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Assessment of Radioactivity Levels for ^{40}K, ^{238}U and ^{232}Th in Edible Leaves of *Vernonia amygdalina* and *Vernonia calvoana*

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Abstract

The focus of this study was to investigate the activity concentrations of ^{40}K, ^{238}U and ^{232}Th in edible leaves of *Vernonia amygdalina* (VA) and *Vernonia calvoana* (VC) and to derive from these activity concentrations, the associated Radium equivalent, the gamma dose rates, and the Internal hazard index (Hin). The average activity concentrations for ^{40}K, ^{238U} and ^{232}Th found in the fresh leaves of VA and VC was: 29.10 ± 3.01; 4.25 ± 1.56; 1.67 ± 0.76 Bq.Kg^{-1} and 32.56 ± 3.50; 4.50 ± 1.68; 1.68 ± 0.72 Bq.Kg^{-1} respectively. The average radium equivalent activities obtained for fresh leaves were 6.79 ± 2.84 Bq/kg and 6.79 ± 2.97 Bq/kg. The gamma dose rate of 3.94 and 4.12 then the hazard index of 0.026 were found both in VA and VC leaves samples.

***Keywords: Vernonia*; Radioactivity; Radium equivalent; Annual effective dose; Internal hazard index**

Introduction

In Cameroon leaves collected from different species of asteraceae plants like *Vernonia amygdalina* (VA), *Vernonia calvoana* (VC), *Vernonia richardiana* and *Vernonia hymenolepis* (VH) are used in folk medicine or consumed by millions of people as legumes. The estimated annual production of *Vernonia* in 2010 was 150 thousand metric tons of fresh leaves and about 75% is locally used for food while 25% is exported. These figures show both the growing needs of consumption of *Vernonia* leaves and the necessity to carry out a continuous monitoring of their radioactivity levels.

The soil-plant-human transfer of radionuclides is a frequent channel for human exposure to radiation coming from cosmic, terrestrial or anthropogenic sources [1,2]. It is also known that human exposure to high radiation doses induces acute or deterministic effects as reported by IAEA [3]. However, non-deterministic or stochastic effects due to low radiation doses are not predictable. According to UNSCEAR (2000) the global average annual human exposure from natural sources is 2.4 µ Sv/year [4].

The aim of this study is to investigate the activity concentrations of gamma emitting radionuclides contained in edible leaves of VA and VC and to derive the associated Radium Equivalent (Raeq), the annual gamma dose rates (Dy) and the internal hazard index (Hin) due to intake from nutrition of naturally occurring radionuclides ^{40}K, ^{238}U and ^{232}Th. Such parameters could be used as benchmark values for radiological protection of populations and quality control of foodstuffs.

Materials and Methods

Samples preparation

The study area shown in FIG. 1 covers the entire southern Cameroon, between the latitude and N and longitude and E. We have identified in this

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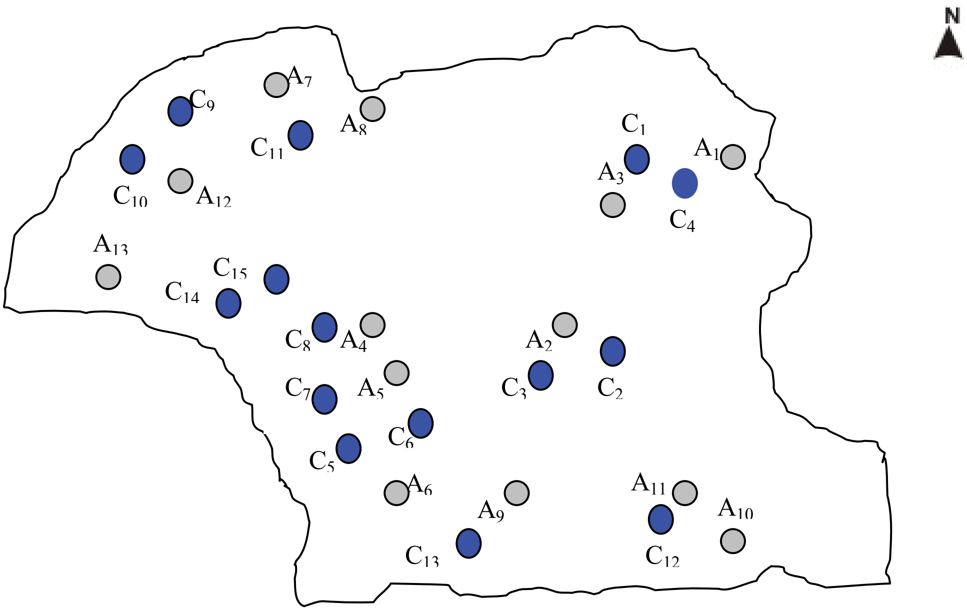

Figure 1: Sampling sites for VA (A1-A13) and VC (C1-C15) cultivated in south Cameroon.

study area 28 sampling sites depicted in FIG. 1 by alphanumerical labels A_1 -A_{13} (blue dots) and C_1-C_{15} (black dots) for VA and VC respectively. The characteristics of these two plants species are described by Biholong [5] and Farombi [6]. Samples of 2 kg of fresh collected in each site were oven-dried at a temperature of 87°C for 24 hours, then crushed and sieved over 2 mm mesh. The material obtained was kept in sealed Marinelli beakers for at least four weeks to allow a sufficient time for ^{238}U and ^{232}Th to attain a state of secular radioactive equilibrium with their corresponding progenies prior to gamma spectroscopy [7,8].

Analysis

The activity concentrations for ^{238}U, ^{232}Th and ^{40}K were determined using an HPGe gamma-ray spectrometer. This counting equipment was a Canberra vertical high-purity coaxial germanium (HPGe) crystal detector, model GC 2018-7500, serial number b87063 enclosed in a 100 mm thick lead shield and coupled to a Canberra Multichannel Analysing (MCA) computer system [9,10]. The energy and efficiency calibrations were done using a well calibrated standard sources supplied by the International Atomic Energy Agency (IAEA) [11].

The activity concentration of ^{40}K was determined directly by its γ–line of 1460.8 keV, while that of ^{238}U and ^{232}Th were estimated by measuring the γ-ray lines of 609.3 keV and 1120.3 keV for ^{214}Bi; 969.0 keV for ^{228}Ac and 583.0 keV for ^{208}Tl respectively. The ^{137}Cs was determined directly from its 661.6 keV gamma-emission. The leaves samples after attaining secular equilibrium were placed on the detector and counted for 36000s FIG. 2.

The activity concentration $A_{i,d}^{j}$ of the dry sample *i* for the plant species *j* is derived from Eq 1:

$$A_{i,d}^{j} = \frac{N_{c,i}^{j}}{I_{\gamma} \times E_{eff}\left(E_{\gamma}\right) \times m_{i,d}^{j}} \quad (1)$$

where I_{γ} is the total net counts under the selected photopeak of sample *i* for the plant species *j* and I_{γ} is the absolute gamma decay intensity for the specific energy photopeak; $E_{eff}\left(E_{\gamma}\right)$ is the absolute efficiency of the detector at this energy E_{γ} and $A_{i,f}^{j}$ is the mass in kg of the dry sample *i* for the plant species *j*. The activity concentration for fresh leaves $A_{i,f}^{j}$ is derived from Equation 2:

$$A_{i,f}^{j} = 0.1\, A_{i,d}^{j} \quad (2)$$

Where $A_{i,d}^{j}$ is the measured activity on dry weight basis for radionuclide *i* in the sample of the plant species *j* and 0.1 is the mean ratio of the dry to the fresh weight of the leaves for the *Vernonia type j*.

The radium equivalent

The radium equivalent activity (Ra_{eq}) expressed in Bq/kg, is a parameter used to evaluate the hazard associated with materials containing ^{238}U, ^{232}Th and ^{40}K. The radium equivalent of the activity concentrations for these three radionuclides were calculated from Equation 3 below, in order to compare the aggregated health risk due to the ingestion by populations of these three radionuclides via the consumption of VA and VC leaves:

$$Ra_{eq,f}^{j} = A_{U,f}^{j} + 1.43 A_{Th,f}^{j} + 0.077 A_{K,f}^{j} \quad (3)$$

The annual effective dose rates (D_y) for adults consuming fresh VA or VC leaves are derived from Equation 4:

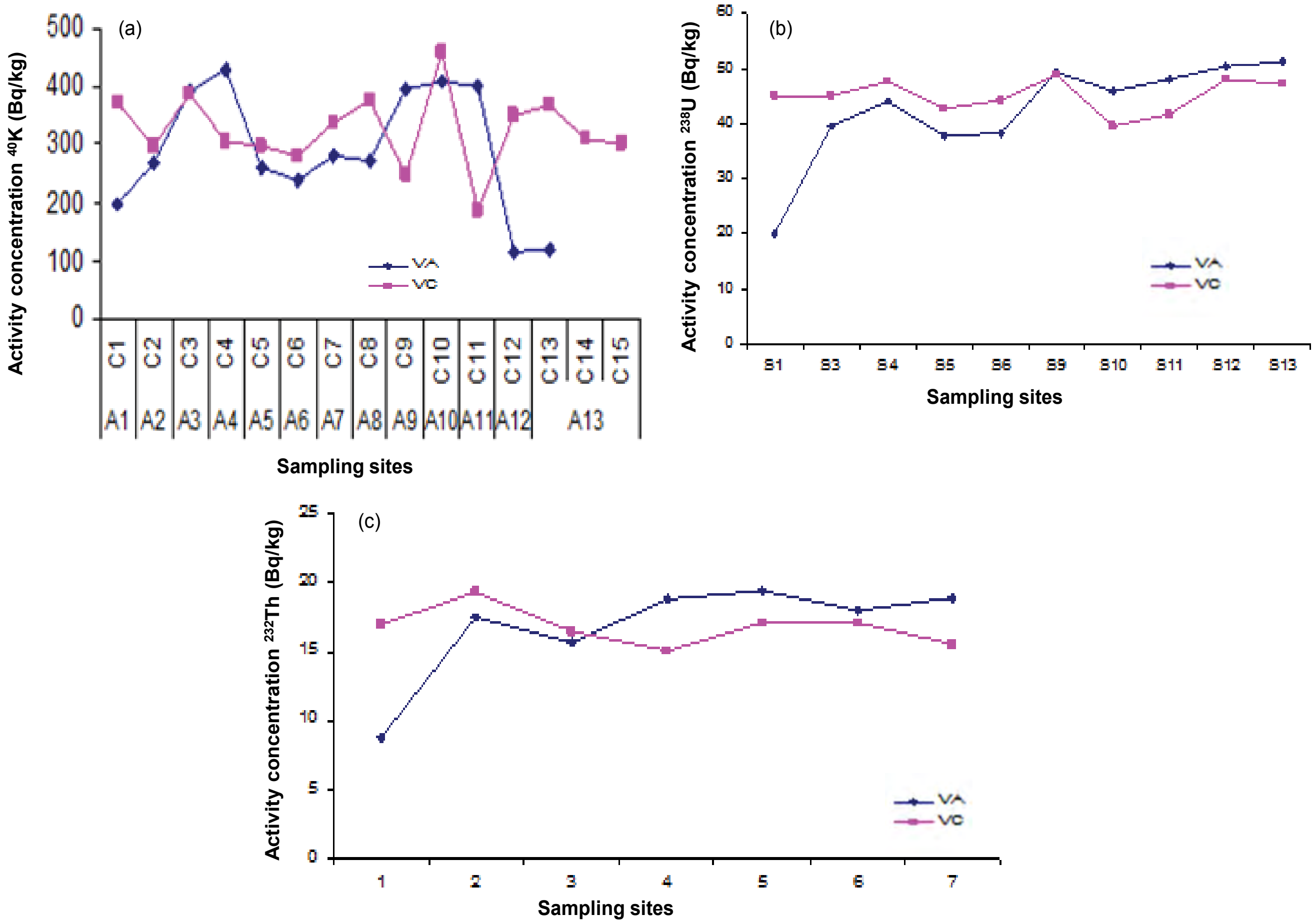

Figure 2: Variations of activity concentrations in VA and VC leaves: (a) 40K, (b) 238U and (c) 232Th.

$$D_j\left(\mu Sv.y^{-1}\right)=\sum_i w_i\times A_{i,f}^j\times Q_j=\left(w_K A_{K,f}^j+w_U A_{U,f}^j+w_{Th}A_{Th,f}^j\right)Q_j \tag{4}$$

Where Q_j is the activity concentration ($Bq.kg^{-1}$) on fresh weight basis for the radionuclide i in the Vernonia specie j while Q_j = 5.2 kg/year the annual consumption rate of fresh leaves per adult for vernonia type j, was obtained from the Cameroonian food consumption yearbook 2008. The different conversion factors w_i ($Sv. Bq^{-1}$) which are the committed effective dose per ingested unit intake for radionuclide i by an adult, were obtained from Misdaq [12] and ICPR-1996 [13] as follows: $w_K=6.2\times10^{-9}$, $w_U = 4.5\times10^{-8}$ and $w_{Th} = 2.3\times10^{-7}$ $Sv. Bq^{-1}$.

Then the internal hazard index H_{in}^j was calculated from eq.5:

$$H_{in}^j=\frac{2A_{Ra,f}^j}{370}+\frac{10A_{Th,f}^j}{370\times7}+\frac{10A_{K,f}^j}{370\times130}=\frac{A_{Ra,f}^j}{185}+\frac{A_{Th,f}^j}{259}+\frac{A_{K,f}^j}{4810}\leq1 \tag{5}$$

Where $A_{U,f}^j=A_{Ra,f}^j$, $A_{K,f}^j$ and $A_{K,f}^j$, are the activity concentrations on fresh weight basis for ^{238}U, ^{232}Th and ^{40}K. The value of this index must be less than unity to keep the radiation hazard insignificant.

Verification of compliance with dose limits

The concept commonly used in dosimetry for calculating the internal dose following an intake of a radionuclide i is the Committed Effective Dose Equivalent $(CEDE)_i$ calculated from Eq 6:

$$CEDE\left(H_{50}\right)_i=0.05\frac{\overline{A}_i^{j,an}}{ALI_i^{ing}}=0.05\frac{\overline{A}_{i,f}^j\times Q_j}{ALI_i^{ing}}\ (Sv) \tag{6}$$

Where $A_{i,f}^j\times Q_j$ and Q_j are as previously defined and where the annual limit intake for radionuclide i denoted ALI_i^{ing} (in Bq) obtained from Delacroix [14] are:

$ALI_{^{40}K}^{ing}$ =3.2 $10^6 Bq$, $ALI_{^{232}Th}^{ing}$ =5.9 $10^7 Bq$ and $ALI_{^{232}Th}^{ing}$ =5.9 $10^7 Bq$ = 2.6×10^4 Bq.

The annual body burden is the product ($A_i^{j,an} = A_{i,f}^j \times Q_j$) for an adult consuming per year Q_j kg of *Vernonia j*. The CEDE is a parameter used in dosimetry for occupational workers to verify the compliance of the internal dose received with the total body limit dose of 0.05 Sv (5 rems) recommended in the BSS [15]. The annual CEDE resulting from the consumption of VA or VC is derived from eq.7 hereafter, where *i = K, U* and *Th:*

$$\begin{aligned} CEDE\left(H_{50}\right)_{an,j} &= 0.05\sum_i\left(\frac{\overline{A}_i^{j,an}}{ALI_i^{ing}}\right) = 0.05Q_j\sum_i\left(\frac{\overline{A}_{i,f}^j}{ALI_i^{ing}}\right) \\ &= 0.05Q_j\left[\frac{\overline{A}_{K,f}^j}{ALI_K^{ing}} + \frac{\overline{A}_{U,f}^j}{ALI_U^{ing}} + \frac{\overline{A}_{Th,f}^j}{ALI_{Th}^{ing}}\right] \end{aligned} \quad (7)$$

Results and Discussions

The results obtained from gamma spectrometry analysis presented in Tables 1 and 2 have shown that ^{238}U, ^{232}Th and ^{40}K are bio accumulated in the leaves of vernonia harvested in southern Cameroon with respective average activity concentrations on dry weight basis of: 291.01 ± 30.14; 42.49 ± 15.64 and 16.66 ± 7.36 Bq/kg for VA and 325.55 ± 34.95; 45.03 ± 16.82 and 16.75 ± 7.17 Bq/kg for VC. These values reveal the slight discrepancies of the intake of these three types of radionuclides in the leaves of VA and VC**.** This trend is explained by the similarity of variation of the activity concentrations of each type of radionuclide in VA and VC leaves depicted in FIG. 2 throughout the sampling sites.

To compare the aggregated risk of radiation exposure due to the intake of these three radionuclides via the consumption of VA and VC leaves we have calculated and presented in Table 3, the radium equivalent, the committed effective dose equivalent (CEDE) and the internal hazard index (H_{in}). The average radium equivalent activities obtained for fresh leaves were 6.79 ± 2.85 Bq/Kg and 6.79 ± 2.97 Bq/Kg for VA and VC respectively. The CEDE of 21.56 and 22.08 (*µSv/y*) and the average values of internal hazard index (H_{in}) were found to be 0.027 and 0.026 for VA and VC leaves samples, respectively.

Conclusion

The low values of the activity concentrations of *Vernonia* leaves leading consequently to low values of the annual effective dose rates (*D*=3.94-4.12 µ Sv/year), the annual committed effective dose equivalent (CEDE=21.56 -22.08 µ Sv/year) and the internal hazard index (H_{in}=0.026) respectively for VA and VC indicate that populations of southern Cameroon consuming these legumes were not at this moment submitted to the hazard of radiation exposure associated to the ingestion of *Cameroonian vernonia.*

Table 1: Activity concentrations (Bq/Kg dry weight) for ^{238}U, ^{232}Th and ^{40}K in the leaves of *VA*.

Sampling code	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)
$_A1$	198,08 ± 30,67	19,94 ± 6,58	8,83 ± 2,92
A_2	269,08 ± 20,95	<MDA	<MDA
A_3	393,83 ± 33,79	39,59 ± 13,77	17,53 ± 6,10
A_4	428,88 ± 67,72	44,13 ± 16,02	<MDA
A_5	259,29 ± 25,06	37,89 ± 14,41	15,71 ± 6,20
A_6	240,05 ± 20,87	38,26 ± 10,21	<MDA
A_7	279,54 ± 33,41	<MDA	<MDA
A_8	272,28 ± 31,06	<MDA	18,85 ± 8,99
A_9	395,85 ± 38,27	49,36 ± 22,78	19,39 ± 9,74
A_{10}	408,42 ± 34,42	45,87 ± 12,04	18,02 ± 9
A_{11}	401,83 ± 33,48	48,13 ± 19,84	18,93 ± 10,23
A_{12}	114,71 ± 10,53	50,38 ± 20,01	<MDA
A_{13}	121,28 ± 11,61	51,35 ± 20,73	<MDA
Mean	**291.01 ± 30.14**	**42.49 ± 15.64**	**16.66 ± 7.36**

Table 2: Activity concentrations (Bq/Kg dry weight) for ^{238}U, ^{232}Th and ^{40}K in the leaves of *VC*.

Sampling Code	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)
C_1	371,97 ± 35,08	45,16 ± 19,21	17,01 ± 7,02
C_2	295,75 ± 37,16	45,09 ± 15,77	19,32 ± 8,24
C_3	389,11 ± 56,61	<MDA	<MDA
C_4	304,85 ± 33,20	<MDA	<MDA
C_5	296,83 ± 33,25	47,57 ± 17,82	16,5 ± 5,12
C_6	280,31 ± 30,69	42,76 ± 15,47	15,04 ± 4,28
C_7	340,41 ± 33,92	44,22 ± 15,27	<MDA
C_8	375,31 ± 31,16	48,89 ± 18,52	<MDA
C_9	247,12 ± 27,24	<MDA	17,09 ± 8,14
C_{10}	459,84 ± 47,64	<MDA	<MDA
C_{11}	187,01 ± 18,56	<MDA	17,11 ± 8,80
C_{12}	352,71 ± 28,92	39,57 ± 17,95	15,54 ± 7,68
C_{13}	369,49 ± 25,16	41,73 ± 13,59	16,37 ± 8,07
C_{14}	309,58 ± 43,49	48,15 ± 17,58	<MDA
C_{15}	303,02 ± 42,24	47,19 ± 17,01	<MDA
Mean	**325.55 ± 34.95**	**45.03 ± 16.82**	**16.75 ± 7.17**

Table 3: Radium equivalent activity (Ra_{eq}), annual effective dose (D_y), committed effective dose equivalent (CEDE) and internal hazard index (H_{in}) for VA and VC.

	Vernonia Amygdalina (VA)				*Vernonia calvoana (VC)*			
Sampling Sites	***Ra_{eq}* (Bq/kg)**	*D_y* (µSv/y)	CEDE (µSv/y)	***H_{in}***	***Ra_{eq}* (Bq/kg)**	*D_y* (µSv/y)	CEDE (µSv/y)	***H_{in}***
A_1	4.78	2.17	11.64	0.018	9.81	4,31	22.73	0.039
A_2	2.07	0.87	2.19	0.006	9.55	4,34	24.42	0.038
A_3	9.50	4.31	23.11	0.036	3.00	1,26	3.17	0.008
A_4	7.72	2.42	6.06	0.033	2.35	0,99	2.49	0.006
A_5	8.03	3.62	20.08	0.031	8.59	4,06	21.74	0.035
A_6	5.67	1.68	4.18	0.026	7.04	3,72	19.86	0.031
A_7	2.15	0.90	2.28	0.006	9.40	2,14	5.34	0.038
A_8	4.79	3.14	21.14	0.013	7.78	2,36	5.90	0.034
A_9	10.76	4.77	25.56	0.043	4.35	2,85	19.17	0.012
A_{10}	10.31	4.56	24.08	0.039	3.54	1,49	3.75	0.010
A_{11}	10.52	4.63	24.44	0.041	3.89	2,66	18.70	0.011
A_{12}	5.92	1.55	3.86	0.029	8.90	3,94	20.77	0.035
A_{13}	6.07	1.60	3.97	0.030	9.36	4,14	21.87	0.037
Mean	**6.79**	**3.94**	**21.56**	**0.027**	7.20	2,13	5.32	0.032
					7.05	2.09	5.21	0.032
					6.79	**4.12**	**22.08**	**0.026**

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