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Assessment of Natural Radioactive Nuclides in Different Herbal Remedies Consumed in Katsina Metropolis

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Abstract
The use of herbal remedies to treat various ailments is common practice in developing countries, but the formulation of these products is usually complex, hence, the need for thorough quality checks especially with possible health risks these could pose. The research aimed to assess the natural radioactive nuclides concentrations of herbal formulations sold in Katsina Metropolis and ascertain the radiological implications for humans. Table 4.2 shows the activity concentration of natural radionuclides in the fifteen (15) herbal remedies commonly consumed in the Katsina metropolis. Results from the above table show the activity of natural radionuclides in 15 herbal remedy samples found in the Katsina metropolis. The activity concentration of ^{40}K ranges from 1027.76±51.30, which was found to have the highest radioactivity in the G, and 51.77±2.60 was found to be lowest in K. In ^{238}U which ranges from 72.78±7.67 in O and the lowest concentration of 1.65±0.23 in E then lastly followed by the activity concentration of ^{232}Th activity concentration varies from 20.18±1.13 as the highest and the lowest in B with 1.26±-0.07.

Keywords: Natural, Radionuclides, Activity, Herbal Remedies, Nigeria

Introduction

The sources of natural radioactivity in the environment are natural radionuclides (^{238}U, ^{232}Th, and ^{40}K) and their decay products, which originated from the crust of the earth (Kessaratikoon and Awaekechi, 2008). Naturally occurring radioactive materials (NORMs) are omnipresent in the environment, including air, water, soil, food, and humans. For over two decades, the World Health Organization (WHO) has been advocating the use of traditional medicine worldwide by promoting the integration of its valuable elements into national healthcare systems (WHO, 2009). Medicinal plants are administered either in their raw form or in formulations such as solutions, tablets, or capsules (Ademola *et al.,* 2015). Medicinal flora are botanical entities that are employed to alleviate various ailments and are regarded as being replete with constituents that can be exploited in the development and synthesis of pharmaceuticals. As posited by the World Health Organization, approximately 80% of individuals residing in outlying communities rely solely on curative herbs for the management of illnesses (Sahito *et al*., 2003).

If plants are employed in the treatment of particular maladies, they have the potential to be harmful if not administered under the guidance of a medical professional (Aleksandra *et al*., 2015). It is therefore crucial to regulate the quantities of heavy metals and radionuclides present in medicinal plants to guarantee the safety of their products. Consequently, it is imperative to undertake a comprehensive inquiry into some of the most commonly utilized medicinal plants in Nigeria to ascertain the levels of natural radionuclides and concentrations of heavy metals in these plants. This undertaking is also crucial in the formulation of policies, rules, and regulations on the processing and production of medicinal products in Nigeria.

Herbal remedies have been an integral part of traditional medicine practices for centuries, with a rich history of use in many cultures around the world. These natural products are often perceived by the general public as safe and effective alternatives to conventional pharmaceutical drugs. However, there is growing concern about the potential presence of natural radioactive nuclides in some herbal remedies, which could pose significant health risks to consumers. The Katsina metropolis, located in the northern region of Nigeria, is home to a thriving herbal medicine industry. This region is known for its diverse array of traditional herbal remedies, which are widely used by the local population to treat a variety of ailments, ranging from common illnesses to chronic conditions. These herbal products are typically sourced from various natural sources, such as plants, minerals, and animal-derived materials, which may contain naturally occurring radioactive materials (NORM). The presence of NORM, including isotopes of uranium, thorium, and their decay products, in herbal remedies can be a significant health concern. Exposure to these radioactive nuclides can lead to increased risks of radiation-induced health effects, such as cancer, genetic mutations, and other adverse outcomes. Therefore, it is crucial to assess the levels of natural radioactive nuclides present in the herbal remedies consumed within the Katsina metropolis to ensure the radiological safety of these traditional medicines.

This study aims to investigate the concentrations of NORM in different herbal remedies sold and consumed within the Katsina metropolis. The findings of this research will provide valuable insights into the radiological safety of these traditional medicines, which can inform public health policies, consumer awareness efforts, and the development of appropriate regulatory frameworks in the region.

MATERIAL AND METHODS

Material

Description of Study Area

The study design was a cross-sectional survey that aimed to assess the levels of natural radionuclides in herbal remedies consumed in Katsina, Nigeria which lies 12^0 18 - 13^0 16 N latitude and 7^0 35' 30 – 7^0 40' 11 E longitude. Samples of each kind were homogenized to give ten (10) composite samples of the herbal products. All the herbal products collected have neither been filed nor registered with NAFDAC and were locally packed in black polythene bags.

Chemicals and Equipment

The materials used in this study were obtained from Federal University Dutsin-ma. These are materials include but not limited to weighing balance, polythene bags, and sulphuric acid.

Methods

Sample Collection

Herbal medicinal plants were collect from local vendors in Katsina metropolis in August 2022. These samples were collected in polythene bags and kept at room temperature for further processing.

Sample Preparation and Measurement

The samples procured in powder form were prepared utilizing the procedure developed by Njinga et al. (2015). Subsequently, these samples were tightly packed and hermetically sealed in polythene bags and kept undisturbed for approximately twenty-eight days, to allow for Ra-226 and Ra-228, along with their progenies, to attain secular radioactive

equilibrium (Lordford et al., 2013). The analysis of these samples was conducted at the Centre for Energy Research and Training (CERT), located in Zaria-Nigeria, utilizing the same gamma-ray spectrometric setup employed by Njinga et al. (2015). The measurements of radioactivity concentration were carried out using gamma-ray spectrometry. The detector assembly utilized for this experiment was composed of a NaI (Tl) detector, measuring 7.62 x 7.62 cm, housed in a lead shield, with a thickness of 6 cm, and a cadmium-lined assembly with copper sheets, which served the purpose of reducing background radiation. The entire assembly was coupled with a computer-based Multichannel Analyzer (MCA) card system, MAESTRO programmed specifically for data acquisition and spectra analysis. The quantitative determination of K-40, Ra-226, and Th-232 in the herbal samples was carried out through calibration of the analyzer with the reference materials supplied by the International Atomic Energy Agency, as shown in Table 2. The absolute detection efficiency of the NaI (Tl) detector was determined through the use of standard sources Okunola *et al* (2020). Finally, the activity concentration of radionuclides in the herbal samples was calculated using the methodology developed by Njinga et al. (2015).

Human Health Effect

The levels of natural radionuclides were compared with the recommended limits set by international organizations to assess the potential radiological risk associated with the consumption of herbal remedies (Njinga *et al.,* 2015).

Average Annual Committed Effective Dose (AACED)

The computation of the Average Annual Committed Effective Dose (AACED) due to the consumption of Naturally Occurring Radioactive Materials (NORMs) within the realm of herbal remedies was executed through the utilization of the expression formulated by Lordford et al. (2013), as it is distinctly delineated in equation 2.

$$E_{ave}=\sum Cf\ x DCF_{ing\ x} A_i \ \ldots\ldots\ldots\ldots (1)$$

where

E_{ave} = Average annual committed effective dose,

C_f = Consumption rate of intake of NORMs from the herbal remedies,

DCF_{ing} = Dose conversion coefficient for ingestion of each radionuclide

According to UNCEAR (2000): 2.8 x10-4 mSv/Bq, 2.3 x10-4 mSv/Bq, and 6.2 x 10-6 mSv/Bq for ^{226}Ra, ^{232}Th, and ^{40}K respectively for an adult.

Ai = Specific activity concentration of each radionuclide

There is currently no standardized dosing protocol in place for the utilization of medicinal plants within Nigeria. Despite this, it is important to note that patients who regularly consume these plants for the treatment of common ailments may experience an increase in their average annual committed effective dose. By utilizing Equation (2) and reconfiguring it to make Cr the subject of the formula, it was possible to obtain the threshold consumption rate for each medicinal plant sample. This was accomplished through the use of the following relation.

$$Cr = \frac{3E_{ave}}{\sum_i^3 = 1\ [DCF_i x\ A_i]} \ldots\ldots\ldots\ldots (2)$$

the threshold average annual committed effective dose attributable to ingestion of naturally occurring radioactive materials (NORMs) in medicinal plants, as published by UNSCEAR (2000), is Eave ¼ 0.3 mSv/yr. The specific activity concentrations of ^{40}K, ^{238}U, and ^{232}Th in the samples of medicinal plants are denoted by A1, A2, and A3, respectively. The DCF_{ing} values for ^{40}K, ^{238}U, and ^{232}Th, which are 6.2 106 mSv/Bq, 2.8 104 mSv/Bq, and 2.3 104 mSv/Bq, respectively, are represented by DCF_1, DCF_2, and DCF_3.

a. Radon Equivalent Activity

Radium equivalent activity (Raeq) was determined according to equation 3.

$$Ra_{eq=}C_U + 1.43C_{Th} + 0.077C_K \ \leq 370 \ldots\ldots\ldots\ldots (3)$$

b. Internal Hazard Index (Hin)

The indices for internal hazard, denoted as H_{in}, and external hazard, denoted as H_{ex}, were computed by applying equations 4 and 5.

c. Annual Gonad Equivalent Dose (AGED)

Annual Gonad Equivalent Dose (AGED) (mSvyr-1), as outlined by UNSCEAR (2010), was calculated utilizing equation 6.

$$II_{in} - \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \ldots (4)$$

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \ldots (5)$$

$$AGED = 3.09C_U + 4.18C_{Th} + 0.314\,C_K \leq 300 \ldots\ldots\ldots$$

Where C_U, C_{Th} and C_K are activity concentration of U-238, Th-232 and K-40, respectively

Statistical Analysis of Data

Data acquired from the research work were analysed using analysis of variance (ANOVA) and SPSS.

Results

Plant Type and Parts Used for the Herbal Formulation

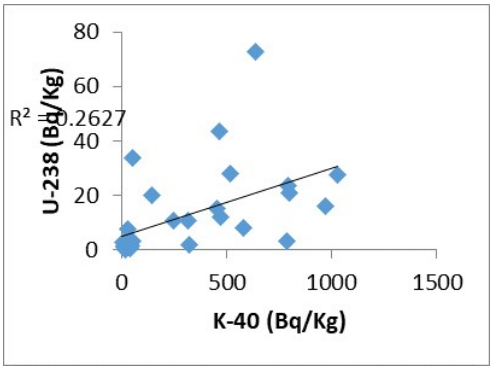

Fig. 1: Relationship between K-40 and U-238 Activity Concentrations of the Samples

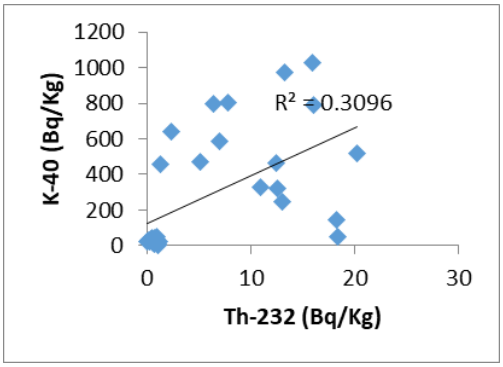

Fig. 2: Relationship between K-40 and Th-232 Activity Concentrations of the Samples

Table 1: Type and parts of medicinal plants used for herbal formulation in Katsina State

	Plant type	Botanical name	Part of Plant
A	Marke, Kirya	*Anogeissus leiocarpus, Prosopis Africana*	Leaves, bark and stem
B	Sabara, Marke	*Guiera senegalensis, Anogeissus leiocarpus*	Leaves, stem
C	Marke	*Anogeissus leiocarpus*	Bark
D	Kalgo, Sabara	*Pillostigma thonningii, Guiera senegalensis*	Leaves, stem
E	Aduwa, Magarya	*Balanites eagyptica, Ziziphus abyssinica*	Bark, stem
F	Magarya, Kirya	*Ziziphus abyssinica, Prosopis Africana*	Leaves
G	Hano, Sabara	*Boswellia odorata, Guiera senegalensis*	Leaves, stem
H	Kirya	*Prosopis Africana*	Leaves, bark
I	Marke, Aduwa	*Anogeissus leiocarpus, Balanites eagyptica*	Bark, stem
J	Kalgo, Sabara	*Pillostigma thonningii, Guiera senegalensis*	Leaves
K	Kirya, Sabara, Kalgo	*Prosopis Africana, Guiera senegalensis, Pillostigma thonningii*	Leaves, bark
L	Sabara, Marke	*Guiera senegalensis, Anogeissus leiocarpus*	Leaves, stem
M	Kirya, Hano, Sabara	*Prosopis Africana, Boswellia odorata, Guiera senegalensis*	Leaves and bark, stem
N	Aduwa, Magarya, Hano	*Balanites eagyptica, Ziziphus abyssinica, Boswellia odorata*	Bark and leaves
O	Aduwa, Sabara	*Balanites eagyptica, Guiera senegalensis*	Bark and leaves

Key: *A to O represent samples from 15 different location at Katsina metropolis*

Activity Concentration of Herbal Formulations

Table 2: Activity Concentration in Herbal Formations in Katsina Metropolis

SAMPLE	K-40 (Bq/kg)	U-238 (Bq/kg)	Th-232 (Bq/kg)
A	637.40±31.86	72.78±7.67	2.30±0.14
B	456.09±22.90	15.32±2.04	1.26±0.07
C	789.16±39.37	3.31±0.42	16.00±0.93
D	249.51±12.02	10.59±0.84	13.01±0.73
E	325.13±16.26	1.65±0.23	10.90±0.63
F	584.00±29.17	8.02±0.99	7.04±0.41
G	1027.76±51.30	27.34±3.18	15.97±0.92
H	793.97±39.78	23.75±2.75	6.42±0.37
I	464.59±23.36	43.42±4.92	12.45±0.72
J	472.12±22.73	12.15±0.96	5.10±0.28
K	51.77±2.60	33.58±3.72	18.36±1.07
L	319.42±16.05	10.57±1.30	12.61±0.73
M	142.77±7.20	20.00±2.61	18.23±1.05
N	970.45⊥46.71	15.93⊥1.25	13.31⊥0.74
O	515.82±24.82	27.77±2.19	20.18±1.13
Minimum			
Maximum			

Discussion

Table 2 shows the activity concentration of natural radionuclides in the fifteen (15) herbal remedies commonly consumed in the Katsina metropolis. Results from the above table show the activity of natural radionuclides in 15 herbal remedies samples found in Katsina metropolis. The activity concentration of ^{40}K ranges from 1027.76±51.30, was found to have the highest radioactivity in the G and 51.77±2.60 was found to be lowest in K. in ^{238}U which ranges from 72.78±7.67 in O and the lowest concentration of 1.65±0.23 in E then lastly followed by the activity concentration of ^{232}Th activity concentration varies from 20.18±1.13 as the highest and the lowest in B with 1.26±-0.07. Generally, from the results recorded, the activity concentration of 40K was observed to be the highest among other natural radionuclides (^{238}U and ^{232}Th) recorded in all the samples. The high activity concentration of ^{40}K compared to ^{238}U and ^{232}Th could be due to the high absorption of 40K by the sampled plants from the soil relative to other elements (Lordford et al., 2013). The implication is that the samples could aid in therapeutic treatment for high blood pressure in the consumer (HBPI, 2012). The synergistic characteristics between the NORMs, using linear regression. The r^2 values indicated in the figures revealed a positive relationship between the NORMs. The correlation between K-40 and U-238, K-40 and Th-232, and U-238 and Th-232 are 0.2627, 0.3096, and 0.1784, respectively, with the highest r2 value between K-40 and Th-232. The recent attention on the implications of uranium as a metallic substance, as opposed to its role as a radionuclide, has been intensified due to concerns regarding the potential health issues arising from the accumulation of uranium in the human body following exposure to dust containing depleted uranium (DU) or injuries caused by shrapnel (Steenkamp *et al.,* 2005). The activity of U-238 in this study ranges from 1.65±0.23 Bq/Kg to 72.78±7.67 Bq/Kg as opposed to that by Adeleye and Chetty (2023) where herbal remedies in South Africa were analyzed and discovered to contain U-238 in the range of 20 to 57 Bq/Kg. This is in the same range as the result reported by Lordford et al., (2013) for herbal plant samples in Ghana where the average Uranium concentration was 31.8±2.8 $Bq\ kg^{-1}$. Naturally occurring radionuclides in herbal remedies in Nigeria are a topic of concern due to their potential health implications. Studies have examined this issue, and the overall consensus is that while radionuclides are present in some medicinal plants, their levels are generally considered to be insignificant and unlikely to pose a significant health risk (Tettey-Lerbi *et al.,* 2013). Several studies, such as those by Kolapo (2018) and Saudi et al (2022), have explored the health effects of exposure to naturally occurring radioactive materials (*NORMs*) from medicinal plants and herbal preparations in Nigeria. These studies have found that the radiological hazard associated with the intake of natural radionuclides in medicinal plants is considered to be low and insignificant.

The present study's Annual Gonad Equivalent Dose (AGED), as displayed in Table 2, exhibited a range of 165.45 $mSvyr^{-1}$ (sample L) to 473.99 $mSvyr^{-1}$ (sample F), with an average of 278.469 $mSvyr^{-1}$. The samples A, A2, C, F, G, H, and N surpassed the recommended limit of 300 $mSvyr^{-1}$. This observation suggests that vulnerable cells such as the gonad, bone surface cells, and bone marrow may be susceptible to health risks for individuals utilizing herbal remedies for disease treatment (Okunola *et al*., 2020).

Considering the trend of increasing global interest in medicinal plants, and global health many herbs may seriously do grazing extremely (Craib, 1999). Fortunately, many of these plants were in various climates and tropical steppe areas of growth and have been used by humans. Given the trend of increasing global interest in medicinal plants and global health (Franz, 1993), this is very necessary to

properly use and protect them. Pasture utilization and conservation of medicinal plants should be done beside others. Then this principle should be used with other herbs in the meadows of the ability to apply to the use they should keep trying. And simply do not keep hay meadows with a view of the protection measures such as byproducts also they are important for many purposes. Knowledge and identification is the initial steps to using medicinal plants in any region. While you have no sufficient science about plants, don't use them. The efficacy of any plant as medicine can be determined through guessing. Despite the acknowledged importance of medicinal plants in Ezinihitte Mbaise Local Area, the application of medicinal plants to health problems is still generally unknown, poorly organized, and regulated while most are being exploited with little or no regard for the future (Emereonye, 2007). However utilization and consumption should be done based on ecological principles to ensure sustainability and conservation of the resources, according following tips: Non--destructive harvesting; Setting aside, reserve areas and cultivation of botanical gardens; Conservation and recovery of threatened medicinal plant species; Also because some medicinal plant may be overused and in undesired and destructive condition, this is necessary to identify the ecological needs of them and propagation. Therefore medicinal plant consumption should be spread in society, especially in local communities which has been destroyed, and harvesting methods must be introduced also in this area and similar communities.

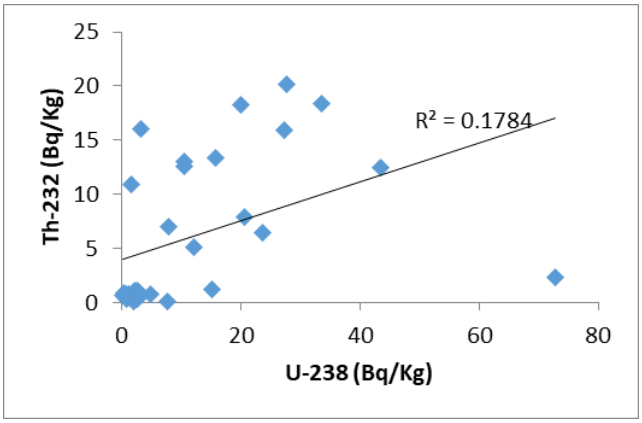

Fig. 3: Relationship between Th-232 and U-238 Activity Concentrations of the Samples

Conclusion

This research work provides baseline data for regulations and quality control of herbal remedies used in the Katsina metropolis. The activity of natural radionuclides in 15 herbal remedies samples found in Katsina metropolis. The activity concentration of ^{40}k ranges from 1027.76+51.30, which was found to have the highest radioactivity in the G, and 51.77+ 2.60 was found to be lowest in K. In ^{232}U which ranges from 72.78+7.67 in O and the lowest concentration of 1.65+0.23 in E then lastly followed by the activity concentration of ^{232}Th activity concentration varies from 20.18+1.13 as the highest and lowest in B with 1.26+0.07. The activity concentrations of naturally occurring radionuclides are higher than the average worldwide ranges in all leave samples. ^{40}K recorded significantly higher concentrations in all samples. Also, all radiological parameters are higher in leave samples than the recommended allowable limit. Zn and Pb have the highest concentrations in all six samples under investigation. Hence, caution must be taken into consideration for workers in these plant stores. Additionally, the data presented in this study will be used as a baseline to gauge how much radiation and toxic elements residents have been exposed to. The study revealed that there are no relations were found between

radon concentrations and toxic elements in some investigated samples.

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