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Uranium and Thorium contaminations of Baobab leave powder consumed in selected locations of Katsina State, North West Nigeria

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ABSTRACT
The need to know the level of contamination of radioactive elements, especially uranium and thorium, in the common foods we eat particularly the Baobab leave powder is highly significant due to their detrimental health effects. The presence of uranium and thorium in significant concentrations also presents an exposure risk to populations. In this study, six (6) baobab samples were collected from Daura, Funtua, Dustin-Ma, Kankia, Katsina, and Malumfashi in Katsina State. The samples were analysed for uranium and thorium content using Instrumental Neutron Activation Analysis (INAA). Concentration level of uranium was detected to be (0.068±0.011 mg/kg) in Daura and (0.021 mg/kg) in Dustin-Ma, while in Funtua, Kankiya, Katsina and Malumfashi, the concentrations were below detection limit. Thorium concentration was equally found to be (0.66±0.07 mg/kg) in Malumfashi, (0.54±0.07 mg/kg) in Daura, (0.43±0.06 mg/kg) in Dutsin-Ma, (0.25±0.04 mg/kg) in Kankia and (0.33±0.05 mg/kg) in Funtua. The concentration level of uranium and thorium accumulated in the baobab leave when analysed was found to be far below joint ATSDR/NIH safe limit of Uranium and Thorium intake in the human body. From this present work, we can see that the Baobab leave powder is safe for human consumption even though care should be taken to avoid contaminations.

Keywords:
Health risk,
INAA,
NIRR-1,
Concentration,
Consumption.

INTRODUCTION

Baobab tree (Adonsonia digitata), known as "*Kuka*" in Hausa language, "*Karkara*" in Kanuri language, and "*Bakko*" in Fulfulde (Fulani) language. In Nigeria and its neighbouring countries, the leaves are locally used to make soup especially in the northern part of Nigeria. The baobab tree leaves are stable source of food for rural population in many parts of Africa. In many West Africa countries, particularly in Nigeria, the young tender leaves are commonly consumed fresh as a substitute for commercial vegetables or used to prepare sauces or even as dried powder (Gebauer et al. 2002). Every part of the baobab tree is reported to be useful for mankind as well as livestock, used as food and medicinal purposes. African baobab (Adansonia digitata) which belong to the family Meliaceae, is a deciduous tree native arid central Africa and it is a largely spread across large area of sub – Saharan, semi-arid and sub-humid regions of African countries (Belewu and Ojo-Alokomaro, 2007; Bocsh and Sic Asafa, 2004). It is an important indigenous fruit tree species important for food security, nutrition, and income generation for the rural population in Africa (Van Boekel et al., 2009). Baobab seeds are roasted and eaten, or they are used as thickening agents when powdered, or as flavour enhancers when fermented. In Burkina Faso, the fermented Baobab seeds are commonly known as Maari, which are known to be rich in fatty acids and essential amino acids, and form part of the local diet (Tano-Debrah, and Diawara, 2015). Baobabs play an important role in providing a balanced nutrition because their edible parts supply vitamins, mineral, proteins, and energy that are not commonly obtained from the cereal- dominated diets of dry lands of Africa, most especially in Northern Nigeria. In Nigeria and Senegal, baobab fruits are reputed to be effective against microbial diseases. This has been confirmed in tests against certain bacteria and fungi, although the active constituents responsible for these effects have yet to be isolated (Hussain & Deeni, 1991).

The Baobab trees are generally stated to be found in countries of Southern Africa, such as Botswana, Mozambique, Namibia, South Africa and Zimbabwe (Palgrave et al., 2000) and other African countries including Nigeria. Baobab occurs in the dry lands of sub-Saharan Africa, and it is a representative of the wooden "Big Five," which also include Tamarindus indica, Zizyphus mauritiana, Sclerocarya birrea, and Mangifera indica (Mohamed et al., 2008).

The baobab is a multi-purpose tree as previously reported with products having numerous food uses and medicinal properties, and a fibrous bark that is used for various applications (Sidibe and Williams, 2002; Codjia et al., 2001; Wickens, 1982). The fact is that every part of the plant is useful - pulp of the fruit, the seeds, and the leaves are all well utilized and are very essentially wild gathered foods. The edible parts of baobab (leaves, seeds, and fruit pulp) are consumed mostly by rural communities who also sell them in local markets, whereas the non-food parts (timber, fodder, and fibers) are mainly used for income generation in sub-Saharan Africa (Gebauer et al., 2016). Moreover, the species has several roles in traditional medicine and cultural and religious beliefs, which often consider the tree as sacred (Kamatou., 2011), The naturally dry fruit pulp is rich in vitamin C. calcium, potassium, and dietary fiber (Baidu-Forson et al., 2012). The pulp is usually used in the preparation of fruit juice, snacks, sweets, as a fermenting agent in local brews (Gebauer et al., 2014), porridge, and in food recipes (Sidibé and Williams, 2002).

Consumption of food crops contaminated with heavy metals is a major food chain route for human exposure (Bocsh and Sic Asafa, 2004; Bustwat et al., 1997; Fischbein, 1992; Sharma et al., 2000). The distribution of heavy metals in plant body depends upon availability and concentration of heavy metals as well as particular plant species and its population (Ashiq et al., 2013; Akan et al., 2012; Awofolu, 2005; Bhata, 2002). Many researchers have shown that some common vegetables are capable of accumulating high levels of metals from the soil (Cobb et al., 2000; Huges et al., 1980). Certain species of these vegetables (e.g., cabbage) are hyper-accumulators of heavy metals into the edible tissues of plants (Wikipedia, 2008). Out of the one hundred and twelve (112) elements in nature, about eighty (80) are metals, most of which are found only in trace amounts in the biosphere and biological materials. Some metals or metal like elements which do give rise to well organize toxic effects in man and his ecological associates (Ashiq et al., 2013; Bunce, 1990; Csuros & Csuros, 2002). These elements include; arsenic, antimony, beryllium, cobalt, chromium, lead, manganese, nickel, etc. (Fischbein, 1992; Lide, 2006). In the recent years, an increasing number of reports appeared which indicated the rapid increase in the number of child cancer, genetically caused malbirths, repetitive abortions and adult cancer cases in Iraqi cities (Anonymous, 2017; Dahr, 2012; Anonymous, 2012). Thus, it was necessary to inspect the possible causes of these epidemic-like diseases, among which food contamination with radioactive material is suspected. This type of contamination was found to be an effective cause of health hazards according to the studies carried out by different centers of nuclear research (Sheppard et al., 1989; Apps et al., 1988; Ibrahim & Whicker, 1988).

Some researchers have studied heavy metal contaminations and health risk assessment of the baobab leaves in Nigeria (Mohammed et al., 2016; Ogbaga et al., 2017; Abdus-Salam & Adekola, 2018; Yaradua et al., 2019; Akintola et al., 2019; Rabiu et al., 2021; Mamman et al., 2021; Yaradua et al., 2023; Joseph et al., 2024), Ghana (Bempah et al., 2011; Agbemafle et al., 2012), Kenya (Stadlmayr et al., 2020), Malawi (Muthai et al., 2017; Kamanua et al., 2018), Zimbabwe (Gabaza, et al., 2018), among others. However, reviewed literatures have revealed a great variation in reported values of nutritional contents of baobab parts. Many reasons had been assigned to these variations (Chadare et al., 2009); he further recommended that, more attention should be given to accuracy and precision of the analytical methods employed for the analysis in order to streamline critical comparison and analysis. It is on this basis that Instrumental neutron Activation Analysis was employed for more accurate and independent (mineral analysis free from pre-chemical treatment or addition) elemental analysis of Baobab leave powder in order to inform proper regulation or proportionate usage or supplement recommendation. Despite some literatures reporting on the chemical, nutritional values, and health properties of baobab, there is no literatures on the analysis of uranium and thorium of Baobab leave powder in Nigeria and particularly in North Western Nigeria using Instrumental Neutron Activation Analysis (INAA), hence the need to embark on this study.

The determination of the composition and concentrations of uranium and thorium in the Baobab leave powder was performed using the Instrumental Neutron Activation Analysis (INAA). INAA is a very precise technique mainly used to determine trace concentrations of elements in samples and/or to acquire information on the spatial distribution of a neutron field via neutron activation detectors (Majerle, 2006; Joseph et al., 2019). This technique is based upon the conversion of stable nuclei to other, mostly radioactive nuclei via nuclear reactions, and measurement of the reaction products. The use of the INAA (relative) method for the calculation of the concentration of each element in the sample irradiated with reactor thermal neutron reduces the NAA equation to the simplest form (IAEA, 1990):

$$\frac{w}{w_{st}} = \frac{N_s D_{st}}{N_{st} D} = \frac{N_s e^{-\lambda td(st)}}{N_{st} e^{-\lambda td}} \qquad (1)$$

Where N_s= net photo peak area of radionuclide of interest in sample, N_{st} = net photo peak area of radionuclide of interest in standard, W= weight of element in sample irradiated, W_{st}= weight of the element in standard irradiated, $D = e^{-\lambda td}$ =decay factor for sample, $D_{st} = e^{-\lambda td(st)}$ =decay factor for standard, t_d= decay time for sample, $td(st)$= decay time for standard, λ= decay constant for radionuclide of interest

The concentration of the unknown element in the sample denoted by Cs is given by

$$C_s = \frac{W}{M} \quad (2)$$

where M= known weight of the irradiated sample containing the unknown weight of the element irradiated, W= unknown weight of the element irradiated.

MATERIALS AND METHODS

Sampling and Sample Preparation

Six Baobab leave powders were sampled at random from different locations during local market days of Dustin-ma, Kankia, Funtua, Malunfashi, Daura and Katsina all of Katsina State North Western Nigeria. Samples of about 1 kg each were collected in polythene bags that were previously cleaned by soaking in 1:1 HNO_3 for 3 days and washed with de-ionized water, then thoroughly mixed and transferred into clean and labelled plastic containers for analysis in the laboratory. The samples were prepared for irradiation without further treatment at the sample preparation laboratory, Centre for Energy Research and Training, CERT, Ahmadu Bello University, Zaria, alongside with the certified reference materials CRMs-NIST 1515 (apple leaves) supplied by U.S. National Institute of Standards and Technology for verification and quality control purposes for analysis by INAA. The samples were placed in a high-density polythene vial, and weighted using a Mettler Toledo balance model AE 240. These measured samples were double heat sealed in small pieces of cleaned polythene sheets using heat from a modern drier.

The irradiation facility used for this study is the Nigeria Research Reactor-1 (NIRR-1) at the Centre for Energy Research and Training, Ahmadu Bello University, (CERT, ABU), Zaria which is a Miniature Neutron Source Reactor (MNSR) facility, specifically designed for neutron activation analysis (NAA) and commissioned for operation in February 2004. The detailed description of the reactor and the irradiation facility as well as the theory, methodology, among other of INAA have been well documented (Jonah et al., 2004; Jonah et al., 2005 Jonah et al., 2006; Jonah et al., 2012; Adeleye et al., 2012; Njinga et al., 2012; Joseph et al., 2011; Joseph et al., 2013; Joseph et al., 2015a; Joseph et al., 2015b; Joseph et al., 2017; Joseph et al., 2019; Abubakar & Joseph, 2023). The Radioactivity Measurements Device is a gamma-ray data acquisition system which consists of a horizontal dip-stick High-Purity Germanium (HPGe) detector with a relative efficiency of 10 % at 1332.5 keV gamma-ray line, the MAESTRO emulation software compatible with the ADCAM® multi-channel analyzer (MCA) card, associated with electronic modules all made by EG & G ORTEC and a personal computer whose details has been described by Jonah *et al*. (2006) and later by Joseph & Nasiru (2013). The efficiency curves of this detector system at both near and far source-detector geometries have been determined by standard gamma-ray sources in the energy range of 59.5–2254 keV and were later extended to 4000 keV by a semi empirical method (Jonah and Sadiq, 2006). The gamma-ray spectrum analysis software ***WINSPAN 2004*** (Liyu, 2004), a software developed at CIAE, Beijing, China, was used for the peak identification, spectra analysis, and quantification of the elements present.

RESULTS AND DISCUSSION

The result of NIST 1515 (apple leaves) obtained which is to serve as the quality control in order to ensure the accuracy of the technique is presented in table 1. To assess the laboratory performance, we determined the U-score, Z-score and the relative bias (RB) of the NIST 1515 (apple leaves) which details had been discussed (Abubakar & Joseph, 2023).

The illustrated results in table 1 show that, all the element concentrations are in good agreement with the certified values. Also, the statistical parameters U-score and Relative bias calculated for all elements are acceptable only Z-score shows no satisfaction in concentrations of some elements.

It is therefore evident that there is a good agreement between the values obtained in our work with the certified values within the limit of experimental errors, hence we can conclude that our methodology can be applied in the evaluation of the elemental composition of biological sample such as the Baobab leave powder

Table 1: Quality control Data for NIST 1515 (apple leaves) (in mg/kg)

Elements mg/kg	NIST 1515 Certified Values	NIST 1515 Recorded Values	U-score	Z-score	R-bias
K	1.61±0.02	1.433±1.19	0.15	8.85	10.99
Al	286±9	300.3±17.3	0.73	1.59	5
Ba	49±2	53.9±7.3	0.65	2.45	9.6
Cl	579±23	573.21±23.9	0.18	0.25	-1
Mn	54±3	57.24±7.7	0.39	1.08	6
Br	1.8	1.49±1.2	0.26	-	-17.2
La	20	27.6±5.3	1.43	-	38
Sm	3	4.38±2.09	0.66	-	46

Values represent mean ± standard deviation.

Table 2: Concentration (mg/kg) of Uranium and Thorium of Six (6) Boabab Samples and

Location/ Elements	Daura	Dutsin-Ma	Funtua	Kankia	Katsina	Malumfashi
U	0.068 ± 0.011	0.021 ± 0.00	BDL	0.25 ± 0.04	BDL	BDL
Th	0.54 ± 0.07	0.43 ± 0.06	0.33 ± 0.05	0.25 ± 0.04	0.33 ± 0.05	0.66 ± 0.07

As seen in Table 2, the concentration of uranium (U) was found to be 0.068 ± 0.011 mg/kg in Daura, 0.021 ± 0.00 mg/kg in Dutsin-Ma, 0.25 ± 0.04 mg/kg in Kankia, but below detection limit (BDL) in Funtua, Katsina and Malumfashi. The average concentration value for U is 0.056 ± 0.009 mg/kg. Similarly, as indicated in Table 2, the concentration of thorium (Th) was found to be 0.54 ± 0.07 mg/kg in Daura, 0.43 ± 0.06 mg/kg in Dutsin-Ma, 0.33 ± 0.05 mg/kg in Funtua, 0.25 ± 0.04 mg/kg in Kankia, 0.33 ± 0.05 mg/kg in Katsina and 0.66 ± 0.07 in Malumfashi. The average concentration value for Th is 0.423 ± 0.06 mg/kg.

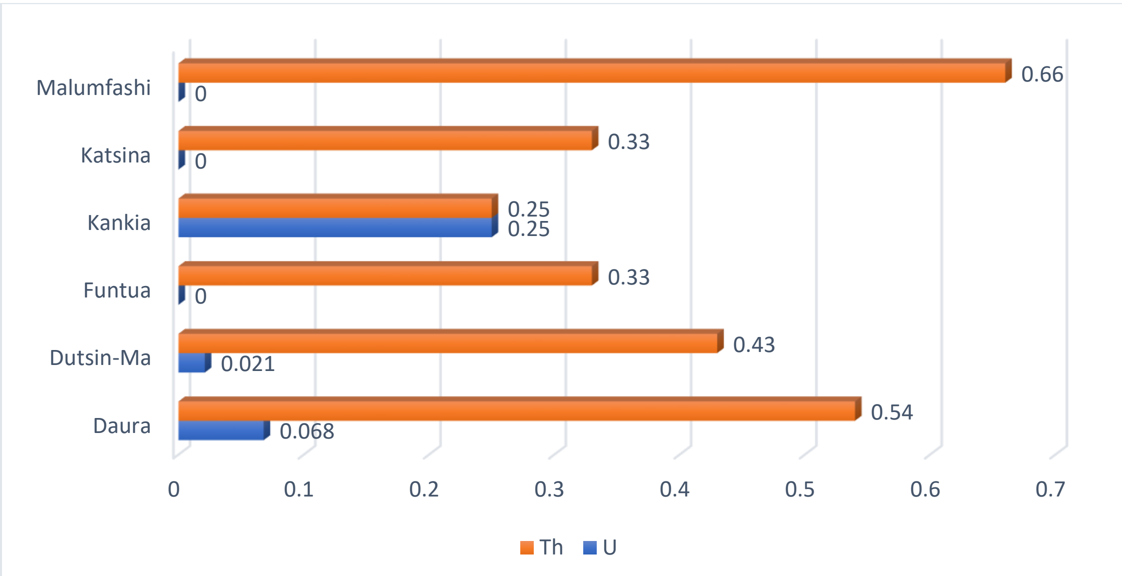

Figure 1: The concentrations of Th and U

In figure 1, we can see that the Kankia has the highest value of natural uranium followed by Daura with Dutsin-Ma been the least, while Malumfashi on the other side has the highest value of natural thorium followed by Daura, with Kankia been the least. The results from table 1 and figure 1 show that Th is more abundant in the Baobab leave powder in the study area than U.

The uranium mean value in this present study which is 0.056 ± 0.009 mg/kg is in agreement with that previously reported for Germany by Müller et al. (1997) and below the world value by (Kabata-Pendias and Pendias, 2001) which are greatly influenced by the granite weathering soil for vegetable forage and foods. Our result is also below the safety limit for the radioactive contamination of food and drinking water was defined as 1.63 mg/kg calculated as uranium element (UNSCEAR, 1994).

The most toxicologically important of the 22 currently recognized uranium isotopes are anthropogenic uranium-232 (^{232}U) and uranium-233 (^{233}U) and naturally occurring uranium-234 (^{234}U), uranium-235 (^{235}U), and uranium-238 (^{238}U). When an atom of any of these five isotopes decays, it emits an alpha particle (the nucleus of a helium atom) and transforms into a radioactive isotope of another element (NNDC, 2011). The process continues through a series of radionuclides until reaching a stable, non-radioactive isotope of lead (or bismuth in the case of ^{233}U). The radionuclides in these transformation series (such as isotopes of radium and radon), emit alpha or beta particles, as well as gamma and x-rays, with energies and intensities that are unique to the individual radionuclide. More so, there are three basic categories of uranium isotope mixtures (based on the mass percentage of ^{235}U relative to that of the earth's crust): natural uranium, enriched uranium, and depleted uranium. Natural uranium in the earth's crust is comprised of 99.2742% ^{238}U, 0.7204% ^{235}U, and 0.0054% ^{234}U by mass. Natural uranium in the environment can vary somewhat from these ratios due to physical and environmental factors, as shown by the varying ratios of natural uranium in air (EPA 2008).

Uranium's main target is the kidneys. Kidney damage has been seen in humans and animals after inhaling or ingesting uranium compounds. Ingesting water-soluble uranium compounds will result in kidney effects at lower

doses than following exposure to insoluble uranium compounds. Pinkerton et al. (2004) reported significantly increased mortality from nonmalignant respiratory disease (100 observed vs. 70.16 expected; SMR 1.43, 95% CI 1.16–1.73) within a cohort of 1,485 workers employed in uranium mills in the Colorado Plateau region (Arizona, Colorado, New Mexico) when worker mortality was compared to mortality within the U.S. population. Boice et al. (2008) examined mortality in a cohort of mining (1,867 males and females) and milling (759 males and females) workers at Grants, New Mexico, compared to mortality rates for the U.S. population. The pulmonary toxicity of uranium compounds varies in animals. Reports of pulmonary toxicity in animals after acute-duration exposure to uranium are limited to experiments with uranium hexafluoride. Gasping and severe irritation to the nasal passages were reported after 10-minute exposures at 637 mg U/mg^3 in rats and mice (Spiegl 1949) and nasal hemorrhage in rats after a 5-minute exposure to 54,503 mg/m^3 (Leach et al. 1984).
The average concentration value for Th is 0.423 ± 0.06 mg/kg in this current study agrees with Sathyapriya et al. (2011), as well as the values reported from Cochin, a coastal town in southern India where the natural background levels are low (Sathyapriya et al., 2010). Thorium is a naturally occurring radioactive metal found at trace levels in soil, rocks, water, plants and animals (EPA, 2023). Thorium is solid under normal conditions. There are natural and man-made forms of thorium, all of which are radioactive. In general, naturally occurring thorium exists as Th-232, Th-230 or Th-228. 232Th is a long lived naturally occurring radionuclides present in earth's crust and forms 100% of thorium present in the earth crust and three times more abundant than uranium. Once in human body thorium accumulates in lungs, liver, and skeleton. The principal site of deposition of thorium in the body is the skeleton (70%). In addition, thorium is deposited in the liver (4%), other soft tissue (16%), and the gonads (<1%). The remaining activity (10%) is considered to be promptly excreted in urine via the bladder (ICRP, 1995).

CONCLUSION

The study shows INAA can be used for estimation of uranium and thorium concentrations in Baobab leave powder samples effectively. The average values obtained for uranium and thorium are 0.056 ± 0.009 mg/kg and 0.423 ± 0.06 mg/kg respectively which are below the tolerable levels. These results shows that Baobab leave powder samples studied from the six local government areas in Katsina State, North Western Nigeria are safe for human consumptions. However, efforts should be made to reduce the exposure and contamination of Baobab leave powder to these naturally occurring elements.

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