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A Radiological Risk Assessment of ^{226}Ra, ^{228}Ra and ^{40}K Isotopes in Tilapia Fish and its Granitic Environment in Singida Municipality, Tanzania

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Abstract: Consumption of fish containing elevated levels of radionuclides can lead to undesirable health effects for consumers. People in the Singida Municipality harvest fish from lakes and ponds of granite rocks which are linked with hazardous radioisotopes that may be bio-concentrated by fishes they consume. Currently, no study has ascertained the levels of radioisotopes in fish from these environments. This study was carried out to analyse the radioactivity levels of ^{226}Ra, ^{228}Ra and ^{40}K isotopes in order to assess the radiological risk associated with Tilapia fish consumption and its environment in Singida Municipality. Some 51 samples, which included water (20), sediment (20), Nile tilapia (8) and Manyara tilapia (3), were randomly sampled and composited; then, they were analysed using a high-purity germanium (HPGe) detector, between May and June 2022. The results revealed that (i) the activity levels of ^{228}Ra were below the detection limit for fish and water samples, while in sediment, the combined activity of ^{228}Ra was within the acceptable international levels; (ii) the mean activity concentrations of ^{226}Ra and ^{40}K in all other samples were within the recommended levels; (iii) the activities of radionuclides in the samples analysed were high in sediments, followed by fish, and lastly water; (iv) the bioaccumulation results show that only ^{40}K was bio-accumulated (with 1.26 in Nile tilapia), while other radionuclides (^{226}Ra, ^{228}Ra) were not bio-accumulated; (vi) the radionuclide transfer from water to fish was higher compared to the radionuclide transfer from sediment to fish; (vii) the human effective doses due to consumption of Nile tilapia and Manyara tilapia were 0.00973 and 0.005 mSv/y, respectively, which is below the 1 mSv/y international limit. These findings therefore show that the current levels of radioactivity in fish in the study area do not pose a significant radiological risk to fish consumers. However, more studies on other types of fish are recommended.

Keywords: sediment; water; radionuclides; gamma spectrometry; annual effective dose

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1. Introduction

The living environment is continuously exposed to radionuclides such as ^{226}Ra, ^{228}Ra and ^{40}K, which are widely distributed throughout rocks, soil, sediment, food, air and water, with different concentration levels. Their existence in the environment can be attributed to naturally occurring and artificially induced sources [1]. Natural radionuclides are mainly released from rocks, such as granite rock, as naturally occurring radioactive materials (NORMs), or as products of human activities such as mining operations; they are referred to as technically enhanced naturally occurring radioactive materials (TENORMs). Thus, radionuclides are transported to the aquatic environment and bio-concentrated by aquatic organisms including Tilapia fish [2]. The radionuclides found in igneous rocks including granite rocks may be concentrated by physical, biological, and chemical processes before entering the food chain [3].

Studies on NORMs, TENORMs and artificial radionuclides are usually carried out around the world to attain data about the radioactivity levels of ionizing radiation in the environment [4–6]. Similarly, many studies have been carried out in Tanzania on the distribution of natural radioactivity in sediments, soils, surface water, and foodstuffs [7–9]. However, information on the health risks associated with natural radionuclides in tilapia fish is still limited to the best of the authors' knowledge, particularly in this study area. It is estimated that living organisms receive at least one eighth of the total effective dose resulting from radionuclides, through consumption of food materials including fish [6]. Thus, it is important to measure the radioactivity concentrations of fish species and their surrounding source of radionuclides; this will then help us to take appropriate precautions to reduce radionuclide intake.

The Singida municipality is known to be predominantly granitic, with higher levels of natural radioactive and various water bodies. Moreover, a number of anthropogenic activities such as agriculture and mining are taking place in the environs of Singida, and this may enhance levels of natural radioactivity in the surrounding environment. This area drew the attention of this research study, as its water bodies are most vulnerable to NORMs and TENORMs [10].

Therefore, this study was designed to assess the radiological risk of ^{226}Ra, ^{228}Ra, and ^{40}K in Tilapia fish, which is a preferred protein food, in relation to its granitic environment. The information obtained may be used to estimate the annual effective dose due to consumption of fish. The results of this research will benefit the regulatory authorities on fish and environmental monitoring. Furthermore, upon publication, this study will increase societal knowledge of fish and their environment.

2. Materials and Methods

2.1. Study Area

The Singida municipality is located in Tanzania at the coordinate of 4°40′ East and 4°53′ South of equator, and longitude 34°30′ and 34°53′ East of Greenwich Mean Time (GMT). The geology of the area is dominated by the Tanzanian granite rock formation, which incorporates an closed internal drainage system with Uranium deposits reported within the same region [11]. Its climatic condition is semi-arid with a long dry season. The common social economic activities in the area include activities like fishing, agriculture and mining, as shown in Figure 1.

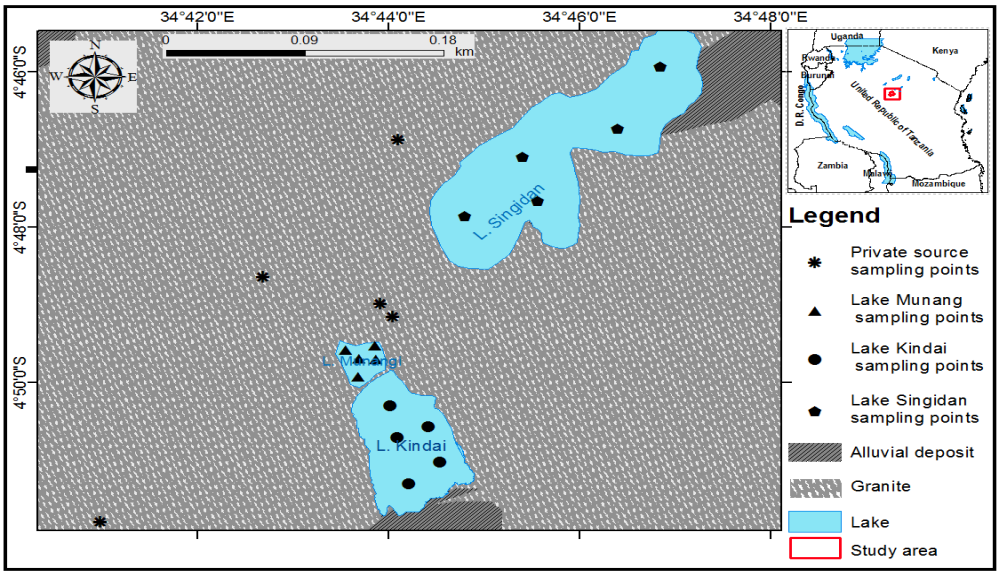

Figure 1. A geological map showing the domination of granite rocks and sampling points in study area at Singida Municipality.

2.2. Sampling and Sample Preparation for Gamma Spectrometry

Fifty-one samples were first randomised to minimise impartiality and then composited to reduce objectivity, and then manageable samples were obtained. Thus, samples were collected from eight areas which were then divided into two major groups including ponds (5) and lakes (3). A total of 51 samples comprising water (20), sediment (20), Nile tilapia (8) and Manyara tilapia (3) were collected. Nile tilapia were obtained in all water bodies but Manyara tilapia were obtained only from Lake Kindai, Lake Munang and Lake Singidani. In each sampling station, samples of sediments and water were collected, and fish samples were randomly sampled from fishermen around water bodies. Data such as coordinates were taken onsite using a Global Positioning System (GPS) machine from Garmin. Guidelines regarding the measurement of radionuclides in food and the environment were observed throughout, as guided by IAEA 1989.

The details of the study area and sampling points are indicated in Figure 1.

2.3. Sediment Sampling and Laboratory Preparations

A grab sampler (a bucket of 10 litres with stone of about 4 kg) was used by a diver to collect 15 sediment samples (about 1 kg each) from Kindai Lake, Munang Lake and Singidani Lake, with an average depth of 1.5 m. Then, from the bottom of the three lakes, sediments from around 0–10 cm depth were collected under minimum disturbance. A trowel was used to collect five sediment samples from each of the five ponds at approximately 0.2 to 0.5 m depth on average; thereafter, these were merged to obtain about 1 kg of sediment subsampled from each pond. Therefore; five sub-samples from five ponds were collected after random sampling and stored in a cool box to maintain a temperature of about 4 °C. These samples were then transported to Tanzania Atomic Energy commission (TAEC) laboratories for further laboratory work.

Upon arriving in the laboratory, the sediment samples were air dried under shade for about six hours then dried in an automatic oven for about two days at a temperature of 105 °C until a constant weight was reached, in order to evaporate water until only the dried sediment remained. The dried samples were ground using a Retch milling machine; the samples were cleaned well with ethanol after each sample milling and sieved in 2 mm to obtain homogeneous samples [12]. Thereafter, each prepared sample was packed in canisters with a specific geometry (100 mm in diameter and 40 mm in height) similar to that of the calibration source provided by the International Atomic Energy Agency (IAEA). Consequently, these canisters were used for packing, and they were well labelled then closed tightly with cello tape to avoid air exchange and escape of radon gas. Samples were then stored for more than 28 days for the uranium and thorium radionuclide daughters to attain secular equilibrium between short-lived progeny and the respective long-lived (Ra-226 from U-238) and (Ra-228 from Th-232), before gamma spectrometric analysis [13].

2.4. Water Sampling and Laboratory Preparations

Fifteen water samples from the three lakes (Kindai, Munang, and Singidani) were collected using a glass bottle (Coke bottle 350 mL) tied with a stone of about 0.2 kg and a rope of about 3 m to collect deep water samples from bottom, middle and surface water; these were then mixed up to obtain about 1.125 L of a composite sample of each sampling station. Water samples from the fish ponds were also collected using a glass bottle; these samples included deep, middle, and surface water, with a depth between 0.2–0.5 m. Samples were taken in the middle and near the bank of the pond from five points, then composited to obtain a minimum of 1.125 L per sample. Therefore, five sub-samples from five ponds were collected and stored in a cool box to maintain a temperature of about 4 °C. Generally, twenty samples of water were collected from the ponds and lakes. These water samples were then pre-treated with drops of concentrated hydrochloric acid (HCl) acid, then transported to TAEC laboratories for further laboratory preparation [13].

In the laboratory, water samples were transferred without filtration to a one litre Minnelli (with a similar geometry to that of the calibration source), and were pre-washed

with distilled water, dried, and rinsed with acetone to avoid impurities. The twenty water samples were then labelled and closed tightly with the cello tape to avoid any escape of radon gas. Samples were then stored for more than 28 days for uranium and thorium radionuclide daughters to attain secular equilibrium between short-lived progeny and the respective long-lived (Ra-226 from U-238) and (Ra-228 from Th-232), before gamma spectrometric analysis [8].

2.5. Fish Sampling and Laboratory Preparations

Eight Nile tilapia fish samples were sampled around the lakes and ponds, whereas three samples of Manyara tilapia were sampled from the three lakes. The collected fish samples were of approximately the same size, and were identified with the help of Singida fishery officers and fishermen [14]. These samples were well labelled and packed in special plastic bags, and stored in ice-cool boxes. During transportation to the TAEC laboratories, they were kept at approximately 4 °C by storage with ice cubes in cool boxes.

Laboratory preparations of fish started by taking them out of the cool box for thawing for about six hours. Nile tilapia were then dissected in order to obtain the edible parts of the fish (the fish muscles). Whole Manyara tilapia (i.e., how local residents consume them) were air-dried and then oven-dried at a temperature of 70 °C for about two days until they reached a constant weight (to ensure that they were completely dried). These samples were then crushed down with a grinding machine and cleaned with acetone to avoid contaminations; they were then sieved in a 2 mm mesh to obtain a homogeneous powder [15]. Thereafter, samples were packed in canisters with a specific geometry similar to that of the calibration source provided by the IAEA. They were well labelled then closed tightly with cello tape to prevent radon from escaping. Samples were then stored for more than 28 days, so that Uranium and thorium radionuclide daughters could attain secular equilibrium between short-lived progeny and the respective long-lived (Ra-226 from U-238) and (Ra-228 from Th-232), before gamma spectrometric analysis [16].

2.6. Gamma Spectrometric Analysis

Some 51 samples, which included water (20), sediment (20), Nile tilapia (8) and Manyara tilapia (3), were analysed using a high-purity germanium (HPGe) detector. During analysis, the photo-peaks regularly observed in the samples were identified, and belong to the natural radioactive decay series led by ^{228}Ra, ^{226}Ra, and a third natural radionuclide, ^{40}K. The activity of ^{226}Ra was determined using the gamma-lines of ^{214}Pb (295.2 and 351.9 keV) and ^{214}Bi (609.3 keV), which represented the ^{226}Ra. The activity for ^{228}Ra was measured from ^{228}Ac (338.3 and 911.1 keV). The ^{40}K was measured from its gamma-line energy of 1460.8 keV. Efficiency calibration was calculated by subjecting it to expression (1) and using a standard source with known parameters, while energy calibration was observed from the energy levels of the standard source [12].

2.7. Determination of Activity Levels, Concentration Factor, and Annual Effective Dose

The activity concentrations of specific radionuclides (^{226}Ra, ^{228}Ra and ^{40}K) in the sediment, water, and fish muscle samples were calculated using the following analytical expression, as shown in Equation (1), which was also used in similar studies by other researchers, such as [8].

$$\mathrm{Asp} = \mathrm{N_{sam}}/\mathrm{PE}\varepsilon(\mathrm{E}) \quad (1)$$

where Asp is the specific activity concentration of the radionuclide in the sample, $\mathrm{N_{sam}}$ is the net counts of the radionuclide in the sample (sediment, water, and fish), PE is the gamma-ray emission probability, ε (E) is the absolute counting efficiency of the detector system, Tc is the sample counting time, and M is the mass/volume of the fish, sediment, or water sample in kilograms or litres. The equations used to calculate the radionuclide

transfer factor CF (BCF and BASAF), given by expression (2), have also been used by other researchers [14].

$$\mathrm{CF} = \frac{\text{Activity concentrations in biota concetrations } (\mathrm{Bqkg}-1)(\text{dry weight})}{\text{Activity concentrations of reference medium } (\mathrm{Bqkg}-1)} \quad (2)$$

The effective committed dose due to dietary intake is provided by the following Equation (3), adapted from ICRP reports [17] .

$$H_{T,r} = \sum \left(U^i \times \mathrm{C}_r^i \right) \times \mathrm{gT_r} \quad (3)$$

where i denotes the food group, U^i is the consumption rate per capital (kg/y), C_r^i the activity concentration of a radionuclide r of interest (Bq/kg), and $\mathrm{gT_r}$ is the dose conversion coefficient for the ingestion of the radionuclide r (Sv/Bq) in tissue. This is given as ^{226}Ra, ^{228}Ra, and ^{40}K, with conversion factors of 2.8×10^{-7}, 2.3×10^{-7}, and 6.2×10^{-9} in Sv/Bq, respectively. In order to obtain reliable data on the consumption rate in kilograms per person per year, we interviewed more than 100 people in the study area. The amounts that each adult consumes annually on average were 24.5 and 20.6 kg/p/y for Nile tilapia and Manyara tilapia, respectively.

3. Results

3.1. Activity Concentrations in Sediment

The activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K in the sediment samples were calculated using Equation (1), and are presented in Table 1. The mean activity concentrations of radionuclides in Lake Kindai were 79.95 ± 0.04 Bq/kg, 105.48 ± 0.06 Bq/kg, and 472.36 ± 0.31 Bq/kg for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively. The mean activity concentrations of radionuclides in Lake Munang were 87.96 ± 0.07 Bq/kg, 105.48 ± 0.06 Bq/kg, and 434.16 ± 0.24 Bq/kg for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively. The mean activity concentrations of radionuclides in sediment ponds were 44.1.96 ± 0.07 Bq/kg, 68.20.48 ± 0.10 Bq/kg, and 541.09 ± 1.04 Bq/kg for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively. The mean activity concentrations of radionuclides in Lake Singidani were 113.42 ± 0.10 Bq/kg, 116.74 ± 0.9 Bq/kg, and 574.91 ± 0.40 Bq/kg for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively.

Generally, radionuclides' activities in sediment varied as follows. ^{40}K had combined mean of 505.63 ± 21.79 Bq/kg, which ranged from (224.63–951.8) Bq/kg; this was followed by ^{228}Ra (98.91 ± 3.43 Bq/kg), which ranged from (29.35–134.19) Bq/kg. The lowest combined mean of radionuclide was observed in ^{226}Ra (113.42 ± 0.10 Bq/kg), which ranged from (22.57–143.54) Bq/kg. The concentration of the three radionuclides in the sediment samples varied from (22.57 to 143.54) Bq kg^{-1}, from (29.35 to 134.19) Bq/kg, and from (224.63 to 951.8) Bq kg^{-1} for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively, with average values of 88.05 ± 4.41 Bq/kg, 98.91 ± 3.43 Bq/kg, and 505.63 ± 21.79 Bq/kg for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively.

Table 1. Activity concentrations (Bq per kg) in sediment from sampled water bodies; values are expressed as mean ± SEM. ($p > 0.05$ and sample size $n = 20$).

Water Body	Sample ID	^{226}Ra	^{228}Ra	^{40}K
Lake Kindai	SK1	89.43 ± 8.32	133.52 ± 12.63	539.18 ± 50.17
	SK2	75.06 ± 7.53	95.44 ± 9.34	504.04 ± 50.30
	SK3	92.57 ± 8.86	108.94 ± 10.35	493.53 ± 47.43
	SK4	79.37 ± 7.85	112.96 ± 10.88	502.66 ± 49.91
	SK5	63.34 ± 6.00	75.25 ± 7.05	322.41 ± 33.35
Mean ± SEM		79.95 ± 0.04	105.22 ± 0.08	472.36 ± 0.31

Table 1. *Cont.*

Water Body	Sample ID	^{226}Ra	^{228}Ra	^{40}K
Lake Munang	SM1	95.67 ± 9.32	81.63 ± 8.01	393.16 ± 40.00
	SM2	135.55 ± 13.20	112.41 ± 11.50	432.44 ± 45.18
	SM3	114.83 ± 10.65	111.75 ± 10.37	432.54 ± 43.16
	SM4	137.34 ± 13.08	99.21 ± 9.89	372.15 ± 39.55
	SM5	88.41 ± 8.29	122.43 ± 11.52	540.53 ± 50.95
Mean ± SEM		87.96 ± 0.07	105.48 ± 0.06	434.16 ± 0.24
Ponds	SP1	36.28 ± 3.67	73.51 ± 8.27	947.06 ± 88.49
	SP2	76.12 ± 6.97	99.56 ± 9.39	273.68 ± 26.47
	SP3	56.65 ± 5.48	95.40 ± 9.17	411.46 ± 39.24
	SP4	28.77 ± 3.02	43.06 ± 4.49	225.76 ± 23.04
	SP5	22.68 ± 2.50	29.50 ± 3.63	847.49 ± 81.76
Mean ± SEM		44.1 ± 0.07	68.20 ± 0.10	541.09 ± 1.04
Lake Singidani	SS1	88.57 ± 8.54	99.68 ± 10.04	523.31 ± 50.98
	SS2	103.97 ± 9.69	122.98 ± 11.37	545.96 ± 53.15
	SS3	108.36 ± 10.21	126.75 ± 11.71	582.03 ± 55.84
	SS4	142.83 ± 13.60	122.72 ± 11.91	626.47 ± 62.18
	SS5	124.80 ± 11.94	111.23 ± 10.84	594.80 ± 59.36
Mean ± SEM		113.42 ± 0.10	116.74 ±0.09	574.91 ± 0.40
Overall mean ± SEM		88.05 ± 4.41	98.91 ± 3.43	505.63 ± 21.79
Overall min–max		(22.57–143.54)	(29.35–134.19)	(224.63–951.8)
World average (UNSCEAR)		35	30	400

3.2. Activity Concentrations in Water

The activity levels of ^{226}Ra, ^{228}Ra, and ^{40}K in the water samples were estimated using expression (1), and are presented in Table 2. The results show that ^{228}Ra was below the detection limit (BDL) in all samples from Lake Kindai, Lake Munang and Lake Singidani. Lake Kindai had a mean activity of 0.64 ± 0.018 Bq/L and 3.13 ± 0.87 Bq/L for ^{226}Ra and ^{40}K, respectively; for Lake Munang, both ^{226}Ra and ^{228}Ra were at BDL. However, the isotope ^{40}K was detected to have a mean radioactivity of 3.67 ± 1.41 Bq/L. The mean activity concentration of ^{226}Ra was noted to be 0.58 ± 0.20 Bq/L and ^{40}K (5.20 ± 0.531 Bq/L) in the ponds. All radionuclides were below detectable activity in Lake Singidani.

Table 2. Activity concentrations (Bq/L) from different sampled water bodies; values are expressed as mean ± SEM and sample size n = 20.

Water Body	Sample ID	^{226}Ra	^{228}Ra	^{40}K
Lake Kindai	WK1	BDL	BDL	2.60 ± 0.07
	WK2	BDL	BDL	BDL
	WK3	BDL	BDL	3.14 ± 0.09
	WK4	0.64 ± 0.01	BDL	6.19 ± 0.17
	WK5	BDL	BDL	2.97 ± 0.09
Mean ± SEM		0.64 ± 0.01	-	3.13 ± 0.87
Lake Munang	WM1	BDL	BDL	BDL
	WM2	BDL	BDL	BDL
	WM3	BDL	BDL	3.23 ± 2.20
	WM4	BDL	BDL	5.05 ± 2.74
	WM5	1.02 ± 0.02	BDL	BDL
Mean ± SEM		1.02± 0.02	-	3.67 ± 1.41

Table 2. *Cont.*

Water Body	Sample ID	^{226}Ra	^{228}Ra	^{40}K
Ponds	WP1	0.43 ± 0.01	BDL	5.40 ± 0.15
	WP2	BDL	BDL	3.18 ± 0.09
	WP3	0.43 ± 0.012	BDL	5.40 ± 0.15
	WP4	0.64 ± 0.02	BDL	5.71 ± 0.16
	WP5	1.34 ± 0.03	BDL	6.31 ± 0.18
Mean ± SEM		0.58 ± 0.20	-	5.20 ± 0.53
Lake Singidani	WS1	BDL	BDL	BDL
	WS2	BDL	BDL	BDL
	WS3	BDL	BDL	BDL
	WS4	BDL	BDL	BDL
	WS5	BDL	BDL	BDL
Mean ± SEM		-	-	-
Overall Mean ± SEM		0.31 ± 0.04	-	3.25 ± 0.30
Min–Max		BDL–1.41	BDL	BDL–8.3

3.3. Activity Concentrations in Fish

The radioactivity concentrations of fish samples were calculated using Equation (1), and are presented on Table 3, showing each fish type with their mean radioactivity concentration. Nile tilapia from Lake Kindai had activity levels of 11.23 ± 0.324 Bq/kg and 540.61 ± 15.606 Bq/kg for ^{226}Ra and ^{40}K, respectively. This level is higher compared to the Nile tilapia activity of Lake Singidani, which was 4.56 ± 1.33 Bq/kg and 5.98 ± 0.41 Bq/kg for ^{40}K and ^{226}Ra, respectively. Nile tilapia from ponds had combined mean activity levels of 8.51 ± 0.12 Bq/kg and 691.39 ± 3.72 Bq/kg for ^{226}Ra and ^{40}K, respectively, which are a little bit higher than all samples from Lake Kindai, Lake Munang, and Lake Singidani, which had radioactivity levels of 5.49 ± 0.96 Bq/kg and 637.51 ± 21.62 Bq/kg for ^{226}Ra and ^{40}K, respectively. Generally, the activity of Nile tilapia ranged from 0.71 Bq/kg to 14.70 Bq/kg for ^{226}Ra, and 479.37 Bq/kg to 845.79 Bq/kg for ^{40}K.

Manyara tilapia from Lake Munang had the highest activity concentrations of ^{226}Ra (7.61 ± 0.21 Bq/kg). However, Manyara tilapia samples from Lake Kindai showed the highest ability to uptake ^{40}K, with an activity of 381.81 ± 11.02 Bq/kg compared to the mean activity level of 352.23 ± 9.58 Bq/kg for ^{40}K.

Table 3. Activity concentrations (Bq/kg) in Nile tilapia and Manyara tilapia from sampled water bodies; values are expressed as mean ± SEM. ($p > 0.05$, and sample size $n = 11$).

Water Body	Type of Fish	Sample ID	^{226}Ra	^{228}Ra	^{40}K
Lake Kindai	Nile tilapia	TK	11.23 ± 0.32	BDL	540.61 ± 15.60
Lake Munang		TM	BDL	BDL	597.92 ± 17.26
Lake Singidani		TS	4.56 ± 1.33	BDL	504.60 ± 50.46
Ponds		TP1	BDL	BDL	693.87 ± 20.00
		TP2	5.65 ± 0.16	BDL	547.39 ± 15.80
		TP3	14 ± 0.40	BDL	805.51 ± 23.25
		TP4	BDL	BDL	745.17 ± 21.51
		TP5	5.89 ± 0.17	BDL	665.05 ± 19.19
Mean ± SEM			8.51 ± 0.12	-	691.39 ± 3.72
	Overall Mean ± SEM		5.49 ± 0.96	-	637.51 ± 21.62
	Min–Max		0.71–14.70	-	479.37–845.79

Table 3. *Cont.*

Water Body	Type of Fish	Sample ID	^{226}Ra	^{228}Ra	^{40}K
Lake Singidani	Manyara tilapia	TS0	4.97 ± 0.14	BDL	349.00 ± 10.07
Lake Kindai		TK0	5.38 ± 0.15	BDL	381.81 ± 11.02
Lake Munang		TM0	7.61 ± 0.21	BDL	325.88 ± 9.40
	Overall Mean ± SEM		5.98 ± 0.41	-	352.23 ± 9.58
	Min–Max		4.72–7.99	-	309.90–352.23

3.4. Activity Concentrations (Bq/kg) Per Location

The mean activity levels of each sample location are presented in Figure 2, whereby ^{40}K had the highest concentrations for all locations. Other radionuclides of ^{226}Ra and ^{228}Ra had high activity concentrations in Lake Singidani, Lake Kindai, and Munang, compared to the ponds. Lake Munang had a high level of ^{226}Ra compared to the ponds, and Lake Singidani shows almost the same level of ^{226}Ra and ^{228}Ra.

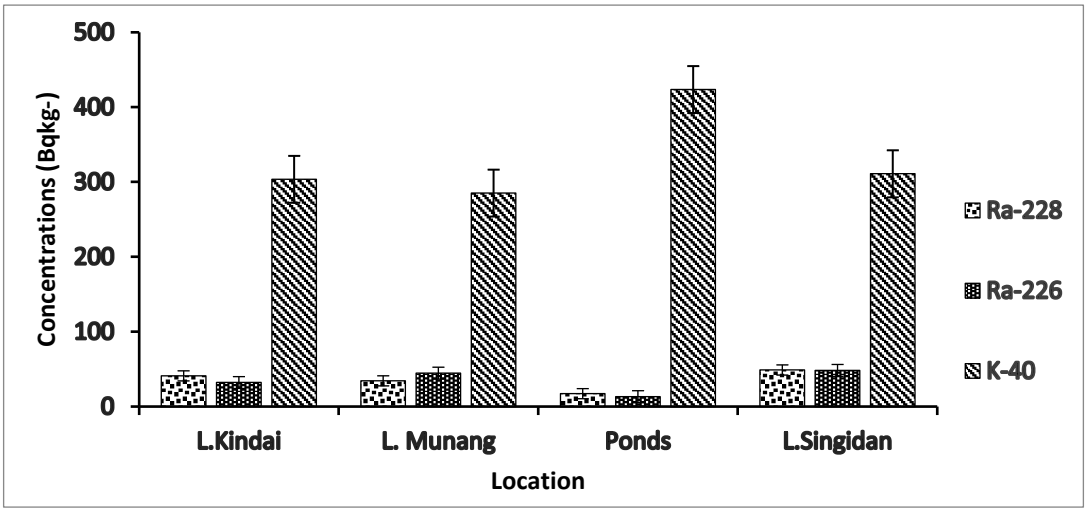

Figure 2. Radioactivity concentrations (Bq/kg) per location.

3.5. Radionuclide Transfer from the Fish Environment to Fish

Radionuclides may be transferred between biota and reference media (water and sediment) in fresh water ecosystems. The accumulation of radionuclides in biota can be represented in simplified ratio by relating the radionuclides' concentration in biota to the radionuclides' concentration in water and sediment. The radionuclides (^{226}Ra, ^{228}Ra, and ^{40}K) bioconcentration factors (BCFs) and biota sediment accumulation factors (BSAF) were calculated using Equation (3), and are presented in Table 4.

Table 4. Radionuclide transfer factor from water to fish (BCF) and from sediment to fish (BSAF), for each type of fish.

	Transfer of Radionuclides from Water to Fish			Transfer of Radionuclides Sediment to Fish		
Sample	^{226}Ra	^{228}Ra	^{40}K	^{226}Ra	^{228}Ra	^{40} K
Manyara tilapia	19.46	3.09	108.39	0.03	0.01	0.70
Nile tilapia	17.87	4.57	196.17	0.06	0.02	1.26

The results show that the transfer of radionuclides from water to fish was higher compared to the radionuclide transfer from sediment to fish. Thus, the results of this equation reveal that Nile tilapia have high levels of radionuclides compared to Manyara tilapia.

The dissolved and particulate phases have also been shown to be in a steady state, with exchanges of nuclides between particles and water being wholly reversible. The BCF of ^{226}Ra (19.46 and 17.87) is higher than that of ^{228}Ra (3.09 and 4.57). Additionally, the ^{40}K transfer factor (108.39 and 196.17) from water to fish was higher five times compared to those of ^{226} Ra (19.46 and1.7.87) and ^{228}Ra (3.09 and 4.57) for Manyara tilapia and Nile tilapia, respectively. However, in gauging the biota sediment accumulation factor of ^{226}Ra, ^{228}Ra, and ^{40}K, only ^{40}K was bio-accumulated, with 1.26 BSAF; the other radionuclides (^{226}Ra, ^{228}Ra) had values less than 1 BSAF, indicating that they were not bio-accumulated.

3.6. Annual Effective Dose (AED)

The radioactivity of fish types obtained from the results was used to estimate the committed dose, which is also the annual effective dose of the population in this study area. The total dose ingested through the consumption of fish can be calculated by summing the doses derived for each radionuclide (^{226}Ra ^{228}Ra and ^{40}K). The annual effective doses due to consumption of contaminated fish were calculated using Equation (3), and are presented in Table 5. The results of this calculation show that Nile tilapia and Manyara tilapia had an AED of 0.009 and 0.005 mSv/y, respectively. The average mean for both Nile tilapia and Manyara tilapia was 0.0074; this value is also less than the international recommended limit of 1 mSv/y.

Table 5. Annual effective dose compared to recommended limits (mSv/y).

Fish Type	AED
Nile tilapia	0.0090
Manyara tilapia	0.0050
Average mean	0.0074
Recommended limits [18,19]	1.0000

4. Discussion

4.1. Activity Concentrations in Sediment

The sediment samples had the lowest mean activity levels of ^{226}Ra compared to the other two radionuclides in this study. This may be attributed to the higher solubility of ^{226}Ra compared with ^{228}Ra and ^{40}K. These properties makes ^{226}Ra less available in sediment compared to ^{228}Ra [19,20]. The radioactivity of ^{40}K dominated over the other isotopes, possibly because it is the most abundant in continental rocks and it is elevated in many light minerals. The radioactivity concentration of three isotopes ranged from (22.57–143.54) Bq/kg for^{226}Ra, 29.35–134.19 Bq/kg for ^{228}Ra, and (224.63–951.80) Bq/kg for ^{40}K; only in ^{40}K are these values are little bit higher than the world average, as indicated in Table 1. These values are due to the geological nature of granite rock and the anthropogenic activities nearby these water bodies. However, a value being higher than world average may not mean it causes direct radiological hazard; this depends on the use of sediment. The sediment in this study area does not directly interact with humans.

A comparison of mean sediment activities from this study and the study conducted by [14] revealed that the Malaysian sediment's mean radioactivity for ^{228}Ra is 249.8 $\pm$ 19 Bq/kg, which is almost twice as high as the value found in the current study ^{228}Ra (98.91 $\pm$ 3.43 Bq/kg). The mean activity concentration of sediment in the present study is almost four times less when compared to that from Malaysia with regard to ^{226} Ra (397.9 $\pm$ 42.1 Bq/kg). At the same time, the mean radioactivity concentrations of ^{40}K (505.63 $\pm$ 21.7 Bq/kg) were three times less than those from Malaysia in ^{40}K (1782 $\pm$ 201 Bq/kg). The world average concentrations are 35 Bq/kg, 30 Bq/kg, and 400 Bq/kg for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively. The results of the current study are higher, creating baseline data for future references. Thus, in general, the averages and ranges of the activity concentration of ^{226}Ra, ^{228}Ra, and ^{40}K in these sediments are higher than the world figures reported in [21].

4.2. Activity Concentrations in Water

Radionuclide levels in water were generally low, reflecting the strong chemical bonding properties of the sediments. The isotope ^{228}Ra is insoluble in water, hence it is difficult to dissociate it from granite rock and make it available in water, compared to ^{226}Ra. The isotope of ^{226}Ra, which is more soluble in water, tends to dissolve from granite rock and form leachate solutions; it is more readily available in water compared to ^{228}Ra [13]. The isotopes of ^{228}Ra in water samples were below the detection limit simply because ^{228}Ra is essentially insoluble in water compared to ^{226}Ra. Additionally, ^{40}K is known to be more soluble in water than ^{226}Ra and ^{228}Ra [22]. The higher P^H levels in Lake Singidani favoured ^{226}Ra forming complex carbonates and thus being unavailable in water [11]. The mean activity concentrations of ^{226}Ra and ^{228}Ra in water were below the guideline levels for radionuclides in drinking water (1.0 Bq/L), as recommended by the World Health Organisation.

The mean radioactivity level of ^{40}K (3.29 $\pm$ 1.20 Bq/L) in this study at Lake Singidani ranged from (1–5) Bq/L, while a study at Bahi swamp (which is found in the same geological area of central Tanzania) found a level that was a little bit higher (3.13 $\pm$ 0.87 Bq/L), ranging from (2.6–6.19) Bq/L [8]. This variation may depend on the underlying type of granite rock and the anthropogenic activities around these water bodies.

4.3. Activity Concentrations in Fish

The result showed that Nile tilapia from Lake Kindai had high radioactivity levels compared to ones from Lake Singidani. This is possibly because of different levels in alkalinity, where Lake Singidani is said to be more alkaline than Kindai [11]. It is also reported that low alkalinity favours the migration or dissolution of ^{226}Ra, making it available in water, and then available for fish. These results indicate that Manyara tilapia exceed Nile tilapia by 0.57 Bq/kg. The concentrations of ^{226}Ra Nile tilapia from the current study have a high mean level of ^{40}K concentration (637.51 $\pm$ 21.62 Bq/kg), while Manyara tilapia had a mean ^{40}K concentration of 352.23 $\pm$ 9.58 Bq/kg. The activity concentration of ^{40}K in Manyara tilapia (352.23 $\pm$ 9.58 Bq/kg) is almost half the ^{40}K activity concentration of Nile tilapia (637.51 $\pm$ 21.62 Bq/kg). This study was challenged by the limited number of similar studies available for comparing their results with the present results. However, [12] carried out a related study on whole Nile tilapia, and the results were ^{226}Ra (25.6 $\pm$ 7.4 Bq/kg), ^{228}Ra (52.4 $\pm$ 28.7 Bq/kg), and ^{40}K (462 $\pm$ 80 Bq/kg). These differences may vary due to the differences in underlying rocks and the nature of the water bodies. The AED for this study was 0.00973 mSv/y, which is three times higher than the results obtained by [16] from Nigeria, although both were below the limit of 1 mSv/y proposed by the ICRP.

4.4. Comparisons of Activity Concentrations Levels in Different Water Bodies

Higher ^{40}K concentrations in ponds may be attributed to many factors; these may include the use of phosphate-containing fertilisers, which are known to have high radioactivity levels [23]. The concentrations of ^{226}Ra and ^{228}Ra in Singidani Lake had high radioactivity levels compared to other water bodies. These levels may be attributed to factors such as geological nature, and the depth of water bodies being closer to the earth's surface. The activity level of ^{226}Ra in the water of Lake Kindai is a little bit lower than the activity level of ^{226}Ra in the water of Lake Munang. This may be because Lake Munang receives more water from nearby upstream ponds in which the water activity level of ^{226}Ra is higher, per this study.

4.5. Radionuclides Transfer from Fish Environment to Fish

The radioactivity of ^{226}Ra in Manyara tilapia (whole fish) is a little bit higher than that in Nile tilapia, showing a difference of 0.57 Bq/kg. This difference may be due to ^{226}Ra deposited in the Manyara tilapia bones, which increased the activity levels compared to Nile tilapia samples (which were analysed without bones). Thus, it may be safer to eat fish fillets rather than whole fish with bones in case of contamination, because radium, like other alkaline earth metals, tends to be deposited in bones. The radioactivity level of ^{228}Ra

was not detected in fish compared to ^{226}Ra, probably because ^{228}Ra was below the BDL in water; thus, it could also not be detected in fish. ^{226}Ra was available in water compared to ^{228}Ra; thus, it was more easily ingested by fish during food ingestion and gaseous exchange. The radioactivity concentrations of ^{40}K in fish were higher in all samples analysed in this study. Bio-accumulations of ^{40}K in Manyara tilapia may be influenced by different factors such as fish species characteristics, the physical chemical properties of the radionuclides, and the fish food chain aquatic environment [14].

4.6. Annual Effective Dose (AED)

The AED values of the two varieties of fish are approximately 100 times less when compared to the maximum AED limit (1 mSv/y) recommended by the International Commission on Radiological Protection (ICRP). Additionally, they are three times higher compared to those in a study conducted by [16] in Nigeria. The differences in radioactivity levels in this study and that of [16] may be attributed to many factors, like the geological nature of underlying rocks found in lakes. These results imply that fish consumers are radiologically safe; however, the consumption of fish bones will increase their level of radioactivity, especially their level of ^{226}Ra.

5. Conclusion and Recommendations

All samples analysed in this study had activity concentrations that followed following trend: Sediment > fish > water. All radionuclides' activity followed the trend ^{40}K > ^{228}Ra > ^{226}Ra. The only bio-accumulated radioisotope in fish (Nile tilapia) was ^{40}K, which is among the elements essential for human health. The movement of radionuclides from water to fish was higher compared to the movement of radionuclides from sediment to fish. Additionally, all sampling site activity levels followed the trend of Lake Singidani > Lake Kindai > Lake Munang > ponds. Thus, the reverse trend in the fish supply of these areas may lower the radiological risk of dose exposure. The annual effective doses of Nile tilapia and Manyara tilapia were 0.00937 and 0.005 mSv/y, respectively, which are much below the 1 mSv/y recommendation of the ICRP. Therefore, this study has shown that the current level of radioactivity in fish consumed in this study area does not pose a significant radiological risk to fish consumers. The study recommends that research be extended to other species of fish.

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