An Assessment of Radiological Hazard Levels in Vegetables and Condiments Obtained from Ile-Ife Main Market, Ile-Ife, Nigeria

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Abstract- This study evaluated the radiological hazard levels of the naturally occurring radionuclides in samples of commonly consumed vegetable and condiment samples collected from Ile-Ife main market. Gamma Ray Spectrometer was used to profile ^{238}U, ^{232}Th, and ^{40}K levels in the samples. Activity Concentrations, Radium equivalent activity concentration index (Ra_{eq}), Absorbed Gamma Dose Rate (D_R), External Hazard Index (H_{ex}), Internal Hazard Index (H_{in}), Excess Lifetime Cancer Risk (ELCR) and Annual Effective Dose Equivalent (AEDE) were subsequently evaluated. In the vegetables, the range for the activity concentrations were 78.93 to 118.18 Bq/kg for ^{40}K, 44.04 to 62.87 Bq/kg for ^{238}U, while for ^{232}Th, the range was 4.45 to 6.92 Bq/kg. In condiment samples, ^{40}K ranged between 59.68 and 72.83 Bq/kg, ^{238}U had a range of 33.33 to 40.91 Bq/kg, while ^{232}Th was found within the range 3.42 to 4.26 Bq/kg. Although the ELCR calculated indicated higher values of 100 to 200 folds compared to acceptable value of 2.9 x 10^{-3} stipulated by UNSCEAR the values of D_R, Ra_{eq}, H_{ex} and H_{in} were significantly lower ($P \leq 0.05$) than or matched well with the permissible values indicating consumption safety for those who do not rely heavily on the consumption of the items investigated.

Index Terms- Radiological hazards, Vegetables, Condiments, Activity concentrations, Excess Life Time Cancer Risk, Ile-Ife

I. Introduction

A condiment is a dried seed, fruit, root, bark, or vegetative substance primarily used for flavouring, colouring or preserving food (by killing or preventing the growth of harmful bacteria) and can sometimes be used to hide other flavours (Ononugbo *et al*., 2016), while vegetables are a large group of plant parts (leaves, fruits, roots, and so on) usually consumed as food. Condiments are excellent sources of antioxidants, whereas several important minerals, dietary fibres and vitamins are often derived from vegetables (Anas and Yusuf, 2017).

Natural background radiation can originate from terrestrial and extraterrestrial sources. Sources of terrestrial radiation are radioactive nuclides from environmental materials including soils, building materials, water, rocks and atmosphere, while outer space as primary cosmic rays is responsible for extraterrestrial radiation (Mason and Moore, 1982; Faure, 1986; Me'nager *et al*. 1993; Rudnick and Gao, 2003).

Apparently, radionuclides detected in plants are originally contained in soils from where they get translocated via the root system to different plant parts or transported either by direct fallout of radionuclides and resuspension of contaminated soils followed by deposition on plant leaves or soils within the vicinity of the plant (Noordijk *et al*., 1992). Sometimes, man-made radionuclides contaminate the food chains as a result of fallout from nuclear weapons tests in the atmosphere or from routine and accidental releases of nuclear wastes (IAEA, 1989). As the spatial distribution of radionuclides varies with respect to the parent soils from region to region (Keser, *et al*., 2013), so also their uptake by plants varies from place to place even among similar cultivars (Pendo and Leonid, 2017). Thus, an assessment of radionuclides concentration and distribution in frequently consumed food items is of particular interest in order to have clear and reliable pieces of information about the potential hazard of radiation exposure that could result from incorporating such items into our dietary requirements (Pendo and Leonid, 2017).

In the past, people suffered avoidable health hazards because of little or improper knowledge on the effect of ionizing radiation from various sources in the environment. Knowledge about the need to protect humans from excessive radioactive sources can positively and apparently enhance their safety and better well being (Banzi *et* al., 2000).

According to Jibiri *et al*. (2007), to make sure that all people have access to sufficient, nutritionally adequate and safe food is one of the three cardinal goals of the United Nations for sustainable food security. Thus, in the present work, levels of ^{238}U, ^{232}Th, and ^{40}K in samples of garden egg leaf, water leaf, bitter leaf, pumpkin leaf, amaranth, cayenne pepper, Scotch bonnet, curry, nutmeg and thyme collected from Ile-Ife main market were determined. This was done with a view to establishing the radiological hazards that could be associated with consistent incorporation of the sampled materials as part of human diets.

II. MATERIALS AND METHODS

Sample Collection and Sample Pre-treatment

Five condiment samples (cayenne pepper, Scotch bonnet, curry, nutmeg and thyme) and five types of fresh vegetables (Garden egg leaf, Water leaf, Bitter leaf, Pumpkin leaf and Amaranth) used in this study were obtained from Ile-Ife central market, Osun state, Nigeria. Each vegetable sample was properly rinsed with distilled water to remove radionuclide bearing particulates that might have been deposited on their outer surfaces. The vegetables were further sliced in a way similar to how they are normally prepared for human consumption, placed in properly labelled crucible dishes and dried to a constant weight in the laboratory using the Gallenkamp oven at a temperature of 60^0C. Similarly, each of the condiment samples was dried to a constant weight to ensure that all the samples were similarly pretreated.

Sample Preparation for Gamma Ray Spectroscopy Analysis

To ensure homogeneity, the dried samples were crushed and pulverized to fine powder using agate mortar and pestle. Each of the pulverized samples was weighed by the use of an electronic balance and placed in a labelled container and reweighed. The samples were kept in air tight plastic containers which had been washed thoroughly with soap and rinsed with distilled water and incubated for a period of 28 days in order to attain secular equilibrium between parents and progenies radionuclides. Gamma ray analysis was carried out by counting each sample for 10 hours using a Caesium Iodide (CsI) scintillation detector. The gamma ray spectrometric analysis was carried out using a 1 inch by 1 inch Cesium Iodide (CsI) detector enclosed in a thick lead shield at the Biological Trace Element Research Laboratory, Department of Physics, Obafemi Awolowo University, Ile-Ife, Nigeria.

Activity Calculation

The activity (A) in Bq/kg of the radionuclides in the samples was calculated after decay correction using the expression:

$$C\ (Bq/kg) = \frac{C_n}{\in_{P_\gamma M_s}} \quad [1]$$

where C is the activity concentration of the radionuclide in the sample given in Bq kg^{-1}, C_n is the count rate under the corresponding peak, $\in$ is the detector efficiency at the specific γ-ray energy, $P\gamma$ is the absolute transition probability of the specific c-ray, and M_s is the mass (kg) of the sample.

Determination of Radiological Hazard Index

***Radium equivalent activity concentration index* (Ra_{eq})**

Radium equivalent (Ra_{eq}) index in Bq/kg is a widely used radiological hazard index tool. It is a convinient index to compare the specific activities of samples containning different concentrations of ^{238}U,^{232}Th and ^{40}K (Beretka and Matthew,1995; Ravinsankar *et al.,* 2014). For the present study, Ra_{eq} was calculated using the formula:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \quad [2]$$

where A_U, A_{Th} and A_K are the specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/kg respectively.

The Absorbed Gamma Dose Rate

The input of natural radionuclides to the absorbed dose rate in air (D_R) at average height of one meter above the surface of ground depends on the natural specific activity concentration of ^{238}U, ^{232}Th and ^{40}K. This was estimated using the formular given by Kurnaz *et al.* (2007) and Ravinsankar *et al*. (2014):

$$D_R\ (nGyh^{-1}) = 0.43A_U + 0.666A_{Th} + 0.042A_K \quad [3]$$

where A_{Ra}, A_{Th} and A_K are the specific activity concentrations (in Bq kg^{-1}) of Ra, Th and K, respectively.

Annual Effective Dose Rate or Equivalent (AEDR or AEDE)

Annual estimated average effective dose equivalent is calculated using a conversion factor of 0.7 $SvGy^{-1}$, which is used to convert the absorbed rate to human effective dose equivalent. The annual effective dose rate was determined using the relationship:

$$AEDR = D \times T \times F \quad [4]$$

where D is the calculated dose rate (in $nGyh^{-1}$), T is the outdoor occupancy time (0.2 x 24 h x 365.25 d ≈ 1753 hy^{-1}), F is the conversion factor (0.7 x 10^{-6} $SvGy^{-1}$).

Excess Lifetime Cancer Risk (ELCR)

Excess lifetime cancer risk (ELCR) is calculated using the following equation.

$$ELCR = AEDE \times DL \times RF \quad [5]$$

where AEDE is the Annual Equivalent Dose Equivalent, DL is the average duration of life (estimated to 54 years), and RF is the Risk Factor (Sv^{-1}), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for public (Taskin *et a*l., 2009).

External Hazard Index (H_{ex})

Radiation exposure due to ^{226}Ra, ^{232}Th and ^{40}K may be external. This hazard, defined in terms of external hazard index or outdoor radiation hazard index and denoted by H_{ex}, can be calculated using the equation:

$$H_{ex} = C_{Ra/}370 + C_{Th}/259 + C_K/4810 \quad [6]$$

(Beretka and Mathew, 1985)

where C_{Ra}, C_{Th} and C_K are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bq/kg. The value of this index should be less than 1 $mSvy^{-1}$ in order for the radiation hazard to be considered acceptable to the public (Beretka and Mathew, 1985).

Internal Hazard Index (H_{in})

The internal hazard index (H_{in}) is a measure of the internal exposure to carcinogenic radon and according to Beretka and Mathew (1985) can be evaluated using the formula:

$$H_{in} = C_{Ru}/185 + C_{Th}/259 + C_K/4810 \quad [7]$$

where C_{Ra}, C_{Th} and C_K are activity concentrations (in Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

The value of this index should be less than 1 $mSvy^{-1}$ in order for the radiation hazard to have negligible hazardous effects to the respiratory and other internal organs of the public (Beretka and Mathew, 1985).

III. RESULTS AND DISCUSSION

Activity Concentrations

Tables 1 and 2 summarize the activity concentrations (in Bq/kg) of the radionuclides ^{40}K, ^{238}U and ^{232}Th in the vegetable and condiment samples, respectively. The activity concentrations in vegetable samples ranged from 78.93 ± 0.91 to 118.18 ± 6.20 for ^{40}K, 44.04 ± 0.44 to 62.87 ± 4.32 for ^{238}U, and 4.45 ± 0.39 to 6.92 ± 1.52 for ^{232}Th. The overall mean activity levels for ^{40}K, ^{238}U and ^{232}Th in vegetable samples were 92.97 ± 15.04, 51.93 ± 7.37 and 5.35 ± 0.95, respectively. These value were higher than the 0.6-2.6, 24, 35 recommended as the permissible levels by the Jordan, USA, and UNSCEAR (2000) respectively, but lower than the 61-72 acceptable value of India (Pendo and Leonid, 2017). Total activity concentrations ($^{40}K + ^{238}U + ^{232}Th$) in the vegetables indicated the order: Water leaf (127.42) < Bitter leaf (140.24) < Garden egg leaf (141.67) < Amaranth (153.95) < Pumpkin leaf (243.97). In all the cases, water leaf had the least activity concentrations, while pumpkin leaf had the highest. This observation probably suggests that apart from the geological formations and compositions of the soil on which a given plant grows, plant species might have some notable roles to play in the amount of radionuclides that could get translocated into the plant system. Thus, it is considered a paramount health safety measure to consume vegetables with more activity concentrations less regularly than those with less activity concentrations.

Similarly, the radionuclide activity concentrations due to ^{40}K, ^{238}U and ^{232}Th in the condiments respectively ranged from 59.68 ± 4.48 to 72.83 ± 2.63, 33.33 ± 3.30 to 40.91 ± 1.82 and 3.42 ± 1.09 to 4.26 ± 0.58 with Curry and Scotch bonnet having the lowest and highest values respectively in all the cases. Their respective mean values were 66.54 ± 6.44, 37.13 ± 3.26 and 3.85 ± 0.35. These values were lower than either the 400 or 698 – 1439 Bg/kg maximum acceptable levels stipulated by UNSCEAR (2000) or Jordan, respectively. Apart from the 61-72 India allowable value, the levels of ^{238}U in the condiments, with a mean value of 37.13 ± 3.26, were significantly higher than the 0.6-2.6 Bq/kg (Jordan), 24 Bq/kg (USA) or 35 Bq/kg (UNSCEAR, 2000) values. For ^{232}Th, the values were significantly higher ($p \leq 0.05$) than the Jordan permissible levels (0.7 – 3.4 Bq/kg), but much lower than the values 18 and 30 Bq/kg recommended by UNSCEAR (2000). Total activity concentration values indicated that curry had the least radionuclide concentrations (96.43 Bq/kg), while Scotch bonnet had the highest value (118.00 Bq/kg). The implication is that using Curry as food seasoning agent ensures less exposure to radionuclides than other condiments investigated in this study.

These results were higher than those (400, 35 and 30 for ^{40}K, ^{238}U and ^{232}Th respectively) obtained by Pendo and Leonid (2017). Compared with the UNSCEAR (2000) permissible value of 35 Bq/kg, the activities of ^{238}U in the vegetable and condiment samples were found to be higher than the world average value, while the activities of ^{232}Th and ^{40}K were found to be lower than their respective world permissible values of 30 and 400 Bq/kg.

According to Qabas *et al*. (2014), high values of radionuclides in plant samples could be due to geographical and geological factors. Thus, the same reason could be partly responsible for the activities of ^{238}U detected in the present study. However, the uptake of radionuclides by vegetables and condiments from the soil into plants is a complex combination of interwoven factors including plant species, soil conditions, the concentration of radionuclides in soil, bioavailability of the radionuclides in soil and the types and quantity of agrochemicals, such as phosphate fertilizers, used (Ferdous *et al*., 2013).

The calculated radium equivalent (Ra_{eq}), absorbed dose rate (D_R), annual effective dose equivalent (AEDE), effective life cancer risk (ELCR), external hazard (H_{ex}) and internal hazard (H_{in}) indices are presented in Table 3. The Ra_{eq} for vegetable and condiment samples varied from 56.48 - 81.87 $Bqkg^{-1}$ (mean = 66.74 Bq kg^{-1}), and 42.82 - 50.91 Bq kg^{-1} (average = 47.77 Bq kg^{-1}), respectively. These values were far below the allowable limit (370 Bq kg^{-1}) recommended by the International Atomic Energy Agency (IAEA, 1989). The absorbed dose rates in the present study indicated that the dose rate due to ^{238}U in the vegetable samples varied from 25.22 to 36.61 $nGyh^{-1}$ (mean = 29.80 $nGyh^{-1}$) and in the condiment samples, the absorbed dose rate ranged between 19.12 and 23.49 with an average value 21.33 Bg/kg, these values when compared to the world average value of 55 $nGyh^{-1}$ (IAEA, 1989) were lower.

The value of annual effective dose equivalent (AEDE) was observed to fall within the permissible limit of 0.48. The possible human carcinogenic effect was carried out by estimating the likelihood of cancer occurrence in a population of individuals for a specific lifetime from projected intakes (and exposures). The calculated values of the excess lifetime cancer risk (ELCR) in the vegetable samples ranged between 0.43×10^{-3} and 0.62×10^{-3} with an average value 0.52×10^{-3} and in the condiment samples, the effective life cancer risk varied from 0.32×10^{-3} to 0.40×10^{-3} with a mean value of 0.36×10^{-3}. When compared with the recommended safe limit of 2.9×10^{-3} by the United Nations Scientific Committee on the Effect of Atomic Radiation (2000), it could be inferred that the ELCR values would not lead to respiratory diseases, such as asthma, cancer and external diseases, such as erythema, skin cancer and cataracts.

In the case of vegetable samples, the H_{ex} values obtained (Table 3) were 0.19 to 0.27 with an average value 0.27, while the values were 0.14 to 0.17 with a mean value 0.16 for condiments. The calculated H_{ex} values for all samples were below unity, which implied that their consumption would not pose radioactive related harm to the populations of the consumers. The H_{in} values for vegetable samples (Table 3) were 0.34 to 0.49 with an average value 0.40, and for the condiment samples, the values were 0.26 to 0.32 having a mean value 0.29. The calculated H_{in} values for all samples were significantly below unity at $P \leq 0.05$ confidence level.

IV. CONCLUSION

The radionuclide activity concentrations of ^{40}K, ^{238}U, ^{232}Th in both the vegetable and condiment samples frequently consumed in Ile-Ife and its environs were determined in this study. Except for the activity concentrations of ^{238}U, which were higher than the permissible levels probably as a result of the geographical and geological factors, the values obtained for ^{40}K and ^{232}Th were found to be lower than the permissible values. The activity concentrations, gamma absorbed dose rates (D_R), radium equivalent activity (Ra_{eq}), annual effective dose equivalent, effective life cancer risk and hazard indices (H_{ex} and H_{in})

indicated good level of consumption safety when compared with the world permissible values.

Table 1: Activity Concentration of Radionuclides in Vegetables (Bq/kg) Samples

Sample	^{40}K	^{238}U	^{232}Th	Total activity concentrations
Garden egg leaf	85.97±2.49	50.27±2.46	5.43±0.91	141.67±5.86
Water leaf	78.93±0.91	44.04±0.44	4.45±0.39	127.42±1.74
Bitter leaf	88.20±2.91	47.25±1.74	4.79±0.40	140.24±5.05
Pumpkin leaf	118.18±6.20	62.87±4.32	6.92±1.52	243.97±12.04
Amaranth	93.55±3.71	55.22±2.83	5.18±0.76	153.95±7.3
Mean ± s.d.	92.97±15.04	51.93±7.37	5.35±0.95	161.45 ± 23.36
Range	78.93-118.18	44.04 – 62.87	4.45 - 6.92	127.42 – 243.97
India	-	61-72	-	
Jordan	698-1439	0.6-2.6	0.7-3.4	
USA	-	24	18	
UNSCEAR (2000) *	400	35	30	

*UNSCEAR (2000) Acceptable Value

Table 2: Activity Concentration of Radionuclides in Condiment Samples (Bq/kg)

Sample	^{40}K	^{238}U	^{232}Th	Total activity concentrations
Cayenne pepper	72.30 ± 2.73	39.44 ± 2.19	4.13 ± 0.68	111.74
Scotch Bonnet	72.83 ± 2.63	40.91 ± 1.82	4.26 ± 0.58	118.00
Curry	59.68 ± 4.48	33.33 ± 3.30	3.42 ± 1.09	96.43
Nut meg	67.98 ± 3.43	37.72 ± 2.55	3.86 ± 0.86	109.56
Thyme	59.89 ± 4.46	34.27 ± 3.16	3.60 ± 0.99	97.76
Mean ± s.d.	66.54 ± 6.44	37.13 ± 3.26	3.85 ± 0.35	106 ± 9.31
Range	59.68-72.83	33.33 - 40.91	3.42 - 4.26	96.43 – 118.00
India	-	61-72	-	
Jordan	698-1439	0.6-2.6	0.7-3.4	
USA	-	24	18	
UNSCEAR, 2000*	400	35	30	

(Pendo and Leonid, 2017) *UNSCEAR (2000) Acceptable Value

Table 3: Radiological Hazard Values for Vegetables and Condiments

Sample	Ra_{eq}	D_R ($nGyh^{-1}$)	AEDE	ELCR	H_{ex}	H_{in}
Garden egg leaf	64.65	28.84	0.18	0.49	0.21	0.39
Water leaf	56.48	25.22	0.15	0.43	0.19	0.34
Bitter leaf	60.89	27.21	0.17	0.46	0.20	0.37
Pumpkin leaf	81.87	36.61	0.22	0.62	0.27	0.49
Amaranth	69.83	31.12	0.19	0.53	0.23	0.42
Mean ± s.d.	66.74±9.78	29.80±4.38	0.18±0.03	0.52±0.07	0.27±0.03	0.40±0.06
Cayenne pepper	50.91	22.75	0.14	0.38	0.17	0.31
Scotch Bonnet	52.61	23.49	0.14	0.40	0.17	0.32
Curry	42.82	19.12	0.12	0.32	0.14	0.26
Nut meg	48.47	21.65	0.13	0.37	0.16	0.29
Thyme	44.03	19.65	0.12	0.33	0.15	0.26
Mean ± s.d.	47.77±4.25	21.33±1.90	0.13±0.01	0.36±0.03	0.16±0.01	0.29±0.03
UNSCEAR (2000)*	370	55 $nGyh^{-1}$	0.48	2.9×10^{-3}	≤ 1	≤ 1

*UNSCEAR (2000) Acceptable Value

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COVERING LETTER

Dear Editor,

This paper titled "An Assessment of Radiological Hazard Levels in Vegetables and Condiments Obtained from Ile-Ife Main Market, Ile-Ife, Nigeria" on behalf of all the authors declare this

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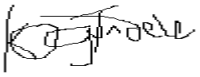

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