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Estimation of Effective Dose and Excess Lifetime Cancer Risks due to Ingestion of Natural Radionuclides in Rice Samples from Selected Farms in Southwestern Nigeria

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ABSTRACT

This study was carried out to evaluate the radioactivity concentrations of ⁴⁰K, ²³²Th and ²³⁸U in ten rice samples of the same species collected from different rice farms across Ondo and Ekiti States, southwestern Nigeria. The activity concentration measurements were determined using n-type co-axial HPGe detectors. In samples from one of the states (Ondo), the activity concentration was found to vary from 36.04 - 2139.35 Bq·kg⁻¹ for ⁴⁰K, 0.12 - 79.23 Bq·kg⁻¹ for ²³²Th and 0.22 - 83.36 Bq·kg⁻¹ for ²³⁸U. In the other state (Ekiti), the value was found to vary from 3.99 - 2078.91 Bq·kg⁻¹ for ⁴⁰K, 5.18 - 73.82 Bq·kg⁻¹ for ²³²Th and 14.50 - 83.21 Bq·kg⁻¹ for ²³⁸U. The mean annual effective dose to adults from the consumption of the rice were obtained as 0.93 and 0.84 mSv·y⁻¹ with excess lifetime cancer risk (ELCR) calculated to be 2.98 x 10⁻¹⁰ and 2.56 x 10⁻¹⁰ for Ondo and Ekiti, respectively. The values were compared with the internationally acceptable limits and were found to be lower compared to the acceptable world average ELCR value of 0.29 x 10⁻³ for radiological risk to the public by UNSCEAR. The rice samples from the locations are safe for human consumption.

Keywords: Radioactivity, Rice, Effective ingestion dose, Gamma spectroscopy, Excess life time cancer risk

1. Introduction

Life on earth has developed under the presence of environmental, gamma, and charged particle radiations [1,2]. It has however been established that ionizing radiation may harm life and biological systems. Natural radionuclides entering the food chain are mostly derived from the soil and as a result, high levels of radionuclides content in soils may be a prime source of radioactive contamination of food with plant uptake of radionuclides varying from species to species [3, 4, 5]. ⁴⁰K, ²³²Th, and ²³⁸U are the long-lived, naturally occurring radionuclides present in the earth's crust since the formation of the earth and they have significant contributions to the

ingestion dose owing to their presence in the biotic systems of plants and animals, in soil, water and air [6,7].

The distribution of radionuclides in different parts of plants depends on the chemical characteristic of the plants and soil in relation to the biological and physiological processes, climate, and agricultural practices [8, 9]. Radionuclides can be released into the environment because of the human activities including energy production and military operations such as nuclear weapon testing or through nuclear accidents [10]. Agricultural activities such as fertilizer application and spraying with pesticides can release technologically enhanced, naturally occurring radioactive materials into the environment as well [11]. When the radionuclides are

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released into the environment, they can be ingested or inhaled and could be deposited on the leaves of plants, soil, and water in varying concentrations, which are later, transferred and cycled through natural processes. The radionuclides eventually get into the human system through the food chain and thereby constituting a great radiological impact on man.

Rice is one of the most important cereal crops for human consumption in many parts of the world. It is the staple food for over 3 billion people (mostly the poor) constituting about half of the world's population [12, 13]. It is the only crop grown in the soil that is poorly drained and it is adapted to both wet and dry soil conditions if there is water [14]. The consumption of rice components that are high in natural radioactivity content can contribute to internal exposure of humans to ionizing radiation. Using gamma ray spectrometry with High Purity Germanium (HPGe) detector, Nahar et al. [15] reported the average activity levels of natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K in rice samples from Bangladesh to be 1.09 ± 0.31 , 0.17 ± 0.21 and 4.70 ± 1.59 $\text{Bq}\cdot\text{kg}^{-1}$, respectively. They further estimated the effective doses due to the consumption of the cultivated rice for the respective radionuclides to be 43.69, 16.39 and $4.15 \mu\text{Sv}\cdot\text{y}^{-1}$. Rice samples from Iraq were similarly investigated for ^{238}U , ^{232}Th , and ^{40}K but with NaI(Tl) detector. ^{238}U , ^{232}Th , and ^{40}K were reported to be of average values of 5.94, 2.65, and $16.84 \text{ Bq}\cdot\text{kg}^{-1}$ respectively [16]. In a German brand rice sample from Kuwait, anthropogenic ^{137}Cs was detected ($0.10 \pm 0.012 \text{ Bq}\cdot\text{kg}^{-1}$). This rare occurrence that does not come from natural sources was however attributed to the Chernobyl fallout [17]. Plants have been known to absorb radionuclides alongside nutrients from soils through their roots after which they are transported to other parts of the plant [18]. This transference is modelled using the soil-plant transfer ratio (TF) which is defined as the ratio of radioactivity unit per dry crop mass to that of unit per dry soil mass. Alsaffar et al. [19] analysed the effect of soil physico-chemical characteristics like pH, cation exchange capacity, electrical conductivity, organic matter, and soil texture on soil-grain TF in some rice samples from Malaysia. ^{40}K showed the highest TF factor of ($0.74 - 4.72 \times 10^{-1}$) ahead of ^{226}Ra ($0.06 - 0.36 \times 10^{-1}$) and ^{232}Th ($0.04 - 0.14 \times 10^{-1}$) with uptake discovered to be dependent on soil physico-chemical characteristics and plant component type (root, straw, husk, and grain). Grains were found to have the least activity concentrations for all three radionuclides. Although several studies on radioactivity levels in rice have been carried out in various countries across the world [20, 21], the literature reveals a small number of studies on the radionuclide content of rice grown in Nigeria [22, 23, 24]. Ugbede et al. [25] assessed the concentrations of ^{238}U , ^{232}Th and ^{40}K in rice samples and

soil from a cultivated paddy rice field in southeastern Nigeria and at different stages in the cultivation. ^{40}K had the highest TF of 0.72, higher than that of ^{238}U and ^{232}Th with TF of 0.27 and 0.21 respectively. This was attributed to accumulation of radionuclides in soil from previous farming seasons; a direct consequence of the continuous use of fertilizers which also has led to high ^{40}K activity concentration in the soil ($^{40}\text{K} = 68.45 \pm 20.40 \text{ Bq}\cdot\text{kg}^{-1}$, $^{238}\text{U} = 6.84 \pm 3.76 \text{ Bq}\cdot\text{kg}^{-1}$, $^{232}\text{Th} = 3.44 \pm 1.52 \text{ Bq}\cdot\text{kg}^{-1}$).

In this study, the investigation of the natural radioactivity due to ^{238}U , ^{232}Th , and ^{40}K has been carried out in order to quantify the radiological safety of rice grown from the selected farms in both Ondo and Ekiti states. The annual effective doses to the general public due to this consumption of the rice and the excess lifetime cancer risk was also estimated. It is expected that the results will serve as a baseline study in these locations for consequent radiological investigations.

2. Material and Methods

2.1 Sample collection and preparation

A total number of 10 rice samples were collected directly from ten rice farm settlements as shown in Figs.1, 2 and 3, five samples each from Ondo and Ekiti. All the samples were oven-dried at a temperature of 110°C until each sample attained a constant weight of 250 g. The rice grains were milled to remove the husk. The brown rice supplied from the milling process was then crushed by the grinding machine and sieved using a 2 mm sieve to obtain a fine texture of the samples. Each sample was placed in a Marinelli beaker of 250 ml, which was previously washed, rinsed with diluted HCl, and dried. Then, each sample in the Marinelli beaker was sealed for a month to allow sufficient time for both ^{232}Th and ^{238}U to attain a state of secular equilibrium with their corresponding progenies before the gamma spectroscopy analysis.

consumption per annum or annual intake of the foodstuff ($\text{kg}\cdot\text{y}^{-1}$) with rice having $U_r = 32 \text{ kg}\cdot\text{y}^{-1}$ as given by United States Department of Agriculture (USDA) [30], and G_{rf} is the ingested dose conversion factor for radionuclides ($\text{Sv}\cdot\text{Bq}^{-1}$). It should be noted that G_{rf} varies from one radionuclide to another. The total annual effective dose H_{rf} ($\text{Sv}\cdot\text{y}^{-1}$) to an individual was established by summing the contributions from all radionuclides present in the food samples. Thus, Eq. 2 can be rewritten as

$$H_{rf} = \sum G_{rf} U_r C_{rf} + \dots \quad (3)$$

Radiation doses ingested were obtained by measuring the radionuclides activities in the foodstuff ($\text{Bq}\cdot\text{kg}^{-1}$) and multiplying this by the masses of the food consumed over a period of time ($\text{kg}\cdot\text{y}^{-1}$). A dose conversion factor can then be multiplied to give an estimate of the ingested dose. The dose calculations were based on the assumption that each person obtained the food according to the consumption defined in the food balance sheet [31] and the radionuclides dose conversion factors were 2.8×10^{-7} for ^{238}U , 2.3×10^{-7} for ^{232}Th , and 6.2×10^{-9} for ^{40}K for the adult members of the public [32].

2.4 Excess Lifetime Cancer Risk (ELCR)

Excess lifetime cancer risk (ELCR) was obtained on the basis of the relation given in Eq.4:

$$ELCR = E_f \times D_L \times R_c \quad (4)$$

where $ELCR$, E_f , D_L , and R_c are the excess lifetime cancer risk, annual intake of the radionuclides (Bq), average duration of life (70 years) and mortality risk coefficient (Bq^{-1}). The mortality risk coefficient in (Bq^{-1}) are 9.56×10^{-9} for ^{238}U , 2.45×10^{-9} for ^{232}Th and 5.89×10^{-10} for ^{40}K [33].

3. Results and Discussion

Table 1 and 2 summarize the results of the measured concentrations of the naturally occurring radioactive elements ^{40}K , ^{232}Th , and ^{238}U and shows estimated dose values for the rice samples at the various locations. The specific activities of ^{40}K , ^{232}Th , and ^{238}U of Ondo rice samples varied between 36.04 – 2139.35, 0.12 - 79.23 and 0.22 - 83.36 $\text{Bq}\cdot\text{kg}^{-1}$ while those of Ekiti were between 3.99 - 2078.91, 5.18 – 73.82 and 14.50 – 83.21 $\text{Bq}\cdot\text{kg}^{-1}$, respectively. Compared to ^{232}Th , and ^{238}U , ^{40}K had the highest activity concentration in all the rice samples except in one location (Ado). The highest concentration of ^{40}K in Ondo rice samples (2139.35

$\text{Bq}\cdot\text{kg}^{-1}$) was found in Isua (Fig. 3), while ^{232}Th (79.23 $\text{Bq}\cdot\text{kg}^{-1}$) and ^{238}U (83.36 $\text{Bq}\cdot\text{kg}^{-1}$) concentrations were found to be the highest in the rice samples from Isua and Ile-Oluji, respectively. Ekiti had the highest activity concentrations of ^{40}K in Iye (2078.91 $\text{Bq}\cdot\text{kg}^{-1}$) rice sample while ^{232}Th (73.82 $\text{Bq}\cdot\text{kg}^{-1}$) was found to be the highest in Omuo rice sample. ^{238}U (83.21 $\text{Bq}\cdot\text{kg}^{-1}$) was found to be the highest in Omuo rice sample as well.

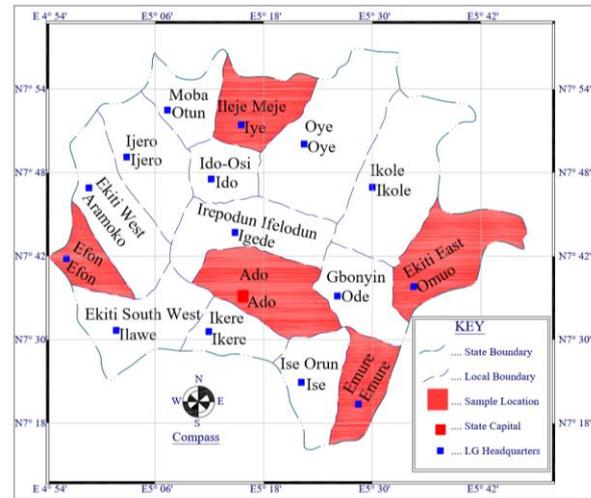


Fig.3 Map of Ekiti showing sample locations

The increase of ^{232}Th and ^{238}U in Isua and Ikaramu rice samples from Ondo State, and also in Omuo and Ado (Ekiti State) rice samples might be due to the local geology of the area [4]. Besides, the increase in ^{40}K in the rice samples from both Ondo and Ekiti States might as well be due to the extensive phosphate fertilizer application involved to improve the crop yields. The fact that the natural abundance of ^{40}K in soil is only about 0.012% shows that fertilizer use is the most probable reason for its high level in food [25]. Its solubility and high mobility make it easier for the radionuclide to be absorbed by plants via their roots [34].

The annual effective dose calculated using Eq. 2 had the highest values in Isua (Ondo state) and Omuo (Ekiti state) samples as 1.44 and 1.53 $\text{mSv}\cdot\text{y}^{-1}$, respectively. The annual effective dose values for all of the rice samples from the two states had a higher contribution to the dose when compared with the globally recommended value of 0.29 $\text{mSv}\cdot\text{y}^{-1}$ by UNSCEAR [35].

The total annual effective dose using Eq.3 was also determined for Ondo and Ekiti states with the mean annual effective dose values being 0.93 and 0.84 $\text{mSv}\cdot\text{y}^{-1}$, respectively, which were less than the limit of 1 $\text{mSv}\cdot\text{y}^{-1}$ set by the International Commission on Radiological

Table 1. Locations, coordinates, activity concentrations (Bq·kg⁻¹), annual effective doses (mSv·y⁻¹) and ELCR calculated for samples from Ondo

Locations	Lat.	Lon.	⁴⁰ K (Bq·kg ⁻¹)	²³² Th (Bq·kg ⁻¹)	²³⁸ U (Bq·kg ⁻¹)	AEDE (mSv·y ⁻¹)	ELCR
Isua	7°45'N	5° 87'E	2139.35	79.23	54.71	1.44	4.15x10 ⁻¹⁰
Ikaramu	7°39'N	5° 51'E	1786.09	52.56	21.43	0.89	1.83x10 ⁻¹⁰
Iju	7° 34'N	5° 18'E	36.04	0.12	0.22	0.00	1.50 x10 ⁻¹²
Ile-Oluji	7° 20'N	4° 87'E	1356.70	22.52	83.36	1.18	5.39 x 10 ⁻¹⁰
Okitipupa	6° 50'N	4° 78'E	1375.61	56.01	48.20	1.12	3.51 x10 ⁻¹⁰
Total			6693.79	210.44	207.92	4.64	1.49 x10 ⁻⁹
Mean ±S.E			1338.76 ± 318.74	42.09 ± 12.37	41.58 ± 12.77	0.93	2.98 x10 ⁻¹⁰

Table 2. Locations, coordinates, activity concentrations (Bq·kg⁻¹), annual effective doses (mSv·y⁻¹) and ELCR calculated for samples from Ondo

Locations	Lat.	Lon.	⁴⁰ K (Bq·kg ⁻¹)	²³² Th (Bq·kg ⁻¹)	²³⁸ U (Bq·kg ⁻¹)	AEDE (mSv·y ⁻¹)	ELCR
Iye	7° 95'N	5° 24'E	2078.91	28.24	27.15	0.86	2.16 x10 ⁻¹⁰
Efon	7° 65'N	4° 87'E	829.95	17.06	15.42	0.43	1.21 x10 ⁻¹⁰
Omuo	7° 46'N	5° 43'E	1237.39	73.82	83.21	1.53	5.98 x 10 ⁻¹⁰
Emure	7° 44'N	5° 46'E	699.45	5.18	14.50	0.31	9.92 x 10 ⁻¹¹
Ado	7° 37'N	5° 13'E	3.99	60.34	33.04	0.70	2.47 x 10 ⁻¹⁰
Total			4849.60	184.60	173.30	4.19	1.28 x10 ⁻⁹
Mean ±S.E			969.90 ± 305.10	36.90 ± 11.64	34.70 ± 11.30	0.84	2.56 x10 ⁻¹⁰

Protection [36]. The calculated values for the excess lifetime cancer risk for the two states varied from 1.50 x 10⁻¹² to 5.98 x 10⁻¹⁰. The average ELCR for the two states (i.e Ondo and Ekiti) were 2.98 x 10⁻¹⁰ and 2.56 x 10⁻¹⁰, respectively which were lower compared with the world average of ELCR limit of 0.29 x 10⁻³ for radiological risk in general [35]. For both states under study, Figure 4 and 5 show a comparison. This showed a similar trend between the samples among the neighboring states.

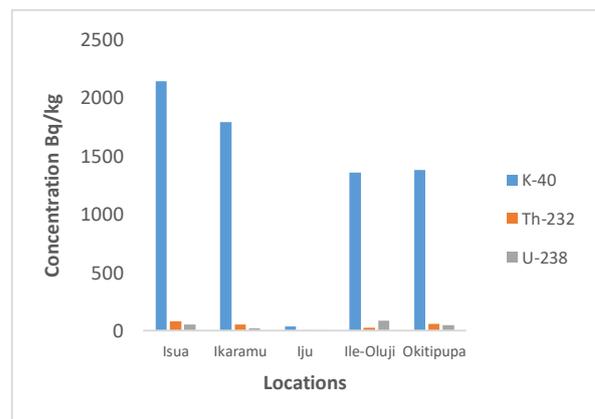


Fig.4 Radionuclides Concentration (Bq·kg⁻¹) of Rice Samples from Ondo State

Table 3. Comparison of activity concentrations of ^{40}K , ^{232}Th and ^{238}U in rice samples from different parts of the world

Country	Activity Concentrations (Bq·kg ⁻¹)			Reference
	^{40}K	^{232}Th	^{238}U	
India	70.30	0.15	-	[39]
Brazil	14.70	0.20	0.10	[40]
Thailand	22.00-23.00	0.02-0.30	-	[20]
Pakistan	7.00- 50.00	0.06-0.08	-	[20]
India	120.80	34.00	3.07	[7]
Malaysia	65.00-110.00	35.0-65.0	18.00-25.00	[28]
Ghana	104.00	4.00	5.00	[41]
Egypt	36.00	0.60	0.80	[17]
France	51.00	0.30	-	[17]
Germany	87.00- 101.00	0.40-0.50	-	[17]
Iran	-	-	37.00	[42]
Bangladesh	4.70	0.17	1.09	[43]
Iraq	16.80	2.70	5.90	[44]
Nigeria (Kogi)	41.15 ± 5.41	10.36 ± 1.72	12.73 ± 3.77	[22]
Nigeria (Ebonyi)	229.04	34.04	41.43	[25]
Nigeria (Ondo)	1339 ± 319	42.1± 12.4	41.6± 12.8	Present study
Nigeria (Ekiti)	970 ± 305	36.9 ± 11.6	34.7± 11.3	Present study
World Average	310.0	82.0	67.0	[35]

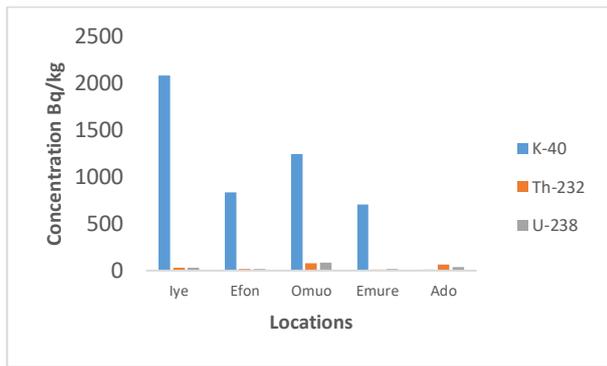


Fig.5 Radionuclides Concentration (Bq·kg⁻¹) of Rice samples from Ekiti State

The present results are compared to those reported in the literature. Table 3 shows the values of the activity concentrations of the present study which were higher with respect to the cases reported in the literature and the world average value of ⁴⁰K (310 Bq·kg⁻¹), but in tandem with the findings by Ajayi et al. [37] and Ayodele et al. [38] on the soils of both states. In the same vein, it is evident that the mean activity concentrations of ²³²Th and ²³⁸U in the rice samples from both states were a bit higher when compared with the results obtained from the other countries but still found to be within the recommended international limits of 82 and 67 Bq·kg⁻¹, for ²³²Th and ²³⁸U respectively [36]. The exceptions are in the rice samples from Isua and Ile-Oluji (Ondo State), and Omuo (Ekiti State) which were also higher than the internationally acceptable values (Table 1) [35]. However, the average concentrations of ²³²Th and ²³⁸U found in this study for the two states were below the UNSCEAR recommended values.

4. Conclusion

This study investigated the activity concentrations of radionuclides found in ten rice samples grown in Ondo and Ekiti States, Nigeria using gamma spectrometry with HPGe detector. The values of ⁴⁰K in this study seemed a bit higher compared to values reported in other parts of the world and the world in average. This observation in the radionuclides has been attributed to the excessive use of potassium rich phosphate fertilizers used by farmers to improve their crop yield. This result is an indicator that relevant agricultural agencies need to pay attention to the regulation of the use of soil enhancing chemicals as used by farmers.

The annual effective dose equivalent from the rice consumption by adults in the area was estimated using dose conversion factors for ⁴⁰K, ²³²Th and ²³⁸U. The results were found to be less than 1.0 mSv·y⁻¹ recommended by ICRP. Also, the excess lifetime cancer risk for the two states were lower compared to the acceptable ELCR limit of 0.29 × 10⁻³ for radiological risk in general. These values were significantly low to result in biological health risk of the farmers and the consumers. Hence, the rice sample are safe for human consumption. However, farmers should be sensitized on the danger of planting on high background areas and indiscriminate use of fertilizers.

Conflict of Interest

The authors have no conflict of interest.

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