Nigerian Journal of Science and Environment 2024 Volume 22 (3) 107 – 117 ISSN: 3043 – 4440
https://doi.org/10.61448/njse223248

Surveying of Radioactivity Concentration of U-238, Th-232 and K- 40 and Evaluation of Radiological Health Parameters on Nigeria Coastal Line

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Abstract

Activities around the Atlantic Ocean coastline region have greatly increased of resent due to industrial and commercial establishments. This study quantified the natural radionuclide contents in the waters along the Atlantic Ocean in West Africa, with special interest in the Nigerian coastline region. The study then evaluated the health risks associated with these natural radionuclides for each population living along the coastline. Several water samples were collected offshore and onshore from five different locations from south-west to south-south in clean plastic bottles and kept in the laboratory for 30 days. In preparation for the use of the HPG detector used, which is of serial number 9744, model GC8023 has a relative efficiency of 80% and a resolution of 2.3 keV full width at half maximum (FWHM) at 1.33 MeV. The results show variation in concentration levels in U238, Th232, and K40 across the coastline. The absorbed dose increases from the south-west down to the south-south of the coastline, ranging from 14.73 and 28.78 $nGyhr^{-1}$ in the south-west, while in the south-south it is 18.75 and 26.18 $nGyhr^{-1}$. The indoor effective dose is in the range 0.072 to 0.141 mSv/y and the outdoor effective dose ranges from 0.018 - 0.035 mSv/y. The increase in absorbed dose is due to K-40 from the agricultural activities in the region and also the crude oil exploration use of radionuclides in oil fields. The excess lifetime cancer risk within the region is slightly comparable to the tolerable values set by regulatory bodies.

Keywords: oceans; absorbed dose; carcinogenic; off shore; radionuclides; regions; coastline.

Introduction

Pollution in the Ocean is a very significant environmental issue that can be a great health problem. Human activities and naturally occurring radioactive materials are the major sources of radioactive contamination in marine and oceans all over the world. The International Atomic Energy Agency has compiled the inventory of radioactivity released into the oceans IAEA. (1999). Thus, primarily, most of the ocean contamination is due to the release of anthropogenic radionuclides into the ocean beds as a result of dumping radioactive waste. Previous works show that such radioactive material does travel with ocean currents, is deposited in marine sediment, and does climb the marine food web RIFE _13, (2008) and discharges of the same from nuclear facilities around the world. Specifically, accidental discharge of nuclear materials and nuclear waste products due to natural disasters as experienced in Chernobyl and Fukushima accidents. Waste discharges such as those from phosphate, oil, and gas industries enhance radioactivity levels in the coastline areas Hurtado-Bermúdez *et al*. (2021), unethical deposits of nuclear materials or waste into the deep ocean, and many more unpredictable sources of radioisotopes in the ocean that contribute to the radioisotope contamination of the ocean. So also oceans are polluted through rivers, runoff, atmospheric deposition and direct industrial discharges, (Colford *et al*., 2007; Haile *et al*., 1999; Small and Nicholls 2003; Hugo *et al*., 2011). The range of anthropogenic radionuclides released from weapons testing is quite large Buesseler, (2014). Consequences of ocean pollution are far reaching in our ecosystem, with negative outcomes in all segments of the ecosystem. Radiation doses from ingestion of natural

radionuclides in food chains have been estimated from measured concentrations of the radionuclides in body tissues or organs UNSC-EAR, (2000). Ionising radiation can affect living organisms, for example, by damaging the genetic material DNA, which can lead to cancer or other. Radionuclide pollutants in the ocean may eventually concentrate in the tissues of many organisms and be transferred along the food chain; this may cause harmful biological effects on a given population and ecosystem RIFE _13, (2008). All of these activities have contributed to the increase in radioactivity levels in oceans (Livingston *et al*., 2000; Carvalho *et al*., 2014). Even though the levels of radioactive contamination in marine fish and shellfish are generally very low, partly because the contamination is diluted in these large bodies of water, human industrialization, marine pollution is an inevitable global problem yet to be understood the effects of contamination are still a concern worldwide Cabral *et al.,(2019*); Piarulli *et al.,* 2021). For this reason, better assessment and understanding of the radioactivity levels in the oceans is required. There are several causes of ocean pollution, but this study is concerned with radionuclide pollutants. Therefore, this study aimed to measure the concentrations of 238U, 234Th, and 40K in water samples along the Nigerian Atlantic Ocean coastline and estimate the effective doses, lifetime cancer risk, and other hazard parameters associated with the coastline pollution as related to global warming. The obtained data will also form part of the data baseline for a reliable assessment of the ocean radionuclide pollutants for critical ecosystem health issues.

Materials and Methods

Study Area

In all the coastline of the country, five sampling locations were conveniently selected and randomly sampled. These locations were on the coastline of south-west Olokun, which is in Ondo State, and Ebute-Lekki and Akodo, both on the coastline of Lagos State, with their longitude and latitude indicated in Table 1 below, and three locations were sampled on the coastline of south-south: Obabebe in Delta State and Amatu-Ijaw in Bayelsa State of Nigeria.

Table. 1: Sample locations and the coded names

Location	CODED	Longitude	Latitude
Ebute Lekki; Lagos	LKK	4º 05’ 56.2’’	6º 25’ 15.0’’
Akodo ; Lagos	AKO	3º 55’ 51.2’’	6º 26’ 16.3’’
Amatu-Ijaw; Bayelsa	AMI	5º 04’ 52.5’’	5º 23’ 52.3’’
Obabebe; Delta	OBA	5º 25’ 56.1’’	5º 16’ 00.8’’
Olokun; Ondo	OLO	4º 36’ 36.0’’	6º 14’ 30.1’’

These locations have been coded as indicated in Table 1 for easy reference. The sampling locations are as illustrated in Figure 1 below, showing coastline areas.

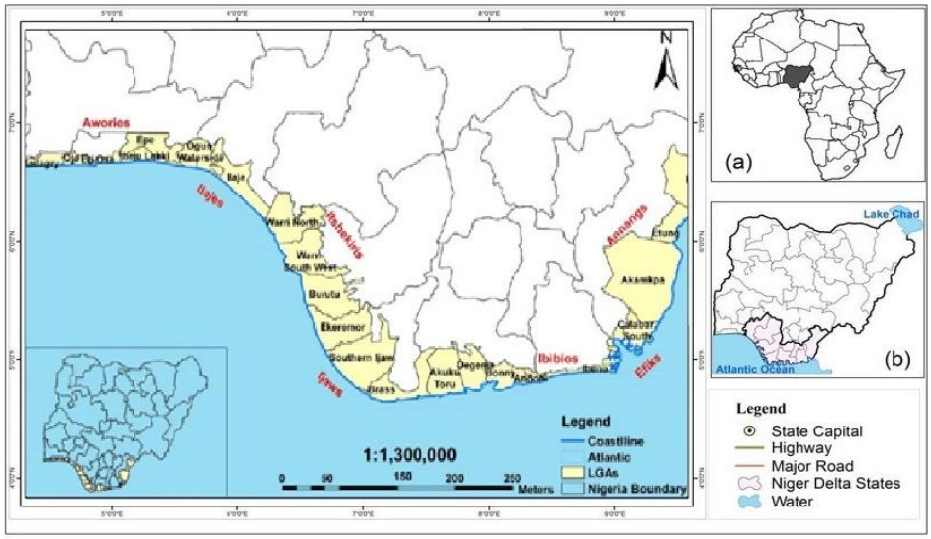

Figure 1: Showing the coastline of the sampling locations Fabiyi, (2019)

The span of the Atlantic coastline in Nigeria is as shown in Figure 1. The coastline extends from Lagos to Ogun to Ondo in the south-west of the country, then to all parts of the south-south of the country, and this is one of the motivations for the sampling across the coastline in Nigeria. There are many human activities, such as fishing, transportation, sand-filling for land reclamation from the seas, etc., ongoing at the various locations along the coastlines.

Sample collection and preparation techniques:

From a distance of about 10 meters off shoreline, two water samples (10 litres) each from five different locations were randomly collected (indicated as points 1 in each case), and the same was repeated onshore (indicated as point 2 in each case) along the Atlantic coastline of the Nigerian ecosystem. Afterwards, the samples were collected in clean plastic bottles of capacity 750 cl, acidified, marked, and sealed. Then, the samples were sent to the laboratory, where they were kept for 25–30 days to allow 226Ra and its short-lived progenies to attain secular equilibrium (Chen *et al*., 2005; Jibiri *et al*., 2007) before the analysis.

Sample Analysis

Samples were analysed with a gamma spectrometry device. The HPG detector used is of serial number 9744; model GC8023 has a relative efficiency of 80% and a resolution of 2.3 keV full width at half maximum (FWHM) at 1.33 MeV. The detector is constantly cooled with liquid nitrogen at -196 °C to reduce leakage current to acceptable levels. The detector is coupled to a Canberra Series 10 Plus multi-channel analyser (MCA). It is housed in a 5 cm thick lead shield that minimises the effect of natural background radiation. The detector is interfaced with the MCA, which consists of an analogue to digital converter (ADC), an internal amplifier, and an in-built high-voltage power supply (HVPS). The efficiency of the detector is the proportionality relationship that relates the activity of the source being counted and the number of counts observed. The efficiency of the detector is calibrated regularly using a standard reference source sample supplied by IAEA (No. MGS6M315) and an efficiency curve generated using GENIE 2000 software. Efficiency is such that the centroids of photo peaks energies are matched with the corresponding channel numbers continuously. The absolute photo peak efficiency of the detector for 238U, 234Th, and 40K gamma-ray spectrum lines was derived from the relationship between the gamma photo peak efficiency of the detector and the photo peak energy E. The

sampled data were also analysed statistically to empirically authenticate our findings.

Analysis and Instrumentation

For water samples, gamma spectra were accumulated for a counting time of 28800s for each sample, and the activity concentrations of 238U, 234Th, and 40K in water were obtained from the count rates from photo-peaks of interest. For 238U, the photo-peaks considered were photo-peaks of 214Pb and 214Bi of energies 295.21 keV and 609.31 keV, respectively. For 232Th, the photo-peaks considered were the photo-peaks of 212Pb, 228Ac, and 208Tl of energies 238.63 keV, 911.21 keV, and 2614.55 keV. The activity concentration of 40K was determined from its photo-peak of energy 1460.8 keV. Activity concentration in water was determined using Equation (1),

$$A_{(Bq/l)} = \frac{C}{T \times v \times \epsilon \times Y} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

where A is the activity concentration of the radionuclide, C is the net count for each radionuclide, which is the gross count minus the background, is the counting lifetime in seconds, v is the volume of the water in litres, ε is the detector energy dependent efficiency for each radionuclide, and Y is the gamma-ray yield per disintegration of the nuclide (emission probability).

Health Risk Assessment Indices

Evaluating the health risk from the samples, the radiological parameters used are the annual effective dose, radium equivalent, gamma index, the hazard index, and the excess lifetime cancer risk.

Radium Equivalent Activities

Radium equivalent is computed from the Equation 2 shown below Dia *et al*. (2008)

$$Ra_{eq}\left(\frac{Bq}{l}\right) = A_U + 1.43A_{Th} + 0.077A_K$$

……. (2)

where A_U, A_{Th}, A_K represent activity concentration of a series of Uranium and series of Thorium and Potassium, respectively Absorbed Dose Rate Total rate of the absorbed dose in the air is calculated in terms of the concentrations of Uranium, Thorium, and Potassium through the Equation (3) shown below according to Shawkat (2000).

$$D_R\left(\frac{nGy}{h}\right) = 0.462A_U + 0.604A_{Th} + 0.0417A_K$$

…….. (3)

Gamma Index

Gamma activity index for the water sample has be computed according to Al-Taher and Makhluf (2010) in Equation (4) shown below

$$I_\gamma = \frac{A_U}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

Annual Effective Dose (E_{in} *or* E_{out})

In totally evaluating the annual effective dose, the conversion factor of absorbed dose to effective dose and the internal occupation factor must be taking into consideration. 0.7 Sv is the conversion factor from the absorbed dose in the air to the annual effective dose. 0.80 and 0.20 are the internal and external time factors, respectively; 8760 is the number of hours a year; and 0.48 mSv is the global average annual effective dose Al-Taher and Makhluf (2010) as seen in Equations (5) and (6):

Nigerian Journal of Science and Environment 2024 Volume 22 (3) 107 – 117 ISSN: 3043 – 4440
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$$ED_{in}\left(\frac{mSv}{y}\right) = D_R\left(\frac{nGy}{h}\right) \times 10^{-6} \times 8760\left(\frac{h}{y}\right) \times 0.80 \times 0.7\left(\frac{Sv}{Gy}\right) \quad \text{.....(5)}$$

$$ED_{out}\left(\frac{mSv}{y}\right) = D_R\left(\frac{nGy}{h}\right) \times 10^{-6} \times 8760\left(\frac{h}{y}\right) \times 0.20 \times 0.7\left(\frac{Sv}{Gy}\right) \quad \text{.....(6)}$$

Hazard index internal and external both the internal and external hazards have been evaluated in this work. The internal hazard index (H_{in}) is the internal exposure caused by inhalation of radon gas and its relevant progenies. While the external hazard assessment required the external hazard index (H_{ex}). The needed computation has been carried out using the following Equations (7) and (8):

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad \text{.....(7)}$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad \text{.....(8)}$$

Excess lifetime cancer risk (ELCR).

This indicates the risk of death in a population brought on by cancer due to exposure to background radiation in excess resulting from a lifetime exposure to carcinogens. That is the expected number of cases of cancer in a population for an average life of about 70 years.

$$ELCR = ED_{out}\left(\frac{Sv}{y}\right) \times LD \times CRF\left(\frac{1}{Sv}\right) \quad \text{..... (9)}$$

where ED_{out} is the annual effective dose outside and *LD* is the lifetime duration of 70 years, and *CRF* is the risk factor, which is 0.05 (Sv^{-1}) Taskin H. *et al*., (2009).

Results and Discussion

Water sampling from the different locations along the coastline (offshore and onshore sampling) from the southwest to the south-south of Nigeria was collected for analysis. The 238U, 232Th, and 40K concentrations were analysed from the samples collected and then used to evaluate the radiological risk due to exposure to the radiation from 238U, 232Th, and 40K.

Table. 2: The mean concentrations (Bq/L) of radionuclides in each sampling location

Sampling locations	**Mean radionuclides concentration (Bq\L)**		
	238-U	232-Th	40-K
LKK1	3.87±0.34	0.38±0.01	641.67±9.21
LKK2	4.72±0.71	0.59±0.11	510.03±6.49
AKO1	4.53±1.05	3.46±0.18	549.35±3.01
AKO2	4.89±0.97	0.42±0.08	491.18±7.03
OLO1	4.26±0.84	0.63±0.08	461.81±4.09
OLO2	5.77±0.72	0.65±0.03	280.04±9.66
OBA1	5.67±1.05	4.93±0.22	370.01±6.33
OBA2	4.98±1.11	3.18±0.95	348.25±8.21
AMI1	4.99±0.14	4.85±0.68	502.33±7.05
AMI2	5.16±0.54	2.64±0.83	388.33±3.61

From Table 2 above, the mean concentrations of the 238U and 232Th from the southwestern coastline were generally higher than those of the south-south coastline, and for the 40K, the reverse is the case. The mean concentrations of 40K have been found to be higher in the south-west coastline than the south-south coastline. These disparities in the mean concentrations of these radionuclides may be due to many reasons, which could include the ocean tides along the coastline, or they may be caused by the fact that in the south-south coastline there are many oil exploration activities in that region as compared to that of the south-west coastline.

The radionuclides used on the oil fields during oil prospect are not properly disposed of have been washed away into the ocean, thereby causing more of the 238U and 232Th concentrations in that region. Nevertheless, the observed mean concentration of 40K is higher in the south-west coastline than in the south-south coastline due to agricultural activities in the south-west, in which the use of fertilisers in the soil might have increased the amount of 40K that may have been washed into the ocean. The obtained results have been thoroughly analysed in this work. The data from all the sampling locations were analysed using the descriptive statistics shown in Table 3. For the period of sampling, 238U exhibits a mean, maximum, and minimum of 4.89, 5.33, and 4.30, respectively. The standard deviation is 0.395 at a 5% level of significance. From the analysis, 232Th concentrations show the mean, maximum, and minimum of 1.89, 4.06, and 0.49, respectively. The standard deviation is 1.84 at a 5% level of significance. The 40-Potassium also exhibits the mean, maximum, and minimum of 445.54, 575.85, and 359.13, respectively. The standard deviation is 88.0015 at a 5% level of significance. Thus, the concentrations of 238-U and 232-Th have been compared in all the sampling locations, as shown in Figure 2 below.

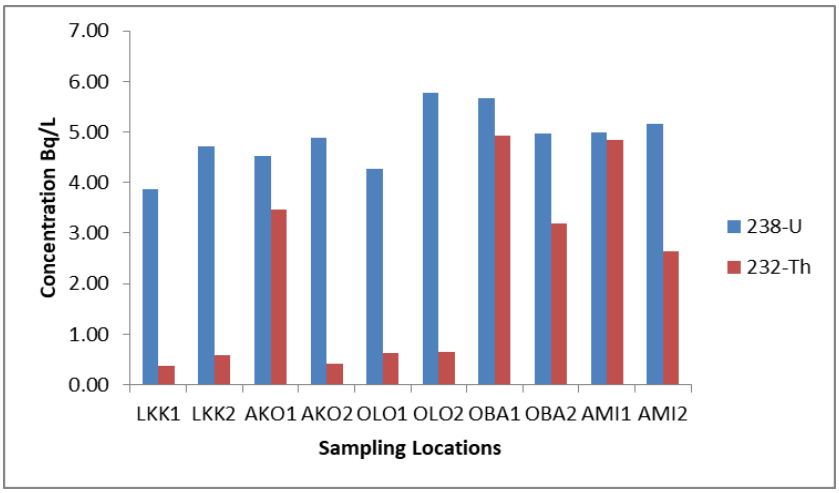

Figure 2: 238U and 232Th concentrations per location

The concentration of 238U is higher in all the sampling locations as indicated in Figure 2 above; the coastline along the south-west has slightly lower values when compared to the other coastline along the southern part of Nigeria. Whereas the 232-Th has a higher value of concentration in the south-south coastline as compared to the south-west coastline; this may be the result of many oil exploration activities ongoing in that part since radionuclide isotopes are used in oil wells for gauging purposes. The Olokun in Ondo on longitude 4^{o} 36’ 36.0’’ and latitude 6^{o} 14’ 30.1’’ in the south-west coastline has a relatively lower value of 232-Th concentration. Akodo longitude 3^{o} 55’ 51.2’’ and latitude 6^{o} 26’ 16.3’’ have the lowest value

of 232-Th concentration. The highest concentration values of 232-Th have been recorded from Amatu-Ijaw on longitude 5° 04' 52.5'' and latitude 5° 23' 52.3'' and Obabebe on longitude 5° 25' 56.1'' and latitude 5° 16' 00.8'' both from the south-south coastline. The distribution of 40-K concentrations is quite different from that of 238-U and 232-Th, and its concentration values are far higher than those of 238-U and 232-Th from all the sampling locations, as indicated in Figure 3 below.

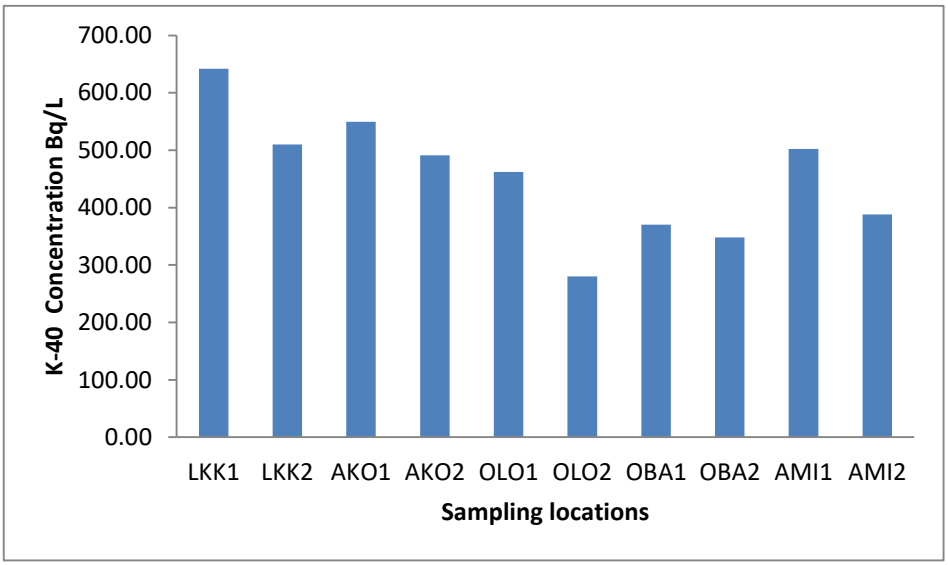

Figure 3: 40K concentrations per location

The highest concentration of 40K has been recorded from the south-west coastline as compared to the south-south coastline. The reason may be due to the agricultural activities in which the use of various types of fertilisers with 40K as part of the chemical components is used as a means of improving crop yielding; thus, these fertilisers are then washed down to the Atlantic Ocean, which then increased the level of 40K in that part of the coastline. The evaluated absorbed dose rate, as shown in Table 3 below, shows that the results from the south-west sampling locations have even distributions of dose rate across the coastline, in which these values are slightly greater than those of the south-south coastline. These values of the absorbed dose obtained from the southwest coaster line are within the range of 14.73 and 28.78 $nGyhr^{-1}$; and the south-south absorbed dose is in the range of 18.75 and 26.18 $nGyhr^{-1}$.

Table. 3: Showing the highest and lowest values of 238U, 232Th, and 40K.

Sampling Locations	Absorbed Dose (nGy/h)	Annual Effective Dose Indoor (mSv/y)	Annual Effective Dose Outdoor (mSv/y)	Radium-Equivalent (Bq/l)	Gamma Index (Bq/l)	Hazard Index indoor (Bq/l)	Hazard Index outdoor (Bq/l)	ELCR
LKK 1	28.779	0.141	0.035	53.830	0.229	0.156	0.145	1.24×10^{-6}
LKK 2	23.805	0.117	0.029	44.836	0.189	0.134	0.121	1.02×10^{-6}
AKO 1	27.090	0.133	0.033	51.777	0.216	0.152	0.140	1.16×10^{-6}
AKO 2	22.999	0.113	0.028	43.320	0.182	0.130	0.117	9.87×10^{-7}

OLO 1	21.604	0.106	0.026	40.715	0.171	0.121	0.110	$9.27x\ 10^{-7}$
OLO 2	14.731	0.072	0.018	28.251	0.116	0.092	0.076	$6.32\ x\ 10^{-7}$
OBA 1	21.027	0.103	0.026	41.211	0.167	0.127	0.111	$9.03\ x\ 10^{-7}$
OBA 2	18.747	0.092	0.023	36.350	0.149	0.112	0.098	$8.05\ x\ 10^{-7}$
AMI 1	26.184	0.128	0.032	50.609	0.208	0.150	0.137	$1.12x\ 10^{-6}$
AMI 2	20.176	0.099	0.025	38.845	0.160	0.119	0.105	$8.66\ x\ 10^{-6}$

The indoor effective dose from the southwest coastline ranges from 0.072 to 0.141mSv/y, while the outdoor effective dose recorded is between 0.018 and 0.035 mSv/y. For the south-south coaster region, both the indoor and outdoor annual effective doses are in the range 0.092 – 0.128 and 0.023 – 0.032 mSv/y, respectively, as indicated in Table 3 above. However, the health risk indicators are also evaluated along the coastline regions in Table 3 above. The radium equivalent is one of the parameters used for the assessment of radiological hazards of radioactivity in environmental materials; thus, it is the parameter that enables the expression of gamma output from the radionuclides of interest. The radium equivalent has been evaluated in this study. In the south-west, the mean radiation equivalent from the Lekki sampling location is the highest; this is then followed by the Akodo sampling location and then the Amatu-Ijaw sampling location, while the lowest mean of the radiation equivalent is obtained from both the Olokun on longitude 4º 36’ 36.0’’ and latitude 6º 14’ 30.1’’, and Ababebe on longitude 5º 25’ 56.1’’ and latitude 5º 16’ 00.8’’. These two locations are midway between the south-west and south-south coastlines. The gamma index is usually correlated with the annual dose due to the excess external gamma radiation caused by environmental materials; hence, this study evaluated the gamma index as shown in Table 3 from each sampling location and shows that the mean value of the gamma index decreases as it moves from the south-west coastline to the south-south coastline.

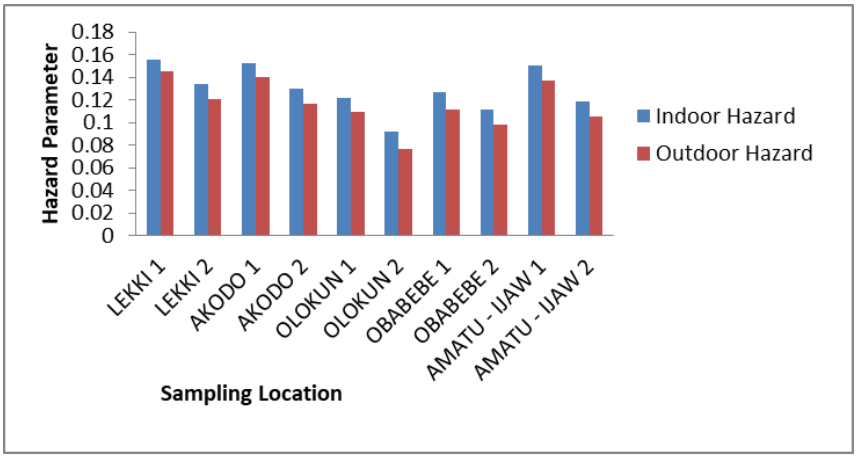

Figure 4: Internal and External Hazard Parameters for Each Location Sampled

The variations in hazard index parameters for both the indoor and outdoor hazards are recorded in this study as in Figure 4 above. In each of the sampled locations from the coastline, both the indoor and outdoor hazards are less than unity from all the locations; however, the trend is that from the southwest, the indoor hazard from Lekki is about 1.56 and the outdoor is about 1.45. These values decrease as one moves towards the south-south coastline regions, in which the indoor hazard has been recorded as 0.12 and the outdoor has 0.11. Excess lifetime cancer risk is the probability that a person may develop cancer in a lifetime

because of the exposure to a contaminated environment. As this also speaks to the health parameter issue in the coastal line, this study evaluated the excessive lifetime cancer risk as shown in Figure 5 below.

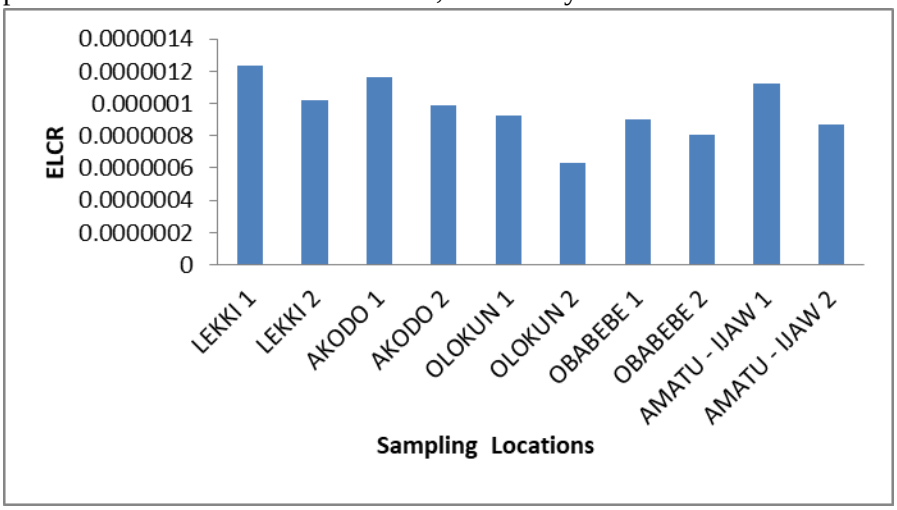

Figure 5: Excess lifetime cancer risk per location sampled

The excess lifetime cancer risk obtained from the coaster line in the southwest is in the range $6.32 \times 10^{-7} – 1.24 \times 10^{-6}$, and the range from the south-south coaster line is in the range $8.05 \times 10^{-7} – 8.66 \times 10^{-6}$.

Conclusion

The offshore and onshore sampling of the Atlantic Ocean's waters has been analysed. There have been variations in the concentration levels of U-238, Th-232 and K-40 across the sampling locations from these two main reasons have been established in this study. Firstly, the higher absorbed dose in the south-west of the coastline has been attributed to the agricultural activities in the area and industrial waste, which have led to great increase in 40-K in the samples. U-238 values were higher than that of the Th-232 in general and this may be attributed to wash away natural radionuclide from the beds of the sea. Secondly, the number of crude oil explorations going on in the south-south coastline due to the use of radionuclides in oil fields during exploration processes. Both the indoor and outdoor hazard quotients vary from locations to locations and may be as different in the concentration levels of the radionuclides along the coastal line. Notwithstanding, the excessive lifetime cancer risk have been compared within the sampling locations. The lifetime cancer risk may be increased in the population of the residents around the banks of the oceans in these areas in a long term.

Acknowledgement

We acknowledged Miss Uche Uba and Mr Williams for assisting in partly the collection of part of the samples used in this study.

Statements and Declarations

Consent for Publication

We, the above-named authors, give our consent for the publication of this document, including all the diagrams, tables, and details within the text to be published in the journal.

Data Availability and Materials

The data that supports the findings of this study are available within the article.

Conflict of Interest

There is no known conflict of interest in this research work. This work was purely the academic and personal efforts of the authors to contribute knowledge to the scientific community and the society at large.

Funding

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. This research work is not funded by any organization.

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