ChemSearch Journal 11(1): 118 – 125, June, 2020
Publication of Chemical Society of Nigeria, Kano Chapter

Received: 22/05/2020 Accepted: 09/06/2020
http://www.ajol.info/index.php/csj

Plant Source Apportionments and Radiological Risk Using Natural Radionuclides of Herbal Remedies Consumed in Katsina, Nigeria

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ABSTRACT

The use of herbal remedies for treatment of various ailment are common practice in developing countries, but the formulation of these products is usually complex, hence, the need for thorough quality check especially with possible health risk these could pose. Therefore, this study examined the plant mix of *Anogeissus leiocarpus, Prosopis Africana, Boswellia odorata and Guiera senagalensis among the samples* in ten (10) herbal remedies sold in Katsina, Nigeria using cluster analysis of the natural radionuclides (K-40, Ra-226 and Th-232) and further evaluates the radiological hazard due to consumption of the herbal remedies. The activity concentration of the K-40, Ra-226 and Th-232 was determined using gamma spectroscopic analysis and the Radium equivalent activity (Ra_{eq}), Average Annual Committed Effective Dose (AACED) and Annual Gonad Equivalent Dose (AGED) due to consumption of radionuclides in the herbal remedies were calculated. The results recorded the activity concentration of K-40 ranges from 63.92±2.78 - 210.43±6.54 Bq/Kg, Ra-226 varied from 8.55±4.07 - 41.19±2.71 Bq/Kg and Th-232 activity concentration ranges from 30.51±0.27 - 157.31±1.29 Bq/Kg. The exposure of human consuming the herbal remedies using the AACED showed ingestion of K-40, Ra-226 and Th-232 in the herbal remedies is below the standard average radiation dose of 0.3 mSv. However, the indexes, Ra_{eq} and AGED in three samples are above 370 and 300 $mSvyr^{-1}$ recommended limits, respectively. Hence these herbal remedies are not safe for consumption based on radiolological hazard.

Keywords: Activity, Herbal Remedies, Natural Radionuclides, Nigeria

INTRODUCTION

The use of medicinal plants by man has existed for many years and their successes gave birth to the modern medicine. However, at the moment orthodox pharmaceuticals are not successful to offer general health benefits to the dream and goal of World Health Organization (WHO, 2002), hence, it has become imperative to assess both the beneficial and hazardous contents of some of the common alternative and traditional medicine employed in the treatment of diseases. About 80% of the developing countries rely on herbal drugs in their primary healthcare requirement (Desideri *et al*., 2010; Njinga *et al*., 2015).

Herbal drugs are generally made up of combinations of many components which eventually result in the expected efficacy. However, these products suffer from a range of shortcomings because of unacceptable evidences of safety, due to availability of natural occurring radioactive materials (NORMs). According to the International Food Safety Authorities Network (INFOSAN, 2011) and World Nuclear Association (WNA, 2014), natural radionuclides; ^{40}K, ^{232}Th and ^{238}U and their progenies are commonly found in plants, which may also be found in plants used for medicinal purposes.

The use of herbal drugs in Nigeria today especially in rural areas is based on the fact that medicinal plants are cheap, readily available and widely distributed which can be collected for the treatment of various diseases (Oni *et al*,. 2011; Njinga *et al*., 2015). In Nigeria, there are many registered and unregistered herbal medicines sold without restriction by license practioners.

According to Center for Disease Control and Prevention (CDCP, 2003), in 1999, following the use of lavalactone-containing herbal products, severe hepatic toxicity was reported and several of the patients required liver transplant. This report corroborate with Lordford *et al*. (2013) that possible side effects of NORMs in medicinal plants has not been considered because herbal plants are not considered in the group of edible plants that have been studied in the past by nutritionist. Hence, need for more research on safety of herbal formulation containing many plants.

This research work is focused on determining the activity concentration of naturally occurring radionuclides in herbal formulations sold in Nigeria using Sodium Iodide (NaI) Gamma-Ray spectrometer. This study determined the specific activity concentration of NORMs due to Ra-226, Th-232 and K-40 present in the selected medicinal plants and evaluate the annual effective doses due to ingestion of Ra-226, Th-232 and K-40.

The studies would be to determine the specific activity concentration of NORMs due to ^{226}Ra, ^{232}Th and ^{40}K present in the selected medicinal plants and evaluate the annual effective doses (**AACED)** due to ingestion of Ra-226, Th-232 and K-40, threshold consumption rate and annual gonad equivalent dose (AGED).

MATERIALS AND METHODS

Sample Collection

In December 2018, a total of 30 herbal remedies samples comprising three (3) samples each of ten (10) different herbal formulations were purchased from ten (10) herbalist outlets in Katsina metropolis 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 has neither being filed nor registered with NAFDAC and were locally packed in black polythene bags. The sample code and sampling locations coordinates are shown in Table 1. During sampling, medicinal use and names of plants used in formulation of the herbal remedies were disclosed by the herbalist and plants identified by botanist as presented in Table 2. The information on the sample composition was further validated using cluster analysis. The medicinal use of the herbal remedies includes typhoid, yellow fever, pile and fatigue.

Table 1: Coordinate of Sampling Locations

Sample Code	Latitude (N)	Longitude (E)
LR	12^0 50' 50	7^0 40' 11
BR	12^0 59' 39	7^0 35' 30
DR	13^0 16	7^0 36' 24
PK	12^0 59' 54	7^0 36' 20
GG	12^0 59' 31	7^0 36' 18
KM	12^0 59' 19	7^0 36' 26
SL	12^0 58' 36	7^0 36' 35
YK	12^0 58' 15	7^0 35' 59
ST	12^0 58' 4	7^0 36' 1
KK	12^0 18	7^0 37' 17

Table 2: Information of Plants used for the Sample Formulation

Sample Code	Botanical Name of Plants	Local Name of Plant	Part of the Plant
YK	*Anogeissus leiocarpus*	Marke	Leaves
	Prosopis Africana	Kirya	Bark
LR	*Same as above*	*Same as above*	*Same as above*
ST	*Prosopis Africana*	Kirya	Bark and leaves
SL	*Anogeissus leiocarpus*	Marke	Bark and leaves
BR	*Prosopis Africana*	Kirya	Bark
	Boswellia odorata	Hano	leaves
KK	*Anogeissus leiocarpus*	Marke	Bark and leaves
KM	*Same as above*	*Same as above*	*Same as above*
DR	*Guiera senagalensis*	Sabara	Leaves, Bark
	Prosopis Africana	Kirya	Stem
PK	*Anogeissus leiocarpus*	Marke	Leaves
	Prosopis Africana	Kirya	Bark
GG	*Prosopis Africana*	Kirya	Stem
	Anogeissus leiocarpus	Marke	Leaves
	Guiera senagalensis	Sabara	Bark

Sample preparation and Measurement

The samples collected are available in powder form. The samples were prepared using the procedure of Njinga *et al*. (2015). The samples were packed and sealed in air tight polythene bags and kept for about 28 days period to allow Ra-226 and Ra-228, and their progenies in the samples attained secular radioactive equilibrium (Lordford *et al*., 2013).

The analyses of the samples were carried out at the Centre for Energy Research and Training (CERT), Zaria-Nigeria using the same gamma ray spectrometric set up as employed by Njinga *et al*. (2015). Gamma ray spectrometry was employed for the radioactivity concentration measurements. The used detector assembly consisted of a 7.62 x 7.62 cm NaI (Tl) detector housed in a 6 cm thick lead shield, cadmium-lined assembly with copper sheets for the reduction of background radiation. The entire assembly was coupled to a computer-based Multichannel Analyzer (MCA) card system MAESTRO programmed used for the data acquisition and spectra analysis. The quantitative determination of K-40, Ra-226 and Th-232 in the herbal samples was done by calibration of the analyzer with the IAEA supplied reference materials as shown in Table 2. The absolute detection efficiency of the NaI (Tl) detector was determined using the standard sources according to Varier (2009). The activity concentration of radionuclides in the herbal samples were calculated using the expression according to Njinga *et al*. (2015).

Table 2: Spectra Energy Windows used in the Analysis

Isotope	Gamma energy (Kev)	Energy window (Kev)
R-226	1764.0	1620 - 1820
Th-232	2614.5	2480 - 2820
K-40	1460.0	1380 - 1550

The Average Annual Committed Effective Dose (AACED) from ingestion of NORMs in the herbal remedies was calculated using Lordford *et al*. (2013) expression as presented in equation 1:

$$E_{ave} = \sum C_f \; x \, DCF_{ing} \, x \, A_i \quad (1)$$

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.

A_i = Specific activity concentration of each radionuclides

In Nigeria, there is no standardized dosage for the use of herbal remedies, rather a table spoon is recommended twice daily. Therefore, according to Lordford *et al*. (2013), assuming 5g is equivalent to two table spoon consumed per day by individual, then, 1.8Kg/yr is assumed as the consumption rate for this study.

Using Equation 1, C_f the threshold consumption rate for each of the medicinal plant samples was obtained using equation 2.

$$C_f = \frac{3E_{ave}}{\sum_{i=1}^{3} (DCF_{ing} \; x \, A_i)} \quad (2)$$

where; E_{ave} = 0.3 mSv/yr is the threshold average annual committed effective dose due to ingestion of NORMs in medicinal plants according to UNSCEAR (2000),

A_1 A_2 and A_3 are Specific activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th respectively in the medicinal plant samples,

DCF_1, DCF_2 and DCF_3 are the DCF_{ing} for ^{40}K, ^{226}Ra and ^{232}Th

Radium equivalent activity (Ra_{eq}) was determined according to equation 3.

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

Internal hazard index (H_{in}) and external hazard index (H_{ex}) were determined using equations 4 and 5. While Annual Gonad Equivalent Dose (AGED) (mSvyr^{-1}) was determined using equation 6 in accordance with UNSCEAR (2010).

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (4)$$

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

$$AGED = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \leq 300 \quad (6)$$

Where C_{Ra}, C_{Th} and C_K are activity concentration of Ra-226, Th-232 and K-40, respectively

RESULTS AND DISCUSSION

Table 3 showed the activity concentration of natural radionuclides in the ten (10) different herbal remedies commonly used in Katsina. As shown, the activity concentration of ^{40}K in the Herbal remedies ranges from 63.92±2.78 Bq/Kg (LR) to 210.43±6.54 Bq/Kg (DR) with a mean value of 137.84±5.31Bq/Kg. For the activity concentration of ^{226}Ra it varied from 8.55±4.07

Bq/Kg (LR) to 41.19±2.71 Bq/Kg (KK) with an average value of 28.00±2.59 Bq/Kg. ^{232}Th activity concentration ranges from 30.51±0.27Bq/Kg to 157.31±1.29 Bq/Kg with average value of 59.93±1.09 Bq/Kg. The lowest activity was obtained for sample ST while the highest activity was obtained for sample PK. Generally, from the results recorded, the activity concentration of ^{40}K was observed to be the highest among other natural radionuclides (^{226}Ra and ^{232}Th) recorded in all the samples with exception of sample PK where ^{232}Th recorded the highest activity. The high activity concentration of ^{40}K compared to ^{226}Ra and ^{232}Th could be due to high absorption of ^{40}K by the sampled plants from the soil relative to other elements (Lordford *et al.*, 2013). The implication is that the samples could aid therapeutic treatment for high blood pressure in the consumer (HBPI, 2012). Also from the Table, the profile of activity concentration of the natural radionuclides follows the order: K-40 > Th-232 > Ra-226

Table 3: Activity Concentration in the Herbal Remedies

Sample Code	^{40}K (Bq/Kg)	^{226}Ra (Bq/Kg)	^{232}Th (Bq/Kg)
YK	129.4±2.61	22.45±1.87	49.18±0.78
LR	63.92±2.78	8.55±4.07	55.00±0.58
ST	103.34±5.30	30.36±1.99	30.51±0.27
SL	85.64±8.31	40.83±0.39	44.70±1.96
BR	173.59±8.09	14.62±1.55	39.20±1.73
KK	166.14±6.97	41.19±2.71	43.76±2.16
KM	140.67±4.29	31.88±2.71	66.37±0.78
DR	210.43±6.54	40.27±6.11	81.94±0.23
PK	129.4±7.61	26.17±3.91	157.31±1.29
GG	175.89±0.66	23.76±0.67	31.37±1.12
Mean	137.84±5.31	28.00±2.59	59.93±1.09

Comparison of the results from this study with other similar studies in Nigeria and other countries are presented in Table 4. The table below shows that the activity concentration of ^{40}K is the least compared to published reports in Nigeria and other countries. However, the activity concentration of ^{226}Ra and ^{232}Th are higher compared to data obtained from similar studied as presented in Table 5.

Table 4: Comparison of the Activity Concentration in the Herbal Remedies

	^{40}K (Bq/Kg)		^{226}Ra (Bq/Kg)		^{232}Th (Bq/Kg)	
Country	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**
[a]Nigeria (Katsina State)	63.92 - 210.43	137.84	8.55 - 41.19	28.00	30.51- 157.31	59.93
[b]Nigeria (Niger State)	74.6 - 324.2	171.7	10.8 - 42.5	25	27.8 - 41.1	35.1
[c]Ghana	566.4 - 1093.1	839.8			42.0 - 70.6	56.2
[d]Brazil	666.0 - 1216.0	976.3			<11 - 43.0	21.7
[e]Italy	5.4 - 3582.0	654.7			-	-
[f]Serbia	126.0 - 1243.7	589.6			17 - 15.1	7.4

[a]This Study; [b]Njinga *et al*. (2015); [c]Lordford *et al*. (2013); [d]Scheibel and Appoloni, 2007; [e]Desideri *et al*. (2010) [f]Jevremovic *et al*. (2011).

Figures 1 (a - c) shows the synergistic characteristics between the NORMs, using linear regression. The r^2 values indicated in the figures revealed positive relationship between the NORMs. The correlation between K-40 and Ra-226, K-40 and Th-232, and Ra-226 and Th-232 are 0.093, 0.002 and 0.002, respectively, with the highest r^2 val;ue between K-40 and Ra-226.

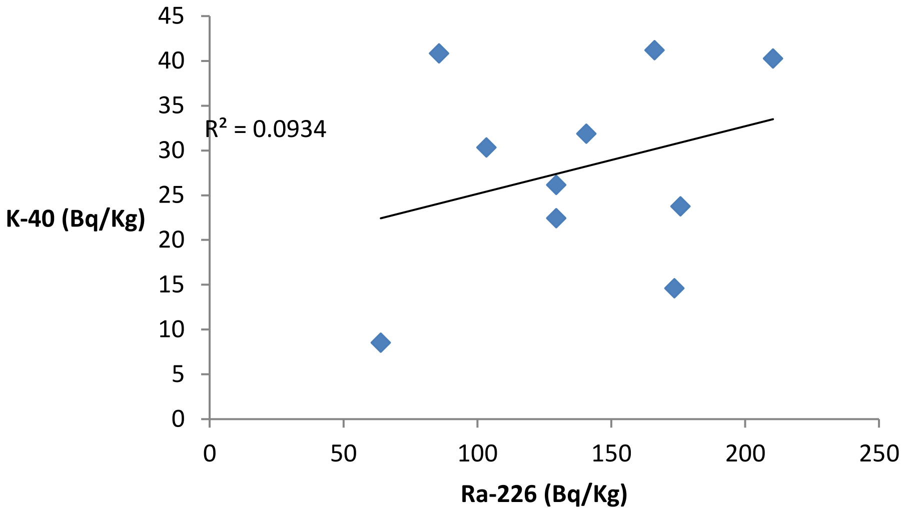

Figure 1a: Relationship between K-40 and Ra-226 Activity Concentrations of the Samples

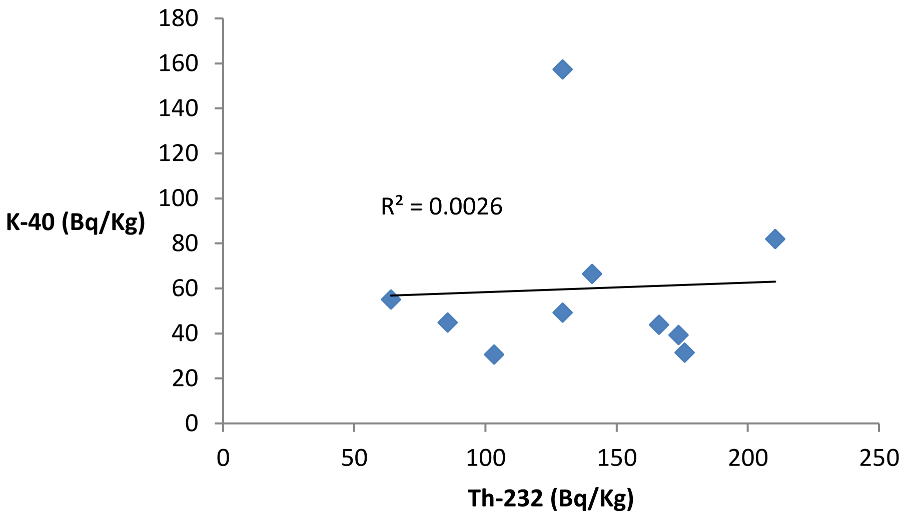

Figure 1b: Relationship between K-40 and Th-232n Activity Concentrations of the Samples

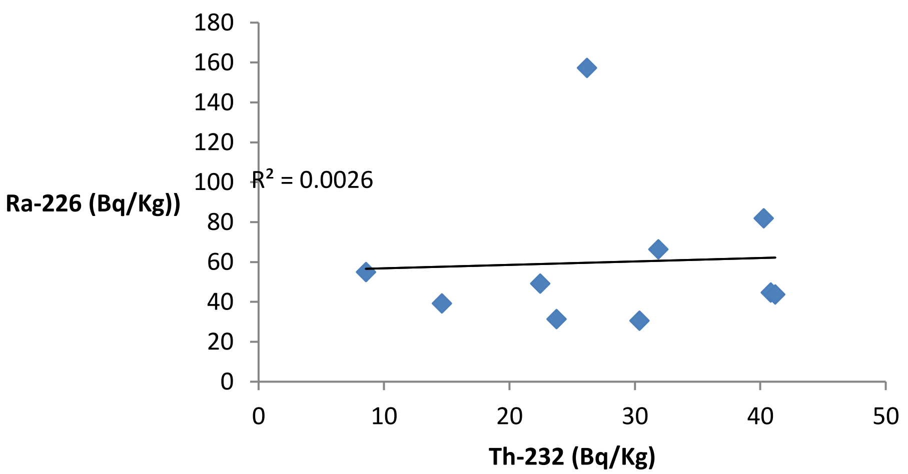

Figure 1c: Relationship between Ra-226 and Th-232 Activity Concentrations of the Samples

Based on the estimation that consumption rate of 1.8 Kg per annum of the herbal remedies were used, the AACED of the NORMs due to ^{40}K, ^{226}Ra and ^{232}Th were calculated as shown in Table 5. From the table, the Average Annual Committed Effective Doses varied from 0.00851±0.00052 mSvyr^{-1} (BR) to 0.02658±0.00086 mSvyr^{-1} (PK) with an average value of 0.01348±0.00060 mSvyr^{-1}. The highest value observed for sample PK is due to high activity concentration of ^{232}Th obtained for the sample compared to activity concentrations of ^{226}Ra and ^{40}K in the sample as shown in Figure 2. As shown, 40 – 70 % of the AACED is contributed by ^{232}Th compared to ^{40}K and ^{226}Ra. This observation corroborated to Lordford *et al*. (2013). Also, the value is higher than maximum values of 0.00686 ± 0.00044 and 0.014 ± 0.002 reported for medicinal plants in Nigeria (Njinga *et al*., 2015) and Ghana (Lordford *et al*., 2013), respectively.

Table 5: Radiological Indices for the Consumption of the Herbal Remedies

Sample Code	AACED (mSvyr^{-1})	Threshold consumption rate (Kgyr^{-1})	Internal hazard index (H_{int})	External hazard index (H_{ext})	Radium Equivalent Activity Ra_{eq} (Bq/Kg)	AGED (mSvyr^{-1})
YK	0.0110±0.0004	48.9139	0.3381	0.2775	102.7412	315.5745
LR	0.0093±0.0008	58.2869	0.2719	0.2488	92.1238	276.3966
ST	0.0097±0.0004	55.6972	0.3034	0.2213	81.9465	253.7930
SL	0.0133±0.0004	40.4597	0.4111	0.3007	111.3453	339.9017
BR	0.0085±0.0005	63.4435	0.2665	0.2270	84.0424	263.5391
KK	0.0136±0.0008	39.7737	0.4261	0.3148	116.5596	362.3619
KM	0.0150±0.0006	35.9086	0.4578	0.3717	137.6207	420.1062
DR	0.0189±0.0011	28.6383	0.5778	0.4690	173.6473	533.0185
PK	0.0266±0.0009	20.311	0.7757	0.7050	261.0871	779.0527
GG	0.0090±0.0003	60.1668	0.2861	0.2219	82.1626	259.7745
Mean	0.0135±0.0006	45.1599	0.4115	0.3358	124.3277	380.3518

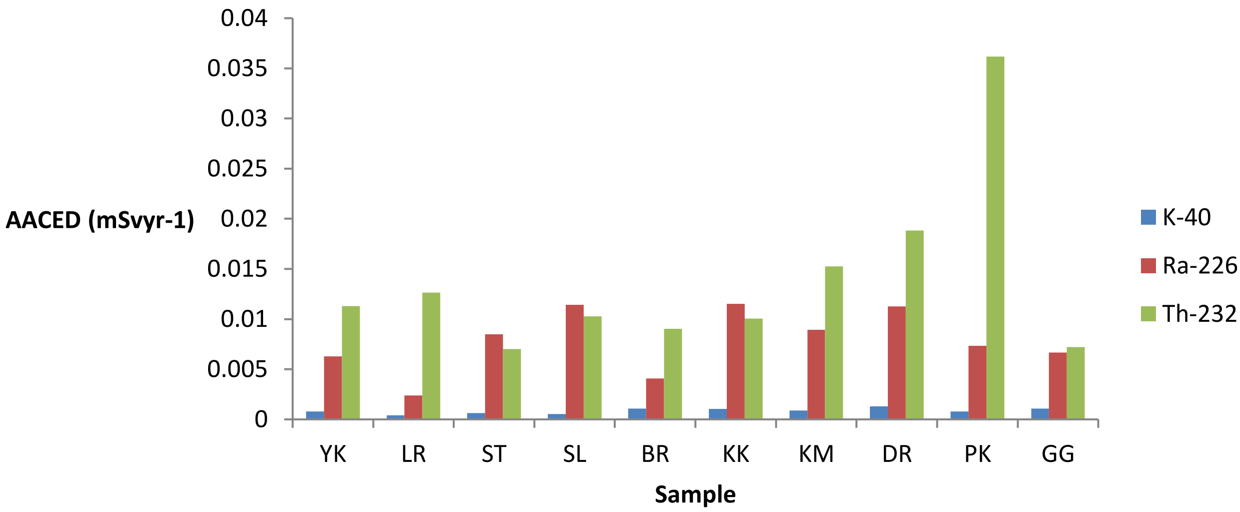

Figure 2: Average Annual Committed Effective Dose (AACED) from ingestion of the natural radionuclides in the herbal remedies

Also, as shown in Table 5 above, the threshold consumption rate varied from 20.3109 Kgyr^{-1} to 63.44347 Kgyr^{-1}. The highest and lowest threshold consumption rates were recorded for sample BR and sample PK, respectively. The trend in the values compared to AACED shows inverse relationship which implies the greater the AACED the lower the threshold value for the herbal remedies and *vis visa*. The highest value recorded in this study is lower than 70.37 Kgyr^{-1} reported by Njinga *et al*. (2015). Since threshold consumption rate is the minimum value above which AACED becomes greater than the 0.3 mSv, the values obtained in this study indicated that patients consuming the herbal remedies within this consumption rate would be exposed to insignificant radiological health risk. Similar, the effective equivalent dose due to radium, internal and external hazard index as shown in Table 5 revealed H_{int} ranged from 0.2665 for ample BR to 0.7757 for sample PK with average value of 0.4115. The H_{ext} varied from 0.2213 (sample ST) to 0.7050 (sample PK) with a mean value of 0.3359. Similar to H_{ext}, effective equivalent dose due to radium (Ra_{eq}) showed lowest value of 253.7930 and highest value of 779.0527 for samples ST and PK, respectively. The H_{in} and H_{ex} values for samples in this study is higher than 0.18 reported for medicinal plants in south-west Nigeria (Ademola and Omoboyede, 2018) which may be due to the complexity of the samples, however, the values were lower than 1 recommended standard. Moreso, the Ra_{eq} for samples KM, DR and PK are higher than 370 Bq/Kg recommended limits. Hence, the radiation hazard effect from these samples are significant, a serious health implication for consumer of the herbal remedies.

The Annual Gonad Equivalent Dose (AGED) in the present study as shown in Table 5 ranged from 253.7930 $mSvyr^{-1}$ (sample ST) **t**o 779.0527 $mSvyr^{-1}$ (sample PK) with an average value of 380.3518 $mSvyr^{-1}$. The samples KM, DR and PK are higher than 300 $mSvyr^{-1}$ recommended limits. This indicates that the sensitive cells including the gonad, bone surface cells and bone marrow could be susceptible to health risk for those using the herbal remedies for disease treatment.

Analysis of Similarities among Samples

Cluster Analysis of the samples based on the radioactivity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the samples were investigated to validate the composition of the herbal remedies Figure 3 (a and b) below show the dendrograms resulting from the cluster analysis of the samples based on the similarity in the NORMs concentrations in the samples. It can be seen in Figure 3a that the samples were divided into two separate groups, labeled groups I and II. Falling into group I are samples YK, KM, BR, GG, KK, and DR whereas Group II consist of samples LR, ST and SL. Sample PK is an outlier here due to its ^{232}Th value which is almost doubling or tripling the value of radioactivity due to ^{232}Th in the other samples investigated. Sample PK was removed and the analysis was repeated. Figure 3b shows a better defined relationship between the samples.

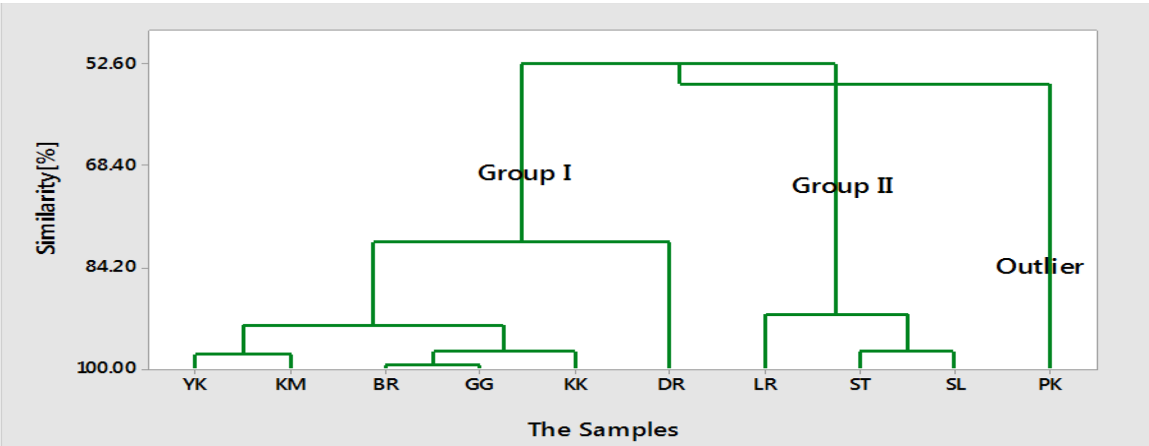

Figure 3a: Cluster Analysis of Medicinal Samples

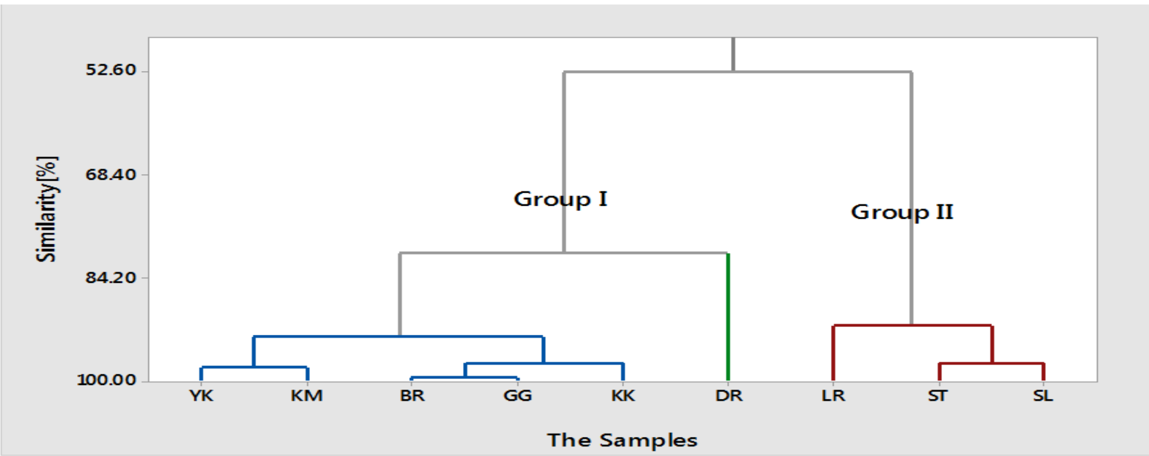

Figure 3b: Cluster Analysis of Medicinal Samples

CONCLUSION

This study provides confirmation of the plants mixed of *Anogeissus leiocarpus, Prosopis Africana, Boswellia odorata and Guiera senagalensis among the samples* using cluster analysis of activity concentrations of K-40, Ra-226 and Th-232 of the herbal remedies consumed in Nigeria. The statistical analysis showed the similarity in the samples which further establish the information provided by the herbalist. From the results, the activity concentration of ^{40}K was observed to be the highest among other natural radionuclides analysed (^{226}Ra and ^{232}Th) recorded in all the samples with exception of sample PK where ^{232}Th recorded the highest activity. Also, 40 – 70 % of the AACED is contributed by ^{232}Th compared to ^{40}K and ^{226}Ra. Other indexes, H_{int},and H_{ext} showed that consumption the herbal remedies do not pose any radiological risk to the consumers. However, samples KM, DR and PK among all the samples studied has higher Ra_{eq} and AGED above 370 and 300 $mSvyr^{-1}$ recommended limits, respectively. Hence the herbal remedies (KM, DR and PK) are not safe for consumption based on radiolological hazard analysis of the present work.

CONFLICT OF INTEREST

There is no conflict of interest among the authors.

ACKNOWLEDGMENT

The authors wish to acknowledge the assistance and support provide by Professor Adeyemo and Mr. I. A. Bappah of the Center for Energy Research and Training (CERT), Zaria-Nigeria in sample analysis and data interpretation.

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