Natural Radioactivity Levels of Some Herbal Plants With Antimalaria Potency In Ibadan South-West Local Government Area Of Oyo State, Nigeria

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Abstract: Natural radioactivity levels of twelve herbal plants with antimalarial potency in Ibadan South-West Local Government Area of Oyo State, Nigeria were investigated by gamma-ray spectroscopy (NaI(TI)) to determine the activity concentration of 238 U, 232 Th and 40 K. Relevant radiological hazard parameters due to ingestion of the herbal plants were also evaluated. Results of the study show the mean activity concentrations of 238 U, 232 Th and 40 K. Relevant radiological hazard parameters due to ingestion of the herbal plants were also evaluated. Results of the study show the mean activity concentrations of 238 U, 232 Th and 40 K as 5.79 ± 1.51 , 4.13 ± 0.55 and 630.03 ± 52.9 Bqkg⁻¹ respectively.Estimation of the radiological risk parameters showed: average annual committed dose (0.001 - 0.01mSvy⁻¹) with a mean value of 0.005 ± 0.004 mSvy⁻¹, radium equivalent (7.50 - 145.48 Bqkg⁻¹) with a mean value of 59.9 ± 42.65 Bqkg⁻¹ and internal hazard indices (0.03 - 0.43) with a mean value of 0.18 ± 0.12 ; these values lower than the world average values of 0.3mSvy⁻¹, 370Bqkg⁻¹ and unity respectively for ingestion of natural radionuclides provided by the United Nations Committee on the effects of Atomic Radiation (UNSCEAR)reports for any individual indicate that the associated radiological health risk resulting from the ingestion of radionuclides in the herbal plants is insignificant. Consequently, the herbal plants of this study are considered safe in terms of the radiological health hazards. The results therefore provide baseline values which may be useful in establishing rules and regulations relating to radiation protection as well as developing standards and guidelines for the use of medicinal plants to the appropriate authorities.

Key words: Natural radioactivity, herbal plants, Gamma spectrometry, Average annual effective dose, radium equivalent, internal hazard index

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I. Introduction

Malaria is a disease which primarily affects poor populations in tropical and subtropical areas, where the temperature and rainfall are suitable for the development of vectors and parasites (Greenwood *et al.*, 2008). The therapeutic use of natural products from indigenous plants for treatment of malaria is common in the rural areas of developing countries where commercial drugs are mostly unaffordable or unavailable; furthermore, traditional medications are readily available and more culturally acceptable (Okatch *et al.*, 2012). In Nigeria today, the use of herbal medicines for therapeutic purposes has increased drastically due to the fact that they are cheap, readily available and widely distributed. Besides, Nigeria being in the tropics, has forest that are full of cheap, easily available and sustainable medicinal plants which can be used and have always been used for the treatment of various diseases (Oni *et al.*, 2011).

Natural occurring radioactive materials (NORMs) are found in every constituent; air, water, soil, food and in humans (Tettey-Larbi*et al.*, 2013). According to International Food Safety Authorities Network (INFOSAN, 2011), plants used as food commonly have ⁴⁰K, ²³²Th and ²³⁸U and their progenies. It is expected that similarities would be found in plants used for therapeutic purposes since plants are the primary pathway of natural radionuclides entering into the human body through the food chain. Ingestion of plants containing radionuclides is the primary pathway of being exposed to internal radiation (Mollah, 2014; Krisananuwat*et al.*, 2015; Al-Masri*et al.*, 2008). Most of the radionuclides found in plants could be as a result of direct deposition from the atmosphere to the external surfaces of the plant, re-suspension of soil on aerial parts and direct uptake via the root (Al-Masri*et al.*, 2008; Al-Masri*et al.*, 2015; Vera *et al.*, 2003). The consumption of the plants containing the radionuclides by man could then lead to continuous radiation dose (Krisananuwat*et al.*, 2017).

Many indigenous plants are being used in herbal medicine for the treatment of malaria. Such plants include *Lawsonia inermis, Vernoniaamygdalina, Enantiachlorontia, Azadirachtaindica, Curcuma longa, Nauclealucida, Khayagrandifoliola, Alstoniaboonei, Manigeraindica, Spathodeacampanulata, Dacryodesedulis and Azadirachtaindica.* To the best of our knowledge, no documented reports on the specific activity of the natural radionuclides of the above listed herbal plants with antimalarial potency in Ibadan South-West Local Government Area (LGA), Oyo State, Nigeria has been published. Therefore, this work is aimed at establishing a baseline radioactive data of the specific activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th of twelve herbal plants used for the treatment of malaria in selected areas of Ibadan South-West Local Government Area, Oyo State, Nigeria.In addition to this, is to estimate the radiological hazard parameters due to the ingestion of ²³⁸U, ²³²Th and ⁴⁰K in the herbal plants.

II. Materials and Method

2.1 Study area

This study was conducted at Ibadan South -West Local Government Area (LGA). It is one of the thirty three LGAs in Oyo State in Nigeria and has an area of 40km² and a population of 282,585 at the 2006 population census (Makinde*et al.*, 2016). It lies between longitude 7.3694 and latitude 3.8596 with several settlements. An ethno-botanical survey was conducted in Bode market, one of predominantly herbal markets of Ibadan South-West LGA between December 2016 and January 2017. The herbal market is distinct for having a high proportion of herb sellers, aged locals or elders and traditional herbal medicine practitioners. These people treat ailments using plant remedies on the basis of their rich ethno-botanical knowledge.

2.2 Sample collection

Twelve (12) different medicinal plants parts samples (Table 1) used extensively for the treatment of malaria fever in Ibadan South-West Local Government Area were sampled. The various plants considered are; *Lawsonia inermis, Vernoniaamygdalina, Enantiachlorontia, Azadirachtaindica, Curcuma longa, Nauclealucida, Khayagrandifoliola, Alstoniaboonei, Manigeraindica, Spathodeacampanulata, Dacryodesedulisand Azadirachtaindica.* The plants were purchased from Bode market. The samples were transferred into the laboratory after they were labelled accordingly.

2.3 Sample preparation

The samples were then first washed under running water and then with distilled water to rid it of contaminants and air-dried on trays at a room temperature for a period of six weeks. The samples were then milled into fine powder and filtered with a sieve so as to obtain uniformly homogenous sample matrix. The powdered materials were stored in air-tight containers prior to further analysis.

2.4 Laboratory procedure

Measurements are carried out by adopting systems of gamma spectrometry from CANBERRA, equipped with a high efficiency scintillation detector, an NaI(Tl) detector of $(3"\times3")$ crystal dimension, with resolution 7.5% for ¹³⁷Cs (661.7 keV). An aluminium shield of 0.5 mm thickness was put around the detector to lessen the background, with a 0.3 cm layer of copper sheets to weaken x-rays emitted. The spectra are analyzed off-line using the GENIE 2000 data acquisition and analysis system. The activity concentration is expressed in (BqKg⁻¹) dry weight. Necessary energy and efficiency calibrations of the detector were done on the detector using the IAEA-385 supplied reference materials for the quantitative determination of ⁴⁰K, ²³²Th and ²³⁸U. The system was set at a working energy range of 0 - 3000 keV, which accommodated the energy range of interest in the study. The specific activity of ⁴⁰K was directly identified from the peak areas at 1460 keV. The activity concentrations of ²³⁸U and ²³²Th were measured presuming secular equilibrium with their decay products. To measure the activity concentration of radioisotope in the ²³⁸U-series, gamma transition lines of ²¹⁴Bi (1765 keV) were employed. Also, radioisotope activity concentrations in the ²³²Th-series were identified by applying gamma transition lines of ²⁰⁸T1 (2614 keV). The average counting time is 10,800 s for each sample in the set geometry, to ensure a good statistical significance.

2.5 Calculations

The activity concentration of the samples in $BqKg^{-1}$ was evaluated by the expression given by Ebaid, (2010):

$$A_{sp}(E,i) = \frac{N_{sam}(E,i)}{E_{V}(E)T_{e}P_{v}(E,i)M_{sam}}$$

Where N_{sam} (E,i) is the net counts for the radionuclide *i* at energy *E*, $\varepsilon_{\gamma}(E)$ is the photo peak efficiency at energy *E*, T_c is the counting live-time (secs), P γ (E, i) is the gamma emission probability of the radionuclide *i* for a transition at energy *E*, M_{sam} is the dry-weight of samples in kg.

(1)

Having obtained the values for the specific activity concentrations of the individual naturally occurring radionuclides in the herbal plants, the average annual committed effective dose, (2)

 $AACED = C_rDCF_{ing}A_{sp}$

Where AACED = average annual committed effective dose, C_r = consumption rate of intake of NORMs in the plant sample, DCF_{ing =} dose conversion coefficient for each radionuclide (i.e. 4.5 x10⁻⁵ mSv/Bq, 2.3 x 10⁻⁴ mSv/Bq and 6.2 x 10⁻⁶ mSv/Bq for ²³⁸U, ²³²Th and ⁴⁰K respectively) (UNSCEAR, 2000) and A_{sp} = specific activity of radionuclide in the plant sample. The appropriate dose of herbal plant depends on several factors. A consumption rate (C_r) of 1kgy⁻¹ reported by Nginja*et al.*, (2015) was used in this study. The maximum value for insignificant radiological health risk has been reported to be 0.3mSvy⁻¹ by UNSCEAR (2000).

Radium equivalent activity represents a weighted sum of activities of ²³⁸U, ²³²Th and ⁴⁰K and is based on the estimation that 1Bq kg⁻¹ of 238 U, 0.7Bq kg⁻¹ of 232 Th and 13Bq kg⁻¹ of 40 K produce the same radiation dose rates (Tripathi*et al.*, 2013). The index in equation (3) is given by UNSCEAR(2000): $\begin{aligned} &\kappa a_{eq}(BqKg^{-}) = A_{U} + 1.43A_{Th} + 0.077A_{K} \end{aligned} (3) \\ & \text{Where, } A_{U}, A_{Th} \text{ and } A_{K} \text{ are activity concentrations of } ^{238}\text{U}, ~^{232}\text{Th and } ^{40}\text{K} \text{ respectively.} \end{aligned}$

Taking into consideration the hazardous effects of radon a progeny of radium and its short-lived products to the respiratory organs, the internal exposure is quantified by the internal hazard index (H_{in}) using the expression given by Tufailet al., (2000):

 $H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}$ (4) Where, A_U, A_{Th}, and A_K are the radioactivity concentration of ²³⁸U, ²³²Th and ⁴⁰K in the samples.

The internal hazard index has to be less than unity as well to provide safe radionuclide levels in medicinal plants (Diabet al., 2008).

Table 1: Herbal plants examined including their scientific names, family names, common names and parts

S/N	Scientific Name	Family Name	Common Name	Part sampled
1	Lawsonia inermis	Lythraceae	Henna plant (Ewe laali)	Leaf
2	Enantiachlorontia	Annonaceae	African yellow wood (Yanni)	Bark
3	Curcuma longa	Zingiberaceae	Tumeric (Atale pupa)	Corm
4	Nauclealatifolia	Rebiaceae	African peach (Egbesi)	Bark
5	Dacryodesedulis	Rosaceae	Weeping pear (Ewe pear)	Leaf
6	Morindalucida	Rubiaceae	Brimstone tree (Oruwo)	Bark
7	Khayagrandifoliola	Neliaceae	African mahogany (Oganwo)	Bark
8	Alstoniaboonei	Apocynaceae	Stoolwood (Ahun)	Bark
9	Magniferaindica	Ancardiaceae	Mango (Mongoro)	Bark
10	Vernonia amygdalina	Asteraceae	Bitter leaf (Ewuro)	Whole plant
11	Spathodeacampanulata	Bigloniaceae	Family tree/tree of life (Akoko)	Leaf
12	Azadirachtaindica	Meliaceae	Neem tree (Dongoyaro)	Bark

III. **Results and Discussions**

3.1 Activity concentrations in the herbal plants

The activity concentrations due to 40 K, 238 U and 232 Th in twelve herbal plants of antimalarial potency in Ibadan South-West LGA have been determined and the results are presented in Table 2. Activity concentrations of radionuclides less than the corresponding detectable levels of 0.0975 Bq/kg, 0.0255 Bq/kg and 0.00787 Bq/kg for ⁴⁰K, ²³⁸U and ²³²Th respectively are termed as below detectable limit (BDL) for each radionuclide. The 238 U activity concentration in the herbal plants ranged from (BDL)Bqkg⁻¹to 17.54 ± 4.52 Bqkg⁻¹with an average value of 5.79 ± 1.51 Bqkg⁻¹. The highest activity concentration was recorded for *Alstoniaboonei* while Dacryodesedulis, Khayagrandifoliola, Spathodeacampanulata and Azadirachtaindica had the lowest activity concentration. For the activity concentration of 232 Th, it varied from 0.16 ± 0.02 Bqkg⁻¹ to 11.20 ± 1.25Bqkg⁻¹ with an average value of $4.13 \pm 0.55 \text{Bqkg}^{-1}$. The highest and lowest activity concentration was recorded for *Curcuma longa* and *Spathodeacampanulata*respectively.⁴⁰K recorded the highest activity concentration in all the herbal plants compared to the activity concentration of 238 U and 232 Th observed. The activity concentration varied from(BDL)Bqkg⁻¹ to 1658.97 ± 132.73 Bqkg⁻¹ with an average value of 630.02 ± 52.9Bqkg⁻¹. Morindalucida recorded the highest activity concentration, followed by Vernonia amygdalina while the lowest was recorded in*Enantiachlorontia*. The activity concentration have been compared and represented in Figure 1. The variation of the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in the different medicinal plant samples may be due to the fact that, the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K differ geographically from one soil of cultivation to another and some plants also absorbs certain elements more than others (Tettey-Larbiet al., 2013).

3.2 Radiological Hazard Risk Assessment

The average annual committed effective doses due to the ingestion of 238 U, 232 Th and 40 K in the herbal plants are also presented in Table 2. The average annual committed effective dose of 238 U, 232 Th and 40 K varied from 0.001 ± 0.00 to 0.01 ± 0.00 mSvy⁻¹ with an average of 0.005 ± 0.004mSvy⁻¹. The highest value was recorded for *Curcuma longa*, *Morindalucida*, *Khayagrandifoliola*, *Magniferaindica*, *Vernoniaamygdalina* and *Spathodeacampanulata* while *Enantiachlorontia* and *Dacryodesedulis* recorded the lowest as shown in Figure 2. The AACED due to ingestion of radionuclide in the herbal plant samples are far below the world average of 0.3 mSvy⁻¹ for ingestion of natural radionuclides provided in UNSCEAR (2000) report.

The radium equivalent activity (Ra_{eq}) (Table 3, column 1) of the study was found in the range of 7.50 ± 0.94 to 145.48 ± 14.04 Bqkg⁻¹ with an average value of 59.9 ± 42.65 Bqkg⁻¹; the highest value obtained in*Morindalucida* and the least value in*Enantiachlorontia*. These values are considered safe since they are well below limit of 370 Bqkg⁻¹ recommended by UNSCEAR(2008).

The estimated internal hazard indices (Table 3, column 2) of the study ranged from 0.02 ± 0.00 in *Enantiachlorontia* to 0.43 ± 0.02 in *Morindalucida* with an average value of 0.18 ± 0.12 . The internal hazard indices value of this study is far below the international accepted value of unity (UNSCEAR, 2000). The values of the internal radiation hazard indices less than unity confirms that it is safe to use the herbal plants for curative purposes.

		sa	liipies		
Samples	Acti	vity concentration	Average annual committed dose		
⁴⁰ K 23	²³² Th			(mSvy ⁻¹)	
Lawsonia inermis	356.96 ± 32.84	12.32 ± 3.24	2.71 ± 0.34	0.003 ± 0.00	
Enantiachlorontia	BDL	0.19 ± 0.08	5.11 ± 0.60	0.001 ± 0.00	
Curcuma longa	1021.28 ± 85.36	4.48 ± 1.17	11.20 ± 1.25	0.01 ± 0.00	
Nauclealatifolia	354.54 ± 33.13	3.73 ± 0.99	3.77 ± 0.44	0.003 ± 0.00	
Dacryodesedulis	75.02 ± 6.70	BDL	4.41 ± 0.52	0.001 ± 0.00	
Morindalucida	1658.97±132.73	12.13 ± 3.13	3.92 ± 0.48	0.01 ± 0.00	
Khayagrandifoliola	733.29 ± 62.44	BDL	3.93 ± 0.48	0.01 ± 0.00	
Alstoniaboonei	304.93 ± 28.53	17.54 ± 4.52	2.45 ± 0.98	0.003 ± 0.00	
Magniferaindica	389.63 ± 34.42	15.86 ± 4.13	1.82 ± 0.22	0.01 ± 0.00	
Vernonia amygdalina	1604.52±127.07	3.17 ± 0.89	5.33 ± 0.62	0.01 ± 0.00	
Spathodeacampanula	ta 861.55 ± 73.90	BDL	0.16 ± 0.02	0.01 ± 0.00	
Azadirachtaindica	199.66 ± 17.83	BDL	4.76 ± 0 .59	0.002 ± 0.00	

Table 2: Activity concentration and average annual committed doses	of ²³⁸ U,	, ²³² Th and	⁴⁰ K in th	e herbal plant
somplas				

BDL = Below Detection Limit



Figure 1: Comparison of the activity concentration (A_{sp}) of ²³⁸U, ²³²Th, and ⁴⁰K in the various species of the herbal plant samples

Table 4 shows previously published work with the activity concentration of NORMs in medicinal plants by various researches and their average values compared with this study. It was observed that the average concentration of 238 U is lower than that of Tetty-larbi*et al.*, (2013) and Olatunde*et al.*, (2011). For 232 Th, the average concentration was lower than Tetty-larbi*et al.*, (2013); Olatunde*et al.*, (2011); Jevremovic*et al.*, (2011). 40 K of the study was found to be higher than the previous work except for Tetty-larbi*et al.*, (2013). The variation in the activity concentration may be due to differences in geographical location of the plants and the radiochemical composition of the soils in which these medicinal plants were cultivated since the levels of activity concentration of natural radionuclides are not normalized across the world and the plants have the ability to absorb particular elements more than others.

Table 3: Radium equivalent and internal hazard index in herbal plants in this stud	Table 3: Radium equivalent and internal hazard index	in herbal plants in this study
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Sample	Ra _{eq} (Bqkg ⁻¹)	$\mathbf{H}_{\mathbf{in}}$
Lawsonia inermis	43.68 ± 5.65	0.15 ± 0.03
Enantiachlorontia	7.50 ± 0.94	0.02 ± 0.00
Curcuma longa	95.75 ± 8.89	0.28 ± 0.03
Nauclealatifolia	36.42 ± 0.87	0.11 ± 0.01
Dacryodesedulis	12.08 ± 0.56	0.03 ± 0.00
Morindalucida	145.48 ± 14.04	0.43 ± 0.02
Khayagrandifoliola	62.08 ± 6.76	0.17 ± 0.01
Alstoniaboonei	44.52 ± 5.55	0.17 ± 0.03
Magniferaindica	48.46 ± 5.67	0.17 ± 0.03
Vernonia amygdalina	134.34 ± 12.98	0.37 ± 0.03
Spathodeacampanulata	66.57 ± 7.65	0.18 ± 0.01
Azadirachtaindica	22.18 ± 9.43	0.06 ± 0.00
Average	59.9 ± 42.7	0.18 ± 0.12
UNSCEAR, 2008	≤ 370 Bqkg ⁻¹	≤1

238U		²³² Th	²³² Th ⁴⁰ K		Reference	
Range	Average	Range	Average	Range	Average	
BDL - 17.54	5.79	0.16 - 11.20	4.13	BDL - 1658.97	630.03	This work
BDL - 12.59	4.68	BDL - 14.63	2.91	78.56 - 579.32	219.13	Abojassimet al., 2016
20.4 - 46.9	31.8	42.0 - 70.6	56.2	566.40 - 1093.1	839.8	Tetty-Larbiet al., 2013
14.7- 16.2	15.6	7.0 - 11.4	8.5	66.8 - 70.2	67.9	Olatundeet al., 2011
0.6-8.2	2.6	1.7 - 15.1	7.4	126.0 - 1243.7	589.6	Jevremovicet al., 2011

Table 4: Comparison of the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K (Bqkg⁻¹) in the herbal plants of this study with previous work done.

BDL = Below Detection Limit

IV. Conclusion

This study assessed the natural radionuclide concentrations of twelve herbal plants used for treating malaria in Ibadan South-West Local Government Area of Oyo State, Nigeria. This study proves the presence of activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th in the herbal plants of study. ⁴⁰K was significantly large compared to any other radionuclides in the samples. It is quite natural to say that these radionuclides may be infused into the human body when the herbal plants are used for the treatment of malaria. The values of activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in samples of the herbal plants are found to be lower than the world average allowed maximum values 32, 30 and 400Bqkg⁻¹ respectively, except the activity concentration of ⁴⁰K that found to be higher in samples Curcuma longa, Morindalucida, Khayagrandifoliola, Vernoniaamydalinaand Spathodeacampanulata. This can be explained by the soil that comes as a result of an abundance of this isotope concentration. The corresponding average annual effective dose determinedin this study to any individual's organ or tissue in the population group due to the ingestion of natural radionuclidesin the medicinal plants is far below the averageradiation dose of 0.3 mSvy^{-1} received per head worldwidedue to the ingestion of natural radionuclide (UNSCEAR2000). The values for the radium equivalent activity (Ra_{eq}) are turned to be within the international average allowed maximum value of 370 Bqkg⁻¹. The value of hazard internal is lower than the international permissible value of unity. The result presents insignificant radiological risks associated with the ingestion of the herbal plants in the LGA of study and hence may be considered safe for human use.

Competing interests

All authors declare no financial competing interests.

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