Original Research Article

Natural Radioactivity in Vegetables from Selected Areas in Manyoni, Central Tanzania

ABSTRACT

The study aimed at determining the mean concentrations of natural radionuclides, the daily intake of radionuclides and annual effective dose due to the ingestion of Vegetables from High background radiation areas of Manyoni, in Central Tanzania. A total of 30 leaf vegetable samples grouped into five categories were collected randomly from different locations of the study area. The levels of U-238, Th-238, and K-40 were measured by direct γ-ray spectrometry using HPGe detector by Compton suppression method. The radioactivity in vegetables ranged from 2.22 Bq/kg – 36.76 Bq/kg for U-238, 4.06 Bq/kg – 68.29 Bq/kg for Th-232 and 700.20 Bq/kg – 2523.90 Bq/kg for K-40, respectively. Except for K-40, the activity levels reported into this study were lower than the activity levels of vegetables reported from various parts of Tanzania. However, the concentrations of radionuclides in the vegetables samples found in this study were higher than the world average values suggested by the UNSCEAR. The annual effective dose due to intake of vegetables was 2.73 mSv/year. The annual effective ingestion dose was found 9.4 times higher than total exposure per person resulting from the ingestion of terrestrial radionuclides. The daily intakes of nuclides were within the allowable limits.

Key Worlds: Uranium deposit; Radioactivity; Effective dose; Daily intake of radionuclides

1. INTRODUCTION

The knowledge of radionuclides concentration and distribution in vegetation is of interest (in environmental radiological protection) not only for an accurate determination of activity but also to assess and estimate the effects/potential hazard (monitoring of environmental radioactivity) of radiation exposure inherent in the measured activity to human and biota. More interests have been arisen to minimize the hazards caused by ionizing radiation from manmade or naturally occurring radioactive materials. In the past people paid the price for little or improper knowledge on the effect of ionizing radiation. However the emphasis on radiation protection, safety and security of radioactive sources has positively contributed to reduce such hazards as the people’s awareness has apparently been enhanced [1].

Natural radionuclides occur in soil and they are incorporated metabolically into plants, and ultimately find their way into food crops and water. Man-made radionuclides behave in a similar manner, and worldwide contamination of the food chains by radionuclides produced during tests of nuclear weapons in the atmosphere has taken place during the past half century [2]. In the recent years, extensive uranium exploration and feasibility studies in Tanzania have found several sites with economically viable uranium deposits. In 2009, deposits of uranium were discovered at Mkuju, Namtumbo district, southern Tanzania [3]. This discovery was followed by Manyoni uranium deposits (Singida region) in Central Tanzania [4]. These discoveries of uranium deposit at Mkuju in Ruvuma, Bahi in Dodoma and Manyoni in Singida have brought concern about the levels of natural radioactivity in soil and in locally grown food crops at the areas in the neighbourhood of the deposits.

Green leafy vegetables are very prone to external contamination during their growing season. Other vegetables, including root vegetables, may also become contaminated. In the early stages of fallout, green vegetables can be a very significant pathway for short lived radionuclides. The radioactivity levels play a vital role as they can be used for decision making in economic, legal or environmental management. So far there is no information regarding radioactivity levels in leaf vegetables from the area of study (Manyoni) which have been established. Therefore this study is aimed to establish a baseline radioactivity levels in leaf vegetables in the selected area of study before the commencements of Uranium mining and milling.

In the present work, radioactivity levels of U-238, Th-232, and K-40 in daily diets were determined in selected leaf vegetables from Manyoni. The obtained results were used for the estimation of intakes, and annual effective doses of these radionuclides in the adult population of Manyoni District, Tanzania.

2. MATERIALS AND METHODS

2.1 Description of the study area

Manyoni District (Figure 1) is located in the central part of Tanzania. Its geographical coordinates are 5 45' 0" South, 34 50' 0" East. It has an area of 28,620 sq.km, which is about 58 % of the entire of Singida region. The district has a population of 296,763 people [10]. The area under the study is mainly used for cultivation of different types of crops (like maize, sorghum, vegetables, and Pulses) and animal grazing. The district is underlying on the uranium deposit which extends from Bahi district in Dodoma, the capital of Tanzania. The uranium targets in the area are described as calcrete-hosted uranium mineralization near to the surface and sandstone hosted deposits within buried fluvial channel systems [5].

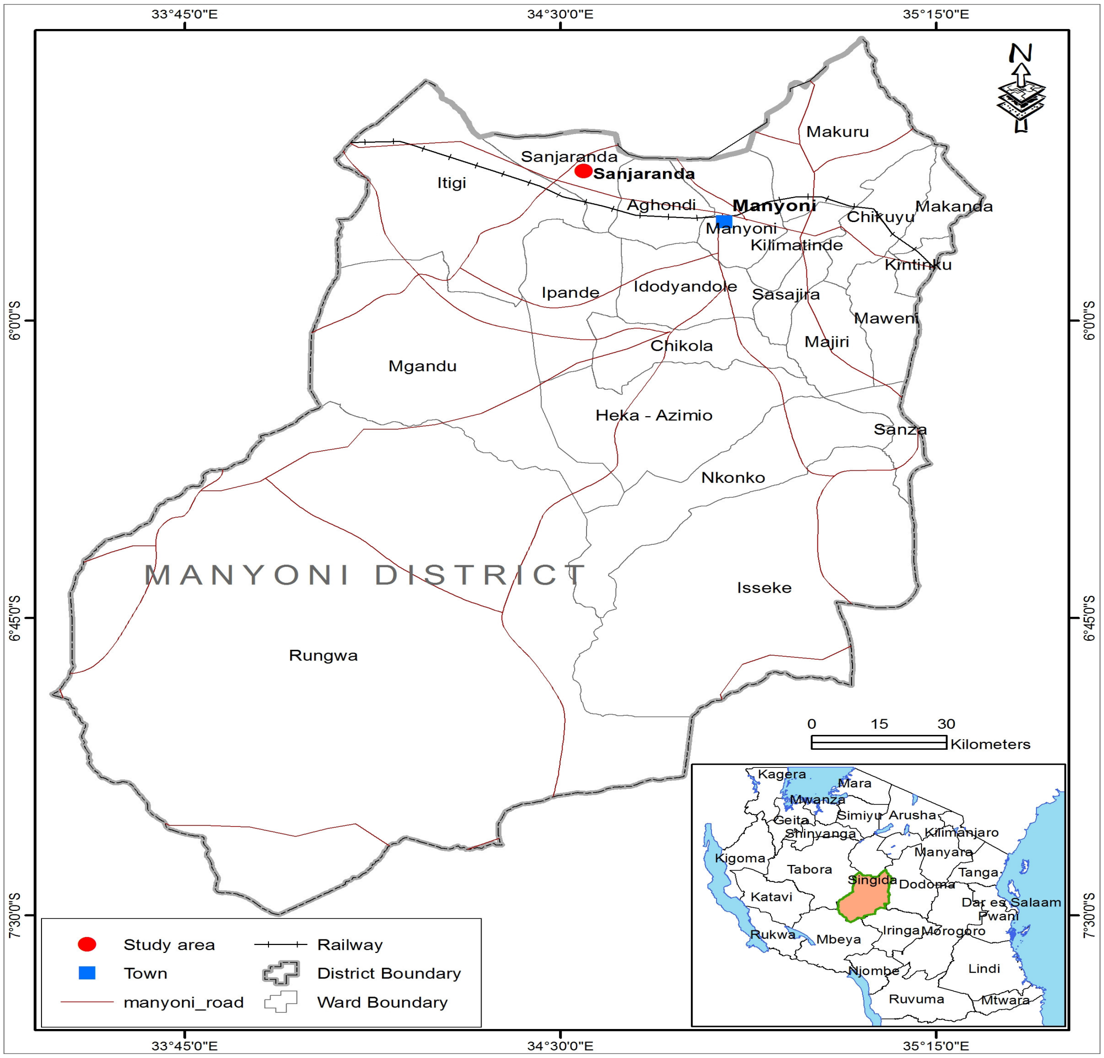

Figure 1: A map showing Manyoni District: The Insert shows its relative position in Tanzania

2.2 Sample collection and Preparation

The sampling was carried out during rainy season (between January and February). Thirty leaf vegetables samples were collected from the farmlands in selected areas in Manyoni district as shown in Table 1. The samples were washed with normal water, as for human consumption, weighed and chopped into small parts, oven dried at a temperature of 80℃ for 4 days, milled and sieved to collect the appropriate fractional mesh size**.** All the samples were packed in plastic (polyethylene) cylindrical containers of approximately 500 cm^3 volume and the containers were completely sealed for at least 4 weeks to allow radioactive equilibrium to be reached [1]. This step ensured that radon gas and its daughters remained in the sample. The table below indicates the type of samples and their place of origin.

Table 1: A list of various samples collected from different areas in Tanzania.

Sample ID	Sample name	Latitude	Longitude	Area
V1 (n=5)	African Spinach leaves (*Amaranthus ssp*)	S 05^0 41' 55.3"	E 34^0 44' 56.4"	kamenyanga
V2 (n=7)	Sweet potato leaves (*Ipomea batata*)	S 05^0 41' 55.3"	E 34^0 44' 56.4"	Kamenyanga
V3 (n=5)	Cowpea leaves (*Vigna unguiculata*)	S 05^0 42' 51.2"	E 34^0 52' 25.1"	Membeta
V4 (n=8)	Pumpkin leaves (*Curcubita moschata*)	S 05^0 43' 37.5"	E 34^0 35' 01.8"	Kitopeni
V5 (n=5)	Okra (*Abelmoschus esculentus*)	S 05^0 41’ 55.3”	E 34^0 44’ 56.4”	Kamenyanga

2.4 Instrumentation

The measurements were carried out at the Egyptian Second Research Reactor (ETRR-2). The gamma ray spectrometry technique was applied for determination of activity concentration in the vegetable samples. The radionuclides were determined by Compton suppression system, high-resolution gamma ray spectrometry using n-type HPGe detector Model GMP-100 250-S and Serial No. 38-N31278A coupled to a computer based PCA-MR 8192 Multi-Channel Analyzer (MCA) mounted in a cylindrical lead shield (100 mm thick) and cooled in liquid nitrogen. The detector has a relative efficiency of 100 % and resolution of 2.1 keV at 1.33 MeV of Co-60 line and a peak-to-Compton ratio of 64:1. The descriptions on the techniques used for energy and efficiency calibrations of the gamma-spectrometry system are well documented elsewhere [5, 6].

The specific radioactivity of K-40 was measured directly by its own gamma ray at 1460.8 keV (10.7), while activities of U-238/Ra-226 and Th-232 were calculated based on the weighted mean value of their respective decay products in equilibrium. The specific radioactivity of Ra-226/U-238 was measured using the 295.2 (18.2), 351.9 (35.1) keV gamma rays from Pb-214 and the 609.3 (44.6), 1764.5 (15.1) keV from Bi-214. The specific radioactivity of Th-232 was measured using the 911.2 (26.6) keV from 228-Ac, and the 583.2 (30.6) keV from 208-Tl. And the specific radioactivity of K-40 was measured directly by its own gamma ray at 1460.8 keV. The values inside the parentheses following gamma-ray energy indicate the absolute emission probability of the gamma decay.

2.4.1 Activity Calculation

The activity (A) in Bq/kg of the radionuclides in the samples was calculated after decay correction using the expression below [7] and presented in Table 3.

$$A = \frac{N}{T_L P_\gamma \varepsilon M} \quad (1)$$

Where M is the dry-weight of sample (kg), N is the net Peak area for the sample in the peak range, P_γ is the gamma emission probability, T_L is the counting live time, and ε is the photo peak efficiency [8].

2.4.2 Detection Limit

Radioactivity measurements are characterized by a variable zero level due to background. This situation obliges one to work with detection and determination limits when the radioactivity of the source is very low. For the purpose of accuracy and reliability of data in gamma spectrometry it is necessary in the measurement process to introduce two specific levels:

i) Detection limit – indicates if an analytical process leads to a quantitative detection.

ii) Decision limit – that allows one to deduce whether the result of the analysis indicates that the sample is radioactive or not radioactive.

Due to the fact that the detection limit is not always significant, another parameter (L_D) was introduced, which is the lower limit of detection taking into account 95 % level of confidence as 5 % presence of real activity (false negative) and 5 % no real activity (false positive) [8, 9, 10]. L_D is given by the following expression:

$$L_D = 2L_C + 2.706 \rightarrow 2(2.326\, \delta_{NB}) + 2.706 \quad (3)$$

Where, L_C is a critical level above which the degree of confidence is acceptable for the net count. ($L_C = 2.326\, \delta_{NB}$).

$$L_D = 4.652\, \delta_{NB} + 2.706 \quad (4)$$

The minimum detectable activity (MDA) was then calculated using the formula below:

$$MDA = \frac{L_D}{\varepsilon T P_\gamma} \quad (5)$$

Whereby P_γ is the gamma emission probability, T is the counting live time; ε is the photo peak efficiency. The minimum detectable activity (MDA) of the γ-ray measurement system was calculated according to the equation 2 above [10]. The MDA for each radionuclide was calculated and summarized in the Table 2 below:

Table 2: The Minimum Detectable Activity (Bq/kg) of detected radionuclides at specific energies

Daughter nuclide	Energy(KeV)	MDA (Bq/kg)
Pb-214	295.22	0.42
Bi-214	609.32	0.45
Ac-228	911.2	0.73
Tl-208	583.19	0.18
K-40	1460.75	5.02

3. RESULTS AND DISCUSSION

3.1 Activity Concentration of Vegetables

The activity concentrations of the natural radionuclides of the Uranium and Thorium series and K-40 were investigated in the Vegetables. The results obtained are shown in the Table 3 below. The total uncertainty value (Table 3) is composed of the random and systematic errors in all the factors involved in producing the final nuclide concentration result [11].

The radioactivity levels in selected vegetables ranged from 2.2 Bq/kg– 36.8 Bq/Kg for U-238, 4.06 Bq/kg – 68.29 Bq/kg for Th-232, 700.20 Bq/Kg – 2523.90 Bq/Kg for K-40. As shown in figure 2, of all the vegetable samples, highest levels of U-238 were found in Cowpea leaves from Membeta followed by Pumpkin leaves from Kitopeni. Sweet potato leaves and Okra recorded lowest concentration levels of about 5.76 Bq/kg and 2.22 Bq/kg respectively. For Th-232, highest activity concentration levels were found in Amaranthus and Pumpkin leaves from Kitopeni respectively. Sweet potato leaves from Kamenyanga occupied a third place. The lowest activity concentration was reported from Cowpea leaves from Membeta (Table 3).

Table 3: Activity concentration (Bq/kg) obtained in different analyzed samples

Name	Area	U-238	Th-232	K-40
African Spinach leaves (*Amaranthus ssp*)	Kamenyanga	19.7 ± 0.4	30.1 ± 0.9	2520.0 ± 50.5
Sweet potato leaves (*Ipomea batata*)	Kamenyanga	5.8 ± 0.5	12.4 ± 1.0	1660.0 ± 33.5
Cowpea leaves (*Vigna unguiculata*)	Membeta	36.8 ± 0.7	4.1 ± 0.6	700.0 ± 14.1
Pumpkin leaves (*Curcubita moschata*)	Kitopeni	30.8 ± 0.7	17.6 ± 1.1	2050.0 ± 41.2
Okra (*Abelmoschus esculentus*)	Kamenyanga	2.2 ± 0.3	6.5 ± 0.7	772.0 ± 15.6

The activity concentration of K-40 was found to be high in all vegetables. This can be attributed to the use of fertilizers in large extent affecting the radionuclides concentrations, especially potassium. But also the high value of K-40 may also be due to the soil origin and the nature of some vegetables.

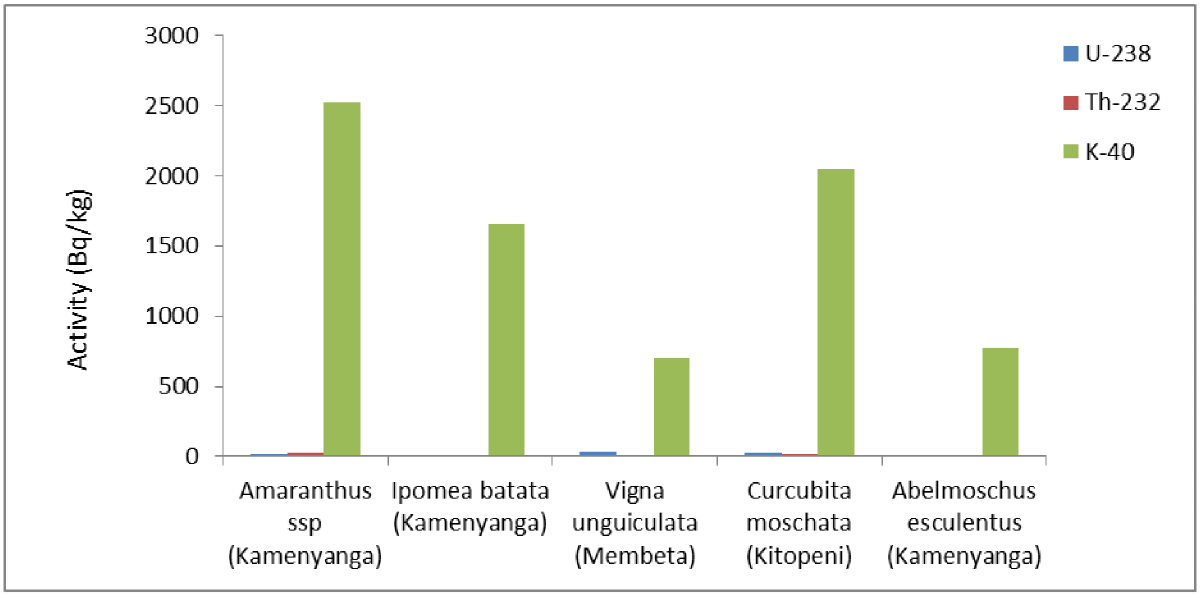

Figure 2: Activity Conc. (Bq/kg) in Vegetable from Manyoni

The activity concentrations of U-238, Th-232 and K-40 in vegetable samples found in this study were compared with studies of radioactivity in vegetable samples within the country (Tanzania) and from other countries as shown in Table 4. Literatures reviewed in this work includes radioactivity studies in vegetables from both low and high background radiation areas or from areas where soils have been contaminated with phosphate fertilizers as well as other means of contamination.

Table 4: Comparison of activity concentration (Bq/kg) in leaf vegetables obtained in this study with that from different parts of Tanzania and around the World

Place	U-238	Th-232	K-40	Reference
TANZANIA (Manyoni)	22.2 – 36.8 (19.06)	4.06 – 30.1 (14.13)	700.0 – 2520.0 (1540.4)	Present Study
TANZANIA (Minjingu)	393	318	1568.0	[1]
TANZANIA (Iringa)	11.3 – 58.9 (29.6)	18.4 – 40.3 (27.8)	437 – 1281 (960)	[12]
SUDAN	0.51 – 3.42 (1.44)	9.04 – 11.06 (10.05)	43.97 – 211.24 (143.47)	[13]
NIGERIA	15.9	20.8	232	[14]
CAMEROON	42	17	302	[15]

IRAN	BDL – 6.99 (5.21)	2.22 – 10.56 (4.76)	108.99 – 319.21 (186.15)	[16]
CHINA	16	23	-	[17]
JORDAN	0.6 – 2.6	0.7 – 3.4	698 - 1439	[18]
USA	24	18	-	[17]
WORD MEAN	35	30	400	UNSCER 21[19]
REF VALUE	20	15	-	[17]

Activity of U-238 in vegetables from Minjingu and Iringa were higher by factors of 20 and 1.5 compared to that reported in Manyoni. The reason for this difference is that, soils from Minjingu are contaminated due to presence of active phosphate mine and deposit [1] also farmers from Iringa use phosphate fertilizers [12]. When compared with activities from other countries, the activity of U-238 from Manyoni was higher than that reported from Jordan, Poland and Iran (Table 4). U-238 levels in vegetables from Manyoni were lower than the values reported from India and United States (Table 4). However, it was within the reference value of 20 Bq/kg [17].

The activity of Th-232 reported in vegetables from Minjingu and Iringa both in Tanzania were higher by folds of 22 and 2 when compared to that of this study. When compared with activity levels from other countries, the activity of Th-232 from this study was higher than the levels reported in vegetables from Sudan, Iran, Poland and Jordan (Table 4), but much lower than the levels reported from United States and World average. However it was the same as the reference value of 15 Bq/kg [17].

Activity of K-40 in vegetables from this study was the same as that reported in vegetables from Minjingu. The presence of phosphate mine and deposit in Minjingu as well as the Uranium deposit in Manyoni might be the reason. However, it was much higher than the all mean activities of K-40 reported in literature reviewed into this study. High concentration of K-40 in food is attributed to concentrations of K-40 in the soils, which may be associated with the geological properties of the area [20].

3.2. Calculation of the Annual Effective Dose

The annual effective ingestion dose due to vegetables consumption depends on the vegetables consumption rate and radioactivity in the vegetables. Calculation of the annual effective dose due to the ingestion of foods was performed based on the metabolic model developed by the International Commission of Radiological Protection [21]. The effective dose from a radionuclide in a foodstuff can be determined by the equation below [22].

$$E_{(\tau)_{ing,p}} = D_{ing} F_p C_{p,i} \qquad 6$$

Where $E_{(\tau)\ ing,\ p}$ is the annual effective dose by ingestion of the radionuclide *i* in foodstuff *p* (mSv/year), D_{ing} is the dose coefficient (dose conversion factor) by ingestion of radionuclide *i* (mSv Bq^{-1}) given by ICRP (1996), which varies with both radionuclides and the age of individuals. $C_{p,\,i}$ is the concentration of nuclide *i* in the ingested foodstuff *p* at the time of consumption (Bq/kg) and F_p is the consumption rate for foodstuff *p* (kg/year). In this study, the dose calculations are based on the assumption described in Table 5.

In this study, it was not possible to compute the vegetables consumption rate because, in Tanzania during the rainy season, vegetables are usually consumed daily to several times a week, while during the dry season frequency of consumption spans from once a week to several times a week [23]. Therefore, the consumption rate of 40 kg/person/year given by FAO was adopted [24]

Table 5: Dose conversion factors and Vegetable consumption rate in Tanzania

Radionuclide	Dose conversion Factor (Sv/Bq)	Vegetable Consumption rates (Kg/person/year) [24]
U-238	4.5 E-08	40
Th-232	2.3 E-07	
K-40	6.2 E-09	

The annual effective ingestion dose due to intake of radio-nuclides is shown in Table 6 and 7. Results show that the total average effective ingestion dose due to annual intake of U-238, Th-232 and K-40 from vegetables was 2.73 mSv/year of which 1.91 mSv/year was from 40K and 0.82 mSv/year was from U-238, Th-232 and K-40.

According to a report by UNSCER [25], the total exposure per person resulting from ingestion of terrestrial radionuclides should be less or equal to 0.29 mSv/year, of which 0.17 mSv/years is from K-40 and 0.12 mSv/year is from thorium (Th-232) and uranium series (U-238). The total annual effective ingestion dose due to intake of vegetables was 2.73 ± 0.08 mSv/year which was found 9.4 times higher than world safe value of total exposure per person resulting from the ingestion of terrestrial radionuclides (Table 7).

Table 6: Annual effective dose (Sv/year)

Name	Radionuclide	Activity (Bq/kg)	Annual effective dose (mSv/year)
African Spinach leaves (*Amaranthus ssp*)	U-238	19.7 ± 0.42	0.036 ± 0.001
	Th-232	30.1 ± 0.89	0.277 ± 0.008
	K-40	2520.0 ± 50.54	0.625 ± 0.013
Sweet potato leaves (*Ipomea batata*)	U-238	5.76 ± 0.49	0.011 ± 0.001
	Th-232	12.4 ± 1.04	0.114 ± 0.009

	K-40	1660.0 ± 33.45	0.412 ± 0.008
Cowpea leaves (*Vigna unguiculata*)	U-238	36.8 ±0.65	0.066 ± 0.001
	Th-232	4.06 ± 0.62	0.037 ± 0.006
	K-40	700.0 ± 14.13	0.174 ± 0.004
Pumpkin leaves (*Curcubita moschata*)	U-238	30.84± 0.68	0.056 ± 0.001
	Th-232	17.6 ± 1.08	0.162 ± 0.010
	K-40	2050.0 ± 41.17	0.508 ± 0.010
Okra (*Abelmoschus esculentus*)	U-238	2.22 ± 0.33	0.004 ± 0.001
	Th-232	6.49 ± 0.67	0.060 ± 0.006
	K-40	772.0 ± 15.55	0.191 ± 0.004

Table 7: Contribution of each nuclide in annual effective dose (mSv/year)

Radionuclide	**Annual Effective doses**		**Allowable Limits [25]**
U-238	0.173 ± 0.005	0.823 ± 0.044	**0.12**
Th-232	0.650 ± 0.039		
K-40	1.910 ± 0.039	1.910 ± 0.039	**0.17**
Total Annual Effective Dose		2.733 ± 0.083	**0.29**

3.3 Calculation of Total daily intake of radionuclides

For the total daily intake evaluation, the weighted average for each radionuclide in each vegetable category (ω) was calculated, and then multiplied by the respective consumption rate (F_p), and then presented in Table 8 and compared with the allowable limits.

$$D_{intake,\, i} = \omega \times F_p \qquad (7)$$

Table 8: Total daily intake of radionuclides (Bq) via ingestion of vegetables

Radionuclide	Weighted average radioactivity (ω) (Bq/kg)	Estimated Daily Intake (Bq)	Allowable Daily Intake of nuclides (Bq) [17]
U-238	19.06 ± 0.51	2.09 ± 0.06	5.7
Th-232	14.13 ± 0.86	1.55 ± 0.09	1.7
K-40	1540.40 ± 30.97	168.8 ± 3.39	-

The daily intake of nuclide especially U-238 and Th-232 were below the allowable daily intake as shown in Table 8. These daily intake values were much lower compared to the daily intake of U-238 and Th-232 reported from China, Germany, India and United States of America [17].

4. CONCLUSION

A radiological study was performed to determine the radioactivity levels in vegetables and the annual effective dose of natural radionuclides (U-238, Th-232 and K-40) into human body due to intake of vegetables from high background radiation areas in Manyoni District, Central part of Tanzania. The radioactivity concentrations in vegetable samples were higher than the world average value. The annual effective ingestion dose to human body due to intake of vegetables was also found 9.4 times higher than the world safe value of total exposure per person resulting from the ingestion of terrestrial radioisotopes. The daily intakes of nuclides (U-238 and Th-232) were within the allowable daily intake. Since this study aimed to establish a baseline database of radioactivity background levels and radiation dose due to ingestion, the results can be used as reference information to assess any changes in the radiological background levels due to any geological processes that may occur in future. This will facilitate comparative studies between these results and those, which might occur in future when the country decides to mine and mill the uranium ore.

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