

## The Annual Intake and the Annual Effective Dose due to $^{226}\text{Ra}$ and $^{228}\text{Ra}$ In Drinking Water in Yaoundé Area, Cameroon

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**Abstract:** The average concentrations of radionuclides of reservoir, tap and Spring water in the Yaoundé area, with a population of 3000 000 inhabitants were estimated from measurements of mean specific activity using a well calibrated Canberra NaI(Tl) and HPGe detector systems. Water samples from reservoirs and taps were collected during the dry and the rainy seasons, respectively in December 2002 and July 2003 and spring samples water were collected in August 2010. The radionuclides observed with regularity belonged to the series decay naturally occurring radionuclides headed by  $^{238}\text{U}$  and  $^{232}\text{Th}$ . The average annual intake for these populations is 3354 Bq/y for reservoir water, 3821 Bq/y for tap water and 1161 Bq/y for spring water. The annual effective dose received by Yaoundé adult population as a result of ingestion of this drinking water is respectively 0.925 mSv for the reservoir water, 1.052 mSv for the tap water and 0.255 mSv for spring water.

**Key words:** Natural radionuclide • Concentration • Annual intake • Annual effective dose

### INTRODUCTION

There are different forms (isotopes) of uranium but  $^{238}\text{U}$  is the predominant contributor to natural radioactivity. The average  $^{238}\text{U}$  contents in the earth's crust have been estimated to be 2.7 mg/kg and the concentration may be as high as 120 mg/kg in phosphate rocks [1]. Meanwhile, the average  $^{232}\text{Th}$  content of the earth's crust is about 9.6 mg/kg [2]. Enhanced levels of uranium, thorium and their fission products might be present in water in areas that are rich in natural radioactivity. As groundwater moves through fractures in the bedrock that contain these deposits radioactive minerals can leach out into the groundwater system. Uranium isotopes ( $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$ ) have a non-negligible radiotoxicity [3]. Several radionuclides coming from the radioactive decay chain starting from  $^{238}\text{U}$  and  $^{235}\text{U}$  are highly radiotoxic. The most radiotoxic and most important among them is radium, which is a known carcinogen and exists in several isotopic forms. The predominant radium isotopes in ground water are  $^{226}\text{Ra}$ , an alpha emitter with a half-life of 1600 years and  $^{228}\text{Ra}$ , a beta emitter with a half-life of 5.8 years [4]. When radium is

taken into the body, its metabolic behaviour is similar to that of calcium and an appreciable fraction being distributed almost uniformly in soft tissues [5].

The purpose of the present work was firstly to investigate the types and concentrations of natural radionuclides in the reservoir and tap water of Yaoundé town, capital city of Cameroon, and secondly to estimate the annual effective dose exposure of the population submitted to the consumption of this water.

**Sampling:** The study was conducted over the Yaoundé area which lies at the latitude of  $3^{\circ}52'\text{N}$  and longitude of  $11^{\circ}31'\text{E}$ , covering a total area of 297 km<sup>2</sup>, with an average altitude 740 m. The study area was partitioned into 17 units and water samples were collected on December 2002 and July 2003, corresponding respectively to the dry season and to the rainy season, in taps and reservoirs. 3 sampling locations were reservoirs namely Ngoaekelle, Mimbomane and Njoungolo. For instance, 5 samples locations were spring water collected in August 2010. The major bedrock types of the river Nyong where the water of these three reservoirs comes from include gneiss, pegmatite, pegmatite schist and undifferentiated schist

[6]. Others locations were taps whose water is used for drinking, washing clothes, cleaning of food, for irrigation and for various domestic uses. The sampling locations of tap water were chosen based on such factors as population density, hospital, educational institutions, etc. The water taps were first turned on at full capacity for several minutes to purge the plumbing system of any water which might have been there for some time. The taps were turned down to a low rate to reduce turbulence and, thus, reduce radon loss [7]. After the water samples were collected as mentioned above, they were transferred to 1 litre kegs prior to processing for  $\gamma$ -spectrometry analysis. The water reservoirs, which have been connected to a network of pipes that carry water into the residences, were equally collected using 1 litre plastic kegs.

All the samples of water were acidified with 11 M of ( $\text{H}_3\text{O}^+$ ,  $\text{Cl}^-$ ) at the rate of 10 ml per litre of sample as soon as possible after sampling to avoid absorption of radionuclides on to the walls of the containers as documented by the International Atomic Energy Agency [8]. Marinelli beakers previously washed, rinsed with a dilute sulphuric acid and dried to avoid contamination were filled with known volume of the various water samples and later firmly sealed for, at least, four weeks to ensure that no loss of radon occurs thereby ensuring a state of secular equilibrium to be reached between radium isotopes and their respective daughters. From each location, four samples were made from water collected.

**Instrumentation:** The first Gamma-counting equipment was a Canberra sodium iodide thallium activated NaI(Tl) crystal detector model GC2018-7500, serial number b 87063. This crystal used has an excellent energy resolution. The multichannel analyser (MCA) used for this work contains 8192 channels, each channel was capable of storing up to  $10^5$ - $10^6$  counts per second. The contents of the memory after a measurement can be recorded or pulse height spectrum [9]. The selected bias voltage of 4500 Vdc (dc: deviation current) used for NaI(Tl) detector employed for this work; the typical measurement time was 36000 seconds. The second Gamma counting system was a Canberra High Purity Germanium (HPGe) detector, model Gx3019-7500SL, serial number 11026235; the selected bias voltage of 4000 Vdc used for this crystal with a counting time of 86400 seconds. Because of the cosmic radiation that continuously bombards the earth's atmosphere and the existence of natural radioactivity in environment, radiation detectors records some background signal which varies with the

size and type of the detector as well as the extent of shield. Hence the knowledge of the net peak area (without the background) under the full-energy peak that appears in its spectrum is important to apply the peak efficiency data for any detector. In gamma-ray spectroscopy with NaI(Tl) and HPGe detectors, for instance, the pulse height scale must be calibrated in terms of absolute gamma-ray energy if various peaks in the spectrum are to be properly identified. Also, any measurement of absolute gamma-ray emission rates requires knowledge of the detector efficiency. Thus, the detector system has to be calibrated in terms of energy and absolute efficiency. The energy and efficiency calibrations were done using a well calibrated standard water source supplied by the International Atomic Energy Agency (IAEA), Vienna, Austria for the NaI(Tl) detector and supplied by Laboratory standard activity (LEA-Pierrelatte) for HPGe detector. The techniques used are well described elsewhere. The photopeaks observed with regularity in the water samples were identified to belong to the naturally occurring series decay radionuclides headed by  $^{238}\text{U}$  and  $^{232}\text{Th}$ . Other radionuclides, if present, appeared rather infrequently at low levels or occurred at levels below the maximum detectable limits (MDL), statistically determined at two-standard deviation analytical error.

The activity concentrations of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were indirectly obtained from the  $\gamma$ -rays emitted by their progenies which were in secular equilibrium with them.  $^{226}\text{Ra}$  concentration was determined by measuring the 609.3 keV  $\gamma$ -rays from  $^{214}\text{Bi}$ . The 583.0 keV  $\gamma$ -rays of  $^{208}\text{Tl}$  was used to determine that of  $^{228}\text{Ra}$ . The gamma spectroscopy analysis with NaI(Tl) crystal was carried out by a sophisticated spectra-analysis program, SAMPO 90 [10] which matched  $\gamma$ -energies at various energy levels to a library of possible isotopes. Another sophisticated spectra-analysis program named Genie 2000 was used for the HPGe detector. This data analysis routine subtracted a linear background distribution from the pulse-height spectra of the samples and the background in addition to the net background peak area being subtracted from the corresponding net peak area for a particular radionuclide. The activities of the radionuclides were calculated from the difference between net peak and net background areas, accumulation time, absolute peak efficiency, absolute  $\gamma$ -ray emission probability ( $\gamma$ -ray intensity) and the sample volume. Triplicate analyses were conducted on all the water samples to check on the reproducibility of results and the stability of the counting system. The overall uncertainty in the measured concentrations was estimated from the parameters contained in the above

mentioned relation, the calibration procedure, the peak area determination and the background.

**Determination of the Specific Activity and the Annual Effective Dose:** Each radionuclide activity per unit volume A, in each water sample was evaluated using the relation:

$$A = \frac{N}{\varepsilon \times I_y \times V \times t_c} \quad (1)$$

where

N: net peak area of the radionuclide of interest

$\varepsilon$ : efficiency of the detector for the energy  $E_y$

$I_y$ : intensity per decay for the energy  $E_y$

V: volume of the water sample

$t_c$ : total counting time in seconds

When analysing the total annual effective dose to the human population from natural sources, the dose received by ingestion of long-lived natural radionuclides must be considered. Effective doses resulting from the intake of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  may be determined directly from external measurements of their concentrations in the body or estimated from concentrations intake materials such as air, food and water. Intakes of the natural radionuclides  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  through water in Yaoundé taps and reservoirs were calculated. Assuming the volume of drinking water for adult to be 1 litre/day, these intakes of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  through reservoir and tap water in all locations are presented in table 2. The annual effective dose was calculated with the intake of individual radionuclide and ingestion dose coefficients ( $\text{Sv.Bq}^{-1}$ ) reported by the International Commission on Radiological Protection. In Yaoundé, we have two seasons, the dry season which covered 91 days and the rainy season which covered 274 days; the equations for calculating the effective dose ( $H_{\text{dry season}}$  and  $H_{\text{rainy season}}$ ) and the annual effective dose (H) per person are [11]:

$$H = \sum_i I_i \times 365.25 \times D_i \quad (2)$$

$$H_{\text{dry season}} = \sum_i I_d \times 91 \times D_i \quad (3)$$

$$H_{\text{rainy season}} = \sum_i I_r \times 274 \times D_i \quad (4)$$

$$H = \sum_i I_d \times 91 \times D_i + \sum_i I_r \times 274 \times D_i \quad (5)$$

Where,

$I_d$  and  $I_r$  are respectively the daily intakes of radionuclide i ( $\text{Bq.d}^{-1}$ ) in dry and rainy season.

The ingestion dose coefficient  $D_i$  for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  is respectively  $2.2 \times 10^{-7}$  and  $2.8 \times 10^{-7} \text{ Sv.Bq}^{-1}$ .

The annual intake ( $\text{Bq/y}$ ) can be express by:

$$A(\text{Bq/y}) = \sum I_i(\text{Bq/d}) \times 365.25 \quad (6)$$

## RESULTS AND DISCUSSIONS

Table 1 shows the summary of the  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations and table 2 shows the annual intake and the annual effective dose due to ingestion of sampled drinking water.

The mean concentration of  $^{226}\text{Ra}$  agreed with a range of values obtained by many investigators namely Buchli and Burkart [12] in drinking water: 10 - 43  $\text{BqL}^{-1}$ ; Mc Curdy and Russel [13]: 0.08 - 36.4  $\text{BqL}^{-1}$ , in imported bottled water and Dana [14]: 1.5 -124  $\text{BqL}^{-1}$  in public water supplies in North Carolina. The  $^{228}\text{Ra}$  concentrations recorded for this work fell within the wide range of values 0.05- 4.6  $\text{BqL}^{-1}$  quoted for the USA's imported bottled water by Mc Curdy and Russel [13]. The concentration values are relatively low during the rainy season; this could be due to the dilution effects of rain water since the river Nyong where reservoir and tap water studied comes. The lowest concentration was recorded in reservoir water due to treatment. The mean specific activity of uranium in

Table 1: Mean specific activity content ( $\text{BqL}^{-1}$ ) in drinking water in Yaoundé area

Type of Water sample	Number of Samples	$^{226}\text{Ra}$		$^{228}\text{Ra}$	
		Range	Mean	Range	Mean
Reservoir water(dry season)	12	6.2 -10.1	$8.7 \pm 3.5$	0.4 - 0.8	$0.6 \pm 0.2$
Reservoir water(rainy season)	12	6.1-11	$8.5 \pm 3.7$	0.6 - 0.7	$0.6 \pm 0.2$
Tap water (dry season)	36	8.4 -13.8	$11.4 \pm 3.7$	0.3 - 1.6	$1 \pm 0.3$
Tap water(rainy season)	36	4.3 -12.2	$9 \pm 3.5$	0.2 - 1.1	$0.7 \pm 0.2$
Spring water(rainy season)	10	1.7 - 5.2	$3.2 \pm 0.6$	Traces	

Table 2: The annual effective doses for the present investigation

Nuclide	Type of sample	I <sub>i</sub> (Bq.d <sup>-1</sup> ) / person	H (mSv y <sup>-1</sup> )
<sup>226</sup> Ra	Reservoir water	8.7 (dry season)	0.877
		8.5 (rainy season)	
	Tap water	11.4 (dry season)	0.991
		9 (rainy season)	
	Spring water	3.2 (rainy season)	0.255
<sup>228</sup> Ra	Reservoir water	0.6 (dry season)	0.048
		0.6 (rainy season)	
	Tap water	1 (dry season)	0.061
		0.7 (rainy season)	
	Spring water	-	-
	Spring water	-	-
Annual intake (Bq/y)	Reservoir water: 3354 Bq/y		
	Tap water: 3821 Bq/y		
	Spring water : 1161 Bq/y		
Annual effective dose for each sample type (mSv/y)	Reservoir water: 0.877 + 0.048 = 0.925 mSv/y		
	Tap water: 0.991 + 0.061 = 1.052 mSv/y		
	Spring water: 0.255 + 0 = 0.255 mSv/y		

this type of water samples is lower than the results ( $10.4 \pm 1.7$  BqL<sup>-1</sup>) of Tchokossa [15] in the reservoir water of Mukuro, in Nigeria. <sup>226</sup>Ra activity is not too different to the result ( $620 \pm 10$  m BqL<sup>-1</sup>) obtained by Hakam *et al.* [16] in the drinking water from Fez locality in Morocco. While they are higher than 0.20 - 135 pCiL<sup>-1</sup> equivalent to 0.007 - 0.05 BqL<sup>-1</sup>, obtained by Nour Khalifa [11] in tap water from Qena locality in Egypt. They are still within the range of 0.00 - 8.75 BqL<sup>-1</sup> reported by David *et al.* [13], quoted by Mc Curdy and Russel on 1981 for domestic bottled water marketed and consumed in USA.

The specific activity due to natural thorium is relatively low in all the water samples investigated; this is because <sup>238</sup>U is more mobile than <sup>232</sup>Th. Slight variation in the radioactivity content in water of the same type and from the same source can be observed in different locations and even worldwide, mainly due to potential changes occurring in the pipe during distribution, the oxidation state of the water, the concentration of suitable complexing agents which can increase the solubility of uranium or thorium [17].

The annual intake by the populations of Yaoundé area as a result of both <sup>226</sup>Ra and <sup>228</sup>Ra in those drinking waters is estimated to be for reservoir water 3354 Bq/y, for tap water 3821 Bq/y and for spring water 1161 Bq/y. The annual effective dose received by inhabitant of Yaoundé is 0.925 mSv/y for reservoir water, 1.052 mSv/y-for tap water and 0.255 mSv/y for spring water. According to ICRP recommendations [18] the limit effective dose for public exposure is 1 mSv.y<sup>-1</sup>. The doses obtained in our study are nearer this recommendation.

## CONCLUSION AND RECOMMENDATIONS

The results of the study has indicated that the average specific activity concentration of <sup>226</sup>Ra and <sup>228</sup>Ra in the reservoir and tap water in this area have consistent values than spring water and still in the range of those reported for many other countries in the world. This study has shown that the naturally occurring radionuclides in Yaoundé differ in quantity and from location to location. These observations demonstrate that the radionuclide concentrations are a function of the geology of the area and it could be greatly influenced by the water transportation, precipitation and other numerous factors. The annual effective dose received by Yaoundé adults as a result of ingestion of this drinking water are 0.925 mSv/y for reservoir water, 1.052 mSv/y for tap water and 0.255mSv/y for spring water. Knowing that they are nearer the limit of 1mSv.y<sup>-1</sup> fixed by ICRP on 1991, there is a need for regular monitoring the water quality in the country as a whole since high radiation doses as well as low radiation doses could induce serious health effects. People can then filter this water before consumption, because by filtrating, the radioactive substances that couldn't be dissolved have been eliminated.

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