion dose calculations.

Bunkering activities and crude method of refining crude oil along Imo river course has introduced a lot of hazardous waste into the water bodies. Recently the entire Rivers state is experiencing massive air pollution (black soot). Some speculations are pointing towards the illegal refining of crude oil in all those oil bunkering centres which produces some kind of explosions in the process. The inadvertent discharges of petroleum hydrocarbons or petroleum derived wastes streams from oil and gas productions activities are toxic to the coastal waters, soils and
sediment near the discharge point [6]. For human race, water is essential to life as air to breath. Thus, the importance of investigating the levels of radionuclide element in river water is very important [1] as river water serves as a major source of drinking water for the human race. Estimation of radiation dose distribution is vital in assessing the health risk to a population and serves as a reference for documenting changes in environmental radioactivity due to anthropogenic activities [7]. Hence, the aim of this work is to determine the radiological health risk of the populace from the activity concentration of $^{238}$U, $^{232}$Th and $^{40}$K in river water collected from Imo River near the bunkering sites in Rivers state. The result will help in assessment of the health impact of oil bunkering activities in Rivers state.

2. MATERIALS AND METHODS

2.1 Study Area

The Imo River is located in the northern part of Rivers State in South eastern Nigeria. The study area is the boundary between Abia State and River State in the Niger Delta region. It lies between longitude 007° 08' 11.9" and 007° 11' 35.5" East and latitudes 04° 54' 11.9" and 04° 51' 37.8" North of equator (Fig. 1). It flows 240 km into the Atlantic Ocean with an estuary of about 40 km wide, it has an annual discharge of 4 km$^3$ with 26,000 hectares of wetlands. Its tributaries are the Otamiri and Oramirukwa [8]. The River serves as drinking water sources for the surrounding communities.

Two geologic formations are covered in the study area, namely: Imo shale and Ameki formations. Imo shale consists of a thick sequence of blue and dark grey shales with occasional bands of clay-ironstones and subordinate sandstones [9]. It dips at angles 17° to 25° to the south-west and South [10]. It includes three constituent sandstones: the Igbabu, Ebenebe and Umuna Sandstones with the last two outcropping in the Imo River Basin. The Umuna sand stone is composed of thick sandstone units and minor shales and is generally less than 70 m thick. The Ebenebe Sand stone occurs as a lens in the northwestern extremity of the Imo River Basin. It is similar in lithology to the Umuna sandstone but is relatively thicker with a maximum thickness of 130 m [10]. Ameki Formation (Eocene) consists of sand and sandstones. The lithologic units of the Ameki Formation fall into two general groups [11,12,13]; an upper grey-green sandstones and sandy clay and a lower unit with fine to coarse sandstones, and intercalations of calcareous shale and thin shelly limestone.
2.2 Sample Collection and Preparation

Sampling started from the meeting point of Otamiri tributary and Imo River at the Abia/Rivers boundary to over seven kilometers along the Imo River. The water samples (20 altogether) were collected with 1.5 l polypropylene bottles which were carefully washed using detergent and then rinsed with freshly distilled Hydrochloric acid (HCl) to remove an inorganic material that might have stuck to the walls of the container as 20 ml of 1 M HNO₃ added immediately to each sample in the containers so as to fix the contained radioactive elements [14]. The samples were taken to the National Institute of Radiation Protection and Research (NIRPR) University of Ibadan. 250 ml of each of the samples were collected with 1.5 l linear polypropylene bottles and 2.0 l/d for infants, children and adults (≥ 17 years) respectively, were used for calculations. In this work, the rate of water intake rates based on UNSCEAR [19] recommendation of 0.5, 1.0 and 2.0 l/d for infants, children and adults (≥ 17 years) respectively, were used for calculations. The conversion factors for 238U, 232Th and 40K as reported by ICRP [20] and presented in Table 1 were used for all the age groups.

2.3 Gamma Spectroscopy

Activity count of the radionuclides contained in the samples were performed using gamma spectroscopy system having a thallium activated 3” × 3” Sodium Iodide (NaI(Tl)) detector connected to an ORTEC 456 amplifier of the spectrometry system [15,16]. Energy and efficiency calibration of this system were carried out using 137Cs and 60Co, standard sources from IAEA, Vienna and the energy resolution was 39.5 and 22.2%. The analysis was performed using a Canberra S 100 computer analyzer. Standard of natural origin were prepared in the same manner as the samples, these standards are uranyl nitrate (UO(2)₃(NO₃)₂ 6H₂O) 502.18 mol/g, potassium chloride (Kcl) 74.55 mol/g and thorium nitrate (Th(NO₃)₄ .5H₂O) 570.13 mol/g. One gram of each of the standard was taken and dissolved into a 200 ml distilled water to form a standard solution. It is subtle that 1 g of uranyl nitrate contains 0.474 g of uranium which has activity of 0.0294 Bq/l, also 1 g of potassium chloride contains 0.534 g of potassium which has activity of 0.706 Bq/l and 1g of thorium nitrate contains 0.859 g of thorium with activity of 0.0175 Bq/l [17]. The standard solution was kept to equilibrate before counting. The peak energy of 1764 kev gamma-line of Bi-214 is used to estimate the activity concentration of uranium in samples. Also the energy of 2614.5 kev gamma line of Ti-208 is used to estimate the activity concentration of thorium in the samples. The single energy of 1460 kev gamma line of potassium-40 gives the direct activity concentration measurement of potassium-40. The operational voltage was set at 900 v and preset time 29,000 seconds maintained [18].

The configuration and detector geometry was maintained throughout the analysis. The individual radionuclide concentration calculated using relative method as in equation (1) (Onoja, 2011).

\[
\frac{Activity \text{ of } U1}{Activity \text{ of } S1} = \frac{\sum U1 - \sum b}{\sum S1 - \sum b}
\]  

Where \( U1 \) = The unknown sample activity concentration in the unit of Bq⁻¹, \( S1 \) = activity of the standard source, \( \sum U1 \) = sum under the peak of \( U1 \) in cps, \( \sum S1 \) = the sum under the peak \( S1 \) in cps.

3. RADIOLOGICAL RISK ESTIMATION

The annual effective dose from ingestion of radionuclide in water samples was estimated using the obtained mean activity concentrations of the identified radionuclides. Assumptions on the rate of ingestion of water were made. In this work, the rate of water intake rates based on UNSCEAR [19] recommendation of 0.5, 1.0 and 2.0 l/d for infants, children and adults (≥ 17 years) respectively, were used for calculations. The conversion factors for 238U, 232Th and 40K as reported by ICRP [20] and presented in Table 1 were used for all the age groups.

The total annual effective dose due to ingestion of water was computed using the following formula [21,22] (ICRP, 1996, Ndontchueng et al. [22]).

\[
H_{ing} \text{ (mSv} y^{-1}) = \sum_{i=1}^{3} DCF_{ing} \text{ (i) } \times A_i \times I
\]  

Where \( DCF_{ing} \text{ (i) } \) is the dose coefficient of a particular radionuclide in Sv/Bq for a particular age categories, \( A_i \) is the specific activity concentration of radionuclide in the water sample measured in Bq/l and I, the radionuclide intake in liters per year for each age categories.

In addition to the estimated annual effective dose, the cancer and hereditary risk due to low dose without any threshold doses known as stochastic effect were estimated using the ICRP cancer risk methodology [23]. Radiation risks to members of the public results from exposure to low dose radiation are normally known as chronic risk of somatic or hereditary damage of human tissues, thus much emphasis is always placed on
and drinking water are 0.26, 0.20 and 0.1 mSv y\(^{-1}\) corresponding to one year consumption of effective dose for infants, children and adults respectively.

The recommended reference levels of the Bq L\(^{-1}\) (Table 2 and ranges from BDL to 4.36 ± 1.07 Bq L\(^{-1}\)). These values were estimated at 0.2 × 10\(^{-6}\) for stochastic effects after exposure to low dose rates was determined in river water from Imo River. The activity concentration of 238\(^{\text{U}}\), 232\(^{\text{Th}}\) and 40\(^{\text{K}}\) in the river water was relatively higher than that of 238\(^{\text{U}}\) because thorium is very insoluble [24]. The activity concentration of 232\(^{\text{Th}}\) and 40\(^{\text{K}}\) are slightly higher than the reference levels of 1.0 and 10.0 Bq L\(^{-1}\) while that for 238\(^{\text{U}}\) is within the reference levels. The results obtained in this work was compared with other works done in a similar environment within this country and other countries of the world as presented in Table 4. Figs. 2 and 3 shows the comparison of the activity concentration of 238\(^{\text{U}}, 232^{\text{Th}}\) and 40\(^{\text{K}}\) in river water with standard value prescribed by ICRP [20]. It shows that 238\(^{\text{U}}\) activity concentrations are lower than the standard value in all the locations while about six locations, activity concentration of 232\(^{\text{Th}}\) exceeded the standard value. The ICRP [20] and WHO, [25] regulations for drinking water quality does not include a listing for 40\(^{\text{K}}\) but specifies that the maximum allowable concentration limit for beta and photon emitters should correspond to a committed effective dose of 1.0 mSv y\(^{-1}\) from annual intake at the rate of two liters’ of drinking water per day [26].

| S/N | Radioisotopes | Infant ≤ 1 year | Children 10 years | Adult > 17 years |
|-----|--------------|----------------|------------------|-----------------|
| 1   | 238\(^{\text{U}}\) | 1.4 E-07       | 6.8 E-08         | 4.5 E-08        |
| 2   | 232\(^{\text{Th}}\) | 1.6 E-06       | 2.9 E-07         | 2.3 E-07        |
| 3   | 40\(^{\text{K}}\)  | 5.2 E-05       | 1.3 E-08         | 6.2 E-09        |
|     | Water intake  | 0.5 L/day      | 1.0 L/day        | 2.0 L/day       |

The reduction of these radiological risks to natural radiation. The nominal lifetime risk coefficient of fatal cancer recommended in the 2007 recommendations of the members of the public is 5.5× 10\(^{-2}\) Sv\(^{-1}\). For hereditary effects, the detriment-adjusted nominal risk coefficient for the whole population was stated in [23] for stochastic effects after exposure to low dose rates was estimated at 0.2 × 10\(^{-5}\) Sv\(^{-1}\).

The risk to population was then estimated using the 2007 recommended risk coefficient of ICRP report and assumed 70 years lifetime of continuous exposure of the population to low level radiation. According to ICRP methodology:

\[
\text{Cancer Risk} = \text{Total annual Effective Dose (Sv)} \times \text{Cancer risk factor (Sv}^{-1}) \quad (3)
\]

\[
\text{Hereditary Effects} = \text{Total annual Effective Dose (Sv)} \times \text{Hereditary effect factor (Sv}^{-1}) \quad (4)
\]

The recommended reference levels of the effective dose for infants, children and adults corresponding to one year consumption of drinking water are 0.26, 0.20 and 0.1 mSv\(^{-1}\) respectively.

4. RESULTS AND DISCUSSION

The activity concentration of 238\(^{\text{U}}, 232^{\text{Th}}\) and 40\(^{\text{K}}\) determined in river water from Imo River and the associated annual effective dose to infant, children and adult population of the communities are presented in Table 2 while the estimated cancer risks and hereditary effects of adult member of the public are shown in Table 3.

4.1 Specific Activity Concentration of 238\(^{\text{U}}, 232^{\text{Th}}\) and 40\(^{\text{K}}\) in River Waters

The specific activity concentration of 238\(^{\text{U}}, 232^{\text{Th}}\) and 40\(^{\text{K}}\) in the river water samples are shown in Table 2 and ranges from BDL to 4.36 ± 1.07 Bq L\(^{-1}\) with an average of 2.02 ± 0.02 Bq L\(^{-1}\), BDL to 7.89 ± 0.76 Bq L\(^{-1}\) with an average value of 3.59 Bq L\(^{-1}\) and BDL to 22.11 ± 1.71 Bq L\(^{-1}\) with an average value of 10.43 Bq L\(^{-1}\). The result clearly show that 238\(^{\text{U}}\) are sparsely distributed along the coastal shore. This could be due to high mobility of uranium-238 in river water. Uranium -238 were below detectable limit in most of the locations along the shore. This is in agreement with the fact that uranium in natural environment are variable in uranium content, depending mainly on factors such as contact time with uranium bearing rocks, uranium content of the contact rock, amounts of evaporation and availability of complex ions. The ability of uranium to undergo inorganic dissociation and re-precipitation is probably the most important process in the natural environment to cause disequilibrium between the nuclides in the decay chains. The large variation of uranium observed in this work could be due to PH values which cause precipitation of uranium from the solution along the flow direction [4].

The activity concentration of 40\(^{\text{K}}\) is highest at the old Imo River basin due to illegal oil and gas bunkering activities that releases its wastes into the river. The activity concentration of 232\(^{\text{Th}}\) in river water was relatively higher than that of 238\(^{\text{U}}\) because thorium is very insoluble [24]. The activity concentration of 232\(^{\text{Th}}\) and 40\(^{\text{K}}\) are slightly higher than the reference levels of 1.0 and 10.0 Bq L\(^{-1}\) while that for 238\(^{\text{U}}\) is within the reference levels. The results obtained in this work was compared with other works done in a similar environment within this country and other countries of the world as presented in Table 4. Figs. 2 and 3 shows the comparison of the activity concentration of 238\(^{\text{U}}, 232^{\text{Th}}\) and 40\(^{\text{K}}\) in river water with standard value prescribed by ICRP, [20]. It shows that 238\(^{\text{U}}\) activity concentrations are lower than the standard value in all the locations while about six locations, activity concentration of 232\(^{\text{Th}}\) exceeded the standard value. The ICRP [20] and WHO, [25] regulations for drinking water quality does not include a listing for 40\(^{\text{K}}\) but specifies that the maximum allowable concentration limit for beta and photon emitters should correspond to a committed effective dose of 1.0 mSv y\(^{-1}\) from annual intake at the rate of two liters’ of drinking water per day [26].

Table 1. Dose coefficients (Sv/Bq) for ingestion of radionuclides for members of the public to 70 years of age (ICRP, [20]; Publication 119) and water intake

| S/N | Radioisotopes | Infant ≤ 1 year | Children 10 years | Adult > 17 years |
|-----|--------------|----------------|------------------|-----------------|
| 1   | 238\(^{\text{U}}\) | 1.4 E-07       | 6.8 E-08         | 4.5 E-08        |
| 2   | 232\(^{\text{Th}}\) | 1.6 E-06       | 2.9 E-07         | 2.3 E-07        |
| 3   | 40\(^{\text{K}}\)  | 5.2 E-05       | 1.3 E-08         | 6.2 E-09        |
|     | Water intake  | 0.5 L/day      | 1.0 L/day        | 2.0 L/day       |
Table 2. Activity concentration of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in water samples and total annual effective dose for different age categories

| S/N | Sample ID | Location                | Activity concentration (Bq/l$^{-1}$) | Total annual effective dose (mSv) |
|-----|-----------|-------------------------|--------------------------------------|----------------------------------|
|     |           |                         | $^{238}\text{U}$ | $^{232}\text{Th}$ | $^{40}\text{K}$ | Infant | Children | Adult |
| 1   | SW1       | Otamiri-Imo River       | BDL                    | BDL                     | BDL              | 0.00    | 0.00     | 0.00   |
| 2   | SW 2      | NNPC-Alscon             | BDL                    | BDL                     | 3.50±0.27        | 33.0    | 0.0167   | 0.0158 |
| 3   | SW 3      | Obigbo Bridge           | BDL                    | 0.85±0.089              | 20.33±1.50       | 193.0   | 0.186    | 0.235  |
| 4   | SW 4      | Mama Town               | BDL                    | BDL                     | 14.36±1.08       | 137.0   | 0.068    | 0.0650 |
| 5   | SW 5      | Old Imo River           | BDL                    | 3.77±0.37               | 22.11±1.71       | 212.0   | 0.504    | 0.0417 |
| 6   | SW 6      | Imo River Village       | 1.93±0.50              | 7.89±0.76               | BDL               | 0.169   | 0.883    | 1.388  |
| 7   | SW 7      | Back of Kom-Kom         | BDL                    | BDL                     | 14.36±1.08       | 137.0   | 0.068    | 0.0650 |
| 8   | SW 8      | Imo River Railway       | 3.08±0.84              | BDL                     | 3.84±0.30        | 13.63   | 0.077    | 0.0604 |
| 9   | SW 9      | NNPC Pipeline           | 4.36±1.07              | BDL                     | 3.84±0.30        | 13.63   | 0.077    | 0.0604 |
| 10  | SW 10     | Imo River               | BDL                    | BDL                     | 13.34±0.95       | 9.018   | 0.064    | 0.061  |
| 11  | SW 11     | Imo River Division 1    | BDL                    | 4.34±0.43               | BDL               | 1.271   | 0.459    | 0.728  |
| 12  | SW 12     | Imo River Division 3    | 1.10±0.30              | BDL                     | 1.43±0.10        | 13.63   | 0.034    | 0.0426 |
| 13  | SW 13     | Imo River Division 5    | BDL                    | 4.12±0.41               | BDL               | 1.206   | 0.436    | 0.692  |
| 14  | SW 14     | Imo River Division 7    | BDL                    | 7.99±0.60               | BDL               | 76.0    | 0.038    | 0.0362 |
| 15  | SW 15     | Imo River Banks 1       | 1.27±0.34              | BDL                     | 2.17±0.17        | 20.60   | 0.042    | 0.0515 |
| 16  | SW 16     | Imo River Banks 2       | BDL                    | 4.02±0.40               | BDL               | 1.177   | 0.425    | 0.675  |
| 17  | SW 17     | Imo River Banks 3       | BDL                    | BDL                     | 14.98±1.11       | 142.55  | 0.071    | 0.068  |
| 18  | SW 18     | Imo River Banks 4       | 0.39±0.09              | BDL                     | 9.81±0.73        | 93.40   | 0.056    | 0.057  |
| 19  | SW 19     | Imo River Banks 5       | BDL                    | 11.32±0.82              | BDL               | 107.72  | 0.054    | 0.0512 |
| 20  | SW 20     | Mmiri-Nwayi Division 14 | BDL                    | 0.50±0.05               | BDL               | 0.146   | 0.053    | 0.084  |
|     |           |                         | Mean                   | 2.02                    | 3.59             | 10.43   | 58.64    | 0.19   | 0.24 |

WHO, 2008 Standard: 10.0, 1.0, 10.0, 0.26, 0.20, 0.10
Table 3. Estimated cancer risks and hereditary effects of adult member of the public

| S/N | Sample ID | Total annual effective dose (mSv) | Fatality cancer risk to adult per year \( \times 10^{-6} \) | Lifetime fatality cancer risk \( \times 10^{-4} \) | Severe hereditary effects in adult per/y \( \times 10^{-7} \) | Estimated lifetime hereditary effects \( \times 10^{-6} \) |
|-----|-----------|-----------------------------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|
| 1   | SW1       | 0.00                              | 0.00                                          | 0.00                                          | 0.00                                          | 0.00                                          |
| 2   | SW2       | 33.0                              | 0.017                                         | 0.016                                         | 0.61                                          | 0.32                                          | 2.22                                          |
| 3   | SW3       | 193.0                             | 0.186                                         | 0.235                                         | 12.93                                         | 9.05                                          | 4.70                                          | 32.90                                        |
| 4   | SW4       | 137.0                             | 0.068                                         | 0.065                                         | 3.57                                          | 2.50                                          | 1.30                                          | 9.09                                         |
| 5   | SW5       | 212.0                             | 0.504                                         | 0.042                                         | 2.29                                          | 1.60                                          | 0.83                                          | 5.83                                         |
| 6   | SW6       | 0.169                             | 0.883                                         | 1.388                                         | 76.35                                         | 53.44                                         | 27.76                                         | 194.30                                       |
| 7   | SW7       | 0.00                              | 0.00                                          | 0.00                                          | 0.00                                          | 0.00                                          | 0.00                                          | 0.00                                         |
| 8   | SW8       | 0.079                             | 0.077                                         | 0.060                                         | 3.32                                          | 2.32                                          | 1.21                                          | 8.45                                         |
| 9   | SW9       | 13.63                             | 0.034                                         | 0.043                                         | 2.34                                          | 1.64                                          | 0.85                                          | 5.96                                         |
| 10  | SW10      | 9.018                             | 0.064                                         | 0.061                                         | 3.35                                          | 2.34                                          | 1.22                                          | 8.522                                        |
| 11  | SW11      | 1.271                             | 0.459                                         | 0.728                                         | 40.06                                         | 28.04                                         | 1.46                                          | 101.98                                       |
| 12  | SW12      | 13.63                             | 0.034                                         | 0.043                                         | 2.34                                          | 1.64                                          | 0.85                                          | 596.4                                        |
| 13  | SW13      | 1.206                             | 0.436                                         | 0.692                                         | 38.03                                         | 26.60                                         | 13.83                                         | 96.80                                        |
| 14  | SW14      | 76.0                              | 0.038                                         | 0.036                                         | 1.99                                          | 1.39                                          | 0.72                                          | 5.06                                         |
| 15  | SW15      | 20.60                             | 0.042                                         | 0.052                                         | 2.84                                          | 1.98                                          | 1.03                                          | 7.22                                         |
| 16  | SW16      | 1.177                             | 0.425                                         | 0.675                                         | 37.12                                         | 25.98                                         | 1.35                                          | 9.45                                         |
| 17  | SW17      | 142.55                            | 0.071                                         | 0.068                                         | 3.73                                          | 2.61                                          | 1.35                                          | 9.48                                         |
| 18  | SW18      | 93.40                             | 0.056                                         | 0.057                                         | 3.15                                          | 2.20                                          | 1.14                                          | 8.01                                         |
| 19  | SW19      | 107.72                            | 0.054                                         | 0.051                                         | 2.82                                          | 1.97                                          | 1.03                                          | 7.17                                         |
| 20  | SW20      | 0.146                             | 0.053                                         | 0.084                                         | 4.62                                          | 3.23                                          | 1.68                                          | 1.18                                         |
| **Mean** |       | 58.64                             | 0.19                                          | 0.24                                          | 13.43                                         | 9.40                                          | 3.39                                          | 61.67                                        |

Fig. 2. Comparison of activity concentration of \( ^{232} \text{Th} \) with ICRP, 2012 standard

The annual effective dose due to ingestion of the river water sampled was estimated for three different age groups: Infants, children and adults. The calculated total annual effective dose for different age groups as shown in Table 2 range from 0.00 to 212.0 mSv\(^{-1}\) for infants, 0.00 to 0.883 mSv\(^{-1}\) for children and from 0.00 to 1.388 mSv\(^{-1}\) for adult with average values of 58.64, 0.19 and 0.24 mSv\(^{-1}\) respectively. It can be observed from Fig. 5 that the radiation dose received by infants is relatively higher than that received by children and adults. The WHO [25] and UNSCEAR [19] reference levels of the effective dose for infants, children and adult due to one year continuous ingestion of various drinking water are 0.26, 0.20 and 0.10 mSv\(^{-1}\) respectively. The effective doses obtained were higher than the reference values for infants and adults that consume river water but that for children are within the reference level of 0.2 mSv\(^{-1}\). From the radiation protection point of view, life-long ingestion of these sampled river waters may cause significant radiological health problems.
In order to determine the radiation risk due to ingestion of $^{238}$U, $^{232}$Th and $^{40}$K in river water sampled, ICRP [20] methodology was adopted in the study and the results are shown in Table 3. The results of the cancer and non-cancer risk components were evaluated from the estimated total annual effective dose of the various age groups. The result of the estimated fatal cancer risk to adult per year in each of the stream water sampled ranged from $0.87 \times 10^{-6}$ to $76.35 \times 10^{-6}$ with its associated lifetime fatality cancer risk in adult of $1.18 \times 10^{-6}$ to $596.40 \times 10^{-6}$. This means that in terms of the lifetime fatality cancer risk to adult approximately 53 out of 10,000 may suffer some form of cancer fatality and for the lifetime hereditary effect approximately 596 out of 1000,000 may suffer some hereditary effects. The negligible cancer fatality risk value recommended by USEPA is in the range of $1.0 \times 10^{-6}$ to $1.0 \times 10^{-4}$ (i.e. 1 person out of 1 million to 10,000 persons suffering from some form of cancer fatality is considered trivial).
Comparing the estimated results of the lifetime fatality cancer risk in the present study with the acceptable risk factor, it can be seen that all estimated results of the lifetime fatality risk in adult member of the Nigerian population due to ingestion of radionuclide in the studied stream water are higher than the range of acceptable risk value recommended by USEPA.

4.2 Statistical Analysis

Basic statistics with statistical software package SPSS version 11.0 for windows was used to demonstrate the distribution and behavior of the measured radionuclide in stream water sand presented in Table 5. The statistical parameters determined includes the range (minimum-maximum), arithmetic mean (AM), arithmetic standard deviation (SD), median, mode, skewness, kurtosis and the type of frequency distribution for the three radionuclides for all the water samples.

The frequency distribution curves of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ are shown in Fig. 5. From Table 5, all the radiological parameters have positive skewness which shows that $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ have asymmetric distribution and only $^{40}\text{K}$ has a negative kurtosis indicating relatively flat distribution. Pearson’s correlation analysis was also carried out to ascertain if there are mutual relationship between the pairs of variables by calculating their linear correlation coefficient $R^2$. It is important to note that a positive correlation among variables indicates similar source and behavior in the given environment.

Results of the Pearson correlation coefficient among all the three studied radionuclide and the associated radiological parameters are presented in Table 6. From Table 6, it can be observed that positive correlation exists among the three radionuclides and all the radiological parameters except $^{238}\text{U}$ having a negative

### Table 4. Comparison of activity concentration of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in water samples of Imo River Rivers State Nigeria and other studies in different parts of the world

| Samples                  | Country       | $^{238}\text{U}$ (Bq l$^{-1}$) | $^{232}\text{Th}$ (Bq l$^{-1}$) | $^{40}\text{K}$ (Bq l$^{-1}$) | References |
|--------------------------|---------------|-------------------------------|-------------------------------|-------------------------------|------------|
| Stream OD W (Nigeria)    | Nigeria       | 0.59                          | 1.8                           | 27.7                          | [27]       |
| Stream OW                | Nigeria       | 4.62                          | 4.06                          | 42.57                         | [27]       |
| Stream water              | Nigeria       | 9.04±3.11                     | 2.28±0.57                     | 100.37±23.47                  | [1]        |
| Well OD                  | Nigeria       | 3.16                          | 2.38                          | 235.64                        | [27]       |
| Mineral bottled water    | Cameroon      | 0.022                         | 0.035                         | 0.107                         | [22]       |
| Portable water            | Nigeria       | 0.000833                      | 0.0005039                     | 0.4191                        | [26]       |
| Borehole water            | Nigeria       | 0.49                          | 0.30                          | 7.40                          | [28]       |
| Stream water              | Nigeria       | 2.02                          | 3.59                          | 10.43                         | This study |

### Table 5. Descriptive statistics of radiological parameters

|          | U-238 | Th-232 | K-40 | AEDE$_{\text{Infant}}$ | AEDE$_{\text{Children}}$ | AEDE$_{\text{Adult}}$ |
|----------|-------|--------|------|------------------------|--------------------------|------------------------|
| N Valid  | 18    | 18     | 18   | 18                     | 18                       | 18                     |
| missing  | 0     | 0      | 0    | 0                      | 0                        | 0                      |
| Mean     | .6739 | 1.9161 | 6.9544 | 58.6442               | .1945                    | .2442                  |
| Std. error of mean | .29716 | .68676 | 1.77320 | 16.90533 | .05726 | .08878 |
| Median   | .1800$^a$ | .3636$^a$ | 3.6700$^a$ | 18.2767$^a$ | .0660$^a$ | .0607$^a$ |
| Mode     | .00   | .00    | .00   | .00                    | .03                      | .04                    |
| Std. Deviation | 1.26076 | 2.91367 | 7.52305 | 71.72324 | .24293 | .37666 |
| Variance | 1.590 | 8.489  | 56.596 | 5144.224 | .059 | .142 |
| Skewness | 2.067 | 1.431 | .746 | 1.027 | 1.679 | 2.087 |
| Std. Error of Skewness | .536 | .536 | .536 | .536 | .536 | .536 |
| Kurtosis | 3.795 | 1.065 | -.741 | -.261 | 2.420 | 4.132 |
| Std. Error of Kurtosis | 1.038 | 1.038 | 1.038 | 1.038 | 1.038 | 1.038 |
| Range    | 4.36  | 9.00   | 22.11 | 211.92    | .87                       | 1.37                   |
| Minimum  | .00   | .00    | .00   | .08        | .02                      | .02                    |
| Maximum  | 4.36  | 9.00   | 22.11 | 212.00    | .88                      | 1.39                   |
| Sum      | 12.13 | 34.49  | 125.18 | 1055.60   | 3.50                     | 4.40                   |

$^a$ Calculated from grouped data
Fig. 5. Frequency distribution of $^{238}$U, $^{232}$Th and $^{40}$K in stream water

Table 6. Pearson correlations of measured parameters

|       | $^{238}$U | $^{232}$Th | $^{40}$K | AEDEinfant | AEDEchild | AEDEadult |
|-------|-----------|------------|-----------|------------|-----------|-----------|
| $^{238}$U | 1         |            |           |            |           |           |
| $^{232}$Th | -0.12834 | 1          |           |            |           |           |
| $^{40}$K  | -0.36877  | -0.18064  | 1         |            |           |           |
| AEDEinfant | -0.37325 | -0.10098  | 0.923147  | 1          |           |           |
| AEDEchild | -0.03014 | 0.680037  | -0.152366 | -0.0826631 | 1         |           |
| AEDEadult | 0.027668 | 0.642057  | -0.42099  | -0.3598336 | 0.89566772 | 1         |

Correlation with AEDE$_{\text{children}}$ and AEDE$_{\text{adult}}$ indicating that uranium did not contribute to gamma emission on children and adult. Strong correlation were observed between $^{232}$Th and $^{40}$K while $^{238}$U is weakly correlated with $^{232}$Th and $^{40}$K.

The strong positive correlation between $^{232}$Th and $^{40}$K shows that their origin and behavior in the coastal environment are the same while weak positive relationship between $^{238}$U and the other two indicates that they may have the same origin but their behavior in the river environment differs. All the three radionuclides have strong positive correlation coefficient with the radiological parameters except for Uranium-$^{238}$ that showed negative correlation with AEDE$_{\text{children}}$ and AEDE$_{\text{adult}}$. This means that...
two of the radionuclide only contributed significantly to gamma-ray emission at the sampling points.

5. CONCLUSION

The activity concentrations of $^{232}$Th and $^{40}$K measured in river water collected from Otamiri tributary and Imo River at the Abia/Rivers boundary to over seven kilometers along the Imo River exceeded the reference level of 1.0 and 10.0 Bq$^{-1}$ while the activity concentration of $^{238}$U measured are within the reference level of 10.0 Bq$^{-1}$. The mean total annual effective dose determined for infant, children and adult population that drink river water from the Imo River are 58.64, 0.19 and 0.24 mSv$^{-1}$ respectively. AEDE estimated for infant are 94% higher than the reference Level of 0.26 mSv$^{-1}$ and also higher than that for children and adult.

The estimated fatal cancer risk to adult per year and the lifetime hereditary effect shows that 53 out of 10,000 population may suffer some form of cancer fatality and approximately 596 out of 1000,000 might suffer some hereditary effects. Statistically all the radionuclide showed positive skewness and kurtosis except $^{238}$U. Pearson correlation of the radionuclides and all the radiological parameters showed positive correlation between $^{232}$Th and $^{40}$K which indicate same origin and behavior in the coastal environment. $^{238}$U showed negative correlation with the radiological parameters which shows that $^{238}$U did not contribute to gamma emission and probably had a different origin.

The result of this study showed that the activity of oil bunkering along the creeks, river shore has impacted negatively on the river water which in turn might lead to radiation related health challenges to infants and adult citizens of the area. Therefore, citizens of the area should desist from drinking water from Imo river and its tributaries.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Ajayi JO, Adedokun O, Balogun BB. Levels of radionuclide content in stream water of some selected rivers in Ogbomosho land, South West Nigeria. Journal of Environmental and Earth Sciences. 2012;4(a):835–837.
2. Arogunjo MA, Farai IP, Furape IA. Impact of oil and gas industry to the natural radioactivity distribution in the Delta region of Nigeria. Nigerian Journal of Physics. 2004;16:131-136.
3. Awirio GO, Agbalagba OE. Survey of gross alpha and gross beta radionuclide activity in Okpara creek, Delta state, Nigeria. Asia Network for science Information Journal of Applied Science. 2007;7(22):3542–3547.
4. Kulmun AV, Wonde SN, Mafejeana A. Radioactivity dose calculation and water quality evaluation guidelines for domestic water use. Department of Water Affairs and Forestry Document; 2002.
5. UNSCEAR. United Nations scientific committee on the effects of atomic radiation. Sources and Effects of Ionizing Radiation (Report to the General Assembly, United Nations, New York); 2000.
6. Apecza OS, Szabó Zoltán. Radioactivity in ground water a review. U.S. Geological Survey Water Supply Paper. 1988;2325: 50–57.
7. El-Tahaway MS, Farouk MA, Ibraheim MN, El Mongery SAM. Natural and Artificial Radioactivity in the Suez Canal bottom sediment and stream water. Journal of Environmental Radioactivity. 1994;47(2): 201-212.
8. Hany El-Gamal, Abdallah IA, EL- Mageed. Natural radioactivity in water samples from Assiut city, Egypt. International Journal of Pure and Applied Science and Technology. 2014;22(1):44-52.
9. Obed RI, Farai IP, Jibiri NN. Population dose distribution due to soil radioactivity concentration levels in 18 cities across Nigeria. Journal of Radioactivity Protection. 2005;2.
10. Kalin M, Wheeler WN, Meinrath G. The removal of uranium from mining waste water using algal/microbial biomass. Journal of Environmental Radioactivity. 2005;78:151–177.
11. West Larry. World water day: A billion people worldwide lack safe drinking water. Wikipedia; 2006.
12. Alfahia AA Osman, Isam Salih, Ibrahim A Shaddad, Saif El Din, Siddeeg MB, Hatem Eltaheb, Hajoldriss, Wadi Hamza, Yousif EH. Investigation of natural radioactivity...
levels in water around Kadugli, Sudan. Applied Radiation and Isotopes. 2008;68: 1650–1653.

13. Taskin H, Karavus MA, Topuzoglu BA, Hindiroglu S, Karahan G. Radionuclide concentrations in soil and life-time cancer risk due to radioactivity in Kirklareli, Turkey. Journal of Environmental Radioactivity. 2009;100:49-53.

14. Ahmed NK. Natural radioactivity of ground and drinking water in some areas of upper Egypt. Turk. J. Eng. Environ. Sci. 2004;28: 344-354.

15. Jibiri NN, Mabawonku AA, Oriade Ujiaegbedion SJ. Natural radionuclide levels in soil and water around a cement factory in Ewekoro, Ogun State, Nigeria. J. Phy. 1999;5(11):12-16.

16. Guogang J, Giancario T. Estimation of radiation doses to members of the public from intakes of some important naturally occurring radionuclides. J. Environ. Rad. 2007;76:654-72.

17. ICRP. Publication 26. Recommendation of the International Commission on radiological protection. Annals of the ICRP; 1977.

18. Oseji JO. Aquifer systems of Ndokwaland, Delta State, Nigeria. IJRRAS. 2010;5(3): 344-354.

19. UNSCEAR. Sources and effect of ionizing radiation. Unscear, 2008 Report to the General Assembly with Scientific Annesvol II; 2008.

20. ICRP Publication. Annals of the ICRP. Compendium of Dose Coefficients Based on ICRP Publication 119; 2012.

21. Isinkaye MO, Emelue HU. Natural radioactivity measurements and evaluation of radiological hazards in sediment of Oguta Lake, South East Nigeria. Journal of Radiation Research and Applied Sciences. 2015;8:459-469.

22. Ndontchuen MM, Simo A, Njuelem EJM, Beyala JF, Kryeziu D. Preliminary study of natural radioactivity and radiological risk assessment in some mineral bottled water produced in Cameroon. International Journal of Science and Technology. 2013;3(5):271–276.

23. Jibiri NN, Mabawonku AA, Oriade Ujiaegbedion SJ. Natural radio nuclide levels in soil and water around a cement factory in Ewekoro, Ogun State, Nigeria. Journal of Physics. 1999;5(11):12–16.

24. Senthilkumar B, Dharamani V, Ramkumar S, Philominathan P. Measurement of gamma radiation levels in soils samples from Thanjavur using x-ray spectrometry and estimation of population exposure. Journal of Medical Physics. 2010;35(1):48-53.

25. WHO. Guidelines for drinking water quality. Incorporating First Addendum. Recommendations 3rd Radiological Aspect Geneva. 2008;1.

26. Onoja RA, Adeyemo DJ, Okoh S. Determination of natural radionuclide concentrations in portable water supply of Northern part of Kaduna state. Research Journal of Applied Sciences, Engineering and Technology. 2014;7(14):2905-2907.

27. Jibiri NN, Chijioke MN, George OA. Radionuclide contents and physico-chemical water quality indicators in stream, well and borehole water sources in high radiation area of Abeokuta, Southwestern Nigeria. Journal of Water resources and Protection. 2010;2:291–297.

28. Meltem D, Gursel K. Natural radioactivity in various surface waters in Adana, Turkey. Desalination. 2010;261:126-130.

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