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ACTIVITY CONCENTRATION OF NATURAL RADIONUCLIDES AND ASSESSMENT OF THE ASSOCIATED RADIOLOGICAL HAZARDS IN THE MARINE CROAKER (*PSEUDOTOLITUS TYPUS*) FISH FROM TWO COASTAL AREAS OF NIGERIA

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ABSTRACT
The risk assessment associated with radionuclide contamination of Marine Croaker fish widely consumed in two coastal areas of Nigeria was carried out. The activity concentrations of the three radionuclides Uranium-238 (^{238}U), Thorium-232 (^{232}Th), and Potassium-40 (^{40}K) present in the fish samples were determined using gamma-ray spectrometry. The radiological hazard of consuming this fish was assessed by calculating the radium equivalent activity (Ra_{eq}), internal hazard index (H_{in}) of the radionuclides as well as the ingestion effective dose values due to ingestion of radionuclides from fish per year. Comparison of the values obtained from the radiation hazard assessments with the maximum permissible levels of 370 Bq kg^{-1} for Ra_{eq} and 1 for H_{in} recommended by UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) (UNSCEAR, 2000) shows that consumption of the contaminated Croaker fish from these two coastal areas studied poses great radiological hazards to the public.

Keywords: Radioactivity concentration; Croaker fish; radiological hazards; gamma spectrometry

INTRODUCTION
It has been established that fish consumption has been very beneficial to man as a major source of protein in the diet and as the main source of livelihood for many fishermen, especially those living in the coastal areas like Lagos and Port-Harcourt in the Niger Delta region of Nigeria. In contrast, fish consumption could also contribute to human health hazards if exposed to some radioactive and chemical contaminants. The proximity of Lagos and Port-Harcourt in Nigeria to water bodies has influenced the dietary habits of the inhabitants of these two cities making them to have some of the highest marine fish consumption in the Country. This also translates to increased chances of higher intake of radionuclides from contaminated fish which could have undesired detrimental effect on human health. Food and Agriculture Organization of the United Nations (FAO, 2010) reported that this health risks could be more pronounced in women of reproductive age, pregnant or nursing mothers, breastfed infants and young children. While there are a number of potential contaminants of concern in fish, radionuclides and heavy metals pose the greatest threats, hence radionuclides are the focus of this study. Radioactivity is all about unstable isotopes/nuclides changing to stable isotopes/nuclides. The disintegration of these unstable nucleus into another nucleus in order to become more stable is always accompanied by the emission of an α or a β particle, and sometimes electromagnetic radiations (γ or x-ray photons). If the transition is spontaneous, the process is called natural radioactivity but If the transition is human-induced by the bombardment of particles or radiation, then it is called artificial radioactivity. The emitted particles from these radionuclides are called ionizing radiations and ionizing radiations can destroy the cells in tissues and upset the natural chemistry of the body when they are ingested or inhaled. The severity of the damages depends on the absorbing tissue or organ, the nature of the radiation and the dose (Orosun *et al*., 2018). The time scale of decay processes varies widely, ranging from a small fraction of a second to billions of years, so the radiological hazard could persist for a long time, and the body organ or tissue is continuously irradiated if the radionuclides have medium to long half-lives. Radioactive nuclides in the environment come from various sources; some are produced naturally from weathered rock, while others are artificially produced as a result of human activities such as mining, fertilizers application, agrochemicals, research, and medical facilities. ^{226}Ra, ^{232}Th and ^{40}K are three naturally abundant radionuclides in the environment. Some of these radionuclides often find their way into the water bodies, indirectly into the food chain and ultimately into the human body via sea food such as fish with potential adverse effects on human health. Similarly, careless disposal of spent radioactive sources, nuclear reactor spent fuel elements and the indiscriminate discharge of untreated industrial effluents into water bodies pose significant risks to human health. The effluents sometimes contain radioactive nuclides and heavy metal contaminants which can reach dangerous levels of accumulation in the environment especially receiving water bodies such as Lagos Lagoon. The heavy metals are largely non-biodegradable and their persistence in the environment degrades water quality and soil constituting potential hazards to human health as they accumulate in animal and fish tissue. Feeding on fish from such polluted water body poses long-term risks to public health. In extreme cases, leading to failure of vital organs such as liver, kidneys or causing cancer of the lungs (Adeleye, 2013). Metals like Cd, Hg, As, Pb, Cr, etc. which have been found in human diets have been reported to be carcinogenic and/or mutagenic in a broad spectrum of animal studies (Khandaker *et al*., 2015). Lagos being the focal point of

industrial activities in Nigeria has had more than its own fair share of pollutions from industrial wastes most of these wastes end up in the lagoon as the receiving water body.

Continuous striking of body tissues and organs by ionizing radiations produced from the decay of ingested radionuclides are known to have triggered or induced cancer in living tissues such as kidney or brain (Fasae and Isinkaye, 2018; Orosun, *et al*., 2018). It is therefore imperative to evaluate the extent of contamination of this fish and assess potential hazards it poses to the public consuming it in these two study areas.

These radionuclides in ocean show a complex behavior, some like caesium and uranium form compounds which are quite soluble in sea water while others such as thorium are almost totally insoluble in water. So, they can be transferred in the marine environment when dissolved in the seawater, or attached to planktons suspended in the seawater and attached to sediment on the seabed leading to the contamination of the marine organisms, including fish (UNSCEAR, 2008; Khandaker *et al*., 2015).

Export of crude oil produced mainly in the Niger Delta region of Nigeria including Port-Harcourt city contributes more than 90% of the nation's foreign exchange earnings (Uwakonye *et al*., 2006), but this production processes involves the use of radionuclides for natural gas and oil prospecting. Such anthropogenic activities including testing of nuclear weapons, electric power production, particle accelerators, production of natural gas and oil, mining and processing of ores, industrial discharge of untreated effluents into water bodies, etc. have been reported to enhance the radioactivity in the marine environment. (Cherry and Heyraudm., 1981; Khandaker *et al*., 2015; Alayande *et al*., 2016). Many radionuclides have short half-lives and therefore decay rapidly. However, limited artificial radionuclides have been reported in the marine environment (UNEP/IAEA 1992; IAEA, 1999). According to International Atomic Energy Agency, total radioactivity in marine environment is predominantly due to potassium (UNEP/IAEA, 1992; IAEA 1999). Also, hydrosphere (marine environment) has a unique property in which daughter nuclide can be separated from parent due to change in chemical properties. This leads to distortion in equilibrium of radiological isotopes between parent and daughter nuclides. High radiation doses in some marine organisms are based on this phenomenon.

In Nigeria, fish community can be classified into following units: estuarine and creek sciaenid, off shore suprathermocline sciaenid, shallow suprathermocline sparid and deep subthermocline sparid sub-communities (FAO, 1986). Creeks, estuaries and brackish water are inhabited by marine and fresh water fish species. A common catch in this sub-community is *Pseudotolithus typus (P. typus)*, commonly called long neck croaker. At maturity, it can attain length of 100 cm but 50 cm is common in catches. It inhabits sandy and mud bottoms to a depth of 150 m (FAO, 1986). It is a very important commercial sciaenid specie in Nigeria, fished by industrial fleets and local artisans (FAO, 1986). To the best of our knowledge, there has been no radiological study on the specie in Nigeria. In view of its importance, measurement of activity concentration of natural radionuclides in this fish is logical and crucial to ascertaining radionuclides level, bioaccumulation of toxic elements, and current level of pollution of the habitat based on either natural or anthropogenic activities. In this study, radiology hazard was assessed in *P. typus* from Lagos and Borokiri (Port Harcourt), two coastal regions in Nigeria based on Ingestion Effective Dose (D); Internal Hazard Index (H_{in}) and Radium Equivalent Activity (Ra_{eq}).

MATERIALS AND METHODS

The Study Area

Lagos is a city located in the southwestern geopolitical zone of Nigeria with latitude of 6.465422ºN, and longitude of 3.406448ºE. The city, with its suburbs makes the Lagos State with Ikeja as its capital. Lagos is classified into two main geographical *areas*—the "Island" and the "Mainland". The two major urban islands in Lagos are Lagos Island and Victoria Island, which are separated from the mainland by the main channel draining the lagoon into the Atlantic, which forms Lagos Harbor. The islands are connected to the mainland by bridges. The Lagos metropolis occupies a total 1,171.28 km^2 of which 999.6 km^2 is land and 171.68 km^2 is water (Wikipedia, 2020). With estimated population of 21 million in 2016 by the National Population Commission of Nigeria based on 2006 Census, Lagos is Nigeria's largest city and most populous city in Africa (NPC, 2007). It is bounded by the state of Ogun to the north and east, by the Bight of Benin to the south, and by the Republic of Benin to the west. Many of the residents are into farming and fishing with their products collected in the lagoon ports of Badagry, Epe, and Ikorodu shipped to markets in Lagos city.

The second area of the study, Borokiri is a neighborhood of the city of Port Harcourt situated just south of Old Government Reserved Area (GRA) in Rivers State, Nigeria. It is an approximate area of 255 hectares of arable agricultural land and fishing waters. It lies at latitude 4.749° N and longitude 7.035° E. The neighborhood is bounded by Ahoada Street to the north, Okrika Island to the east, Orubiri oilfield to the south and Ship Builders Road to the west. Port Harcourt is the capital and largest city of Rivers State, Nigeria. It lies along the Bonny River and is located in the Niger Delta. As of 2016, the Port Harcourt urban area has an estimated population of 1,865,000 inhabitants. The area that became Port Harcourt in 1912 was before that part of fishing settlements called Borokiri in Okrika language. (NPC, 2007).

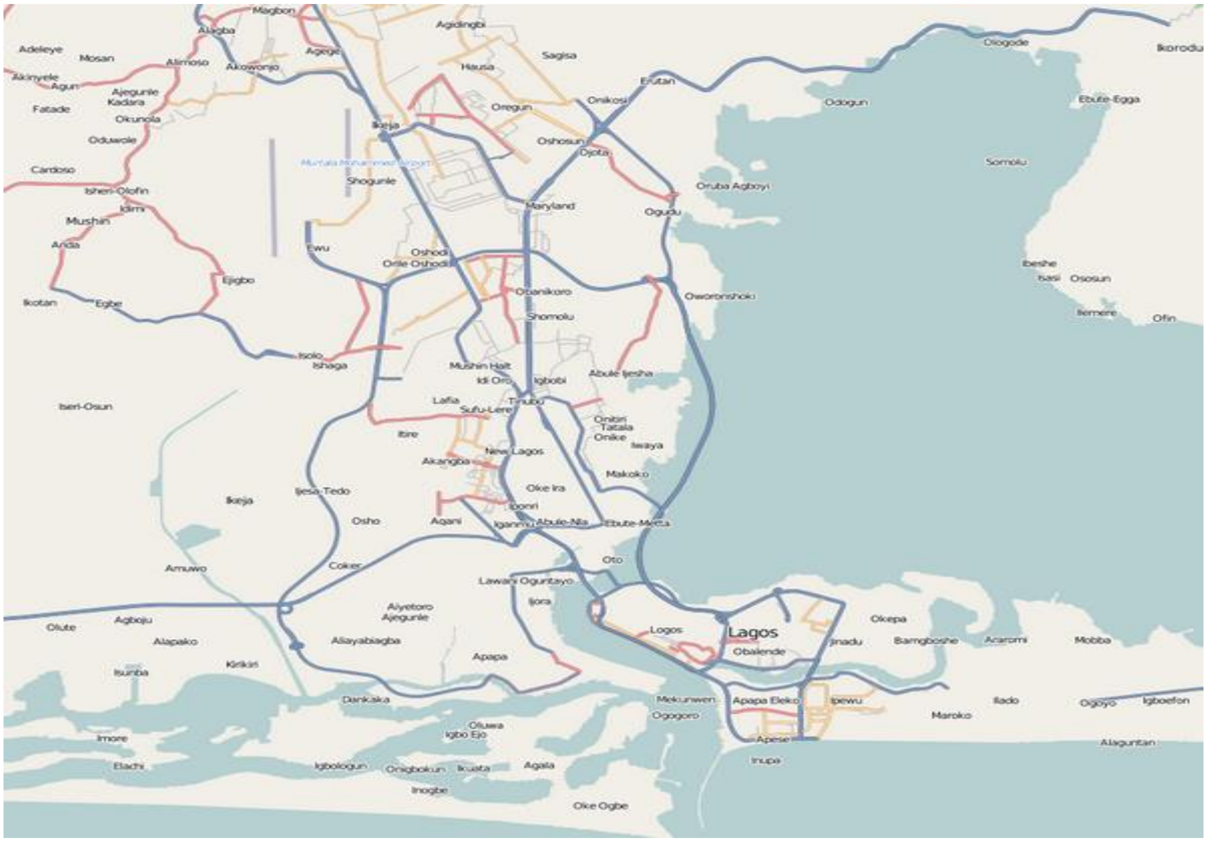

Figure 1: Map of Lagos State (Source: Wikipedia, 2020)

Figure 2: Map of Port Harcourt, Rivers State (Source: Google, 2020)

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Sample Collection and Preparation

Live samples of demersal fish (long-neck Croakers - *Pseudotolitus typus*) were collected from the jetties/landings of industrial fleets and coastal artisanal fishermen from Lagos and Borokiri, Port-Harcourt respectively using random sampling technique. The samples were killed, rinsed with distilled water to remove dust or any other extraneous materials inadvertently collected along with the samples, cut into smaller pieces and then oven-dried at a temperature of 60^{o}C to 80^{o}C for several hours depending on the moisture contents until a constant weight was attained. The dried samples were then pulverized to obtain a fine powder and sieved for homogeneity, then packaged for analysis at the gamma laboratory of the Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile-Ife.

Between 2 and 3 g of each sample weighed using digital balance was added into the polythene bag and heat-sealed with the sample identity on it. The sample was then placed into a cylindrical vial 5 cm high and 1.5 cm diameter, the top stocked with cotton wool, vials cap snapped closed, sealed with cello tape and the ID number of the sample written on the vial. The packaged samples were then re-arranged to composite samples based on region and then sealed for a minimum of 28 days to achieve secular radioactive equilibrium before gamma spectroscopy measurements.

Measurements of radioactivity

The measurement was done using gamma-ray spectrometer fitted with a 7.62 cm x 7.62 cm Thallium-Activated Sodium Iodide detector system (Alayande *et al*., 2016). The detector was coupled to a multi-channel analyzer (MCA) through a photomultiplier. The spectrum was acquired for 18,000 s and the multi-channel analyzer (MCA) with 4096 channels was used to determine the photo-peak energies of interest in the γ-ray spectrum and the photo-peak areas analyzed by Maestro software. Prior to the spectral analysis, routine energy and photo peak detection efficiency calibrations of the spectrometer were carried out using standard gamma-ray calibration sources whose activities are well known, prepared by the Isotope Products Laboratories, Burbank California, USA. (Adeleye *et al*., 2014). Blank counts for an empty vial was also carried out to determine the background radiation in order to enable the spectrometer determine the net count rates from the gross. Updated recommended decay data for nuclides were obtained from literature (Arzu *et al*., 2010, 2011).

Radiological Hazard Study

In order to relate the effect of activity level of radionuclides in the samples from the different regions to bio-system, some important radiological hazard indices such as the absorbed dose rate, annual effective dose and external hazard index were calculated in this study as follows:

Radium Equivalent Activity (Ra_{eq}):

The Ra_{eq} measures the hazard associated with the presence of Uranium-238 (^{238}U), Thorium-232 (^{232}Th), and Potassium-40 (^{40}K) radionuclides in a material. It is based on the assumption that 370 Bq kg^{-1} of ^{226}Ra or 260 Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same gamma (γ) dose rate. This was calculated using equation 1 (Abojassim *et al.,* 2014).

$$Ra_{eq}\,(Bq\ kg^{-1}) = A_u + 1.43A_{Th} + 0.077A_k \qquad (1)$$

Where,

A_u, A_{Th}, and A_k are the Activity Concentrations (Bq kg^{-1}) of Uranium, Thorium, and Potassium respectively in the fish samples.

Internal Hazard Index (H_{in}):

The H_{in} measures the internal exposure to carcinogenic radon in the fish samples and was computed using equation 2 (Nasim *et al*., 2012).

$$H_{in} = (A_u/185) + (A_{Th}/259) + (A_k/4810) \qquad (2)$$

Ingestion Effective Dose (D):

The D value in fish food was calculated using equation 3 (Samavat *et al*., 2006; Jibiri *et al*., 2007).

$$D\ (\mu Sv\ y^{-1}) = D_f \times U \times (C_d \times h) \qquad (3)$$

where,

D_f is the dose coefficient (Sv Bq^{-1}) which are 2.8×10^{-7}, 2.2×10^{-7}, and 6.2×10^{-9} Sv Bq^{-1} for ^{238}U, ^{232}Th, and ^{40}K respectively (IAEA, 1996), U is the annual per capita fish consumption in Nigeria taken as 9.8 kg y^{-1} (USAID, 2010), C_d is the activity concentration of the radionuclide in dried fish sample (Bq kg^{-1}), h is the ratio of dried to fresh fish consumption in Nigeria estimated at 30:70 (Adamu *et al.,* 2013).

RESULTS AND DISCUSSION

Radioactivity Level

The result obtained for the activity concentrations in Bq kg^{-1} for ^{238}U, ^{232}Th, and ^{40}K in the composite samples are presented in Table 1. The activity concentrations for ^{238}U were 74.75±2.55 Bq kg^{-1} and 54.42±2.29 Bq kg^{-1}, ^{232}Th were 10.43±4.5 Bq kg^{-1} and 299.33±22.28 Bq kg^{-1}, and ^{40}K were 2305.8±5.61 Bq kg^{-1} and 1767.19±4.91 Bq kg^{-1} for fish samples from Borokiri (Port-Harcourt) and Lagos State, respectively. The results show that the activity concentrations of ^{40}K in the fish samples from both locations were higher compared to those of ^{232}Th and ^{238}U respectively.

The activity concentration values were excessively higher when compared with values earlier reported in fishes worldwide (Table 2). This could be attributed to differences in the feeding habits of the fishes and geographical location conditions (UNSCEAR, 2000). Radionuclide accumulation rate in fishes have been observed to be influenced by their feeding habits (Khan *et al*., 2007). A number of studies conducted previously established lung cancer have been induced in experimental animals after inhaling radon-222 or ingesting alpha-emitting radionuclides such as uranium-238 or beta–emitting radionuclide such as thorium-234 (ICRP, 1980; UNSCEAR, 2016). There are also high chances of the transfer of some of these radionuclides in the food chain from soil to plants, animals and man.

Table 1: Radioactivity Concentrations of Radionuclides in the Fish samples from Borokiri (Port-Harcourt, Niger Delta region) and Lagos State, Nigeria (Bq kg^{-1})

Location	^{238}U	^{232}Th	^{40}K
Borokiri	74.75±2.55	10.43±4.5	2305.84±5.61
Lagos	54.42±2.29	299.33±22.28	1767.19±4.91

Table 2: Comparison of Activity Concentration Values with some Radionuclides in fish samples from some Countries and UNSCEAR Report (2000)

Location	Activity Concentrations in Bg kg^{-1}			Reference
	^{238}U	^{232}Th	^{40}K	
Bangladesh	0.11-1.94	0.24-2.28	4.93-77.1	Alam *et al*., 1995
Hong Kong, China		0.064-0.19	41.2-111	Yu *et al*., 1997
India			15.6-360.7	Khan *et al*., 2007
Netherlands	0.05-1.1			Aten *et al*., 1961
Oman	0.14-2.66	0.06-4.68	38-570	Goddard *et al*., 2003
UK	0.0025			Hamilton, 1972
USA		0.0012	34-170	Fisenne *et al*., 1987 and Klement, 1965
UNSCEAR	0.03	10		UNSCEAR, 2000
Nigeria	54.42-74.75	10.43-299.33	1767.19-2305.84	This study

Assessment of Radiation Hazards

The radium equivalent activity (Ra_{eq}) and internal hazard index (H_{in}) values were 267.21 Bq kg^{-1}and 0.92 for fish samples from Borokiri (Port-Harcourt) and 618.56 Bq kg^{-1}and 1.82 for fish samples from Lagos States respectively (Table 3). These values when compared to the maximum permissible level of 370 Bq kg^{-1} for Ra_{eq} and 1 for H_{in} recommended by UNSCEAR (UNSCEAR, 2000) shows that the fish from Borokiri (Port-Harcourt) is lower while that from Lagos is higher. The Ra_{eq} and H_{in} values obtained for the fish samples from Lagos is twice the value for fish samples from Borokiri (Port-Harcourt). This should be a great concern about the safety of the populace consuming this fish particularly in Lagos State. Anthropogenic activities such as movement of heavy vessels, ships and channeling of untreated effluent into Lagos coasts might be responsible for high Ra_{eq} and H_{in} in croaker fish found in this region. *P. typus* inhabit photic zone in the marine, natural radionuclide due to anthropogenic activities might be abundant in this zone. Secondly, since over 90% of the nation's foreign exchange earnings is through exportation of crude oil (Uwakonye *et al*., 2006), produced mainly in the Niger Delta region of the country including Borokiri (Port-Harcourt), Rivers State, slight release of substances known to contain natural radionuclides can penetrate into the food chain when fish feeds on Phytoplankton and Zooplankton.

Table 4 shows the results of the Ingestion effective dose (D) in µSv y^{-1} due to the specific activities of ^{238}U, ^{232}Th, and ^{40}K in the fish samples. The summation of D values obtained was 157.8 µSv y^{-1} and 388.4 µSv y^{-1} respectively for fish samples from Borokiri (Port-Harcourt) and Lagos State. It is important to note that the D value of 278 µSv y^{-1} due to ^{232}Th obtained for fish samples from Lagos is far higher than 9.7 µSv y^{-1}value for fish samples from Borokiri (Port-Harcourt). The summation values obtained from both locations were significantly higher by two orders of magnitude than the permissible limits of 1 µSv y^{-1} recommendation by ICRP (1996). In addition, the values are higher than 0.14 µSv y^{-1} for age-weighted Ingestion effective dose (UNSCEAR, 2000) and 0.2 – 1 µSv y^{-1} for ingestion effective dose due to radionuclides of natural origin in food diets (UNSCEAR, 2008). It should be noted that higher consumption rates of this fish may therefore cause greater public health risk since the calculations were based on 9.8 kg y^{-1} annual per capita consumption rate in Nigeria which is lower than 25 kg y^{-1} reference value for adults (UNSCEAR, 2000). Besides, inhabitants of these two cities being in the coastal areas have one of the highest marine fish consumption in the Country.

Table 3: Radium equivalent activity and internal hazard index in the fish samples from Borokiri (Port-Harcourt, Niger Delta region) and Lagos State respectively.

Location	Ra_{eq} (Bq kg^{-1})	H_{in}
Borokiri	267.21	0.92
Lagos	618.56	1.82

Table 4: Ingestion effective dose values due to ingestion of radionuclides from fish per year

Location	Ingestion effective dose (µSv y^{-1})			Sum
	^{238}U	^{232}Th	^{40}K	
Borokiri	87.9	9.7	60.2	157.8
Lagos	64.2	278	46.2	388.4

Conclusion

Consumption of radionuclide contaminated Croaker fish from these two locations may constitute a significant health hazard to the public and are therefore not safe for human consumption. In modern food safety, assessment of radionuclide activity concentrations and risk of exposure is considered important. Hence, the findings from this study show that this fish is not safe for consumption and urgent mitigation is required while proper advice is given to the public on the risks involved before increasing levels of contaminations become a significant health issue.

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