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Research Article

Assessment of Radioactivity Levels and Radiological Hazards Indices in Sediments and Fish from Coastal Marine Areas of Tanzania

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Abstract

Environmental radioactivity monitoring is essential for assessing potential exposure risks to humans and marine ecosystem, particularly in coastal marine areas where anthropogenic and natural radionuclides may accumulate. However, research studies on natural radioactivity in Tanzania's coastal marine areas remain scarce. This study was therefore conducted to assess the radioactivity levels of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in marine sediments and fish samples from the Tanzanian coastal areas, and to estimate the associated radiological health hazards for both humans and marine ecosystem. A total of 49 samples comprising both sediment and fish were randomly collected from 12 coastal sites and composited for analysis. Radioactivity concentrations were measured using gamma spectrometry. For fish samples, the mean activity concentrations were 4.9 ± 1.9 Bq kg^{-1} dry weight (dw) for ^{226}Ra, 488.8 ± 34.1 Bq kg^{-1} dw for ^{40}K, while ^{232}Th was below the detection limit. In sediments, the corresponding mean values were 15.1 ± 2.2, 14.9 ± 2.0, and 320.8 ± 27.4 Bq kg^{-1} dw for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Based on these data, a comprehensive set of radiological hazard indices was computed, including radium equivalent activity (Ra_eq), absorbed dose rate (D), annual effective dose equivalent (E_eff), external and internal hazard indices (H_{ex} and H_{in}), representative gamma index (I_γ), annual gonadal dose equivalent (AGDE), excess lifetime cancer risk (ELCR), and annual effective dose (AED). The results showed that the computed total annual effective committed dose for fish consumption was below the 1 mSv y^{-1} as recommended by the ICRP, although the mean ^{40}K activity in fish exceeded the global average of 420 Bq kg^{-1}. All evaluated hazard indices (Ra_{eq}, H_{ex}, D, E_{eff}, I_γ, AGDE, and ELCR) for sediments, as well as H_{in} and ELCR for fish, were within permissible levels values. In conclusion, the current radioactivity levels in the studied coastal area do not pose a significant radiological risk to human health or the marine ecosystem. Nevertheless, it is advisable to keep monitoring for purpose of detecting any future changes due to natural or anthropogenic influences.

Keywords

Natural Radioactivity, Gamma Spectrometry, Fish, Marine Sediments, Radiological Hazards Indices, 226Ra, 40K, Tanzania Coastal Areas

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1. Introduction

Coastal areas are in many cases considered highly favorable for various socio-economic activities including manufacturing, mining, agriculture, oil and gas exploration, tourism and fisheries [1, 2]. It is for this reason coastal areas are generally highly populated. Because of the large population coupled with different economic activities, these areas are prone to environmental pollution caused by organic, chemicals and mineral contaminants produced from the economic activities [3]. Among the chemical contaminants of concern are the radionuclides. These are elements that reside in different environments including soil, underground water, marine sediment, sand beach and biota (both flora and fauna).

Although radionuclides are naturally present in the environment at low concentrations, their concentration may be significantly escalated by the release of radioactive wastes from different economic activities [4, 5]. The released radionuclides may enters the marine environment directly or indirectly through different pathways, including rivers and rainwater which discharge the waste into the sea. It follows that, radionuclides in the coastal areas may be classified as primordial, cosmogenic and anthropogenic radionuclides. Anthropogenic radionuclides mostly originate from socio-economical activities such as gas and oil production, fertilizer production, mineral exploration and mining [6, 7]. Anthropogenic radionuclides may also originate from activities like nuclear tests and operation of nuclear reactors. Nuclear reactor accidents, such as those of Chernobyl (1986) and Fukushima (2011) that happened in the past are also a considerable source of anthropogenic radionuclides [8].

The presence of radionuclides, irrespective of their category, in the environment may lead to radiation exposure to both human beings and animals [4, 9]. According to UNSCEAR (United Nations Scientific Committee on the Effect of Atomic Radiation) [10], natural radionuclides contribute the largest share of radiation exposure to the world population, giving an average annual effective dose of about 2.4 mSv. Of this, decay series radionuclides (mainly from the Uranium (^{238}U) and Thorium (^{232}Th) series, including radon and its progeny) contribute roughly 70-80% of the total dose, while primordial radionuclides such as Potassium (^{40}K) account for about 15-20%. Continuous exposure to ionizing radiation may pose serious harmful biological effects such as DNA (deoxyribonucleic acid) damage or cancer [11, 12]. Hence, there is need to monitor and control the concentrations of radionuclides in the environment in order to maintain the safety of populations.

As coastal areas host large populations, it is important to have a clear knowledge of the concentrations and distribution of radionuclides in the seafood, soil, water, air and sediments [6, 13]. Although the distribution and level of natural radionuclides has been studied widely over the world, a few studies are available in sub-Saharan Africa focusing heavily on countries bordering the Mediterranean and Red seas [14-16]. In Tanzania no data on radionuclide pollution have been published along the coastal areas except some studies that have been reported on radioactivity focus on soils, surface water, fish and foodstuff around mining and phosphate industries [17-21]. This lack of data hinders the development of an effective regulatory framework for radiation protection in this study area.

Tanzania's coastal area has recently been experiencing rapid changes due to urbanization and industrialization in such way there is a significant increase of buildings and road construction as well as the use of fertilizer and pesticides in farming [22, 24]. Also, activities like mining, gas and oil exploration and production have all increased significantly. Despite the economic benefits of these activities, it is envisaged that the generation of solid wastes in coastal areas will reach about 26 million tons by the end of 2030 [23]. This implies that there some possibility of elevation of radioactivity concentrations in the marine environment, which may lead to detrimental effect to the coastal habitats. Therefore, monitoring the radioactivity pollution in these areas is of paramount importance in order to ensure the population safety. Hence, this study is set to assess the radioactivity levels and the associated radiological risks in Tanzania's coastal marine environment and then estimating the annual effective dose due to consumption of fish. Understanding the radiological risk associated with anthropogenic activities and annual effective dose due to consumption of fish is essential for improving safety measures and environmental practices. Also the result of this study will help the regulatory bodies in formulating radiation protection policies and determine the suitability of using sediments material for various applications.

2. Materials and Methods

2.1. Study Area

Tanzania coastal area covers a distance of over 800 km stretching from latitude 4° 49' S to latitude 10° 28' S and respective longitudes 38° and 42° E as shown Figure 1. The study area is characterized with primarily of Mesozoic to Cenozoic sedimentary rocks basins (sandstone, shale limestone). The sedimentary rocks, vary in age from Jurassic, cretaceous to tertiary and quaternary. Also, the study area is featured shallow, unconfined, and semi-confined alluvial and limestone aquifer with vulnerable to saltwater intrusion and major river deltas. The drainage pattern is of perennial and seasonal rivers draining into the Indian Ocean basin. The major rivers including Pangani, Wami, Ruvu, Rufiji and Ruvuma. The other small rivers are Matandu, Mbwemkuru, Lukuledi, and Msimbazi [25]. Other features of the study area include estuaries, mangrove forests, coral reefs, sandy beaches, cliffs, seagrass beds and muddy tidal flats. The study area is also characterized by three big ports which are Dar es Salaam port, Tanga

port and Mtwara port as displayed in Figure 1. This area was of interest to this study because of the activities in the region. For instance along rivers Wami, Ruvu and Pangani there are a big farms of rice, maize and other crops. Meanwhile, natural gas extraction is taking place at Songosongo Island in Lindi region, as well as Mnazi Bay and Madimba in Mtwara region. These activities produce wastes that are discharged to marine environments through rivers such as Matandu, Mbwemkuru, Lukuledi and Ruvuma. Besides, all industries operating in Dar es Salaam, Tanga, Lindi and Mtwara are discharge their waste to marine areas through either of the mentioned rivers.

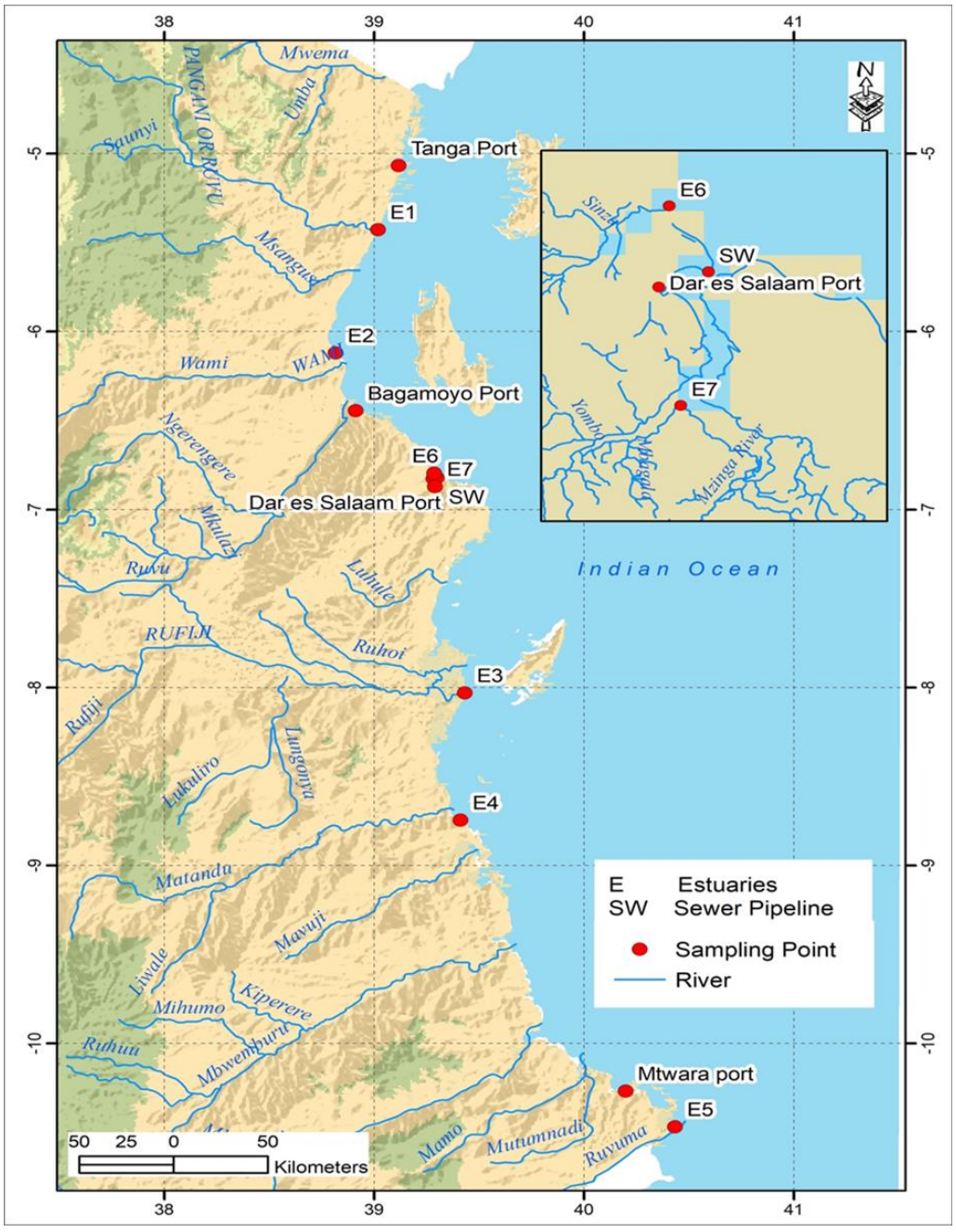

Figure 1. *Map of the Coastal area of Tanzania showing the sampling Stations (Source: IRA GIS Lab).*

2.2. Sample Collection and Preparation

A total of 49 samples were first randomized to minimize impartiality and then composited to reduce objectivity and then manageable samples were collected under the study area. Out of these, 12 fish samples were randomly collected from fishermen in the fishing areas around ferries at Dar es Salaam, Tanga and Mtwara. Table 1 shows the fish species names and their sources.

Table 1. The sampled fish species and their source.

S/No.	Species Name	Common name	Local name	Sample Code	Source
1	Scarus Sordidus	Parrot fish	Pono	F 001	Marine water
				FT 002	Marine water
				FM 002	Marine water
2	Epinephelus Caeruleopunctatus	Grouper fish	Chewa	F 002	Marine water
				FT 001	Marine water
				FM 003	Marine water
3	Panaeus Monodon	Prawns	Kamba Mti	F 003	Marine water
				FT 003	Marine water
				FM 001	Marine water
4	Caranx Melampygus	Jack Fish	Kolekole	F 004	Marine water
				FT 004	Marine water
				FM 004	Marine water

Meanwhile, 37 sediment samples were randomly collected from estuaries, sewage pipelines, areas around ports and ocean during low tide. These samples were collected along the coastline at points separated by a distance of 200 m. Each sample was collected at a depth of 5 - 10 cm using open end stainless-steel hand-held Auger corer with a 70 mm diameter. Table 2 shows the sediment samples with their identity and locations. One should note that, in some areas samples were not collected due to several challenges at the sites.

Table 2. Samples Identity and locations in Estuaries, Sewage discharge and Port areas along the Coastal Marine areas of Tanzania.

S/No.	Sample Code	Type of Sample	Location
1	F 001	Fish	Dar es Salaam Ferry
2	F 002	Fish	Dar es Salaam Ferry
3	F 003	Fish	Dar es Salaam Ferry
4	F 004	Fish	Dar es Salaam Ferry
5	FT 001	Fish	Deep Sea Tanga Ferry
6	FT 002	Fish	Deep Sea Tanga Ferry
7	FT 003	Fish	Deep Sea Tanga Ferry
8	FT 004	Fish	Deep Sea Tanga Ferry
9	FM 001	Fish	Mtwara Ferry
10	FM 002	Fish	Mtwara Ferry
11	FM 003	Fish	Mtwara Ferry
12	FM 004	Fish	Mtwara Ferry
13	E6 001	Sediments	Upper Msimbazi River
14	E6 002	Sediments	Down Msimbazi River
15	E6 003	Sediments	Midpoint Msimbazi & Ocean

S/No.	Sample Code	Type of Sample	Location
16	E6 004	Sediments	Upper Msimbazi River
17	E6 005	Sediments	Along the Ocean
18	E7 001	Sediments	Midpoint Mzinga & Kizinga
19	E7 002	Sediments	Upper Mzinga river
20	E7 003	Sediments	Upper Kizinga river
21	SWP 001	Sediments	Sewage discharge point Dar
22	SWP 002	Sediments	Sewage discharge point Dar
23	SWP 003	Sediments	Sewage discharge point Dar
24	SWP 004	Sediments	Sewage discharge point Dar
25	DP 001	Sediments	Dar port near Ferry
26	DP 002	Sediments	Dar port near Ferry
27	DP 003	Sediments	Dar point near Ferry
28	E1 001	Sediments	Midpoint Pangani & Ocean
29	E1 002	Sediments	Down Pangani river
30	E1 003	Sediments	Down Pangani river
31	E1 004	Sediments	Upper Pangani river
32	SWPT 001	Sediments	Sewage discharge point Tanga
33	SWPT 002	Sediments	Sewage discharge point Tanga
34	SWPT 003	Sediments	Sewage discharge point Tanga
35	SWPT 004	Sediments	Sewage discharge point Tanga
36	SWPT 005	Sediments	Sewage discharge point Tanga
37	TP 001	Sediments	Baggage point at Tanga port
38	TP 002	Sediments	Dredging area at Tanga port
39	TP 003	Sediments	Dredging area at Tanga port
40	TP 004	Sediments	Dredging area at Tanga port
41	TP 005	Sediments	Dredging area at Tanga port
42	E4 001	Sediments	Upper Matandu river
43	E4 002	Sediments	Down Matandu river
44	E4 003	Sediments	Midpoint Matandu & Ocean
45	E4 004	Sediments	Upper Matandu river
46	MP 001	Sediments	Mtwara Ferry near port
47	MP 002	Sediments	Mtwara Ferry near port
48	MP 003	Sediments	Mtwara Ferry near port
49	MP 004	Sediments	Mtwara Ferry at cargo point

All these samples were packed in air tight polyethylene bags, stored in ice box and transported to TAEC (Tanzania Atomic Energy Commission) laboratories for preparation and analysis. The sediments samples were air dried under shade for about seven hours then dried in an automatic oven for about three days at a temperature 80°C until constant weight was reached, in order to evaporate water until only the dried

sediment remained. Then, the samples were ground with machine and cleaned with acetone to avoid contamination; then sieved to powder of 500 µm grain size. The samples were then weighed and packed in well labelled and air tight canisters of a specific geometry (100mm diameter and 40 mm in height) similar to that of the calibration source provided by IAEA (International Atomic Energy Agency). All the canisters containing samples were then stored for 30 days for uranium and thorium radionuclide daughters to attain secular equilibrium between short- lived and respective long-lived (^{226}Ra from ^{238}U) and (^{228}Ra from ^{232}Th) before gamma spectrometric analysis [26]. The guidelines regarding the measurement of radionuclides in food and the environment were observed throughout, as guided by IAEA [27].

In the case of fish samples, the collected fishes were of approximately the same size and identified with the help of ferry fishery officers and fishermen. First, fish samples were melted naturally at room temperature, then rinsed using deionized water. Afterwards, each fish was dissected to remove edible tissues. The tissues were air-dried and then oven-dried at a temperature of 80°C for about three days until they reached constant weight (to ensure that they were completely dried). The dry samples were now ground using an agate mortar before being sieved to powder of 500 µm grain size mesh to obtain homogeneous fish powder [26]. Thereafter, the samples were weighed and packed in clean 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 30 days so that uranium and thorium radionuclide daughters could attain secular equilibrium between short-lived progeny and the respective long-lived (^{226}Ra from ^{238}U) and (^{228}Ra from ^{232}Th) before gamma spectrometric analysis [27]. After 30 days, all the samples were analyzed to determine the activity concentrations of radionuclides using the HPGe (High - Purity Germanium) gamma spectrometry as well.

2.3. Sample Analysis

After 30 days, all the samples were analyzed to determine the activity concentrations of the present radionuclides. This was accomplished by using p-type coaxial high-purity germanium detector (ORTEC®GEM40-83SMP) inside lead shielding and connected to a multichannel analyzer. The detector, with 40% relative efficiency and specific energy resolutions, was calibrated with a CBSS2 multi-nuclide standard source which containing ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, and ^{51}Cr. The data acquisition and analysis were performed using an ORTEC®DSPEC-LF digital signal processor. The Gamma Vision® software was used in spectrum analysis [28].

The specific activity concentration of ^{226}Ra radionuclide in sample was estimated using the 351.9 keV photo peak from the decay of ^{214}Pb as well as the 609.3 keV, 1120.3 keV, 1728.6 keV and 1764 keV photo peaks from the decay of ^{214}Bi. Likewise, the activity concentration of ^{232}Th radionuclide was estimated using the 911.2 keV photo peak from the decay of ^{228}Ac and the 583.1.6 keV photo peak from the decay of ^{208}Tl. Finally, the activity concentration of ^{40}K was determined using the 1460.8 keV photo peak. The actual time used for measurement of every sample was 172800 seconds. Therefore, activity concentration for each radionuclide of interest was computed using equation (1).

Data analysis was performed using gamma vision software and Origin Pro 2018 64-bit software through descriptive approach whereby means, standard deviation and charts/graphs were used in describing the data pattern to meet the objective of the study. Further, inferential analysis was done using an independent sample t-test in comparing means at the significance level of 0.05.

3. Analytical Methods

3.1. Activity Concentration and Uncertainty of the Activity

The activity concentrations (A_s) of the natural radionuclides of interest in this work were computed using equation (1) [29].

$$A_s\ (Bqkg^{-1}) = C_n / \varepsilon P_\gamma M_s \quad (1)$$

where: C_n is the net counting rate under the corresponding peak, ε is the detector efficiency at the specific gamma-ray energy, P_γ is the absolute transition probability of the specific gamma-ray and M_s is the mass of the sample (kg). The obtained values of activity concentrations were then used to compute other analytical parameters as discussed here.

3.2. Radium Equivalent Activity (Raeq)

Radium equivalent activity (Ra_{eq}) is the most widely used for radiation hazard index and common used to assess the consistency of radiation exposure. Ra_{eq} is a weighted sum of activities of the three natural radionuclides in fish and sediment based on the estimation that 370 Bqkg^{-1} of ^{226}Ra, 259 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K will produce the gamma-ray dose rate. Ra_{eq} was computed using equation (2) [30].

$$Ra_{eq}\ (Bqkg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where: A_{Ra}, A_{Th} and A_k are activity concentrations (Bqkg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K respectively.

3.3. Absorbed Gamma Dose Rates

The absorbed gamma dose rate (D) is the index used to describe the intensity of the energy deposited in a small amount of a tissue anywhere in the body and is used to assess the potential damage to a particular organ, tissue or material. The

absorbed gamma dose rates due to gamma radiations in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) was calculated using equation (3) based on guidelines provided by UNSCEAR for fish and sediments samples [4]. The conversion factors used to compute the absorbed gamma dose rate in air per unit activity concentration in Bq/kg (dry weight) corresponds to 0.427nGy/h for ^{226}Ra, 0.623nGy/h for ^{232}Th and 0.043 nGy/h for ^{40}K.

$$D\ (nGyh^{-1}) = 0.427A_{Ra} + 0.623A_{Th} + 0.043A_k \quad (3)$$

where: A_{Ra}, A_{Th} and A_k are activity concentrations ($Bqkg^{-1}$) of ^{226}Ra, ^{232}Th and ^{40}K respectively.

3.4. External and Internal Hazard Index

The external hazard index was calculated from Ra_{eq} expression through assumption that its maximum allowed value (equal to unity) matches the upper limit of Ra_{eq} (370 $Bqkg^{-1}$) for safe use. The external hazard index (H_{ex}) is used to indicate the significance of external radiation exposure while internal hazard index (H_{in}) is used to assess the hazardous effects to the human respiratory organ through inhalation process. So both index are used to predict the effect of radiation on human health and should be less than unity for the purpose of keeping the radiation hazard insignificant. In this work, the external hazard index (H_{ex}) was computed using Equation (4) while the internal hazard index (H_{in}) was computed using equation (5) [31].

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad (4)$$

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \quad (5)$$

where: A_{Ra}, A_{Th} and A_k are activity concentrations in ($Bqkg^{-1}$) of ^{226}Ra, ^{232}Th and ^{40}K respectively.

3.5. Representative Gamma Level Index

Representative gamma level index (I_γ) was used to examine the level of gamma radiation hazard associated with the characteristics radionuclides in the sediment samples collected in the coastal marine areas of Tanzania. Addition, gamma level index normally help to indicate the radiological safety of the building materials and must be equal or less than unity for low to negligible level of radiation exposure [32]. This index was computed using equation (6) [33].

$$I_\gamma = A_{Ra}/150 + A_{Th}/100 + A_K/1500 \quad (6)$$

where: A_{Ra}, A_{Th} and A_k are activity concentrations ($Bqkg^{-1}$) of ^{226}Ra, ^{232}Th and ^{40}K respectively.

3.6. Annual Gonadal Dose Equivalent

The AGDE (Annual gonadal dose equivalent) is the radiation index used to assess potential radiation effect to gonads since are more relative sensitive to ionizing radiation when you compare to other organs of the body and hence they are considered to be at a high risk of radiation exposure [34]. The AGDE was computed using equation (7) to estimate the potential radiation dose that gonads may receive from ^{226}R, ^{232}Th and ^{40}K [7, 35].

$$AGDE\ (nGyh^{-1}) = 0.3.09A_{Ra} + 4.18A_{Th} + 0.314A_k \quad (7)$$

where: A_{Ra}, A_{Th} and A_k are activity concentrations in $Bqkg^{-1}$ of ^{226}Ra, ^{232}Th and ^{40}K respectively.

3.7. Annual Effective Dose Equivalent

The annual effective dose equivalent (E_{eff}) is another parameter which define the radiation hazards. The E_{eff} was computed from absorbed dose by applying the dose conversion factor of $0.7SvGy^{-1}$ with the outdoor and indoor occupancy factors of 0.2 and 0.8 respectively. The E_{eff}, was computed using equation (8) [31], and the limit of 1mSv/y is considered acceptable for the general public [4].

$$E_{eff}\ (uSvy^{-1}) = D\ (nGyh^{-1}) \times 24h \times 365.25\ days \times 0.2 \times 0.7\ Sv\ Gyy^{-1} \times 10^{-3} \quad (8)$$

3.8. Excess Lifetime Cancer Risk

The ELCR (Excess lifetime cancer risk) is another parameter which describes the effect of radiation exposure. The ELCR can be defined as the probability of developing stochastic effect at a lifetime due to low doses of ionizing radiation to human being over a given period [4]. The ELCR in this work was calculated using equation (9) [36].

$$ELCR = E_{eff}\ X\ DL\ X\ RF \quad (9)$$

where: E_{eff} is the annual effective dose equivalent, DL represents the average duration of life (estimated to be 70 years) and RF is a risk factor (Sv^{-1}). The risk factor can be defined as the fatal cancer risk per Sievert. For stochastic effects, the value for RF = 0.05 Sv^{-1} for public members [4].

3.9. Dose Assessment

The radioactivity of fish obtained from the study was used to estimate the total annual effective committed dose (H_T) of the population in the study area. The total dose ingested through the consumption of fish can be calculated by summing the doses derived from each radionuclides (^{226}Ra, ^{232}Th, and ^{40}K). However, ^{40}K is an essential biological element distributed throughout the body and its concentration in human tissue are under metabolic (homeostatic) control. In estimating the total annual effective committed dose we used the annual fish consumption rate value that is 8.5 kg/y reported in 2020

[37, 38]. Therefore, the total annual effective committed dose was computed using equation (10) adopted from ICRP (International Commission on Radiological Protection) report [39].

$$H_{T,r} = \sum (U^i \times C^i_r) \times gT_r \quad (10)$$

Where; i denotes the food group, U^i is the consumption rate per capital (kg/y), C^i_r the activity concentration of a radionuclides r of interest (Bq/kg), and gT_r is the dose conversion coefficient for the ingestion of radionuclide r (Sv/Bq) in tissue. This is given as ^{226}Ra, ^{232}Th, and ^{40}K, with conversion factors of 2.8 x 10^{-7}, 2.3 x 10^{-7}, and 6.2 x 10-9 in Sv / Bq respectively [39].

4. Results and Discussion

4.1. Radioactivity Levels in Fish

The analysis of radionuclides concentrations in fish samples yielded the results presented in Table 3. These results shows that the activity concentrations of ^{40}K in almost all fish samples were higher than the world average value [4]. Meanwhile, the activity concentrations of ^{226}Ra were lower than the world average values. Besides, it was observed that the activity concentrations of ^{232}Th in all samples were BDL (below detection limit) of the gamma spectrometry system used for sample analysis. From the result it shows that the species with code F002, FT001 and FM 003 accumulate more ^{226}Ra while species with code FT001 accumulate more ^{40}K. This may be due to the different feeding habits of these species, geographical location and conditions as well as solubility of ^{40}K in water [4]. This observation is supported by literature like a study by Khan *et al.* [40], which reported that radionuclide accumulation rates in fishes are influenced by their feeding habits and geographical location conditions. Also from the result, it has shown that concentration of ^{40}K in fish samples was so high than the recommended value, but the computed total annual effective committed dose (Table 4) due to the consumption of fish was below the recommended limit [39]. This implies that consumers of fish from marine coastal areas are safe. Besides, it has been reported that the ^{40}K level in human body are not affected by variations in the environmental level, as result its radiation dose within the body remain constant [4, 41].

Table 3. *Activity Concentration of radionuclides (Bq/kg) in fish samples from coastal marine areas in Tanzania; values are expressed as mean ±SEM (p>0.05 and sample size n=12).*

S/No.	Sample Code	Activity (Bq/kg) ^{226}Ra	Activity (Bq/kg) ^{232}Th	Activity (Bq/g) ^{40}K
1	F001	BDL	BDL	573.1 ± 72.3
2	F 002	4.6 ± 1.5	BDL	286.0 ± 30.3
3	F003	1.5 ± 0.8	BDL	353.3 ± 34.5
4	F 004	BDL	BDL	620.3 ± 78.8
5	FT 001	10.4 ± 2.4	BDL	693.6 ± 48.9
6	FT 002	BDL	BDL	574.2 ± 63.2
7	FT 003	BDL	BDL	438.4 ± 50.6
8	FT 004	BDL	BDL	492.6 ± 30.4
9	FM 001	BDL	BDL	460.6 ± 28.6
10	FM 002	BDL	BDL	543.3 ± 64.3
11	FM 003	3.0 ± 1.3	BDL	373.9 ± 39.1
12	FM 004	BDL	BDL	456.6 ± 50.1
	Min	1.5 ± 0.8		286.0 ± 30.3
	Max	10.4 ± 2.4		693.6 ± 48.9
	Mean ± SEM	4.9 ± 1.9	BDL	488.8 ± 34.1

BDL- Below Detection Limit

Table 4. *Total Annual Effective Dose from 226Ra and 40K compared to recommended limits (mSv/y).*

Radionuclides	Effective dose (mSv/y)	References
^{226}Ra	0.0117	
^{40}K	0.0258	
Total	0.0375	This study
Recommended limits	1	[39]

The mean activity concentration of ^{40}K reported in this study was higher than the values reported in other parts of the world like Nigeria [42], Libya [43], Bangladesh [44], Pakistan [45], and Malaysia [46]. Figure 2 shows the comparison of radionuclides concentrations in fish observed in this study with the concentrations reported elsewhere as well as the recommended average values.

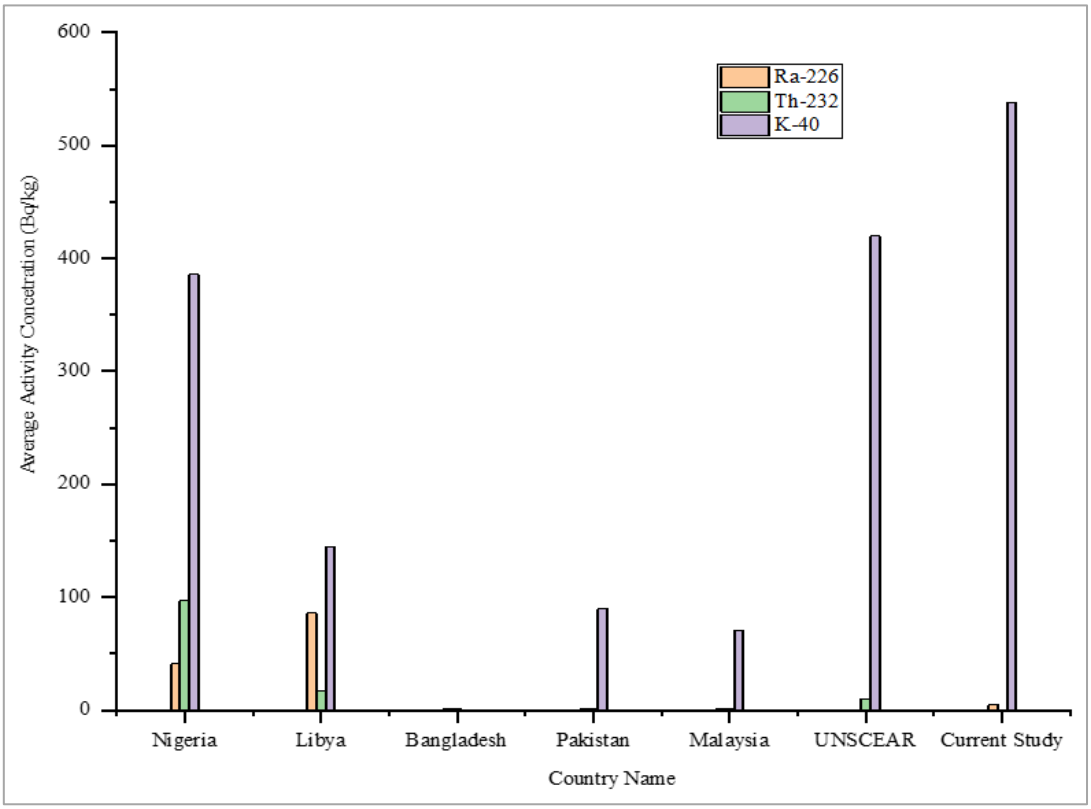

Figure 2. *Shows the comparison of radionuclides concentration in fish observed in the study area and parts of the world as well as recommended average value.*

4.2. Radiological Risk Indices in Fish

Following the high concentrations of radionuclides in fish, it was important to compute the radiological risk indices associated with consumption of fish in the study area. The H_{in} and ELCR were determined as presented in Table 5. The average values of all these indices were observed to be below the values recommended by UNSCEAR [4], as shown in Table 5. Hence, although some fish samples appeared to have high activity concentrations of ^{40}K, the computation of radiological risk indices and total annual effective committed dose (Table 4) revealed no significant health risks posed by consumption of fish from the marine coastal areas in Tanzania.

Table 5. *Computed radiological risk indices due to consumption of fish in the study area; values are expressed as mean ± SEM ($p>0.05$ and*

sample size n=12).

S/No.	Sample code	H_{in}	ELCR X 10-6
1	F001	0.1	105.7
2	F 002	0.06	52.9
3	F003	0.08	67.9
4	F 004	0.1	114.5
5	FT 001	0.2	147.4
6	FT 002	0.1	106.1
7	FT 003	0.09	81.2
8	FT 004	0.1	90.7
9	FM 001	0.1	85.1
10	FM 002	0.1	100.5
11	FM 003	0.09	74.6
12	FM 004	0.09	84
	Min	0.06	52.9
	Max	0.2	147.4
	Mean±SEM	0.1±0.01	92.6±7.1
Recommended mean values [4]		1	290

4.3. Radioactivity Levels in Sediments

The activity concentration of natural radionuclides in sediments samples was computed and presented in Table 6. It was found that activity concentration of ^{40}K was higher than other radionuclides in the study area which may be influenced by organic enrichment in sediments as a result of the waste discharges draining by rivers from mainland or coastal activities like farming, industrial and domestic municipal waste. The result also shows that the samples collected upper stream of the rivers, E_6 001, E_6 004, E_7 002, E_7 003, E_1 004, E_4 001 and E_4 004 have more activity concentration when compare with those sample collected downstream, midpoint and along the ocean in each river and sewage pipeline except sample E_7 001 and SWPT 003. This may justify that the natural radionuclide are draining from farming places, industrial places, domestic waste discharge, mineral exploration activities, oil and fuel extraction in the mainland and coastal itself and not from Ocean activities.

Again from Table 6, it shows that the activity concentration of natural radionuclides of sample collected from port station and ferry station were higher than those from river estuaries and sewage pipeline. The samples collected from Tanga port station were containing more activity than other port stations and ferry stations except sample with code MP002 because there was a dredging excise which was undertaken and samples were collected there. The higher concentration observed in port stations and ferry stations may be due to the geological and morphological of the rock formation. This observation was supported from the literature by Ramli *et al.* [47], and Yii *et al.* [5]. Also, it is reported that the radioactivity of ^{226}Ra and ^{232}Th is linked with heavy minerals, while that of ^{40}K is associated with clay minerals [48].

Moreover, from Table 6 it was observed that the ratio of $^{226}Ra/^{40}K$, $^{232}Th/^{40}K$ and $^{226}Ra/^{232}Th$ were comparable with recommended average by UNSCEAR [10]. The ratio of ^{226}Ra and ^{232}Th shows that there was disequilibria of radionuclides in the study area which may be caused by anthropogenic activities. This observation can be supported from the literature by Higgy [49].

Table 6. *Activity Concentration of radionuclides in the sediment samples from the coastal marine areas in Tanzania; values are expressed as*

mean ±SEM (sample size n= 37).

S/No.	Sample code	Activity Bq/Kg Ra-226	Activity in Bq/Kg Th-232	Activity in Bq/Kg K-40	Ra-226 / K-40	Th-232 / K-40	Ra- 226 / Th-232
1	E6 001	15.4±1.8	15.0 ± 1.9	568.3 ± 54.5	0.03	0.03	1.03
2	E6 002	9.3 ± 1.4	14.1 ± 1.6	285.4 ± 29.1	0.03	0.05	0.66
3	E6 003	3.7 ± 0.8	3.2 ± 0.9	370.2 ± 36.5	0.01	0.01	1.16
4	E6 004	7.6 ± 0.5	6.0 ± 0.7	643.8 ± 15.1	0.01	0.01	1.27
5	E6 005	3.6 ± 0.6	2.4 ± 0.5	180.4 ± 17.5	0.02	0.01	1.5
6	E7 001	11.6±1.7	10.3 ± 1.3	405.5 ± 41.0	0.03	0.03	1.13
7	E7 002	8.7 ± 1.2	10.2 ± 1.3	488.7 ± 48.9	0.02	0.02	0.85
8	E7 003	14.3±1.6	20.2 ± 2.0	372.5 ± 35.1	0.04	0.05	0.71
9	SWP 001	4.5 ± 0.8	< MDA	202.9 ± 19.1	0.02		
10	SWP 002	7.8 ± 0.6	7.6 ± 0.7	100.5 ± 6.2	0.08	0.08	1.03
11	SWP 003	5.8 ± 0.9	6.2 ± 0.8	311.5 ± 30.0	0.02	0.02	0.94
12	SWP 004	4.2 ± 1.1	3.4 ± 1.0	399.1 ± 40.7	0.01	0.01	1.24
13	DP 001	7.2 ± 0.3	4.6 ± 0.3	177.3 ± 4.8	0.04	0.03	1.57
14	DP 002	7.5 ± 0.5	4.2 ± 0.6	195.9 ± 6.6	0.04	0.02	1.79
15	DP 003	12.6±1.0	14.7 ± 1.0	314.9 ± 13.4	0.04	0.05	0.86
16	E1 001	7.6 ± 0.4	< MDA	420.2 ± 11.4	0.02		
17	E1 002	8.4 ± 0.5	< MDA	396.0 ± 10.2	0.02		
18	E1 003	7.1 ±1.6	7.6 ± 1.2	467.6 ± 46.7	0.02	0.02	0.93
19	E1 004	17.0±2.0	25.8 ± 3.6	395.8 ± 39.8	0.04	0.07	0.66
20	SWPT01	9.6 ± 1.6	7.4 ± 1.2	63.0 ± 11.4	0.15	0.12	1.29
21	SWPT02	7.7 ±0.7	7.6 ± 0.4	92.1 ± 4.1	0.08	0.08	1.01
22	SWPT03	46.6±6.9	47.1 ± 5.5	312.9 ± 44.0	0.15	0.15	0.99
23	SWPT04	9.0 ± 0.4	< MDA	86.9 ± 3.0	0.1		
24	SWPT05	11.8±1.1	7.4 ± 0.7	153.6 ± 12.1	0.08	0.05	1.59
25	TP 001	15.4±0.8	12.7 ± 0.8	324.2 ± 14.7	0.05	0.04	1.21
26	TP 002	32.1±0.7	19.5 ± 0.7	210.9 ± 5.1	0.15	0.09	1.65
27	TP 003	31.2±2.2	21.9 ± 1.4	238.1 ± 16.4	0.13	0.09	1.42
28	TP 004	28.6±3.1	23.1 ± 2.4	196.1 ± 21.4	0.15	0.12	1.24
29	TP 005	27.9±1.5	22.7 ± 1.1	267.5 ± 14.1	0.1	0.08	1.23
30	E4 001	16.0±1.2	25.5 ± 1.5	607.0 ± 28.7	0.03	0.04	0.63
31	E4 002	10.3±1.4	21.5 ± 2.3	560.8 ± 55.4	0.02	0.04	0.48
32	E4 003	6.9 ± 1.1	10.1 ± 1.2	524.0 ± 50.3	0.01	0.02	0.68
33	E4 004	16.3±1.9	41.1 ± 4.1	641.9 ± 62.2	0.03	0.06	0.39
34	MP 001	12.3±0.5	12.3 ± 0.9	285.2 ± 9.1	0.04	0.04	1
35	MP 002	64.7±3.0	(117.2 ± 5.4)	234.9 ± 11.2	0.28		
36	MP 003	12.5±0.6	9.5 ± 0.6	281.2 ± 8.0	0.04	0.03	1.32
37	MP 004	17.3±2.3	20.9 ± 2.7	354.2 ± 36.4	0.05	0.06	0.83

S/No.	Sample code	Activity Bq/Kg Ra-226	Activity in Bq/Kg Th-232	Activity in Bq/Kg K-40	Ra-226 / K-40	Th-232 / K-40	Ra- 226 / Th-232
	Min.	3.6 ± 0.6	2.4 ± 0.5	63.0 ± 11.4	0.01	0.01	0.39
	Max.	64.7±3.0	47.1 ± 5.5	643.8 ± 15.1	0.28	0.15	1.79
Mean±SEM		15.1±2.2	14.9 ±2.02	320.8±27.4	0.06±0	0.05±0	1.08±0.1
Recommended mean values [10]					0.067	0.067	1.0

The result in the brackets () are outliers and excluded from average.

Figure 3 shows bar chart graph illustrating the distribution of mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the study area. It was observed that the mean activity concentration of ^{40}K in river estuaries (E_6, E_7, E_4, E_1) are so high compared to sewage pipelines and port/ferry site. The Tanga sewage pipeline (SWPT) contain the least mean activity concentration of ^{40}K. The other radionuclides ^{226}Ra and ^{232}Th at the TP, MP, E_4 and SWPT site contain a bit higher mean activity concentration compared to other sites.

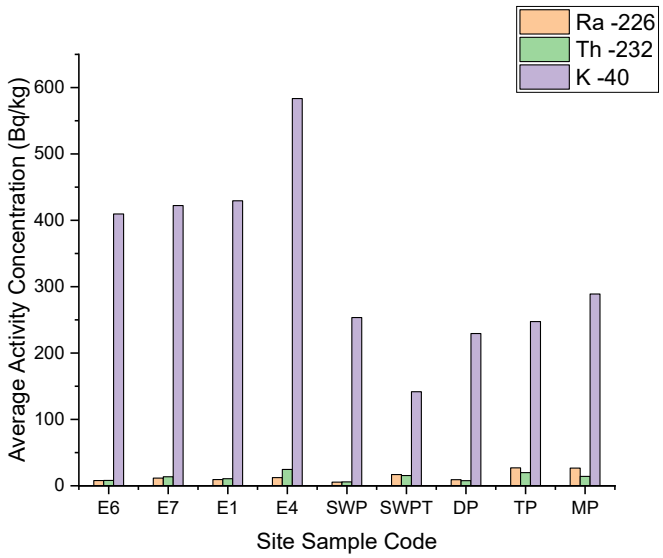

Figure 3. *The average activity concentration of 226Ra, 232Th and 40K in Bq/kg of the sediment sample for the locations under the study area.*

Figure 4 and Figure 5 were plotted to test for correlation between ^{232}Th and ^{226}Ra and ^{40}K and ^{226}Ra respectively. It is noted that a moderate to strong good correlation between ^{232}Th and ^{226}Ra was observed with correlation coefficient of 0.79 (r= 0.79, p< 0.0001), this indicate that the correlation is highly statistically significant and hence the two radionuclide are in the same origin. Also the correlation was observed between ^{226}Ra and ^{40}K with correlation coefficient of -0.103 (r= -0.103, p≈ 0.545) which is very weak negative correlation that means no statistically significant correlation and suggest that no significant linear relationship between activity concentration of ^{40}K and ^{226}Ra.

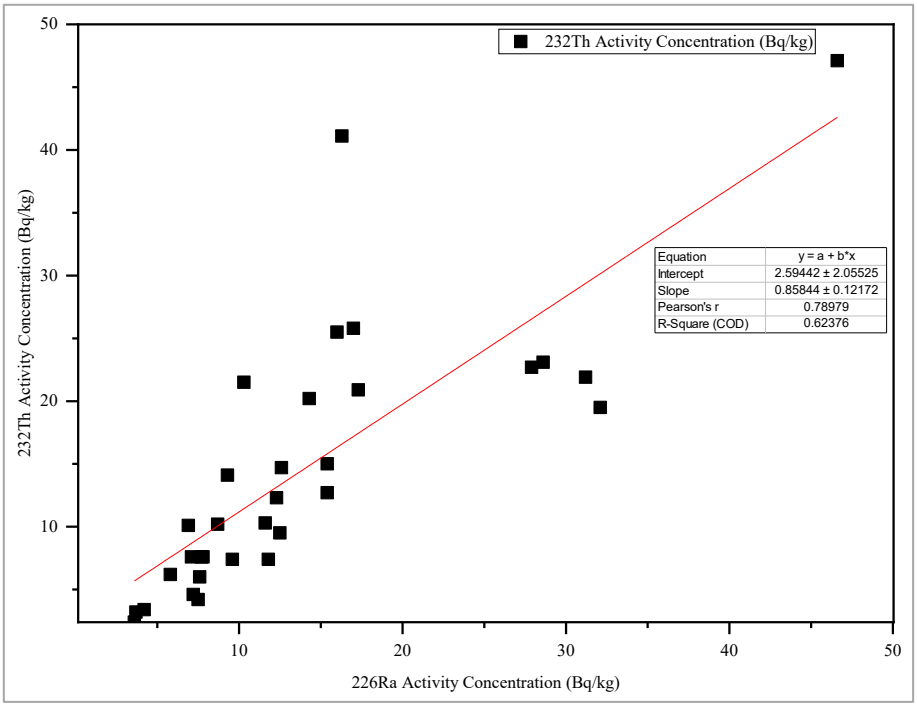

Figure 4. *The correlation between activity concentrations of 232Th and 226R.*

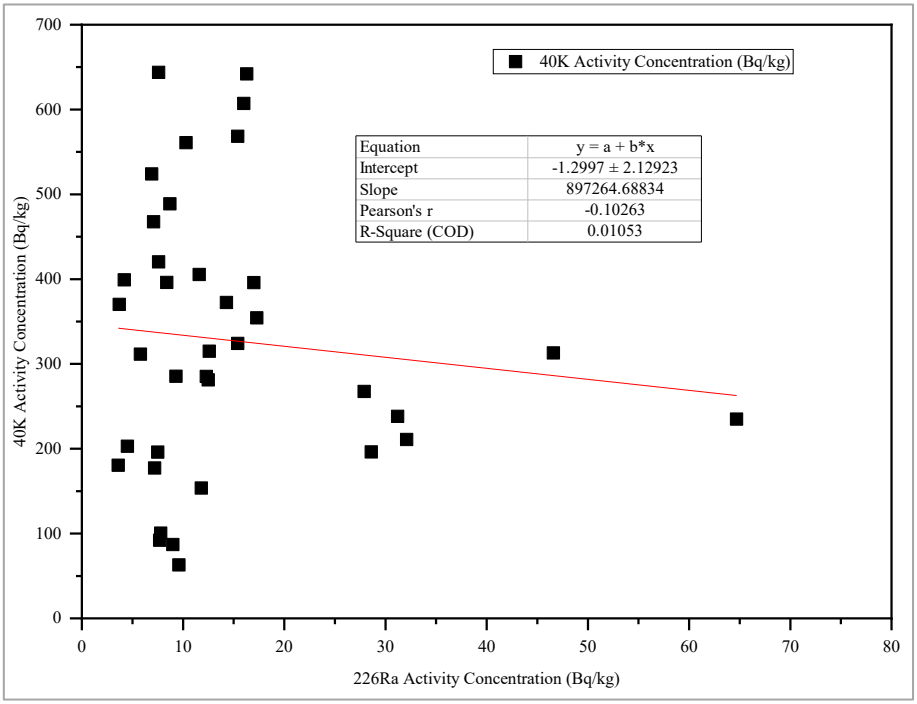

Figure 5. *The correlation between activity concentrations of 40K and 226Ra.*

Following the comparison between the ranges and mean activity concentration of radionuclide (^{226}Ra, ^{232}Th and ^{40}K) in sediments samples collected in coastal marine areas of Tanzania and some other countries of the world was presented in Table 7. The radioactivity levels in the coastal marine areas of Tanzania sediments was found to be higher than the levels reported for the Mediterranean coast of Egypt [50], coastal structure in Kuwait [51], coast of Oman sea, Iran [52], Qatar [53], and coast of Greater Accra, Ghana [54], except ^{226}Ra in Kuwait and ^{232}Th in Ghana were higher compared to the current study. Also for Patras Harbour, Greece [55], Mumbai Harbour, India [56], Red Seashore sediment, Egypt [57], Coastal Marine Sediments, Gulf of Oman [58], Yangtze estuary, China [59], Izmit Bay, Turkey [60], and Malaysia Coast,

South China sea [61], the levels are comparable with the current study, except the radioactivity of ^{40}K in Greece, India, Egypt, Turkey and China is higher than the current study.

Moreover, from Table 7, when you compare the ranges, the radioactivity levels of the current study was found to be higher than the levels reported for Qatar [53], and northern coast of Oman Sea, Iran [52]. The Yangtze Estuary, China [59], was found to be comparable with the current study except the ranges of ^{232}Th and ^{40}K was higher than the current study. Therefore, the mean and ranges of the radioactivity levels in sediment samples of the current study were lower than recommended values reported worldwide by UNSCEAR [10], but mostly comparable with other part of the world. Hence the sediments collected from coastal marine areas of Tanzania cannot pose radiological effect to human being as well as biota animals.

Table 7. *Comparison of average /ranges of Activity Concentration of 40K, 232Th and 226Ra (Bq/kg) from study area, other parts of the world and the recommended values.*

S/No.	Location	Average/ranges of Activity Concentration in Bq/kg			Reference
		Ra - 226	Th - 232	K - 40	
1	Kuwait	36	6	227	[53]
2	Greece	22.6	24.5	497	[55]
3	India	10.6		436	[56]
4	Egypt	5	2.1	46	[50]
5	Ghana	22.04	108.6	29.78	[54]
6	Egypt	24.7	31.4	427.5	[57]
7	Iran	11.8 -22.7	10.7 - 25.0	223 - 535	[52]
8	Qatar	4.2 - 19.5	1.0 - 6.0	11 - 188	[53]
9	China	13.7 - 52	26.1 - 71.9	392 - 898	[59]
10	Turkey	0.0 - 18.0		568	[60]
11	Oman	0.0 - 16.2	0.0 - 34.5	54.7	[58]
12	Malaysia	27.7	73.3		[61]
13	World range	17.0 - 60.0	11.0 -64.0	140.0 - 850.0	[10]
14	World average	35	30	400	[10]
15	Tanzania, range	3.6 - 64.7	2.4 - 47.1	63.0 - 643.8	Current study
16	Tanzania, average	14.6	14.6	327.9	Current study

4.4. Radiological Risk Indices in Sediments

The radiological indices and hazard indices for sediments samples collected from study area were computed and presented in Table 8. Generally it observed that the radiological hazards indices were below the recommended values as reported by UNSCEAR [4], as shown in Table 8.

Moreover, when you compare the current study with the reported study recorded in Table 8, it shows that the Suliman *et al.* [57] in Egypt and Al-Trabulsy *et al.* [61] in Saudi Arabia have higher values for Ra_{eq} and D compared with current study. At the same times Papeefthymiou *et al.* [54] in Ghana reported to have lower values for Ra_{eq} and E_{eff} and Ramasamy *et al.* [62] in India reported to have lower value for ELCR compared to the current study.

Hence, in view the radiological hazard indices calculated in sediments samples and compared to the recommended values and other studies reported over the world it shows that the radioactivity levels in the coastal marine areas of Tanzania sediments have no radiological concern for human health and biotic environment. Therefore, the sediment around the study can be used for building materials or other various applications.

Table 8. Using Average Activity Concentration to Computed Raeq, Hex, D, Eeff, Iγ, AGDE and ELCR for Sediments Samples from present study and sediments from other parts of world as well the recommended value; values are expressed as mean± SEM (p<0.05 and sample size n=9).

S/No.	Sample code	Ra_{eq} Bq/kg	H_{ex}	D nGy/h	Eeff µSv/y	I_γ	AGDE µSv/y	ELCR x 10-6	Reference
1	E6	51	0.1	26	31.9	0.4	164.9	111.7	
2	E7	63.5	0.2	31.5	38.6	0.5	193	135.1	
3	SWP	33.4	0.1	16.9	20.7	0.3	105.6	72.5	
4	DP	37.9	0.1	18.6	22.8	0.3	107.5	79.8	
5	E1	57.6	0.2	29.1	35.7	0.5	182.1	124.9	
6	SWPT	49.8	0.1	22.9	28.1	0.4	114.1	98.4	
7	TP	74.4	0.2	34.5	42.3	0.5	168.9	148.1	
8	E4	92.5	0.2	45.7	56	0.7	289.9	196	
9	MP	69.3	0.2	32.7	40.1	0.3	105.6	72.5	
	Min	33.4	0.1	16.9	20.7	0.1	55.1	32.6	
	Max	92.5	0.2	45.7	56	0.7	289.9	196	
	Mean±SEM	58.8±6.2	0.2±0	28.7±2.9	35.1±3.6	0.4±0	159.1±20.2	115.4±13.6	
Mean values, others areas									
Red Seashore, Egypt		101		42					[57]
Coast of Accra, Ghana		9	0.5	77	0.1				[54]
India								0.002	[62]
Saudi Arabia		92.9	0.1	45.6	56	0.3			[61]
Recommended value		370	1	59	70	1	300	290	[4]

5. Conclusion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of fish and sediments samples along the coastal areas of Tanzania was determined using the gamma spectrometry. The mean activity concentrations of ^{226}Ra in fish (4.9 ± 1.9 Bq kg^{-1}) and sediments (15.1 ± 2.2 Bq kg^{-1}), ^{232}Th in fish was below detection limit (BDL) and sediments (14.9 ± 2.0 Bq kg^{-1}) were compared to the global averages of 35 Bq kg^{-1}, and 45 Bqkg^{-1}respectively. The ^{40}K in fish (488.8 ± 34.1 Bq kg^{-1}) and sediments (320.8 ± 27.4 Bq kg^{-1}) were compared to the global average value of 420 Bq kg^{-1}. The study also revealed that the computed annual effective committed dose was below the recommended limit by ICRP although the mean activity concentration of ^{40}K in fish samples was higher than the value recommended worldwide. Additionally, the current study also has shown that the radiological hazards indices computed for fish and sediments samples were below recommended values. Likewise, the study also has shown strong good correlation between ^{226}Ra and ^{232}Th, while a weak correlation was observed between ^{40}K and ^{226}Ra as well as ^{232}Th in sediments samples.

Therefore, this study may conclude that the level of radioactivity in fish consumed in this study areas does not pose a significant radiological risk to fish consumers as well as sediments if it will be used for any purpose. This study also will be used as baseline for coastal marine areas of Tanzania for radioactivity and the data may be used by regulatory bodies during enforcing radiation protection policies. Lastly, the study recommends that research should be extended to other species of fish and the areas left behind during sample collection and extend the methodology to include further radionuclides of interest to nuclear security and safeguards.

Abbreviations

UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
DNA	Deoxyribonucleic Acid
TAEC	Tanzania Atomic Energy Commission

IAEA	International Atomic Energy Agency
HPGe	High - Purity Germanium
AGDE	Annual Gonadal Dose Equivalent
ELCR	Excess Lifetime Cancer Risk
ICRP	International Commission on Radiological
BDL	Below Detection Limit

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Author Contributions

Melchior Ludovick Mungubariki: Conceptualization, Data curation, Formal Analysis, Methodology, Resources, Validation, Visualization, Poject administration, Writing– original draft, Writing– review & editing

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Innocent Jimmy Lugendo: Methodology, Supervision, Validation, Visualization, Writing – review & editing

Conflicts of Interest

The authors declares that they have no conflicts of interest.

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