DETERMINATION OF RADIOACTIVITY IN MAIZE AND MUNG BEANS GROWN IN THE NEIGHBORHOOD OF MINJINGU PHOSPHATE MINE, TANZANIA

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ABSTRACT

Two staple foods (maize and mung beans) which were cultivated in Minjingu village, where there is phosphate deposit in Tanzania, were collected directly from the farms. The activity concentrations of $^{226}\text{Ra}$, $^{228}\text{Th}$ and $^{40}\text{K}$ were determined in the maize and mung beans samples using $\gamma$ ray spectrometry employing HPGe detector of relative efficiency of 51.0 %. The mean radioactivity level in the food samples were found to be $21.01 \pm 0.8 \, \text{Bq/kg (mung beans)}$, $25.6 \pm 0.7 \, \text{Bq/kg (maize) for } ^{226}\text{Ra}$, $62.6 \pm 1.1 \, \text{Bq/kg (mung beans)}$, $72.9 \pm 1.0 \, \text{Bq/kg (maize) for } ^{228}\text{Th}$ and $542.9 \pm 8.6 \, \text{Bq/kg (mung beans)}$, $434.6 \pm 18.7 \, \text{Bq/kg (maize) for } ^{40}\text{K}$. The radioactivity content of the maize and mung beans from Minjingu village were higher than that of similar food samples collected from Bukombe district in Geita Region in Tanzania. The total annual effective dose for consumption of $^{226}\text{Ra}$ and $^{228}\text{Th}$ by adults was calculated to be $2.003 \pm 0.044 \, \text{mSv/year}$, which is higher than the annual dose limit of $1 \, \text{mSv/year}$ recommended by the ICRP for the general public. Hence a conclusion could be made that food crops cultivated at Minjingu village might expose the population to high radiation dose which might be detrimental to their health.

Key Words: Radioactivity, annual effective dose, Minjingu Phosphate mine, High Background Radiation Area

1. INTRODUCTION

Food crops from contaminated environment may accumulate radioactivity that could form a direct route of exposure to human population when consumed. Therefore, the knowledge of radioactivity levels in grown food crops is very important in order to establish the dose received by populations. The main radioactive environmental contamination in Tanzania are from the natural radioactive materials that could enter the environment either through uncontrolled mining activities or from the use of phosphate fertilizers which are manufactured from phosphate rocks (Banzi et al. 2002). Phosphate rocks are reported to contain radioactivity an order of magnitude higher than normal soil and other rocks (Shukla et al. 1982).

The phosphate rock from Minjingu has shown to have higher activities of $^{238}\text{U}$ ($4641 \, \text{Bq/kg}$) and $^{226}\text{Ra}$ ($5022 \, \text{Bq/kg}$) and $^{232}\text{Th}$ in contrast to many other rocks found in the world (Shukla et al. 1982, Banzi et al. 2002, Ogunleye et al. 2002). Because of that, Minjingu was termed as a high background radiation area in Tanzania (Bianconi 1987, Banzi et al. 2000, 2002). The phosphate in Minjingu is mined to be exported to the neighboring countries as well as be used to manufacture fertilizer for local use. The mine, which is also carrying the processing of
the phosphate is within the village of Minjingu, which is located in semi-arid zone with approximately 11,000 inhabitants (Nkaiti Ward Census 2010). The soil in Minjingu, which might have high activity due to phosphate deposit in the area may as well be contaminated through ore dust resulting from mining process. Therefore, food crops grown in the neighbourhood of the Minjingu phosphate mine may also be contaminated.

The radioactivity level in soil can plausible be used to show the magnitude of contamination in locally grown food crops, but it cannot describe the biological effects of radiation exposure to individuals who consume that food. Therefore the estimation of doses is usually carried out for assessing health safety of an individual undergoing radiation exposure through ingestion of contaminated food. The intake of radionuclide within food is dependent on the concentration of radionuclides in various food crops and on the food consumption rates. The risks associated with an intake of radionuclides in the body are proportional to the total dose delivered by radionuclides while staying in various organs. In general, it is assumed that stochastic effects occur linearly with dose and usually the annual effective dose quantities ($E_{(a)}$) are usually used to define those risks when prolonged exposure to a single individual from a single intake of a radionuclide is being considered. Therefore, the present study determined the specific activities from maize and mung beans grown in the neighbourhood of Minjingu phosphate mine and from the activities the annual effective dose received by population was calculated.

2. Materials and Methods

2.1 Description of the Study Area

The Minjingu phosphate mine in Minjingu village is located to the east of the saline Lake Manyara along the rift valley escamement at Manyara region in the northern Tanzania at latitude $03^\circ 42' 30.9''$ S and longitude $035^\circ 54' 56.3''$ E (Fig.1). The Minjingu village occupies approximately a land area of 24,000 hectares with a population of about 11,000 people (URT 2012). The phosphate rock at Minjingu is mined by the open dry process, a method that can lead to the dispersion of large amounts of dust to the environment. Such emission may give rise to enhanced exposure to naturally occurring radionuclides through air, soil, vegetation and water.

Agriculture, pastoralism and phosphate mining are the land uses development in Minjingu village. Crops cultivated for subsistence living in the area includes maize, mung beans and cowpeas, while for income generation the crops include water melon, cotton and sesame.

2.2 Sample collection and Preparation

In this study, samples were collected in farms within an area covered with a radius of 6.82 km from the Minjingu phosphate mine to the North and West of the village. Maize and mung beans were identified as the main staple foods hence were collected for analysis. A total of 20 samples of maize (mass 1000 g each) and 13 samples of mung beans (mass of 1000 g each) were collected from the farms in different locations within an area covered with a radius of 6.82 km from the Minjingu phosphate mine to the North and West of the village. Two samples (one for maize and the other for mung beans) taken from Bukombe district in Geita Region, which is far from Minjingu area were used as control to make comparison.

Samples were sun dried for 2 days and then oven dried at $45 - 50\, ^\circ C$ for 48 hours (Holynscsa and Jasion 1986). They were then crushed into small grains using mortar and pestle, and by using MonoMill Pulverizer, the samples were...
pulverized into powder, then sieved to reduce particle size to the recommended size of 2 mm (IAEA 1989), and finally the samples were homogenized. The homogeneous powdered samples were packed (100 g for maize and 130 g for mung beans) into cylindrical stainless steel canister to a height of 1.8 cm. They were then sealed using glycerin and wrapped by using gas tightness insulation tape to avoid escape of radon gas and stored for more than 21 days to allow attainment of the radioactive equilibrium stage between $^{226}$Ra and its short-lived decay products (Banzi et al. 2000).

2.3 Selection of $\gamma$ Lines
The activity of $^{226}$Ra was obtained through the peak intensities of $\gamma$ lines of $^{214}$Pb (295.21 keV and 351.92 keV) and $^{214}$Bi (609.31 keV and 1120.29 keV) respectively. The activities of $^{228}$Ac (338.32 keV, 911.60 keV and 969.11 keV), and $^{208}$Tl (583.19 keV and 860.50 keV) were considered to represent the $^{228}$Th activity. And measurement of $^{40}$K activity concentration was determined directly by 1460.81 keV $\gamma$ line emissions.

2.4 Accuracy and Precision of Results
For quality assurance, the IAEA Soil 375 standard reference material (SRM) was 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 1 shows, the

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**Figure:** The map of Minjingu Village showing the sampling sites.
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experimental values agreed well with the recommended values approximately within ± 7 % accuracy for $^{226}$Ra, ±11% for $^{228}$Th and ±10% for $^{40}$K.

3. Results and Discussion

3.1 Radioactivity of Maize samples

The mean values of natural radionuclide activity concentrations in maize samples collected from Minjingu Village are reported in Table 2. Specific activities concentrations of $^{226}$Ra, $^{228}$Th and $^{40}$K are reported in Bq/kg dry weight and the errors are the statistical uncertainties of 20 maize samples.

The radionuclides $^{226}$Ra, $^{228}$Th and $^{40}$K were detected in all samples of maize analysed in this study. As it is seen in Table 2, the mean activity concentration of $^{226}$Ra, $^{228}$Th in maize samples from Minjingu village were higher than the concentration in control sample collected from Bukombe district in Geita Region. These values were higher by factors of 3 and 12 for $^{226}$Ra and $^{228}$Th, respectively, than their concentrations in control sample.

The activity concentrations in maize samples collected from Minjingu village ranged from 21.2 Bq/kg to 31.8 Bq/kg, 64.1 Bq/kg to 79.8 Bq/kg, 260.7 Bq/kg to 548.4 Bq/kg with standard deviations values of 3.0 Bq/kg, 4.4 Bq/kg and 83.8 Bq/kg for $^{226}$Ra, $^{228}$Th and $^{40}$K, respectively. The concentration of radionuclides in the food crops depends on the concentration in the soil (Jibiri et al. 2007). Therefore, low SD values for both $^{226}$Ra and $^{228}$Th in maize samples might indicate evenly distribution of the radionuclides in the soil within a big area of Minjingu involved in this study.

The activity concentrations of all three radionuclides in this study were higher than the concentrations reported in maize samples collected in other areas of Tanzania. The mean activity of $^{226}$Ra was 8 times higher than the mean value reported in maize from Likuyu Village (Mohammed and Mazunga 2013). However, the lower extreme value of the range obtained in this study was similar to the upper value of the range reported in maize from Mbeya cultivated with the extensive applications of phosphate fertilizer from Minjingu (Mlwilo et al. 2007). The activity of $^{228}$Th in samples analysed the present study was higher than its values in maize from other places in Tanzania reported by Mlwilo et al. (2007) and Mohammed and Mazunga (2013) by factors of 15 and 18, respectively. $^{40}$K in maize from Minjingu had higher activity concentration by a factor of 9 than the mean activity in maize sample collected from Mbeya region reported by Mlwilo et al. (2007) and 16 times higher than the value reported from Likuyu village (Mohammed and Mazunga 2013).

Moreover, the mean concentration values for $^{226}$Ra in maize from Minjingu were higher than the value reported in 4 out of 5 literatures surveyed in this work (Asefi et al. 2005, Romilton dos Santos et al. 2005, Jibiri and Abiodun 2012). The mean activity value was 14 times higher than the mean activity reported in maize samples from Southwestern Nigeria (Jibiri and Abiodun 2012). This activity value of $^{226}$Ra in maize from Minjingu was also about 20 times higher than the value reported in maize from Bangladesh, Iran and Brazil (Asefi 2005, Romilton dos Santos et al. 2005). At the same time, the concentration of $^{226}$Ra found in maize samples from Minjingu was lower than the value (34.1 ± 14.2 Bq/kg) reported by Jibiri et al. (2007) in maize samples from a high background area (HBRA) of Bitsichi, Josh Plateau in Nigeria.

3.2 Radioactivity of Mung beans samples

Natural radioactivity concentrations in mung beans samples collected from Minjingu village
are reported in Tables 3. The errors shown in the results are the statistical uncertainties of 13 mung beans samples and are expressed as the standard errors of the mean (SEM).

The radionuclides $^{226}$Ra, $^{228}$Th and $^{40}$K were detected in all 13 samples of mung beans analysed in this work. The activity concentrations in mung beans samples were higher than the concentrations in control sample collected from Bukombe, Geita Region. It was found that the minimum activity value of mung beans sample from Minjingu was higher by factors of 3 and 12 for $^{226}$Ra and $^{228}$Th, respectively, than that of control sample. Whilst the value of $^{40}$K was approximately the same as control samples for mung beans.

Table 4 shows the comparison of concentration obtained in this study with those obtained in food samples inside and outside Tanzania. Minjingu is reported to be High Background Radiation Area (Bianconi 1987). Hence the maize and mung beans cultivated in this area were expected to have elevated concentrations of radionuclides than maize and mung beans cultivated from other regions in Tanzania. However, the mean concentration values for $^{226}$Ra in both maize and mung beans analysed in this study were much lower than the mean value in edible vegetations from Minjingu reported by Banzi et al (2000).

The activity of $^{226}$Ra and $^{228}$Th in mung beans from this study were higher than the activity of beans collected in Likuyu village by factors of 7 and 20, respectively (Mhammed and Mazunga, 2013). In surveyed literature outside Tanzania, concentration of $^{226}$Ra in mung beans collected

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Experimental Activity (Bq/kg)</th>
<th>Certified Values (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{226}$Ra</td>
<td>21.5</td>
<td>20.0</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>22.7</td>
<td>20.5</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>466.4</td>
<td>424</td>
</tr>
</tbody>
</table>

Table 2: The activity concentrations (Bq/kg ± SEM) of $^{226}$Ra, $^{228}$Th and $^{40}$K on maize samples collected from Minjingu Village

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Mean Activity</th>
<th>Minimum Activity</th>
<th>Maximum Activity</th>
<th>Standard Deviation (SD)</th>
<th>Control Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{226}$Ra</td>
<td>25.6 ± 0.7</td>
<td>21.2</td>
<td>31.8</td>
<td>3.0</td>
<td>6.5</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>72.9 ± 1.0</td>
<td>64.1</td>
<td>79.8</td>
<td>4.4</td>
<td>5.3</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>434.6 ± 18.7</td>
<td>260.7</td>
<td>548.4</td>
<td>83.8</td>
<td>346.7</td>
</tr>
</tbody>
</table>

Table 3: The activity concentrations (Bq/kg ± SEM) of $^{226}$Ra, $^{228}$Th and $^{40}$K on mung beans samples collected from Minjingu Village

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Mean Activity</th>
<th>Minimum Activity</th>
<th>Maximum Activity</th>
<th>Standard Deviation (SD)</th>
<th>Control Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{226}$Ra</td>
<td>21.0 ± 0.8</td>
<td>16.2</td>
<td>26.7</td>
<td>2.8</td>
<td>5.5</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>62.6 ± 1.0</td>
<td>53.2</td>
<td>68.2</td>
<td>3.8</td>
<td>4.5</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>542.9 ± 8.5</td>
<td>453.7</td>
<td>574.3</td>
<td>30.8</td>
<td>423.1</td>
</tr>
</tbody>
</table>
from Minjingu were also higher than the means reported elsewhere. Mung beans from Minjingu had activity concentration 2 times higher than
be higher than those reported in the surveyed literature. This might be because the Minjingu phosphate rock has shown higher activities of

Table 4: Activity levels (Bq/kg) of $^{226}\text{Ra}$, $^{228}\text{Th}$, and $^{40}\text{K}$ on maize and beans samples from Minjingu and similar samples from other areas (around the world)

<table>
<thead>
<tr>
<th>Study</th>
<th>Maize</th>
<th>Beans</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ra-226</td>
<td>Th-228</td>
</tr>
<tr>
<td>Average Range Present Study</td>
<td>25.6 ± 3.1  21.2–31.8</td>
<td>72.9 ± 4.3  64.1–79.8</td>
</tr>
<tr>
<td>Mbeya-Tanzania (Mlwilo et al. 2007)</td>
<td>9.7 ± 5  6.2–20.0</td>
<td>17.5 ± 5.5  MDL – 23.0</td>
</tr>
<tr>
<td>Dar es Salaam-Tanzania (Mlwilo et al. 2007)</td>
<td>11 ± 7.2  4.0–22.0</td>
<td>MDL</td>
</tr>
<tr>
<td>Likuyu Village-Tanzania (Mohammed and Mazunga, 2013)</td>
<td>3.2 ± 0.2</td>
<td>5.0 ± 0.3</td>
</tr>
<tr>
<td>Nigeria (Jibiri et al. 2007)</td>
<td>34.1 ±14.2</td>
<td>MDL</td>
</tr>
<tr>
<td>Nigeria (Jibiri et al. 2012)</td>
<td>1.8 ± 0.7</td>
<td>6.6 ± 0.7</td>
</tr>
<tr>
<td>Minjingu edible leaf (Banzi et al. 2000)</td>
<td>393 ± 9</td>
<td>318 ± 2.6</td>
</tr>
</tbody>
</table>

that from Bitsichi, Jos plateau in Nigeria, 39 times higher than that reported from HBRA of the Posos de Caldas Plateau in Brazil (Amaral et al. 1992, Jibiri et al., 2007). The mean concentration of $^{226}\text{Th}$ in maize was found to be higher than the value reported in maize from Likuyu and from Mbeya (Mlwilo et al. 2007).

The natural radioactivity of $^{226}\text{Ra}$, $^{228}\text{Th}$ and $^{40}\text{K}$ in maize and mung beans samples from Minjingu analyzed in this study were found to $^{226}\text{Ra}$, $^{228}\text{Th}$ and $^{40}\text{K}$ in contrast to many other rocks found in the world (Shukla et al. 1982; Banzi et al. 2002; Ogunleye et al. 2002). Banzi et al. (2000) reported concentrations of $^{226}\text{Ra}$ in the Minjingu phosphate rock as 5760 ± 107 Bq/kg. The activity concentrations of $^{40}\text{K}$ in both maize and mung beans were found to be higher compared to the concentrations of $^{226}\text{Th}$ and $^{228}\text{Ra}$ in samples from Minjingu. These values were also above the world range of 40 Bq/kg to 240 Bq/kg of $^{40}\text{K}$ in food crops.
reported by Maul and O’Hara (1989). High concentration of $^{40}$K in the food samples might be attributed to concentrations of $^{40}$K in the soil, which may be associated with the geological properties of the area (Saidou et al. 2010).

The radioactivity in foods depends on many factors such as the concentration of radionuclides in soil, the means of agriculture (such as the use of fertilizer) and the uptake of radionuclides by the plant, which is a dependant of the plant species (Jibiri et al. 2007). There is always variation in the accumulation of radionuclides among different species that grow under the same conditions (Kuo et al. 1997). Hence, the differences in radioactivities in the two foods found in this work might be influenced by the differences in uptake of radionuclides.

3.3 Dose Assessment

In this study, a small survey was conducted to collect a food consumption pattern specifically for Minjingu population in order to estimate the food consumption rates for the selected food crops. The food consumption rates were found to be 151.2 kg capita$^{-1}$ year$^{-1}$ and 11.9 kg capita$^{-1}$ year$^{-1}$ for maize and mung beans, respectively. Therefore, using the annual effective dose per intake (dose conversion factors) provided by ICRP (1996), food consumption rates for Minjingu population the and radioactivity concentration found in the analyzed food samples as input to equation 1, the annual committed effective dose, $E_{(\tau)_{\text{ing},\;p}}$ is the annual committed effective dose from consumption of nuclide $i$ in foodstuff $p$ (mSv/year), $C_{p,i}$ is the concentration of radionuclide $i$ in foodstuff $p$ at the time of consumption (Bq/kg), $F_p$ is the consumption rate for foodstuff $p$ (kg/year), and $D_{\text{ing}}$ is the dose coefficient for ingestion of radionuclide $i$ (mSv Bq$^{-1}$) given by ICRP (1996), which varies with both radionuclides and the age of individuals. The total effective dose to an individual from radionuclide $i$ in food crop $p$ was established by summing contributions from all radionuclides present in food crop.

The total calculated annual effective dose for adults (>17 years) in this paper is 2.003 ± 0.044 mSv/year (Table 5). This dose value is higher than the reported annual effective dose from HBRAs in Asia from the dietary intake of these two radionuclides (Iyengar et al. 2004; Pradyumna et al. 2012). The dose value is also higher than the annual dose limit of 1 mSv/year recommended by the ICRP for the general public. However, high dose value of 2.38 mSv/year was reported by Jibiri et al. (2007) in HBRA of Bitsichi, Josh Plateau in Nigeria. Among the analysed radionuclides $^{226}$Ra was found to be the major contributor to the dose from maize (Table 5). About 93.8 % the total effective dose for adults (age group > 17 years) due to consumption of the two staple foods was contributed by maize.
This is because maize is consumed much more than mung beans. $^{226}$Ra contributed to about 57.7% of the total committed effective dose, whilst $^{228}$Th contributed 42.3% of the total committed effective dose.

The total annual effective dose calculated in this work might be overestimated. This is because; the calculation of food consumption rates assumed no importation of maize and mung beans from outside the Minjingu village. However, in some cases depending on the climatic conditions of a particular year, the harvest might become insufficient to cover for the whole year, and the village had to import food from other areas. Hence, the annual effective dose received by the population from the food crops grown in Minjingu might be variable.

4. Conclusion

The radioactivity concentrations for $^{226}$Ra and $^{228}$Th in analysed maize and mung beans samples from Minjingu were higher than those of control. This might be because Minjingu village is close to the phosphate mine. In this study, $^{226}$Ra is accumulated more in maize than in mung beans cultivated in the same area. The annual effective dose for adults (>17 years) calculated in this paper is 2.003 ± 0.044 mSv/year. This dose value is higher than the reported annual effective dose from HBRAs in Asia from the dietary intake of these two radionuclides (Iyengar et al. 2004, Pradyumna et al. 2012). The dose value is also higher than the annual dose limit of 1 mSv/year recommended by the ICRP for the general public. Hence a conclusion could be made that maize and mung beans cultivated at Minjingu village expose the population to high radiation dose, which might be detrimental to their health.

Table 5: Total annual effective doses from $^{226}$Ra and $^{228}$Th, for Adults of age > 17, calculated using Minjingu food consumption rates.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Effective Dose (mSv/year ± SEM)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Maize</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>1.08 ± 0.03</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>0.79 ± 0.01</td>
</tr>
<tr>
<td>Total</td>
<td>1.87 ± 0.04</td>
</tr>
</tbody>
</table>

Acknowledgements

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