

## Human Internal Exposure Due to Radionuclides Levels in Foodstuffs Grown Using Fertilizer in Jos Nigeria

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**Abstract:** The activity concentration of naturally occurring radionuclides in food samples grown with fertilizer in an elevated radiation background area of Jos, Nigeria were determined using gamma-ray spectrometry. The present study investigated the impact of Sulphate of ammonia fertilizer on foodstuffs samples. Sixty foodstuffs samples from fields cultivated with fertilizer in tin mining area were collected and analyzed for the activity concentrations of <sup>40</sup>K, <sup>226</sup>Ra, <sup>232</sup>Th employing gamma spectrometry. In the same vein, similarly analysis was undertaken in sixty samples grown with commonly primitive methods without fertilizer. The annual effective doses in the foodstuffs were deduced. The calculated values for foodstuffs grown with fertilizer were 9.2-188, 0.03-0.51 and 0.02-0.22  $\mu\text{Sv y}^{-1}$  for <sup>40</sup>K, <sup>226</sup>Ra, <sup>232</sup>Th. For foodstuffs grown without fertilizer application, the values were 2.7-146, 0-0.43 and 0.01-0.24  $\mu\text{Sv y}^{-1}$ . Based on ICRP recommendation, the level of radionuclides in the foodstuffs grown with fertilizer in tin mining area of the city will not constitute and radiological burden to the populace.

**Keywords:** *Foodstuffs, Tin, Fertilizer, Radioactivity, Jos Nigeria*

### Introduction

The use of different types of fertilizers such as Super Phosphate, Urea Sulphate, and Ammonium Nitrate Sulphate in the agricultural sector for the purpose of improving crop yield has become very common nowadays; fertilizers are usually used in reclaiming the land and improving the properties of crops (Alharbi, 2013). Therefore, fertilizers redistribute naturally occurring radionuclides at trace levels throughout the environment and become a source of radioactivity because of the phosphates in them. This phenomenon may result in potential radiological risks owing to external exposure during resident time in the farms and internal exposure through ingestion of food grown on fertilizer soils (Rehman et al, 2006). The phosphorous percentage of fertilizer is taken from phosphate rocks, which contains enhanced concentration of natural radionuclides (Skorovarov et al, 2000, Ashraf et al, 2001). It is essential to know the radioactivity in these foodstuffs keeping in view of human's health and radiation protection (Quindos et al, 1994, El-Taher and Makhluaf, 2010). One of the most important contributors to high level of radioactivity in Jos Plateau State Nigeria is the presence of tin, columbite and gypsum ores (Babalola, 1984). Tin mining activities began in 1902 under British and continue to the present day (Wikipedia, 2014a). Tin ore contains Monazite, Thorite and Zircon which are radioactive (Oresegun & Babalola, 1990, Oresegun & Babalola, 1993). There are numerous numbers of abandoned tin pits that are filled with water. These serve as sources of water for drinking, domestic uses and irrigation. Natural radionuclides exist in all human environments such as soil, air, food and human bodies. Every food has some small amount of radioactivity in it. The ingestion of fruits and vegetables is one of the pathways leading to internal retention and contribution to human exposure from natural and man-made sources (USEPA, 2006). Dose accrue to humans depends directly on the level of radioactivity in their environment and food they consume. Conversant among the radionuclides in food is <sup>40</sup>K, <sup>226</sup>Ra, <sup>232</sup>Th and their progenies (Eisenbud & Gesell, 1997). Agricultural practices such as irrigation and fertilizer (being produced from phosphate rocks) applications that aid the movement of radionuclides within fruit bearing plants should not be overlooked. Therefore, care should be given to the period between cultivation, domestic and industrial consumption (Carini, 1998). Several studies of naturally occurring radionuclides concentrations in foodstuffs, fruits and vegetables from normal and high background radiation area had been carried out (Fisenne *et al*, 1987, Long,

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1987, Shukla et al, 1994, Yu and Mao, 1995, Radhakrishna, et al, 1996, Pietrzak-Flis et al, 1997, EFSA, 2009). The study is carried out to determine, the suitability of these foodstuffs cultivated in Jos Plateau due to the fact that the area has

- (i) Large deposit of monazites
- (ii) Witnessed used of the water from abandoned tin site for irrigation
- (iii) Being confirmed to be highly radioactive (Oresegun and Babalola, 1990, Oresegun and Babalola, 1993)
- (iv) Witnessed the used of fertilizer by farmers for cultivation
- (v) Witnessed series of bombings by insurgents using improved explosive devices in recent times (BBC, 2010, Vanguard, 2010, Aljazeera, 2010, BBC, 2012, Reuters, 2014)
- (vi) 9.8m Gy annual average dose to people and tin mill workers (Oresegun and Babalola, 1990, Oresegun and Babalola, 1993)
- (vii) 0.08-1.57 Gy cumulative dose to tin mill workers for a period of 8-30 years (Oresegun and Babalola, 1990, Oresegun & Babalola, 1993).

### **Area under study**

Jos is located on a granitic plateau about 1500 m above the sea level in the North Central of Nigeria (Balogun, 2003) on coordinates:  $9^{\circ} 56'$ ,  $8^{\circ} 53'$  E/ $9.933^{\circ}$ N  $8.75^{\circ}$ E (Balogun, 2003, Wikipedia, 2014b). The phases of volcanic activities involve in formation of volcanic features (Emielu, 2004). Typical among these features is Riyom rock at Riyom Local Government Area of the State, the Shere hills and Kurra falls found east of Jos, Assop falls on the road to Abuja (Wikipedia, 2014a, Wikipedia, 2014b). The total annual rainfall in the area is 1422 mm. The lithological formation in the area is basement complex with lithosols soil types. The area has a population of about 900,000 people (NPC, 2006). The mean monthly temperature is between  $21-25^{\circ}$ C (Balogun, 2003). It drops to  $11^{\circ}$ C around mid-November to January resulting in chilly nights (Wikipedia, 2014b). The main resources of the area are agricultural products and mining activities. The climate is ideal for the growth of several fruits, cereal and cash crops as seen in the analyzed foodstuffs samples. Soil in the area is not heavily leached due to lack of heavy rainfall (Emielu, 2004). Farmers in the area use fertilizer and water from the abandoned tin pits for irrigation and therefore have high agricultural output. These products in commercial quantity are sold in the major markets in southern Nigeria.

### **Materials and Methods**

#### **Sample collection, preparation and counting**

Foodstuffs samples grown with fertilizer were obtained from Jos (tin mining city) in Plateau state. Control samples grown without fertilizer were obtained in Ekiti state southern part of Nigeria. Ten samples of one hundred gramme (100 g) each of foodstuffs were collected. Periods of collection were not classified based on the fact that farmers practiced irrigation system of farming; therefore foodstuffs are available throughout the year. The species are *Lycopersicum esculentum*, *Solanum icanum*, *Capsicum annum var. abbreviatum*, *Capsicum annum var. acuminatum*, *Capsicum annum var. grossum*, *Citrullus lanatus* and *Abelmoschus esculentus*. In all sixty samples were collected from each area (Table 1). Samples were washed, cleaned, cut into pieces and dried in air. They were then oven-dried at  $80^{\circ}$ C for 16 hrs to remove the moisture content (Santos *et al*, 2002). Dried samples were grounded separately using a domestic blender. Thirty (30 g) of each dried foodstuffs were placed in polyethylene beakers previously cleaned with 10 % nitric acid. The beakers were then sealed and allowed to stand for at least 4 weeks so that the  $^{226}\text{Ra}$  series was able to reach radioactive secular equilibrium. The activity concentrations of natural radionuclides  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the samples were determined using a  $3 \times 3$  inch NaI (Tl)  $\gamma$ -ray spectrometer system with a 1024 channel computer analyzer. The detector has a peak efficiency of  $1.2 \times 10^{-5}$  at 1.33 MeV Co-60 and an energy resolution (FWHM) of 8 % for 0.66 MeV, detector employed with adequate lead shielding which reduces the background radiation. The  $^{226}\text{Ra}$  activity determination was based on 1.76 MeV gamma rays from  $^{214}\text{Bi}$ . The activity of  $^{232}\text{Th}$  was determined through its 2.62 MeV gamma rays from  $^{208}\text{Tl}$ . The activity of  $^{40}\text{K}$  (non-series) radionuclides was determined through its 1.46 MeV gamma rays.

## Theoretical Calculations

### Activity concentrations

Activity concentrations in each sample were determined using the following relation (Ibrahim, 1998, Abbady, 2004).

$$A_s (\text{Bq. kg}^{-1}) = \frac{C_a}{\epsilon P_\gamma M_s} \quad 1$$

Where  $C_a$  is the net gamma counting rate (counts per second),  $\epsilon$  is the detector efficiency of the specific  $\gamma$ -ray,  $P_\gamma$  is the absolute transition probability of gamma-decay and  $M_s$  is the mass of the sample in kg.

### Internal dose from ingested foodstuffs

The annual consumption (Q) for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were determined in foodstuffs using the activity level  $A_s$  in foodstuffs and the annual food consumption ( $C_{ra}$ ). The calculated annual intake and the internal dose conversion factors (IDCF) of  $6.2 \times 10^{-3}$ ,  $0.28 \times 10^{-3}$  and  $0.23 \times 10^{-3}$   $\mu\text{Sv Bq}^{-1}$  of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were used to estimate the annual internal effective dose (H) (Saleh et al, 2007) using the following relations.

$$Q = A_s \times C_{ra} \quad 2$$

$$H = Q \times \text{IDCF} \quad 3$$

## Results and Discussion

The measured concentrations of natural radionuclides  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the foodstuffs samples are shown in Table 1. In foodstuffs with Sulphate of Ammonia application (SA), the mean activity concentrations of  $^{40}\text{K}$  varies from 222.2 in *Solanum icanum* to 421.9  $\text{Bq kg}^{-1}$  in *Citrullus lanatus*, the mean concentration of  $^{226}\text{Ra}$  varies from 14.6 in *Capsicum annum var. abbreviatum* to 20.1  $\text{Bq kg}^{-1}$  in *Capsicum annum var. grossum*, whereas the mean concentration of  $^{232}\text{Th}$  exists in the range from 7.5 in *Capsicum annum var. acuminatum* to 13.6  $\text{Bq kg}^{-1}$  in *Citrullus lanatus*. For foodstuffs without Sulphate of Ammonia application (WSA), the mean activity concentrations of  $^{40}\text{K}$  varies from 213.1 in *Solanum icanum* to 367.8  $\text{Bq kg}^{-1}$  in *Abelmoschus esculentus*, the mean concentration of  $^{226}\text{Ra}$  varies from 1.4 in *Abelmoschus esculentus* to 17.8  $\text{Bq kg}^{-1}$  in *Solanum icanum*, whereas the mean concentration of  $^{232}\text{Th}$  exists in the range from 8.4 in *Capsicum annum var. acuminatum* to 12.4  $\text{Bq kg}^{-1}$  in *Capsicum annum var. abbreviatum*.

**Table 1.** The mean activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the foodstuffs (SA\* and WSA<sup>+</sup>)

Foodstuffs	Sample no	$^{40}\text{K}$ (Bq kg <sup>-1</sup> )	$^{226}\text{Ra}$ (Bq kg <sup>-1</sup> )	$^{232}\text{Th}$ (Bq kg <sup>-1</sup> )
<i>Lycopersicon esculentum</i> (tomatoes)	10	331.27 ± 70.12*	19.81 ± 8.34*	10.43 ± 3.76*
<i>Solanum icanum</i> (garden egg)	10	257.28 ± 58.08 <sup>+</sup>	16.91 ± 4.98 <sup>+</sup>	9.68 ± 4.01 <sup>+</sup>
<i>Capsicum annum var. abbreviatum</i> (pepper 1)	10	222.18 ± 49.39*	18.74 ± 7.63*	9.78 ± 3.76*
<i>Capsicum annum var. acuminatum</i> (pepper 2)	10	213.12 ± 74.92 <sup>+</sup>	17.80 ± 6.43 <sup>+</sup>	12.32 ± 4.22 <sup>+</sup>
<i>Capsicum annum var. grossum</i> (pepper 3)	10	298.22 ± 57.32*	14.57 ± 7.01*	11.43 ± 3.78*
<i>Citrullus lanatus</i> (water melon)	10	252.13 ± 49.63 <sup>+</sup>	13.21 ± 5.11 <sup>+</sup>	12.43 ± 4.87 <sup>+</sup>
<i>Abelmoschus esculentus</i> (okra)	10	324.13 ± 48.52*	14.81 ± 5.99*	7.45 ± 2.37*
	10	298.48 ± 60.02 <sup>+</sup>	16.50 ± 6.11 <sup>+</sup>	8.39 ± 3.67 <sup>+</sup>
	10	302.26 ± 65.86*	20.12 ± 5.76*	11.24 ± 4.11*
	10	301.13 ± 61.99 <sup>+</sup>	13.42 ± 4.76 <sup>+</sup>	9.32 ± 3.05 <sup>+</sup>
	10	421.92 ± 78.33*	16.91 ± 6.59*	13.62 ± 5.01*
	NA	NA	NA	NA
	NA	NA	NA	NA
	10	367.76 ± 75.84 <sup>+</sup>	1.42 ± 8.03 <sup>+</sup>	9.78 ± 4.01 <sup>+</sup>

Note: Sulphate of Ammonia\* (SA), Without Sulphate of Ammonia<sup>+</sup> (WSA), NA- Not available.

The results showed that natural radionuclides have the higher emitter of radiation in foodstuffs in SA application compared to WSA. T-test analysis was carried out on the activity concentrations in foodstuffs from both areas. The results obtained showed that  $P > 0.05$  i.e. fertilizer application has no significant effect on the foodstuffs. Table 2, presents the range of estimated daily intake ( $\text{Bq d}^{-1}$ ) and

annual effective dose ( $\mu\text{Sv y}^{-1}$ ) of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ . The minimum and maximum values of annual effective dose in foodstuffs with SA application were found to vary from 9.2-188, 0.03-0.51 and 0.02-0.22  $\mu\text{Sv y}^{-1}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ . For foodstuffs WSA, the minimum and maximum value of annual effective dose found to vary from 2.7-146, 0-0.43, and 0.01-0.24  $\mu\text{Sv y}^{-1}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  respectively.

**Table 2.** The estimated daily intake ( $\text{Bq d}^{-1}$ ) and annual effective dose ( $\mu\text{Sv y}^{-1}$ ) of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and consumption rate (Cra)  $\times 10^{-3}$  ( $\text{kg d}^{-1}$ ) (SA\* and WSA<sup>+</sup>)

Foodstuffs	Cra	$(\text{Bq d}^{-1})$			$(\mu\text{Sv y}^{-1})$		
		$^{40}\text{K}$	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$	$^{226}\text{Ra}$	$^{232}\text{Th}$
<i>Lycopersicon esculentum</i> (tomatoes)	250.10	82.90* 64.30 <sup>+</sup>	4.95 * 4.23 <sup>+</sup>	2.61* 2.42 <sup>+</sup>	188* 146 <sup>+</sup>	0.51* 0.43 <sup>+</sup>	0.22* 0.24 <sup>+</sup>
<i>Solanum icanum</i> (garden egg)	18.21	4.05* 3.88 <sup>+</sup>	0.34* 0.32 <sup>+</sup>	0.18* 0.22 <sup>+</sup>	9.17* 3.68 <sup>+</sup>	0.03* 0.03 <sup>+</sup>	0.02* 0.02 <sup>+</sup>
<i>Capsicum annum var.</i> <i>abbreviatum</i> (pepper 1)	125.50	37.40* 31.60 <sup>+</sup>	1.83* 1.66 <sup>+</sup>	1.43* 1.56 <sup>+</sup>	35.50* 30.00 <sup>+</sup>	0.19* 0.17 <sup>+</sup>	0.12* 0.13 <sup>+</sup>
<i>Capsicum annum var.</i> <i>acuminatum</i> (pepper 2)	125.50	37.90* 37.80 <sup>+</sup>	2.53* 1.68 <sup>+</sup>	1.41* 1.17 <sup>+</sup>	36.00* 35.90 <sup>+</sup>	0.26* 0.17 <sup>+</sup>	0.12* 0.10 <sup>+</sup>
<i>Citrullus lanatus</i> (water melon)	50.92	21.50* NA	0.86* NA	0.69* NA	20.40* NA	0.06* NA	0.06* NA
<i>Abelmoschus esculentus</i> (okro)	7.80	NA 2.87 <sup>+</sup>	NA 0.01 <sup>+</sup>	NA 0.08 <sup>+</sup>	NA 2.72 <sup>+</sup>	NA 0.00 <sup>+</sup>	NA 0.01 <sup>+</sup>

Note: Sulphate of Ammonia\* (SA), Without Sulphate of Ammonia<sup>+</sup> (WSA), NA- Not available.

### Conclusion

The activity concentrations of natural radionuclides have been determined in foodstuffs samples grown with fertilizer in tin mining areas in Jos- Plateau Nigeria using gamma-ray spectroscopy. The activity concentrations of these naturally occurring radionuclides in foodstuffs grown with fertilizer in Jos were a bit higher than those grown without fertilizer in southern part of Nigeria. However, the annual effective doses estimated in both samples are still within the acceptable limit. It suffices to say, there is no significant harmful effect to those that ingest foodstuffs grown with fertilizer in tin mining area of Jos.

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