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Research article

Radiological Analysis of ^{40}K, ^{226}Ra and ^{232}Th in Fish, Crustacean and Sediment Samples from Fresh and Marine Water in Oil Exploration Area of Ondo State, Nigeria

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ABSTRACT: Radiological analysis was carried out on fish, crustacean and sediment samples collected from both fresh and marine water of river Igbokoda in the coastal area of Ondo State, Nigeria. The activity concentrations of radionuclides were determined using gamma spectrometry method. The means of the annual effective ingestion dose varied between 23.3 ± 10.2μSv.y^{-1} (*Oreochromisniloticus*and*Gymnarchusniloticus*) and 34.8 ± 1.7 μSv.y^{-1} (*Parachnnaobscura*) for fresh water fish samples and 6.4 ± 0.7 μSv.y-^{1} (*Chrysithctysnigrodigitatus*)and14.2 ± 1.6 μSv.y-^{1} (*Cynoglossussenegalensis*) for marine water fish samples. That of the crustacean samples (*Peanusmonodon*) was 2.4 ± 0.2 μSv.y^{-1}. The equivalent dose to the skin of humans from contact with sediment was 2.0 μSv.y^{-1} and 1.2 μSv.y^{-1} for the fresh and marine water sediments, respectively. The effective dose was calculated as 0.010 μSv.y^{-1} and 0.006 μSv.y^{-1} for the fresh and marine water sediments, respectively. The equivalent doses obtained are less than the ICRP recommended 50mSv per annum for the public.

Key Word: radiological analysis, aquatic samples, dose, oil exploration.

INTRODUCTION

Ionizing radiation are widely used throughout the world, particularly in medicine, industry, agriculture and research. Natural radiation in environment is augmented by artificial sources, emanating from man-made radionuclide such as those used in medical practices, nuclear power generation and also from fallout in a nuclear explosion (Larmash 1983). Most of these are not usually available to the public, but they may find their way into the environment through routine releases, accidents, thefts, loss and incorrect disposal or misuse. Unlike the natural sources of radiation which are widely distributed around the world, man-made sources are usually localized and affect only a small fraction of the population at any one time (Mettler *et al* 1990).

Both naturally and artificially occurring radionuclides are found in the aquatic environment. The naturally-occurring radionuclides are primordial and of cosmogenic origin. Their presence in the aquatic environment is principally due to their solubility in water, run-off from land, exchange and removal of particulate material with the atmosphere (Pentreath 1977). In water medium of the aquatic environment, ^{40}K is the most abundant radionuclide followed by ^{238}U. Because ^{232}Th is essentially insoluble in water, members of its series are not found in significant amount in sediments where the nuclides have been absorbed to particulate matters of the cosmogenic radionuclide.

Plants and animals in the aquatic environment accumulate radionuclide to concentration greater than those of the ambient (Pentreath 1988). This is because of the physical and chemical properties of the surfaces.

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An aquatic organism such as benthic algae accumulates more radionuclides because of the absorptive properties of their coating which protect them from physical damage and desiccations. Active transport and direct exchange of radionuclide with water occur to maintain homeostatic concentration of a number of radionuclides in tissue.

The major industry in the coastal areas of Nigeria is oil production and exploration. The oil (petroleum) industry is the most important sector of the Nigerian economy. The rate at which modern technology is being introduced in the country by this sector is very high. In Nigeria apart from medical exposure, the petroleum industry is the largest importer and user of radioactive sources, covering both upstream and downstream operations (Elegba 1993). These include nuclear well logging, nuclear level density gauges, industrial radiography and the application of radiotracers in oil well management, reservoir studies and learn detection in pipelines. Radiation from these radioactive sources is used in a number of ways, including non-destructive testing (NDT) of welds and the examination of vital metal components for cracks and other flaws. It is also used as gauges to monitor and sometimes to control the density of mud and cement grout, to monitor and control the density of material in vessels in petrochemical plants. Downhole logging tools incorporate a variety of radioactive sources to log various features of a well.

The coastal areas of Ondo State, Nigeria border the land and the Atlantic Ocean. The coastal fresh water and brackish/marine wet lands are ramified by a network of rivers and creeks. The Ondo State coastal area is bounded to the north by an extensive river (fresh water and marine water) flood plain which is a low lying area with heights not more than 3.5 m above sea level (Keay 1959). For over five years now, the Eseodo and Ilaje local government, a major geomorphic feature in the Ondo State coastal zone has been under intensive exploration and mining of petroleum and other mineral resources. This is connected with the large deposit of oil and gas (htt://www.ondostate.info). These exploration activities involve the use of radioactive materials of different forms, strength and half-lives. Despite various measures that might be put in place by the oil industries to ensure safety, there are possibilities of accidental discharges and leakages of the radioactive material into the aquatic environment. Such pollution will affect all animal and plant species in the rivers and streams. Traces of this will settle in sediments as it is being transported with water current. Apart from the controlled artificial radionuclides applied in oil and gas production, naturally occurring radioactive materials (NORMs) has been identified to be associated with oil and gas production. The first report of NORMs being associated with oil and gas production appeared in 1904 (IAEA 1990). Analysis carried out on the material from many different oil and gas fields showed that the solids found in downhole and surface structures of oil and gas production facilities such as inner walls of production tubular, wall heads, valves, pump and separator consists of nuclides of ^{238}U and ^{232}Th series (Jonkers*et al* 1997). NORM in the oil and gas industry has the potential to cause external exposures during production due to accumulation of gamma emitting radionuclide and internal exposure to workers and other personnel particularly during maintenance, transportation of waste and contaminated equipment (Jonkers*et al* 1997, Silk 1995).

Considering the environmental impacts of oil and gas exploration with radioactive sources there might be a high level of radionuclide presence in the sediment and aquatic animals. And it is feared that further exploration might have some effect on the aquatic environment and the population. The aquatic species in these areas may be exposed to ionizing radiation. It is therefore necessary to investigate radioactivity levels in fresh and marine water animals and sediments in the coastal area of Ondo State. Hence this work seeks to investigate, the extent to which the members of the public living near the oil exploration site in Igbokoda, Ondo State, are exposed to radiation through the consumption of aquatic organisms from fresh and marine water in the area.

MATERIALS AND METHODS

Samples collection and preparation

Ten species of fish and crustacean that are widely consumed in Nigeria were collected from both fresh and marine water of river Igbokoda in Ilaje Local Government Area of Ondo State. Figure 1 shows the map of the study area. The fish and crustacean samples were collected in oil exploration area with the help of fishermen. Three samples of fish and crustacean were collected for each species of fish, making a total of thirty fish samples. The samples were identified at the fisheries section of Zoology Department University of Ibadan, and were grouped based on their types. Thereafter, they were oven dried at a temperature of $80^{O}C$ (Dougherty and Ng 1982, Akinloye*et al* 1999). Table 1 presents the species, family, common names and the source of the fish and crustacean samples. The dried samples were pulverized, weighed and packed 100 g by mass in plastic containers. The containers were sealed and kept for more than 28 days in order to

allow for Ra and its short-lived progeny to attain secular equilibrium.

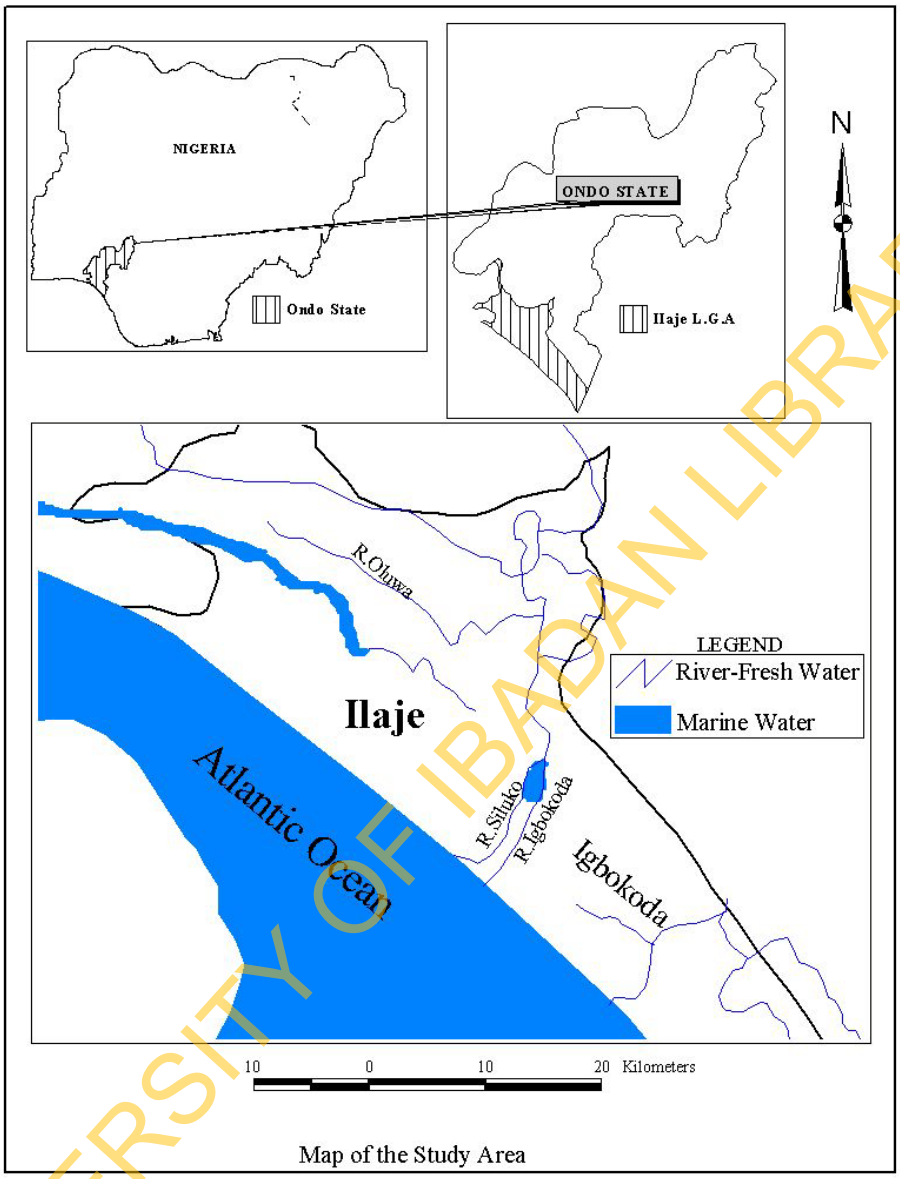

Figure 1:
A map of Nigeria showing the study area

Ten sediment samples each were collected from fresh and marine water, respectively of River Igbokoda in Ilaje Local Government Area of Ondo State. The water in each sediment sample was drained. They were packed, labeled and taken to the laboratory for further processing. The samples were oven dried at a temperature of 105^{O}C before pulverization (Alam*et al* 1997, Godoy *et al* 1998). They were then packed 200 g by mass into labeled cylindrical plastic containers of uniform dimension. The plastic containers were tightly covered, sealed and left for more than 28 days, a period considered sufficient for the attainment of secular equilibrium of Ra and its short- live progeny.

Measurements of activity concentrations

The measuring system employed in this work for the determination of the radionuclides consists of a scintillation detector sealed with photomultiplier tube and connected through a preamplifier base to a Canberra series 10 plus multi-channel analyzer. The detector is a 7.6 cm x 7.6 cm NaI (Tl) (Model No 1102 series). The detector has a resolution of about 8% at the energy 0.662 MeV of ^{137}Cs.

The detection efficiency calibration of the system for the determination of radionuclides in the sediment samples was carried out using reference standard source prepared form Rocketdyne Laboratories, Canoga Park, California, USA, which is traceable to a mixed standard gamma source by Analytic Inc. Atlanta Georgia. For the fish samples, a standard reference dairy sample with reference number IAEA-152 was counted at the same fixed geometry of the spectrometric assembly. The net count A under the photopeak above the background was computed and related to the activity concentration c (Bq kg^{-1}), of the reference source using the relation (Farai and Ademola 2005);

$$E_p = \frac{A}{tcym} \qquad (1)$$

Where E_p is the detection efficiency of the measuring system for each of the radionuclide, t is the counting time, y is the gamma yield and m, the mass of the sample.

The ^{226}Ra content of the samples was determined from the intensity of the 1.760 MeVphotopeak of ^{214}Bi, ^{232}Th content from the 2.614 MeVphotopeak from ^{208}Tl and potassium content from the 1.460 MeVphotopeak following the decay of ^{40}K. Each of the samples was counted for 36,000 seconds (10 h).

RESULTS AND DISCUSSION

Activity concentrations

The activity concentration of each radionuclide in the samples was obtained by relating the detection efficiency to the net count under each photopeak. The results obtained are shown in Tables 2 and 3 for the fish and crustacean samples. The mean activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th of each species of fish samples collected from fresh water varied from 462 ± 80 to 792 ± 107, 21.4 ± 3.8 to 38.6 ± 11.6 and 40.7 ± 25.9 to 84.4 ± 2.3 Bq.kg^{-1}, respectively. For the fish samples collected from marine water, it varied from 688 ± 230 to 791 ± 39, 23.0 ± 4.6 to 49.7 ± 33.1 and 32.1 ± 5.3 to 96.7 ± 19.9 Bq.kg^{-1}, respectively. The mean activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th of the crustacean samples were 525 ± 77, 35.3 ± 14.9 and 31.9 ± 5.2 Bq.kg^{-1}, respectively. The highest content of ^{40}K was obtained for *Gymnarchusniloticus* while the highest contents of ^{226}Ra and ^{232}Th were obtained for *Cynoglossussenegalensis*.

The activity concentrations of the natural radionuclides in the sediment samples are presented in Tables 4 and 5. The lower limits of detection (LLD) of the detector for the radionuclides were determined as 14.9 Bq.kg^{-1} (^{40}K, 1.460 MeV), 4.0 Bq.kg^{-1} (^{226}Ra, 1.760 MeV) and 4.6 Bq.kg^{-1} (^{232}Th, 2.614 MeV). Only values above the LLD were used to calculate the means. The mean activity concentrations of the sediment samples due to the ^{40}K, ^{226}Ra and ^{232}Th contents were 70.0 ± 23.1, 11.2 ± 2.2 and 10.3 ± 3.5 Bq.kg^{-1}, respectively for fresh water and 76.3 ± 32.3, 10.4 ± 3.8 and 10.1 ± 5.7 Bq.kg^{-1}, respectively for samples collected from marine water. From the results obtained, there are no significant differences between the radionuclide content of the fresh and marine water sediments.

Table 1:
The sampled fish species and their source

S/N	Species	Family	Common name	Source
1	*Gymnarchusniloticus*	Gymnarchidae	Trunk fish	Fresh water
2	*Parachanaobscura*	Channidae	Snake head fish	Fresh water
3	*Clariasanguillaris*	Claridae	Cat fish	Fresh water
4	*Heterotiniloticus*	Ostigloccidae	African bony tongue	Fresh water
5	*Oreochromisniloticus*	Chclidae	Tilapia	Fresh water
6	*CynoglossusSenegalensis*	Cynoglossidae	Tongue sole	Marine water
7	*Chrysicthysnigrodigitatus*	Bagridae	Silver cat fish	Marine water
8	*Arius heudelotic*	Arridae	Smooth mouth sea cat fish	Marine water
9	*Dasyatis margarita*	Dayatidae	Sting ray	Marine water
10	*Peanusmonodon*	Crustacean	Prawn	Marine water

Table 2:
Activity concentration of natural radionuclides and annual effective ingestion dose in fish samples from fresh water

SAMPLES/SPECIES		^{40}K (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	$H_{T,r}$ (μSv.y^{-1})
Gymnarchusniloticus	*1*	786	33.9	42.8	23.7
	2	689	30.1	13.8	12.9
	3	902	51.9	65.4	33.4
Mean ± Std. Dev.		*792 ± 107*	38.6 ± 11.6	40.7 ± 25.9	23.3 ± 10.2
Parachnnaobscura	*1*	642	35.4	84.1	36.1
	2	502	17.9	82.2	32.9
	3	424	43.2	86.8	35.4
Mean ± Std. Dev.		523 ± 110	32.2 ±12.9	84.4 ± 2.3	34.8 ±1.7
Heterotisniloticus	*1*	678	31.5	63.5	29.4
	2	582	34.5	49.7	24.1
	3	621	32.8	81.4	34.8
Mean ± Std. Dev		627 ± 48	32.9 ± 1.5	64.9 ± 15.9	29.4 ± 5.4
Clariasanguillaries	*1*	699	25.5	65.2	29.7
	2	573	18.0	61.9	26.9
	3	549	20.6	61.9	26.9
Mean ± Std. Dev		607 ± 81	21.4 ± 3.8	63.0 ± 1.9	27.8 ± 1.6
Oreochromisniloticus	*1*	456	34.0	44.9	21.3
	2	546	22.6	84.1	34.3
	3	385	20.1	28.1	14.2
Mean ± Std. Dev		462 ± 80	25.6 ± 7.4	52.4 ± 28.7	23.3 ± 10.2

Table 3:
Activity concentration of natural radionuclides and annual effective ingestion dose in fish and crustacean samples from marine water

SAMPLES/SPECIES		^{40}K (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	$H_{T,r}$ (μSv.y^{-1})
Cynoglossussenegalensis	*1*	832	23.5	112	15.4
	2	753	86.9	74.2	12.4
	3	788	38.8	104	14.7
Mean ± Std. Dev.		791 ± 39± 1.6	49.7 ± 33.1	96.7 ± 19.9	14.2 ± 1.6
Arius heudolotis	*1*	586	25.2	59.4	8.9
	2	526	26.0	46.1	7.3
	3	952	17.7	26.4	6.3
Mean ± Std. Dev.		688 ± 230	23.0 ± 4.6	44.0 ±16.6	7.5 ± 1.3
Dasyatis margarita	*1*	836	70.0	62.9	11.1
	2	837	25.9	35.9	7.1
	3	586	24.5	61.0	9.1
Mean ± Std. Dev		753 ± 145	40.1 ± 25.9	53.3 ± 15.1	9.1 ± 2.0
Chrysithctysnigrodigitatus	*1*	758	23.3	33.6	6.6
	2	680	26.2	26.2	5.6
	3	731	34.0	36.5	7.0
Mean ± Std. Dev		723 ± 39.6	27.8 ± 5.5	32.1 ± 5.3	6.4 ± 0.7
Peanusmonodon	*1*	515	18.0	32.9	2.3
	2	606	43.4	26.2	2. 3
	3	453	44.4	36.5	2.6
Mean ± Std. Dev		525 ± 77	35.3 ± 14.9	31.9 ± 5.2	2.4 ± 0.2

Annual effective ingestion dose of fish and crustacean samples

The annual effective ingestion dose for an adult member of the public due to the intake of radionuclide through ingestion of food can be calculated based on

the metabolic models developed by the International Commission of Radiological Protection (ICRP 1996);

$$H_{T,r} = \sum \left(U^i \times C_r^i\right) \times gT_r \qquad (2)$$

where i donate a food group, U^i is the consumption rate per capital (kg.y^{-1}), C_r^i is the activity concentrations of radionuclide r of interest (Bq.kg^{-1}) and gT_r is the dose conversion coefficient for ingestion of radionuclide r (Sv.Bq^{-1}) in tissue. The dose conversion coefficients for the radionuclides determined in this work are given as6.2 × 10^{-9}, 2.8 × 10^{-7} and 2.2 × 10^{-7}Sv.Bq^{-1}for ^{40}K, ^{226}Ra and ^{232}Th, respectively(IAEA 1996). Consumption rate per capital in Nigeria is given as 1.5 kg.y^{-1}, 0.5 kg.y^{-1} and 0.2 kg.y^{-1} for fresh water fish, marine water fish and crustacean, respectively (FOS, 2006). The means of the annual effective ingestion dose due to the content of the three radionuclides in the fresh water fish samples varied between 23.3 ± 10.2µSv.y^{-1} (*Oreochromisniloticus* and *Gymnarchusniloticus*) and 34.8 ± 1.7 µSv.y^{-1} (*Parachnnaobscura*).

Table 4:
Activity concentration of natural radionuclides in fresh water river sediment

S/N	^{40}K (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})
1	61.3 ± 5.9	10.4 ± 1.5	11.4 ± 1.9
2	52.6 ± 5.6	8.1 ± 1.4	BDL
3	46.6 ± 5.4	11.8 ± 1.5	15.8 ± 2.2
4	114.6 ± 5.8	9.6 ± 1.5	BDL
5	52.8 ± 5.6	13.7 ± 1.5	7.0 ± 1.7
6	49.9 ± 5.5	10.1 ± 1.5	12.0 ± 2.0
7	92.5 ± 6.9	14.0 ± 1.5	7.1 ± 1.7
8	89.8 ± 6.9	8.5 ± 1.4	12.6 ± 2.0
9	81.7 ± 6.6	14.3 ± 1.6	BDL
10	58.2 ± 5.8	11.0 ± 1.5	6.4 ± 1.6
Mean ± SD	70.0 ± 23.1	11.2 ± 2.2	10.3 ± 3.5

BDL indicates below detectable limit

For the marine water fish samples, it varied between6.4 ± 0.7 µSv.y^{-1} (*Chrysithctysnigrodigitatus*)and14.2 ± 1.6 µSv.y^{-1} (*Cynoglossussenegalensis*).The mean annual ingestion dose for crustacean (*Peanusmonodon*) is 2.4 ± 0.2 µSv.y^{-1}. The annual effective dose calculated for all the samples are less than 1 mSv.y^{-1}. Potassium-40 which is an isotope of essential element is usually of limited interest. It is homeostatically controlled in humans and as a result the body content of ^{40}K is determined largely by its physiological characteristics rather than by its intake. From the results obtained the radiation dose incurred from the ingestion of the fish and crustacean samples pose no significant health effect to the population from a radiological point of view.

Table 5:
Activity concentration of natural radionuclides in marine water sediment

S/N	^{40}K (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})
1	51.8 ± 5.6	6.6 ± 1.4	13.8 ± 2.1
2	60.6 ± 5.9	10.8 ± 1.5	7.3 ± 1.7
3	21.4 ± 8.1	8.3 ± 1.5	23.6 ± 2.9
4	92.3 ± 6.9	19.4 ± 1.4	11.3 ±1.9
5	84.4 ± 6.6	10.5 ± 1.5	5.7 ±1.6
6	99.8 ± 7.2	11.9 ± 1.5	8.3± 1.4
7	83.8 ± 6.6	13.0 ± 1.5	9.6 ± 1.8
8	BDL	7.9 ± 1.4	BDL
9	17.0 ± 4.8	8.7 ± 1.4	6.2 ± 1.6
10	BDL	7.3 ± 1.4	5.5 ± 1.6
Mean ± SD	76.3 ± 32.3	10.4 ± 3.8	10.1 ± 5.7

BDL indicates below detectable limit

Assessment of skin dose to humans

Equivalent dose to the skin of humans from contact with sediment containing radionuclides can be calculated using a method given by (Little *et al* 1997);

$$E_{skin} = H_{skin,i} + H_{skin,j} + \text{etc.} \qquad (3)$$

where

$$H_{skin,i} = A_{s,i} \times T_s \times (\beta_{skin,i} + \gamma_{skin,i}) \qquad (4)$$

and

$H_{skin,I}$is the contact skin dose from beta and gamma emission from radionuclide i in environment materials (Sv.y^{-1})

$A_{s,I}$is the activity per unit area of radionuclide i on skin (Bq.cm^{-2})

T_s is the skin contact time (h.y^{-1})

$\beta_{skin,i}$ is the skin equivalent dose rate to the basal layer of epidermis (4 mg.cm^{-2}) for beta particle irradiation from radionuclide i (Sv.h^{-1} per Bq.cm^{-2})

$\gamma_{skin,I}$ is the skin equivalent dose rate to the basal layer of epidermis (7 mg.cm^{-2}) for gamma irradiation from radionuclide i (Sv.h^{-1} per Bq.cm^{-2})

The activity per unit area on the skin can be calculated as (EA 2003);

$$A_{s,i} = C_i \times \rho \times d \qquad (5)$$

where C_i is the concentration of radionuclide i in (dry) sediment ($Bq.kg^{-1}$), ρ is the density of dry sediment ($kg.cm^{-3}$) and d is the thickness of dry sediment on the skin (cm). The density of dry sediment, ρ used was 1.5×10^{-3} $kg.cm^{-3}$, and the thickness of dry sediment on the skin d, used was 0.01 cm (EA 2003). The skin contact time was taken as 500 $h.y^{-1}$ and 300 $h.y^{-1}$ for fresh and marine water sediments, respectively (EA 2003). The skin equivalent dose rate for each radionuclide to the basal layer of epidermis for beta particle irradiation (β_{skin}) and gamma irradiation (γ_{skin}) were taken from Harvey *et al* (1993). The mean activity concentrations of the radionuclides in the sediment samples were used to calculate the equivalent dose to the skin from contact with the sediment. The results were 2.0 $\mu Sv.y^{-1}$ and 1.2 $\mu Sv.y^{-1}$, respectively for the fresh and marine water sediments. The results obtained are lower than the ICRP recommended annual equivalent dose of 50 mSv to the public (ICRP 2007). To obtain the effective dose from contact with the sediment, the equivalent dose was multiplied by tissue weighting factor for UV exposed skin, 0.01 (ICRP 1990), and fraction of UV exposed skin in contact with sediment, 0.5 (Oatway and Mobbs, 2003). The effective doses calculated were 0.010 $\mu Sv.y^{-1}$ and 0.006 $\mu Sv.y^{-1}$ for the fresh and marine water sediments, respectively.

Conclusion

The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in fish, crustacean and sediment samples from fresh and marine water in oil exploration area in Ondo State, Nigeria has been investigated using gamma-ray spectroscopy method. The annual effective dose due to intake of radionuclides through the consumption of fish and crustacean was calculated. The highest value of 34.8 ± 1.6 $\mu Sv.y^{-1}$ was obtained for *Parachanaobscura* while the least value 2.4 ± 0.2 $\mu Sv\ y^{-1}$ was obtained for *Peanusmonodon*. The equivalent and effective doses to the skin from contact with the sediments were calculated as 2.0 $\mu Sv.y^{-1}$ and 0.010 $\mu Sv.y^{-1}$, respectively for fresh water samples and 1.2 $\mu Sv.y^{-1}$ and 0.006 $\mu Sv.y^{-1}$ for marine water samples. The results obtained for the equivalent doses are less than the ICRP recommended value of 50 mSv y^{-1} to the public.

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