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Investigation of Naturally Occurring Radioactivity Materials in some Animal parts commonly used in Traditional Medicine in Zaria, Nigeria

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Abstract
Animals and animal derivatives are one of the main components used in traditional medicines in managing health care in rural areas. These products are recognized, sold and have unique stores in the world including Nigeria. Animal products have been implicated to contain naturally occurring radioactive materials (NORMs) and major health concern in the safety of the crude drug as medicines. This study focuses on investigating some of these animal parts commonly used in traditional medicines and their naturally occurring radioactivity levels for safety. Twelve animal and animal parts samples were collected from two major well-known herbal markets in Zaria. The local names were identified, prepared according to the international standard of radioactivity methods and preparation specification using a NaI (Tl) detector (76×76 mm) and crystal optically coupled to a photomultiplier tube (PMT). The data were analysed with Maestro software by Canberra Nuclear Products. The naturally occurring radioactive materials concentrations and radiation health hazard indices were compared with the world average permissible limits. The results showed significant radioactive concentrations in all the twelve animal samples when compared with the world average standards and pose further risks of utilization as medicines. The study concludes possible cautions on the use of animals as natural medicines to avoid increasing carcinogenic potentials among users. We recommend further investigation of animal and animal-derived products used in Nigeria as medicines and possible standardization of the radioactivity concentration levels.

Keywords: Radioactivity; NORMs; Animals; Safety; Health; Nigeria

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Introduction

Traditional medicine practices have existed for many years (Elujoba *et al.,* 2005). These practices have been recognized as a holistic method in treatment, diagnosis and providing possible solutions to human illnesses (Abubakar *et al.,* 2007; WHO, 2011). Among developing countries, Africa traditional medical practitioners have offered cure to defy western medical treatments including spiritual attack, myths and other mysterious diseases and infections (Owumi and Jerome 2008). Patronage of traditional medicines including the use of animal and animal parts in the world is on the increased (Negi and Palyal 2007; Soewu, 2008; Mishra *et al.,* 2011; Yirga *et al.,* 2011). Studies have revealed that many tribes and cultures in the world with structured medicinal system continually utilizes animals and animals derived products as medicines during the process of healing human diseases (Alves and Rosa, 2005; Dedeke *et al*., 2006; Soewu, 2008). The World Health Organization estimated that about 9% of essential chemicals used in drug developments were from animal sources (Dedeke *et al.,* 2006).

The role of naturally occurring radioactive materials (NORMs) in animal and plant metabolism is well established (UNSCEAR, 2000); but their effect and influence on crops, medicinal plants, zoological materials and mineral substances used in traditional medicines had received relatively little attention. The International Food Safety Authority Network reports that naturally occurring radionuclides are present in the food we eat, the air we breathe, the water we drink and have posed serious health hazard among the general public (INFOSAN, 2011). These natural radionuclides are transferred and circulated through natural processes between various compartments entering the ecosystem and food chain through direct or indirect contamination of natural radionuclides (Avwiri and Agbalagba, 2007; Adewumi 2011; Avwiri *et al.,* 2011). It is expected that likewise may be found in animals or animal parts and products used in traditional medicines. The increase in the background ionizing radiation from numerous sources has various health side effects on the populace such as thorium and uranium which are present in the biotic system of plants, animals, soil, water and its ingestion could be harmful (Shanthi *et al.,* 2009). Periodically, radionuclides can enter the body by ingestion, inhalation or injection and once taken into the body, their radiation effects depend on their anatomic distribution, duration of retention in the body, rate of radioactive decay, as well as on the energies of the emitted radiation (Idoko *et al.,* 2016).

The United Nations Scientific Committee on the Effects and Risks of ionizing radiation (UNSCEAR) approximated that 15% of ^{210}Pb and ^{214}Pb ions, 99% of ^{226}Ra and ^{228}Ra, ^{214}Bi (bone seeker) and ^{210}Po (soluble) reach the blood or the lung fluid stream and they are distributed to the whole body (UNSCEAR, 2000). Therefore, this study focuses on investigating some naturally occurring radioactivity levels and their gross radiological health indices of some animal and animal parts used as medicine in Zaria, Kaduna-Nigeria.

Materials and Methods

Study Area

This study was carried out in Zaria, Kaduna State, Nigeria. The city is a metropolitan city which consists of Zaria and Sabon Gari Local Government Areas in the Northern, Nigeria. It is a famous urban with many western and qur'anic educational centres and traditionally known as *Zazzau*. Dahiru *et al.,* (2018) documented that Zaria city is one of the original seven Hausa city-states and its population was about 500,000 people and other dwellers (Jolijn, 2006; NPC, 2007). The study area comprises of two major herbal markets in Zaria, Kaduna State. The selected markets are specialized for the trading and selling of animal and animal products and well known for traditional medicinal materials trading including mineral materials known as "Kasuwan Armaru" (Zaria city) and "Sabon Gari" herbal markets. Salome *et al,* (2018) have also reported and described the market composition and their activities previously.

Collection and Identification

Twelve (12) animal parts samples were collected in Zaria market in Kaduna State, Nigeria. They were identified by local market sellers in Hausa names and also authenticated by an expert (a zoologist) in the Department of Zoology, Ahmadu Bello University Zaria. The cabinet number for the animal's parts were noted for documentation in the study.

Animal samples Preparation

The animal parts procured were collected, labelled, dried and pulverized into fine powder in aseptic conditions using Thomas-Wiley laboratory mill (Model- 4 Arthur H. Thomas Company Philadelphia, PA., USA). The samples were packed into radon-impermeable cylindrical plastic containers which were selected based on the space allocation of the detector vessel which measures 76 mm × 76 mm in dimension (geometry). To target the progenies of unstable isotopes radionuclides (^{40}K, ^{226}Ra and ^{232}Th), the samples were sealed and packed in containers to prevent radon-222 escaping (Idoko *et al.,* 2016). Briefly, the sealing process includes smearing of the inner rim of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container, and tight-sealing lid-container with masking adhesive tape. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days before gamma spectroscopy measurements. All the packaging in each case were replicated.

Evaluation of Naturally Occurring Radioactivity of Samples

The analyses were carried out using a 76 ×76 mm Sodium Iodide (Tl) detector crystal optically coupled to a photomultiplier tube in the Center for Energy Research, Ahmadu Bello University. In brief, the assembly has a pre-amplifier incorporated into it and a 1-kilovolt external source and the detector is enclosed in a 6 cm lead shield with cadmium and copper sheets. This arrangement is aimed at minimizing the effects of background and scattered radiation. The data acquisition software known as Maestro by Canberra Nuclear Products measured each sample for 29,000 seconds and were recorded (Idoko *et al.,* 2016). The peak area of each energy in the spectrum was used to compute the activity concentrations in each sample and the packaging in each sample was triple-sealed for statistical analysis (UNSCEAR 2000). The average determination of naturally occurring radioactive concentration was determined as in (Table 1).

Radiation Health Hazard Indices

These indices are useful in estimating the radiological effects of samples that contain naturally occurring radionuclides for uranium-238 (^{238}U), thorium-232 (^{232}Th) and potassium-40 (^{40}K) which takes into consideration the radiation hazard associated parameters. These indices and the corresponding equations are presented below.

i. Annual Gonadal Equivalent Dose AGED: UNSCEAR considered organs such as the gonads, bone marrow and bone surface cells as target organs since they are mainly sensitive parts of the human body to radiation. AGED has been known to cause bone marrow and destroys red blood cells which are often replaced by white blood cells (UNSCEAR, 1998). This situation results in blood cancer (leukaemia). AGED is calculated with a given activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/Kg) using the relation.

AGED (mSv⁄yr) = 3.09 CRa + 4.18 CTh + 0.314 CK……….…………………… (a)

Where, CRa, CTh, and CK are the radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/Kg) in animal parts and mineral samples respectively.

i. Activity Concentration Index (Representative Gamma Index Iγr):

This estimates the gamma radiation hazard that may be associated with the natural radionuclides in specific investigated samples. Turhan and co-worker reported that an increase in the representative gamma index greater than the universal standard may result in radiation risk leading to the deformation of human cells thereby causing cancer (Turhan *et al.,* 2009). Values of I**γ**r ≤ 1 corresponds to an annual effective dose of less than or equal to 1 mSv, while I**γ**r ≤ 0.5 corresponds to annual effective dose less or equal to 0.3 mSv (Higgy *et al.,* 2000).

The activity concentration index is given as:

$$I\gamma r = \frac{CRa}{150} + \frac{CTh}{100} + \frac{CK}{1500} \quad \text{………………} (b)$$

ii. Annual Effective Dose Equivalent (AEDE):

The annual effective dose equivalent received by a person outdoor is calculated from the absorbed dose rate by applying the dose conversion factor of 0.7 Sv/Gy. Taking into consideration that people on average, spent 20% of their time outdoors, occupancy factor for outdoor and indoor is 0.2 (5/24) and 0.8 (19/24) respectively (UNSCEAR, 2000; Veiga *et al.,* 2006). The AEDE (indoor) occurs within a house whereby the radiation risks due to building materials only are taken into consideration while AEDE (outdoor) involves a consideration of the absorbed dose emitted from radionuclides in the environment such as ^{226}Ra, ^{232}Th and ^{40}K. The standard AEDE (Outdoor) value is 70 µSvyr-1 and that for AEDE (Indoor) is 450 µSvyr-1. These indices measure the risk of stochastic and deterministic effects in irradiated individuals (Alias *et al.*, 2008).

AEDE (Outdoor) (µ Sv⁄y) = Absorbed dose D (nGy⁄h) × 8760h × 0.7 Sv⁄Gy × 0.2 × 10^{-3} …………………………………………..(c)

AEDE (Indoor) (µ Sv⁄y) = Absorbed dose D (nGy⁄h) × 8760h × 0.7 Sv⁄Gy × 0.8 × 10^{-3} ………………………………………….. (d)

iii. Excess Lifetime Cancer Risk (ELCR):

This is associated with the probability of developing cancer over a lifetime at a given exposure level. It is a value depicting the number of cancers expected in a given number of people on exposure to a carcinogen at a given dose. An increase in the ELCR causes a proportionate increase in the rate at which an individual can get breast, prostate or even blood cancers (Taskin *et al*., 2009).

ELCR (outdoor and indoor) = AEDE × DL × RF …………………… ……………….(e)

Where; AEDE is the Annual Effective Dose Equivalent (outdoor and indoor dose equivalent)

DL is the average duration of life/life expectancy (estimated as 70 years);

RF is the Risk Factor (Sv-1), i.e. fatal cancer risk per Sievert. For stochastic effects, the International Commission on Radiological Protection (ICRP) uses RF as 0.05 Sv-1 for the public with the ELCR UNSCEAR standard being 0.29 X10^{-3}. (Taskin *et al.*, 2009).

iv. External Hazard Index (Hex): Beretka and Matthew defined external hazard index (Hex) by the equation below,

$$Hex = \frac{CRa}{370} + \frac{CTh}{259} + \frac{CK}{4810} \quad \text{.......................... (f)}$$

Where, CRa, CTh and CK are the radioactivity concentration in Bq/kg of 232Th, and 40K. The value of this index must be less than unity for the radiation hazard to be insignificant. The maximum value of Hex equal to unity corresponds to the upper limit of Raeq (370 Bq/Kg) (Beretka and Matthew, 1985).

v. Internal Hazard Index (H_{in}):

The internal hazard index is given as H_{in} should be less than unity for the radiation hazard to be insignificant. Internal exposure to radon and its daughter products are very hazardous and can lead to respiratory diseases like asthma and cancer (Beretka and Matthew, 1985).

$$H_{in} = \frac{CRa}{370185} + \frac{CTh}{259} + \frac{CK}{4810} \quad \text{.................... (g)}$$

vi. Radium Equivalent Activity (Ra_{eq}): According to UNSCEAR, (2000); Radium Equivalent Activity (in Bq/Kg) is estimated using the equation given below:

Ra_{eq} = CRa + 1.43CTh + 0.077Ck …….. (h)

Ra_{eq} is a single parameter used to represent the radionuclide concentrations of Ra-226, Th-232 and K-40 taking into account their respective radiation hazards.

vii. Absorbed dose rate D (nGy/h)**:** The gamma-radiation absorbed dose rate in air DR (nGy h21) was calculated by using the conversion factor of 0.461 nGy h–1 per Bq kg–1 for 226 Ra and 0.623 nGyh–1 per Bq kg–1 for 232Th and 0.0414 nGyh–1 per Bq kg–1 for 40K (Shanthi *et al.*, 2010).

D (nGy/h)= 0.461CRa + 0.623CTh + 0.0414CK………………………………(i)

Where, CRa, CTh, and CK are the radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/Kg) in animal parts and mineral samples respectively.

Data Analysis

The data acquired were analysed with Maestro by Canberra Nuclear Products. The samples were measured for 29,000 seconds, for each sample. The peak area of each energy in the spectrum was used to compute the activity concentrations in each sample and the packaging in each sample was triple-sealed for triplicate and statistical analysis. The average determination of naturally occurring radioactive concentration was determined.

Results and Discussion

In this study, most animal and animal products used in traditional medicines in Zaria, Kaduna, Nigeria are mainly wild animals that could only be tamed and collected by hunters who have expatriate abilities. This result confirms the folkloric usages of wild animals as medicine and other secret belief of potency associated therein as medicines (Alves and Rosa, 2005).

The physical description of the method of processing showed that these animal and animal parts used as traditional medicines were dried, almost in a decomposed state, presences of whitish spores probably fungi growths, specks of dust, loads of contamination and possible infections during the process of storage and identification. Also, it was noticeable that animal dungs are also being used as traditional medicines (Figure 1) and this could lead to infections and unhealthy consumption of recipes as medicines. Although, further studies on the antimicrobial load of these animals and chemical profiles should be carried out to ascertain its safety utilization. Salome and co-worker documented some of the folkloric usages of these animal parts as protection from the nightmare, ability to tame wayward women, worn for shield and strength, used as an aphrodisiac, and tendencies to prevent promiscuity (Salome *et al.,* 2018).

The World Health Organization on crude drug evaluation and quality stated that no crude drug or medicines should contain faeces or other faecal quantity (WHO, 2011). It was noticed that the mode of application of the animal products was through an oral application and sometimes to be burnt into ashes to be inhaled or consumed this could suggest its therapeutic potential as a natural therapy.

The results from the naturally occurring radioactive materials (NORMs) evaluated from the animal parts showed the presence of ^{40}K, ^{226}Ra and ^{232}Th (Table 1). This confirms that radioactive materials could enter the animal systems through the food chain and concentrated in their bodies before being used as medicines to cure diseases (Avwiri*et al.,* 2011; INFOSAN, 2011). Wild animals are prone to roam around radioactive sites and contaminated environments since they are not restricted, which could increase their potential and high deposits of NORMs. The potassium-40 concentration levels in the animal samples were within the world permissible limits (412.00 Bq/kg) probably because potassium is an abundant element in the earth crust and widely utilized to control homeostatic balance in nature while the ^{226}Ra and ^{232}Th were significantly higher when compared with Radium (45.00Bq/kg) and thorium (32.00Bq/kg) permissible limits respectively (Shittu *et al*., 2015). Potassium did not constitute a threat because it is within the permissible limits and is also an essential element in human cells (Jwanbot *et al.,* 2012) but these findings observed the high concentrations levels of thorium-232 and radium-226 in the animal samples investigated. Cherry *et al., (*2012) reported that abundance of the radium and thorium in the animal parts and that the alpha particle emitter can settle in the lining of bones and may cause bone cancer or bone marrow disorder. Generally, there is a high indication that most of the radioactive concentrations deposit in the samples were not during the mode of processing and preparation by the traditional practitioners but possibly as a long-time deposit in the wild animals used as medicines.

The radiation health hazard indices showed a highly significant health risk, when compared with the world averaged estimated specific activity in the earth crust (UNSCEAR 2000; Anekwe and Avwiri 2016; Idoko *et al.*, 2016). The noticeable influence of consumption and application of these animal parts could grossly affect the gonad as well as increase the excess lifetime cancer risk over a certain period of years. The health effects of radiation exposures to NORMs from the intake of medicinal plants and herbal preparations to their level are associated with most forms of

Figure 1: Examples of animals and animal parts sold in Zaria Local Government Area, Kaduna State, Nigeria

A. Skins of different animals display
B. The shell of tortoise, snail and other products mixed.
C. A view of the traditional practitioners and animal material being sold
D. Different dried faeces and dungs of animals being sold
E. A sighted feather and a pen for list supply to a user
F. The traditional practitioners selling a dead (rat) dried animal part to a user.

Table 1: Naturally occurring radioactive materials (NORMs) of the animal parts used in traditional medicines in Zaria Nigeria

Scientific names (English common names)	**Hausa names**	**Part used**	**K-40 ± (Bq/Kg)**	**Ra-226± (Bq/Kg)**	**Th-232 ± (Bq/Kg)**
Mormyrus bane Lacépède, (Electric fish)	*Wuta Kifi*	Whole dried	192.4706 ± 8.6877	57.8575 ± 0.7192	39.8301 ± 0.3539
Crocodylus niloticus Laurenti (Nile crocodile)	*Kada*	Skin	195.4202 ± 4.5584	57.6577 ± 0.6793	48.4410 ± 1.1796
Archachatina marginata Swainson (Giant African snail)	*Dodon kodi*	Shell	114.4420 ± 6.9180	122.1481 ± 1.9579	63.2249 ± 2.3591
Loxodonta africana Linn (African Elephant)	*Giwa (katon Nama)*	Bone	197.6189 ± 6.7035	50.6653 ± 4.6350	61.6915 ± 0.4718
Python sebae Gmelin (Rock python)	*Mesa/Mai hadiya*	Whole dried	195.5811 ± 6.7571	51.6642 ± 3.6361	112.61 ± 2.4771
Panthera leo Linneaus (Lion)	*Zaki*	Bone	69.9844 ± 4.3975	79.4342 ± .2775	28.9781 ± 0.8257
Poelagus marjorita St. Leger (Rabbit)	*Zomo*	Bone	61.5112 ± 4.1294	67.9266 ± 1.4384	14.2335 ± 0.7077
Panthera tigris Linneaus (Tiger)	*Damisa*	Shell/Bark	123.3979 ± 7.5079	37.8791 ± 4.1555	77.4584 ± 1.6121
Equus ferus caballus Linneaus (Horse)	*Doki*	Bone	185.6063 ± 4.2902	26.2916 ± 2.4773	46.3178 ± 1.6121
Varanus niloticus Linn (Bosch monitor lizard)	*Patan tsari*	Skin/head	41.7762 ± 2.3060	55.9795 ± 2.4773	55.7937 ± 2.5951
Aldabrachelys gigantean Schweigger (Tortoise)	*Kunkuru*	Shell	33.1957 ± 3.0032	58.0573 ± 4.0756	15.7276 ± 1.3368
Naja nigricollis Hallowell (Black cobra)	*Bakin maciji*	Whole	89.1296 ± 5.2555	48.2279 ± 3.8359	8.7288 ± 0.2359
World average (UNSCEAR 2000)			412.00 ± 0.00	45.00 ± 0.00	32.00 ± 0.00

Values are means ±Error (Bq/kg) of triplicate samples
Bq/kg = Becquerel per kilogram

Table 2: Radiation Health Hazard Indices of the animal and animal parts used in traditional medicines in Zaria Nigeria.

Scientific names (English common names)	Annual effective dose rate AEDE Outdoor) (μ Sv⁄y	Annual effective dose rate AEDE indoor) (μ Sv⁄y	Absorbed dose rate D (nGy/h)	Radium equivalent activity index Ra_{eq} (Bq/Kg)	Annual gonad equivalent dose AGED mSv/yr	Representative level index I_{γ} (Bq/Kg)	External hazard index H_{ex}(mSv/y)	Internal hazard index H_{in} (mSv/y)	Excess lifetime cancer risk (*E*-03) outdoor	Excess lifetime cancer risk (*E*-03) indoor
Mormyrus bane Lacépède (Electric fish)	72.91	291.64	59.45	129.63	405.71	0.91	0.35	0.19	0.26	1.021
Crocodylus niloticus Laurenti (Nile crocodile)	79.53	318.13	64.85	141.98	442.01	0.99	0.38	0.23	0.28	1.11
Archachatina marginata Swainson (Giant African snail)	123.18	492.72	100.44	221.37	677.65	1.52	0.60	0.27	0.43	1.72
Loxodonta africana Linn (African Elephant)	74.77	343.24	69.97	154.10	476.48	1.09	1.02	0.28	0.26	1.20
Python sebae Gmelin (Rock python)	125.18	500.71	102.07	227.76	691.76	1.60	0.62	0.48	0.43	1.75
Panthera leo Linneaus (Lion)	70.60	282.42	57.57	126.26	388.56	0.87	0.34	0.13	0.25	0.99
Poelagus marjorita St. Leger (Rabbit)	52.40	209.62	42.73	93.02	288.70	0.63	0.25	0.07	0.18	0.73
Panthera tigris Linneaus (Tiger)	86.87	347.46	70.83	158.15	479.57	1.11	0.43	0.32	0.30	1.22
Equus ferus caballus Linneaus (Horse)	59.68	238.71	48.66	106.82	333.13	0.76	0.29	0.22	0.21	0.84
Varanus niloticus Linn (Bosch monitor lizard)	76.40	305.62	62.30	138.98	419.31	0.96	0.38	0.22	0.27	1.07
Aldabrachelys gigantean Schweigger (Tortoise)	46.53	186.12	37.94	83.10	255.56	0.57	0.22	0.07	0.16	0.65
Naja nigricollis Hallowell (Black cobra)	38.46	153.84	31.36	67.57	213.50	0.47	0.18	0.05	0.13	0.54

leukaemia and other carcinogens of many organs such as the bone, lung, breast and thyroid in the long term (CNSC, 2013). Likewise, ^{238}U has an affinity for electron donor and could deposit itself in the tissues of the lung and lining on the bones marrows which may lead to leukaemia condition. Also, ^{232}Th an alpha particle emitter when settled in the lining of the bones may lead to bone cancer (Cherry *et al.,* 2012). Conversely, potassium - 40 is not considered to be of radiological significance since the human body control system homeostatically and the volume is dependent on the activity concentration of potassium in the body.

Conclusion

The radioactive concentrations in the animals sold as medicines in Zaria, Kaduna state, Nigeria might cause possible health damage and further deteriorate the wellbeing of the populace when consumed and inhaled. This study concludes that animal and animal parts usages in traditional medicines might not be as safe as medicine since they are potential sources of illness and diseases to human. We recommend further investigation of animal and animal-derived products used in Nigeria as medicines and possible standardization of the radioactivity concentration levels towards safe utilization.

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