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Estimation of Naturl Radioactivity Levels in Some Food Spices Commonly Used in Nigeria and Its Radiological Risks

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Authors' contributions

This work was carried out in collaboration between all authors. Author CPO designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript. Authors GOA and CPO managed the analyses of the study. Author SOI managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

A spice is a vegetable substance of indigenous or exotic origin, which has a hot pigment taste that could be used to enhance taste of food or add to food some necessary stimulant ingredients. Knowledge of radioactivity levels in human diet is of particular concern for the estimation of possible radiological hazards to human health. In this study, the radioactivity concentrations of ^{40}K, ^{238}U and ^{232}Th in some selected natural food spices commonly used in Nigeria were determined using gamma spectrometry and the ingested doses via food consumption were also assessed. The average activity concentration of ^{40}K, ^{238}U and ^{232}Th in the samples are 729.85±1.44 $Bqkg^{-1}$, 2.19±1.38 $Bqkg^{-1}$ and 46.73±0.27 $BqKg^{-1}$ respectively. The highest activity concentration of ^{40}K, ^{238}U and ^{232}Th was recorded in Tatashey (2097.17±183) for ^{40}K, Tomatoes (21.90±4.8) for ^{238}U and Red onions (176.38±19) for ^{232}Th. The estimated total annual effective dose received from ^{40}K, ^{238}U and ^{232}Th due to consumption of natural (fresh) spices by the inhabitants of Port Harcourt ranges from 0.85 to 46.76 $\mu Svy^{-1}$and this is far below the average radiation dose of 290 μSvy^{-1} received per caput worldwide due to ingestion of natural radionuclides in food spices. The results indicate

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insignificant radiological health hazard to the public due to the consumption of spices via foods. The excess lifetime cancer risk due to ingestion of radionuclide in fresh food spices were evaluated and the values obtained were compared with standards. Based on these results it can be concluded that the foods spices analyzed in this study do not present any significant radiological concerns.

Keywords: Radioactivity; radiation; spectroscopy; dose; effective dose; spices.

1. INTRODUCTION

Humans are continuously exposed to ionizing radiation of natural sources [1]. Naturally Occurring Radioactive Materials (NORMS) from terrestrial and extraterrestrial origin, and artificial sources used for various applications in medicine, agriculture, industry, research and teaching [2,3]. The extent of exposure if significant could lead to detrimental health effects. Exposure to high levels above background radiation could lead to somatic and genetic effects that tend to damage critical and/or radiosensitive organs of the body, which ultimately can lead to death [1]. Naturally occurring radioactive materials (NORM) can be found in the materials that make up the environment i.e. air, water, soil, food and even in humans. The quantity or amount of naturally-occurring radionuclides changes depending on some factors such as local geology, climate, agricultural practice, [2,1]. Contamination in leafy vegetables, tubers and roots are rampant during the raining season, this can happen as direct fallout or washed down by rain and picked up by roots, tubers and even grains through plant growth process [3,4]. The process by which radionuclides find their way into our body is through a complex mechanism which may also include through food stuff and through food chain from natural radiation sources. A correlation exist between seasons of the year and the extent and magnitude of contamination in the different food and food spices [5].

Spices come from the bark, roots, seeds, and fruit of plants and trees and they are used as recipes to add flavor to dishes and they play an important role in the general makeup of culinary art [6]. Foods and spices can be contaminated either through deposition in the atmosphere or transferred by water through the soil, the introduction of these radionuclide contaminants into the food chain poses a great risk on human health as they get into humans through these food spices primarily by ingestion. Edible plants contains ^{40}K, ^{238}U, ^{232}Th and their progenies [7]. The use of fertilizers especially phosphorus fertilizers for plants can also introduce radioactive isotopes to the plant. When ingested, these radionuclides depending on the type and energy of the emitted radiation and half-life may result in some biological effects. Organs such as kidney, gonad, skin, liver, lungs, circulatory system, etc., can be affected. Chronic effects accompanied by signs such as cancer, tumor, genetic effects, kidney failure and vascular diseases can also be observed [8]. According to [9] one-eighth (1/8) of the annual effective dose (mean) that comes apparently on account of natural sources is caused by foodstuff consumption.

A spice is a dried seed, fruit, root, bark, or vegetative substance primarily used for flavoring, coloring or preserving food and can sometimes be used to hide other flavors. It is sometimes used as a preservative by killing or preventing the growth of harmful bacteria. Spices are distinguished from herbs, which are parts of leafy green plants also used for flavoring or as garnish. Many spices have antimicrobial properties and may explain why spices are more commonly used in warmer climates, which have more infectious disease, and why use of spices is especially prominent in meat, which is particularly susceptible to spoiling [10]. There are two groups of spices consumed by Nigerians and they are the natural or the fresh ones and those that have been processed into powdered or solid forms. Some people prefer the processed ones to the fresh ones. This could be due to storage or convenience. Assessment of contamination of spices consume by the population is very important to know the baseline value, or the level of radiation dose of both natural and artificial radionuclides received by them. Knowledge of the concentrations and distributions of the radionuclides in spices are of interest since it provides useful information in the monitoring of environmental radioactivity. Studies on the natural and artificial radioactivity levels in spices via foods assume importance as it is necessary to estimate the ingestion dose to the public. However, in Nigeria no study has been performed to determine the radionuclides

concentrations in spices and the dose assessment from consumption of these spices by the Nigerian population.

A great deal of research work has been done in different states, countries and continents of the world to analyze food samples for radionuclide concentration and also dose assessment that is due to the consumption of the foodstuff and spices by the public [11,12]. Al-Ghamdi [11] investigated 14 different food spices which were randomly collected from markets located in Jeddah, Saudi Arabia. Gamma spectrometry was used for the analysis using NaI(Tl) detector and found out that total effective dose ranges from 5.63 μSvy^{-1} in Nankhan to 64.92 μSvy^{-1} in Cloves with mean value of 23.6 μSvy^{-1} and cancer risk of $0.55x10^{-4}$. The result showed that food spices are safe radiologically in comparison with international standard. Kansaana et al. [13] also carried out the analysis of activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in 15 food spices commonly consumed in Accra metropolis by Ghana's inhabitants. These food spices included Onions, Ashanti Pepper, Garlic and Green Pepper, Ginger which consists of natural and processed form. The result indicated that the average concentration of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs present in the unprocessed (natural) spices were greater than the concentration in the processed food.

In this study, the radioactivity levels of ^{40}K, ^{238}U and ^{232}Th in fresh natural food spices were estimated using gamma spectrometry. The assessment of the ingested dose in these radionuclides via food consumption was also made. The natural spices considered in the study were Onion, Garlic, Ginger, Ashanti Pepper, Green Pepper, Achi (Brachystegia eurycoma), Calabash nutmeg, Ethiopian/Negro pepper, Offor (Detarium microcarpum), African bush mango, African black pepper, Bell pepper, Nutmeg, black pepper, Tomatoes, melon seed, ground nut oil, palm oil, white onions and the vegetable oil. The study focused on these spices because they are commonly available and used in Nigeria. These spices were purchased from different traders at the mile one market in the Port Harcourt Metropolis of Rivers state, Nigeria. This study, therefore, investigated the extent of the exposure of the general public due to intake of spices as part of the national effort to establish baseline values for the Regulatory Authorities to control the exposure of the public to natural and artificial radionuclides due to the consumption of spices in Nigeria.

2. MATERIALS AND METHODS

2.1 Study Area

Mile one market is located latitudes N4°47′20″.0 to N4°47′55.5″ and longitude E6°54′0.07″ to E6°59′.921″ within Port Harcourt metropolis. Two geologic formations are covered in the study area, namely: Imo shale and Ameki formations respectively. Imo shale consists of a thick sequence of blue and dark grey shales with occasional bands of clay-ironstones and subordinate sandstones [9]. It dips at angles 17° to 25° to the south-west and South [10]. It includes three constituent sandstones: the Igbabu, Ebenebe and Umuna Sandstones with the last two outcropping in the Imo River Basin. The Umuna sandstone is composed of thick sandstone units and minor shales and is generally less than 70m thick. The Ebenebe Sandstone occurs as a lens in the northwestern extremity of the Imo River Basin. It is similar in lithology to the Umuna sandstone but is relatively thicker with a maximum thickness of 130 m [10]. Ameki Formation (Eocene) consists of sand and sandstones. The lithologic units of the Ameki Formation fall into two general groups [14]; an upper grey-green sandstones and sandy clay and a lower unit with fine to coarse sandstones, and intercalations of calcareous shales and thin shelly limestone.

The specific activity concentration of the different radionuclides present in the samples was analyzed using sodium iodide detector. Gamma spectroscopy is ideal for this kind of analysis because it has the ability to measure the radiation emitters directly in their pristine form in the original sample without first going through the rigorous task of chemical separation hence it allows for qualitative and quantitative determination of the various radionuclides in the samples, other materials used includes:

Weighing instrument for taking the mass of the samples, Tray for drying the samples, Masking tape for labeling the samples, Plastic containers for packaging the samples, one litre marginally sample container to store the samples ready for analysis .The samples were broken down into two categories according to their different forms i.e. Solid and liquid samples. Nineteen of the samples are solid and were sliced into the smallest bits possible and spread on the different labeled tray and was sun-dried for several days after which the

samples were pulverized into powder, 500grams measured each and then they were packaged in nylons, labeled and taken to the radiological laboratory for analysis.

2.2 Sampling and Sample Preparation

A total of nineteen natural food spices samples were purchased from different traders at the mile 1 market. They includes; Achi (Brachystegia eurycoma), Tomatoes, Calabash Nutmeg, Garlic, Ginger, Ashanti Pepper, Greenbell Pepper, Ethiopian/Negro pepper, Offor (Detarium microcarpum), African bush mango, African black pepper, Black pepper, Red Onions, Short pepper, Tatashey, White Onions, Chobo, Nutmeg, Egusi, and four liquid natural samples includes Vegetable oil , groundnut oil and Palm oil. These samples were taken to National Institute for Radiation Protection and Research (NIRPR), University of Ibadan, Nigeria. The samples were open air dried on trays for a period of one week and then oven dried at a temperature of 105°C (± 5) to constant weight. The oven dried samples were then grounded into fine powder with a stainless steel ball grinder. The prepared samples, in powdered form, were packed into weighed one liter Marinelli plastic beaker, hermetically sealed, reweighed and stored prior to counting [15,16,17]. The liquid samples were then put into marginally sample container, sealed and kept for four (4) weeks to allow for secular equilibrium between the thorium and the radium content of the sample and their progenies before counting was done. The samples after attaining secular equilibrium was placed on the Sodium Iodide detector and allowed to count for 10 hours.

The containers were sealed to avoid any possibility of out-gassing of radon and kept for a period of 1 month to make sure the samples attained radioactive equilibrium between Ra-226 and its decay products in the uranium series, and Ra-228 and its decay products in the thorium series [18].

2.3 Gamma Spectroscopy

The samples were counted using a gamma-ray spectrometry. The gamma-ray spectrometry system consists 3" × 3" Thallium-activated Sodium Iodide [NaI(Tl)] detector and installed in a 100mm thick lead castle. The detector is connected to an amplifier linked to a computer program GENIE 2 K Window that correlated gamma energies to a number of possible isotopes. The sample was placed the marinelli beaker and then made to sit on the NaI(Tl) detector. Shielding from background (environmental) radiation was achieved by counting in camberra 100 mm thick lead castle. The energy resolution for the detector using Cs-137 from International Atomic Energy Agency (IAEA) is 7.5% at 662KeV Cs-137 line [19]. The energy and efficiency calibration of the system was carried out before sample analysis using the multinuclide reference standard solution supplied by the International Atomic Energy Agency, IAEA. This was to enable identification and quantification of the radionuclides. The standard and the sample were counted for a period of 36,000 seconds to acquire spectral data for a better counting statistics and evaluation. The activity concentration of ^{238}U, ^{232}Th and ^{40}K were determined after correction for background and inhomogeneity [20,21].

The specific activity concentration of ^{238}U, ^{232}Th and ^{40}K in the medicinal plants were determined from the quantitative analysis of the spectra acquired from the Gamma-ray spectrometry using the Gamma-ray spectrum analysis software, Ortec MAESTRO–32 at specific energies. ^{238}U was calculated from the average of ^{214}Pb at energies of 251.9 Kev and 295.2 Kev and ^{214}Bi at energies of 609.3 Kev and 1764.5 Kev. ^{232}Th was determined from the average of ^{208}Tl at energies of 2614.5 Kev and 583.2 Kev, ^{212}Pb at the energy of 238.6 Kev and ^{228}Ac at the energy of 911.2 Kev and ^{40}K at 1460.0 Kev. The specific activity ($A_{sp(E,i)}$ in Bq kg^{-1}) of the radionuclide *i* in the samples were calculated after decay correction using the expression in equation [22].

$$A_{sp}(E,i) = \frac{N_{sam}\,(E,i)}{\varepsilon\gamma(E)\,Tc\,P\gamma(E,i)\,M_{sam}} \quad (1)$$

where; N_{sam} $(E_{,i})$ is the net counts for the radionuclide *i* at energy *E*, $\varepsilon_{\gamma(E)}$ is the photo peak efficiency at energy *E*, T_c is the counting live-time (s), Pγ(E, i) is the gamma emission probability of the radionuclide *i* for a transition at energy *E*, M_{sam} is the dry-weight of samples (kg).

3. RESULTS AND DISCUSSION

3.1 Specific Activity Concentrations in All the Food Spices Samples

The activity concentration of ^{238}U, ^{232}Th and ^{40}K in samples of food spices commonly consumed by the Nigerian population are presented in

Table 1. The activity concentration for ^{40}K in the spices samples ranged from 13.86 Bqkg^{-1} in Calabash nutmeg to 2097.17±183.38 Bqkg^{-1} in Bell pepper and the mean value of ^{40}K in the samples was 839.33±76.17 Bq/kg. The activity concentration of ^{238}U in the food spice samples ranged from 3.85±0.91 Bqkg^{-1} in Calabash nutmeg to 21.90±4.57 Bqkg^{-1} in tomatoes and the mean value of ^{238}U in the samples was 8.4±1.65 Bqkg^{-1}. The activity concentration of ^{232}Th ranged from 0.2±0.03 Bqkg^{-1} in African black pepper to 176.38±19.77 Bq/kg in Red onion with a mean value of 48.86±5.87 Bqkg^{-1}. ^{40}K was detected in all the food spices with reasonable activity levels except in Achi, ground nut oil and palm oil. The concentration of potassium was found to be very high compared to Uranium and Thorium due to the high concentration of ^{40}K in the soil and high transfer ratio as seen in Fig. 1.

Potassium is a micronutrient and it may be expected that soil characteristics favor the mobilization of potassium and its subsequent migration into the plant [23,11]. However, ^{40}K is an essential biological element and its concentration in human tissue is under close metabolic control. Fig. 1 represent the activity concentration of ^{238}U, ^{232}Th and ^{40}K in various food spices.

3.2 Estimation of Annual Effective Dose and Dose Rate from Ingested Food Spices

The effective dose due to consumption of food spices comes in handy for the possibility of summing up different radionuclides which of course may come from different radioactive sources. These ingested radiation doses can be quantified by measuring the activity concentration (Bqkg^{-1}) of the radionuclide in the food spices and then multiplied by the masses of these food spices consumed over a given time frame (kg/day or kg/year) and a dose conversion factor (Sv/Bq) given for each radionuclide and can be appropriately applied [13].

$$E = \Sigma\,(As\ x\ Is\ x\ DCF) \qquad (2)$$

Where, E = Annual Effective Dose equivalent by ingestion of the radionuclides (Sv/y) A_s = Activity concentration of radionuclides in the sample (Bq/kg) I_s = Annual intake of the food spices (kg/y), DCF = internal dose conversion factor by ingestion of the radionuclides (Sv/Bq). This is given as 0.28 µSv/Bq for ^{238}U, 0.23 µSv/Bq for ^{232}Th and 0.0062 µSv/Bq [24]. In this study the consumption values for locally produced food spices corresponds to those of the adult citizen. The scale of the annual intake (1 kg/y) was used. The risk associated with an intake of radionuclides in the body is proportional to the total internal dose delivered by the radionuclides.

Table 1, presents the annual effective dose equivalent of ^{238}U, ^{232}Th and ^{40}K radionuclides and the total dose due to the three radionuclides in Nigerian food spices samples estimated and compared with the reported global dose due to ingestion of naturally occurring radionuclide. From Table 1, the annual effective ingestion doses due to intake of ^{238}U varied from 0.00 µSvy^{-1} in calabash nutmeg, Garlic, ginger, long pepper, Ethiopian pepper, Offor (Detarium microcarpum), black pepper, melon seed, nutmeg and oil to 6.13 µSvy^{-1} in Tomatoes. Most samples show insignificant low ingestion doses over the values of 6.3 µSvy^{-1}` reported by UNSCEAR, [14]. The dose received from ^{232}Th due to consumption of food spices varied from 0.00 in African black pepper to 40.57 µSvy^{-1} in red onions which is 10676.3 % of the total ingestion dose of 0.38 µSvy^{-1} as reported by UNSCEAR, [14].

The values of effective dose from ingestion of ^{40}K ranged from 0.00 in Achi (Brachystegia eurycima), Ground nut oil and palm oil to 13.00 µSvy^{-1} in Tatashey. Thus the contribution to dose from the ingestion of ^{40}K in food spices with its relatively low dose conversion factor will be much less than that for the thorium (^{232}Th) but higher than that for Uranium (^{238}U). The mean annual effective dose from ^{238}U, ^{232}Th and ^{40}K in food spices were estimated to be 0.61, 10.75 and 4.52 µSvy^{-1} respectively. The highest mean annual internal dose was ^{232}Th and all their mean doses are less than annual dose limit of 1 mSvy^{-1} for the general public. The UNSCEAR, [14] report shows that the sub-total ingestion dose of uranium and thorium series is given as 120 µSvy^{-1} which is higher than the sub-total dose for the results reported in this study for uranium and thorium. This difference comes from ^{210}Po, ^{210}Pb and ^{228}Ra, which were not detected in our measurement.

The total effective dose ranged from 0.85 µSvy^{-1} (vegetable oil) to 41.38 µSvy^{-1} tomatoes) with average value of 15.89 µSvy^{-1}. These values are much less than the world total food dose value of 290 µSvy-1 for all foods [14]. The low values of effective dose due to intake of spices is due to

low annual intake of only 1 kgy^{-1} when compared to a few hundred kgy^{-1} for the total food intake. The relative contribution to the total dose due to ^{238}U, ^{232}Th and ^{40}K was 4%, 68% and 28% respectively which is presented in Fig. 2.

Table 1. Activity concentration of ^{40}K, ^{232}Th and ^{238}U in food spices in Nigerian market and their radiological risk parameters

S/N	Samples	^{40}K ($Bqkg^{-1}$)	^{238}U ($Bqkg^{-1}$)	^{232}Th ($Bqkg^{-1}$)	Annual effective dose equivalent (AEDE) (μSvy^{-1})			Total AEDE (μSvy^{-1})	ELCR × 10^{-4}
					^{40}K	^{238}U	^{232}Th		
1	Achi (Brachystegia eurycoma)	BDL	3.85±0.91	100.67±11	0	1.08	23.15	24.23	0.61
2	Tomatoes	1763.19±163	21.90±4.8	105.71±13	10.93	6.13	24.31	41.38	1.03
3	Calabash nutmeg	13.86±1.44	BDL	127.42±15	0.09	0	29.31	29.39	0.73
4	Garlic	714.71±63	BDL	12.79±1.6	4.43	0	2.94	7.37	0.18
5	Ginger	1254.97±110	BDL	31.29±3.9	7.78	0	7.20	14.98	0.37
6	Green bell Pepper	349.28±32.4	18.46±2.9	0.2±0.03	2.17	5.17	0.05	7.38	0.18
7	Long Pepper	1288.03±113	BDL	34.9±4.3	7.99	0	8.03	16.01	0.40
8	Ethiopian pepper	688.34±62.9	BDL	39.652±4.9	4.27	0	9.12	13.39	0.33
9	Offor(Detarium microcarpum)	285.93±28.7	BDL	47.32±5.8	1.77	0	10.88	12.66	0.32
10	African bush mango	454.51±45.1	BDL	3.62±0.47	2.82	0	0.83	3.65	0.09
11	Oil 1 (Vegetable)	76.873±7.7	BDL	1.61±0.21	0.48	0	0.37	0.85	0.02
12	Oil 2 (Vegetable)	120.53±11.96	BDL	24.56±3.10	0.75	0	5.65	6.40	0.16
13	Oil 3 (Groundnut)	BDL	BDL	14.7±1.89	0	0	3.38	3.38	0.08
14	African black pepper	1337.32±116	BDL	BDL	8.29	0	0	8.29	0.21
15	Palm Oil	BDL	1.81±0.41	17.11±2.16	0	0.51	3.94	4.44	0.11
16	African black Pepper	1818.31±153	BDL	42.83±5.23	11.27	0	9.85	21.12	0.53
17	Red Onion	947.08±86.5	1.13	176.38±19	5.87	0.32	40.57	46.76	1.17
18	Short Pepper	891.86±84.7	3.25	18.52±2.4	5.53	0.91	4.26	10.70	0.27
19	Bell pepper	2097.17±183	BDL	26.46±3.3	13.00	0	6.09	19.09	0.48
20	White Onion	553.34±53.3	BDL	82.84±9.96	3.43	0	19.05	22.48	0.56
21	Black pepper	1544.89±145.6	BDL	48.9±6.22	9.58	0	11.25	20.83	0.52
22	Melon seed	412.61±41.4	BDL	25.89±3.22	2.56	0	5.95	8.51	0.21
23	Nutmeg	173.779±18.2	BDL	91.47±11.0	1.08	0	21.04	22.11	0.55
	Mean	**729.85**	**2.19**	**46.73**	**4.52**	**0.61**	**10.75**	**15.89**	**0.32**

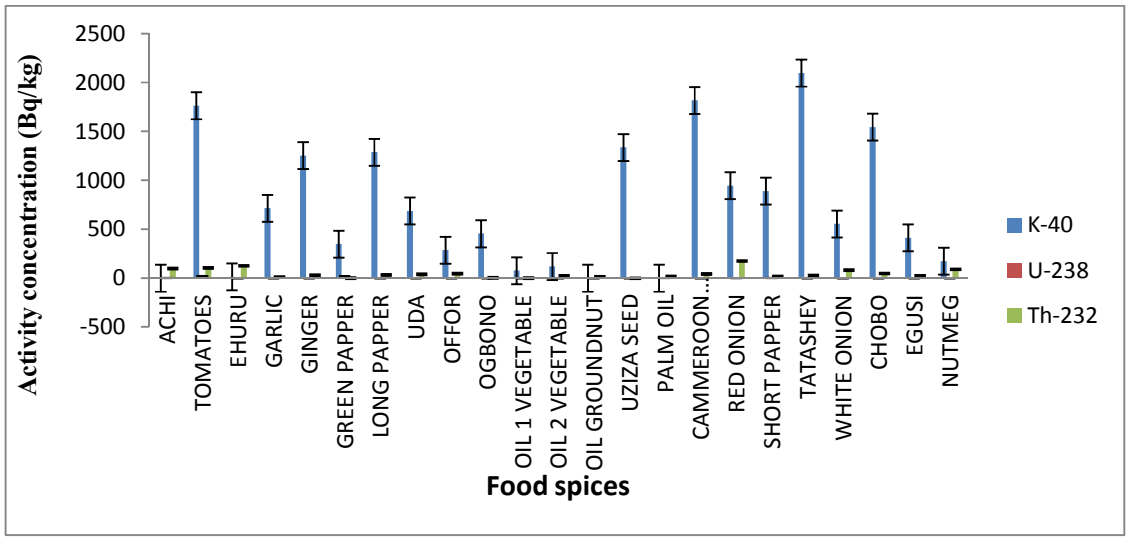

Fig. 1. Activity concentration of ^{238}U, ^{232}Th and ^{40}K in Nigerian food spices

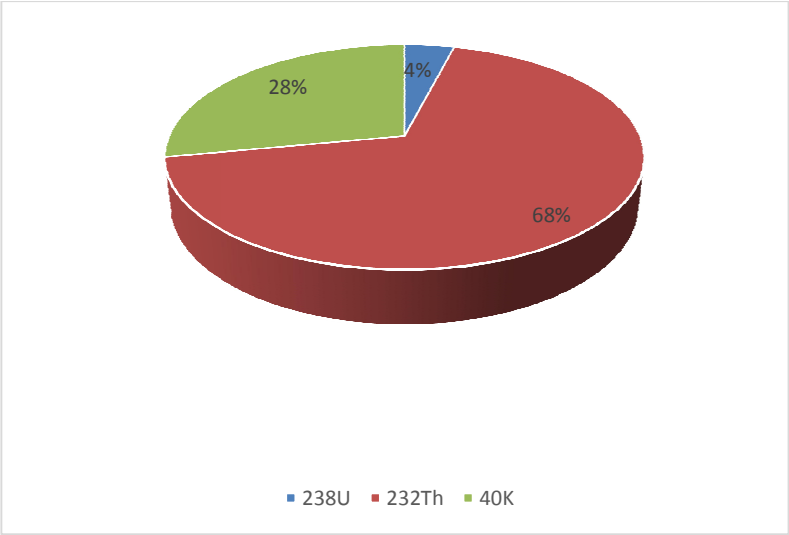

Fig. 2. % contribution of ^{238}U, ^{232}Th and ^{40}K to the total effective dose from food spices

3.3 Excess Lifetime Cancer Risk (ELCR)

The risk incurred by a population is estimated by assuming a linear dose-effect relationship with no Threshold as per ICRP practice. For low doses ICRP fatal cancer risk factor is 0.05 Sv^{-1} [26]. The risk factor states the probability of a person dying of cancer increases by 5% for a total dose of 1 Sv received during his lifetime. Therefore, the probability of death from cancer due to 'natural incidence' increases from about 25% to 30% following a total lifetime exposure of 1 Sievert. To estimate cancer risk for an adult person using the following relationship [25,26]:

$$ELCR = Cd \times RF\ (Sv^{-1}) \quad (3)$$

where: - RF is risk factor (Sv^{-1}), fatal cancer risk per Sever. For stochastic effects, ICRP 60 uses values of 0.05 for the public [26]. Cd is the life time effective dose which is a measure of the total effective dose received over an average lifetime of 50y following ingestion of a radionuclide was calculated using [14]:

$$Cd = 50 \times D \quad (4)$$

where D is the total effective dose to an individual.

Table 1, shows the calculated cancer risk due to ingestion of food spices which ranged from 0.02 $\times 10^{-4}$ (Vegetable oil) to 1.16 $\times 10^{-4}$ (red Onions) with an average value of 0.32 $\times 10^{-4}$. The average value is comparable with other kinds of health risks which gives a risk factor of 0.48 $\times 10^{-4}$ due to food spices [27,28]. The mean value of ELCR is lower than the world average value of 2.9 $\times 10^{-4}$ based on annual dose limit of 1 mSv for general public [14].

4. CONCLUSION

Estimation of natural radioactivity in some food spices commonly used in Nigeria and its radiological risk has been done using gamma spectroscopy. The activity concentration of ^{238}U, ^{232}Th and ^{40}K found in all the sampled food spices are within their permissible values. The total annual effective dose received by members of the public from food spices were also lower than the safe value of 290 μSvy^{-1} The estimated excess lifetime cancer risk are slightly higher than the world safe value of 0.29 $\times 10^{-3}$ though it has no immediate significant health hazard. This study therefore reveals that all the natural food spices sampled are radiologically safe for consumption. This result serves as a baseline radiological data on food spices for future studies.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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