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Radionuclide Analysis of Some Locally Consumed Herbal Plants Commonly Used in Katsina State, Nigeria

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ABSTRACT

The need to investigate the presence of radionuclides in herbs is paramount as they have direct link to the soil which is the main source of terrestrial radionuclides and heavy metals. Ten (10) medicinal plants commonly sold and consumed in Katsina Urban, were studied using thallium activated sodium iodide detector gamma spectrometric set up to measure the natural radionuclides present in the samples. Potassium-40 (^{40}K), Uranium-238 (^{238}U), and Thorium-232 (^{232}Th) were measured in all the samples. The activity concentration of K-40 ranged between 64.57 - 777.06 *Bq/kg*, U-238 ranged between 175.88 *Bq/kg* - 369.10 *Bq/kg*, while Th-232 ranged between 0.20 - 9.52 *Bq/kg*. The absorbed dose ranged between 108.02 *nGy/h* – 198.88 *nGy/h* while the annual effective dose due to the radionuclide's intake ranged between 0.13 μSv/y – 0.24 *μSv/y*. The annual effective dose for K-40, U-238 and Th-232 was found to be lower than the limit set by United Nation Scientific Committee on Energy and Atomic Research of 0.48 *mSv/y*. The radiological hazard indices calculated ranged between 1.09 - 2.14 and 0.62- 1.5 for the internal and external hazard indices respectively. The results of our findings indicates that there is serious radiological risk in consuming these plants for medicinal purposes.

Keywords:
Measurement,
Radionuclides,
Herbal plants,
Radioactivity.

INTRODUCTION

Man has used plants as his source of medicine for various ailments over the years, long before the arrival of conventional medicine Njinga et al. (2015). Due to the fact that the use of such plants has produced immense results, they are now used in the production of modern medicines which is used in places like the hospitals Okunola et al. (2020). Organic constituents in plants such as vitamins, oil, glycosides and essential oil gives rise to the therapeutic efficacy of medicinal plants Desideri et al. (2009), Njinga et al. (2015). The high cost of procuring allopathic medicines contributes largely to the populace preferring the local remedies compared to the conventional medicines Njinga et al. (2015). People hardly consider the side effects these plants may have.

Considering the fact that radionuclides are found everywhere and in almost anything around us Joseph et al. (2018), it is expected that they will also be found in the plants used for medicinal purposes. These radionuclides, if taken into the body in unsafe quantities can cause irreversible damage to vital organs of the body like the lungs and liver Njinga et al. (2015) hence, the need for thorough investigation on the detailed constituents of Plants used as herbal remedies.

Radioactive elements which are unstable, and inorganic elements, all find their way into the plants during the process of photosynthesis (Adewumi, 2006). The absorption of these elements by plants through their roots, during nutrient intake, enables the plants to transport the absorbed nutrients to other parts of the plants using their phloem. The concentration of the radionuclides intake largely depends on the activity concentration in IAEA, (2006).

Excess Uranium suggests danger to cancer of the blood, that is, leukemia Baba-Kutigi et al. (2016). This is because the electron donor usually relaxes finally at the bone marrows Cherry et al. (2012). It also results in renal failure, refractory anemia, and liver dysfunction Pavlakis et al. (1996). The same danger applies to an individual with high intake of Thorium, an alpha particle emitter, which also deposits itself in the lining of the bone which can lead to bone cancer Cherry et al (2012). It is known to also induce liver cancer and fibrosis several decades after ingestion Manjoor et al. (2013). Excess of potassium however can lead to temporary or long-term health issues in kidneys,

hyperkalemia, diabetes and burns. It also results in stroke in women with hypertension Arjun et al. (2014).
This study was therefore based on the measurement of the radioactivity levels of ten (10) selected herbal plants commonly sold and consumed in selected locations of Katsina State, Nigeria using the thallium activated sodium iodide detector gamma spectrometric set up, at National Institute of Radiation protection And Research, Ibadan. The study was particularly focused on the activity concentration of the naturally occurring radionuclides ^{40}K, ^{238}Th, and ^{232}U which were embedded in the plants that were selected. The absorbed and annual effective doses were also calculated to ascertain the level of ingestion.

MATERIALS AND METHOD

Sample preparation and measurement

Ten (10) different herbal remedies were bought at an herbalist outlet at Latitude N12^{0}59'27.47" and Longitude E7^{0}36'14.81" of Katsina metropolis. The unregistered samples were kept in large basins while some were primitively sealed in polythene bags. The gamma ray spectrometry method was adopted using a Canberra co-axial sodium iodide doped with thallium, with active diameter 8cm. The names and curative properties of the herbal plants used is represented in table1.
The herbal plants were obtained from a shop, already pulverized. They were weighed using a triple beam balance, not less than 300g for each plant sample. After weighing, they were sealed, labelled and transported to the laboratory for analysis.

Specifications and Calibration of the Measuring System

The detector used for the radionuclide's measurements was 8cm Canberra co-axial NaI detector dopped with thallium and a pinnacle 900H spectrometer with serial number PHC18070501. It is located at the National Institute of Radiation Protection and Research, University of Ibadan campus, Ibadan. The efficiency calibration of the detector was determined by using a 650g mixed CANBERRA soil standard containing ^{125}Sb, ^{155}Eu, ^{54}Mn, ^{65}Zn and ^{40}K in Marinelli beaker in the energy range of 35.5 keV to 1460.8 keV. The standard reference sample and the experimental samples' containers were geometrically identical. The detection absolute efficiency was determined for each of the gamma energies under consideration by using the equation below (National Institute of Radiation Protection and Research, Ibadan):

$$\varepsilon = \frac{NC_i}{A_i \times y_i \times M \times T} \quad (1)$$

where, ε is the efficiency of the NaI at the energy of the i^{th} radionuclide, NC_i is the net counts of the i^{th} radionuclide (background subtracted) in the corresponding photo peak, A_i is the activity concentration of the i^{th} radionuclide in Bq/kg, y_i is the emission probability of the i^{th} radionuclide, M is the mass of the plant sample in kg, T is the counting time (18000 seconds).
The Thallium-activated NaI detector responds to the gamma-ray by producing a flash of light or a scintillation. The scintillation occurs when scintillator electrons, excited by the energy of the photon return to their ground state. Sodium iodide activated with a trace amount of thallium converts about 15% of fast particle energy into light, a figure that is the highest among commonly used scintillation materials. The high atomic number of the iodine components ensures that large crystals of sodium iodide will fully absorb the energy of a large fraction of all incident gamma rays. The detection efficiency for gamma rays in large scintillators is generally much larger than for germanium detector (Gbadebo, 2011).

Radiological Analysis of Samples

Each sealed sample was placed on the NaI(TI) detector crystal and counted for 18,000s. The samples containers have the same geometry material as that of the IAEA reference sample. THE IAEA-375 PLANT REFERENCE MATERIAL was used. An empty container of the same geometry and dimension was counted for the same counting time of 18,000s to determine the background distribution spectrum.
The choice of radionuclides to be detected was predicted on the fact that the NaI(TI) detector used for this study has a modest energy resolution. Hence, the photons emitted by them would only be sufficiently discriminated if their emission probability and their energy were high enough, and the surrounding background continuum is low enough. Thus, the activity concentration of ^{214}Bi (determined from its 1120 keV and 609 keV γ-ray peaks) was chosen to provide an estimate of ^{238}U in the samples, while that of the daughter radionuclide ^{228}Ac determined from its 911 keV γ-ray peak was chosen as an indicator of ^{232}Th. ^{40}K was determined by measuring the 1460 keV γ-rays emitted during its decay. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks.

Table 1: Names and Curative Properties of the Herbal Plants Analyzed

English Name	Local Name (Hausa)	Botanical Name	Curative Property
Natural Pomegranate	Ruman	*Punica Granatum*	It is used as an antioxidant,anti-viral and ani-tumour properties. It is also a good source of vitamins especially vitamins A, E and C as well as folic acid.
Natural Moringa	Zogale	*Moringa Oliefera*	It is a powerful anti- inflammatory agent.
Natural Garlic	Taparnuwa	*Allium Sativum*	This is commonly used for conditions related to the heart and blood system which includes high blood pressure, high levels of cholesterol and hardening of the arteries (atherosclerosis)
Natural Olive	Zaitun	*Olea europaea*	It is used for prevention or treatment of inflammation, infections for example, common cold, influenza, candidiasis, urinary tract infections, diarrhoea
Natural Fig	Baure	*Ficus Carica*	This is used as a laxative to relieve constipation, it is also used for treatment of diabetes, skin conditions such as eczema, psoriasis e.t.c
Black Seed	Habbatu Sauda	*Nigella Sativa*	This is used for the treatment of digestive tract conditions including gas, colic, diarrhoea, constipation, dysentery e.t.c.
Baobab	Kuka	*Adansonia*	This is used for the treatment of fever, diarrhoea, measles, small pox and as a pain killer.
Ginger	Chitta	*Zingiber Officinale*	Cold, nausea, arthritis, migraines, and hypertension.
Fenugreek	Hulba	*Trigonella Foenum-graecum*	It solves digestive problems like loss of appetite, stomach upset, constipation, painful menstruation and obesity.
Anise	Yansu	*Pimpinella Anisum*	It helps soothe intestinal gas, runny nose e.t.c. it is also an appetite stimulant for stomach upset.

Source: (https://mysasun.com/blogs/bloglearning-bytes/30-common-nigerian-spices-and-their-uses)

Activity Concentration

The activity concentration in the plant samples was calculated using the following:

$$(Bq/kg) = kC_n \quad (2)$$

where, $k = 1/\varepsilon P_\gamma M_s$, C is the activity concentration of the radionuclide in the sample given in (Bq/kg), C_n is the count rate under the corresponding peak, ε is the detector efficiency at the specific γ-ray energy, P_γ is the absolute transition probability of the specific γ-ray, and M_s is the mass of the sample given in (kg) (Jibrin & Esen, 2007).

Absorbed Dose

The total absorbed dose rate in air was calculated using the dose conversion factors (0.462, 0.604 and 0.0417 for ^{238}U, ^{232}Th and ^{40}K, respectively) as given in Equation 2 UNSCEAR (2000a).

$$(n\,Gy/h) = (0.462C_U + 0.604C_{Th} + 0.0417C_K) \quad (3)$$

where, C_U, C_{Th} and C_K are the activity concentration (*Bq/kg*) of uranium, thorium and potassium, respectively in the samples and (nGy/h) is the absorbed dose.

Annual effective dose

The annual effective dose equivalent to the population due to the activity concentrations of the radionuclides in the plant was determined using the outdoor annual effective dose only, which depends on conversion coefficient from absorbed dose to effective dose as 0.7 Sv/Gy and outdoor occupancy factor of 0.2 as stated by (UNSCEAR 2000a). The annual effective dose equivalent in units of *mSv/y* was calculated using the equation below:

$$(msv/y) = (n\,Gy/h) \text{ x } 24 \text{ x } 365 \text{ x } 0.2 \text{ x } 0.7 \text{ x } 10^{-6} \quad (4)$$

Internal hazard index (Hin)

Internal hazard index gives the value of internal radiation which is the radiation that occurs when humans consume something that emits radiation. The radiation results in radiological hazards (Purnama & Damayanti, 2020).The internal exposure was calculated using the equation below:

$$H_{in} = \frac{A_u}{370} + \frac{A_{Th}}{258} + \frac{A_K}{4810} \quad (5)$$

The value of this index should be less than one (1) in order for the radiation hazard to have negligible effect to the respiratory organs of the public (Bretka & Mathew, 1995). Higher internal hazard indices indicates that continuous exposure could lead to some level of health risk (Nwankwo et al., 2015).

External hazard index (Hex)

The external hazard index or indoor radiation hazard index which is denoted by (Hex), is the hazard due to external exposure to radiation. The main objective of the hazard index is to ascertain if the value is ≤ 1 (ICRP 2004). The external hazard index was calculated using the equation below:

$$H_{ex} = C_{Ra}/370 + C_{Th}/370 + C_K/4810 \leq 1 \quad (6)$$

RESULTS AND DISCUSSION

The activity concentration of potassium-40 (^{40}K) in all the plant samples ranges between 64.57 - 777.06 Bq/kg. Anise was found to contain the lowest concentration of Potassium with 64.57 Bq/kg, which is in agreement with the result obtained by Okunola et al. (2020). Natural fig contained the highest concentration of potassium with 777.06 Bq/kg, which agrees with the work done by Nura, (2015) and Adeleke et al. (2021) but disagrees with that of Ademola (2021). The high level of potassium in ginger could be attributed to the ubiquitous nature of potassium. Potassium constitutes about 2.1–2.3% of the earth's crust and thus is the seventh or eighth most abundant element (Joseph & Nasiru, 2013). Hence, it's soil reserves are generally large fertilizers in intensive agriculture (Schroeder, 1978). Potassium is very essential in the body and deficiency of it can cause health problems in man (Deborah, 2019). It is found in foods and vegetables that man consume and therefore expected to also be found in herbal remedies ingested by man Joseph et al. (2016). Excess of potassium however can lead to temporary or long term health issues in kidneys, hyperkalemia, diabetes and burns. Arjun et al. (2014) documented that excess potassium results in stroke in women with hypertension.

Ginger exhibited the highest concentration of Uranium with a value of 369.10 Bq/kg, while the baobab was the lowest in Uranium with 175.88 Bq/kg. Uranium may be transported to vegetation by air or by water. It can be deposited on the plants themselves by direct deposition or re suspension, or it can adhere to the outer membrane of the plant's root system with potential limited absorption. The plants, aquatic or terrestrial, may be eaten directly by humans or consumed by land or aquatic animals, which provide food for humans ATSDR, (2013). Excessive ingestion of Uranium results in renal failure, refractory anaemia, rhabdomyolysis, myocarditis, and liver dysfunction Paylakis *et al.* (1996). This assertion further agrees with the review made by Elena et al (2010) that uranium can be toxic to many bodily systems like the normal functioning of the kidney, brain, liver, and the heart. The high values of uranium obtained in the plant samples used in this research work is therefore a great concern.

Thorium ranged between 0.20 - 9.52 Bq/kg, with moringa having the lowest concentration of 0.20 Bq/kg, contrary to the report of Joel et al. (2019). Olive had the lowest concentration of 9.52 Bq/kg. More than 50% of total injected thorotrast is found in liver and it is known to induce liver cancer and fibrosis several decades after injection Manjoor et al. (2013). Human intoxication with ^{232}Th occurs by oral, dermal, wound or respiratory routes. Whatever the means of internal contamination, ^{232}Th is absorbed in the blood and deposited in the target organs (Kumar et al). Thorium is termed as a radionuclide with particularly high level of radio toxicity (Poliakova, 2017) and is a primary radioactive hazard in humans Melissa et al. (2001).

The health effects of radiation exposures to natural occurring radioactive materials from consumption of herbal plants or herbal preparations (Joseph et al., 2017) may be linked to most forms of cancer of the blood and of many organs such as the bone, lung, breast and thyroid as time goes on (Ademola *et al* 2018). The activity concentration of ^{238}U, ^{232}Th and ^{40}K for each plant type has been summarized in Table 2 and represented in figure 1.

Table 2: Activity Concentration in the Medicinal Plant Samples

Sample Names	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	Absorbed Dose (nGy/h)	Annual Effective Dose (μSv/y)	Internal Hazard Index (H_{in})	External Hazard Index (H_{ex})
Punica granatum	621.63	211.37	4.67	126.39	0.20	1.29	0.71
Moringa Oliefera	710.70	340.32	0.20	186.99	0.23	1.99	1.07
Allium Sativum	640.24	280.77	7.85	161.15	0.20	1.68	0.92
Olea europaea	546.52	368.71	9.52	198.88	0.24	2.14	1.5
Fiscus carica	777.06	299.30	3.7	172.91	0.21	1.79	0.98
Nigella Sativa	481.26	221.22	6.07	125.94	0.15	1.31	0.71
Adansonia	511.15	175.88	9.02	108.02	0.13	1.09	0.62
Zingiber Officinale	523.13	369.10	5.91	195.90	0.20	2.12	1.12
Trigonella foenum	565.95	194.41	6.19	117.16	0.23	1.19	0.67
Pimpinella anasum	64.57	272.09	6.27	132.19	0.20	1.5	0.76

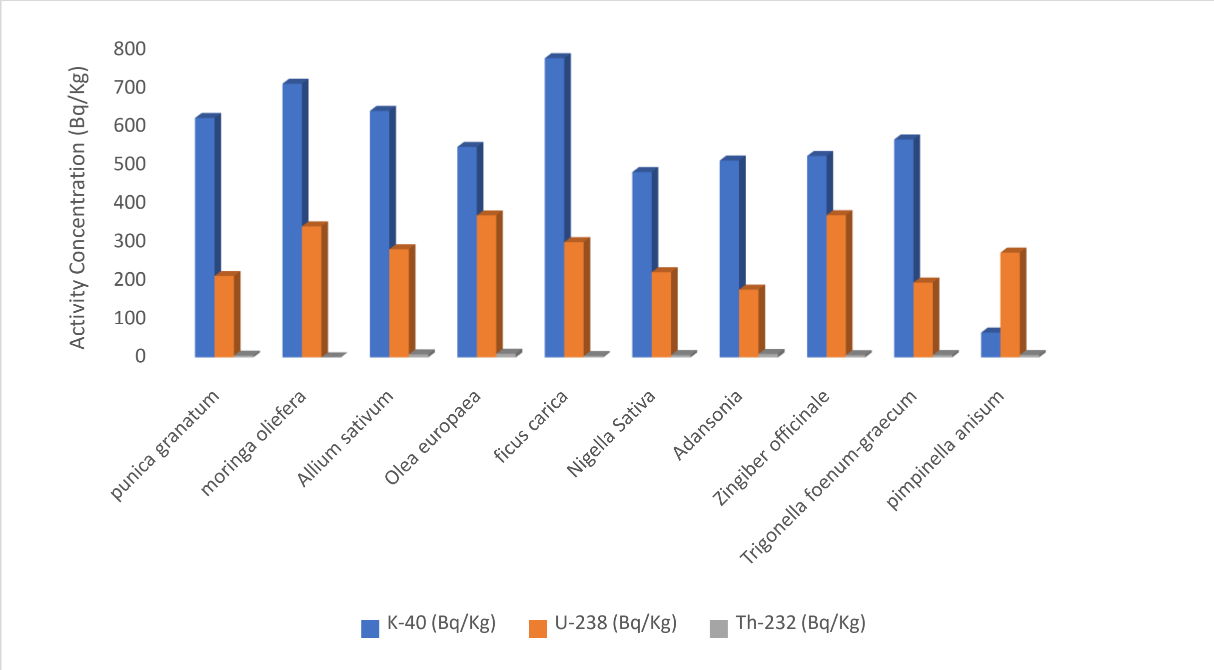

Figure 1: Activity concentration of ^{40}K, ^{238}U and ^{232}Th for each plant type.

Assessment of Radiological Hazards

The radiological hazards determined in this research work are the absorbed dose, the annual effective dose and the external hazard indices. The results obtained, are shown below.

Absorbed Dose

The absorbed dose was determined from the plant samples using Equation 2. The results ranged from 108.02 - 198.88 nGy/h. *Adansonia* was found to have the lowest value of 108.02 nGy/h while *Olea europaea* had the highest value of 198.88 nGy/h. The Absorbed dose rate according to the plant types are presented in Table 2.

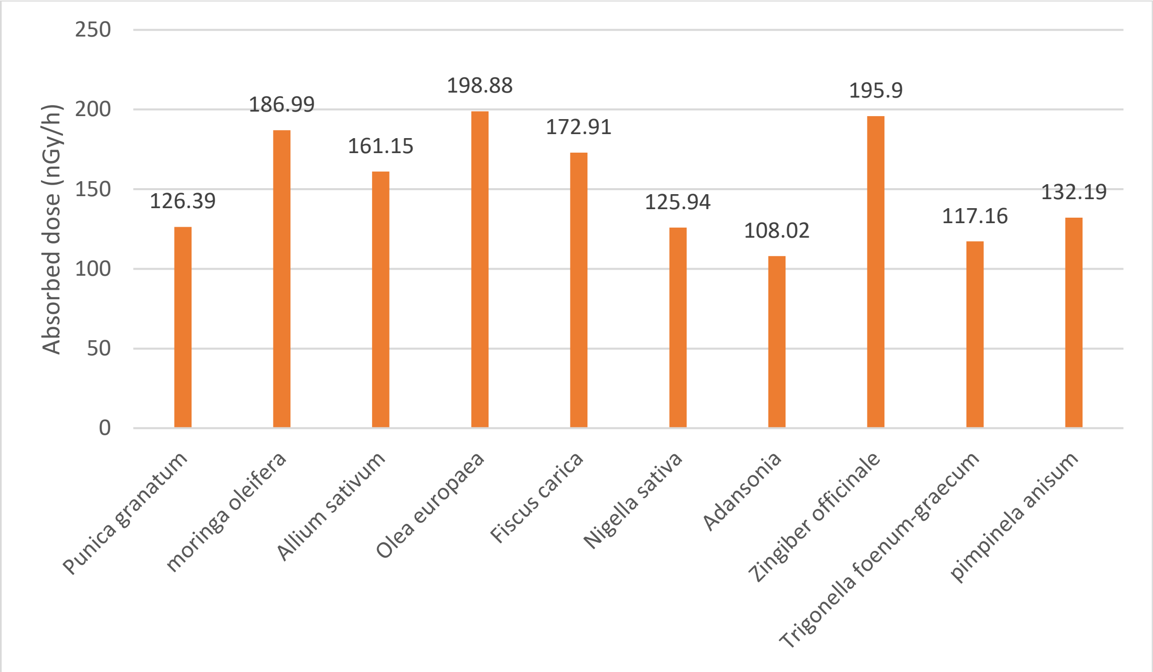

Figure 2: Range of values for absorbed dose for the plant types.

Annual Effective Dose

The values of the absorbed dose were used to evaluate the annual effective dose for all the samples. Equation 4 was used and the result is presented in table 4. The values ranged from 0.13 - 0.24 µSv/y. *Olea europaea* and *zingiber officinale* had the highest values of 0.24 µSv/y for the annual effective dose and *adansonia* had the lowest value of 0.13 µSv/y.

Internal and External Hazard Indices

The internal and external hazard indices was calculated using equation (4 and 5) respectively**.** The values of the internal hazard ranged between 1.0 9 and 2.14 with *Adansonia* being the lowest and *Olea europaea* being the highest. The external hazard indices ranged between 0.62 and 1.5 in which *olea europaea* also peaked the highest, while *adansonia* was lowest. The result shows that most of the values exceeded the recommended threshold of (≤1) (ICRP 2004), which indicates a high risk of damaging the internal organs of humans. Summary of the results are presented in the table 2.

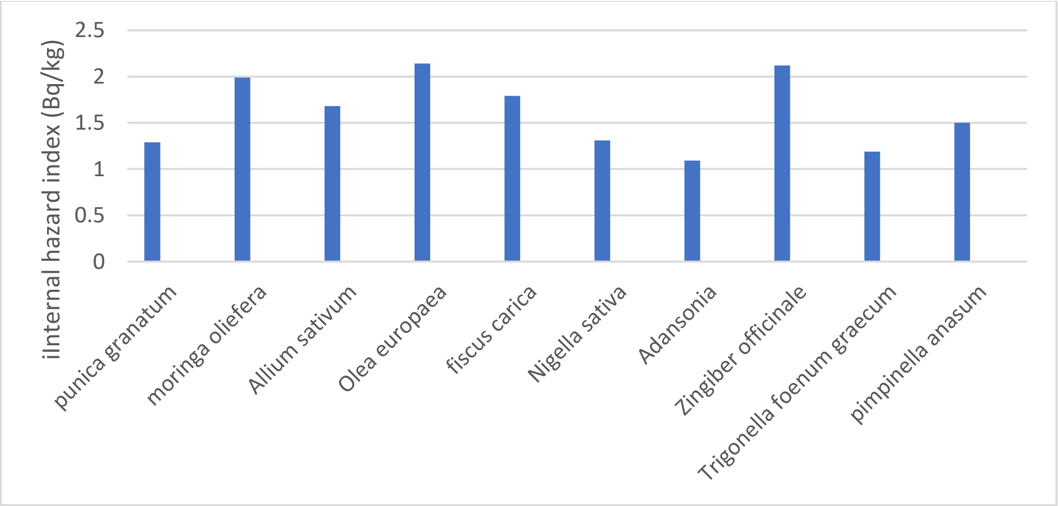

Figure 3: Range of internal hazard index for each plant type

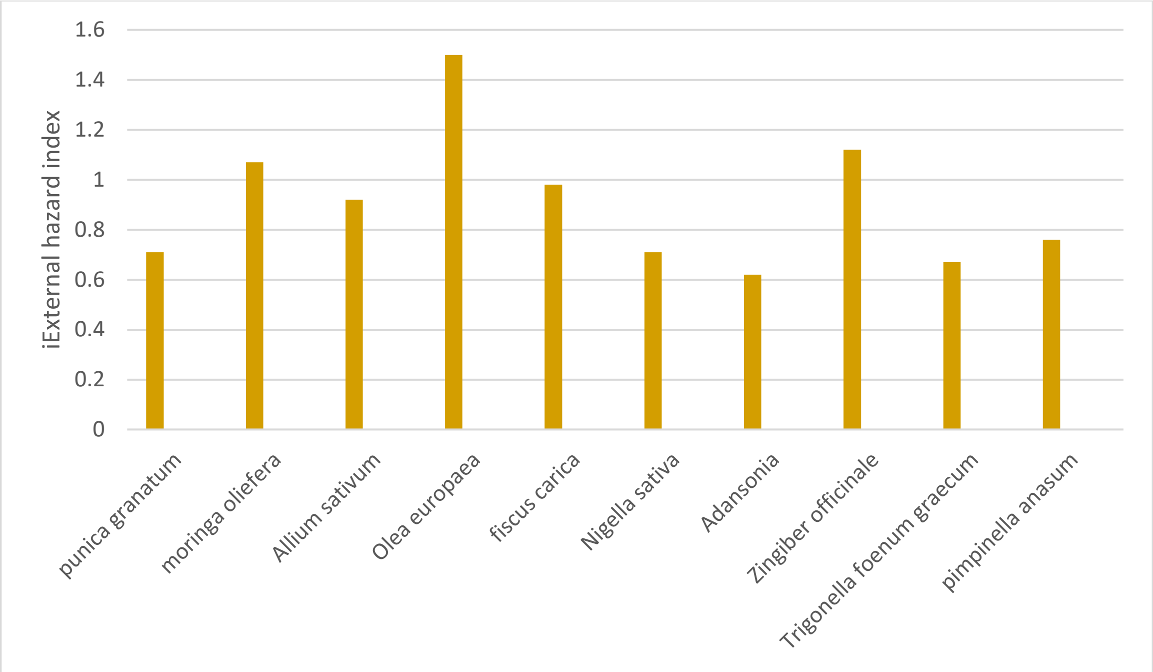

Figure 4: Range of External hazard index for each plant type

CONCLUSION

The radioactivity concentration of the radionuclides potassium, uranium and thorium in some medicinal plants in Urban Katsina was determined with NaI detector using the gamma-ray spectroscopy. Potassium was found to have the highest concentration of 777.06 Bq/kg in *fiscus carica* while it was lowest in *pimpinella anasum* with a value of 64.57 Bq/kg. Uranium peaked in *zingiber officinale* (369.10 Bq/kg) and was lowest in *adansonia* (175.88 Bq/kg). Thorium on the other hand was highest in *olea europaea* (9.52 Bq/kg) and lowest in *moringa oliefera* (0.20 Bq/kg). The absorbed and annual effective dose due to the activities in the samples were also estimated and olive had the highest absorbed dose of 198.88 nGy/h, while *adansonia* had the lowest value of 108.012 nGy/h. The annual effective dose was also calculated and *olea europaea* and *zingiber officinale* had the same highest values of 0.24 μSv/y while *adansonia* had the lowest value of 0.13 μSv/y. Consequently, the internal and external hazard indexes were calculated and the values ranged between 1.09 - 2.14 Bq/kg and 0.62 – 1.5 Bq/kg respectively. The hazard indexes were higher than the recommended permissible limit of ≤1 which suggests health risk if such plants are ingested. It can be concluded that some herbal plants sold in herbal outlets in Katsina urban are not safe for consumption.

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