Risk Assessment of Natural Radionuclides in Surface and Ground Water of Oil and Gas Producing Communities, Rivers State, Nigeria

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Abstract: The natural radioactivity and radiological health risk associated with the use of water from hand-dug wells, tap/boreholes and river/creeks in three communities with history of oil spillage, gas flaring, oil bunkering and operation of illegal artisanal oil refining activities in Rivers State, Nigeria was here assessed and measured with gamma ray spectroscopy. The results showed that the mean activity concentration of water samples for hand-dug well water ranged from 11.94±1.12 to 12.77± 1.12Bq/l for ^{238}U, 7.88±1.05 to 9.20±1.82Bq/l for ^{232}Th and 13.50±0.62 to 20.13±3.88Bq/l for ^{40}K. The mean activity concentration for the tap/borehole water ranged from 2.42±0.45Bq/l to 5.31±0.80Bq/l for ^{238}U, 1.02±1.01 to 2.24±0.90Bq/l for ^{232}Th and 9.71±5.04 to 11.73±3.77Bq/l for ^{40}K. The ^{238}U, ^{232}Th and ^{40}K activity concentrations in the river/creek waters ranged respectively from 6.81±0.80 to 8.82±1.20Bq/l, 4.25±0.65 to 9.52±1.16Bq/l and 25.01±3.23 to 30.03±1.96Bq/l. These values are higher than the mean activity concentrations of the 3 radionuclides in the control samples and some are higher than the international permissible standards. Results further revealed that there was statistically significant difference at $p<0.05$ in the mean variations of the activity concentrations of natural radionuclides in the water supply sources within the three communities and the control. The committed annual effective dose and excess lifetime cancer risks are radiologically higher than the permissible limits. Therefore, children and adults in the three communities are susceptible to high dose related diseases and have high chances of contracting cancer when water is ingested from the three water supply sources since the committed annual effective dose and excess lifetime cancer risk are respectively over 1.16 times and 8 times higher than the UNSCEAR and USEPA international permissible limits of 0.29mSv/y and $1x10^{-4}$ mSv/y.

Keywords: Artisanal oil Refining, Committed Annual Effective Dose, Excess Lifetime Cancer Risk, Oil Bunkering, Radionuclides

1. Introduction

Water as a universal solvent is a very important parameter of environmental science because it is indispensible to human life. Water sources like rivers, springs, wells, boreholes and other fresh water bodies provide water for drinking and domestic purposes including cooking and will generally constitute an exposure pathway for contaminants to reach the population (Fasunwon et. al., 2008). Water, no matter its source does not exist in pure form for appreciable length of time in nature. Even while waterfalls as rain, it picks up small amount of contaminants from the atmosphere and moves as it filters through the ground (Ajayi et. al., 2012). 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 (Ononugbo and Ogan, 2017). Naturally occurring radionuclides are present in the food we eat, in the water we drink and use for domestic purposes through weathering and recycling of terrestrial minerals and rocks. Radionuclides may be deposited from the air or may be released to the water through oil spill in surface water, oil exploration and exploitation that takes place in an environment and/or human activities such as bunkering and illegal crude method of crude oil refining along river course. Radionuclides may also be released to water from the ground through erosion where they are washed away from soils and seeps into underground water. Water becomes contaminated as it picks up radioactive materials from the surrounding rocks, soils or cracked cement as it flows past (Ononugbo and Ogan, 2017). Domestic sewage, feedlots and surface runoff, and other pollution sources get surface and groundwater contaminated. The subterranean aquifers may become contaminated in areas where the subsurface geology permits

rapid downward movement of water sources from the surface or where groundwater/well water sources are tapped near the surface (Jibiri et. al., 2010). Ingestion of radionuclides through drinking water and food intake accounts for a substantial part of the average radiation doses to various organs of the body and also represents one of the important pathways for long-term health considerations (UNSCEAR, 2000). Long-term exposure to radionuclide like thorium through inhalation and ingestion has severe health effects such as chronic lung diseases and bone cancer, bone weakening, cranial leucopoenia, and necrosis of the mouth and nasal tumors (Jibiri et. al., 2011). While long-term exposure to radium increases the risk of developing several diseases like lymphoma, bone cancer and diseases that affect the formation of blood (leukemia and aplastic anemia), lung cancer, cancer of the pancreas, cataracts, sterility and atrophy of the kidney (Taskin et. al., 2009).

Between 2016 and 2021 Rivers State which is the oil and gas hub of Nigeria experienced air pollution in the form of dispersed particulate matter known as black soot. This soot contains radionuclides and when deposited in any of the environmental media, could elevate the concentration of naturally occurring radionuclides in the environment. Naku (2022) asserted that the black soot were products of emission from operators of illegal crude oil refining in all the oil bunkering centres located within the communities in the State. Aside from being a threat to air quality leading to respiratory diseases, the soot can affect farming activities as acid rainfall could potentially impact on crop yield and harvest. Again, the soot poses a threat to animals, aquatic lives, as well as rivers and streams which are sources of water supply for most households in the communities rendering the waters unclean arising from deposition of soot into these surface water bodies (Alikor, 2022). The inadvertent discharges of petroleum hydrocarbons or petroleum derived wastes streams from activities of oil and gas production operating companies in the communities are toxic to the coastal waters, soils and sediments near the discharge point (Azpecza and Szabo, 1988). Besides surface water bodies, groundwater is very important as it accounts for about 88% of safe drinking water in rural areas where population is widely dispersed and the infrastructure needed for treatment and transportation of surface water does not exist (Amakom, 2007). Hand-dug wells however, are still the most common source of groundwater in rural communities, and a large percentage of the rural population depends on these wells for their water supply (USEPA, 2006). River water also serves as a major source of drinking water in the rural area for the human race, thus it is important to investigate the levels of radionuclide element in river water (Ajayi et. al., 2012). This study therefore assesses the radionuclides content in the different sources of water for drinking and domestic use by the inhabitants of three communities with history of crude oil spill, oil bunkering/illegal oil refining activities, and to evaluate the radiological health risk associated with these activities when water from these different sources is ingested by the inhabitants.

2. The Study Area

The study areas are three communities comprising Eleme, Bunu-Tai and Bodo. Eleme lies within Latitude 04°46'37.6"N and Longitude 007°07'51.0"E, Bunu-Tai lies within Latitude 04°45'41.0"N and Longitude 007°14'29.4"E while Bodo lies within Latitude 04°44'46.2"N and Longitude 007°06'32.1"E. The three communities respectively belong to Eleme Local Government Area (LGA), Tai LGA and Gokana LGA of Rivers State, Nigeria (Figure 1). The general topography is relatively flat lying and consists of terrestrial and marine environment. Due to previous crude oil spills that cover the land and surface water bodies, the terrestrial environment has patchy regenerating vegetation which consisted mostly of scanty and secondary type residual grasses while the surface water bodies have oil films with dark to brownish colouration. Each of the three communities is within 1,000m radius of the spilled sites, gas flaring at flow stations, oil bunkering and illegal artisanal oil refining activities.

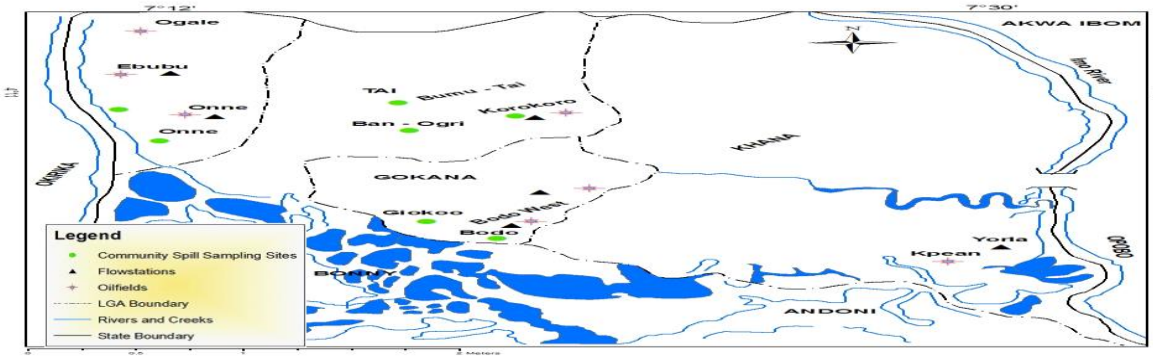

Figure 1: Map of the Study Area (NGSA, 2010)

2.1 Regional Geology

The study area falls within the Niger Delta region which is made up of thick clastic sedimentary sequence with age ranging from Eocene to Recent and it sits astride the Niger flood plains, which overlies the Benin formation that is often called the coastal plain sand (Tattam, 1943). This formation consists predominantly of coarse grained sandy soils with few shale intercalations. The unconsolidated, highly porous sands of the Benin formation is a fresh water bearing sands zone (Amajor, 1991), and all aquifers in this region are located within this lithio-stratigraphic unit. The Benin formation comprises multiple layers of clay, clay conglomerates, peat and/or lignite all of variable thickness and texture and covered by overburden soil (Short and Stauble, 1967).

3. Materials and Methods

3.1 Sample Collection and Preparation

Eight water samples were collected from three communities with history of crude oil spill, gas flaring, illegal artisanal oil refining and oil bunkering activities (table 1 and figure 2). The simple random sampling method was adopted. Water samples were collected from rivers/creeks, taps/boreholes and hand-dug wells that serve as community water supply sources. Selection of sample locations was based on proximity of water source to spill site and industrial areas. Three water samples used as control were collected from river, tap/borehole and hand-dug well from non-oil bearing communities (ie with no history of oil production and oil spillages) located at 55km away from Ogoniland. Well water samples were collected manually in the early hours of the day while river/creek water samples were collected near the middle of the water body in the early hours using wading gear and a 2-metre extendable metal grab. Samples were collected against the flow of water by submerging a water container to a depth of 10-20 cm under the surface. Tap/borehole water samples were collected with treated containers at laminar flow rate after first turn on at full capacity for 5 minutes to purge the plumbing system of any water that might contaminate the sample to reduce radon loss.

To minimize contamination, the collection of the water samples from the three water supply sources was done each with 2-litre container. The 2-litres containers were first rinsed three times with water and acid washed with 20ml±1ml Conc. HNO_3 before using it to collect the samples so as to avoid adsorption of the radionuclides on the walls of the containers. For each sample, about 1% airspace was left for thermal expansion before it was tightly sealed and taken to the laboratory to store for 30 days to ensure that no radon loss occurs, and to reach a state of circular equilibrium between radium isotopes and their daughters. Samples` were then transferred into a 1-litre Marinelli beaker after filtration to remove all solid particles in the water. It was further processed through evaporation until 0.5 litres remained in the beaker and was then stored in a desiccator to allow it cool and to prevent it from absorbing moisture.

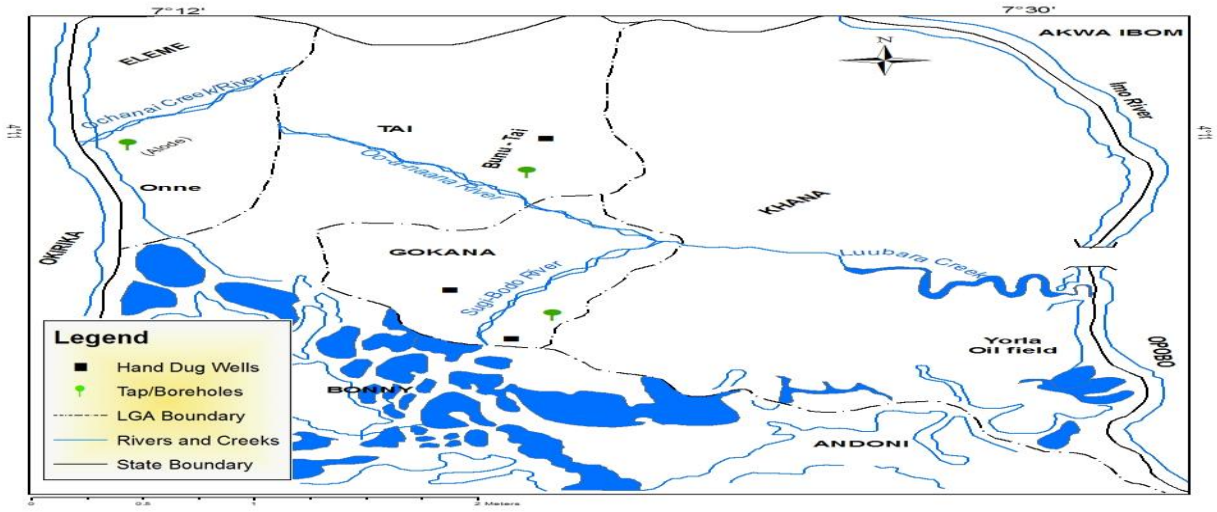

Figure 2: Map Showing Water Sampling Points in the Study Area

Table 1: Communities where water samples were collected

Community	River Water	Tap/borehole	Hand-dug well
Bunu-Tai	Oo-a-naana River	Bunu-Tai Tapwater	Bunu-Tai Well
Bodo	Sugi-Bodo River	Bodo Tapwater	Bodo Well
Eleme	Ochanai creek	Alode Tapwater	-

3.2 Sample Analysis

The activity of natural radionuclides of the prepared water samples were counted at the Centre for Energy Research and Training, Zaria with gamma ray spectrometer detector for 29000 seconds at 900V to produce strong peaks at gamma emmiting energies so as to assess the natural radionuclide contents in the water samples. Due to the smaller life time of the daughter radionuclides in the decay series of ^{232}Th and ^{238}U, the activities of ^{238}U was determined from the average activities of ^{214}Pb at 352kev and ^{214}Bi at 609Kev while that of ^{232}Th was determined from average activities of the decay products of ^{208}Ti at 583kev and ^{228}Ac at 911Kev. The activity of ^{40}K content was determined from strong peaks at gamma emitting energies of 1,460Kev (1.460 MeV). The background spectra measured under the same conditions for both the standard and sample measurements were used to correct the calculated sample activity concentrations. The activity concentration (C_w) in Bq/l of ^{238}U, ^{232}Th and ^{40}K in the water samples was calculated after subtracting decay correction using the expression:

$$C_w\,(Bq/l) = \frac{C_a}{\varepsilon_y \times M_s \times t_c \times P_\gamma} \quad \ldots\ldots (1)$$

Where: C_w is the sample concentration, C_a is the net peak area of a peak at energy, ε_y is the efficiency of the detector for a gamma energy of interest, M_s is the sample mass, t_c is the total counting time, P_γ is the abundance of the γ-line in a radionuclide.

3.3 Radiation Hazard Indices

To assess the health status of irradiated persons in an environment, UNSCEAR (2008) and ICRP (2012) recommended the following hazard indices for radiological risk assessment:

(i) Radium Equivalent Activity (Ra_{eq}): Since 98.5% of the radiological hazard of uranium series is due to radium and its decay products, ^{238}U is replaced with concentration of ^{226}Ra in hazard assessment. As a result of non-uniform distribution of ^{226}Ra, ^{232}Th and ^{40}K in environmental samples, uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) to compare the activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. This index makes possible the use of a single regulatory limit on radionuclide rather than having to limit uranium, thorium and potassium separately (Farai and Ademola, 2005). This index also provides a useful guideline in regulating the safety standards on radiation protection for the general public residing in an area. The Ra_{eq} index represents a weighted sum of activities of ^{238}U, ^{232}Th and ^{40}K and it is based on the estimation that 370Bq/kg of ^{238}U, 259Bq/kg of ^{232}Th and 4810Bq/kg of ^{40}K provide the same gamma radiation dose rates and it is given as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \ldots\ldots (2)$$

Where A_{Ra}, A_{Th} and A_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively measured in Bq/l. The allowable limit of Ra_{eq} in environmental samples is 370 Bq/l (UNSCEAR, 2000, 2008).

(ii) Absorbed Dose Rate (D): It is imperative to calculate the absorbed dose rate based on the fact that radiation exposure pathways involved inhalation and ingestion of radioactive pollutants. The absorbed dose rates (D) due to gamma radiation from ingested radionuclide in water was calculated from ^{238}U, ^{232}Th and ^{40}K activity concentration values assuming that other radionuclides such as ^{137}Cs, ^{90}Sr and ^{235}U decay series are neglected since they contribute very little to the total dose from environmental background.

$$D = 0.462A_u + 0.604A_{Th} + 0.0417A_K \quad \ldots\ldots (3)$$

Where: A_u, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/kg respectively. The unit of D is ηGyh^{-1}.

(iii) Annual Gonadal Dose Equivalent (AGDE): The gonads, the bone marrow and the bone surface cells are considered as organs of interest by UNSCEAR (2000, 2008) and ICRP (2012) because of their sensitivity to radiation. An increase in AGDE has been known to affect the bone marrow, causing destruction of the red blood cells that are then replaced by white blood cells. This situation results in a blood cancer called leukemia which is fatal (Avwiri et. al., 2014). Therefore, the AGDE due to the activities of ^{238}U, ^{232}Th and ^{40}K in the water that is consumed by inhabitants in a community or by using it for various purposes was evaluated by the following equation (Avwiri et. al., 2012; UNSCEAR, 2008):

$$AGDE\ (mSv/y) = 3.09A_U + 4.18A_{Th} + 0.314A_K \quad (4)$$

Where A_u, A_{Th} and A_K are respectively the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/l.

(iv)The External Hazard Index (H_{ex}): Most radionuclides occur naturally in terrestrial soils and rocks and upon decay, these radionuclides finds its way into surface and underground water bodies thus producing an external radiation field to which all human beings are exposed to. In terms of dose, the principal primordial radionuclides are ^{232}Th, ^{238}U and ^{40}K. Thorium and uranium head series of radionuclides that produce significant human exposure. Therefore the external hazard index becomes another parameter of interest. It is defined as (UNSCEAR, 2000; Beretka and Mathew, 1985):

$$H_{Ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

Where A_u, A_{Th} and A_K are respectively the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/l. The value of this index (H_{ex}) must be less than unity for the radiation hazard to be negligible (UNSCEAR, 2008) while H_{ex} equal to unity corresponds to the upper limit of radium equivalent dose of 370Bq/kg (Beretka and Mathew, 1985).

(v) The Internal Hazard Index: Radon (Rn) is the product of natural decay of radium (Ra), which is nearly in all rocks and finds its way into water bodies when it weathers and the soils are blown or eroded/carried into surface waters. The internal exposure to radon is very hazardous to the respiratory organs and can lead to respiratory diseases like asthma and cancer (Tufail et. al., 2007). Hence it has become imperative to evaluate and quantify the internal hazard index H_{in} by the relation (UNSCEAR, 2000; Beretka and Mathew, 1985):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

Again, the value of the internal hazard index (H_{in}) must be less than unity for the radiation hazard to be negligible (UNSCEAR, 2000).

(vi)The Representative Gamma Index ($I_{\gamma r}$): This is also known as gamma radiation representative level index (RLI). This index is used to estimate the gamma radiation hazard associated with the natural radionuclide in specific investigated samples. It is a screening tool for identifying materials that become of health concern when used for construction and other purposes (Tufail et. al., 2007). The representative gamma index is given by OCED/NEA (1979):

$$I_{\gamma r} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \quad (7)$$

(vii)The Committed Annual Effective Dose (AEDE): The committed annual effective dose to an individual due to the consumption of radionuclides from water is the arithmetic summation of the effective dose of the three measured radionuclides and was estimated using the relation (UNSCEAR, 2008):

$$AEDE = I_w . A_w . \Sigma\, C(U, Th, K); \text{ OR} \quad (8)$$

$$AEDE = I_w\, [A_{wU}.C_U + A_{wTh}.C_{Th} + A_{wK}.C_K] \quad (9)$$

Where I_w is the daily intake or consumption of water per person. WHO (2012) considers 3 age groups for the assessment of radiation doses. These are infants aged 1 year old, children aged 10 years old and below, and adults aged 17 years old and above. The daily intake or consumption of water is taken as 2 litres per day for adults (≥ 17 years), 1 litre per day for children (≤ 10 years) and 0.75 litres per day for infants (ICRP, 2012; IAEA, 2004; WHO, 2011) multiplied by 365 days (e.g 2x365= 730 litres per year for adults). A_w is the activity concentration of radionuclides in water samples (Bq/l); A_{wU}, A_{wTh} and A_{wK} are respectively the activity concentration of ^{238}U, ^{232}Th and ^{40}K in Bq/l in the water samples, and C (i.e C_U, C_{Th} and C_K) is the ingestion coefficient or the dose conversion factors of the specific radionuclide (Bq/l) which were extracted from ICRP (2012) and IAEA (2004).

(viii) Excess Lifetime Cancer Risk (ELCR): This is the probability of developing cancer over a lifetime at a given exposure level and it is presented as a value representing the number of extra cancers expected in a given number of people on exposure to a carcinogen at a given dose (Ononugbo et. al, 2013). The Excess lifetime cancer risk (ELCR) is given as (Taskin et. al., 2009):

ELCR = AEDE x DL x RF ………………………………………………………………….. (10)

Where AEDE is the annual committed effective equivalent dose, DL is average duration of life (estimated to be 70 years for Nigeria) and RF is the risk factor (Sv^{-1}) i.e fatal cancer risk per sievert. For stochastic effects, ICRP (2012) uses RF as 0.05 for members of the public. In this study, the probability of children (≤ 10 years) and adults (≥ 17 years) in developing cancer over a lifetime at a given exposure rate from ^{238}U, ^{232}Th and ^{40}K was computed.

3.4 Statistical Analysis

Due to the oil bunkering/artisanal oil refining activities, previous occurrence of oil spillages and gaseous emission of particulate matter from flare stacks at flow stations in the investigated communities, statistical analysis using SPSS version 20 software was performed on the water samples data of the mean activity concentration among the three communities. The essence of the statistical analysis was to establish if the different activities associated with oil mining operations in the area has in any way affected the activity concentration levels of the radionuclides in the water sample sources. A one way Analysis of Variance (ANOVA) was employed to ascertain if there is a significant difference in the mean activity concentrations of radionuclides in the water samples from the various communities and the control. In using this statistical tool, two hypotheses were propounded, the null (H_o) and the alternative hypothesis (H_1) which states as follows:

H_o: There is no significant difference in the mean activity concentration values of natural radionuclides in the water samples collected from various sources in the three communities and the control

H_1: There is a significant difference in the mean activity concentration values of natural radionuclides in the water samples collected from various sources in the three communities and the control

The statistical analysis were performed at 95% confidence interval and the Duncan post hoc test was applied to determine statistically significant differences among individual means at $p<0.05$.

4. Results and Discussion

4.1 Statistical Analysis Assessment of Water Radioactivity: The mean activity concentration of water samples for the hand-dug well water ranged from 11.94±1.12 to 12.77± 1.12Bq/l for ^{238}U; 7.88±1.05 to 9.20±1.82Bq/l for ^{232}Th; and 13.50±0.62 to 20.13±3.88Bq/l for ^{40}K. The mean activity concentration of water samples for the tap/borehole water ranged from 2.42±0.45Bq/l to 5.31±0.80Bq/l for ^{238}U; 1.02±1.01 to 2.24±0.90Bq/l for ^{232}Th; and 9.71±5.04 to 11.73±3.77Bq/l for ^{40}K. While the ^{238}U, ^{232}Th and ^{40}K activity concentrations in the river/creek waters ranged respectively from 6.81±0.80 to 8.82±1.20Bq/l, 4.25±0.65 to 9.52±1.16Bq/l and 25.01±3.23 to 30.03±1.96Bq/l. These values are higher than the mean activity concentrations of the three radionuclides in the control samples (Table 2). The mean activity of ^{238}U, ^{232}Th and ^{40}K in the hand-dug well water samples at Bunu-Tai and Bodo, and the mean activity concentration of ^{232}Th and ^{40}K in the tap/borehole water respectively at Bunu-Tai and Bodo communities were above the WHO (2011) recommended standards of 10Bq/l for ^{238}U, 1.0Bq/l for ^{232}Th and 10Bq/l for ^{40}K. Again, the mean activity values of ^{232}Th and ^{40}K in the river/creek water samples at Eleme,

Bunu-Tai and Bodo communities were higher than the international standard. The implication is that the hand-dug well water, tap/borehole water and water from river/creek in those communities are radiologically not fit and safe for consumption because they are contaminated by natural radionuclides from anthropogenic sources. These results are higher than those obtained by Avwiri et. al (2014) on the level of natural radioactivity in river bank and the river water along Mini-Okoro/Oginigba creek in PortHarcourt. It is also higher than the works of Maxwel et. al (2015) on the radioactivity levels of groundwater at Dei-Dei and Kubwa areas of Abuja, North central Nigeria. Nevertheless, the radioactivity result of this study is lower than the works of Ononugbo et al. (2013) on the assessment of surface and groundwater around oilfields and host communities at Ogba/Egbema/Ndoni LGA of Rivers State. It is also lower than the values from the works of Chad-Umoren and Nwali (2013).

Analysis of variance (ANOVA) was employed to determine if the activity concentration of radionuclides in water samples has been elevated in the three communities as compared to the control sample, hence the data in table 2 were subjected to statistical testing. The result of the one way ANOVA test at 95% confidence interval revealed that there was a statistically significant difference in the mean activity concentration of natural radionuclides in the water samples from various water supply sources in the three communities compared to the control (Table 3). The null hypothesis was therefore rejected. This is an indication that the community water supply sources have been enriched with the three radionuclides. Multiple comparison tests identified the communities whose water supply source contributed to this significant difference and the result showed that: (1) ^{238}U radionuclide contents in the water samples at the control well, Bunu-Tai borehole, Bodo borehole, Eleme river, Bodo well and Bunu-Tai well are statistically significantly different from each other at $p<0.05$ in the following order: Control well<Bunu-Tai borehole<Bodo borehole<Eleme river<Bodo well< Bunu-Tai well; (2) ^{232}Th radionuclide contents in the water samples at the control well, Eleme borehole, Bunu-Tai river, Bodo river and Bodo well are statistically significantly different from each other at $p<0.05$ in the following order: Control well<Eleme borehole<Bunu-Tai river<Bodo river<Bodo well; and (3) ^{40}K radionuclide contents in the water samples at the control well, Bunu-Tai borehole, Bodo borehole, Eleme borehole, Bodo well, Bunu-Tai well, Eleme river, Bunu-Tai river and Bodo river are statistically significantly different from each other at $p<0.05$ in the following order: Control well<Bunu-Tai borehole<Bodo borehole<Eleme borehole<Bodo well<Bunu-Tai well<Eleme river<Bunu-Tai river<Bodo river.

Table 2: Mean Activity of Radionuclides in the community Water Samples with Control

S/No	Sample Name	Specific Activity Concentration (Bq/l)		
		^{238}U	^{232}Th	^{40}K
1	Eleme Well Water	-	-	-
2	Bunu-Tai Well Water	12.77±1.12	9.20±1.82	20.13±3.88
3	Bodo Well	11.94±1.12	7.88±1.05	13.50±0.62
4	Alode (Eleme) Tap/borehole	2.42±0.45	2.24±0.90	11.73±3.77
5	Bunu-Tai Tap/borehole	3.82±2.20	1.02±1.01	9.71±5.04
6	Bodo Tap/borehole	5.31±0.80	1.20±0.51	11.21±2.10
7	Ochanai Creek (Eleme)	6.81±0.80	9.52±1.16	25.01±3.23
8	Oo-a-naana River (Bunu-Tai)	8.48±1.84	4.25±0.65	27.83±1.30
9	Sugi-Bodo River (Bodo)	8.82±1.20	5.31±0.91	30.03±1.96
	CONTROL WATER SAMPLES			
1	Hand-dug Well	1.32±0.14	0.90±0.01	5.30±0.07
2	Tap/borehole	2.26±0.08	0.43±0.08	5.58±0.03
3	River/Creek	2.94±0.10	0.53±0.01	6.23±0.06
WHO (2011)		**10.0**	**1.0**	**10.0**

Table 3: ANOVA for ^{238}U, ^{232}Th and ^{40}K in the Water Samples

		Sum of Squares	df	Mean Square	F	Sig.
Uranium-238	Between Groups Within Groups	477.273 2.416	10 22	47.727 .110	434.615	.000

	Total	479.689	32			
Thorium-232	Between Groups Within Groups Total	258.564 4.349 262.913	10 22 32	25.856 .198	130.786	.000
Potassium-40	Between Groups Within Groups Total	3517.058 .864 3517.921	10 22 32	351.706 .039	8959.619	.000

4.2 Radiological Hazard Assessment of Water Samples: The mean values of radium equivalent activity, absorbed dose rates, annual gonadal equivalent dose, internal and external hazard indices, and gamma representative index due to the presence of ^{238}U, ^{232}Th and ^{40}K in the water samples are all respectively within the permissible standards (Table 4). These mean values of absorbed dose rate of water samples from the three supply sources are lower than the 28.79±11.10ηGy/h obtained by Chad-Umoren and Nwali (2013) at Okrika creek due to discharge of wastes from the PortHarcourt refinery. The result of the absorbed dose rate for river/creek water in this study is 6.6 times higher than the value obtained for the water around Mini-Okoro/Oginigba creek in PortHarcourt by Avwiri et. al. (2014). The fact that the values of the absorbed dose rate is higher in the hand-dug well water than the river/creek and tap/borehole waters implies that those drinking or using the water from the hand-dug well are more exposed to radiation of the three natural radionuclides present in the water. The contribution of ^{238}U, ^{232}Th and ^{40}K to the absorbed dose in the hand-dug well water samples are 49%, 45% and 6% respectively with ^{238}U contributing the highest to the absorbed dose rates (Figure 3) while the contribution of ^{238}U, ^{232}Th and ^{40}K to the absorbed dose in the tap/borehole water samples are 56%, 29% and15% respectively with ^{238}U contributing the highest to the absorbed dose rates (Figure 4). The contribution of ^{238}U, ^{232}Th and ^{40}K to the absorbed dose for the river/creek water samples are 44%, 43% and 13% respectively with ^{238}U also contributing the highest to the absorbed dose rates (Figure 5). The percentage of total absorbed dose of ^{238}U, ^{232}Th and ^{40}K in water when ingested from the three water supply sources in the study area are respectively 50%, 38% and 12% (Figure 6).

The AGED values for the river/creek water in this study is higher than the 10.18mSv/y reported for the water around Mini-Okoro/Oginigba creek in PortHarcourt obtained by Avwiri et. al. (2014). The values of committed annual effective dose due to intake of water by the children and adults from hand-dug well waters, tap/borehole waters and rivers/creek waters are well above the 0.29mSv/y recommended international permissible standard by UNSCEAR (2008) as shown in figure 7. It therefore means that children and adults are very much susceptible to high dose related diseases through intake of water from these water supply sources in the study area. The result of this study also compares with the values obtained by Ononugbo et. al. (2013) on the evaluation of natural radionuclide content in surface and groundwater and excess lifetime cancer risk due to gamma radioactivity for the Ogba, Egema and Ndoni oilfields and host communities in Rivers State where they recorded high mean values of committed annual effective dose above permissible standards for the children, teenagers, babies and adults due to intake of tapwater, well water and river water. The excess lifetime cancer risks (ELCR) values due to intake of water by the children and adults from hand-dug well waters, tap/borehole waters and rivers/creek waters are well above the 0.0001mSv/y ($1x10^{-4}$mSv/y) worldwide permissible limit by USEPA (2012) (Figure 8). This means that the chances of having cancer by the children and adults in the study area as well as the people living around the area who depend on these water sources are very high. Therefore all the sources of drinking water in this area must be treated before consumption so as to avert the likely health implications. This result compares well with the works of Ononugbo et. al (2013) who recorded high mean values of ELCR that are above the USEPA world average permissible limit obtained for children, teenagers, babies and adults due to intake of tap water, well water and river waters at the Ogba, Egbema and Ndoni oilfield host communities in Rivers State. The ELCR values obtained in this study is also higher than the $5x10^{-6}$mSv/y obtained by Avwiri et. al. (2014) in the water around Mini-Okoro/Oginigba creek at PortHarcourt, Rivers State.

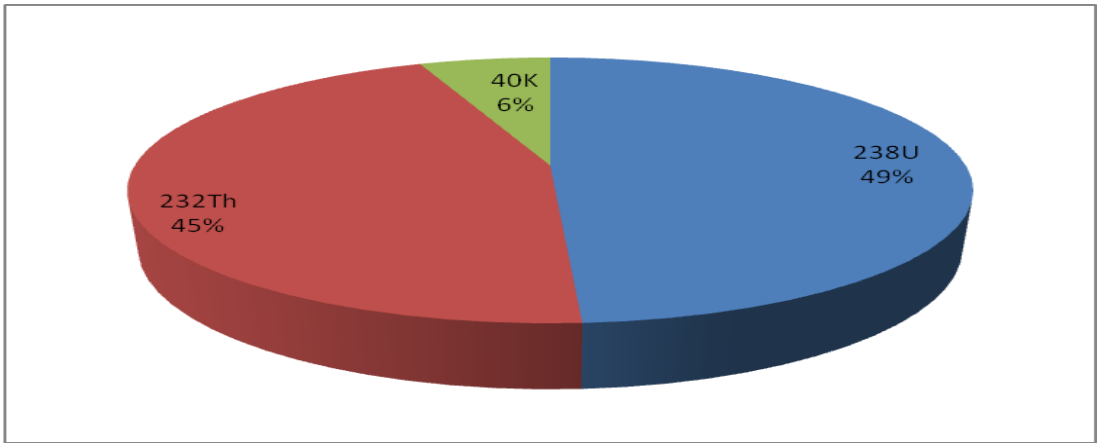

Figure 3: Percentage Absorbed Dose of the three Radionuclides in Hand-dug well waters

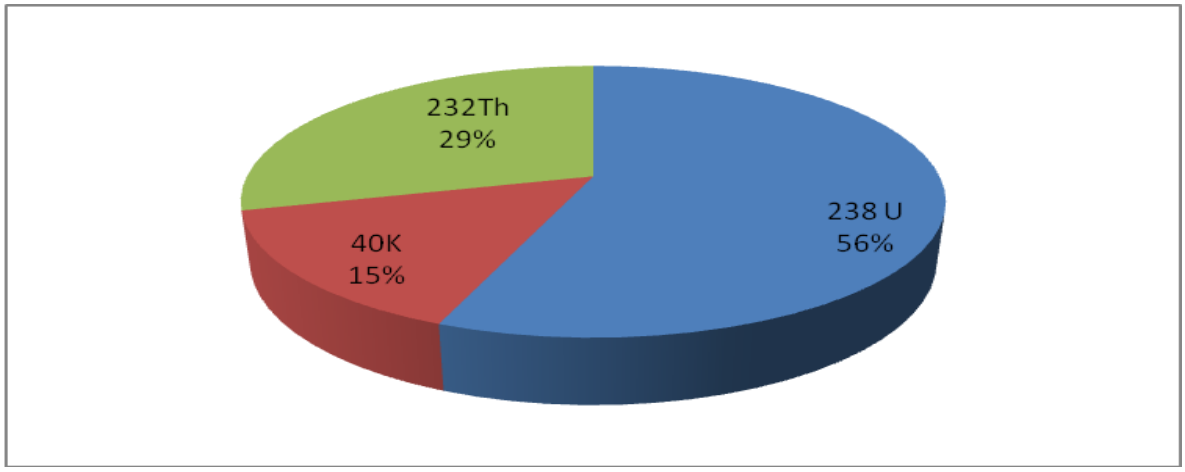

Figure 4: Percentage Absorbed Dose of the three Radionuclides in Tap/borehole waters

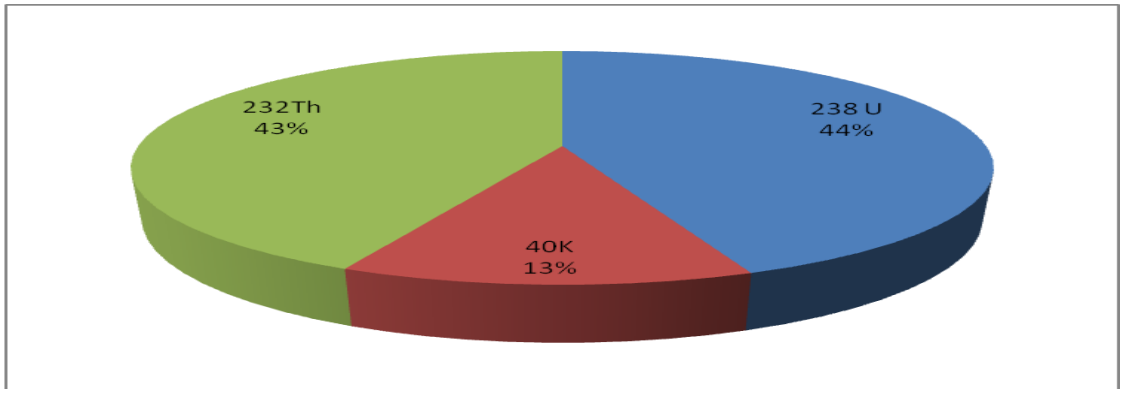

Figure 5: Percentage Absorbed Dose of the three Radionuclides in River/Creek waters

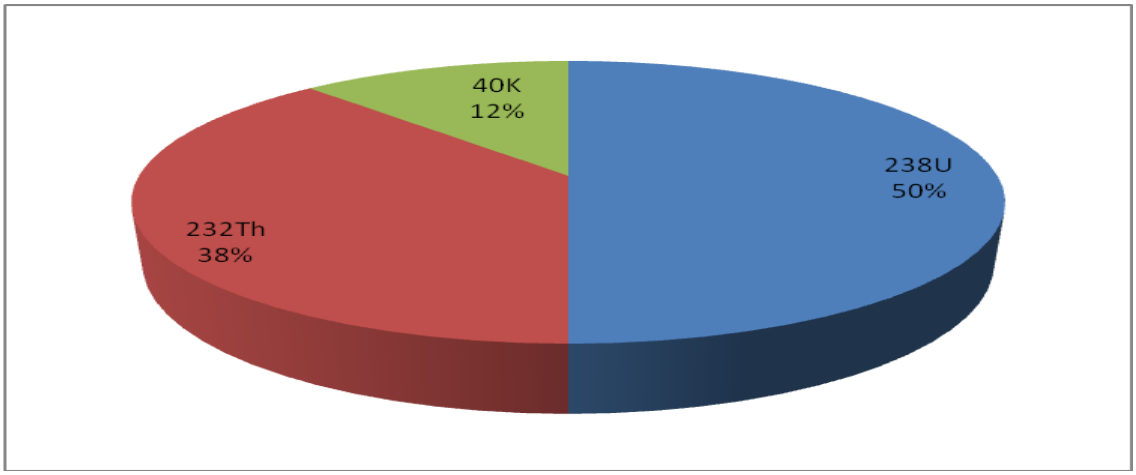

Figure 6: Percentage Absorbed Dose of the ^{238}U, ^{232}Th and ^{40}K in water from the three water supply sources in the study area

Table 4: Activity Concentration of Natural Radionuclides (^{238}U, ^{232}Th and ^{40}K) in the Water Samples and the associated Radiation Hazard Indices

S/No	Sample Name	Specific Activity (Bq/l)			D (ηGy/h)	$Ra_{(eq)}$ (Bq/l)	AGED (mSv/y)	H_{in}	H_{ex}	Iγ	Committed Annual Effective Dose (mSv/y)		ELCR (mSv/y)	
		^{238}U	^{232}Th	^{40}K							Children ≤10years	Adult ≥17 yrs	Children ≤10years	Adult ≥17yrs
1	Bunu-Tai Well Water	12.77±1.12	9.20±1.82	20.13±3.88	12.29	27.48	84.24	0.109	0.074	0.191	1.39	2.06	0.0048	0.0072
2	Bodo Well Water	11.94±1.12	7.88±1.05	13.50±0.62	10.84	24.25	74.07	0.098	0.066	0.167	1.19	1.78	0.0042	0.0062
3	Alode (Eleme) Tap/borehole	2.42±0.45	2.24±0.90	11.73±3.77	2.96	6.53	20.52	0.024	0.018	0.046	0.35	0.51	0.0012	0.0018
4	Bunu-Tai Tap/borehole	3.82±2.20	1.02±1.00	9.71±5.04	2.79	6.03	19.12	0.027	0.016	0.042	0.25	0.34	0.0008	0.0012
5	Bodo Tap/borehole	5.31±0.80	1.20±0.51	11.21±2.10	3.65	7.89	24.94	0.036	0.021	0.055	0.31	0.43	0.0011	0.0015
6	Ochanai Creek (Eleme)	6.81±0.80	9.52±1.16	25.01±3.23	9.94	22.35	68.69	0.079	0.060	0.157	1.29	1.94	0.0045	0.0068
7	Oo-a-naana River (Bunu-	8.48±1.84	4.25±0.65	27.83±1.30	7.65	16.70	52.71	0.068	0.045	0.118	0.79	1.12	0.0028	0.0039

	Tai)													
8	Sugi-Bodo River (Bodo)	8.82±1.20	5.31±0.91	30.03±1.96	8.53	18.73	58.88	0.074	0.051	0.132	0.92	1.32	0.0032	0.0046
WHO (2011); UNSCEAR (2008); OECD (1979), IAEA (2007); USEPA (2012)		**10.0**	**1.0**	**10.0**	**59**	**≤ 370**	**300**	**≤ 1**	**≤ 1**	**≤1**	**0.29**	**0.29**	**0.0001**	**0.00001**

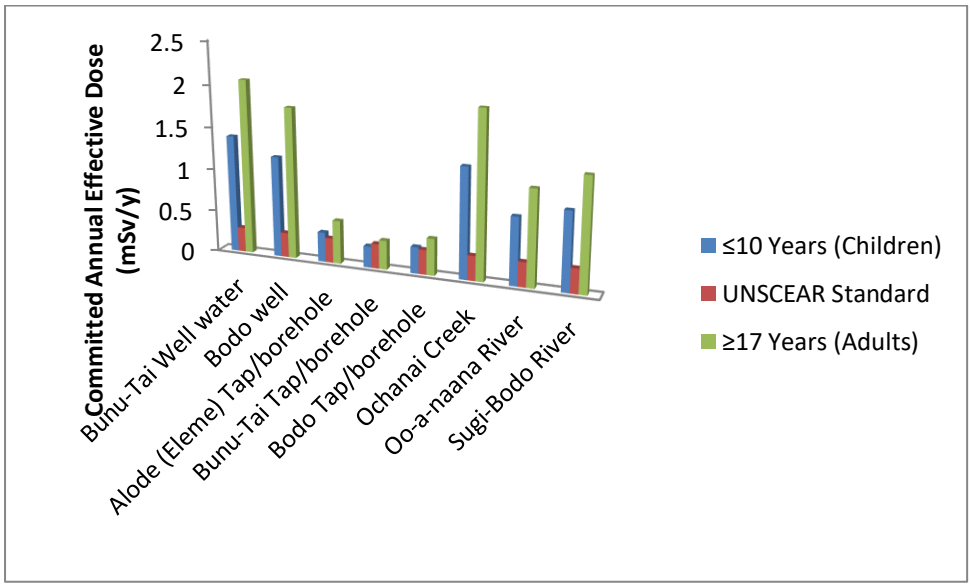

Figure 7: Committed Annual Effective Dose for children and adults due to intake of water from the three sources of water supply

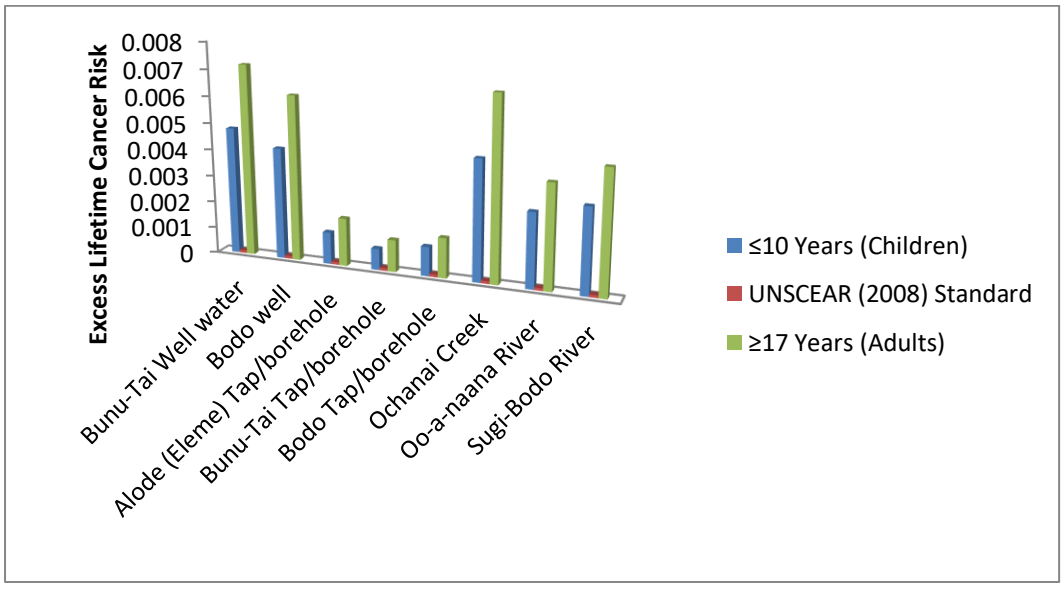

Figure 8: Excess Lifetime Cancer Risks for children and adults due to intake of water from the three sources of water supply

5. Conclusion

The activity concentration of natural radionuclides in the water samples from three water supply sources in an environment with history of crude oil spillages, illegal artisanal oil refining/bunkering activities and gas flaring have been measured with gamma ray spectroscopy. Results showed that the mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in the hand-dug well water samples at Bunu-Tai and Bodo communities as well as the mean activity concentration of ^{232}Th and ^{40}K in the tap/borehole water respectively at Bunu-Tai and Bodo communities were above the recommended standards of 10Bq/l for ^{238}U, 1.0Bq/l for ^{232}Th and 10Bq/l for ^{40}K. The study also revealed that the mean activity values of ^{232}Th and ^{40}K in the river/creek water samples at Eleme, Bunu-Tai and Bodo communities were higher than the international standard. ANOVA statistical testing showed that there was statistically significant difference at $p<0.05$ in the mean variations of the activity concentrations of natural radionuclides in the water supply sources within the three communities considered and the control thus confirming radionuclide enrichment in the water samples collected from the three water supply sources. The study also revealed that children and adults in the three communities are susceptible to high dose related diseases and have high chances of contracting cancer due to the consumption of water from the three water supply sources because the values of committed annual effective dose and excess lifetime cancer risk are respectively higher than the international permissible limits of 0.29mSv/y and $1x10^{-4}$ mSv/y. community based awareness programme by relevant authorities on the issue of environmental contamination is advised. It is recommended that inhabitants of the communities should discontinue the use and consumption of water from these water supply sources and a large reservoir be constructed where clean potable water would be supplied daily from unpolluted environment. The relevant environmental agencies and other stakeholders should also ensure that anthropogenic discharges into surface and groundwater be reduced and all defaulting industries sanctioned for violating industrial effluent standards.

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