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Determination of ^{40}K, ^{226}Ra and ^{228}Ra Concentrations with Dose Rates in Crayfish from Ode Omi River and Radiological Implications to the Consumers

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Abstract

Natural radionuclides such as ^{40}K, ^{226}Ra and ^{228}Ra are found naturally in water and sediments of rivers, likewise in soils. They are transferred to aquatic animals through ingestion. Radioactivity concentrations and dose rates of ^{40}K, ^{226}Ra and ^{228}Ra in crayfish species from Ode Omi River in coastal region, Ogun State Southwest of Nigeria were determined by gamma spectrometry using NaI (TI) detector coupled with a pre-amplifier base to a multiple channel analyzer (MCA). 15 samples of crayfish were collected. *Cherax tenuimanus* had the highest mean concentration and dose rate of values 106.87 ± 9.11 Bq kg^{-1} and 0.00960 mGy hr^{-1} respectively for ^{40}K. Moreso, for ^{226}Ra, *Cherax quadricarinatus* had the highest mean concentration and dose rate of values 2.57 ± 0.82 Bq kg^{-1} and 4.50 x 10^{-7} mGy hr^{-1} respectively. Concerning^{228}Ra, *Astacopsis gouldi* had the highest mean concentration and dose rate of values 5.23 ± 0.46 Bq kg^{-1} and 5.86 x 10^{-13} mGy hr^{-1}, respectively. The average dose rate of the radionuclides in all the crayfish was calculated to be 4.62 x 10^{-3} mGy hr^{-1} which was below the 0.4 mGy hr^{-1} limit recommended. The average annual committed effective dose and average excess lifetime cancer risk of all the radionuclides to the consumers were 0.0807 mSv yr^{-1} and 0.589 x 10^{-4}, respectively, which were below global limits of 1.0 mSv yr^{-1} and 0.29 x 10^{-3}, respectively, therefore, the ingestion of these natural radionuclides could not pose any radiological health hazards to the aquatic animals, likewise man the consumers.

Keywords: Radionuclide concentration, Dose rate, Cancer Risk.

Introduction

Crayfish as aquatic animal is found in rivers of most towns in western, eastern and southern parts of Nigeria, and is being fed on by human beings due to the fact that is a good source of protein and contains good quantity of calcium. Radionuclides are chemical elements with unstable atomic structures called radioactive isotopes. The unstable structures breakdown to release or emit radiation energy from the nuclei or other parts of the atom. Radionuclides find their ways into the aquatic animals through the river water, sediment, plants, insects and other things that are present in the river. Three types of radiation can be released: alpha particles, beta particles and gamma rays (photons). Most naturally occurring radionuclides are alpha particle emitters (uranium and radium-226), but some beta particle emitters also occur naturally (radium-228 and potassium-40). Manmade radionuclides are mainly beta and photon (gamma) emitters. Tritium is a beta particle emitter that may be formed naturally in the atmosphere or by human activities (OEPA

2005). Radiation being energy emitted when a radionuclide decays. It can affect living tissues only when the energy is absorbed in that tissue. Radionuclides can be hazardous to living tissues when they are inside an organism where radiation released can be immediately absorbed. They may also be hazardous when they are outside of the organism but close enough for some radiation to be absorbed by the tissues. Radionuclides move through the environment and into the body through different pathways: through the air, in water and through the food chains. Knowing these pathways make it possible to take necessary control measures to reduce their intake by aquatic animals, terrestrial animals and human beings to minimal levels.

A study of the radionuclides: ^{40}K, ^{226}Ra and ^{228}Ra concentration levels was carried out by Sowole et al. (2019) along with their dose rates in species of fish from Victoria Island lagoon in Lagos State, Southwest of Nigeria. The average dose rates of ^{40}K, ^{226}Ra and ^{228}Ra in the fishes were found to be 0.0049 mGy hr^{-1}, 5.32 x 10^{-7} mGy hr^{-1} and 8.96 x 10^{-13} mGy hr^{-1}, respectively which were below the limit of 0.4 mGy hr^{-1} recommended by NCRP (1991) as reported by Blaylock et al. (1993). The annual dose rate in man consuming them was calculated to be 0.216 mSv hr^{-1} which was below the limit of 1.0 mSv hr^{-1} (ICRP 2007); therefore could not pose radiological health problems to the aquatic animals and the consumers. Also, 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 by Ademola and Ehiedu (2010), the activity concentrations of natural radionuclides: ^{40}K, ^{226}Ra and ^{232}Th were determined using gamma spectrometry method. The means of the annual effective ingestion dose varied between 23.3 ± 10.2 μSv yr^{-1} (*Oreochromis niloticus* and *Gymnarchus niloticus*) and 34.8 ± 1.7 μSv yr^{-1}(*Parachanna obscura*) for fresh water fish samples and 6.4 ± 0.7 μSv yr^{-1}(*Chrysithctys nigrodigitatus*) and 14.2 ± 1.6 μSv yr^{-1}(*Cynoglossus senegalensis*) for marine water fish samples. That of the crustacean samples (*Penaeus monodon*) was 2.4 ± 0.2 μSv yr^{-1}.

This research work was carried out to determine the activity concentrations and dose rates of ^{40}K, ^{226}Ra and ^{228}Ra in crayfish samples from Ode Omi River in coastal region of Southwest of Nigeria. It was also intended to calculate the annual committed effective doses and excess lifetime cancer risk to man that consumes them.

Study Area

Ode Omi is a town located in Ogun Waterside, Ogun State, Southwest of Nigeria. Its geographical coordinates are 6° 24' 0" North, 4° 20' 0" East. Ode Omi River falls in the Southwest coastal region of Nigeria. Ode Omi is a community of more than 6,000 people; their major sources of occupation are cassava cultivation, palm oil making and fishing. Figure 1 shows the study area.

Materials and Methods

A total of 15 samples of crayfish from Ode Omi River were collected in November 2020, and 4 species were obtained as shown in Table 1. This was done by the use of fishing nets at a distance of about 300 m from each location along the River, after which they were preserved in 40% formaldehyde in labelled containers. They were identified and grouped into their species. The groups were then oven dried at 80 °C (Akinloye et al. 1999). The dried animal samples were later pulverized, weighed, packed 100.0 g by mass in plastic containers and carefully sealed for 4 weeks in order to establish secular radioactive equilibrium between the natural radionuclides and their respective progenies.

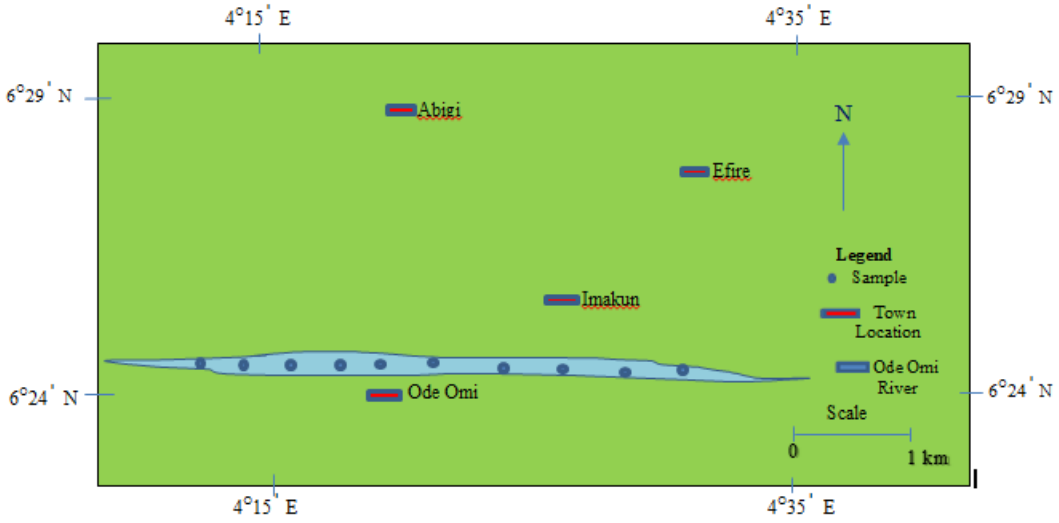

Figure 1: Map of study area showing samples collection along the River.

Table 1: Species and number of samples collected from Ode Omi River

Name of species	Number of samples
Austropotamobius pallipes	5
Cherax tenuimanus	4
Cherax quadricarinatus	3
Astacopsis gouldi	3

The method of gamma spectrometry was adopted for the analysis of the samples collected in order to obtain data on ^{40}K, ^{226}Ra and ^{228}Ra. The spectrometer used was a Canberra lead shielded 7.6 cm x 7.6 cm NaI (Tl) detector coupled to a multichannel analyzer (MCA) through a preamplifier base. The spectrometer was calibrated using known concentrations and of the same geometry as the samples. The linearity of the NaI (Tl) detector system used in this work was examined at fixed values of the lower limit discrimination (LLD) and analog to digital conversion (ADC). A linear relationship was obtained between the channel number and the γ-energy E in MeV, for the purpose of identifying the various radionuclides that were present in the samples based on the gamma energies they emit. The spectrometer used was calibrated by determining the detection efficiencies of the radionuclides ^{40}K, ^{226}Ra, ^{228}Ra and ^{137}Cs, keeping the detector–sample arrangement constant throughout the period of the analyses of the samples with the same counting time. The radionuclides emit gamma rays having energy range of 0.662 to 2.615 MeV. In order to obtain detection efficiencies $\varepsilon(E_\gamma)$ for the range of energies of the radiations in the samples, a standard reference source at a fixed geometry were counted for 10 hours in order to obtain $N(E\gamma)$ at each gamma energy, and evaluating $\varepsilon(E_\gamma)$ from Equation 1 (IAEA 1989):

$$\varepsilon\,(E_\gamma) = \frac{N(E\gamma)}{AI\gamma Mtc} \qquad 1$$

Where: $N(E_\gamma)$ = Net peak area of the radionuclide of interest, $\varepsilon(E_\gamma)$ = Efficiency of the detector for the γ- energy of interest, I_γ = Intensity per decay for the γ- energy of interest, M = Mass of the sample, and t_c = Total counting time in seconds (36000 s). For the aquatic animals, this standard sample had been prepared from Rocketdyne Laboratories

California, and it was traceable to mixed standard gamma source (No 48722-356) by Analytics Inc. Atlanta, USA. The efficiencies were determined using a standard aquatic sample Ref. No IAEA-MA-B-3/RN (AQCS, 1998). The reference sample was counted at the same fixed geometry of the spectrometric assembly.

The resolution of the detector is about 8% at 0.662 MeV of ^{137}Cs. For the analysis of ^{40}K, ^{226}Ra and ^{228}Ra, the photo peak regions of ^{40}K (1.46 MeV), ^{214}Bi (1.76 MeV) and ^{208}TI (2.615 MeV) were, respectively used.

The cylindrical plastic containers holding the samples were put to sit on the high geometry 7.6 cm x 7.6 cm NaI (TI) detector. High level shielding against the environmental background radiation was achieved by counting in a Canberra 10 cm thick lead castle. The counting of each sample was done for 10 hrs because of suspected low activities of the radionuclides in the samples. The areas under the photo-peaks of ^{40}K, ^{226}Ra and ^{228}Ra were computed using the Multichannel Analyzer system. The concentrations of the radionuclides were calculated based on the measured efficiency of the detector and the net count rate under each photopeak over a period of 10 hours using Equation 2 (IAEA 1989):

$$A = \frac{N\,(E\gamma)}{\varepsilon(E\gamma) I\gamma M t c} \qquad 2$$

Radiological indicators that were used to assess the radiological risks to the aquatic animals and the consumers were: dose rate (D), annual committed effective dose (ACED) and excess lifetime cancer risk (ELCR).

Dose rate (D): the dose rates of the radionuclides in the aquatic species were calculated using Equation 3 (Blaylock et al. 1993):

$$D = 5.76 \times 10^{-4}\, En\Phi C \qquad 3$$

Where: E was the average emitted energy for gamma radiations (MeV), n was the proportion of transitions producing an emission of energy E, Φ was the fraction of the emitted energy absorbed and the values of the parameters were obtained from Blaylock et al. (1993), C was the concentration of the radionuclide of consideration and D was the dose rate of radionuclide of consideration.

Annual committed effective dose (ACED): For ingestion of radionuclides in the aquatic animals by man, this was determined using the following expression (Tettey-Larby et al. 2013):

$$ACED = C\ x\ DCF\ x\ CR \qquad 4$$

Where: C = Concentration of each radionuclide, DCF = Dose conversion factor for ingestion of the natural radionuclides, obtained from ICRP (2012); 6.2 x 10^{-6} mSv Bq^{-1}, 4.4 x 10^{-4} mSv Bq^{-1} and 2.2 x 10^{-4} mSv Bq^{-1} for ^{40}K, ^{238}U and ^{232}Th, respectively, and CR = Consumption rate of intake of naturally occurring radioactive materials from the foodstuffs of value 144 kg yr^{-1}.

Excess lifetime cancer risk (ELCR): This was determined based on the value of the annual committed effective dose using Equation 5 (ICRP 2007):

$$ELCR = ACED\ x\ LE\ x\ RF \qquad 5$$

Where: LE is life expectancy taken to be 70 years and RF is fatal risk factor per Sievert which was 0.05 (ICRP 2007).

Results and Discussion

Radioactivity concentrations of radionuclides in the crayfish are shown in Table 2. Ranges mean values of activity concentrations of ^{40}K and dose rates in aquatic species were shown in Table 3; *Cherax tenuimanus* had the highest mean concentration of ^{40}K and dose rate of values 106.87 ± 9.11 Bq kg^{-1} and 0.00960 mGy hr^{-1}, respectively. Moreso for ^{226}Ra, *Cherax quadricarinatus* had the highest mean concentration and dose rate values 2.57 ± 0.82 Bq kg^{-1} and 4.50 x 10^{-7} mGy hr^{-1}, respectively.

Table 2: Activity concentrations of radionuclides in crayfish (Bq kg^{-1}**)** from Ode Omi River

Species	Sample codes	Activity concentrations of radionuclides		
		^{40}K	^{226}Ra	^{228}Ra
Austropotamobius pallipes	CFS_1	99.32 ± 8.43	2.05 ± 0.32	4.16 ± 0.30
Austropotamobius pallipes	CFS_2	106.82 ± 9.65	3.96 ± 0.25	5.24 ± 0.38
Austropotamobius pallipes	CFS_3	89.73 ± 5.84	1.47 ± 0.64	2.63 ± 0.14
Austropotamobius pallipes	CFS_4	87.07 ± 5.83	1.72 ± 0.34	4.86 ± 0.35
Austropotamobius pallipes	CFS_5	103.14 ± 8.45	0.82 ± 0.09	3.12 ± 0.20
Astacopsis gouldi	CFS_6	105.49 ± 8.89	3.15 ± 0.52	7.36 ± 0.76
Astacopsis gouldi	CFS_7	114.82 ± 9.92	0.69 ± 0.15	4.16 ± 0.30
Astacopsis gouldi	CFS_8	100.04 ± 8.43	1.19 ± 0.62	4.17± 0.31
Cherax quadricarinatus	CFS_9	98.68 ± 7.86	3.17 ± 0.64	1.17 ± 0.06
Cherax quadricarinatus	CFS_{10}	102.58 ± 8.45	2.06 ± 0.87	4.37 ± 0.33
Cherax quadricarinatus	CFS_{11}	99.04 ± 8.34	2.47 ± 1.06	5.02 ± 0.36
Cherax tenuimanus	CFS_{12}	114.47 ± 9.86	1.58 ± 0.61	6.06 ± 0.72
Cherax tenuimanus	CFS_{13}	103.18 ± 8.45	2.86 ± 0.37	4.06 ± 0.30
Cherax tenuimanus	CFS_{14}	102.48 ± 8.45	1.04 ± 0.22	5.76 ± 0.47
Cherax tenuimanus	CFS_{15}	107.36 ± 9.68	3.02 ± 0.81	4.18 ± 0.31

Note: CFS was crayfish sample. The confidence of interval was 99%.

Table 3: Range, mean values of activity concentrations of ^{40}K, dose rate in crayfish, annual committed effective dose and excess lifetime cancer risks

Species	Range (Bq kg^{-1})	Mean (Bq kg^{-1})	Dose rate (mGy hr^{-1})	ACED (mSv yr^{-1})	ELCR x 10^{-3}
Austropotamobius pallipes	87.07–106.82	97.22 ± 7.64	0.00874	0.0072	0.0252
Astacopsis gouldi	100.04–114.82	106.78 ± 9.08	0.00959	0.0079	0.0277
Cherax quadricarinatus	98.68–102.58	100.10 ± 8.23	0.00899	0.0075	0.0263
Cherax tenuimanus	102.48–114.47	106.87 ± 9.11	0.00960	0.0080	0.0280

Concerning ^{228}Ra as shown in Table 5, *Astacopsis gouldi* had the highest mean concentration and dose rate of values 5.23 ± 0.46 Bq kg^{-1} and 6.24 x 10^{-13} mGy hr^{-1} respectively. The average dose rates of ^{40}K, ^{226}Ra and ^{228}Ra in all the crayfish were calculated to be 9.23 x 10^{-3} mGy hr^{-1}, 3.67 x 10^{-7} mGy hr^{-1} and 5.30 x 10^{-13} mGy hr^{-1}, respectively.

The average dose rate of all the radionuclides in the crayfish was 3.08 x 10^{-3} mGy hr^{-1}. The values were all below the limit of 0.4 mGy hr^{-1} recommended by NCRP (1991) as reported by Blaylock et al. (1993). Highest annual committed effective dose intake (Table 4) of ^{40}K by man was from *Cherax tenuimanus* of value 0.0080 mSv yr^{-1} with average value of 0.0077 mSv yr^{-1}, that of ^{226}Ra was from *Cherax quadricarinatus* of value 0.0086 mSv yr^{-1} with average value of 0.0071 mSv $yr^{-1}$and that of ^{228}Ra was 0.0421 mSv yr^{-1} with average value of 0.0358mSv yr^{-1}. Furthermore, average annual committed effective dose of all the radionuclides to the consumers of the aquatic animals was calculated to be 0.0169 mSv yr^{-1} which was higher than the value obtained by Ademola and Ehiedu (2010), but was below the limit of 1.0 mSv yr^{-1} (ICRP 2007). The average excess lifetime cancer risk of all the radionuclides to the consumers was calculated to be 0.589 x 10^{-4} which was below the average value of 0.29 x 10^{-3} (UNSCEAR 2000).

Table 4: Range, mean values of activity concentration of ^{226}Ra, dose rate in crayfish, annual committed effective dose and excess lifetime cancer risk

Species	Range (Bq kg^{-1})	Mean (Bq kg^{-1})	Dose rate (mGy hr^{-1})	ACED (mSv yr^{-1})	ELCR x10^{-3}
Austropotamobius pallipes	0.82–3.96	2.00 ± 0.33	3.50 x 10^{-7}	0.0067	0.0235
Astacopsis gouldi	0.69–3.15	1.68 ± 0.43	2.94 x 10^{-7}	0.0057	0.0200
Cherax tenuimanus	1.04–3.02	2.13 ± 0.50	3.73 x 10^{-7}	0.0072	0.0252
Cherax quadricarinatus	2.06–3.17	2.57 ± 0.82	4.50 x 10^{-7}	0.0086	0.0301

Table 5: Range, mean values of activity concentration of ^{228}Ra, dose rate in crayfish, annual committed effective dose and excess lifetime cancer risk

Species	Range (Bq kg^{-1})	Mean (Bq kg^{-1})	Dose rate (mGy hr^{-1})	ACED (mSv yr^{-1})	ELCR x 10^{-3}
Austropotamobius pallipes	2.63–5.24	4.00 ± 0.27	4.77 x 10^{-13}	0.0322	0.1127
Astacopsis gouldi	4.16–7.36	5.23 ± 0.46	6.24 x 10^{-13}	0.0421	0.1474
Cherax quadricarinatus	1.17–5.02	3.52 ± 0.25	4.20 x 10^{-13}	0.0283	0.0991
Cherax tenuimanus	4.06–6.06	5.02 ± 0.45	5.99 x 10^{-13}	0.0404	0.1414

Conclusion

The study of the radionuclides concentration levels had been carried out along with their dose rates in the aquatic animals. The average dose rate of all the radionuclides in all the crayfish species was found to be below the limit recommended. The average annual committed effective dose of all the radionuclides to the consumers of the aquatic animals was also determined to be within the limit recommended globally. Furthermore, average excess lifetime cancer risk of all the radionuclides to the consumers was computed to be within the limit recommended. Therefore, the ingestion of these natural radionuclides did not pose any radiological health hazards to the aquatic animals likewise man the consumer.

Conflict of Interest

The authors declare no conflict of interest.

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