EVALUATION OF NATURAL RADIONUCLIDES IN SOME FOODS CONSUMED IN NORTHERN PART OF DELTA STATE NIGERIA

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Abstract: Food ingestion has played a vital role to human by sustaining lives and radionuclide contamination is very possible. The objectives of this study was to evaluate the natural radionuclides like (^{238}U, ^{232}TH, ^{40}K) in some foods consumed in northern part of Delta State Nigeria and its radiological risks, the analysis result was determined using NaI(TI) gamma spectrometry detector and the highest activity concentration for ^{238}U, ^{232}Th, ^{40}K were 1095.95 ± 210.96 Bqkg^{-1} , 42.36 ± 15.28 Bqkg^{-1}, 67.26 ± 12.28 Bqkg^{-1} respectively. All values are from the food samples based on the balance between input and output rates of radionuclides in the food samples with the mean values for 709.42 ± 131.46 Bq/kg for ^{40}K , 28.59 ± 2.82 Bq/kg for ^{238}U, 31.49 ± 5.08 Bq/kg for ^{232}TH , the annual effective dose of all natural radionuclides for the consumers of the food stuffs were determine as follows (0.05, 0.12 , 0.08)mSvy^{-1}, The radiological hazard risk indices is below the world average permissible and recommended safe limit of <1,<1 and 0.29×10^{-3} respectively**.** Therefore, it is an indication that the study area is safe for human activity.

Keywords: Absorbed dose rate, Excess life cancer risk, Gamma spectrometer detector, Hazard risk, Natural radionuclide.

Introduction

Mokobia (2022) has indicated that some of the foods available to humans contains natural occurring radioactive materials (NORM) such as ^{40}K, ^{210}Po, ^{238}U and ^{232}Th and these foods therefore, possess varying degrees of radioactive materials. The radiation dose to humans consuming the foods depends on the location and geology. Thus in places having high uranium and thorium soil contents, one would expect greater dosage (Daley, 1995). The pathway showing the three stages through which radiation gets to humans in shown in (Figure 1)

The awareness and concern of the radiological contamination of food have motivated a number of investigations in different climes. The 'National Centre for Scientific Research (NCRS) (NCRS, 2020) after some investigations has provided data on the radioactivity levels as determined for different foodstuffs (Figure 2). In Nigeria for instance, Adedokun et al (2019), Arogunjo *et al* (2005) and Onunugbo *et al* (2017) had done some work. This concern prompted the production of "Guideline Levels" by the Joint Commission of the World Health Organization (WHO) and the United States Food and Agricultural Organization (FAO) (NCRS, 2020)

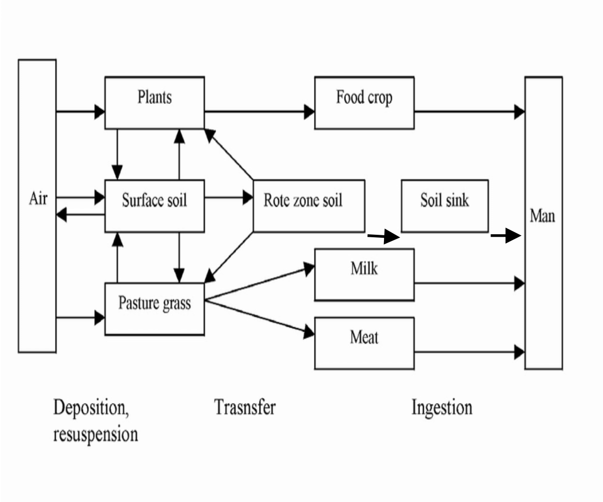

Fig. 1: Pathway of radiation transfer to humans (Carlos, 2004)

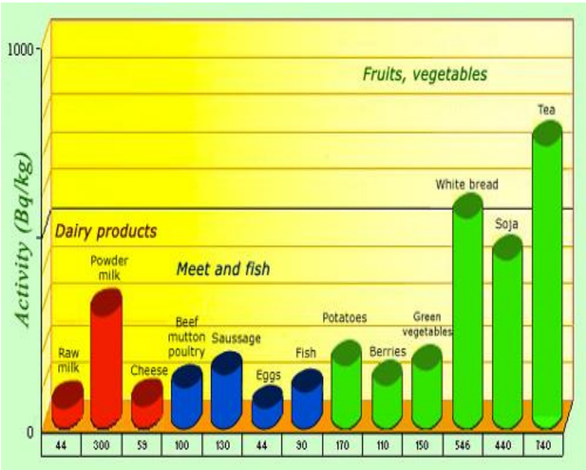

Fig. 2: NCRS data on radioactivity on some foodstuffs (NCRS, 2020)

A large portion of the Oshimili North Local Government area of Delta State, Nigeria is agrarian with most of the food produced being consumed within Asaba, the state Capital and the environs (John, 2014). With the probable non-existent literature regarding this study in this part of the State, this investigation which is aimed at evaluating the radionuclide contaminations of foodstuffs cultivated in this environment intends to provide base line data.

Methods

Study Area

The study area which is estimated to have 304,800 inhabitants (ManPower, 2022) is mostly inhabited by the Aniomas. Figure 3 is the map of Delta State showing the location of Delta North and its town of sample collection. It covers an approximate land area of 1084 km^2 which is partly riverine with large fertile land and a few mangrove forests. It lies between latitude of 6^0 19' 21.83"N and longitude of 6^0, 38'40.02" E for their DMS coordinates lat (long (dec) 6.32273, 6.64445), climate type tropical, The soil more conducive for agriculture the community is known for its farming culture and boast of a prosperous market the people are good in cultivation of cassava, yam potatoes and vegetables most of which are mostly consumed in Asaba and beyond (John., 2014).

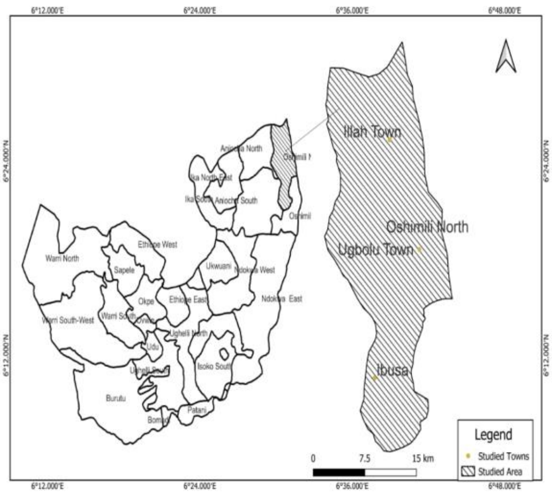

Fig. 3: Map of the study area (ManPower, 2022).

Sample Collection and preparation

The list of foodstuffs collected from the towns indicated in Figure 3 is presented as FS1- FS17, These samples were purchased from the local markets in the arbitrarily chosen towns. Pumpkin leaf, bitter leaf, okro, lemon grass and cocoyam were dried under sun for 24hrs while the other samples were oven dried at a temperature of 87°C. The samples were dried differently because of the nature and pattern of the food stuffs, the dried samples were blended using an industrial electric blender. The blender was cleaned using water and baking soda after each blending of samples to avoid contamination. Known weights of each the powdery samples were separately poured into empty Marinelli beakers. Each filled beaker was tightly sealed using masking tape and labelled appropriately. The purpose of the sealing is to ensure that radiation contaminated air in the laboratory environment does not find its way into the sample within the beakers. The sealed sample containers were stored in the environmental radioactivity laboratory in the Department of Physics and engineering in Obafemi Awolowo University (OAU), Ile–Ife, Nigeria where the radioactivity counting was carried out. The duration of the storage is 30 days. This is to ensure that the parent and daughter radionuclides in the samples achieved secular equilibrium which is a necessary requirement for natural radioactivity measurements (IAEA, 1989; Balogun *et al.*; 2003; Mokobia *et al.*; 2006).An empty beaker was only sealed and stored for the same period. This was used as background.

Table 1: Sampled foodstuffs with codes

Foodstuff	Botanical Name	Code
Okro	Abelmoschus esculentus	F1
Pepper	Capsiocum	F2
Cassava (raw)	Manihot esculenta	F3
Plantain	Musa paradisiacal	F4
Coconut	Cocos nucifera	F5
Melon	Cucumis melo	F6
Ogbono	Irvingia gabonensis	F7
Lemon grass	Citrus limon	F8
Beans	Phaseolus vulgaris	F9
Cocoyam	Colocasia esculenta	F10
Bread fruit	Artocarpus altilis	F11
Pumpkin	Cucurbita	F12
Yam	Dioscorea	F13
Cassava (processed)	Manihot esculenta	F14
Maize	Zea mays	F15
Rice	Oryza sativa	F16
Bitter leaf	Vermonia amygdalina	F17

Radioactivity Counting and Radionuclide Identification A NaI(Tl) gamma (γ) spectrometer was used to count the radioactivity in each of the stored food samples in the sealed Marinelli beakers. Prior to the counting, the spectrometer was calibrated both for energy and efficiency using appropriate standard calibration sources A specific spectrum having photopeaks representing the energies of the contained radionuclides was acquired. A corresponding spectrum was also obtained for the background. The counting and spectra accumulation time for all sample containers as well as that of the background was 36000s as specified by the international regulations (IAEA, 1989).A SAMPO 90 computer software coupled to the spectrometer was employed to highlight the energies of the γ photo peaksof the radio-contaminants within the samples. These radionuclides were then identified using the standard γenergies available in the literature (Gilmore,. 2008)

Quantification

The identified radionuclides were quantified using the formula (Oluyide *et al.*, 2019)

$$A_i(Bqkg^{-1}) = \frac{NC_i}{\varepsilon \times P_i \times M \times t} \qquad (1)$$

A_i is the specific activity of the ith radionuclide in the specific sample, ε : the efficiency of the detector in the spectrometer, NC_i : the net counts of the photopeak of the ith radionuclide (the net photo peak area), P_i : the γ % emission of the ith radionuclide in a specific sample, M : the mass of the sample and t: the time of counting (3600 s) .

The uncertainties in these calculations were determined using the equation (Sohiab and Sobieh, 2010):

$$\Delta A_i = \left\{\left[\frac{\Delta NC_i}{NC_i}\right]^2 + \left[\frac{\Delta\varepsilon}{\varepsilon}\right]^2 + \left[\frac{\Delta P_\gamma}{P_{\gamma i}}\right]^2 + \left[\frac{\Delta M}{M}\right]^2 + \left[\frac{\Delta t}{t}\right]^2\right\}^{\frac{1}{2}} \qquad (2)$$

Where ΔA_i is the error in A_i ΔNC_i : the error in NC_i $\Delta\varepsilon$: the error in ε , ΔP_γ : the error in P_γ . ΔM and Δt are the errors in M and t respectively.

Calculation of the Radiological Health Parameters

Radium equivalent activity (Ra_{eq}), this parameter represents the activity levels of ^{238}U, ^{232}Th and ^{40}K that will result in a radiological hazard (Beretka, 1985). It *is* based on the definition that 4810, 370, and 259 $Bqkg^{-1}$ of ^{40}K, ^{238}U and ^{232}Th, respectively, produce the same $\boldsymbol{\gamma}$ radiation dose effects. It was determined applying the equation the equation in Oluyide *et al.*, (2019) and UNSCEAR, (2000).

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \qquad (3)$$

Where A_U , A_{Th} and A_K are the respective specific activities of the specified radionuclides. .

Radiation absorbed dose rate (DR): to quantify the radiation energy absorbed by a potentially exposed individual, the radiation absorbed dose rate, DR was computed using the following formula as given by Oluyide *et al*. (2019).

$$D_R = RuAu + R_{Th}A_{Th} + R_K A_K$$

(4)

Where $Ru = 0.462$, $R_{Th} = 0.604$ and $R_K = 0.0417$ and A_U, A_{Th} and A_K are the Activity Concentrations for ^{238}U, ^{232}Th and ^{40}K ($Bqkg^{-1}$)

Annual effective dose equivalent (AEDE):

The annual effective dose rate (AEDR) in $mSvy^{-1}$resulting from the absorbed dose values (*DR*) was calculated using the following formula (UNSCEAR, 2000; Omenikono *et al.*, 2017; Sultana *et al*., 2020):

$$AEDE = D_R \times 1.2264 \times 10^{-3} \qquad (5)$$

Using a conversion factor of 0.7 SvGy-1 , which converts the absorbed dose rate in air to human effective dose and 0.2 for the outdoor occupancy factor proposed by the UNSCEAR 2000 report were used.

Hazard risk indices

A widely used hazard index (reflecting the external exposure) called the External Hazard Index, H_{ex} can be determined using the equation (5) (UNSCEAR, 2008)

$$H_{ex} = \frac{A_u}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (6)$$

In addition to External Hazard Index, radon and its short-lived isotopes are also hazardous to the respiratory organs. The internal exposure to radon and its daughter isotopes is quantified by the Internal Hazard Index, H_{in}, which is determined by equation (7)

$$H_{in} = \frac{A_u}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (7)$$

According to the International Commission on Radiological Protection (ICRP, 1994), the primary objective of H_{ex} and H_{in} is to limit the radiation dose to dose equivalent limit of 1 mSv y^{-1}. This implies that their values must be less than unity for the radiation hazards to be without any effect.

Excess lifetime cancer risk (ELCR)

$$ELCR = AED \times DL \times RF \qquad (2.6)$$

Where DL and RF are called the average lifetime duration and fatal cancer risk factor per sievert taken to be 54.5 years and 0.05 Sv^{-1} respectively.

Results

The tables below shows the results of the activity concentration and radiological health indices both hazard external and hazard internal

Table 2. The activity concentration of some staple foods in the study area

S/N	Sample Code	In-situ measurement (μSv/hr)	Activity concentration (Bq kg^{-1}) ^{40}K	^{238}U	^{232}Th
1	Okro	0.28	1095.94± 210.91	42.36±4.15	54.30±9.91
2	Pepper	0.20	875.34±168.46	25.22± 2.47	57.92±10.57
3	Cassava (raw)	0.26	785.62±151.19	37.21±3.65	67.26±12.28
4	Plantain	0.17	769.84 ±113.85	28.30 ±3.08	21.17 ±3.59
5	Coconut	0.15	615.27±118.41	32.93±3.23	45.72±8.35
6	Melon	0.25	1025.44±197.35	32.93± 3.23	31.41 ± 3.67
7	Ogbono	0.16	664.75± 107.75	29.44 ±2.97	24.94 ±3.07
8	Lemon Grass	0.23	548.74 ± 110.53	31.70±3.11	23.34 ±2.73
9	Beans	0.22	835.00±160.70	20.75± 2.03	22.42 ±3.02
10	Cocoyam	0.18	678.39 ± 151.25	29.95 ±3.02	21.79 ±3.15
11	Breadfruit	0.14	566.38±109.00	22.28 ±2.18	19.96±2.51
12	Pumpkin	0.24	508.26±97.82	41.23±4.04	33.98±11.51
13	Yam	0.15	562.41±108.24	21.17± 2.08	25.41 ±4.01
14	Cassava (Processed)	0.21	768.66 ±118.65	23.45 ±2.50	29.29 ±3.28
15	Maize	0.16	572.02 ±96.15	17.34 ± 3.21	20.75 ± 3.36
16	Rice	0.19	708.65±112.10	15.28 ±3.10	18.09 ±2.59
17	Bitter leaf	0.25	506.62± 102.10	34.46± 4.10	17.65±2.10

Table 3. The mean calculation of the activity concentration and radiological hazards indices in staple food samples in the study area.

	Activity concentration (Bq kg^{-1}) ^{40}K	^{238}U	^{232}Th	Ra_{eq} (Bq/Kg)	Hazard risk indices DR (nGyh^{-1})	AEDE (mSvy^{-1})	Hin	Hex	ELCR ($\times10^{-3}$)
Minimum value	508.26±97.82	15.34±4.15	17.65±2.10	44.40	44.40	0.05	0.29	0.26	0.16
Maximum value	1095.94±210.96	42.36±15.28	67.26±12.28	204.40	98.07	0.12	0.67	0.55	0.33
Mean value	709.42±131.46	28.59±2.82	31.49±5.08	129.77	62.53	0.08	0.43	0.35	0.20
(UNSCEAR,2000)	**420**	**33**	**45**	**370**	**59**	**0.10**	**≤1**	**≤1**	**0.29**

Table 4. Results of the absorbed dose rate (*D*R), the radium equivalent activity (Ra_{eq}), the index of external and internal radiation hazard (*H*ex, *H*in) for staple foods in the study area.

Food samples	Code	Ra_{eq} (Bq/Kg)	Radiological hazard risk indices in food samples				
			DR ($nGyh^{-1}$)	AEDE ($mSvy^{-1}$)	Hazard indices		ELCR ($\times 10^{-3}$)
					Hex	Hin	
Okro	FS1	204.40	98.07	0.12	0.55	0.67	0.33
Pepper	FS2	187.41	88.87	0.09	0.51	0.62	0.24
Cassava	FS3	193.88	90.58	0.11	0.52	0.62	0.30
Plantain	FS4	119.39	58.80	0.07	0.32	0.40	0.19
Coconut	FS5	145.69	68.49	0.08	0.39	0.48	0.23
Melon	FS6	156.81	76.95	0.09	0.42	0.51	0.26
Ogbono	FS7	127.49	61.07	0.07	0.34	0.43	0.19
Lemon	FS8	107.33	51.63	0.06	0.29	0.38	0.17
Beans	FS9	117.11	57.95	0.07	0.32	0.37	0.19
Cocoyam	FS10	113.30	55.27	0.07	0.31	0.39	0.18
Breadfruit	FS11	93.92	45.72	0.06	0.25	0.31	0.15
Pumpkin	FS12	128.52	60.77	0.07	0.34	0.40	0.20
Yam	FS13	100.81	48.58	0.06	0.27	0.33	0.16
Cassava	FS14	124.52	60.58	0.07	0.34	0.40	0.20
Maize	FS15	91.06	44.40	0.05	0.25	0.29	0.15
Rice	FS16	95.77	47.56	0.06	0.26	0.30	0.16
Bitter leaves	FS17	98.71	47.71	0.06	0.27	0.36	0.17
(UNSCEAR,2000)		**370**	**59**	**0.80**	**≤1**	**≤1**	**0.29**

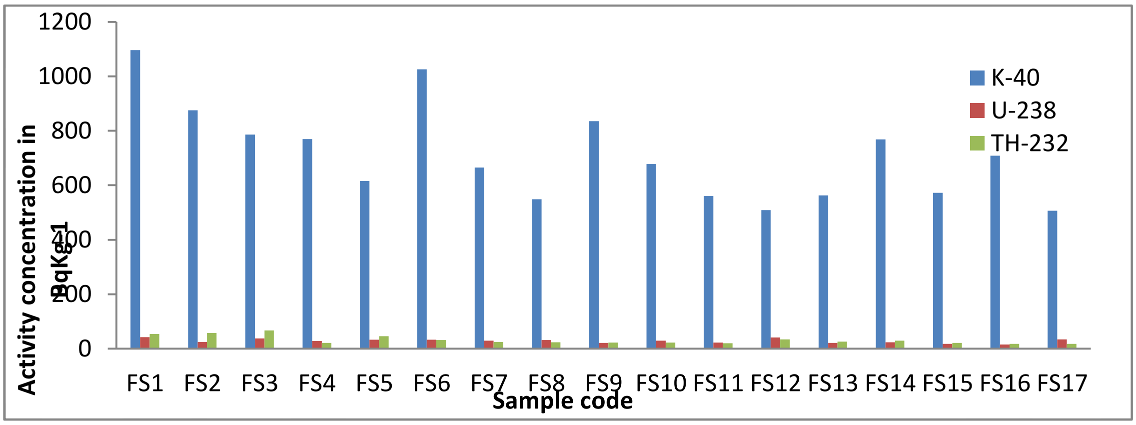

Fig.4. A statistical comparison of the activity concentration of radionuclides in various food samples in the study area.

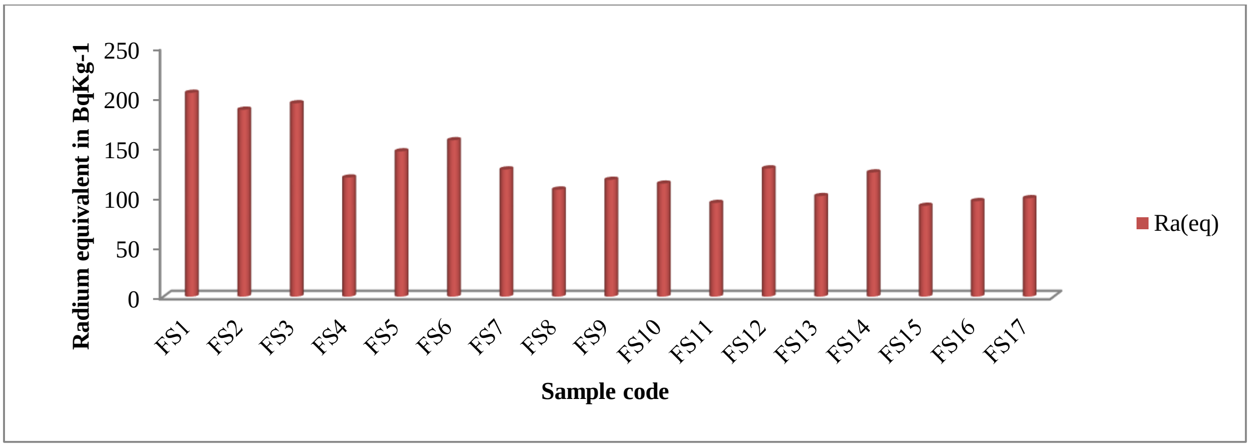

Fig. 5. A statistical analysis of the Radium equivalent of food samples in the study area.

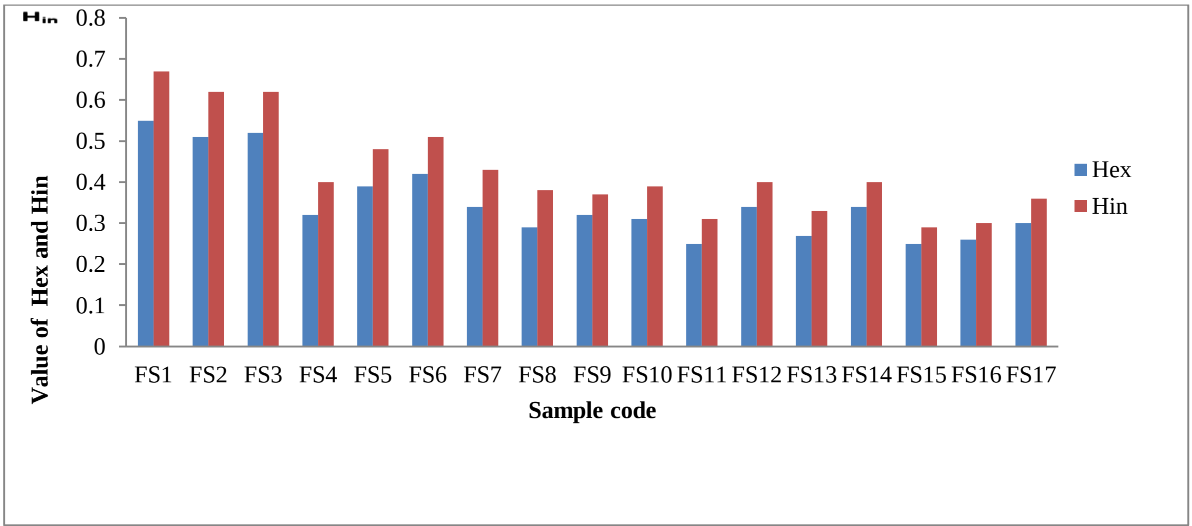

Fig. 6. A statistical analysis of radiological risk hazards in food samples in the study area

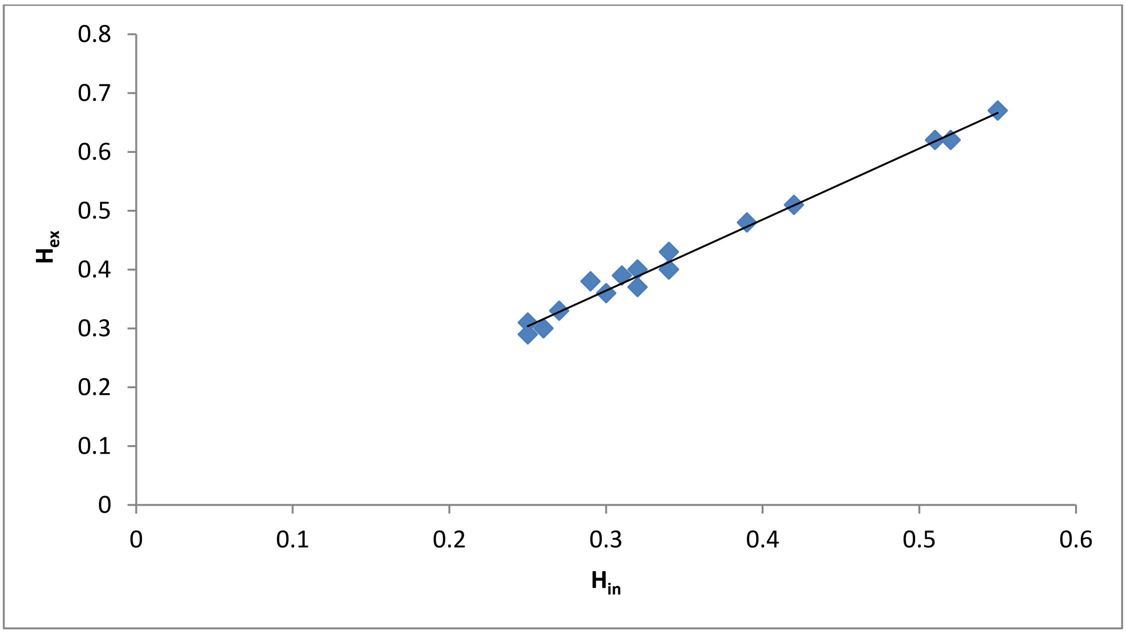

Fig 7. A graph of H_{ex} against H_{in}

Discussion

Table 1 shows the food samples with codes and their botanical names, table 2 depicts the activity concentrations of ^{40}K, ^{238}U and ^{232}Th in the studies found in foods consumed in Northern part of Delta state. The minimum and maximum values of the determined specific activities in Table 2 are 508.26 to 1095.94, 15.34 to 42.36 and 17.65 to 67.27 BqKg^{-1} for ^{40}K, ^{238}U and ^{232}Th respectively. The mean values are 709.42, 28.59 and 31.49 Bqkg^{-1}. The mean activity concentration as seen from figure 4 are in the order of ^{40}K > ^{232}Th > ^{238}U. This obtained activity values for ^{40}K is higher than world average limit of 420 Bqkg^{-1} .The concentration values obtained for ^{238}U and ^{232}Th are below the limit of 33Bqkg^{-1} and 45 Bqkg^{-1}. The activity concentration of ^{40}K is the highest in comparison to ^{238}U and ^{232}Th and this shows that much of the radioactivity concentration in food crops in the study area is due to the presence of this radionuclide which agrees with the result obtained by Tchokossa et al. (2013) in radioactivity assessment of food crops in Delta state, Nigeria.

Using the values of the activity concentration, Table 3 shows the results obtained for the absorbed dose rate (DR), the radium equivalent activity (Ra_{eq}), the index of external and internal radiation hazard (Hex, Hin) for staple foods consumed in Oshimili North. From Table 3, the radium equivalent in food samples ranges between 44.40 to 204.40 BqKg^{-1} and has mean value of 129.77 BqKg^{-1}. The mean value as compared with the world average permissible limit of 370 BqKg^{-1} is lower in value. The radiological hazard risk indices such as the absorbed dose rate, annual effective dose, the internal and external radiation hazard risk indices and the excess lifetime cancer risk index were presented in table 4. The absorbed dose rate as shown from Table 3 has a minimum value of 44.40 nGy·h^{-1} and maximum value of 98.07 nGy·h^{-1} with a mean value of 62.23 nGy·h^{-1}which is less than the recommended value of International Atomic Energy Agency (IAEA) (Guidebook, 1989) which is 1.0 mSvy-1. Furthermore, the Table 4 gives the values of internal radiation hazard risk index to be within the range of 0.29 to 0.67 and with a mean/average value of 0.43 which is needed to be less than unity to be considered negligible (UNSCEAR, 2000).

The annual effective dose equivalent (AEDE) have been measured to estimate the radiological risk due to the presence of ^{238}U, ^{232}Th and ^{40}K in the samples was calculated according to Sultana et al. (2020), to have values ranging from 0.05 to 0.12 representing the minimum and maximum values respectively with a mean value of 0.08, this value is 0.01 above the world permissible limit. In calculating the internal and external radiation hazard risk indices, the equation 7 and were used. In table 3, the internal radiation hazard risk index has value ranging from 0.29 to 0.67 with a mean value of 0.43. Similarly, the external radiation hazard risk index has a value ranging from 0.26 to 0.55 for the minimum and maximum values respectively, with mean value of 0.35. As shown, the mean values of the radiation hazard risk indices are shown to be below unity as recommended for safety by the world average permissible limit UNSCEAR (2000). The internal hazard index should also be less than one to provide safe levels of radon and its short-lived daughters for the respiratory organs of individuals living in the dwellings, the excess lifetime risk index was calculated to be between the range of 0.16 to 0.33 for the minimum and maximum values respectively. These values were less than unity in all the samples that indicate the non-hazardous for human being (Sultana, 2020). The Fig 5 shows a chart of comparison between all the staple food samples depicting the activity concentration of food samples in the study area. From the chart, we found ^{40}K as the highest radionuclide components in each food samples and thorium was the least. The maximum value was seen in Okro sample (FS1) (1095.94±210.91) and minimum value in FS 17 – Bitter leaf (506.62±102.10). The activity concentration of ^{40}K was highest in all the food samples and this could be due to the impact to the use of fertilizers by farmers to improve crop yields on the farms in the study area. The graph shows linearity of direct proportionality between the radiation hazard indices.

Conclusion

The work has shown that these food samples contains radionuclides with different activity concentrations and consequently with their associated radiological hazard risk indices. It is inferred that for all the food samples analyzed, the radium equivalent activity value is well defined within and less the permissible limits of 370 Bq·kg^{-1} irrespective of the high concentration of ^{40}K present in some samples, all which can be attributed to the addition of potassium induced fertilizers during farm practices. The radiological hazard risk indices such as the external, internal hazard index and excess lifetime cancer risk assessment are also below the world average permissible and recommended safe limit of <1,<1 and 0.29×10^{-3} respectively (UNSCEAR, 2000)**.**Therefore, it is an indication that the study area is safe for human activity (Omenikolo et al., 2017).

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