Radioactivity levels in maize from high background radiation areas and dose estimates for the public in Tanzania

Abstract Natural rac

Natural radioactivity levels in maize which is one of the staple foods in various regions in 9 Tanzania have been studied. The radioactivity concentration of ²³⁸U, ²³²Th and ⁴⁰K were 10 determined using y ray spectrometry employing HPGe detector of relative efficiency of 51 %. 11 The average radioactivity concentrations in maize from five regions were ranged from 1.8 ± 12 0.2 to 23.6 \pm 0.7 Bq/kg ²³⁸U, 2.2 \pm 0.1 to 38.9 \pm 1.0 Bq/kg for ²³²Th and 42.0 \pm 0.4 to 434.6 \pm 13 18.7 Bg/kg for ⁴⁰K respectively. Total annual committed effective dose due to total ²³⁸U and 14 ²³²Th intakes as a result of consumption of maize in five Regions were as follows; Manyara 15 (1.46 mSv/y), Mbeya (0.31 mSv/y), Dodoma (0.21 mSv/y), Ruvuma (0.19 mSv/y) and Dar es 16 17 Salaam (0.08 mSv/y). The dose value from Manyara was almost the same to the annual dose guideline for the general public which is 1 mSv/y, where as for other regions the doses 18 are low. Hence a conclusion could be made that food crops cultivated at Minjingu village 19 might expose the population to high radiation dose which might be detrimental to their 20 21 health.

Original Research Article

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Keywords: Radioactivity, Minjingu phosphate deposit, committed effective dose, Uranium
 Deposit

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27 **1. Introduction**

28 The knowledge of natural radioactivity in man and his environment is very important 29 since naturally occurring radionuclides are the major sources of radiation exposure to man [1]. Radionuclides enter the human body through complex mechanism including ingested 30 31 foodstuffs via the food chain from natural sources. Ingestion of radionuclides through food intake accounts for a substantial part of average radiation doses to various organs of the 32 33 body and also represents one of the important pathways for long term health considerations. Radionuclide have always been present in food at various levels depending on factors such 34 as radioactivity contents in soil and the transfer characteristics from the environment medium 35 to food for other regions the dose values are lower stuff and hence to man [2]. The status of 36 37 the soil on which food crops are grown determines, to a significant extent, the quality of food crops produced, the season of the year also determines to a great extent the magnitude of 38 39 contamination of different foods [2].

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41 In Tanzania, the main source of contamination of agricultural soils is through natural occurring radioactive materials (NORMs) and application of fertilizers. All minerals and raw 42 43 material contains radionuclide of natural origin, sometimes the radioactivity of those radionuclides is higher than the normal background and might exceed the world limits. The 44 most important for the purposes of radiation protection are the radionuclides in the ²³⁸U and 45 ²³²Th decay series. In the recent years, extensive uranium exploration and feasibility studies 46 in Tanzania have found several sites with economically viable uranium deposits. In 2009, 47 deposits of uranium were discovered at Mkuju, Namtumbo district, southern Tanzania [3]. 48 49 This discovery was followed by Manyoni uranium deposits (Singida region) and Bahi uranium deposits (Dodoma region) both in Central Tanzania [4]. Furthermore, Tanzania has several phosphate deposits (Figure 1); however, the biggest one is Minjingu phosphate deposit found in Manyara region [5]. These discoveries of uranium deposit at Mkuju in Ruvuma and Bahi in Dodoma region and the presence of phosphate deposit and mine at Minjingu village in Manyara region has 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.

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The soil samples from Likuyu village in Ruvuma region had an average radioactivity as 58 follows. For ²³⁸U the concentration was 51.7 \pm 3.8 Bq/kg, ²³²Th was 36.4 \pm 3.1 Bq/kg and ⁴⁰K 59 was 564.3 ± 9.9 Bq/kg [6]. At Bahi District in Dodoma, the level of radioactivity in soils was 60 reported as for ²²⁶Ra the concentration was 25.2 ± 5.4 Bq/kg, ²³²Th was 37.2 ± 10.7 Bq/kg 61 and ⁴⁰K was 494.9 ± 6.2 Bq/kg [7]. These values were higher than the average activity 62 concentrations of Tanzania soils which are 350.4 ± 18.3, 24.7 ± 2.2 and 34.2 ± 2.0 Bg/kg for 63 ⁴⁰K, ²³²Th and ²³⁸U [8]. However, these radioactivity values were higher than the world 64 average activities of the same radionuclides. Worldwide there are reports which indicate high 65 radioactivity levels in regions near uranium deposits. Reports from Madagascar and India 66 showed that the level of ²³⁸U and ²³²Th and their daughters in the soil around the uranium 67 deposit was high compared to the world average [9]. It has been reported high activity 68 concentration of ²²⁶Ra, ²³²Th and ⁴⁰K in soil around Gurvanbulag uranium deposit in the 69 eastern part of Mongolia [10]. Soils around the phosphate deposit have been reported to 70 contain relatively high concentration of ²³⁸U and ²³²Th and their daughters. The rock 71 phosphate from Minjingu contains uranium concentration of (480 - 1100 ppm) contrast to 72 other rocks in the world [8]. Also the rock phosphate had the activity of 286, 698 and 5022 73 Bq/kg for ⁴⁰K, ²³²Th and ²³⁸U respectively [5]. The fertilizer (Triple superphosphate) 74 manufactured from Minjingu phosphate rock are also reported to have high activities of 362, 75 444 and 3116 for ⁴⁰K, ²³²Th and ²³⁸U, and for Superphosphate, the following activities 491, 433 and 3394 for ⁴⁰K, ²³²Th and ²³⁸U were reported [5]. Therefore, the extensive use of 76 77 phosphate fertilizers for agriculture in Mbeya region might contribute significantly to activity 78 79 levels of farm soils [11] and then to maize via root uptake.

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Studies on radioactivity in food crops grown in high background radiation areas have 81 82 been reported worldwide [12]. In Tanzania, radioactivity levels in edible leaf vegetation, maize and mung beans has been reported in Minjingu [8, 13]. Maize is the food crop that is 83 highly consumed at highest rates and forms the most important part of the daily diet for wide 84 85 range of population ages. For instance, soft plain maize porridge is the most common infant food consumed daily by 94 % of the infants in Tanzania [14]. Based on the understanding of 86 87 agricultural practices that utilizes some phosphate fertilizers and food consumption patterns 88 in mentioned areas in Tanzania, understanding the activity levels for maize crop is wealthy.

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Therefore, the aim of this study is to determine the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in maize from selected regions in Tanzania and to estimate the internal exposure due to ²³²Th and ²³⁸U intakes to individuals (> 17 years) due to maize consuming. The effective dose will also be calculated and compared to the ICRP and UNSCEAR values.

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95 2. Material and Methods

96 2.1 Selection of Sampling sites

97 The sampling sites were identified basing on their characteristics (Figure 1). In Manyara 98 region, maize was sampled from Minjingu village where there is the biggest phosphate 99 deposit and the only active phosphate mine in Tanzania [8]. In Ruvuma region, maize was 100 sampled from Likuyu village, this village is situated about 54 km east of the Mkuju uranium 101 deposit [6]. In Dodoma, maize sample were collected at Bahi Wetlands which is situated 102 about 60 km north-west of the capital Dodoma.



Figure.1. Map of Tanzania showing the discovered Phosphate deposits (the dots) and
 Uranium (Blocks A-G) occurrences in different geological environment. The inset
 shows its relative position in African Continent.

Table 1. Shows block name, Locations and Area coverage of discovered Uranium 112 occurrences in different geological environment in Tanzania.

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Block Name	Deposits Locations	Occurrences Area (Sq. km)
Α	Mkuju, Madaba	135,000
В	Isuna, Bahi, Makutupora	37,000
С	Ndala, Igombe, Kigoma	142,000
	Ugalla River, Mpanda	
D	Minjingu, Gallapo	13,000
E	Monduli, Tarosero	2,000
F	Chimala, Panda, Njombe	19,000
G	Bukoba, Biharamulo	10,000

Through the survey it was revealed that maize available in the city markets of Tanzania were mostly supplied from Mbeya [11]. Therefore, a fourth batch of maize samples was from Mbeya (Mbozi and Usangu), as it was one of the major maize growing area in the country and also uses the phosphate fertilizers from Minjingu [11]. Moreover, Dar es Salaam region, the capital city imports maize originate from several areas in Tanzania, maize samples were collected from different market and mills at Tandika, Manzese and Gongolamboto in Dar es Salaam region, the business city in Tanzania.

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123 2.2 Sample collection and preparation

124 Sampling was carried out between March and June, 2015 in all five regions. A total of 80 125 maize samples were collected. 20 samples from Miniingu (Manyara), 15 samples from 126 Likuyu (Ruvuma), 15 samples from Bahi (Dodoma) and 15 samples from Mbozi and Usangu 127 (Mbeya), and 15 samples from Dar es Salaam. Maize was collect from farms in Minjingu and 128 Likuyu villages. However, in Mbeya and Dar es Salaam city, maize samples were collected 129 from markets and milling areas. Maize samples were prepared as for cooking which include 130 washing. Samples were sun dried for 2 days and then oven dried at 45 - 50 °C for 48 hours. 131 They were then crushed into small grains using mortar and pestle, and by using MonoMill 132 Pulverrizer, the samples were pulverized into powder, then sieved to reduce particle size to 133 the recommended size of (< 50 μ m) [2], and then dried in the desiccator for one week while 134 several series of weight measurement were taken to note when constant weights in samples 135 were achieved. Finally, 100g of each sample was packed into cylindrical stainless steel 136 canister to a height of 1.8 cm, sealed using glycerin and wrapped by using gas tightness insulation tape to avoid escape of radon gas. Samples were stored for more than 21 days to 137 allow attainment of radioactive equilibrium stage between ²²⁶Ra and its short lived decay 138 139 products

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141 2.3 Instrumentation

A lead shielded vertical coaxial (n-type) HPGe detector of relative efficiency of 51 % and 142 resolution of 1.8 keV at 1332 keV (from ⁶⁰Co source) was used for low level counting of 143 144 samples. The HPGe detector was well housed reduce background in three layers of copper, 145 cadmium and lead of 30 mm, 3mm and 100 mm respectively. The detector was connected to 146 a Digital Spectrum Analyzer; DSA100 with built in Multichannel analyzer (MCA) and employed genie 2000 software for analysis. The energy and efficiency calibrations of the γ -147 spectrometry system were performed using five standard radioactive sources (¹⁵⁵Eu, ¹³⁷Cs, 148 ⁶⁰Co, ²²Na and ⁴⁰K). The descriptions of the gamma spectrometry system as well as more 149 150 details on the calibrations and the analytical methods used into this study are well 151 documented elsewhere [2, 15].

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153 An empty cylindrical steel canister with the same geometry as that of the sample was 154 used as background activities of radionuclides as well as their minimum detection limits. The 155 counting time for accumulating spectral for both the samples and background was set at 156 28,800s. However, in order to check the stability of the system, each container was counted 157 twice. The detection limits of the detector for the concentration of radionuclides were 158 determined as documented elsewhere [16]. The detection limits for 295.21 keV (1.23) 159 Bq/kg), 351.92 keV (0.69 Bq/kg), 609.31 keV (0.95 Bq/kg), 1120.29 keV (3.17 Bq/kg), 160 338.32 keV (0.96 Bq/kg), 911.60 keV (3.61 Bq/kg), 969.11 keV (0.74 Bq/kg), 583.19 keV 161 (1.96 Bg/kg), 860.50 keV (2.45 Bg/kg) and 1460.81 keV (18.10 Bg/kg). The radioactivity 162 concentrations below these values have been taken to be below the minimum detection limit 163 (BDL).

164 The specific radioactivity in maize samples were estimated by taking the mean of 165 specific radioactivities obtained from the γ - ray lines from their daughters. ²³⁸U activity 166 concentration in maize samples were determined via its daughters through intensities of the 167 ²¹⁴Pb (295.21 keV and 351.92 keV) and ²¹⁴Bi (609.31 keV and 1120.29 keV) γ lines, 168 respectively. ²³²Th activity concentration were obtained through ²²⁸Ac (338.32 keV, 911.60 169 keV and 969.11 keV) and ²⁰⁸Tl (583.19 keV and 860.50 keV) γ lines, while that of ⁴⁰K was 170 determined directly by the γ - line of energy 1460.81 keV.

For quality assurance, the IAEA Soil 375 standard reference material (SRM) weighed (164 g) in the same method as the samples and packed in cylindrical stainless steel canister at a height of 1.8 cm. As Table 2 shows, the experimental values agreed well with the recommended values approximately within \pm 7.5 % accuracy for ²³⁸U, \pm 10.7 % for ²³²Th and \pm 10 % for ⁴⁰K.

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Table 2: The Standard Reference Values and Experimental Values of the IAEA Reference
 Soil 375 [17]

Radionuclide	Experimental Activity	Certified Value	% Deviation from Certified Value (±)
²³⁸ U	21.5	20.0	7.5
²³² Th	22.7	20.5	10.7
⁴⁰ K	466.4	424	10.0

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181 2.4 Effective dose estimation

The committed effective dose due to the intake of radionuclides through maize was
 calculated using the equation below.

 $E = D_{ing} F C_i$

Where, *E* is the annual committed effective dose from consumption of nuclide *i* in maize (mSv/year), *C_i* is the concentration of radionuclide *i* in maize at the time of consumption (Bq/kg), *F* is the maize consumption rate among Tanzanians (kg/year), and D_{ing} is the dose coefficient for ingestion of radionuclide *i* (mSv/Bq) given by ICRP (1996), which varies with both radionuclides and the age of individuals. The total committed effective dose (*E*) to an individual from radionuclide *i* in maize was established by summing contributions from all nuclides (²³⁸U and ²³²Th) present in maize.

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193 **3. Results and Discussion**

194 *3.1 Radioactivity in maize*

The activity concentrations due to ²³⁸U, ²³²Th and ⁴⁰K in the maize samples collected 195 196 in all five regions in Tanzania are presented in Table 3. The results show that the activity 197 levels of radionuclide is higher for samples collected from Minjingu areas, Manyara Regions where the phosphate mining and factory for phosphate fertilizers is located. Followed on the 198 199 list are samples from Mbeya Regions which can be contributed by use phosphate fertilizers 200 from Minjingu phosphate fertilizers factory. The activity levels of radionuclides in maize from 201 Bahi in Dodoma region occupied a third place while that of Ruvuma in Likuyu village 202 occupied a fourth palace. The values in Bahi and Likuyu village are low because the uranium 203 deposit is not in operation, and hence this result can be used as baseline data. 204

The observed low activity levels for samples from Dar es Salaam markets may be due to the facts that the sample collected could be supplied from other regions where the

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use of fertilizers is low. The average activity levels of these radionuclides in maize were
 found to be compared to those of maize from Nigeria [12, 18], India, Bangladesh, Iran and
 Brazil [19, 20, and 21]. As expected, the activity concentration of ⁴⁰K was higher in all the
 maize samples than the activities of ²³⁸U and ²³²Th.

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 Table 3: Radioactivity concentration (Bq/kg) in Maize samples collected from five

 Regions in Tanzania

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	Activity concentration ($Bq/kg \pm SEM$)			
	²³⁸ U	²³² Th	⁴⁰ K	
MANYARA	< 21.1 – 31.8 > ^a	< 28.7 - 64.1 >	< 260.7 – 548.4 >	
(Minjingu village)	23.6 ± 0.7 ^b	38.9 ± 1.0	434.6 ± 18.7	
MBEYA	< 6.6 - 46.2 >	< BDL – 23.7 >	< 43.2 - 88.0 >	
(Usangu and Mbozi)	16.4 ± 0.2	6.0 ± 0.1	55.6 ± 0.2	
DODOMA	< 1.2 - 4.9 >	< 7.9 - 19.8 >	< 13.8– 38.7 >	
(Bahi Waterlands)	2.4 ± 0.3	5.6 ± 0.2	26.5 ± 4.5	
RUVUMA	< 2.3 - 6.7 >	< 8.6 - 15.2 >	< 18.9 – 44.4 >	
(Likuyu Village)	3.2 ± 0.2	5.0 ± 0.3	32.5 ± 7.2	
DAR ES SALAAM	< BDL – 22.4 >	< BDL - 8.8 >	< 32.1 – 50.8 >	
(Markets and Mills)	1.8 ± 0.2	2.2 ± 0.1	42.0 ± 0.4	

215 216 a: represents the range of radioactivity concentration

b: represents the average radioactivity concentration

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218 *3.2 Effective dose due to ingestion*

219 Effective dose is a useful concept that enables the radiation doses from different 220 radionuclides and from different types and sources of radioactivity to be added. The impact 221 of internal dose risk of the population in Tanzania due to consumption of maize was 222 estimated using the average maize consumption rate (kg/y) and dose coefficients (mSv/Bq), as reported in Table 4. The maize consumption rates in this study were found from the 223 survey to be 151.2 kg capita⁻¹ year⁻¹ in Minjingu and 140.6 kg capita⁻¹ year⁻¹ in Mbozi, 224 respectively. On averaging we have 145.9 kg capita⁻¹ year⁻¹, this value was assumed to 225 226 represent the maize consumption rate in areas of the study.

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Total annual committed effective doses due to ²³²Th and ²³⁸U intakes as a result of 228 consumption of maize in Tanzania are 1.46 mSv/y in Manyara, 0.31 mSv/y in Mbeya, 0.21 229 mSv/y in Dodoma, 0.19 mSv/y in Ruvuma and 0.08 mSv/y in Dar es Salam, respectively 230 (Table 4). The main dose contribution is from ²³²Th. The doses estimated in this paper could 231 232 be refined if specific data on consumption rates were available. These values are below the annual dose guideline of 1mSv/y recommended by the ICRP for the general public except 233 234 the dose value from Manyara (Minjingu village). However, the annual dose limit of 1 mSv/y is 235 strictly applied to controlled releases.

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⁴⁰K is an essential biological element distributed throughout the body and its
 concentration in human tissue is under metabolic (homeostatic) control. Thus the levels in

humans are not normally affected by variations in the environmental levels and as a result its

- radiation dose within the body remain constant [22].
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242 **Table 4:** Annual committed effective dose value due to ²³⁸U and ²³²Th intake in Maize

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Effective dose (mSv/y \pm SEM) REGION (Area) ²³⁸U ²³²Th Total dose MANYARA 0.155 ± 0.005 1.305 ± 0.034 1.460 ± 0.039 (Minjingu village) MBEYA 0.108 ± 0.001 0.201 ± 0.003 0.309 ± 0.004 (Usangu and Mbozi) DODOMA (Bahi Waterlands) 0.016 ± 0.002 0.188 ± 0.007 0.204 ± 0.009 **RUVUMA** 0.021 ± 0.001 0.168 ± 0.010 0.189 ± 0.011 (Likuyu village) DAR ES SALAAM 0.012 ± 0.001 0.074 ± 0.003 0.086 ± 0.004 (Markets and Mills)

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245 4. Conclusion

The radioactivity concentration of ⁴⁰K, ²³⁸U and ²³²Th in maize from high background 246 radiation are in Tanzania has been estimated and discussed. The comparison was made 247 248 with radioactivity of maize around the globe. The annual committed effective dose due to ingestion of maize in Tanzania was also estimated by using the radioactivity concentrations. 249 The annual committed effective doses due to ²³²Th and ²³⁸U intakes are below annual dose 250 251 guideline (1 mSv/y) for the general public from naturally occurring radionuclides [23], except 252 the dose value from Minjingu in Manyara region. However, they are considered to be sufficiently low to result in negligible harmful effects. Further studies on other types of foods 253 and drinking water, as well as the dose contributions from ²²³Ra and its daughters are 254 255 recommended.

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