Determination of Natural Radioactivity in Nuts and Seeds and their Radiological Implications in the Human Body

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ABSTRACT

Naturally occurring radionuclides in nuts and seeds commonly consumed in Windhoek, Namibia were examined. This was done by measuring the activity concentrations of 40K, 226Ra and 232Th in these nuts and seeds resulting in the determination of their associated radiological hazards. Ten samples of nuts and seeds of different species were purchased from shop outlets around Windhoek and a gamma ray spectrometer was used to determine their activity levels. The average activity concentration of the radionuclides 40K and 226Ra were found to be 101.295.55 Bq/kg and 0.80 0.35 Bq/kg respectively, 226Ra was only found in some samples. 40K was found to be very high in most of the samples with the maximum specific activity of 122.22Bq/kg in almond nuts. The maximum specific activity of 226Ra was found to be 4.19 Bq/kg in cashew nuts. However, this did not exceed the permissible limit. The specific activity of 232Th was Below Detection Limit (BDL) in all the samples. The annual effective dose equivalent in all samples was found to be below the limit of 1mSvy-[1]. All the radiological assessments made in this study showed that there were no radiological risks associated with ingestion of nuts and seeds sold in Namibia.

Keywords: Natural radioactivity, radiological hazards, nuts, seeds, annual effective dose.

1.0 INTRODUCTION

Radionuclides have always been present in our environment since the formation of the earth. They can be produced by Naturally Occurring Radionuclide Materials (NORMs) that undergo spontaneous decay to form daughter nuclei. This decay process is accompanied by the emission of one or more types of radiation (Duppen and Andreyev, 2018; ANL, 2005). It can be ionizing or non-ionizing. This ionizing nature of these radiations is what makes them hazardous to health (UNSCEAR, 2000).

Almost all NORMs contain radionuclides that continue to decay forming a decay chain. For instance, the decay chain of ^{238}U contains several radioactive isotopes before ending with a stable isotope of ^{206}Pb. ^{235}U decays in a chain until it produces a stable isotope of ^{207}Pb. The decay chain of ^{232}Th also contains several radioactive isotopes, before ending in another stable isotope of ^{208}Pb (Sanchez-Lorenzo *et-al*, 2015). ^{40}K is another, but non series radionuclide that is ubiquitous in the environment. One of the major sources of human exposure to ionizing radiation is $^{238}U(^{226}Ra)$, that constitute 55.8 %, followed by ^{232}Th that constitute 14 % and lastly ^{40}K that constitute 13.8 % in the environment. (Župunski et al., 2010).

The human body can be exposed to these radionuclides via ingestion, inhalation or dermal pathways. Exposure to these radionuclides may lead to cell transformation, chromosome aberrations, and mutations as a consequence of DNA damage (UNSCEAR, 2000). These radiation effects may eventually lead to cancer of the tissue or organ (Manley et al., 2008). The primary health risk of concern when dealing with NORMs, is the potential for developing cancer. An increase in exposure to ionizing radiation may increase the probability of developing cancer.

NORMs are found in environmental substances such as soils, water, rocks and foodstuffs (Ademola and Morakinyo, 2020; Görür et al., 2012). It is believed that about 15 % of uranium that human beings ingest come from food (Görür et al., 2012). This may lead to kidney damage. Similarly, if thorium and potassium is ingested in large amounts, it will create radiation damage to the tissues and organs of the body (Ashikun et al., 2018). Studies have shown that approximately 12.5 % of the total radiation exposure comes from the food humans consume (Görür et al., 2012). The radionuclides of ^{40}K, ^{226}Ra and ^{232}Th and their daughters are the most common radionuclides found in food and water (Ashikun et al., 2018). In order to understand the possibility of developing any radiological risk, knowledge of radionuclides present in foodstuffs is important. The major foodstuffs that contribute the most to radiation hazard are those that are ingested in large quantities. Such items include water, milk, meat, beans, nuts and some vegetables. In Namibia, people tend to eat nuts extensively in their diet. Therefore, radiological assessment of the levels of radionuclides in nuts and seeds is of paramount importance.

In Namibia, no surveys of radiation dose through the consumption of nuts and seeds has been carried out and no information on NORMS in nuts and seeds has been reported. The results of this study will assist regulatory authorities in creating baseline data to develop future guidelines for radiological protection of the population in the country. It was therefore the aim of study to determine levels of ^{40}K, ^{226}Ra and ^{232}Th in nuts and seeds and their radiological effects in the human body.

2.0 MATERIALS AND METHODS

Five samples each of the seeds and nuts were purchased from the shops around Windhoek, Khomas region Namibia. Windhoek is the capital city of Namibia that covers a land mass of about 5133 km^2. It has a population of about 431 000 according to Namibian population census 2011 (Namibia Statistics Agency, 2013). Figure 1 shows the location of Windhoek where the samples were collected.

Fig 1: Namibian map showing Windhoek

Sample Preparation

The samples of seeds purchased were the redskin's giant, almond, macadamias, cashew and cocktail, while the seeds purchased were linseeds, sunflower, sesame, moringa, and green pumpkin seeds (Table 1). The samples were taken to the laboratory at the Namibia University of Science and Technology (NUST) for further analysis.

Table 1: Names of seeds and nuts and country of origin.

Name of sample	Sample code	Scientific name	Origin
Almond	S_1	Prunus dulcis	South Africa
Cashew	S_2	Anacardium occidentale	South Africa
Cocktail	S_3	Tudy jernstedt	South Africa
Green pumpkin	S_4	Cucurbita maxma	South Africa
Linseed	S_5	Linum usitatissium	South Africa
Macadomias	S_6	Macadamia	South Africa
Moringa	S_7	Moringa Oleifera	Namibia
Redskin giant	S_8	Arachis hypogaea	South Africa
Sesane seeds	S_9	Sesamum indicum	South Africa
Sunflower seeds	S_{10}	Helianthus	South Africa

Sample Analysis and Gamma Spectroscopy

The samples were grinded using a pulveriser and left in an open space in the laboratory to dry due to their high oil content. The samples were then weighted, placed in Nalgene bottles and sealed for more than 31 days to attain secular equilibrium before performing the radioisotope analysis (Onjefu et al., 2017). Every bottle had three key pieces of information written on them: the date of sealing, the sample name and the quantity. For the determination of natural radioactivity concentrations in seeds and nuts samples; a multi-channel analyser (MCA) was connected to the High Purity Germanium detector (HPGe) which has a relative efficiency of 45 % and an energy resolution of 2.0 keV for 1332 keV gamma ray emission of ^{60}Co. To reduce the effects of background radiation the detector was placed in the lead shield 64.80 mm thick, coated with tin and copper (1 mm and 1.6 mm thick). Gamma ray spectrometry system was calibrated using a mixture of radioactive sources ^{60}Co, ^{88}Y, ^{85}Sr, ^{57}Co, ^{113}Sn and ^{137}Cs. Activity concentration of ^{238}U, ^{232}Th and ^{40}K was respectively determined by the energy 609.31 keV (^{214}Bi), 351.92 keV (^{214}Pb), 727.17 keV (^{212}Bi) and 1460.3 keV (^{40}K).

The measurement of gamma radiation is based on the principle that radiation gives off part of its energy to the medium of the detector either by ionizing it directly, or by causing the emission of its charged particle which in turn produces ionization in the medium (Hassan, 2012). After amplification and shaping by the amplification system, the output of the radiation detector will be proportional to the observed energies sent to a pulse height analyzer.

Activity Concentration (A)

The activity concentrations of each radionuclide in the samples was calculated using Equation 1.

$$A = \frac{C_n}{m\,\varepsilon P_\gamma} \quad (1)$$

Where A is the specific activity in (Bq/kg^{-1}) of each radionuclide in foodstuff. C_n is the net count rate of nuclide in counts per second, is the efficiency of specific gamma ray, m is the mass of the sample and is the absolute transition probability of the specific gamma ray. The world accepted criteria of A for ^{226}Ra, ^{232}Th and ^{40}K are 35, 35 and 370 Bqkg^{-1}, respectively (Darwish *et al.*, 2015).

Daily Intake (D_{int})

The activity concentrations of the radionuclides (^{226}Ra and ^{232}Th and ^{40}K) were used to calculate the daily intake of radioactivity due to the consumption of nuts and seeds by using equation 2 proposed by Khandaker *et al.*, (2013).

Where D_{int} is the daily intake of radionuclides in Bq.kg^{-1}, A_{ig} is the activity concentration of radionuclides in Bq.kg^{-1}, A_c is per capital per year consumption of seeds or nuts.

$$D_{int} = \frac{A_c \times A_{ig}}{Y_d} \quad (2)$$

Annual Committed Effective Dose (E_{eff})

Equation 3 was used to compute the annual effective doses due to the consumption of nuts and seeds.

$$E_{eff} = A_C \times A_i \times RF \quad (3)$$

Where E_{eff} is the annual committed effective dose in $mSvy^{-1}$, A_c is the activity concentration ($Bqkg^{-1}$), A_i is the consumption rate per year, RF is the Risk Factor (0.05 Sv^{-1}). It is assumed that the average person consumes 50g of nuts per day or about 1.83 kg/year (Al-Ghamdi, 2018).

Alpha Index (I_α)

The alpha index (internal index) (I_α) measures alpha radiation at a very high intensity. The alpha index was calculated using the equation 4:

$$I_\alpha = \frac{A_{Ra}}{200} \quad (4)$$

Where A_{Ra} is the specific activity of $^{226}R_a$

Gamma Index (I_γ)

The gamma radiation hazard index (I_γ) also known as the representative level index was calculated using equation 5:

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_k}{3000} \quad (5)$$

Where A_{Ra} A_{Th} A_k are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

Radium Equivalent Activity (Ra_{eq})

Gamma radiation exposure is often measured in radium equivalent activity (Ra_{eq}), given by equation 6:

$$Ra_{eq}(Bq\,kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.0417A_k \leq 370 \quad (6)$$

Where A_{Ra} is the activity concentration of radium, A_{Th} is the activity concentration of thorium, A_k is the activity concentration of potassium and 370 Bq kg^{-1} is the the permissible maximum value of the radium equivalent activity. Both the exterior and internal doses from radon and its progeny are connected to the radium equivalent.

Internal Hazard (H_{in})

The internal hazard index regulates the internal exposure of the respiratory organs to ingested radionuclides and their hazardous progenies. H_{in} was calculated from equation 7.

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad (7)$$

Where A_{Ra} is the activity concentration of radium, A_{Th} is the activity concentration of thorium, A_k is the activity concentration of potassium and unity is the permissible maximum value of the internal hazard activity.

Annual Gonadal Dose Equivalent (AGDE)

The Annual Gonadal Dose Equivalent (AGDE) for individuals due to the consumption of certain radionuclides (^{40}K, ^{226}Ra, ^{232}Th) was computed using the following formula in equation 8:

$$AGDE = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_k \quad (8)$$

Where A_{Ra} is the activity concentration of radium, A_{Th} is the activity concentration of thorium, A_K is the activity concentration of potassium.

Excess Lifetime Cancer Risk (ELCR)

This represents the lifetime risk of developing cancer at a given level of exposure, The ELCR has been calculated using equation 9 proposed by (Monica *et al*., 2017).

Where AEDE is the annual committed dose equivalent, DL is the duration of life (70 years) and RF is the Risk Factor (0.05 Sv^{-1}) (Senthilkumar & Narayanaswamy, 2016)

$$ELCR = AEDE \times DL \times RF \quad (9)$$

3.0 RESULTS AND DISCUSSION

Table 2: Activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the nuts and seed samples.

Sample name	Activity concentration (Bq/k		
	^{40}K	^{226}Ra	^{232}Th
Almonds	122.22 ± 4.89	BDL	BDL
Cashew	115.45 ± 4.67	BDL	BDL
Cocktail	103.01 ± 4.43	1.44 ± 0.39	BDL
Green pumpki	116.13 ± 4.62	BDL	BDL
Linseed	101.17 ± 4.00	BDL	BDL
Macadomias	69.22 ± 3.58	BDL	BDL
Moringa	104.51 ± 4.82	1.73 ± 0.36	BDL
Redskin giant	113.79 ± 4.90	4.19 ± 0.29	BDL
Sesane seeds	76.71 ± 3.77	BDL	BDL
Sunflower seec	90.73 ± 4.09	0.60 ± 0.34	BDL
Minimum	69.22 ± 3.58	BDL	BDL
Maximum	122.22 ± 4.89	4.19 ± 0.29	BDL
Mean	101.29± 5.55	0.80 ± 0.35	BDL

BDL Below Detection Limit

The results of the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the ten nuts and seeds samples consumed in Namibia are presented in Table 2. ^{40}K was detected in all samples and showed high activity concentrations. The maximum detectable specific activity of ^{40}K was found in Almond as 122.22 ± 4.89 Bq/kg and the minimum detectable specific activity was found in macadamia nuts as 69.22 ± 3.58 Bq/kg. The activity of ^{226}Ra was found to be very low and it was only detected in four samples (cocktail nuts, moringa seeds, redskin giant and in sunflower seeds). The maximum detectable specific activity of ^{226}Ra was found in redskin giant as 4.190.29 Bq/kg and the minimum detectable specific activity in sunflower seeds as 0.60 0.34 Bq/kg while ^{232}Th was found to be Below Detection Limit (BDL). The highest activity concentrations for potassium were expected, because potassium is an essential microelement for living organisms and is present in all samples from the environment at high levels. The highest activity concentration of ^{40}K in the samples (almond, cashew, cocktail, green pumpkin, linseed, moringa seeds and red skin giant) may be because they have a large capacity to absorb radioisotopes in the soil. However, their concentration does not exceed the global acceptable concentration of 412 Bq/kg as recommended by UNSCEAR 2000.

The activity concentration of ^{226}Ra depends on the radium content of the soil. The mean value for ^{226}Ra was found to be 0.800.35 Bq.kg^{-1}. The activity concentrations for ^{226}Ra detected in all samples were below the world median limit of 42 Bq.kg^{-1} for ^{226}Ra (UNSCEAR, 2000).

Table 3: Annual committed effective dose (E_{eff}) and the Daily intake (D_{in}) of ^{40}K and ^{226}Ra due to the consumption of nuts.

Sample name	Annual committed effective dose, E_{eff} (μSv.y^{-1})		Daily intake (D_{in}) (Bq.d^{-1})	
	^{40}K	^{226}Ra	^{40}K	^{226}Ra
Almonds	$1.93x10^{-3}$	ND	14.73	ND
Cashew	$1.82x10^{-3}$	ND	13.92	ND
Cocktail	$1.62x10^{-3}$	$2.27x10^{-5}$	12.42	0.17
Green pumpkin seeds	$1.83x10^{-3}$	ND	14.00	ND
Linseeds	$1.59x10^{-3}$	ND	12.20	ND
Macadamias	$1.09x10^{-3}$	ND	8.34	ND
Moringa	$1.65x10^{-3}$	$2.73x10^{-5}$	12.60	0.21
Redskin giant	$1.79x10^{-3}$	$6.60x10^{-5}$	13.72	0.51
Sesane seeds	$1.21x10^{-3}$	ND	9.25	ND
Sunflower	$1.43x10^{-3}$	$9.42x10^{-6}$	10.94	0.07
Minimum	$1.09x10^{-3}$	ND	8.34	ND
Maximum	$1.93x10^{-3}$	$6.60x10^{-5}$	14.73	0.51
Mean	$1.60x10^{-3}$	$1.25x10^{-5}$	12.21	0.10

ND – No Detection

Annual committed effective dose and daily intake

The calculated annual committed effective doses (E_{eff}) in nuts and seeds samples varied from $1.09x10^{-3}$ μSv.y^{-1} to $1.93x10^{-3}$ μSv.y^{-1} for ^{40}K and from below detection limit to $6.60x10^{-5}$ μSv.y^{-1} for ^{226}Ra. The annual committed effective dose for ^{232}Th was below detection limit. The values for the annual committed effective doses for both ^{40}K and ^{226}Ra were found to be lower than the recommended limit of 1 mSv.y^{-1}. The Daily Intake due to the consumption of radionuclides (^{40}K and ^{226}Ra) through ingestion of nuts and seeds varied from 8.34 Bq.d^{-1} to 14.73 Bq.d^{-1} for ^{40}K and the minimum for ^{226}Ra was below detection up to 0.51 Bq.d^{-1}. The calculated value for daily intake for potassium-40 was found to be significantly higher than that of ^{226}Ra in all samples.

Table 4: Radium equivalent, internal hazard index, annual gonadal dose equivalent, alpha index Gamma index and excess lifetime cancer risk.

Sample	Radium equivalent (Ra_{eq})	Internal hazard index (H_{in})	Annual Gonadal dose e quivalent (AGDE) (µSv/y)	Alpha index (I_α)	gamma index (I_γ)	Excess lifetime cancer risk (ELCR)x10^{-3}
Almonds	9.41	0.03	38.38	ND	0.082	0.022
Cashew	8.89	0.02	36.25	ND	0.077	0.021
Cocktail	9.37	0.03	36.79	0.007	0.078	0.021
Green pumpkin	8.94	0.02	36.47	ND	0.077	0.021
Linseed	7.79	0.02	31.77	ND	0.067	0.018
Macadamia	5.33	0.01	21.73	ND	0.046	0.012
Moringa	9.78	0.03	38.16	0.009	0.081	0.022
Redskin giant	8.76	0.02	35.73	0.021	0.075	0.020
Sesane seeds	5.91	0.02	24.09	ND	0.051	0.014
Sunflower seeds	7.58	0.02	30.34	0.003	0.064	0.017
Range	5.33-9.78	0.01-0.03	21.73-38.38	ND-0.021	0.046-0.082	0.012-0.022
Mean	8.18	0.02	32.97	0.004	0.07	0.019

Radium equivalent, internal hazard index, annual gonadal dose equivalent, alpha index gamma index and excess lifetime cancer risk.

Calculated values of Ra_{eq} varied from 5.33 Bq/kg in macadamias to 9.78 Bq/kg in moringa. The mean value for Ra_{eq} was 8.18 Bq/kg which is lower than the world average value of 370 Bq/kg (UNSEAR, 2000). The mean value for the internal hazard index was found to be 0.02. This value is lower than unity and thus, the nuts and seeds consumed do not cause any significant radiological effects. Calculated value for the mean for annual gonadal dose equivalent due to the consumption of nuts and seeds was found to be 32.97 µSv/y which was lower than the world average of 300 µSvy^{-1} (UNSCEAR, 2000). When calculated, the values of alpha index varied from 0.003 in sunflower seeds to 0.021 in redskin giants with a mean value of 0.004. The values of gamma index ranged from 0.046 to 0.082 with the mean value of 0.07. The values for all samples were below the critical value for gamma index. The Excess Lifetime Cancer Risk calculated varied from 0.012 to 0.022 in nuts and seeds samples. Its mean was found to be 0.019, a value below the acceptable limit.

Comparison of the activity concentrations in this study with similar studies.

Table 5: Comparison of the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K with those obtained from other countries.

Origin	^{40}K (Bqkg^{-1})	^{226}Ra (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	References
Namibia	101.29	1.99	BDL	Present study
Saudi Arabia	363.82	63.02	50.53	(Al-Ghamdi, 2018)
Nigeria	67.36	37.32	3.88	(Oluwagbenga Inuyomi et al., 2019)
Iraq	308.57	6.71	1.68	(Abojassim & Hashim, 2019)

The results obtained in this study were compared to the results obtained from other studies. The Mean activity concentration for ^{40}K was found to be lower than those of Saudi Arabia and Iraq (Al-Ghamdi, 2018; Abojassim & Hashim, 2019). However, ^{40}K concentration in this study was found to be higher than the value obtained in the study carried out in Nigeria (Oluwagbenga Inuyomi et al., 2019). A comparison of the values for ^{226}Ra showed that the activity concentration of ^{226}Ra was lowest compared to the other studies presented in Table 5 while ^{232}Th was not detected in the present study.

4.0 CONCLUSION

Using gamma-ray spectroscopy, the study estimated the specific activity of radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K in samples of nuts and seeds ingested in Namibia. The mean activity concentrations of ^{40}K and ^{232}Ra were found to be 101.295.55 Bq/kg and 0.80 0.35 Bq/kg, respectively, while for ^{232}Th the mean was found to be below the detection limit. In all the samples, the mean activity concentrations for ^{40}K and ^{226}Ra were below the recommended limit by UNSCEAR 2000. The calculated values for the annual committed effective dose, gamma index, alpha index and excess life cancer risk for ingestion of radionuclides in nuts and seeds samples were lower than internationally acceptable limits. This meant that the results obtained did not show any significant radiological risk to the population of Namibia due to the daily consumption of nuts and seeds. The findings from this study will help provide regulatory bodies with baseline data to be used for developing guidelines for radiation protection due to consumption of nuts and seeds.

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