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Preliminary Study of Natural Radioactivity and Radiological Risk Assessment in Some Mineral Bottledwater Produced in Cameroon

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ABSTRACT

The aim of this work was to present an overview of natural radioactivity and its associated health risk in some mineral bottled water produced in Cameroon by means of using High-Purity Germanium based gamma spectrometry techniques. The water samples were firstly chemically treated by adding nitric acid and then pre-concentrated further by evaporating them up to a certain level. The water residues were transferred to small cylindrical containers and were kept sealed in order to ensure secular equilibrium between U-238, Th-232 and their daughter products. The calculated activity concentration for ^{226}Ra (U-series) and ^{228}Ra (Th-series) were used to estimate the annual effective doses for different age groups infants (age 1-2 years), children (age 7-12 years) and adults (age ≥17years) by taking into account the ingested dose conversion factors obtained from the International Commission on Radiological Protection (ICRP) as well as their yearly average bottled water consumption. The evaluated annual effective dose due to life-long consumption of water for different group ages where below the recommended values by WHO, IAEA and UNSCEAR. The paper presents the overview of the technique used and the summary of findings from this survey.

Keywords: *HPGe Detector, radioactivity, secular equilibrium, bottled water*

1. INTRODUCTION

The mineral water industry in Cameroon is growing and the production of mineral bottled water in the country has also increased over years. In additional to health benefits of drinking mineral water due to the presence of several nutrients the presence of dissolved mineral radioactivity derived from mineralized rocks is also concerned [1]**.** The geological sources of natural mineral water are known as aquifers, which may be of different types and they vary greatly in terms of depth, compositions and their permeability [2]. Depending on the origin of groundwater, it might have high amount of the primordial radionuclide or radioactive elements such as uranium, thorium, potassium, and their radioactive decay products [3,4]. The levels of activity concentrations of radionuclides according to nature in groundwater mainly depend on uranium and thorium bearing soil and rock mineral. Higher amount of activity concentrations of radionuclide in the environment are connected with risk to humans and high radiation damage such as kidney damage, leukaemia as well as cancer of bladder, kidney and lung [5]. Long term exposure to natural radionuclide in drinking water may therefore cause toxic effects to kidney, and can lead to cancer [1]. Measurement of natural radioactivity levels in drinking water is relevant in assessing the contribution to environmental radiation health hazard due to water consumption [4,7].

The natural radioactivity of bottled mineral waters has been a subject of numerous studies but some of them dealt with a part of natural radionuclide activity concentrations. For instance, the measurements of radium isotopes (^{228}Ra, ^{226}Ra), ^{222}Rn and ^{40}K concentrations in the bottled water for Poland, Austria, Northeastern Romania and Algeria were presented by Nguyen *et al* (2009), Wallner *et al* (2007), Elena Botezatu *et al* (2001) and Amrami (2002). In Cameroon, studies related to natural radioactivity monitoring in bottled mineral water have not been carried out so far. The aim of this work was to present a first overview of the radiological situation of all bottled mineral water produced and mostly consumed in Cameroon. This is achieved by first measuring the activity concentrations of ^{226}Ra (from U-238 series), ^{228}Ra (from Th-232 series) and ^{40}K in several samples of bottled mineral waters by means of using high-purity germanium based gamma spectrometry techniques. The annual effective dose for different age groups resulting from consumption of these waters due to their natural radionuclides contents (^{226}Ra, ^{228}Ra and ^{40}K) are estimated since it represents an accurate evaluation of radiation dose received by the population due to intake process. The radiation cancer and non-cancer risks due to intake are also evaluated from the adult annual effective dose.

2. EXPERIMENTAL PROCEDURE

2.1 Sampling and Sample Preparation

In order to measure the natural radioactivity in minerals bottled water produced in Cameroon, a number of six (06) different mineral bottled waters were purchased from different markets in the Central Region of Cameroon. The collected mineral bottled waters are mostly distributed and consumed by the population living in the majority cities of Cameroon.

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The method adopted for the determination of environmental natural radioactivity level required relatively large volume of water up to 20 L in total per each mineral bottled water type. Concentration has been carried out by gradual evaporation of each water sample in an oven at a temperature of 70^0 C and ending up with a volume of 125 ml residue out of 20L of initial mineral water. To prevent adherence of the radionuclide on container walls drops of nitric acid (HNO_3) were added into the sample. After being evaporated, the residues were then transferred into a thoroughly washed and dried 120 mL cylindrical container and hermetically sealed with a plastic tape to ensure air tightness and kept for 30 days to establish secular equilibrium between ^{238}U, ^{232}Th and their daughter products [9].

2.2 Experimental Setup

After the in-growth period, each water sample was subjected to a low background gamma-ray spectrometer consisting of Broad Energy Germanium Detector (BE6530) manufactured by Canberra Industries. As reported by the manufacture it has a resolution of 0.5 keV at 5.9 keV of ^{55}Fe, 0.75 keV at 122 keV of ^{57}Co and 2.2 keV at 1332 keV of ^{60}Co, respectively. To prevent high background counts due to external radioactive sources, with the intention to reduce the counting time and improve the detection limit, the detector is placed in a low-level Canberra Model 747 lead shield having a thickness of 10 cm. Furthermore, a Multiport II Multichannel Analyzer (MCA) was used to generate energy distributions of the radioactive samples. In order to obtain a statistically good computational net peak area, each sample was measured for 86400 s. The background has been evaluated before running the samples and it was measured for 172800 s.

The efficiency calibration files have been generated by means of using Canberra designed LabSOCS (Laboratory Sourceless Object Counting System) mathematical calibration software that incorporates the characterization information of a BEGe6530 high-pure germanium detector. When generating the efficiency calibration file, the LabSOCS calibration software is taking into account all parameters related to these measurements including dimensions of the counting geometries, physical and chemical compositions as well as the distance source-to-detector end-cap. To validate the accuracy of the LabSOCS mathematical efficiency calibration, some test have been conducted comparing the LabSOCS generated efficiency results with the empirical peak efficiency for a ^{60}Co-60 point source positioned at a distance of 25cm from the detector end-cap. The calculated results were in good agreement showing that differences between mathematical and empirical peak efficiencies are within 3-5%. To avoid error due to extrapolating the curve, the calibration curve is plotted in dual mode with cross-over energy at 165.85 keV (^{139}Ce). A fourth order polynomial equation was the best fit for the lower and higher energy curve and the fitting equations are the following:

For the energy less than 165.85 keV the best fit is:

$$\ln(Eff) = -1.193E02 + 1.025E02*\ln(E) - 3.399E01*\ln(E)^2 + 5.046E00*\ln(E)^3 - 2.831E-01*\ln(E)^4 \quad (1)$$

For the energy greater than 165.85 keV the best fit is:

$$\ln(Eff) = -4.851E01 + 3.057E02*\ln(E) - 7.331E00*\ln(E)^2 + 7.550E-01*\ln(E)^3 - 2.893E-02*\ln(E)^4 \quad (2)$$

Genie 2000, Gamma Acquisition V.3.2.1 and Gamma Analysis Software, V.3.2.3 was used for data acquisition and analysis [Genie^{TM} 2000 Spectroscopy Software]. Following the sample analysis, the specific activity concentration for each identified radionuclide has been reported in unit of Becquerel per liter (Bq/L). Furthermore the software is taking care to automatically check and perform the interference correction and calculate also the weighted mean for those radionuclide that emit more than one gamma ray. In addition, CANBERRA's patented Cascade Summing Correction algorithms allows us to correct the nuclide activities for losses or gains due to the presence of cascade summing effect related to these close counting geometries. Assuming secular equilibrium between ^{232}Th, ^{238}U and their decay products the activity of the radionuclides under study has been determined as following:

- ^{226}Ra concentration is calculated based on the gamma ray transitions of ^{214}Pb
- ^{228}Ra concentration is calculated based on the gamma ray transitions of ^{208}Tl and ^{228}Ac
- ^{40}K was directly determined using 1460.83 keV (10.7%) gamma-ray transition.

3. RADIOLOGICAL RISK ASSESSMENT

The annual effective dose (mSv/y) from ingestion of radionuclide in water samples was estimated on the basis of the mean activity concentrations of the radionuclides. This was done for different age categories. Assumptions on the rate of ingestion of bottled mineral water were made. In this paper, the intake rates based on a US national survey [11] are used; 0.6 L/day and 0.8 L/day for infants (age 1-2 year) and children (age 7-12 years) respectively, and 1.3 L/day for adults (age < 17 years). The recent dose conversion factors for ^{226}Ra, ^{228}Ra and ^{238}U ingestion reported by the International Commission on Radiological Protection [12] for three age categories: 1-2 year, 7-12 years and < 17 years were used for calculations. The annual effective dose of the water ($H_{ing}(W)$) was computed by the following formula [12]:

$$H_{Ing}(w) = \sum_{i=1}^{4} DCF_{Ing}(i) \times A_{Spi} \times I \quad (3)$$

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Where: $DCF_{ing}(i)$ - Dose conversion coefficients of a particular radionuclide *i-th* in Sv/Bq for a particular age categorie; Asp_i - Specific activity concentrations of radionuclide i-th in the water samples in Bq/L and I - Radionuclide intake in litres per year for each age category.

In addition to the estimated annual effective dose, the cancer and hereditary risk due to low dose without any threshold dose known as stochastic effect were estimated using the ICRP cancer risk assessment methodology [13]. Radiation risks to population result from exposure to low dose radiation are normally known as chronic risk of somatic or hereditary damage of human tissues, thus much emphasis is always placed on the reduction of these radiological risks to natural radiation. The nominal lifetime risk coefficient of fatal cancer recommended in the 2007 Recommendations of the ICRP for members of the public is 5.5 x 10^{-2} Sv^{-1}. For hereditary effects, the Detriment-adjusted nominal risk coefficient for the whole population as stated in ICRP (2007) for stochastic effects after exposure at low dose rates is estimated at 0.2 x $10^{-2}Sv^{-1}$. The risk to population was then estimated using the 2007 recommended risk coefficients in ICRP report and assumed 70 years lifetime of continuous exposure of population to low level radiation. According to the ICRP methodology:

Cancer Risk = Total Annual Effective Dose (Sv) x Cancer Risk Factor (1)

Hereditary Effects = Total Annual Effective Dose (Sv) x Hereditary Effect Factor (2)

4. RESULTS AND DISCUSSIONS

The activity concentration of ^{40}K, ^{226}Ra and ^{228}Ra in bottled mineral waters used as drinking water in Cameroon as well as the annual effective dose for different age categories are presented below on Table 1.

Table 1. Activity concentration (mBq/L) of ^{40}K, ^{226}Ra and ^{228}Ra in water samples and the annual effective dose (mSv/y) for different age categories

Sample ID	Sample name	Activity concentration (mBq/L)			Annual eff. dose (mSv/y)		
		K-40	Ra-226	Ra-228	Infants	Children	Adults
BW1	SUPERMONT	67.90±14.7	10.40±2.70	23.5±4.81	0.032	0.029	0.009
BW2	SEMME	153±67.10	29.3±4.63	10.2±9.07	0.019	0.018	0.007
BW3	MADIBA	156±67.30	38.2±6.01	121±8.99	0.159	0.147	0.045
BW4	LE FEBE	87.7±26.20	10.40±6.91	5.12±1.53	0.009	0.008	0.003
BW5	PURA	6.69±1.67	6.98±1.72	9.92±1.94	0.014	0.013	0.004
BW6	TANGUI	123±66.70	38.2±5.71	28.1±7.28	0.043	0.041	0.014
Average		**107.30**	**22.25**	**35.96**	**0.050**	**0.046**	**0.015**

As seen, the activity value of ^{40}K concentration varied from 6.69±1.67 to156±67.30 mBq/L with an average value of 107.1 mBq/L. The specific activity concentration of ^{226}Ra ranged from 6.98±1.72 to 38.2±5.71 mBq/L with an average value of 22.25 mBq/L. The activity concentration of ^{228}Ra varied between 5.12±1.53 and 28.10±7.28 mBq/L with an average value of 35.96 mBq/L. This variation in activity concentration of ^{40}K, ^{226}Ra and ^{228}Ra observed in these samples indicate that the origins of these waters are not the same and that they come from different depths and pass through different geological layers. Likewise, this irregular distribution of activity concentrations of the selected nuclides in these minerals water may depend on their contents in rocks or solid aquifers in the areas where the water is located and the residence time of waters/rocks-soils in contact as well. On the other hand, these variations in activity concentration of the selected radionuclides strongly depend on the physical and chemical properties of each water sample.

For comparison purposes, the results for ^{40}K, ^{226}Ra and ^{228}Ra in the present work and the reported values for other countries obtained from the literature of natural radioactivity in water are shown below on Table 2. When comparing these data, the activity concentration of ^{40}K, ^{226}Ra and ^{228}Ra in water samples reported by many authors varies from one country to another and some differences are clearly seen. For instance, the average activities concentrations of ^{226}Ra are found to occur over a wide range from 0.026 to 7.15±6.95 Bq/L. This wide range of ^{226}Ra concentration is in relation to the geological structure and to the characteristics of the areas.

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The average value of ^{40}K (0.107 Bq/L) in the present study is comparatively higher than the average value obtained in Italy (Milano) (0.054 Bq/L) by Roscuni (2003). In the same way the average concentration of ^{228}Ra (0.036 Bq/L) is comparatively lower than those published values selected from the literature. The values obtained in this study were thus compared favourably with the reported average values published by other authors selected from the worldwide investigation of natural radioactivity in different water types.

The activity concentrations of ^{40}K, ^{226}Ra and ^{228}Ra obtained in the present study were also compared with the guideline activity concentration values of the selected radionuclides in drinking water recommended by the WHO and other data obtained from IAEA. This comparison showed that our results were found to be below the guidelines values of the selected radionuclides.

Table 2: Comparison of mean concentrations of natural radionuclides in mineral water with previous measurements performed in different countries

Radionuclides	Country	Number of sample	Mean concentration (Bq/L)	References
^{40}K	Nigeria(Akure)	20	13.54±10.18[a]	[14]
	Nigeria	15	19.09[c]	[15]
	Algeria	08	1.0	[1]
	Bangladesh	30	4.16[c]	[9]
	Italy(Milano)	—	0.052[b]	[16]
	Cameroon	06	0.107	Present study
^{226}Ra	Nigeria(Akure)	20	7.15±6.95[a]	[14]
	Nigeria	15	7.75[c]	[15]
	Algeria	08	0.026	[1]
	Argentina	25	4.4	[17]
	Cameroon	06	0.022	Present study
^{228}Ra	Nigeria(Akure)	20	9.86±12.89[a]	[14]
	Nigeria	15	2.03[c]	[15]
	Cameroon	06	0.036	Present study

[a]well water, [b]tap water, [c]drinking sachet water

Table 3: Estimated Cancer Risks and Hereditary Effects of adult member of the public

Sample N0	sample name	fatality cancer risk to adult per year (10^{-7})	lifetime fatality cancer risk to adult (10^{-5})	severe hereditary effect in adult per year (10^{-8})	estimated lifetime hereditary effect in adult (10^{-6})
01	SUPERMONT	4.95	3.47	1.8	1.26
02	SEMME	3.85	2.70	1.4	0.98
03	MADIBA	24.75	17.33	9.0	6.3
04	LE FEBE	1.65	1.16	0.6	0.42
05	PURA	2.2	1.54	0.8	0.56
06	TANGUI	7.7	5.40	2.8	1.96

The annual effective dose due to ingestion of the monitored bottled water was estimated for different age groups including infants, children and adults; considering only the ingestion of ^{226}Ra and ^{228}Ra as shown in Table 1. Potassium (^{40}K) values were not considered during the calculation of the radiation dose because the absorption of the essential potassium element is under homeostatic control and takes place mainly from ingested food. Thus,

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the potassium contribution to the dose from ingestion in water, with its relatively low dose conversion factor (5×10^{-9}Sv/Bq) will be much less than of many other radionuclides. The calculated radiation dose for different age groups were ranged from 0.009 to 0.159 mSv/year for infants, between 0.008 and 0.147mSv/year for children, and varied from 0.003 to 0.045 mSv/year for adults with average values of 0.050, 0.046 and 0.015mSv/year, respectively. It can be seen that radiation dose received by infants are relatively higher than that received for children and adults.

Following the WHO, IAEA and UNSCEAR [18-20] recommendations, the recommended reference levels of the effective dose for infants, children and adults corresponding to one year consumption of drinking water are 0.26, 0.2 and 0.1mSv/year, respectively. The doses obtained in the present study are low than the recommended reference level and from radiation protection point of view, life-long consumption of these investigated bottled mineral waters may not cause any significant radiological health risk.

In order to evaluate the radiation risk due to ingestion of the selected radionuclides, the ICRP methodology was adopted in this study and the results shown in Table 3. The results of the cancer and non-cancer risk components were evaluated from the estimated annual effective dose of the monitored bottled water. The results of the evaluated fatal cancer risk to adult per year in each bottled water ranged from 1.65×10^{-7} (Le FEBE) to 24.75×10^{-7} (MADIBA) with the associated lifetime fatality cancer risk of 1.16×10^{-5} (LE FEBE) to 17.33×10^{-5} (MADIBA). The evaluated lifetime hereditary effect to adult per year varied from 0.6×10^{-8} (LE FEBE) to 9.0×10^{-8} (MADIBA) with the associated lifetime hereditary effect in adult of 0.42×10^{-6} (LE FEBE) to 6.3×10^{-6} (MADIBA).

This means that in the terms of the lifetime fatality cancer risk to adult approximately 17 out of 100,000 may suffer from some form of cancer fatality and for the lifetime hereditary effect approximately 6 out of 1,000,000 may suffer some hereditary effect. The negligible cancer fatality risk value recommended by USEPA is in the range of 1.0×10^{-6} to 1.0×10^{-4} (i.e 1 person out of 1 million or 10,000 suffering from some form of cancer fatality is considering trivial). Comparing the estimated results of the lifetime fatality cancer risk in the present study with the acceptable risk factor, it can be concluded that, all estimated results of the lifetime fatality risk in adult member of the Cameroonian's population due to ingestion of radionuclides in the monitored mineral bottled water are within the range of acceptable risk values recommended by USEPA.

5. CONCLUSIONS

This work presents the first detailed radioactivity monitoring campaign in some bottled mineral waters produced and available at local markets in Cameroon. The natural radioactivity level of ^{40}K, ^{226}Ra and ^{228}Ra have been measured in several bottled mineral water produced in Cameroon by using high-pure germanium based gamma spectroscopy techniques. The activity profiles of the radionuclides have clearly shown low activity concentrations across the monitored bottled mineral waters. This may explain the low level of natural radioactivity in bedrocks system in the areas where the water in coming from. The estimated lifetime fatality risk in adult member of the Cameroonian's population due to ingestion of radionuclide in the monitored mineral bottled water are within the range of acceptable risk values recommended by USEPA. Based on the results obtained from this survey, we can conclude that Cameroon bottled mineral waters that were subject of this study are suitable for human consumption and do not present any significant radiological risk related to its life-long consumption. Furthermore, the research findings showed an important role in helping us to gain additional knowledge related to both radioecology and environmental radioactivity monitoring issues.

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