

# Activity concentrations of $^{226}\text{Ra}$ , $^{228}\text{Th}$ , and $^{40}\text{K}$ in different food crops from a high background radiation area in Bitsichi, Jos Plateau, Nigeria

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**Abstract** One of the three goals of the United Nations for sustainable food security is to ensure that all people have access to sufficient, nutritionally adequate, and safe food. Decades of tin mining in the Bitsichi area of the Jos Plateau, Nigeria, have left a legacy of polluted water supplies, impoverished agricultural land, and soil containing abnormally high levels of naturally occurring radioactive elements. In order to ascertain the radiological food safety of the population, different crops that constitute the major food nutritive requirements were collected directly across farmlands in the area. The activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{40}\text{K}$  were determined in the food and soil samples using  $\gamma$ -ray spectrometry. Additionally, in situ gamma dose rate measurements were performed on the farms using a pre-calibrated survey meter. The corresponding activity concentrations in the food crops ranged from below detection limit (BDL) to  $684.5 \text{ Bq kg}^{-1}$  for  $^{40}\text{K}$ , from BDL to  $83.5 \text{ Bq kg}^{-1}$  for  $^{226}\text{Ra}$ , and from BDL to  $89.8 \text{ Bq kg}^{-1}$  for  $^{228}\text{Th}$ . Activity concentrations of these radionuclides were found to be lower in cereals than in tubers and vegetables. As for the soil samples, activity concentrations of these radionuclides varied from BDL to  $166.4 \text{ Bq kg}^{-1}$ , from  $10.9$  to  $470.6 \text{ Bq kg}^{-1}$ , and from  $122.7$  to  $2,189.5 \text{ Bq kg}^{-1}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Th}$ , respectively. Average external gamma dose rates were

found to vary across the farms from  $0.50 \pm 0.01$  to  $1.47 \pm 0.04 \mu\text{Sv h}^{-1}$ . Due to past mining activities, the soil radioactivity in the area has been modified and the concentration level of the investigated natural radionuclides in the food crops has also been enhanced. However, the values obtained suggest that the dose from intake of these radionuclides by the food crops is low and that harmful health effects are not expected.

## Introduction

Geologically, the lithological formations of the Jos Plateau, Nigeria, consist of a basement complex, biotite granite, and new basalts [1, 2]. Occurrence of tin and columbite-ore found in this area is usually associated with greisenized biotite granite. Their abundance led to the mining and exploration activities in the area that commenced in 1904 [3, 4]. A map of the geological formations and mineral distributions found at of the Jos Plateau is shown in Fig. 1. These mining operations lasted for more than 50 years, and it was not before 1974 that the radioactive nature of the tin-ore and its tailings was realized [5, 6]. Before 1974, the tailings were treated as non-radioactive and as such were used in building construction, farming, and industries (furnace blasting). During the milling of the Sn-ore to obtain tin, columbite, monazite, zircon, and ilmenite, including electrostatic and magnetic separations and manual processing methods in the open, dust was usually produced, which was widely dispersed into the environment. Decades of tin mining in the area have left a legacy of polluted water supplies, impoverished

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thematic areas identified for priority action and considered relevant to the mandate of both the International Atomic Energy Agency (IAEA) and the Food and Agriculture Organization is plant, animal, and consumer protection. The primary purpose of this study is, therefore, to determine the activity concentration levels of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Th}$  in different varieties of agricultural food crops grown and consumed by the population of Bitsichi town, which is located in an old tin mining area. This is to ensure that food safety is not compromised and the effective doses due to ingestions are within the specified safety limits. The second objective is aimed at providing basic radiometric data for the judgment of potential radioactive releases, and for setting up of a radiometric control of foodstuffs to support the newly established Nigerian Nuclear Regulatory Authority (NNRA) in their food policy, administration and regulatory functions. This is important since data on radioactivity in food is sparse in Nigeria.

## Materials and methods

### Sample collection

The sampling site for this study is Bitsichi, an old tin mining town. It is located in the Barkin-Ladi local government council of Jos Plateau, Nigeria. In order to ensure representative sampling, the entire area was divided into six equal grids of sampling sites, each about  $3\text{ km}^2$  in size. Samples of different food crops were collected from farmlands across the grids. As the investigated area covered a wide variety of agricultural, local, and urban conditions, it was not possible to collect all the identified varieties at a given site. Effort was, however, made to ensure that at least two samples of each variety were collected across the entire grids. Dose rate measurements were performed on the farmlands with a pre-calibrated survey meter at four cardinal points at a height of 1 m above the ground. Additionally, soil samples were collected to a depth of 150 mm below the ground surface at four points, at those farms where samples of food crops were taken. These soil samples were thoroughly mixed together to form a sample representative for that site. The different types of food samples collected and their food group classification is presented in Table 1. Food group classification was done in consultation with a nutritionist, who provided information on the most important food groups and on typical food products belonging to a specific food group.

### Sample preparation

#### *Soil samples*

At the laboratory, all samples were dried at room temperature for many days until a constant weight was reached, and they were crushed to pass a 2-mm mesh sieve. They were then transferred to uncontaminated empty cylindrical plastic containers of uniform size (60 mm in height, 65 mm in diameter) and were sealed for a period of about 4 weeks. This was done in order to allow for radon and its short-lived progenies to reach secular radioactive equilibrium prior to gamma spectroscopy.

#### *Food samples*

Since the present study focuses on the ingestion of foodstuff grown and consumed by the population in the investigated high background radiation area, only the edible parts of the collected plant samples were considered for preparation and analysis. For instance, the peels of cassava tuber, yam tuber, and cocoyam were all discarded. Similar to the soil samples, all plant samples were air dried until they reached a constant weight, homogenized, transferred to the sample containers, and kept sealed for about 4 weeks.

### Measurement technique

Radioactivity measurements were carried out using a lead-shielded  $76\text{ mm} \times 76\text{ mm}$  NaI(Tl) detector crystal (Model No. 802 series, Canberra Inc.) coupled to a Canberra Series 10 plus Multichannel Analyzer (Model No. 1104) through a preamplifier. The detector had a resolution of about 8% at an energy of 0.662 MeV. This was enough to distinguish the  $\gamma$ -ray energies of interest in the present study. The choice of radionuclides to be detected as a reference was made based on the fact that the NaI(Tl) detector used in this study had a poor energy resolution. Hence, the photons emitted by them would be sufficiently discriminated if their emission probability and their energy were high enough and the surrounding background continuum low enough. Therefore, the activity concentration of  $^{214}\text{Bi}$  (determined from its 1.760 MeV  $\gamma$ -ray peak) was chosen to provide an estimate of  $^{226}\text{Ra}$  in the food samples, while that of the daughter radionuclide  $^{208}\text{Tl}$  (determined from its 2.615 MeV  $\gamma$ -ray peak) was chosen as an indicator of  $^{228}\text{Th}$ .  $^{40}\text{K}$  was determined by measuring the 1.460-MeV  $\gamma$ -rays emitted during the decay of  $^{40}\text{K}$ . Finally,

**Table 1** Food samples collected and their corresponding food group

Food group	Sub-food group/food type
Grains/cereals	Millet (3)
	Maize (4)
	Guinea corn (2)
	Acha (4)
	Dyare (2)
Vegetables	Tuberous—sweet potato (3), Irish potato (4)
	Others—okra (3), tomato (3), pepper (3), garden egg (2), kuca (2)
Tubers	Yam (4)
	Cassava (4)
	Cocoyam (3)
Legumes	Groundnut (3)
	Local bean (2)
	Soya beans (3)

( ) indicates the number of samples collected

$^{137}\text{Cs}$  was also quantified (by looking for the 0.662-MeV  $\gamma$ -ray peak) as this isotope is an indicator for any potential environmental contamination due to nuclear accidents and weapon tests [13].

Both soil and food samples were placed symmetrically on top of the detector and measured for a period of 10 h. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks.

From the net area of a certain peak, the activity concentrations in the samples were obtained using Eq. 1 [14–16]:

$$C(\text{Bq kg}^{-1}) = \frac{C_n}{\varepsilon P_\gamma M_s} \quad (1)$$

where  $C$  is the activity concentration of the radionuclide in the sample given in  $\text{Bq kg}^{-1}$ ,  $C_n$  is the count rate under the corresponding peak,  $\varepsilon$  is the detector efficiency at the specific  $\gamma$ -ray energy,  $P_\gamma$  is the absolute transition probability of the specific  $\gamma$ -ray, and  $M_s$  is the mass of the sample (kg).

Equation 1 can be expressed as [17, 18]:

$$C(\text{Bq kg}^{-1}) = \frac{C_k}{A_k} A \quad (2)$$

where,  $C_k$  is the activity concentration of the radionuclide in a standard reference sample having the same geometry as the investigated sample ( $\text{Bq kg}^{-1}$ ),  $A$  is the net area of the corresponding peak in the sample

spectrum, and  $A_k$  is the net area of the peak in the reference sample spectrum.

The standard reference soil sample used was from Rocketdyne Laboratories, California, USA, which is traceable to a mixed standard gamma source (Ref. No. 48722-356) by Analytic Inc., Atlanta, GA, USA. For the food samples, the reference standard sample was obtained from the IAEA traceable to source Ref No. IAEA-312. The dry mass of the food samples used for analysis varied from 80 to 200 g, while for soil samples a uniform mass of 200 g was used.

The detection limit (DL) of a measuring system describes its operating capability without the influence of the sample. The DL given in  $\text{Bq kg}^{-1}$ , which is required to estimate the minimum detectable activity in a sample, was obtained using Eq. 3 [19]:

$$\text{DL}(\text{Bq kg}^{-1}) = \frac{1.96 \left[ \frac{B}{T} + \text{SD}_b^2 \right]^{\frac{1}{2}}}{k \times \varepsilon \times m} \quad (3)$$

where  $\text{SD}_b$  is the estimated standard error of the net background count rate in the peak,  $T$  is the counting time (s),  $\varepsilon$  is the counting efficiency ( $\text{cps/Bq}$ ),  $m$  is the mass of the sample,  $k$  is the factor that converts cps (counts per second) to Bq, and 1.96 represents the 95% confidence level.

**Table 2** Activity concentrations (dry weight) of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Th}$  measured in the investigated food samples

Food items	$^{40}\text{K}$ ( $\text{Bq kg}^{-1}$ )	$^{226}\text{Ra}$ ( $\text{Bq kg}^{-1}$ )	$^{228}\text{Th}$ ( $\text{Bq kg}^{-1}$ )
<b>Cereals/grains</b>			
Maize	243.2 $\pm$ 21.2	34.1 $\pm$ 14.2	BDL
Millet	144.4 $\pm$ 12.8	4.6 $\pm$ 3.4	BDL
Acha (hungry rice)	BDL	BDL	BDL
Dyare	179.4 $\pm$ 25.3	4.7 $\pm$ 1.1	8.1 $\pm$ 3.2
Guinea corn	85.9 $\pm$ 25.6	5.2 $\pm$ 1.3	7.6 $\pm$ 1.6
<b>Tubers</b>			
Yam	684.5 $\pm$ 40.6	85.5 $\pm$ 10.2	89.8 $\pm$ 6.2
Cocoyam	537.1 $\pm$ 18.1	34.0 $\pm$ 15.1	33.3 $\pm$ 7.2
Cassava	539.6 $\pm$ 21.2	27.4 $\pm$ 9.4	22.2 $\pm$ 5.2
Sweet potato	423.7 $\pm$ 30.8	23.6 $\pm$ 11.1	35.6 $\pm$ 12.3
Irish potato	494.4 $\pm$ 22.1	10.7 $\pm$ 3.6	17.1 $\pm$ 9.8
<b>Vegetables</b>			
Okra	213.0 $\pm$ 19.4	BDL	BDL
Tomato	158.9 $\pm$ 28.9	13.9 $\pm$ 6.4	9.6 $\pm$ 4.1
Pepper	132.4 $\pm$ 19.2	4.5 $\pm$ 3.8	BDL
Garden egg	122.3 $\pm$ 22.2	32.1 $\pm$ 19.2	BDL
Kuca	80.6 $\pm$ 17.2	10.4 $\pm$ 7.1	BDL
<b>Legumes</b>			
Soya beans	546.8 $\pm$ 28.6	8.3 $\pm$ 4.2	BDL
Groundnut	398.6 $\pm$ 12.9	7.4 $\pm$ 3.2	9.8 $\pm$ 3.4
Local beans	453.6 $\pm$ 15.8	9.4 $\pm$ 2.4	18.9 $\pm$ 6.4

Measurement errors represent one sigma uncertainty

With the measurement system used in the present work, the DLs obtained were 17.3, 4.2, and 5.1 Bq kg<sup>-1</sup> for <sup>40</sup>K, <sup>226</sup>Ra, and <sup>228</sup>Th, respectively. Values below these numbers are taken in this work as below detection limit (BDL) of the detector.

## Results and discussion

Activity concentrations measured in the different food crop samples are presented in Table 2, while those measured in the soil samples are presented in Table 3, together with the measured gamma dose rates. Radionuclides from artificial sources were not detected in both the food and soil samples. The highest radionuclide concentrations are due to <sup>40</sup>K in all the food samples measured (Table 2). Similar findings have been reported by other authors [11, 20]. In general, measured radionuclide concentrations obtained in food items from this area are observed to be about ten times higher than those obtained for food samples from the southwestern region of Nigeria, where radionuclide levels in tubers were reported to vary from 10.6 to 46.4 Bq kg<sup>-1</sup> for <sup>40</sup>K, 0.5 to 2.7 Bq kg<sup>-1</sup> for <sup>226</sup>Ra, and from BDL to 1.4 Bq kg<sup>-1</sup> for <sup>228</sup>Ra [14], and where radionuclide levels in cereals were reported to vary from 36.4 to 186.9 Bq kg<sup>-1</sup> for <sup>40</sup>K, 0.2 to 1.4 Bq kg<sup>-1</sup> for <sup>238</sup>U, and from 0.3 to 1.8 Bq kg<sup>-1</sup> for <sup>232</sup>Th [15]. The values of <sup>40</sup>K obtained in the present study for tomato (158.9 ± 28.9 Bq kg<sup>-1</sup>), sweet potato (423.7 ± 30.8 Bq kg<sup>-1</sup>), and Irish potato (494.4 ± 22.1 Bq kg<sup>-1</sup>) are also higher than those reported in the literature [11], where values of 59 ± 7, 136 ± 2, and 74 ± 3 Bq kg<sup>-1</sup>, respectively, were obtained, while the values for <sup>226</sup>Ra and <sup>238</sup>U were found to be less than 2 Bq kg<sup>-1</sup>. Higher concentrations were also found in Indian foodstuff, where <sup>40</sup>K, <sup>226</sup>Ra, and <sup>228</sup>Th concentrations vary from 45.9 to 649.0, 0.01 to 1.16, and 0.02 to 1.26 Bq kg<sup>-1</sup>, respectively [12]. However, <sup>226</sup>Ra activity concentrations obtained in the present study are lower than those obtained by Banzi et al. [8], who

reported <sup>226</sup>Ra concentrations of 650 ± 11 and 393 ± 9 Bq kg<sup>-1</sup> for wild leaf vegetations and edible leaf vegetations in Tanzania, respectively.

Generally, <sup>226</sup>Ra concentrations measured in the crops were similar to or higher than those for <sup>228</sup>Th (Table 2), although <sup>226</sup>Ra concentrations in the soil were twice to ten times lower (Table 3). This is so because <sup>226</sup>Ra accumulates more effectively in plants than <sup>238</sup>U and <sup>232</sup>Th, owing to its higher environmental mobility, which results in an increased uptake from soil to plants [21–23]. Additionally, the relatively high concentrations obtained for the investigated radionuclides in this study compared to those reported in the literature may also be due to the heavy applications of nitrogen–phosphorus–potassium (NPK) fertilizers to the soil by the farmers to improve crop yield. This is considered necessary, as soil fertility in the investigated area is rather poor for agricultural purposes; this is due to the decades of mining and the absence of infrastructure for reclamation [24]. It has been reported that fertilizers may be rich in natural radioactive elements and, consequently, their application may enrich these radioactive elements in the plants [25]. The extent to which fertilizer application affected the soil characteristics in the Jos Plateau area was not investigated; this may be a subject for future work.

Except for farm 3, the activity concentration of <sup>228</sup>Th(<sup>232</sup>Th) was highest in all the soil samples investigated, with the highest value of 2,189.5 Bq kg<sup>-1</sup> found at farm 6 (Table 3). In most cases, concentrations for <sup>228</sup>Th are factors 2–10 higher than those for <sup>226</sup>Ra. This observation is in agreement with previous studies that showed that tin tailings are rich in thorium, and that soils in the Jos Plateau are equally rich in this radionuclide [4–6].

External gamma dose rates were measured in situ at those farms where food samples had been collected and values between 0.50 and 1.47 μSv h<sup>-1</sup> were found (Table 3). Assuming an occupancy factor of 0.5 (i.e., a working day of 12 h) for a typical peasant farmer, an annual effective dose due to external exposure can be

**Table 3** Activity concentrations (dry weight) of <sup>40</sup>K, <sup>226</sup>Ra, and <sup>228</sup>Th measured in the investigated soil samples, and the corresponding dose rate levels

Locations	<sup>40</sup> K (Bq kg <sup>-1</sup> )	<sup>226</sup> Ra (Bq kg <sup>-1</sup> )	<sup>228</sup> Th (Bq kg <sup>-1</sup> )	Dose rate (μSv h <sup>-1</sup> )
Farm 1	93.0 ± 9.6	145.2 ± 16.3	373.5 ± 8.9	0.72 ± 0.02
Farm 2	135.7 ± 7.1	175.1 ± 19.6	515.2 ± 10.1	0.73 ± 0.04
Farm 3	166.4 ± 12.4	10.9 ± 15.2	122.7 ± 11.2	0.50 ± 0.01
Farm 4	128.8 ± 17.2	72.5 ± 13.8	168.4 ± 7.8	0.89 ± 0.05
Farm 5	BDL	427.1 ± 12.4	1,036.5 ± 8.8	1.01 ± 0.05
Farm 6	55.1 ± 11.5	470.6 ± 10.9	2,189.5 ± 9.2	1.47 ± 0.05

Measurement errors represent one sigma uncertainty

**Table 4** Annual radionuclide intake and effective dose due to ingestion of  $^{226}\text{Ra}$  and  $^{228}\text{Th}$ 

Food group	Consumption rates (kg year <sup>-1</sup> ) [27]	Annual radionuclide intake (Bq)		Effective ingestion dose (mSv year <sup>-1</sup> )
		$^{226}\text{Ra}$	$^{228}\text{Th}$	
Cereals	50	2,430	785	0.74
Root crops	10	1,469	1,453	0.52
Potatoes	50	1,715	2,635	0.67
Vegetables	15	913	144	0.27
Legumes	20	502	577	0.18

calculated that varies from about 2,200–6,400  $\mu\text{Sv}$ . These values are about 4–13 times higher than the world average value of 500  $\mu\text{Sv year}^{-1}$  [9].

Furthermore, if the concentrations of these radionuclides in a certain food product and its average daily consumption rate are known, the mean daily intake of these radionuclides can be estimated. Daily and yearly effective doses due to the intake of radionuclides through ingestion of food can then be calculated, based on the metabolic models developed by the International Commission of Radiological Protection [26, 27]. To summarize, the effective dose  $H$  to a certain tissue  $T$  due to intake of radionuclide  $r$  is given by:

$$H_{T,r} = \sum (U^i \times C_r^i) \times g_{T,r} \quad (4)$$

where,  $i$  denotes a food group, the coefficients  $U^i$  and  $C_r^i$  denote the consumption rate (kg year<sup>-1</sup>) and activity concentration of the radionuclide  $r$  of interest (Bq kg<sup>-1</sup>), respectively, and  $g_{T,r}$  is the dose conversion coefficient for ingestion of radionuclide  $r$  (Sv Bq<sup>-1</sup>) in tissue  $T$ .

For adult members of the public, the recommended dose conversion coefficient  $g_{T,r}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{137}\text{Cs}$  are  $6.2 \times 10^{-9}$ ,  $2.8 \times 10^{-7}$ ,  $7.2 \times 10^{-8}$ , and  $1.3 \times 10^{-8}$  Sv Bq<sup>-1</sup>, respectively [26]. No scientific data exist at present on specific food consumption rates in the investigated area. Therefore, effective doses due to radionuclide intake were calculated assuming consumption rates published in the literature, for the area [27]. The results are presented in Table 4. The total effective doses calculated for  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  varied from from 0.18 mSv year<sup>-1</sup> (legumes) to 0.74 mSv year<sup>-1</sup> (cereals), respectively, with a total value of 2.38 (~2.4) mSv year<sup>-1</sup>. Potassium-40 is usually of limited interest because, as an isotope of an essential element, it is homeostatically controlled in the human cells. As a result, the body content of  $^{40}\text{K}$  is determined largely by its physiological characteristics, rather than by its intake.

It should be noted that the dose values given in Table 4 most likely represent an upper limit of the real

internal dose due to  $^{226}\text{Ra}$  and  $^{228}\text{Th}$ , as part of the food needed is imported from other areas and, for this reason, probably less contaminated. In addition, for those living in the investigated area, cereals and grains are even more important [28, 29] than assumed in the present study [27], and cereals were found to show quite low concentrations of these radionuclides.

## Conclusion

Activity concentrations of natural radionuclides ( $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Th}$ ) have been determined by means of gamma spectroscopy in different food crops that are regularly being consumed by the population of Bitsichi, an old tin mining town in the Jos Plateau, Nigeria. Activity concentrations of these radionuclides in the soil samples from farms were high probably due to the decades of former mining activities. For improved agricultural crop production, large amounts of NPK fertilizers have been and are still being applied. This is probably another reason for the observed enhanced radionuclide concentrations in the food crops compared to those reported in the literature in samples from other parts of the country. Activity concentrations of these radionuclides were found to be generally lower in cereals than in the other investigated crops. The external gamma dose rates to which an individual may be exposed while working on the farms were found to be somewhat higher (2,200–6,400  $\mu\text{Sv year}^{-1}$ ) in comparison to the world average value of 500  $\mu\text{Sv year}^{-1}$  [9]. The soil radioactivity in the area has been modified due to past mining activities, and the concentration levels of the natural radionuclides in the soil are enhanced. However, the values obtained on the concentration levels of natural radionuclides in the food samples suggest that the dose from the intake of these natural radionuclides is low and that no harmful health effects are likely to occur. Nevertheless, the data shown here represent a small but useful database that will allow further investigations. The data are also seen to be useful for

the newly established NNRA for its food policy, administration and regulatory functions.

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