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Evaluation of the Natural Radioactivity Levels and Radiological Assessment of Nuts Commonly Consumed in Ile-Ife, South West, Nigeria

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Abstract: In this study, the naturally occurring radionuclides and their associated radiological hazards were assessed in commonly consumed nuts in Ile-Ife, South west, Nigeria. Samples of different species of nuts were obtained in Ile-Ife, Osun State, Nigeria and their activity levels were determined using gamma ray spectrometer. The results showed that the mean activity of the identified radionuclides, ^{40}K, ^{232}Th and ^{238}U are 67.36, 37.32 and 3.88 Bq/kg respectively. The radiological indices, ADR (absorbed dose rate), AEDR (annual equivalent dose rate), H_{in} (internal hazard index), H_{ex} (external hazard index), AGDE (annual gonadal dose equivalent), $I_{\gamma r}$ (representative gamma index) and Ra_{eq} (radium equivalent) have values of 22.39 nGy/hr, 0.19 mSv/yr, 0.23 Bq/kg, 0.12 Bq/kg, 152.66 μsvy^{-1}, 0.33 and 48.04 respectively. The radiological assessment suggests that there are no radiological hazards associated with the consumption of the nuts.

Keywords: Activity Concentration, Gamma Index, Gamma Ray Spectrometer, Nuts, Radiation

1. Introduction

Natural radioactivity is caused by the presence of natural occurring radioactive matter (NORM) in the environment. Examples of natural radionuclides include singly isotopes of potassium (^{40}K), uranium (^{238}U and its decay series), and thorium (^{232}Th and its decay series). In addition to being long-lived (in the order of 10^{10} years), these radionuclides are typically present in air, soil, and water in different amounts and levels of activity. Soil not only acts as a source of continuous radiation exposure to humans but also as a medium of migration for the transfer of radionuclides to biological systems [1]. NORM is inherent of many geologic materials and is consequently encountered during geologically related activities. Radiation can cause sterility, making reproduction impossible. It can also cause mutations in developing embryos, which are usually detrimental or even fatal [2]. Natural radionuclides are found in terrestrial and aquatic food chains, with subsequent transfer to humans through ingestion of food. As such, international efforts were brought together collaboratively to apply adequate procedures in investigating radionuclides in food [3], and to set essential guidelines to protect against high levels of internal exposure that may be caused by food consumption [4]. Foodstuffs contain both natural and man-made radionuclides, which after ingestion lead to an effective internal dose. It has been estimated that at least one-eighth of the mean annual effective dose of natural sources occurs by the consumption of foodstuff [5]. For contamination assessment of foodstuff consumed by humans, it is crucial to identify the baseline value, or the level of radiation dose of natural and synthetic radionuclides. Nuts and seeds are popular products consumed all over the world since they are rich in nutrients such as essential minerals, vitamins, proteins, carbohydrates, and unsaturated fatty acids [6]. Nuts of different species are commonly consumed in Nigeria ranging from kola nut to bitter kola to walnut etc. It is presumed that there are radionuclides present in the soils in

which these nuts are cultivated, as a result of the natural radioactivity of the earth. It is therefore imperative to investigate the radioactivity levels of these nuts so as to ascertain the radiological hazards associated with their consumption.

2. Materials and Methods

2.1. Study Collection and Preparation

Five (5) different species of nuts were collected in Ile-Ife, Osun State, Nigeria. Several samples of each species were collected and transported to the laboratory for analysis. To avoid contamination during sampling, transportation and storage, the nut samples were kept in polyethylene bags and taken immediately to the laboratory for analysis. The samples of each species were pulverized using an agate mortar and pestle so as to form a composite sample. The pulverized samples were then passed through a filter sieve to achieve uniform particle size.

2.2. Radioactivity Counting in the Sample

Radioactivity measurement was conducted on the samples. For purposes of the gamma-ray spectrometric study, the samples were homogenized and dried in free air in the laboratory until constant weight was achieved. The samples were properly covered with plain sheet to prevent air particulate contamination. They were then sealed for a minimum of 28 days using a cylindrical plastic container wrapped with a masking tape so as to achieve radiative secular equilibrium before being counted using the gamma-ray spectrometer located at the Department of Physics and Engineering Physics, Obafemi Awolowo University, Ile-Ife, Nigeria. Gamma ray spectrometry was carried out by counting each sample for 10 hours using a well-calibrated Cesium Iodide (CsI) scintillation detector and processing the spectra using a Universal Radiation Spectrum Analyzer (URSA II). The activity concentration of the radionuclides was computed. The obtained result was then used to determine the radiological health impact of the samples.

2.3. Energy Calibration of the Cesium Iodide Detector CsI (Tl)

The term energy calibration simply means assigning a channel number to radionuclides of known energy. The energy calibration is to derive a relationship between peak position in the spectrum and the corresponding gamma-ray energy. Energy calibration is accomplished by measuring the spectrum of a source emitting gamma-rays of precisely known energy and comparing the measured peak position with energy. Efforts were made to ensure that the calibration energies covered the entire range over energies expected in this work. The detector was calibrated in terms of energy using three-point sources (Ba-133, Cs-137 and Co-60) using a gamma emitter sample of known energy that allowed determining a linear equation relating gamma energy to channel number:

$$y = mx + c \quad (1)$$

Where y corresponds to the gamma energy expressed in keV

m is the slope of the calibration line expressed in keV/channel

x is the relative channel

c is the intercept on the energy axis expressed in keV

The efficiency calibration of the detector was done implicitly using reference soil sample (ENV 94084) from Rocketdyne Laboratory which was used as comparator standard for evaluation of activity in the samples

2.4. Determination of Peak Areas

The peak corresponding to 1460 keV for K-40, 352 keV (Pb-214) for U-238 and 240 keV (Ra-224) for Th-232 were used for the estimation of natural radionuclides in all the samples. The integrated counts recorded under the energy peaks 1460, 352 and 240 keV were noted for each spectrum.

2.5. Calculation of Radionuclide Concentration

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, the activity concentrations in the samples were obtained using the methods of Mokobia *et al.,* 2003 and Jibiri and Emelue, 2008 [7, 8].

$$C\ (Bq/kg) = kC_n \quad (2)$$

Where $K = \frac{1}{EP\gamma Ms}$, C is the activity concentration of the radionuclide in the sample given in Bq/kg, Cn is the count rate under the corresponding peak, E is the detector efficiency at the specific gamma-ray energy, P_γ is the absolute transition probability of the specific gamma-ray, and M_s is the mass of the sample (kg).

The detection limit of a measuring system describes its operating capability without the influence of the sample. The detection limit (DL) given in Bq/kg which is required to estimate the minimum detectable activity in a sample, was obtained:

$$DL\ (Bq/Kg) = 4.65\frac{\sqrt{Cb}}{tb}.K \quad (3)$$

Where C_b is the net background count in the corresponding peak, t_b is the background counting time (s), k is the factor that converts cps (counts per second) to activity concentration (Bq/kg) [9].

2.6. Data Treatment

Data treatment for radiological assessment of the samples was done using total absorbed dose rate (ADR), internal hazard index (Hin), external hazard index (Hex), annual gonadal dose equivalent (AGDE), representative level index ($I_{\gamma r}$), annual effective dose rate (AEDR) and radium equivalent (Raeq).

The absorbed dose rate (ADR) in air from external gamma radiation at 1 m above ground level due to the presence of uniformly distributed natural radionuclides in measured samples is calculated according to UNSCEAR, 2000 [4]:

$$ADR\ (nGy/hr) = 0.0417A_K + 0.462A_U + 0.604A_{Th} \quad (4)$$

A_K, A_U and A_{Th} are the activity concentrations (in Bq/kg) for ^{40}K, ^{232}Th (as ^{208}Ac) and ^{238}U (as ^{214}Bi) respectively. Secular equilibrium was assumed for the dose calculation.

The radiological hazard for populations consuming the nuts is evaluated in terms of the Annual Effective Dose Rate (AEDR) which accounts for the dose rate of biological effectiveness in causing damage to human tissues [10]. The annual effective dose due to the natural radionuclides in the nut samples was estimated using the dose conversion coefficient that convert the absorbed dose rate in air to the effective dose (0.7 Sv/Gy) and the outdoor occupancy of 0.2 (average of 4.8 hr spent in the site every day for a year) as proposed by UNSCEAR, 2000 [4]. The AEDR was calculated using the formula given by Jibiri and Emelue, 2008 [8].

$$E = TfQD\varepsilon \quad (5)$$

where E is the effective dose rate (mSv/yr), T is the time in seconds in a year (8760), f is the occupancy factor which corrects the average time spent outdoors in the sites (0.2), Q is the quotient of the effective dose rate and absorbed dose rate in air (0.7 Sv/Gy), ε is the factor converting nano (10^{-9}) into micro (10^{-6}); and D is the absorbed dose rate in air (nGy/hr).

The internal exposure to ^{222}Rn and its hazardous progenies to the respiratory organs is controlled by the internal hazard index (H_{in}) as described below [4]:

$$H_{in} = (A_{Ra}/185 + A_{Th}/259 + A_K/4810) \leq 1 \quad (6)$$

The A_{Ra} is replaced by A_U in the calculations. It may be noted that ^{238}U is replaced by the decay product ^{226}Ra, although there may be disequilibrium between ^{238}U and ^{226}Ra. It is given as 1.03 by UNSCEAR, 2000 [4]. The factors which determine the exposure rate of an individual are the concentration of radionuclides in the nuts and the time spent outdoors [11]. A widely used hazard index reflecting external exposure called the external hazard index H_{ex} is defined as follows:

$$H_{ex} = (A_{Ra}/370 + A_{Th}/259 + A_K/4810) \leq 1\ [4] \quad (7)$$

The gonads, the active bone marrow and the bone surface cells are considered as the organs of interest [4]. Therefore, the Annual Gonadal Dose Equivalent (AGDE) for the residents of the study area due to specific activities of ^{226}Ra, ^{232}Th and ^{40}K is calculated using the following relation:

$$AGDE\ (\mu Svy^{-1}) = 3.09\ A_{Ra} + 4.18\ A_{Th} + 0.314\ A_K\ [12] \quad (8)$$

Another radiation hazard index called the representative gamma index $I_{\gamma r}$ is given by:

$$I_{\gamma r} = A_{Ra}/150\ Bqkg^{-1} + A_{Th}/100\ Bqkg^{-1} + A_K/1500\ Bqkg^{-1} \quad (9)$$

Activity mass concentration due to ^{226}Ra, ^{232}Th and ^{40}K is not uniformly distributed throughout in the nuts [13]. This non-uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}). This is a single quantity that takes into consideration the radiation hazard associated with ^{226}Ra, ^{232}Th and ^{40}K. It is calculated as:

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (10)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg respectively. The A_{Ra} is replaced by A_U in the calculations.

3. Results and Discussion

3.1. Activity Concentrations of Radionuclides in the Nuts

The radioactive daughters/products of ^{238}U are ^{214}Bi, ^{214}Pb and ^{226}Ra; likewise, the daughters of ^{232}Th are ^{208}Ti, ^{224}Ra and ^{228}Ac. ^{40}K is also naturally occurring but a non-decay series radioactive isotope. Using the gamma spectrometric analysis, three naturally occurring radionuclides namely ^{40}K, ^{232}Th (as ^{224}Ra) and ^{238}U (as ^{214}Pb) were determined in the nuts. The result of average activity concentrations with ±σ uncertainty (in Bq/kg) for the non-series of ^{40}K and that of decay series of ^{232}Th (^{224}Ra) and ^{238}U (^{214}Pb) in nuts is shown in Table 1. In the nuts, the mean activity concentrations of ^{40}K, ^{232}Th and ^{238}U are 67.36 ± 1.52 Bq/kg, 3.88 ± 0.41 Bq/Kg and 37.32 ± 1.22 Bq/Kg respectively. Among the three radionuclides, ^{40}K has the highest activity while ^{232}Th has the lowest activity.

Table 1. *Activity Concentrations of the Identified Radionuclides in the Nuts (Bq/kg).*

Nut Samples	^{40}K	^{232}Th	^{238}U
A	66.56 ± 0.90	36.46 ± 0.92	3.81 ± 0.25
B	66.59 ± 0.88	36.26 ± 1.03	4.04 ± 0.41
C	74.43 ± 2.66	41.55 ± 2.06	4.18 ± 0.56
D	63.27 ± 2.02	35.60 ± 1.31	3.45 ± 0.65
E	65.99 ± 1.17	36.72 ± 0.76	3.90 ± 0.18
Mean ± SD	67.36 ± 1.52	37.32 ± 1.22	3.88 ± 0.41

SD = standard deviation

3.2. Radiological Assessment of the Radionuclides in the Nuts

The radiological assessment of the nuts is presented in Table 2. The mean absorbed dose rate of the nuts is 22.39 nGy/hr. The dose rate value is less than the population weighted average absorbed dose rate (60 nGy/hr) in outdoor air from terrestrial gamma radiation [4] and that of the worldwide average of 55 nGy/hr [14]. The mean annual equivalent dose rate is 0.19 mSv/yr which falls lower than the range for individual countries (0.3 - 0.6 mSv/yr range) [4, 15]. World permissive annual dose limit is 1.0 mSv/yr [16]. The annual dose rate of the radionuclides in the nuts is below the world permissive annual dose limit. This indicates that the dose rate

of the nuts, upon consumption, may not lead to respiratory diseases such as asthma and cancer or external diseases such as erythema, skin cancer and cataracts. The internal hazard index is 0.23 Bq/Kg while the external hazard index is 0.12 Bq/Kg on the average. The values of the indices (H_{ex}, H_{in}) must be less than 1.0 for the radiation hazard to be insignificant [17]. The internal hazard and external hazard indices of the studied nuts in this study are both less than 1 and so, the radiation hazard is insignificant. The mean annual gonadal dose equivalent of the radionuclides is 152.66 μSvy^{-1}. This value is less than the world average of 300 μSvy^{-1} [4]. The mean representative gamma index value of the nuts is 0.33. This value is less than unity and this suggests that the nuts exhibit low gamma radiation. The estimated mean value of the Ra_{eq} in this present study is 48.04 Bq/kg. This value is lower than the world average of 370 Bq/kg [4]. The radiological assessment carried out using the above parameters all suggest that the investigated nuts exhibited low gamma radiation.

Table 2. *Radiological Assessment of the Radionuclides in the Nuts.*

Nut samples	ADR (nGy/hr)	AEDR (mSv/yr)	H_{in} (Bq/Kg)	H_{ex} (Bq/kg)	AGDE (μsvy^{-1})	$I_{\gamma r}$	Ra_{eq}
A	21.92	0.19	0.22	0.12	149.48	0.32	47.03
B	21.96	0.19	0.22	0.12	149.83	0.32	47.16
C	24.82	0.21	0.25	0.14	169.23	0.36	53.25
D	21.16	0.18	0.21	0.12	144.29	0.31	45.40
E	22.07	0.19	0.22	0.12	150.48	0.32	47.37
Mean ± SD	22.39	0.19	0.23	0.12	152.66	0.33	48.04

SD = standard deviation

3.3. Comparison of the Activity Concentrations in this Study with Similar Studies

The comparison of the activity concentrations of the radionuclides in the studied nuts with other nuts grown and consumed elsewhere is presented in Table 3. ^{40}K in the present study is less than the nuts reported by Martins *et al.,* 2012 and Ezzulddin, 2017 [18, 19]. The ^{232}Th in this study is greater than those reported by Ezzulddin, 2017 [19] while the ^{238}U is less than those reported by Martins *et al.,* 2012 [18]. This variation observed in the activity concentrations is probably due to the varying climatic and geological properties of the soils in which these nuts are grown. The relatively low ^{40}K observed in this present study could be due to the absence of minerals such as k-feldspars, mica and illite, in the soils in which these nuts are grown, as rare metal pegmatites have been reported to contain a significant amount of ^{40}K.

Table 3. *Comparison of the Activity Concentrations in this Study with Similar Studies.*

Radionuclides	This study	[18]	[19]
^{40}K	67.36	306.31	282.61
^{232}Th	37.32	-	0.151
^{238}U	3.88	56	-

4. Conclusion

Commonly consumed nuts were analyzed for their radioactivity levels using Gamma ray spectrophotometer. The results of the obtained activity concentrations were analyzed using various radiological indices such as absorbed dose rate, annual equivalent dose rate, internal hazard index, external hazard index, annual gonadal dose equivalent, representative gamma index and radium equivalent. The radiological indices confirm that the nuts exhibited low gamma radiation and its consumption may not pose serious radiological effects. However, long-term exposure via consumption should be discouraged to prevent radiation buildup.

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