# Determination of activity concentrations of <sup>40</sup>K, <sup>232</sup>Th and <sup>238</sup>U in some fruit samples commonly consumed in Kano Metropolis using Instrumental Neutron Activation Analysis (INAA)

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#### Abstract

Five fruits samples: Pineapple, Papaya, Plantain, Guava and Okro, frequently consumed in Kano metropolis were analyzed for both short-lived and long-lived elements using NIRR-1 reactor at Centre for Energy Research and Training (CERT), ABU Zaria, to determine the concentration of naturally occurring radioactive elements and hence their fitness for consumption and health of people. The concentrations of Uranium in all the samples were Below the Detection Level (BDL) of NIRR-1 Reactor. However the concentration of Thorium in Pineapple was 2.805±0.081Bq/kg  $(0.690 \pm 0.020 \mu gg^{-1}),$ Pawpaw was 0.365±0.061Bq/kg (0.090±0.015µgg<sup>-1</sup>), and in Guava was 0.256±0.081Bq/kg (0.063±0.020µgg<sup>-1</sup>), while it was Below Detection Level in Plantain and Okro. The highest concentration of  $^{40}K$  of 301.33±0.56Bq/kg (9733±18µgg<sup>-1</sup>) was obtained in Plantain; 210.20±0.53Bq/kg  $(6789 \pm 17 \mu gg^{-1})$  was recorded in Pineapple;  $77.12 \pm 0.84 Bq/kg$  (2491±27 $\mu gg^{-1}$ ) was obtained in Pawpaw;  $64.64\pm0.77Bq/kg$  (2088 $\pm25\mu gg^{-1}$ ) was obtained in Okro while the least concentration of 58.36±0.71Bq/kg (1885±23µgg<sup>-1</sup>) was recorded in Guava. The mean concentration of <sup>40</sup>K in all the samples is 142.32±0.68Bq/kg  $(4597.2\pm 22\mu gg^{-1})$ . The total estimated annual effective dose, exposed to by people in Kano metropolis, as a result of consumption of the five fruits is 52.63µSv.yr<sup>-1</sup>. This is lower than the world average value 290µSv.yr<sup>-1 [1]</sup> and hence poses no health risk to consumers.

Keywords: Activity concentration; Radioactivity; Fruits; Kano Metropolis: INAA.

#### **1.0** Introduction

Natural radionuclides exist in every human environment because they are left over from when the world and universe were created. Even human bodies contain Naturally Occurring Radioactive Materials (NORMs) because humans are products of their environment. Humans are subjected to NORMs on daily basis because of radiations from the environment (soil and air) we live, air we breathe, water we drink, fruits/foods we eat. NORMs are categorized as being of terrestrial or cosmic origin [2]. The exposures of humans to these natural sources can be both internal and external. Internal exposures to terrestrial radionuclides occur from intake of these nuclides through inhalation or ingestion. Inhalation exposure dose results from the existence of dust particles in air, including radionuclides from Uranium ( $^{238}$ U) and Thorium ( $^{232}$ Th) series [3]. The biggest contribution to inhalation exposure comes from short half-life decay product of Radon (222Rn). Naturally occurring radionuclides of Uranium and Thorium are significant contributors of ingestion dose and are present in biotic system of plants, animals, soil, water and air. Distribution of these radionuclides in different parts of the plant depends on the chemical characteristics and several parameters of plant and soil [4]. Ingestion exposure dose mostly results from <sup>238</sup>U and <sup>232</sup>Th radionuclides series and <sup>40</sup>K in drinking water, vegetables, fruits and foodstuff. Presence of radioactivity in plant organs has been reviewed by different scholars. The study of radioactivity concentration in fruits assume importance as it is necessary to estimate the ingestion dose the general public received from the consumption of these fruits daily and/or annually. The aim of this study is to determine the exposure dose of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in some selected fruits which are commonly consumed in Kano metropolis. The significance of the study is that it is one of the first studies to be carried out to determine the background radiation levels in fruits consumed in this region and will provide data for future studies.

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# 2.0 Materials and Methods

# 3.0 Radioactivity Measurements in Fruits Samples

Samples of five fruits: Guava, Okro, Pawpaw, Plantain and Pineapple were purchased directly from the farmers/fruits suppliers who brought these fruits, in large quantities for sale, to Fruits Dealers in Yan-lemu market, a major fruit market in Naibawa, Kumbotso Local Government Area of Kano. This was done so as to be able to identify the source of the fruits. Any soils or foreign materials on the samples were removed so that they were suitable for consumption, cut into small pieces using a sterilized knife, and washed under distilled water. The samples were dried and then taken to the preparatory Laboratory of Centre for Energy Research and Training at Ahmadu Bello Zaria, (ABU), where each of the samples was grounded into powder before irradiation. 8ml polythene vials were sterilized in a mixture of distilled water (200ml) and nitric acid, HNO<sub>3</sub>, (100ml) for 24 hours. The sterilized polythene vials were each weighed empty before putting each of the powdered samples in separate polythene vials and reweighing. Between 0.2500-0.3000g of each of the (biological) samples required were weighed, before the vials were blown and sealed. Using rabbit carriers, the samples and standards were transferred into the Nigeria Research Reactor (NIRR-1) through a Pneumatic Transfer System which uses Pneumatic Pressure. The Standard used in the analysis of this research was the SRMs 1515 (Apple Leaves). The samples were irradiated with NIRR-1 at CERT, Zaria. For the detection of short-lived elements, the samples were irradiated with thermal neutron flux of 2.5 x  $10^{11}$  ncm<sup>-2</sup> s<sup>-1</sup> for 5 minutes duration inside the Reactor. However, for long-lived elements, the samples were irradiated with thermal neutron flux of 5.0 x 10<sup>11</sup> ncm<sup>-2</sup> s<sup>-1</sup> for 6 hours. The whole system is equipped with the electronic timers which help in monitoring the exact irradiation and decay times. The activity of the irradiated samples was allowed to decay/fall within the acceptable handling limit of about 30µSv/h which is well below the initial activity of the samples immediately after its removal from the reactor.

The samples were then taken for Counting. For short-lived elements, the first and second counting were carried- out the same day of exposure of the samples to radiation. The first counting was carried out immediately after the acceptable handling limit is reached and the second counting about two hours after the first. For long-lived elements; the first counting was carried out on the third day after irradiation and the second counting the tenth day after irradiation. The samples were then taken to the Detector. The detecting set-up consists of a High Purity Germanium (HPGe) detector, connected to a PC-based Multi-Channel Analyzer (MCA) in a fixed sample to detector geometry.

## 4.0 Annual Production and Intake of the Selected Fruit Crops

In Nigeria, most horticultural products (fruit crops) are sold fresh with limited packaging and storage, a situation contributing to little or inconsistence in growth of production of fruits (and vegetables). Fruits and vegetables are important sources of minerals and vitamins required for healthy living and source of roughage which aids digestion and prevent constipation. Shown in Table 1 is the individual and total production figures of the five selected fruits sample used in this study from 2007 to 2011 in Nigeria.

Total Annual Production (in tones)								
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**Table 1:** Production of the selected individual fruit crops in Nigeria, in tones, from 2007-2011.

Source: FAO (2012)

Table 2: Individual Annual	Intake of the Selected Fruits.
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Fruits	uits Average Annual		Individual Annual	Individual Annual	
	Production	Intake (in tones)	Intake (in Kilograms)	Intake (to the nearest Kg)	
Mango and Guava	782,400	0.00489	4.89	5	
Papaya	735,800	0.00460	4.60	5	
Plantain	2,848,200	0.01780	17.80	18	
Pineapple	874,600	0.00557	5.57	6	
Okro	1,001,000	0.00626	6.26	6	

In Nigeria, with very little export of these fruit crops, almost all the fruits are consumed locally within the country. With the assumption that the number of these fruits that get spoilt before consumption is also negligible then the average annual intake

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of these fruits, in kilograms, over a period of five years can be obtained by dividing the average annual production by the total population of Nigeria (160 millions). This is shown in Table 2.

## Conversion Factors ppb U, ppb Th, ppm K to Bq/kg

The equations given below were used to convert from parts per billion (ppb) of Uranium and Thorium, and parts per million (ppm) of Potassium to Becquerel per kilogram (Bq/kg). The conversion factors for the primordial nuclides (-radiopurity.in2p3fr/conversion/html) are given by:

- (i)  $1Bq \text{ of } {}^{238}\text{U/kg} = 81ppb \text{ U} = 81 \text{ x } 10^{-3}ppm \text{ U},$
- (ii)  $1Bq \text{ of } {}^{232}\text{Th/kg} = 246\text{ ppbTh} = 246 \text{ x } 10^{-3}\text{ppm Th, and}$
- (iii) 1Bq of  $^{40}$ K/kg = 32.3ppm K.

#### 5.0 Dose Estimation

Ingestion dose occurring through the intake of radionuclides depends on the consumption rate of fruits, vegetables and foodstuff, and the concentration of the radionuclides involved. Ingestion dose is calculated using the following equation [5, 6, 1]

$$H_{Tr} = \sum \left( u^{i} C_{r}^{i} \right) g_{Tr}$$

Where: i is the foodstuff group, U<sup>i</sup> and C<sup>i</sup><sub>r</sub> are annual consumption rate (kg) and radionuclide activity concentration (Bq/kg) for their coefficients and  $g_{T,r}$  is the dose conversion coefficient for r radionuclide in Sv/Bq. The dose conversion coefficients for <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs radionuclides for adult members of the society/public are: 4.5 x 10<sup>-8</sup>, 2.3 x 10<sup>-7</sup>, 6.2 x 10<sup>-9</sup> and 1.3 x 10<sup>-8</sup>Sv/Bq respectively [6, 5, 7].

## 6.0 **Results and Discussions**

Table 3: Concentration	ons of Radior	nuclides in	Selected Frui	ts in pa	arts per	million (	ppm)

Fruit	Fruits	<b>Botanical Name</b>	Concentration of Radionuclides in ppm (parts				
Identification Code			per million)				
			<sup>40</sup> K	<sup>232</sup> Th	<sup>238</sup> U		
GV1	Guava	Psidiumguajava	1885±23	0.063±0.020	BDL		
OK2	Okro	Ahelmoschusesculentus	$2088 \pm 25$	BDL	BDL		
PW3	Pawpaw	Carica papaya	2491±27	$0.090 \pm 0.015$	BDL		
PL4	Plantain	Musa species	9733±18	BDL	BDL		
PN5	Pineapple	Ananascomosus	6789±17	0.690±0.020	BDL		

Table 4: Activity concentrations of Radionuclides in Selected Fruits in Becquerel per kilogram (Bq/kg)

Fruit Identification Code	Fruits	Botanical Name	Activity cond kilogram (Bq/	Becquerel per	
			<sup>40</sup> K	<sup>232</sup> Th	<sup>238</sup> U
GV1	Guava	Psidiumguajava	58.36±0.71	$0.256 \pm 0.081$	BDL
OK2	Okro	Ahelmoschusesculentus	$64.64 \pm 0.77$	BDL	BDL
PW3	Pawpaw	Carica papaya	$77.12 \pm 0.84$	$0.365 \pm 0.061$	BDL
PL4	Plantain	Musa species	301.33±0.56	BDL	BDL
PN5	Pineapple	Ananascomosus	210.20±0.53	$2.801 \pm 0.081$	BDL

Tables 3 and 4 show the concentrations, in ppm ( $\mu g/g = mg/kg$ ), and activity concentrations, in Bq/kg, of naturally occurring radionuclides: <sup>40</sup>K, <sup>232</sup>Th and <sup>238</sup>U in some fruits sample commonly consumed in Kano metropolis and its environs respectively. The concentrations of <sup>40</sup>K in all the fruits samples is generally high compared to other radionuclides detected in the samples. The highest concentration of <sup>40</sup>K was obtained in Plantain, 9733±18µg/g (Table 1) which is equivalent to 301.33±0.56Bq/kg (Table 2) while the least concentration was recorded in Guava, 1885±23µg/g (ppm), which translates to activity concentration of 58.36±0.71Bq/kg. The mean concentration of <sup>40</sup>K in all the samples is 4597.2±22µBq/g (142.32±0.68Bq/kg). This result agrees with the world range reported by Maud and O'Hara (1989) for <sup>40</sup>K concentration from 40Bq/kg to 240Bq/kg. The high concentration of <sup>40</sup>K is due to the fact that it is a macronutrient and that <sup>40</sup>K is the most abundant natural radionuclide present in food samples; its activity concentration varies from 50Bq/kg to 686Bq/kg depending on the kind and nature of the sample. It may be expected that the soil characteristics favour the mobilization of potassium and its subsequent migration into plants. <sup>232</sup>Th was recorded in Pineapple which was 0.690±0.020µg/g (2.801±0.081Bq/kg) and the least concentration of <sup>232</sup>Th was found in Guava fruit. The mean concentration of <sup>232</sup>Th in all the fruit samples is 0.169±0.011µg/g (0.686±0.045Bq/kg). The reason for the presence of <sup>232</sup>Th in the samples is

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largely due to the fact that <sup>232</sup>Th is one of the natural radionuclides found in the soil and during the uptake of nutrients from the soil by fruit bearing plants, some amount of <sup>232</sup>Th are also taken up by the plants and that some of the element are either stored up in the leaves or fruits or tubers of the plants. The concentration and the activity concentration of <sup>238</sup>U in all the fruit samples were below detection limit (BDL) of the analytical method employed in the analysis of the fruits (i.e. Instrumental Neutron Activation Analysis)

S/No	S/No Country List of fruits A				Conc. (Bo	Method of	Reference	
			<sup>238</sup> U	<sup>232</sup> Th	<sup>226</sup> Ra	<sup>40</sup> K	Analysis	
1.	Kinta District, Perak, Malaysia	Banana, Papaya, Jack fruit, Rambotan	0.004	0.002	NA	NA	Gamma ray spectrometr y.	Ramli et. al, (2009)
2.	South-west, India	Mango, Ripe Mango Cherry fruit, Banana, Papaya, Guava, Pomegranate	0.32±0 .03	2.21±0 .16	0.72±0 .16	75.61±2 0.15	Gamma ray spectrometr y.	Shanti et. al., (2009)
3.	Elazig Region, Turkey	Melon, Pear, Quince, Grapes, Apple, Watermelon	BDL	0.65±0 .14	0.64±0 .26	13.98±1 .22	Gamma ray spectrometr y.	Canbazogl u and Dogru (2012)
4.	Ado-Ekiti, Nigeria	Plantain Banana (Desert, cooking and hybrid)	5.74±1 .78	3.64±1 .36	NA	48.75±8 .53	Gamma ray spectrometr y.	Ojo (2013)
5.	Kano, Nigeria	Pawpaw Plantain Pineapple Guava Okro	BDL	0.17±0 .01	NA	142.32± 0.68	Neutron Activation Analysis	Present Study

Table 5: Comparison of Average Activity Concentration of Fruits (Bg/kg) of this Study with other values from Similar Study

Table 5 compares the results of concentration of radionuclides, in Bq/kg, obtained from this study with related studies carried-out on fruits samples by other researchers within and outside this country. While  $^{238}$ U was Below Detection Level (BDL) in the samples used in this study and similar study carried-out in Turkey by [3]; an average concentration of 0.32±0.03Bq/kg was obtained in fruits in Southwest, India [4]; 5.74±1.78Bq/kg was recorded in Musa Species, in Ado Ekiti, Nigeria by [8]; and the least average concentration of 0.004Bq/kg in fruit samples was recorded in Kinta District, Perak in Malaysia [9]. The highest average concentration of  $^{232}$ Th in fruits of 0.65±0.14Bq/kg was recorded in study carried out in Elazig region (Turkey), by [3]; 2.21±0.16Bq/kg in Southwest (India,) by [4]; 0.17±0.01Bq/kg was obtained in this study and the least concentration of 0.002Bq/kg obtained in Kinta District, Perak (Malaysia) by[9].  $^{226}$ Ra was detected with average concentration of 0.72±0.16Bq/kg in fruits in India (Southwest), 0.64±0.26Bq/kg in Turkey (Elazig) but was not analyzed in this study. The highest average concentration of 142.32±0.68Bq/kg for  $^{40}$ K was, however, obtained in this study as compared to other studies carried-out in other countries.

<b>Table 0:</b> Dose coefficients and commuted effective dose values for 0, 111 and K									
Fruits	Annual	Mean	activit	y conc.	Activity intake (Bq)			Committed	
	Intake (kg)	(Bq/kg)					effective dose		
		<sup>238</sup> U	<sup>232</sup> Th	<sup>40</sup> K	<sup>238</sup> U	<sup>232</sup> Th	<sup>40</sup> K	(µSv.yr <sup>-1</sup> )	
Plantain	18	-	-	301.33	-	-	5423.94	33.63	
Pawpaw	5	-	0.365	77.12	-	1.825	385.60	2.81	
Pineapple	6	-	2.801	210.20	-	16.81	1261.20	11.69	
Guava	5	-	0.256	58.36	-	1.28	291.80	2.10	
Okro	6	-	-	64.64	-	-	387.84	2.40	
Total	40kg							52.63	

**Table 6:** Dose coefficients and committed effective dose values for <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K

Table 6 gives the Effective dose values exposed due to natural radionuclides taken into body through the consumption of fruit samples by people in Kano metropolis. Average activity concentration (Bq/kg) was multiplied by fruit consumption rate, and annual activity intake value was determined in Bq unit. The individual annual fruit intake, of the selected samples, for the

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purpose of this study was calculated as: 18kg for plantain; 5kg for Pawpaw; 6kg for Pineapple; 5kg for Guava; and 6kg for Okrototaling 40kg for all the five fruits used in this research for people in Kano metropolis (see Table 2). The effective dose value was then determined by multiplying annual activity intake value by effective dose coefficient. The highest annual exposure of people to naturally occurring radionuclides due to ingestion from fruits was recorded in Plantain (33.63  $\mu$ Sv.yr<sup>-1</sup>), then Pineapple (11.69  $\mu$ Sv.yr<sup>-1</sup>), Pawpaw (2.81  $\mu$ Sv.yr<sup>-1</sup>), Okro (2.40  $\mu$ Sv.yr<sup>-1</sup>) and the least in Guava (2.10  $\mu$ Sv.yr<sup>-1</sup>). This study found that adults living in Kano metropolis intake a radiation dose, due to consumption of these five fruits, of approximately 52.63  $\mu$ Sv yr<sup>-1</sup>. This radiation dose is (52.63  $\mu$ Sv yr<sup>-1</sup>) is lower than the world average value of 290  $\mu$ Sv yr<sup>-1</sup> [1] and therefore present no health risk to consumers and hence public health. Dose values obtained in this study fall within acceptable limit for public as widely reported in literature [8].

#### 7.0 Conclusion

The concentration of radionuclides: <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K was analyzed in five (5) fruit samples consumed in Kano metropolis using Neutron Activation Analysis. The concentration of <sup>238</sup>U in all the fruit samples was Below Detection Limit (BDL) of the reactor NIRR-1, at Centre for Energy Research and Training (CERT), Zaria used for the analysis, <sup>232</sup>Th was detected in three out of the five samples. The concentration of <sup>232</sup>Th in Pineapple was  $2.805\pm0.081Bq/kg$ ; in Pawpaw was  $0.365\pm0.061Bq/kg$  and in Guava was  $0.256\pm0.081Bq/kg$ . The mean/average concentration of <sup>232</sup>Th in all the samples was  $1.141\pm0.074Bq/kg$ . <sup>40</sup>K was detected in all the samples with the highest concentration of  $301.33\pm0.56Bq/kg$  recorded in Plantain;  $210.20\pm0.53Bq/kg$  in Pineapple;  $77.12\pm0.84Bq/kg$  in Pawpaw;  $64.64\pm0.77Bq/kg$  in Okro and  $58.36\pm0.71Bq/kg$  in Guava. The mean concentration of <sup>40</sup>K in all the samples was  $142.32\pm0.68Bq/kg$ . It was found that the radiation dose of  $52.63\mu Sv.yr^{-1}$ , due to consumption of these fruits, was less than the world average value of  $290\mu Sv.yr^{-1}$  [1], and therefore poses no threat to the health of people in Kano metropolis. The results obtained in this research agreed to almost 70% to the existing values in the literature.

#### 8.0 Quality Control

This quality control of the analytical results of Instrumental Neutron Activation Analysis (INAA) obtained in this study was ensured by the agreement between Standard Reference Materials (SRMs) for QAQC used in this research (SRM 1515) and the Certified Values of SRM 1515 Apple Leaves of QAQC of NIST (National Institute of Standard and Technology).

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