

Long-lived gamma-emitting radionuclides in commonly consumed imported coffee brands in Lagos and the total committed effective dose using a high purity germanium detector

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Abstract

In public opinion, most of the imported food products are associated with several health risks relative to their origin. This study was set out to evaluate the long-lived gamma-emitting radionuclides in coffee brands imported from different countries, with steps taken towards assessing the harmfulness due to the level of activity in the each sample. The commonly consumed brands were selected based on a mixed sampling method. The samples were prepared accordingly for gamma analysis and analyzed through hyper pure germanium gamma spectrometry technique. The outcome of the study based on the counting statistics recorded the minimum activity concentrations value of 0.10 ± 0.04 Bq kg⁻¹, 0.34 ± 0.04 Bq kg⁻¹ and 0.34 ± 0.04 Bq kg⁻¹, maximum activity concentrations value 0.41 ± 0.08 Bq kg⁻¹, 2.39 ± 0.32 Bq kg⁻¹ and 985.32 ± 52.25 Bq kg⁻¹, and all-sample mean of (\pm SD) 0.19 ± 0.05 Bq kg⁻¹, 1.31 ± 0.15 Bq kg⁻¹ and 877.49 ± 46.54 Bq kg⁻¹ respectively for ²³⁸U, ²³²Th and ⁴⁰K. While the activity concentrations of ²³⁸U and ²³²Th are well below the acceptable limit, the activity concentrations of ⁴⁰K is dominant and significantly higher than the threshold value irrespective of the origin. The current findings also indicate that activity concentrations for ²³⁸U and ²³²Th lie within the data for other food samples, but that of ⁴⁰K is clearly above the values in other food samples. Potassium, one of the vital micromolecules in the body, is essentially regulated under normal condition in healthy human. In spite its higher value, ⁴⁰K is disregarded as a contributor to the ELCR and was not accounted for in this study. This study presents mortality probability of 6 in 100,000,000.

1 Introduction

Among three common beverages in the world [1] and the second most traded international product after oil [2] is Coffee; a non-alcoholic beverage, a tropical evergreen plant of African origin from the genus *Coffea*, with *C. arabica* and *C. canephora* as the main species used for the beverage production. The latter which is also simply referred to as robusta, is grown in 14 states across Nigeria (Oyo, Ekiti, Ogun, Kwara, Ondo, Kogi, Delta, Edo, Abia, Akwa Ibom, Cross River, Taraba,

Jos and Bauchi), and accounts for 90% of all coffee exports. Arabica is delicate, vulnerable to pests and grows in cool subtropical climate [1]. Commercial production globally is dominated at 70% approximately by arabica [2]. In 2022/2023 coffee year, from October 2022 to May 2023, 83.0 million bags were recorded as against 87.5 million bags in the same period in 2021/2022 coffee year, a noticeable decrease of 5.1% though [3]. The trade statistics for July 2022 from the International Coffee Organization showed 1.98% increase in robusta global export; from 46.92 million bags as at July 2021 to 48.90 million bags as at July 2022; also the world coffee exports was 10.69 million bags as at May 2023 in comparison with 10.54 million as at May 2022 [3]. These are indicators of random rise in acceptance across the world. Coffee beverages though purported to be intoxicating, many people across the globe are ironically attracted to it as a substitute for alcohol [1]. A blog estimated Nigerian consumption in 2020 to be 1,000 tonnes, representing 23% increase from the previous year [4]. This strongly indicate increased acceptance in a predominantly tea and cocoa-drinking nation.

Korkmaz *et al.* [5] noted that sufficient information on the distribution of radionuclides in the environment is crucial for radiation monitoring and protection. Human exposure comes from both natural and manmade radioactive sources. Long-lived naturally occurring radioactive materials (NORM) are present in aquatic and terrestrial environmental samples at different concentrations [6, 7]. These include ^{238}U and its progeny, ^{232}Th and its progeny, and the singly occurring primordial radioisotope of ^{40}K . This enduring and ubiquitous occurrence permits transfer of radioisotopes through the food chain to humans [6, 8]. Similarly, applications of radioisotopes in different human activities like agriculture, mining, construction, medicine, power generation and other processes of industrialization have technically influenced the level of exposure [9, 10]. These releases may be regulated or accidental. Detecting manmade radionuclides in environmental samples is not a usual event during measurements in this environment. This is because identifying manmade radionuclides in any environment depends on the frequency of previous contamination events.

Radionuclides, whether manmade or natural, are drawn by plants through ion channels or specific transporters and through the food chain into humans leading to internal exposure. Therefore, internal radioactive exposure is in direct dependence on the type and amount of food consumed [6, 7]. This dependence has aroused interest in the level of internal exposure resulting from food ingestion around the world [7, 11, 12, 13, 10, 14]. Thus, to determine the quantity of internal exposure, reliable data on radionuclide concentrations in various food samples consumed by members of exposed populations and the general public are important. To this end, the activity of long-lived gamma-emitting radionuclides present in commonly consumed imported coffee brands in Lagos, Nigeria, were assessed using high-purity germanium detector.

2 Materials and Method

Lagos is a metropolis in the geopolitical region of southwestern Nigeria, located between latitude 6.451° and 6.608°N and longitude between 3.3947° and 3.6218°E , covering an area of about 3577 km^2 . Lagos appears on top of the list of most economically viable states in Nigeria. In Lagos, commonly imported coffee brands include bellarom rich roast, alcafe time, marrisons gold roast decaf, rich toast instant grannules, asda smartprice instant coffee medium roast, starcafeinstante, etc. Based on a mixed sampling method involving one-on-one interview with consumers, observations and survey of sales metrics from major traders within Lagos, six of the most preferred imported brands were selected. A pack of each of these imported coffee brands was obtained from major supermarket within the metropolis. The samples were blended with silver crest blender, and sieved with mm sieve to ensure homogeneity of the samples. 500 grammes of each brand were weighed. The weighed samples were kept in plastic bags of an infinitesimal weight for convenience in carrying them to the laboratory. They were later transferred into Marinelli beakers that have been thor-

oughly washed. After ensuring air tightness, the beakers were sealed hermitically with plastic tape, and kept for 28 days to attain secular radioactive equilibrium between ^{232}Th and ^{238}U with their respective progenies. Using Marinelli beakers allows greater geometric detection efficiencies to be obtained since larger quantity of the sample volume are positioned close to the detector. Prepared samples were thus analyzed with high purity germanium (HPGe) detector for reliable and accurate identification of the radionuclides from their passive gamma photon emissions.

The radioactivity measurements were undertaken with a p-type, coaxial, HPGe gamma-ray detector from Canberra (GC8023; serial no. 9744; 78 mm diameter; 69.8 mm length). The spectrometer has energy resolution of 2.3 keV full width at half maximum (FWHM) for the full-energy detection efficiency at 1332.5 keV gamma photopeaks from ^{60}Co and a relative efficiency of 80 percent. Since maintaining low temperature was required for the operation and excellent sensitivity of the HPGe crystal, the detector capsule was continuously cooled at -196°C with liquid nitrogen to minimize the leakage current within acceptable state. The spectrometry system line comprises of a 2002CSL model pre-amplifier with serial number 13000742, the amplifier, high voltage power supply (HVPS), analogue to digital converter (ADC) and a Canberra series 10 plus multichannel analyzer (MCA) model 2024 to register and process each event into pulse spectrum with estimated peak widths for determination of the peak area. Minimizing unwanted interference from ambient background radiation sources, the detector capsule was enclosed vertically in a cylindrical lead shield chamber 5cm in thickness. The efficiency of a detector is said to be a proportionality term which relates the activity of the source to the count number detected [15]. Hence, efficiency calibration of this detector is often carried out with a standard reference source (MGS6M315) from IAEA whilst generating the efficiency curve with GENIE 2000 software.

Determination of the efficiency was such that the centroids of photo peaks energies were fitted with the corresponding channel numbers repeatedly. Absolute photo peak efficiency of the detector for gamma-ray spectrums was determined from the gamma photo peak efficiency and photo peak energy E relationship. Activity concentration of ^{238}U was derived as the average estimation from the photo peaks of ^{214}Bi (609.3 keV; 46.1%) and ^{214}Pb (295.2 keV; 19.3%); that of ^{232}Th was determined as average estimation of the dominant gamma rays of ^{208}Tl (2614.5 keV; 35.7%) and ^{228}Ac (338.3 keV; 11.3%; 911.2 keV; 26.6%). Activity concentration of ^{40}K was derived from 1460.8 keV (10.5%) photo peak. Each sample was counted for a period of 28800 seconds. The activity concentrations, A, of each of these radionuclides present in the coffee samples were deduced with the following equation [7]:

$$A = \frac{N}{mt\varepsilon P_\gamma} \quad 1$$

N is the net counts of the matching photopeak, m is the sample mass in kg, t is the counting time in s, ε is the detector counting efficiency and P_γ is the emission probability per disintegration for specific gamma line. The minimum detectable activity (MDA) was determined with the formula [7]:

$$MDA = \frac{2.71 + 4.66S_b}{\varepsilon P_\gamma mt} \quad 2$$

S_b is the standard error in the net background count rate for the photopeak.

3 Results and Discussion

Based on the counting statistics, the activity concentrations in coffee samples and the uncertainties are presented in Table 1. The activity concentration of ^{238}U were above the minimum detection

(MDA) in all samples except sample 1 with the minimum value of $0.10 \pm 0.04 \text{ Bq kg}^{-1}$ and a maximum value of $0.41 \pm 0.08 \text{ Bq kg}^{-1}$ and all-sample mean of (\pm SD) $0.19 \pm 0.05 \text{ Bq kg}^{-1}$. Founded on the counting statistics also, Table 1 indicates the activity concentrations of ^{232}Th in the coffee samples and the uncertainties. The activity concentrations of ^{232}Th were above MDA in all samples with the minimum value of $0.34 \pm 0.04 \text{ Bq kg}^{-1}$ and a maximum value of $2.39 \pm 0.32 \text{ Bq kg}^{-1}$ and all-sample mean of (\pm SD) $1.31 \pm 0.15 \text{ Bq kg}^{-1}$. The activity concentrations of ^{40}K and the uncertainties based on the counting statistics in the samples of coffee is also shown in Table 1. Activity concentrations of ^{40}K in all samples are above the MDA with maximum value of $985.32 \pm 52.25 \text{ Bq kg}^{-1}$ and minimum value of $778.83 \pm 41.30 \text{ Bq kg}^{-1}$ and all-sample mean of (\pm SD) $877.49 \pm 46.54 \text{ Bq kg}^{-1}$.

The activity concentrations of ^{238}U and ^{232}Th in all the samples are below the respective acceptable limit of 33 Bq kg^{-1} and 45 Bq kg^{-1} according to UNSCEAR [16]. The activity concentration of ^{40}K is however higher than the acceptable limit of 412 Bq kg^{-1} in all the samples by factors ranging from 1.89 – 2.39 indicating that the activity concentrations of natural radionuclides in these coffee samples are dominated by ^{40}K . During beverage extraction processes, significant amount of the micronutrient (potassium) is transferred into the coffee beverage [17]. Hence coffee product may find other useful applications apart from beverages, these include applications such as fertilizer or fertilizer additive for plants. A noticeable different pattern of variations was observed in two samples (sample ID 2 and 4) from different brands of the same origin (Spain); higher activity concentrations for ^{238}U and ^{232}Th in sample with sample ID 2, but lower activity concentrations for ^{40}K compared with sample ID 4. This may result from processing factors inclusive of time, handling, etc which suggests inhomogeneity of coffee brands of same origin.

Table 1: Radionuclide activity concentrations (Bq kg^{-1}) in investigated coffee samples.

Sample ID	Origin	^{238}U	^{232}Th	^{40}K
1	Cyrus	0.41 ± 0.08	1.54 ± 0.11	985.32 ± 52.25
2	Spain	0.16 ± 0.04	2.14 ± 0.28	778.83 ± 41.30
3	Colombia	0.18 ± 0.05	0.34 ± 0.04	940.37 ± 49.87
4	Spain	BDL	0.98 ± 0.07	822.49 ± 43.62
5	United Kingdom	0.10 ± 0.04	0.48 ± 0.06	940.92 ± 49.90
6	Uganda	0.08 ± 0.02	2.39 ± 0.32	796.98 ± 42.27

3.1 Annual intake of radionuclides in coffee (Bq y^{-1})

Annual intake, Y_i , of the radioactivity to an individual through ingestion of coffee is determined with the following formula [18]:

$$Y_i = \frac{A_i \times A_{im} \times F}{P_N} \quad 3$$

Where A_i is the activity concentration of each radionuclide of interest (Bq kg^{-1}), A_{im} is the annual import of coffee ($2,690,000 \text{ kg y}^{-1}$ in 2020), P_N is the population of Nigeria ($208,327,405$ in 2020 according to WHO [19]), F is the fraction of coffee being consumed, taken as 95%.

3.2 Dose Estimation

In principle, there is no way to directly measure the committed dose due to exposure to radionuclides and the accumulation of the radiation dose. Effective dose computing is a convenient way to represent the radiation doses from different radionuclides. This term expresses the risks of radiation induced detriments applying the International Commission on Radiological Protection (ICRP) metabolic model. Applying necessary conversion factors, effective dose resulting from the activity concentrations of radionuclides measured in coffee were estimated as a measure of the ingested doses. The dose conversion factor (*DCF*) of $6.2 \times 10^{-9} Sv Bq^{-1}$ for ^{40}K , $6.9 \times 10^{-7} Sv Bq^{-1}$ for ^{232}Th and $2.8 \times 10^{-7} Sv Bq^{-1}$ for ^{238}U in food samples for adult members of the public [18] were applied in this study. The ingested dose was thus calculated with [18]:

$$D_{eff} = Y_i \times DCF \tag{4}$$

Where D_{eff} ($\mu Sv y^{-1}$) represents the committed effective dose to an individual resulting from consumption of radionuclides over a year. Total committed effective dose resulting from consumption of coffee is simply determined from the summation of the egestion dose due to each radionuclide.

The conversion factors used in the evaluation of the ingestion dose are based on the values suggested by the International Commission on Radiological Protection (ICRP) and United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The ingestion dose from the analyzed samples is lower than the UNSCEAR limit of 120 μSv per year for thorium series and uranium series. This finding also showed that the analyzed samples present lower value of ingestion dose for potassium than the UNSCEAR limit of 170 μSv per year in only one sample. With E_{aveof} 290 $\mu Sv/y$, the coffee samples can be regarded as being within limit of internal exposure to the consumers. It will be of utmost interest to the radiation safety experts, food safety experts, food importers and the general public that further studies are carried out to ascertain the harmfulness of coffee by evaluating the radionuclide contents in coffee beans, coffee powder and the soil where the plant is grown, the transfer ratio, and the level of radiation exposure due to the consumption dose from both coffee beans and coffee beverage, and the effective dose contributed by the soil.

Table 2: Radionuclide activity concentrations ($Bq kg^{-1}$), annual intake ($Bq y^{-1}$) and ingestion Dose ($\mu Sv y^{-1}$) in coffee from different parts of the world.

	Origin	Activity ($Bq kg^{-1}$)			Annual Intake ($Bq y^{-1}$)			Ingestion Dose ($\mu Sv y^{-1}$)		
		^{238}U	^{232}Th	^{40}K	Y_U	Y_{Th}	Y_K	D_U	D_{Th}	D_K
1	Cyrus	0.41	1.54	985.32	5.03 x 10^{-3}	18.89 x 10^{-3}	12.09	1.41 x 10^{-3}	1.30 x 10^{-2}	7.49 x 10^{-2}
2	Spain	0.16	2.14	779.83	1.96 x 10^{-3}	26.25 x 10^{-3}	9.57	5.50 x 10^{-4}	1.81 x 10^{-2}	5.93 x 10^{-2}
3	Colombia	0.18	0.34	940.37	2.21 x 10^{-3}	4.17 x 10^{-3}	11.54	6.18 x 10^{-4}	2.88 x 10^{-3}	7.15 x 10^{-2}
4	Spain	BDL	0.98	822.49	BDL	12.02 x 10^{-3}	10.09		8.29 x 10^{-3}	6.26 x 10^{-2}

	Origin	Activity (Bq kg ⁻¹)			Annual Intake (Bq y ⁻¹)			Ingestion Dose (μSv y ⁻¹)		
		²³⁸ U	²³² Th	⁴⁰ K	Y _U	Y _{Th}	Y _K	D _U	D _{Th}	D _K
5	United Kingdom	0.10	0.48	940.92	1.23 x 10 ⁻³	5.89 x 10 ⁻³	11.54	3.43 x 10 ⁻⁴	4.06 x 10 ⁻³	7.16 x 10 ⁻²
6	Uganda	0.08	2.39	796.98	9.81 x 10 ⁻⁴	29.32 x 10 ⁻³	9.78	2.75 x 10 ⁻⁴	2.02 x 10 ⁻²	6.06 x 10 ⁻²

Different values of activity concentrations for natural radionuclides of ²³⁸U, ²³²Th and ⁴⁰K in different food samples have been reported by literature for different parts of the world (Table 3). Different values of activity concentrations for these natural radionuclides in coffee from different parts of the world are also presented in Table 3. The activity concentrations of ⁴⁰K obtained in this study are higher than that reported for countries like India, Honduras, Guatemala, Mexico, Indonesia, Ethiopia, Uganda, Vietnam, Colombia and Brazil but lower than values recorded in coffee samples from Arab and Turkey. The activity concentrations of ²³⁸U from this study are significantly lower than what was obtained in coffee samples from India, Honduras, Guatemala, Mexico, Indonesia, Ethiopia, Uganda, Vietnam, Colombia, Brazil, Arab and Turkey. It was also observed that sample of coffee of Colombia origin from this study recorded lower activity concentrations when compared with sample of the same origin by a factor of 81.7 for ²³⁸U, 1.9 for ²³²Th but 1.5 higher for ⁴⁰K. The sample of coffee of Uganda origin in this study are however of higher activity concentrations for ²³⁸U, ²³²Th and ⁴⁰K, ranging from BDL to 0.08, and factors of 3.5 and 1.5 respectively than sample of the same origin.

Except for bean, pear and lemon, the Table presents lower activity concentrations for ²³⁸U in all the coffee products in this study than other food samples. Other than coffee sample from Colombia origin, the activity concentrations for ²³²Th in all the samples in this study lie within the data from other food samples. Activity concentrations of ⁴⁰K presented in the Table from this study are clearly above the values obtained in other food samples.

Table 3: Activity concentrations in coffee and other food samples from different origin.

Origin	Ref	Food	Activity concentrations		
			²³⁸ U	²³² Th	⁴⁰ K
Nigeria	[18]	Wheat	3.1	1.2	214
Nigeria	[18]	Rice	1.7	0.7	90
Nigeria	[18]	Millet	2.9	0.9	191
Malaysia	[20]	Rice	2.2	5.4	92
Turkey	[21]	Apple	0.5	0.38	0.34
Turkey	[21]	Grape	8.04	3.22	1.78
Turkey	[21]	Pear	0.29	0.67	40.39
Turkey	[21]	Lemon	0.36	0.45	50.57
Turkey	[21]	Pepper	1.73	0.57	BDL
Turkey	[21]	Bean	0.28	0.78	2.81
Ghana	[22]	Millet	3.91	7.15	216
India	[13]	Coffee	12.30	0.69	666.00

Origin	Ref	Food	Activity concentrations		
			^{238}U	^{232}Th	^{40}K
Honduras	[13]	Coffee	18.20	0.82	622.00
Guatemala	[13]	Coffee	11.40	0.43	574.00
Mexico	[13]	Coffee	10.60	0.75	632.00
Indonesia	[13]	Coffee	26.90	0.97	693.00
Ethiopia	[13]	Coffee	10.00	0.64	564.00
Uganda	[13]	Coffee	BDL	0.69	666.00
Vietnam	[13]	Coffee	13.90	0.77	704.00
Colombia	[13]	Coffee	14.70	0.65	626.00
Brazil	[13]	Coffee	40.90	1.51	718.00
Arab	[23]	Coffee	44.61	4.31	1041.24
Turkey	[23]	Coffee	22.52	3.12	1523.10
Cyrus	Present Study	Coffee	0.41	1.54	985.32
Spain	Present Study	Coffee	0.16	2.14	779.83
Colombia	Present Study	Coffee	0.18	0.34	940.37
Spain	Present Study	Coffee	BDL	0.98	822.49
United Kingdom	Present Study	Coffee	0.10	0.48	940.92
Uganda	Present Study	Coffee	0.08	2.39	796.98
UNSCEAR	(2000)	Food	67	82	310

3.3 Excess lifetime cancer risk (ELCR)

Radiation cancer risks are described by the terms, morbidity and mortality. Carcinogenic detriments characterized by evaluating the probability of cancer in any exposed population for specific lifetime. The carcinogenic risk resulting from coffee consumption was evaluated based on the procedure exemplified in US Environmental Protection Agency, EPA, [23]:

$$ELCR = \sum_{i=^{238}\text{U}, ^{232}\text{Th}} (Y_i \times R_{c,i}) \times A_{ls} \quad 5$$

Based on WHO [19] data, life expectancy at birth has improved from 53.6 years in year 2000 to 62.6 years in year 2019 in Nigeria. Average life expectancy $A_{ls} = 62.6$ was taken to be the average life span in Nigeria. The mortality risk coefficients, $R_{c,i}$, list for all radionuclides resulting from the dietary intake of food indicates the likelihood of radiogenic outcomes per unit intake with the intake average taken over all ages and both sexes. In this study, mortality risk coefficients of $9.56 \times 10^{-9} \text{ Bq}^{-1}$ and $2.74 \times 10^{-8} \text{ Bq}^{-1}$ were taken for ^{238}U and ^{232}Th respectively. Micronutrients are essentially regulated based on amount of intake. The potassium concentration in the human body is homeostatically regulated. This implies that for a healthy person and under normal situations, the concentration of potassium in human system is not a factor of its intake, more so that the intake is accompanied by excretion. Therefore, the intake of ^{40}K does not contribute to the ELCR and was not accounted for in this study. The outcome of this study indicates that the total ELCR resulting from ^{238}U and ^{232}Th is 6.46×10^{-8} which is significantly minimal compared to the 4.5×10^{-3} ICRP cancer risk factor [18]. Based upon the 1 mSv annual above background dose threshold for the general public, annual mortality likelihood of 10^{-5} equivalent to 1 in 100,000, this study presents mortality probability of 1 in 100,000,000.

4 Conclusion

This study reflects the first attempt towards assessing activity concentrations of ^{238}U , ^{232}Th and ^{40}K long-lived gamma-emitting natural radionuclides in coffee beverage brands imported from different origins to Lagos Nigeria through hyper-pure germanium gamma spectrometry. The findings in this study verifies that the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in the studied samples have elevated values for ^{40}K , above the limit suggested by UNSCEAR. This indicate coffee beverage as being rich in potassium and a good source of potassium micronutrient to the body. The outcomes of this study also give credence to the opinion that coffee may find suitable application as plant fertilizer or fertilizer additive. The ingestion dose evaluated for the activity concentrations of ^{40}K suggests higher dose of exposure. Measures should be prioritized in evaluating the radionuclide contents in coffee beans, coffee powder and the soil where the plant is grown, the transfer ratio, and the level of radiation exposure due to the ingestion dose from both beans and beverage, and the effective dose contributed by the soil to ascertain the harmfulness of coffee.

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6 Author contribution statement

Research plan, field works, sample preparation and analysis, and manuscript preparation were carried out by M.A. Olaoye, O.P. Abodunrin and B.A. Okuwobi.

7 Declaration of competing interest

The authors declare that there is no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper.

8 Data availability

Data will be made available on request.

9 Copyright Notice

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