Research Square

Preprints are preliminary reports that have not undergone peer review.
They should not be considered conclusive, used to inform clinical practice,
or referenced by the media as validated information.

Examination of Medicinal Plants for Radionuclides Absorption and Their Health Implications

Felix Adegoke Popoola (felixgoke@gmail.com)
Glorious Vision University
Oladotun Bamiji Isola
Glorious Vision University
Oluseye Daniel Fakeye
University of Ibadan
Tunde Ayobami Owolabi
Igbinedion University
Modupe Eunice Sanyaolu
Redeemer's University
Sheu Isiaq Owoyemi
Adeyemi Federal University of Education
Isaac Oluwafemi Elijah
Glorious Vision University

Research Article

Keywords: radiological hazards, medicinal plants, radionuclides adsorption, health safety, gamma spectrometry

Posted Date: July 7th, 2023

DOI: https://doi.org/10.21203/rs.3.rs-3099147/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License.
Read Full License

Additional Declarations: No competing interests reported.

EXAMINATION OF MEDICINAL PLANTS FOR RADIONUCLIDES ABSORPTION AND THEIR HEALTH IMPLICATIONS

Felix Adegoke Popoola[1]*, Oladotun Bamiji Isola[2], Oluseye Daniel Fakeye[3], Tunde Ayobami Owolabi[4], Modupe Eunice Sanyaolu[5], Sheu Isiaq Owoyemi[6], Isaac Oluwafemi Elijah[7]

[1]*Department of Mathematical & Physical Sciences, Glorious Vision University, Ogwa, Edo State, Nigeria.

[2]Department of Chemical Sciences, Glorious Vision University, Ogwa, Edo State, Nigeria.

[3]Department of Zoology, University of Ibadan, Ibadan, Oyo State, Nigeria.

[4]Department of Pharmacognosy, Igbinedion University, Okada, Edo State, Nigeria.

[5]Department of Physics, Redeemer's University, Ede, Osun State, Nigeria.

[6]Department of Integrated Science, Adeyemi Federal University of Education, Ondo, Ondo State, Nigeria

[7]Department of Works & Physical Planning, Glorious Vision University, Ogwa, Edo State Nigeria

***Corresponding author: Felix Adegoke Popoola**

Glorious Vision University (Formerly Samuel Adegboyega University), Ogwa, Edo State, Nigeria.

E-mail: felixgoke@gmail.com; fpopoola@sau.edu.ng

ORCID: https://www.orcid.org/0000-0003-3484-6883

Abstract

This study tests the concentration of ^{40}K, ^{238}U, and ^{232}Th radionuclides and also evaluates the possible radiological health risks in medicinal plants found in Ewu, Edo State, Nigeria, using a NaI(Tl) gamma spectrometer. The six selected medicinal plants are *Magnifera indica, Dacryodes edulis, Terminalia catappa, Cymbopogon citratus, Anacardium occidentale, and Persea Americana*. The results showed that the activity concentrations for ^{40}K ranged from 146.59 ± 4.81 in *Persea americana* to 296.08 3.42 Bq/kg in *Cymbopogon citratus* with a mean of 209.43 ± 5.14 Bq/kg, 238U ranged from 2.25 ± 0.06 to 5.57 ± 0.15 Bq/kg with a mean of 4.73 ± 0.15 Bq/kg and ^{232}Th varied from 4.50 ± 0.35 to 12.07 ± 0.57 Bq/kg with a mean of 8.00 ± 0.40 Bq/kg. The maximum and minimum activity concentrations of both ^{238}U and ^{232}Th were found in *Magnifera indica* and *Cymbopogon citratus* respectively. The calculated radiological hazards assessment due to the investigated medicinal plants is well within the internationally recommended safe limits. ^{232}Th contributes 54.91% to the total E_{CED}, while 6.35% for ^{238}U is the least.

^{232}Th exhibit a very strong, positive, and significant relationship with (E_{CED} and ELCR), and it contributes largely to the E_{CED} and ELCR due to ingestion of the examined herbal plant. Therefore, the use of these medicinal plants is radiologically safe for human consumption.

Keywords: *radiological hazards; medicinal plants; radionuclides adsorption; health safety; gamma spectrometry*

INTRODUCTION

Safety and quality of medicinal plant materials are becoming increasingly important due to global demand by health authorities, pharmaceutical industries, and also the public at large (WHO, 2007). Probably the oldest strategy still in use today for helping people deal with illness is the use of medicinal herbs. In many traditional medical systems around the world, medicinal plants have long been employed as a form of treatment. (WHO, 2007; Chandrashekara & Somashekarappa, 2016). The World Health Organization (WHO) has promoted the integration of traditional medicine's beneficial components into national health care systems in an effort to stimulate the use of traditional medicine worldwide (Oni *et al.*, 2011). This has served as alternative medical technique although, concerns about estimated dosages and the sanitary state of the medications produced using this method were addressed at various gatherings. (Oni *et al.*, 2011).

Medicinal plants can be found in their natural state or processed (Saudi *et al.*, 2022). Physical, chemical, and biological pollutants can be found in herbal remedies. (WHO, 2007). Human waste, animal manure, and sewage used as fertilizer can produce chemical and microbiological pollutants (WHO, 2007). The contamination of herbal material or products can be at various stages of the manufacturing process (WHO, 2007). The consumption of medicinal plants with high levels of natural radioactivity can cause health problems (Sussa *et al.*, 2013; Kareem *et al.*, 2016). Since most medicinal products are applied topically to the skin and taken orally, radioactive substances present in large amounts may expose people to radiation both internally and externally, which could harm humans (Oni *et*

al., 2011). The particular metabolic character of plant species may lead to the accumulation of radionuclides in their organs, which may depend upon the soil's physicochemical characteristics (Hashim *et al.*, 2019). An individual's annual effective dose from ingestion increases because of increased concentrations of radioactive elements in plants, increasing the risk of radiological harm.

Life-threatening pollution from a nuclear disaster could result from something quite different. WHO has developed protocols to follow in the case of significant radionuclide contamination brought on by a catastrophic nuclear accident (WHO, 2007). Actinides, activation products, and long- and short-lived fission products are a few examples of these radionuclides. For ^{238}U and ^{226}Ra, the lungs and kidneys are typically the sites of deposition; for ^{232}Th, the lungs, liver, or bones may all accumulate; and for ^{40}K, the body as a whole, but mostly in the muscles (Akhter *et al.*, 2007; Nahar *et al.*, 2018; Ugbede *et al.*, 2022). As a result, the immune system of the body may weaken in the face of many illnesses with higher death rates (Nahar *et al.*, 2018; Ugbede *et al.*, 2022). The amount of exposure to radiation depends on the intake of radionuclides, and its significance depends in turn on other variables such as the age, metabolic kinetics, and weight of the individual who ingests them (also known as the dose conversion factor) (WHO, 2007). The level of contamination might be reduced during the manufacturing process (WHO, 2007). To determine if people have been exposed to various radiation levels directly or indirectly in this regard, it is crucial to test the radioactivity of the environment and native herbs.

The radiation levels and radioactivity concentrations in some foodstuffs like vegetables, fruits, tubers, maize, and meat from the oil and gas production areas in Delta State, Nigeria, as reported by (Tchokossa *et al.*, 2013) indicated that the primordial radionuclides were within the internationally recommended safe limits in the food types grown in the area. Jwanbot *et al.*, (2013) obtained results that indicate that the activity concentration of the farmland soils and crops is a

significant health risk for the farmers and consumers of the crops in Barkin Ladi LGA, Plateau State, Nigeria. The assessment done by Sowole & Olaniyi (2018) to determine the concentrations of natural radionuclides in some fruits from markets at Ijebu-Ode town in Nigeria reveals that the highest concentrations of ^{40}K, ^{238}U and ^{232}Th were found in pineapple ($102.36 \pm 10.81\ \mathrm{Bqkg^{-1}}$), orange ($12.18 \pm 4.36\ \mathrm{Bqkg^{-1}}$) and mango ($8.01 \pm 3.25\ \mathrm{Bqkg^{-1}}$) respectively, with an average annual committed effective dose of $0.11\ \mathrm{mSv.yr^{-1}}$, which is lower than the limit of $0.3\ \mathrm{mSv.yr^{-1}}$ recommended by UNSCEAR (2000) implying that the ingestion of these fruits poses no radiological health hazard to consumers. Numerous authors have documented radioactivity in medicinal plants in Nigeria's River State (Port Harcourt), Niger State, and Ogun State (Njinga *et al.*, 2015; Oni *et al.*, 2011; Sowole & Olaniyi, 2018). Accordingly, the findings of this study can be utilized as a baseline value for radioactivity in medicinal plants in Esan Land, Edo State, Nigeria. According to Abojassim & Lawi (2018), it is crucial to assess the radionuclide concentrations in medicinal plants in order to avoid subjecting consumers to radiation. Therefore, this research was conducted to determine the level of natural radionuclide absorption by medicinal plants and their health implication using medicinal herbs grown in Ewu, Edo State, Nigeria.

MATERIALS AND METHODS

The sampling area

The medicinal herbs chosen for this study originated from Ewu in Edo State's Esan-Central Local Government Area (LGA). The town is located 200 feet above sea level in Edo State's plateau region and is 100 kilometres north of Benin City, the state's capital. Agbede, Irrua, and Ekpoma form the northern, southeast, and southwest borders of Ewu, respectively (Omonhinmin, 2017). In spite of the fact that farming constitutes the majority of the population's work, occasionally government employees, students, traders, and other workers also engage in it. The existence of a

modern herbal manufacturing company (Pax Herbals) has greatly influenced interest in the cultivation of herbs for both private and commercial uses; as a result, the majority of these people have extensive native knowledge of traditional medicine as it is their main source of healthcare and income.

Sample collection and preparation

Samples of six (6) different medicinal plant parts (2 kg each) commonly used and available in the localities of Ewu were collected. Table 1 shows medicinal plants and their medical uses.

Table 1: Name and uses of selected medicinal plants

Sample Code	Scientific Name	Common name	Uses	Part used
A	*Magnifera indica*	Mango tree	To treat diabetes, restlessness, gall and kidney stone, respiratory problems, dysentery	Leaves
B	*Dacryodes edulis*	African Pear	To treat wound, skin diseases, dysentery and fever	Leaves
C	*Terminalia catappa*	Umbrella tree	Gonorrhea, ulcers, cough, catarrh, haemoptysis, cholagoghe, astringent, cardiac tonic	Leaves
D	*Cymbopogon citratus*	Lemmon grass	For treating rheumatism, fever, high blood pressure, convulsion, stomachache, vomiting, cough	Leaves
E	*Anacardium occidentale*	Cashew tree	Diarrhhoea, blood pressure, reducing blodd sugar level, oral ulcer, sore throat	Leaves
F	*Persea americana*	Avocado	To improve digestion, prevent cancer, regulate blood pressure, improve vision	Leaves

The fresh leaves of the selected medicinal plants were collected within the locality of Ewu town, Esan LGA, Edo state, Nigeria in the month of September 2022. The plants were identified and authenticated the Professor J.C. Okafor Herbarium, Pax Herbal Clinic and Research Laboratories, Ewu, Nigeria, and voucher specimens were deposited. The leaves were treated separately by sorting, rinsed under running water, and air-dried at room temperature to remove the surface moisture for a period of 5 days. Thereafter, they were separately transferred into an oven maintained at 50˚C for another 2 days before pulverizing into powder using an electric milling machine (Chris Norris, England) (Owolabi *et al.*, 2019), and 300 g of each sample was packaged into a labelled and tightly sealed plastic container and kept for 1 month to ensure secular equilibrium in the ^{238}U and ^{232}Th with their respective progeny (Popoola *et al.*, 2019). The samples were then transported to the Radiation and Health Physics Laboratory of the Ladoke Akintola University of Technology, Ogbomoso, Nigeria, for gamma spectrometric measurement.

Gamma Spectrometry Measurement

A 3" x 3" NaI(Tl) detector made by Princeton Gamma Tech in the USA is the basis of the gamma spectrometry equipment used. The detector is protected from background radiation by a cylindrical lead shield. The Gamma Spectacular (type GS-2000 Pro) multichannel analyzer was connected to the detector, and the computer was connected to the analyzer for display. Theremino software was used to acquire data and analyze gamma-ray spectra.

The NaI(Tl) spectrometry system was calibrated to determine a qualitative and quantitative relationship between the peak position in the spectrum and the related gamma-ray energy. With the use of the RSS8 gamma source set, which can be traced to Spectrum Techniques LLC, USA, the detector's energy was calibrated. It was done by examining the spectra of two-point sources (Cs-137 and Co-60) that generate gamma rays with known energy. A reference source of known

radionuclide activity, including ^{40}K (578.4 Bq/kg), ^{238}U (20.9 Bq/kg), and ^{232}Th (10.47 Bq/kg), was used to calibrate the detector's effectiveness. The standard sources are intended for the detection of radionuclides such as ^{40}K, ^{238}U, and ^{232}Th in the medicinal plants.

The background gamma-ray distribution count was calculated by counting an empty container for 36000s prior to the sample measurement. Once the sealed samples had reached a state of secular equilibrium, they were all sequentially put on the detector for analysis. Every sample was counted for the same amount of time as the container's empty counterpart. The most notable radionuclides found in the samples were identified as having energies of 1460.0 keV (^{40}K), 1764.5 keV of ^{214}Bi (^{238}U), and 2614.7 keV of ^{208}Tl (^{232}Th). Each detected radionuclide in the sample had its activity concentration A ($Bqkg^{-1}$) measured by:

$$A = \frac{C_{net}}{P_\gamma \times \varepsilon \times v \times t} \quad (1)$$

Where C_{net} is the net peak count for each radionuclide present in the sample after subtracting the background count from the gross count, $P\gamma$ is the absolute gamma-ray emission probability of the identified radionuclide, ε is the obtained full energy peak efficiency for each identified radionuclide, v is the volume of the sample and t is the counting time.

Estimation of Radiological Hazard

After determining the values for the individual naturally occurring radionuclides' specific activity concentrations in medicinal plants, the effects of radiation on people's health and the environment are evaluated using the radiation hazard below.

i. **Radium Equivalent Activity, (Ra_{eq})**

The radium equivalent (Ra_{eq}) activity is used to compare the activities of ^{238}U, ^{232}Th and ^{40}K in samples by a single quantity (Ugbede, 2020). It is the weighted sum of activities of ^{238}U, ^{232}Th and

^{40}K. It is calculated that 1 Bq/kg of ^{238}U, 0.7 Bq/kg of ^{232}Th and 13 Bq/kg of ^{40}K provide the same radiation dosage. The radium equivalent activity index was computed using (Ononugbo *et al.*, 2017; Sultana *et al.*, 2020).

$$\mathrm{Ra_{eq}\ (Bqkg^{-1})} = \mathrm{C_U} + 1.43\mathrm{C_{Th}} + 0.077\mathrm{C_K} \quad (2)$$

Where $\mathrm{C_U}$, $\mathrm{C_{Th}}$ and $\mathrm{C_K}$ are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively in $\mathrm{Bqkg^{-1}}$.

ii. **Absorbed Dose Rate**

The amount of energy that ionizing radiation deposits per unit mass in a material is measured by the absorbed dose. The dose rate, D, due to the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K was determined in accordance with (Ofomola *et al.*, 2023; Popoola *et al.*, 2019; UNSCEAR, 2000):

$$\mathrm{D\ (nGy/h)} = 0.462\ \mathrm{C_U} + 0.604\ \mathrm{C_{Th}} + 0.0417\ \mathrm{C_K} \quad (3)$$

Where $\mathrm{C_U}$, $\mathrm{C_{Th}}$ and $\mathrm{C_K}$ are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively, and coefficients are dose rate conversion factors.

iii. **Hazard Indices ($\mathbf{H_{ex}}$ and $\mathbf{H_{in}}$)**

Indicators for external hazard, $\mathrm{H_{ex}}$ and the internal hazard, H_{in} were estimated using (UNSCEAR, 2000; Ononugbo *et al.*, 2017; Ugbede, 2020):

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (4)$$

$$H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} < 1 \quad (5)$$

Where $\mathrm{C_U}$, $\mathrm{C_{Th}}$ and $\mathrm{C_K}$ are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively

iv. **Annual Gonadal Equivalent Dose (AGED)**

The Annual Gonadal Equivalent Dose (AGED) is the risk radiation poses to susceptible cells, such as the gonads, bone marrow, and surface cells. It has been observed that an increase in AGED affects the bone marrow, causing the destruction of red blood cells and their replacement by white blood cells. This syndrome manifests itself as leukemia, a deadly type of blood cancer. (Ononugbo *et al.*, 2017). The equation (Avwiri *et al.*, 2014; Saudi *et al.*, 2022) is used to determine AGED given activity concentrations of ^{238}U, ^{232}Th and ^{40}K:

$$AGED\ (Sv/yr) = 3.09\ C_U + 4.18\ C_{Th} + 0.314\ C_K \quad (6)$$

Where C_U, C_{Th}, and C_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively. The standard UNSCEAR value for AGED is 300 $mSv.yr^{-1}$.

v. **Representative Gamma Index (I_γ):**

This is done to determine the gamma radiation hazards connected to the radionuclides in the samples of medicinal plants under investigation. The representative gamma index is given as follows (Ofomola *et al.*, 2023; Ononugbo *et al.*, 2017):

$$I_\gamma = \frac{C_U}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \quad (7)$$

Values of $I_\gamma \leq 1$ indicate an annual effective dose of less than or equal to 1 mSv, whereas $I_\gamma \leq 0.5$ indicates an annual effective dose less than or equal to 0.3 mSv (Ugbede, 2020).

vi. **Average annual committed effective dose** (E_{CED})

The E_{CED} for ingestion of NORMS in medicinal plants was calculated using the expression in equation (8) (Saudi *et al.*, 2022; Tettey-larbi *et al.*, 2013):

$$E_{CED} = A_c * DCF_{ing} * C_r \quad (8)$$

where E_{CED} is the average annual committed effective dose, A_c is the activity concentration of each radionuclide in the plant sample, DCF_{ing} is the dose conversion coefficient for ingestion for

each radionuclide (i.e. 6.2×10^{-9} mSv/Bq, 4.5×10^{-8} mSv/Bq, and 2.3×10^{-7} mSv/Bq for ^{40}K, ^{238}U, and ^{232}Th respectively for an adult (UNSCEAR 2000), and C_r is the rate of consumption rate from intake of NORMS in medicinal plants.

Although there is no set dosage for medicinal plants in Nigeria, it was assumed that a patient requires 100 mL/day of herbal preparation or product to successfully treat a common condition (Njinga *et al.*, 2015). As a result, it was assumed that the medicinal plants used in the present study had a NORM consumption rate of 1 kg/yr (Alade *et al.*, 2020; Njinga *et al.*, 2015).

vii. **Excess Lifetime Cancer Risk (ELCR)**

This has to do with the potential for a lifelong case of cancer at a specific level of exposure. It is expressed as a number that indicates the number of extra cancers that could be detected in a specified number of individuals. This is following exposure to a carcinogen at a particular dose. According to Orosun *et al.* (2018), Popoola *et al.* (2019), and Saudi *et al.* (2022), it is calculated as follows:

$$ELCR = E_{CED} \times DL \times RF \quad (9)$$

Where E_{CED} is the average annual committed effective dose equivalent, RF is the fatal risk factor and DL is the duration of life. The RF to the public is taken to be $0.05\ \mathrm{Sv/y}$ (Popoola *et al.*, 2019) and DL is 54.5 years, the life expectancy at birth in Nigeria (WHO, 2015).

RESULTS AND DISCUSSION

Activity concentrations in the selected medicinal plants

Table 2: Activity concentration (Bq/L) of natural radionuclides in the medicinal plant samples

Sample Code	Scientific Name	^{40}K	^{238}U	^{232}Th
A	*Magnifera indica*	241.39 ± 6.16	5.57 ± 0.15	12.07 ± 0.57

B	*Dacryodes edulis*	202.14 ± 5.64	4.97 ± 0.29	7.76 ± 0.18
C	*Terminalia catappa*	185.67 ± 5.41	5.31 ± 0.09	9.19 ± 0.50
D	*Cymbopogon citratus*	296.08 ± 3.41	2.25 ± 0.06	4.50 ± 0.35
E	*Anacardium occidentale*	184.71 ± 5.40	5.06 ± 0.08	9.51 ± 0.59
F	*Persea americana*	146.59 ± 4.81	5.21 ± 0.25	4.97 ± 0.19
	Min	146.59 ± 4.81	2.25 ± 0.06	4.50 ± 0.35
	Max	296.08 3.42	5.57 ± 0.15	12.07 ± 0.57
	Mean	209.43 ± 5.14	4.73 ± 0.15	8.00 ± 0.40

The results of activity concentrations of ^{40}K, ^{238}U and ^{232}Th obtained for the medicinal plants samples in Ewu is presented in Table 2. The average activity concentrations in the medicinal plants' samples for ^{40}K ranged from 146.59 ± 4.81 (in *Persea americana*) to 296.08 3.42 Bq/kg (in *Cymbopogon citratus*) with a mean of 209.43 ± 5.14 Bq/kg; for ^{238}U, it ranged from 2.25 ± 0.06 to 5.57 ± 0.15 Bq/kg with a mean of 4.73 ± 0.15 Bq/kg and ^{232}Th varied from 4.50 ± 0.35 to 12.07 ± 0.57 Bq/kg with a mean of 8.00 ± 0.40 Bq/kg. The maximum and minimum activity concentration of both ^{238}U and ^{232}Th were found in *Magnifera indica* and *Cymbopogon citratus* respectively. As can be noted, the radionuclides were present in all the examined samples, though at varying concentrations. Variances in the geological location and radiochemical makeup of the soils where medicinal plants are produced cause variations in natural radionuclide activity concentrations (Tettey-larbi *et al.*, 2013).

The mean concentrations of ^{40}K, ^{238}U, and ^{232}Th radionuclides in the examined medicinal plants are lower than the worldwide average of 400 Bq/kg for ^{40}K, 33 Bq/kg for ^{238}U, and 45 Bq/kg for ^{232}Th (UNSCEAR, 2000). It is obvious that the activity concentration of ^{40}K was the highest of all the samples (Fig. 1).

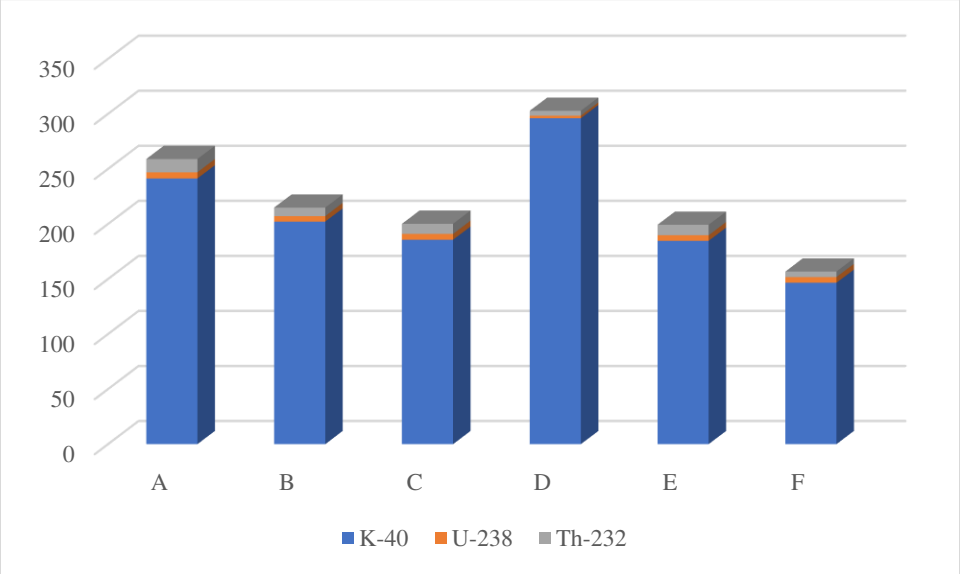

Fig.1: Activity concentration in selected samples of medicinal plants.

According to their metabolic needs and potential regional differences, plants may absorb varied amounts of potassium from soil (Kolapo & Omoboyede, 2018). Popoola *et al.*, (2019) reported radionuclide concentration of 57.80 ± 1.7 Bq/Kg and 30.19 ± 1.22 Bq/Kg in the soil of higher educational institutions in Ogwa and Igueben, respectively. The results of ^{40}K in this study imply that the area under study has experienced some ^{40}K enrichment. These plants have a high potassium activity concentration because they can absorb potassium from the soil more readily than other elements (Tettey-larbi *et al.*, 2013) and maybe as a result of fertilizer application by farmers to increase agricultural yields. When compared to the concentration reported by (Oni *et al.*, 2011) for Ugheli and Ogbomoso, the ^{40}K concentrations for *Cymbopogon citratus* obtained in this study are significantly higher. Similar to this, *Terminalia catappa* had ^{40}K concentrations that were greater than the 145.59 ± 7.19 Bq/kg reported by (Njinga *et al.*, 2015) but with elevated concentration of

^{232}Th. *Magnifera indica's* activity concentration of ^{40}K for this investigation is lower than the 389.63 ± 34.42 Bq/kg reported by (Alade *et al.*, 2020). The high activity concentration of ^{40}K in leaves may be due to the significant function that potassium plays in plant photosynthesis, which causes it to accumulate more in leaves where photosynthesis occurs most frequently. (Saenboonruang *et al.*, 2018).

Table 3: Comparison of activity concentration (Bq/L) of natural radionuclides in medicinal plant samples.

	Activity concentration (Bq.kg^{-1})			**References**
Country	**^{40}K**	**^{238}U**	**^{232}Th**	
Nigeria	**209.43**	**4.73**	**8.00**	This study
Bangladesh	661.10	12.65	7.38	(Sultana *et al.*, 2020)
Iraq	219.1	4.68	2.91	(Kareem et al., 2016)
Turkey	1910	-	5.67	(Cengiz & Çağlar, 2019)
Egypt	471.4	7.25	7.78	(Saudi *et al.*, 2022)
India	230.00	-	36.00	(Monica *et al.*, 2020)
Uganda	359.59	-	9.65	(Biira *et al.*, 2021)
Nigeria	171.72	-	35.09	(Njinga *et al.*, 2015)
Nigeria	630.02	5.79	4.13	(Alade *et al.*, 2020)
Ghana	839.80	31.78	56.16	(Tettey-larbi *et al.*, 2013)
World	400	33	45	UNSCEAR, 2000

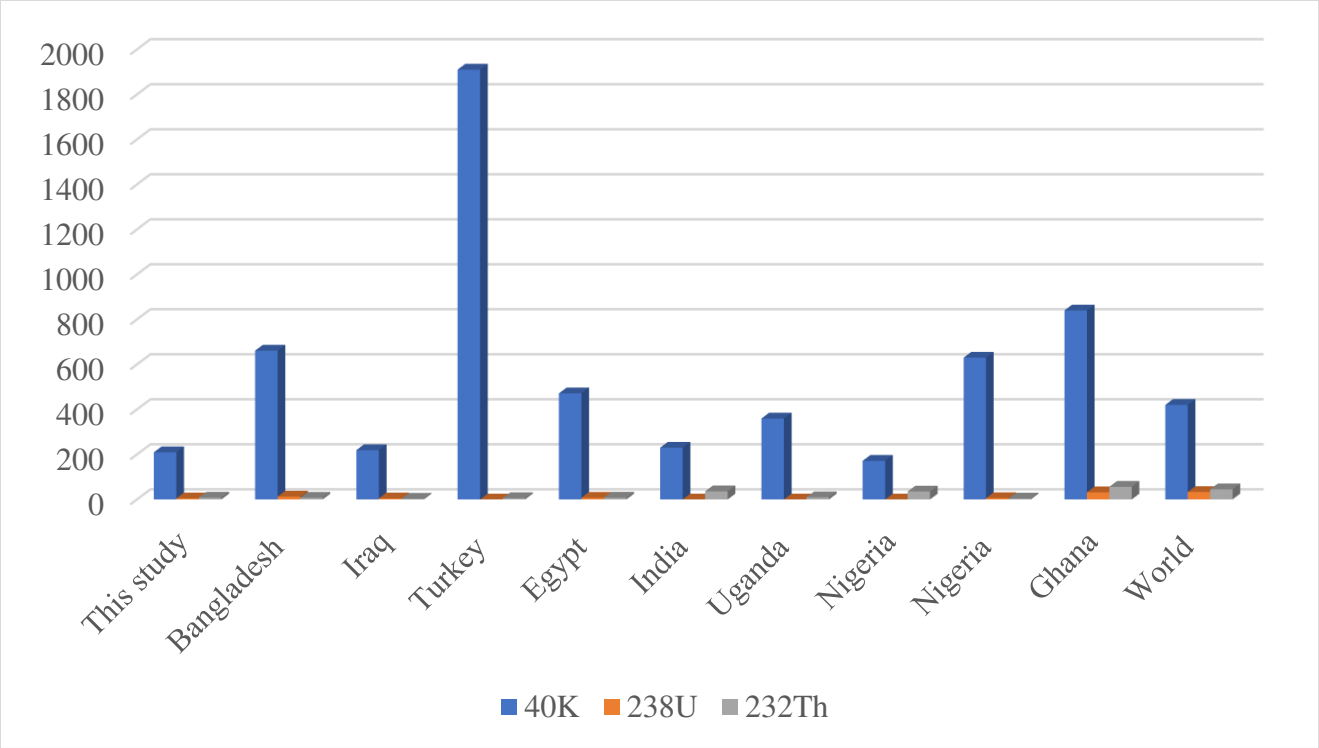

Fig. 2: Comparison of the mean activity concentration (Bq/L) of ^{40}K, ^{238}U and, ^{232}Th radionuclides in medicinal plant samples with other parts of the world.

A comparison of the mean activity concentration of ^{40}K, ^{238}U and, ^{232}Th radionuclides in the examined medicinal plant to other studies in other parts of the world is presented in Table 3 and illustrated in Fig. 2. There is no doubt that 40K is prevalent in medicinal plants around the world, as can be seen from the chart.

The activity concentration of ^{40}K in this study is higher than the results reported by (Njinga *et al.*, 2015) in Lapai, Niger State, Nigeria, and slightly below the results found in Iraq (Kareem *et al.*, 2016) and India (Monica *et al.*, 2020). However, it is more than nine times lower than that found in Turkey (Cengiz & Çağlar, 2019), and four times lower than that recorded in Ghana (Tettey-larbi *et al.*, 2013). It is lower than that of Uganda (Biira *et al.*, 2021) and is more than three times lower than that recorded in Bangladesh (Sultana *et al.*, 2020), and Ibadan (Alade *et al.*, 2020), but more

than twice lower than that obtained in Egypt (Saudi *et al.*, 2022). Our results demonstrate that the level of ^{238}U in medicinal plant samples is comparable to that reported by Kareem *et al.* (2016) in Iraq but lower than that Saudi *et al.* (2022) in Egypt, Sultana *et al.* (2010) in Bangladesh, Alade *et al.* (2010) in Ibadan, Nigeria, and Ghana (Tettey-larbi *et al.* (2013). The findings of the ^{232}Th study demonstrate that our findings are comparable to those of studies conducted in Egypt and Bangladesh, respectively (Saudi *et al.*, 2022) and Bangladesh (Sultana *et al.*, 2020). Also, ^{232}Th is higher than the results found in Iraq (Kareem *et al.*, 2016), Ibadan, Nigeria (Alade *et al.*, 2020), and Turkey (Cengiz & Çağlar, 2019) and lower than the results found in India (Monica *et al.*, 2020), Uganda (Biira *et al.*, 2021), Lapai, Nigeria (Njinga *et al.*, 2015), and Ghana (Tettey-Larbi *et al.*, 2013). It's possible that variances in geography, geology, and the mineral makeup of different sample locations are accountable for the discrepancy in natural radioactivity concentrations across the various sampling sites.

Radiological hazard indices for medicinal plants

Table 4 presents the estimated radiological hazard indices in medicinal plant samples. The samples' results for radium equivalent activity ranged from 23.6 ± 0.89 to 41.42 ± 1.44 Bq/kg, with a mean of 32.29 ± 1.12 Bq/kg. The obtained mean value was less than the Ibadan finding (Alade *et al.*, 2020) and the 370 Bq/kg allowable global threshold (UNSCEAR, 2000). Additionally, it was presented that the absorbed dose rate varied from 11.52 ± 0.43 to 19.93 ± 0.67 nGy/h with a mean of 15.75 ± 0.52 nGy/h which is lower than the public exposure rate of 84 nGy/h (UNSCEAR, 2000). Also, the estimated values of H_{ex} and H_{in} are less than unity, with corresponding mean values of 0.087 ± 0.003 and 0.100 ± 0.003 respectively in the samples. These values do not exceed the 1 upper maximum that is allowed. The values of H_{ex} and H_{in} concur with (Saudi *et al.*, 2022). The annual gonadal equivalent dose (AGED) was also assessed and can be found in Table 4. AGED values have

303 a minimum value of 82.90 ± 3.08 Sv/yr and a maximum of 143.46 ± 4.78 with an average of 113.81
304 ± 3.75 Sv/yr. All values are below the stated value of (Saudi *et al.*, 2022) and are within the range
305 of the 300 Sv/yr global recommendation (UNSCEAR, 2000). Another index evaluated is the gamma
306 representative index, I_γ which is a screening parameter for medicinal plants of possible radiological
307 health burden. The minimum, maximum and mean values are 0.18 ± 0.007, 0.32 ± 0.011 and 0.250
308 ± 0.008. the mean value compares well with (Saudi *et al.*, 2022). All samples of medicinal plants
309 had I_γ values below 1, as observed, suggesting that they pose no radiological risk and may not have
310 any adverse effects.

Table 4: Radiological hazard indices in medicinal plant samples

Sample Code	Scientific Name	Ra_{eq} (Bq/kg)	D (nGy/h)	H_{ex}	H_{in}	AGED mSv/y	Gamma Index
A	*Magnifera indica*	41.42 ± 1.44	19.93 ± 0.67	0.112 ± 0.004	0.127 ± 0.004	143.46 ± 4.78	0.32 ± 0.011
B	*Dacryodes edulis*	31.63 ± 0.98	15.41 ± 0.48	0.085 ± 0.003	0.099 ± 0.003	111.27 ± 3.42	0.25 ± 0.007
C	*Terminalia catappa*	32.75 ± 1.22	15.75 ± 0.57	0.088 ± 0.003	0.103 ± 0.004	113.12 ± 4.07	0.25 ± 0.009
D	*Cymbopogon citratus*	31.48 ± 0.82	16.10 ± 0.38	0.085 ± 0.002	0.091 ± 0.002	118.73 ± 2.72	0.26 ± 0.006
E	*Anacardium occidentale*	32.88 ± 1.34	15.78 ± 0.62	0.089 ± 0.004	0.102 ± 0.004	113.39 ± 4.41	0.25 ± 0.010
F	*Persea americana*	23.6 ± 0.89	11.52 ± 0.43	0.064 ± 0.002	0.078 ± 0.003	82.90 ± 3.08	0.18 ± 0.007
	Minimum	23.6 ± 0.89	11.52 ± 0.43	0.064 ± 0.002	0.078 ± 0.003	82.90 ± 3.08	0.18 ± 0.007
	Maximum	41.42 ± 1.44	19.93 ± 0.67	0.112 ± 0.004	0.127 ± 0.004	143.46 ± 4.78	0.32 ± 0.011
	Mean	32.29 ± 1.12	15.75 ± 0.52	0.087 ± 0.003	0.100 ± 0.003	113.81 ± 3.75	0.250 ± 0.008

Table 5: Radiological hazard indices in medicinal plant samples

Sample Code	Scientific Name	E_{CED} (mSv/yr)	E_{CED} (μSv/yr)	ELCR ($\times 10^{-3}$)
A	*Magnifera indica*	0.00452 ± 0.00018	4.52 ± 0.176	0.01233 ± 0.00048
B	*Dacryodes edulis*	0.00326 ± 0.00009	3.26 ± 0.089	0.00889 ± 0.00024
C	*Terminalia catappa*	0.00350 ± 0.00015	3.50 ± 0.153	0.00955 ± 0.00024
D	*Cymbopogon citratus*	0.00297 ± 0.00010	2.97 ± 0.104	0.00810 ± 0.00028
E	*Anacardium occidentale*	0.00356 ± 0.00017	3.56 ± 0.173	0.00970 ± 0.00047
F	*Persea americana*	0.00229 ± 0.00009	2.29 ± 0.085	0.00623 ± 0.00023
	Minimum	0.00229 ± 0.00009	2.29 ± 0.085	0.00623 ± 0.00023
	Maximum	0.00452 ± 0.00018	4.52 ± 0.176	0.01233 ± 0.00048
	Mean	**0.00335 ± 0.00013**	**3.35 ± 0.130**	**0.00913 ± 0.00035**

The calculated average annual collective dose equivalent, E_{CED} for examined medicinal plants have been presented in Table 5. The values vareied from 2.29 ± 0.085 to 4.52 ± 0.176 μSv/yr, with 3.35 ± 0.130 μSv/yr as the mean. Compared to the comparable global value of 1 mSv/yr, the E_{CED} values are less significant. According to UNSCEAR (2000), the general populace receives an annual effective dose of NORMs from medicinal plants of E_{CED} = 0.3 mSv/yr. ^{232}Th contributes 54.91% to the total average annual collective equivalent dose E_{CED}, followed by ^{40}K with 38.75%, while ^{238}U adds 6.35% to have the least impact on the E_{CED}. In comparison to Alade *et al.* (2020), Njinga *et al.* (2015), and Tettey-larbi *et al.* (2013), our findings for E_{CED} are of lesser value. The consumers' health may not be affected by the yearly effective dosage of the therapeutic herbs in this investigation. The minimum, maximum, and mean values of the estimated ELCR for the medicinal plants are 0.00623 ± 0.00023, 0.01233 ± 0.00048 and 0.00913 ± 0.00035 respectively. Since the results are lower than the UNSCEAR (2000) global average of 0.29×10^{-3}, there is extremely little probability of acquiring stomach cancer when these plant samples are used as medications.

Overall, Sample D, *Cymbopogon citratus*, had the greatest value of all the medicinal plants under examination (^{40}K), whereas Sample A, *Magnifera indica*, had the highest value of all the hazard indices. This is so because *Magnifera indica*, contains the greatest concentration of ^{232}Th and ^{238}U out of all the samples, and ^{232}Th is what mostly contributes to the estimated radiological risk.

3.2 Correlation Analysis

Pearson's correlation analysis was conducted to ascertain the degree of association between the examined radiological parameters in the medicinal plants. The matrix of established correlation coefficient (*r*-values) is shown in Tables 6. Some of the highest correlations observed are discussed. The analysis's findings showed that ^{238}U exhibited significantly strong negative relationships with ^{40}K contents in the dry season (r = -0.75*, p= 0.0859). This result show that the decrease in ^{238}U values leads to an increase in the concentration of ^{40}K. The association between ^{40}K with (D, AGED and I_γ) is strong and not statistically significant, according to the data.

The correlation matrix showed that ^{232}Th exhibited a very strong and significant positive correlation with H_{in} (r= 0.83**, p= 0.0410), with H_{in} (0.92**, p= 0.0095), with E_{CED} (r= 0.92**, p= 0.0087), ELCR (r= 0.92**, p= 0.0087) in the medicinal plants. This could be as a result of the fact that ^{232}Th makes up the majority of the annual effective dose. The E_{CED} perfectly connected to the internal exposure H_{in} and ELCR. Additionally, E_{CED} has a very strong and near perfect positive and significant relationship with Ra_{eq} (r= 0.98***, p= 0.0006), H_{ex} (r= 0.98***, p= 0.0006), AGED (r= 0.92**, p= 0.0088), and I_γ (r= 0.93**, p= 0.0071). Like the association of E_{CED} with the other radiological parameter, a similar trend was observed for ELCR. This implies that there is a direct relationship between the E_{CED} and ELCR.

Table 6: Correlation coefficient matrix for all radiological variables

	^{40}K	^{238}U	^{232}Th	Ra_{eq} (Bq/kg)	D (nGy/h)	H_{ex}	H_{in}	AGED mSv/y	Gamma Index I_γ	E_{CED}	ELCR
^{40}K	1										
^{238}U	-0.75*	1									
^{232}Th	-0.06	0.66	1								
Ra_{eq} (Bq/kg)	0.50	0.17	0.83**	1							
D (nGy/h)	0.62	0.03	0.75*	0.99***	1						
H_{ex}	0.50	0.17	0.83**	1.00***	0.99***	1					
H_{in}	0.32	0.37	0.92**	0.98***	0.94**	0.98***	1				
AGED mSv/y	0.67	-0.03	0.70	0.98***	1.00***	0.98***	0.92**	1			
Gamma Index, I_γ	0.65	0.00	0.72	0.98***	1.00***	0.98***	0.93**	1.00***	1		
E_{CED}	0.33	0.34	0.92**	0.98***	0.94**	0.98***	1.00***	0.92**	0.93**	1	
ELCR	0.33	0.34	0.92**	0.98***	0.95**	0.98***	1.00***	0.92**	0.93**	1.00***	1

r-values significant *p<0.1; **p<0.05; ***p<0.001

CONCLUSION

Natural radionuclide activity concentrations of six (6) different samples of medicinal plants used in the manufacture of herbal products were examined using the gamma spectrometric method. 40K, 238U, and 232Th were detected in all samples. The average concentrations of 209.43 ± 5.14 Bq/kg, 4.73 ± 0.15 Bq/kg, and 8.00 ± 0.40 Bq/kg for ^{40}K, ^{238}U and ^{232}Th, respectively are below the global recommended limit set by UNSCEAR (2000) and all predicted radiological hazards fall within the boundaries that have been established globally. The relationship between ^{232}Th and (E_{CED} and ELCR) was shown to be extremely strong, positive, and significant. This indicates that the ^{232}Th radionuclide has a significant role in both the increased lifetime cancer risk brought on by ingesting the studied herbal plant and the average annual collective effective dose, E_{CED}. Because of this, there is extremely little risk to customers' health and very little natural radiation exposure associated with ingesting these therapeutic plants. This study can therefore be used as baseline values for radioactivity of medicinal plants in Esan Land, Edo State, Nigeria and may help develop standards and guidelines for medicinal plants as well as radiation protection rules and regulations.

Acknowledgement

The authors would like to appreciate the Management of Pax Herbal Clinic and laboratories for the assistance in the processing and preparation of plant materials used in this study.

Competing interest

The authors hereby declare that the study presented in this article is original and that no any conflicting interest exist between us as regard to the study.

Authors' Contribution Statement

Felix Adegoke Popoola conceptualized and proposed the project, created the study strategy, laboratory analysis, and preliminary drafting. **Oladotun Bamiji Isola** was responsible for the manuscript's validation, conclusion, text editing, and critical revision. **Tunde Ayobami Owolabi** collected and prepared field samples, assisted with article composition, and edited the manuscript. **Oluseye Daniel Fakeye** participated in developing the study plan and interpretation of data. **Isaac Oluwafemi Elijah and Sheu Isiaq Owoyemi** conducted the search for relevant literature and contributed to data acquisition. **Modupe Eunice Sanyaolu** performed the statistical analysis and contributed to the article drafting. The article was revised and approved for submission by all authors.

Availability of data and materials

The datasets and other relevant materials developed during the study can be made accessible by the corresponding author upon reasonable request.

Funding

No funding was obtained for this study.

REFERENCES

Abojassim, A. A.; Lawi, D. J., Alpha particles emissions in some samples of medical drugs (capsule) derived from medical plants in Iraq. *Plant Archives*, **2018**, *18* (1), 1137–1143.

Akhter, P., Rahman, K., Orfi, S.D. & Ahmad, N (2007). Radiological impact of dietary intakes of naturally occurring radionuclides on Pakistani adults. *Food Chem. Toxicol*. 45(2), 272-277. https://doi.org/10.1016/j.fct.2006.08.006.

Alade, A. A., O, I. C., & Adekunle, T. A. (2020). Natural radioactivity levels of some herbal plants with antimalaria potency in Ibadan South-West Local Government Area of Oyo State , Nigeria. *IOSR Journal of Applied Chemistry*, *13*(1), 1–7. https://doi.org/10.9790/5736-1301010107

Avwiri G.O, Ononugbo C.P, & Nwokeoji I.E. (2014). Radiation Hazard Indices and Excess Lifetime Cancer Risk in Soil, Sediment and Water Around Mini-Okoro/Oginigba Creek, Port

Harcourt, Rivers State, Nigeria. *Comprehensive Journal of Environment and Earth Sciences*, *3*(1), 38–50. http://www.knowledgebasepublishers.org/ENV SCI.html

Biira, S., Ochom, P., & Oryema, B. (2021). Evaluation of radionuclide concentrations and average annual committed effective dose due to medicinal plants and soils commonly consumed by pregnant women in Osukuru, Tororo (Uganda). *Journal of Environmental Radioactivity*. 227, 106460. http://doi.org/10.1016/j.jenvrad.2020.106460

Cengiz, G. B., & Çağlar, İ. (2019). Estimation of natural radioactivity of some medicinal or herbal plants used in Kars , Turkey. *Acta Pharmaceutica Sciencia*, *57*(4), 133–139. https://doi.org/10.23893/1307-2080.APS.05729

Chandrashekara, K., & Somashekarappa, H. M. (2016). Estimation of radionuclides concentration and average annual committed effective dose due to ingestion for some selected medicinal plants of. *Journal of Radiation Research and Applied Sciences*, *9*(1), 68–77. https://doi.org/10.1016/j.jrras.2015.09.005

Hashim, A. K.; Hamza, A.; Pradesh, H.; Kumari, P.; Kumar, G.; Risk, C.; Mohammed, R. S., Effective radium activity, radon exhalation rate and uranium concentrations in medicinal plants. *IOP Conf. Series: J. of Phy: Conf. Series* **2019**, *1234*, 012002. https://doi.org/10.1088/1742-6596/1234/1/012002

Jwanbot, D. I., Izam, M. M., Nyam, G. G., & John, H. N. (2013). Radionuclides analysis of some soils and food crops in Barkin Ladi LGA, Plateau State, Nigeria. *Journal of Environment and Earth Science*, *3*(3), 79–87.

Kareem, A. A., Hady, H. N., & Abojassim, A. A. (2016). Measurement of natural radioactivity in selected samples of medical plants in Iraq. *International Journal of Physical Sciences*, *11*(14), 178–182. https://doi.org/10.5897/IJPS2016.4507

Kolapo, A. A.; Omoboyede, J. O., Health risk assessment of natural radionuclide and heavy metals in commonly consumed medicinal plants in South-West Nigeria. *Ife J. of Sci.* **2018**, *20* (3), 529–537.

Monica, S., Jojo, P. J., & Khandaker, M. U. (2020).Radionuclide concentrations in medicinal florae and committed effective dose through Ayurvedic medicines. *International journal of Radiation Biology,* 96(8), 1028-1037. http://doi.org/10.1080/09553002.2020.1767816

Nahar, A., Asaduzzaman, K., Islam, M.M, Rahman, M.M. & Begum, M (2018). Assessment of natural radioactivity in rice and theirassociated population dose estimation. *Radiat. Eff. Defect. S,* 173(11–12), 1105-1114. https://doi.org/10.1080/10420150.2018.1542696.

Njinga, R. L., Jonah, S. A., & Gomina, M. (2015). Preliminary investigation of naturally occurring radionuclides in some traditional medicinal plants used in Nigeria. *Journal of Radiation Research and Applied Sciences*, *8*(2), 208–215. https://doi.org/10.1016/j.jrras.2015.01.001

Ofomola, O. M., Ugbede, F. O., & Ochuko, A. (2023). Environmental risk assessment of background radiation, natural radioactivity and toxic elements in rocks and soils of Nkalagu quarry, Southeastern Nigeria. *Journal of Hazardous Materials Advances*, *10*, 100288. https://doi.org/10.1016/j.hazadv.2023.100288

Omonhinmin, G. (2017, February 26). Ewu: Chieftaincy tussle that will not go away. *The Guardian*. https://www.guardian.ng/features/ewu-chieftaincy-tussle-that-will-not-go-away/

Oni, O. M., Isola, G. A., & Sowole, O. (2011). Natural activity concentrations and assessment of radiological dose equivalents in medicinal plants around oil and gas facilities in Ughelli and environs , Nigeria. *Environment and Natural Resources Research*, *1*(1), 201–206. https://doi.org/10.5539/enrr.v1n1p201

Ononugbo, C. P., Avwiri, G. O., & Agbalagba, E. O. (2017). Radioactive pollution and excess

lifetime cancer risk due to gamma exposure of soil and ground water around open landfills in River State, Nigeria. *Canadian Journal of Pure and Applied Sciences*, *11*(1), 4121–4130.

Orosun, M. M.; Alabi, A. B.; Olawepo, A. O.; Orosun, R. O.; Lawal, T. O.; Ige, S. O., Radiological Safety of Water from Hadejia River. *IOP Conf. Series: Earth and Environ. Sci.* **2018**, *173* (1). https://doi.org/10.1088/1755-1315/173/1/012036

Owolabi, T. A.; Ezenwa, K. C.; Olayioye, E. Y.; Iyorhibe, O. C.; Amodu, E.; Aferuan, O. F., Adaptogenic (Anti-Stress) Effect of Aqueous Musanga cecropioides (Urticaceae). *Int. J. Curr. Microbiol. App. Sci*. **2019**, *8* (10), 2558-2565.

Popoola, F. A., Fakeye, O. D., Basiru, Q. B., Adesina, D. A., & Sulola, M. A. (2019). Assessment of aadionuclide concentration in surface soil and human health risk associated with exposure in two higher institutions of Esan land, Edo State, Nigeria. *Journal of Applied Sciences and Environmental Management*, *23*(12), 2279–2284. https://doi.org/10.4314/jasem.v23i12.29

Saenboonruang, K., Phonchanthuek, E., & Prasandee, K. (2018). Determination using gamma spectroscopy of aatural radionuclide activity concentrations and annual committed effective doses in selected Thai medicinal plants. *Chiang Mai J. Sci. 2018;*, *45*(2), 821–831.

Saudi, H. A., Abedelkader, H. T., Issa, S. A. M., Diab, H. M., Alharshan, G. A., & Zakaly, H. M. H. (2022). An in-depth examination of the natural radiation and radioactive dangers associated with regularly used medicinal herbs. *Int. J. Environ. Res. Public Health*, *19*, 8124. https://doi.org/https:// doi.org/10.3390/ijerph19138124

Sowole, O., & Olaniyi, O. E. (2018). Assessment of radioactivity concentrations and effective of radionuclides in selected fruits from major markets at Ijebu – Ode in Ogun State, Southwest of Nigeria. *J. Appl. Sci. Environ. Manage.*, *22*(1), 95–98.

Sultana, S., Ferdous, J., Haque, M. M. (2020). Natural radioactivity and hazards assessment in medicinal plants in Bangladesh, Journal of Health Science, 10(1), 20-27. http://doi.org/10.5923/j.health.20201001.03

Sussa, F. V.; Damatto, S. R.; Alencar, M. M.; Mazzilli, B. P.; Silva,P. S. C., Natural radioactivity determination in samples of Peperomia pellucida commonly used as a medicinal herb. *J. of Environ. Rad.* **2013***,* 116, 148-151, DOI: 10.1016/j.jenvrad.2012.09.012

Tchokossa, P., Olomo, J. B., Balogun, F. A., & Adesanmi, C. A. (2013). Assessment of radioactivity contents of food in the oil and gas producing areas in Delta State , Nigeria. *International Journal of Science and Technology*, *3*(4), 245–250.

Tettey-larbi, L., Darko, E. O., Schandorf, C., & Appiah, A. A. (2013). Natural radioactivity levels of some medicinal plants commonly used in Ghana. *SpringerPlus*, *2*(157), 1–9.

Ugbede, F. O. (2020). Distribution of 40 K , 238 U and 232 Th and associated radiological risks in river sand sediments across Enugu East, Nigeria. *Environmental Nanotechnology, Monitoring & Management*, *14*, 100317. https://doi.org/10.1016/j.enmm.2020.100317

Ugbede, F. O., Akpolile, A. F., Oladele, B. B., Agbajor, G. K., & Popoola, F. A. (2022). Ingestion exposure and committed health risk of natural radioactivity and toxic metals in local rice sold in Enugu urban markets. *International Journal of Environmental Analytical Chemistry*, 1–21. https://doi.org/10.1080/03067319.2022.2036983

UNSCEAR United Nations. Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000. In Sources and Effects of Ionizing Radiation: Sources (Vol. 1); United Nations Publications: New York, NY, USA, 2000.

World Health Organization (WHO) (2007). WHO guidelines for assessing quality of herbal medicines with reference to contaminants and residues. *World Health Organization*, 1–17.

World Health Organization (WHO) (2015). WHO statistical profile. Country Statistics and Global Health Estimates by WHO and UN Partners. Global Health Observatory, World Health Organisation.