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Radioactivity levels of staple foodstuffs and dose estimates for most of the Tanzanian population

N A Mlwilo[1], N K Mohammed and N M Spyrou

Department of Physics, University of Surrey, Guildford GU2 7XH, UK

E-mail: mlwilon@tlabs.ac.za

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Abstract

Levels of natural radioactivity in staple food products (maize and rice) from various localities of Tanzania have been studied. The average activity concentrations of ^{40}K, ^{232}Th and ^{238}U in maize were 48.79 ± 0.11, 4.08 ± 0.01 and 13.23 ± 0.10 Bq kg^{-1}, respectively. In rice the concentrations of ^{40}K, ^{232}Th and ^{238}U were 24.67 ± 0.03, 3.82 ± 0.02 and 5.02 ± 0.02 Bq kg^{-1}, respectively. ^{137}Cs was detected in only one sample collected in Zanzibar. The sample, with activity concentration of 5.57 ± 0.01 Bq kg^{-1}, had been imported from Thailand. The relatively high average concentrations of the radionuclides in maize compared to rice may be attributed to the extensive use of phosphate fertilizers in maize production in Tanzania. Total annual committed effective doses due to total ^{232}Th and ^{238}U intakes as a result of consumption of staple foodstuffs for infants, children and adults were 0.16, 0.29 and 0.36 mSv y^{-1}, respectively, which are lower than the annual dose guideline for the general public.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The naturally occurring radionuclides in food come mainly from natural isotopes of potassium, uranium and thorium and their daughter products. The majority of radionuclides in the environment are present as daughter products of ^{232}Th and ^{238}U, distributed by natural geological and geochemical processes, in addition to the unrelated naturally occurring ^{14}C and ^{40}K (Olomo 1990). There are many human activities which can enhance the level of naturally occurring radioactivity levels in the environment. The use of radioactive materials in modern research and fallout from historic weapons testing have released radionuclides into the

[1] Address for correspondence: Physics Group, iThemba LABS, PO Box 722, Somerset West 7129, South Africa.

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global environment. Enhancements of naturally occurring radionuclides into the environment are made through the burning of fossil fuels and the uncontrolled mining processes (Banzi *et al* 2000). As would be expected, those radionuclides accumulated in arable soil are incorporated metabolically into plants and ultimately can be transferred into the bodies of animals (including humans) when contaminated foods are consumed. Naturally occurring radionuclides and their associated health risks have not been studied to the same extent as their artificial counterparts, but some comprehensive investigations have been carried out in various parts of the world.

Previous studies have reported significant levels of ^{210}Pb and ^{210}Po in terrestrial plants and other environmental samples by using beta counters and alpha spectrometry techniques (Bolca *et al* 2006, Gasco *et al* 2002, Pietrzak-Flis and Skowronska-Smolak 1995). However, the detection of these radionuclides in the samples analysed in this study is limited by the gamma-ray spectrometry technique used. The main source of these radionuclides in the environment is the decay of ^{222}Rn gas evolved from the ground into the atmosphere. These radionuclides deposit on the ground in association with aerosols via washout and sedimentation. Several studies have revealed that uptake of ^{210}Pb and ^{210}Po into plants can occur both indirectly through the root system and via direct deposition from the atmosphere (Hill 1960, Lasinskaya *et al* 1973, McDowell-Boyer *et al* 1980, Parfenov 1974, Popova and Taskaen 1980). However, the interception and retention of radionuclides on plant surfaces will vary according to the physical and chemical form of the deposit, the meteorological conditions at the time of deposition, the type of vegetation and the stage of its growth, and the method of their preparation before consumption (Chamberlain 1970, Chamberlain and Garland 1991). If deposition occurs in sufficiently heavy rain to cause appreciable runoff from leaves, retention will be less than after light showers (Ewers *et al* 2003). In this study, the contributions of ^{210}Pb and ^{210}Po to activity levels, and thereafter effective dose to consumers as a result of their direct deposition on the two food crops, are considered not significant due to the fact that the two food crops analysed require frequent rain feeding or irrigation during farming and at the same time post-harvest preparation includes removing crop outer cover and washing. Hence, this study is going to provide the basic information on levels of ^{40}K, ^{232}Th and ^{238}U radionuclides via root uptake.

In Tanzania, the main source of contamination in soil is through natural radioactivities. Extensive use of phosphate fertilizers in agriculture might contribute significantly to activity levels of farm soils and then to food crops via root uptake. These fertilizers are manufactured from phosphate ore, which are reported to contain potentially relatively high concentrations of uranium, thorium, radium and their daughter products depending on where they originate (Barisic *et al* 1992). Tanzania has several deposits of phosphate rocks, which are used directly in farming as fertilizers: the rocks from Minjingu in the northern region are exported or used internally to manufacture phosphate fertilizers. So far, research into the levels of natural radioactivity have been undertaken only at the Minjingu phosphate mines. The results have shown relatively high concentrations of uranium (480–1100 ppm), in contrast to many other rocks found in the world (Banzi *et al* 2000, Makweba and Holm 1993, Ogunleye *et al* 2002). This research has also shown activities of ^{238}U and its daughter products in the edible leaf vegetation, surface water and chicken feed. The fertilizers manufactured from Minjingu phosphate rocks are also reported to have high activities of ^{40}K, ^{232}Th and ^{238}U (Makweba and Holm 1993). These fertilizers, direct use of phosphate rocks and other phosphate fertilizers imported from other countries which are commonly used in the cultivation of maize may introduce radioactivity into the soil and the food chain.

Human organs are known to exhibit varying degrees of radiosensitivity to radionuclides. Following ingestion or inhalation the metabolism of radionuclides in the body and the organs will alter its uptake into that organ and that, combined with the different radiosensitivities, will

produce different doses to different organs from the same contaminated food source (Cothern *et al* 1986). For instance, ^{137}Cs is very important because it follows the course of potassium in ecosystems and it persists in the environment for many years due to its relatively long half-life, 30.2 years. Furthermore, it is characterised as a potential genetic hazard because it accumulates in many types of human tissues and its penetrating gamma-rays reach all body cells (Whicker and Schultz 1982).

The International Commission on Radiological Protection (ICRP) and the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) through their reports have recommended maximum quantities of various radionuclides within the body and maximum permissible concentrations of nuclides in air and water.

The scope of this study is to identify radionuclides present in staple foodstuffs with the aim to establish their baseline activity concentrations in the products and eventually assess internal exposure due to ^{232}Th and ^{238}U intakes. The effective dose will also be calculated and compared to the ICRP and UNSCEAR values.

2. Materials and methods

2.1. Sampling procedure

Sampling was carried out between May and June 2005, based on a thorough understanding of agricultural practices and food consumption habits in selected areas of interest in Tanzania. Two staple food products (maize and rice) were identified as those being consumed at the highest rates and subsequently forming the most important part of the daily diet for a wide range of population ages. In Tanzania, soft plain maize porridge is the most common infant food consumed daily by 94% of the infants (Poggensee *et al* 2004, Tatala *et al* 1998).

It was also identified from the survey that the staple foods available in the city markets were supplied from different regions of the country. Maize sampling was done in Mbeya region (Mbozi, Mbalizi and Usangu), as it was the main supplier of maize to the country whereas rice was sampled in Zanzibar, Ifakara, Kahama and Mbeya regions (see figure 1). Moreover, representative samples said to originate from the same cultivation areas were randomly collected from different markets and mills in Dar es Salaam, the capital city.

2.2. Sample preparation

The fresh masses of samples were recorded in order to normalise the units of consumption rates (kg y^{-1} relative to fresh mass) and activity concentrations (Bq kg^{-1} relative to dry mass). The samples were then ground after drying at room temperature, sieved through a 100 mesh (149 μm) and then dried in the desiccator for one week while several series of weight measurements (using a weighing balance) were taken to note when constant weights in samples were achieved. After preparation, each sample was packed in a 400 ml plastic container, sealed, weighed by electronic balance ($\pm$0.001 g) and then coded according to its origin or brand name.

The samples were stored for more than 21 days to allow attainment of the radioactive secular equilibrium stage between ^{226}Ra, ^{228}Ra and its short-lived decay products (Bruzzi *et al* 1997, Valkovic 1993).

2.3. Gamma-ray spectrometry

A lead-shielded (100 mm) coaxial HPGe detector of resolution 2.62 keV for 1332 keV ^{60}Co gamma-ray source and relative efficiency of 17.5% was used for low background counting of the samples. The background spectra obtained from an empty bottle counted for 24 h was



Figure 1. A map of Tanzania showing sampling locations.

used to find the background activities of the radionuclides as well as their minimum detection limits. Each sample was then counted for the same time (24 h), in which energy calibration was repeated before each sample counting. The energy and efficiency calibrations of the HPGe spectrometer system and the analytical method used for this study are well documented elsewhere (Mobius 1988, IAEA 1989).

^{238}U activity concentrations of the samples were determined via its daughters (^{214}Pb and ^{214}Bi) through the intensities of the 295 keV and 609.3 keV γ lines, respectively. ^{232}Th activity concentrations were obtained through ^{228}Ac and ^{208}Tl emissions at 338 keV and 860 keV, respectively, whereas ^{40}K activity concentrations were determined by 1461 keV emissions.

The detection limits (DL) of the detector for the concentrations of the radionuclides in the samples were determined at 3σ using the procedure detailed elsewhere (Knoll 1999). The detection limits for 295 keV, 338 keV, 609.3 keV, 860 keV and 1461 keV γ lines in maize samples were 0.14 Bq kg^{-1}, 0.18 Bq kg^{-1}, 0.27 Bq kg^{-1}, 0.01 Bq kg^{-1} and 2.50 Bq kg^{-1}, respectively, whereas in rice samples they were 0.18 Bq kg^{-1}, 0.23 Bq kg^{-1}, 0.35 Bq kg^{-1}, 0.02 Bq kg^{-1} and 3.0 Bq kg^{-1}, respectively. The detection limit of ^{137}Cs (661 keV) in rice samples was 0.02 Bq kg^{-1}. Concentration values below these detection limits have been taken in this work to be below the minimum detection limit (BDL).

3. Results and discussion

The results for the activity concentrations due to ^{40}K, ^{232}Th and ^{238}U are presented in tables 1, 2 and 3. Whenever the concentration of the radionuclide in the sample was below the DL of the method, a mean value equal to one-half of the DL was used when calculating the average activity concentration instead of missing data. This is similar to the approach used by Ndengerio-Ndossi and Cram (2005).

Table 1 shows the concentrations of radionuclides detected in maize samples collected in Mbeya region. The concentrations of ^{238}U ranged from 6.61 $\pm$ 0.02 to 46.17 $\pm$ 0.03 Bq kg^{-1} with an average of 16.38 $\pm$ 0.01 Bq kg^{-1}. The activity concentrations of ^{232}Th ranged from

Table 1. Concentrations of ^{40}K, ^{232}Th and ^{238}U radionuclides detected in maize samples collected in the Mbeya region (dry mass).

	Activity concentration (Bq kg^{-1} ± SD)		
Sample	^{40}K	^{232}Th	^{238}U
Mbozi 1	88.01 ± 0.89	12.07 ± 0.12	19.98 ± 0.02
Mbozi 2	43.42 ± 0.04	BDL[a]	6.75 ± 0.01
Mbalizi 1	42.58 ± 0.03	BDL	6.61 ± 0.01
Mbalizi 2	45.41 ± 0.05	BDL	12.11 ± 0.02
Usangu 1	43.22 ± 0.01	23.71 ± 0.04	6.71 ± 0.01
Usangu 2	70.83 ± 1.06	BDL	46.17 ± 0.03

[a] Below minimum detection limit.

Table 2. Concentrations of ^{40}K, ^{232}Th and ^{238}U in maize samples collected in the Dar es Salaam region (dry mass).

	Activity concentration (Bq kg^{-1} ± SD)		
Sample	^{40}K	^{232}Th	^{238}U
Kasuku 1	41.62 ± 0.05	BDL	12.95 ± 0.05
Kasuku 2	50.84 ± 1.50	BDL	22.39 ± 0.79
Neema 1	43.42 ± 0.04	BDL	BDL
Neema 2	32.09 ± 0.01	8.80 ± 0.01	4.99 ± 0.01

BDL to 23.71 ± 0.04 Bq kg^{-1} with an average of 5.96 ± 0.02 Bq kg^{-1} and ^{40}K ranged from 43.22 ± 0.01 to 88.01 ± 0.89 Bq kg^{-1} with an average of 55.58 ± 0.23 Bq kg^{-1}.

Table 2 shows the concentrations of radionuclides detected in maize samples collected in Dar es Salaam region. The activity concentrations of ^{238}U ranged from BDL to 22.39 ± 0.79 Bq kg^{-1} with an average of 10.08 ± 0.20 Bq kg^{-1}. The concentrations of ^{232}Th ranged from BDL to 8.80 ± 0.01 Bq kg^{-1} with an average of 2.2 ± 0.01 Bq kg^{-1}. ^{40}K concentrations ranged from 32.09 ± 0.01 to 50.84 ± 1.50 Bq kg^{-1} with an average of 42.0 ± 0.4 Bq kg^{-1}. The average activity levels of these radionuclides in maize were found to be comparable to those of white corn from Nigeria (Olomo 1990).

The concentrations of radionuclides in rice samples collected from different regions of the country are presented in table 3. The concentrations of ^{238}U and ^{232}Th ranged from BDL to 30.41 ± 0.02 Bq kg^{-1} (with an average of 5.02 ± 0.02 Bq kg^{-1}) and 8.80 ± 0.01 Bq kg^{-1} (with an average of 3.82 ± 0.02 Bq kg^{-1}), respectively. The concentrations of ^{40}K in rice samples ranged from 3.55 ± 0.01 to 30.89 ± 0.02 Bq kg^{-1} with an average of 24.67 ± 0.03 Bq kg^{-1}. ^{137}Cs, at a concentration of 5.57 ± 0.01 Bq kg^{-1}, was detected in only one sample and that was one of the two collected in Zanzibar. This rice is not cultivated in Tanzania but imported from Thailand, which might be contaminated as a result of atmospheric weapon tests or the Chernobyl accident. However, except for the ^{137}Cs the average concentrations of other radionuclides are comparable to the values reported in a study on natural radionuclides in rice conducted in Nigeria (Arogunjo *et al* 2005).

The mean activity concentration of ^{238}U in maize is approximately greater by a factor of 2 to 3 than its concentration in rice samples, whereas the concentration of ^{40}K in maize is greater than in rice by a factor of 2. The presence of ^{40}K in all samples at relatively higher concentration levels compared to ^{232}Th and ^{238}U is due to its presence in soils, rocks and various organic materials.

The main pathway for radionuclides into the food chain is through plant uptake from the soil. As a general rule, radionuclides present in soil pass into the root system in the same

Table 3. Concentrations of ^{40}K, ^{137}Cs, ^{232}Th and ^{238}U in rice samples collected from different regions in Tanzania (dry mass).

	Activity concentration (Bq kg^{-1} ± SD)			
Sample	^{40}K	^{137}Cs	^{232}Th	^{238}U
Mbozi 1	23.29 ± 0.09	BDL	1.39 ± 0.16	1.68 ± 0.20
Mbozi 2	23.29 ± 0.08	BDL	2.12 ± 0.01	3.81 ± 0.01
Kahama 1	3.55 ± 0.01	BDL	BDL	BDL
Kahama 2	32.09 ± 0.01	BDL	8.80 ± 0.01	4.99 ± 0.01
Ifakara 1	29.26 ± 0.08	BDL	3.03 ± 0.17	BDL
Ifakara 2	29.26 ± 0.08	BDL	8.28 ± 0.01	3.43 ± 0.01
Usangu 1	21.89 ± 0.02	BDL	1.79 ± 0.02	1.45 ± 0.05
Usangu 2	25.81 ± 0.02	BDL	1.79 ± 0.02	BDL
Zanzibar 1	27.51 ± 0.02	5.57 ± 0.01	7.55 ± 0.01	4.27 ± 0.01
Zanzibar 2	30.89 ± 0.02	BDL	3.47 ± 0.01	30.41 ± 0.02

Table 4. Radionuclide contents of the Minjingu ground rock phosphate, phosphate fertilizers and phosphogypsum in Tanzania. (Note: Makweba and Holm (1993).)

	Activity concentration (Bq kg^{-1} ± SD)		
Sample	^{40}K	^{232}Th	^{238}U
Ground rock phosphate (GRP)	286	698	5022
Triple superphosphate (TSP)	362	444	3116
Superphosphate (SSP)	491	433	3394
Phosphogypsum	41	—	3219

manner as non-radioactive isotopes of the same cations. The element may or may not be required for normal metabolism, and some elements like iodine, cobalt, uranium and radium are known to be present in plants although they serve no known metabolic function (Eisenbud 1963).

Phosphate rocks used world-wide in the manufacturing of phosphate fertilizers, phosphoric acid and other chemicals, are known to contain relatively high levels of uranium compared to other environmental materials and, as such, are one of the sources of technologically enhanced natural radiation (TE-NORM), which contribute to radioactivity levels in soils (Schmidt 1993). The direct application of ground rock phosphate has been in practice in many parts of the world. In Tanzania, the ground rock phosphate with ^{238}U and its progenies from the Minjingu phosphate mine has been applied at rates of 3×10^4–6×10^4 kg km^{-2} (Makweba and Holm 1993). Tanzania uses phosphate fertilizers extensively in maize cultivation and rarely in rice farming because rice is grown in wetlands, which do not necessarily need the application of fertilizers. Makweba and Holm (1993), reported the activity concentrations in ground rock phosphate from Minjingu, Tanzania and fertilizers manufactured from this rock (see table 4). These activities are higher than the values from phosphate rocks reported elsewhere (Ogunleye *et al* 2002, Shukla *et al* 1982). On the other hand, the average activity concentrations of Tanzanian soils were found to be 350.4 ± 18.3, 24.7 ± 2.2 and 34.2 ± 2.0 Bq kg^{-1} for ^{40}K, ^{232}Th and ^{238}U, respectively, (Banzi *et al* 2000). These levels are similar to the world average activities reported by Tyka (Tykva 1995). Therefore, the relatively high average concentrations of ^{238}U in maize samples compared to rice samples shown in figures 2 and 3, could be attributed to the usage of phosphate fertilizers in the cultivation of maize.

The impact of internal dose risk of the population in Tanzania due to consumption of maize and rice was estimated using the average consumption rates assumed (kg y^{-1}) of grain products

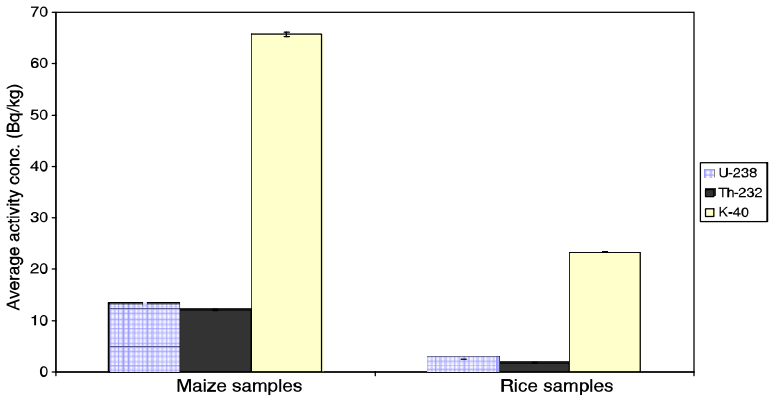


Figure 2. Average activity concentrations of maize and rice samples collected in Mbozi (dry mass).

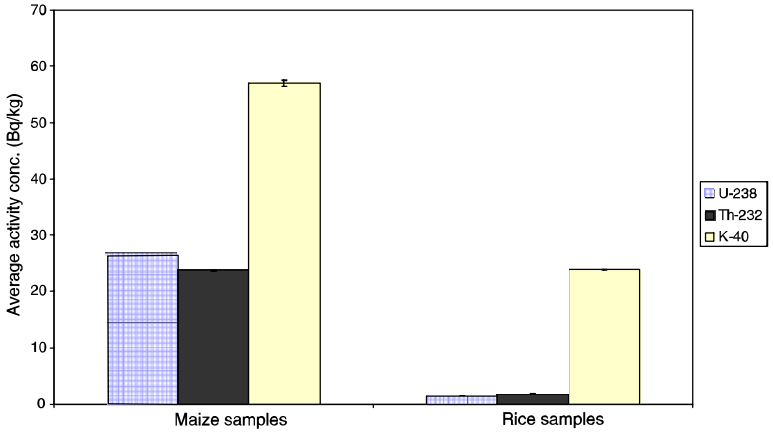


Figure 3. Average activity concentrations of maize and rice samples collected in Usangu (dry mass).

and dose coefficients (mSv Bq^{-1}). The consumption rates for infants, children and adults are 45 kg y^{-1}, 90 kg y^{-1} and 140 kg y^{-1}, respectively, based on fresh masses (ICRP 1996, Sabol *et al* 1995, UNSCEAR 1982). Hence, the masses used to calculate activity concentrations were normalized by a factor to obtain absolute mass units similar to the consumption rates.

Total annual committed effective doses due to ^{232}Th and ^{238}U intakes as a result of consumption of staple foodstuffs for infants, children and adults were 0.16, 0.29 and 0.36 mSv y^{-1}, respectively (table 5). The main contribution is from ^{232}Th, in which for infants, children and adults are 63%, 62% and 68%, respectively. The contributions of maize in annual committed effective dose for infants, children and adults are 56%, 59% and 58%, respectively. These values are well below the annual dose guideline of 1 mSv recommended by the ICRP for the general public from naturally occurring radionuclides (figure 4). However, the annual dose limit of 1 mSv is strictly applied to controlled releases.

^{40}K 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

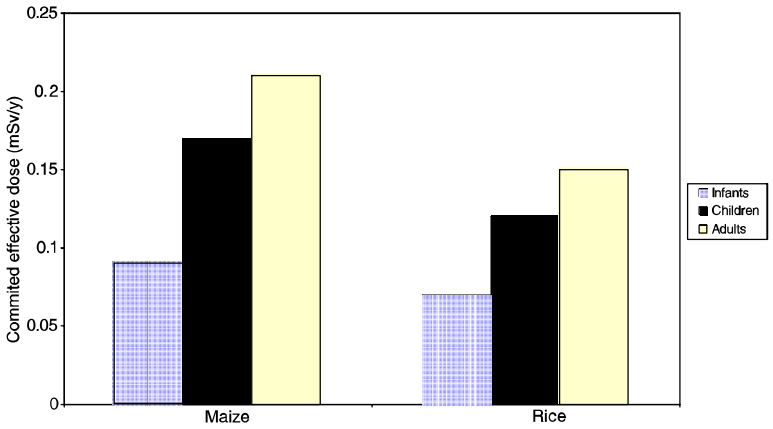


Figure 4. Committed effective dose to each age group from ^{232}Th and ^{238}U intakes in maize and rice.

Table 5. Annual committed effective dose values due to ^{232}Th and ^{238}U intakes for infants, children and adults.

	Committed effective dose (mSv y^{-1}) $\pm$ 0.01					
	Infants		Children		Adults	
Foodstuffs	^{232}Th	^{238}U	^{232}Th	^{238}U	^{232}Th	^{238}U
Maize	0.05	0.04	0.09	0.08	0.13	0.08
Rice	0.05	0.02	0.09	0.03	0.12	0.03
Total	0.10	0.06	0.18	0.11	0.25	0.11

not normally affected by variations in the environmental levels and as a result its radiation dose within the body remain constant (Ioannides *et al* 1997). An average annual dose of 165 μSv to infants and adults has been estimated from measurements of average body content of this radionuclide (ICRP 1975, UN 2000). The average annual dose in younger age groups (10 years old) is higher, at 185 μSv, due to a higher ^{40}K concentration (UN 2000). Lower ^{40}K concentrations in elderly females give rise to an annual dose about half that of young individuals (ICRP 1975, UNSCEAR 1982). The doses estimated in this paper could be refined if specific data on consumption rates were available.

4. Conclusions

Concentrations of radionuclides in two staple foodstuffs, widely consumed in Tanzania, have been determined. ^{232}Th and ^{238}U were detected in 30% and 90%, respectively, of all maize samples analysed, whereas ^{40}K was detected in all maize samples with higher concentrations compared to other radionuclides. In the rice samples, ^{232}Th and ^{238}U were detected in 90% and 70%, respectively, whilst ^{40}K was detected in all samples analysed. ^{137}Cs concentration of 5.57 $\pm$ 0.01 Bq kg^{-1} was detected in only one sample and that was one of the two collected in Zanzibar, which had been imported from Thailand. Since the contribution of activities of these radionuclides from Tanzanian soil is minor, the higher activities found in maize samples could be associated with the application of phosphate fertilizers in their cultivation.

The annual committed effective doses due to ^{232}Th and ^{238}U intakes are below annual dose guideline (1 mSv y^{-1}) for the general public from naturally occurring radionuclides (IAEA 1996). Further studies on other types of foods and drinking water, as well as the dose contributions from the direct deposition of ^{210}Pb and ^{210}Po, are recommended.

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