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The annual effective dose due to natural radionuclides in the reservoir and tap water in Yaoundé area, Cameroon

R. Marie Lydie and R. Martin Nemba

Department of Inorganic Chemistry, University of Yaoundé 1, Cameroon.

Abstract

The average concentrations of radionuclides of reservoir and tap water in the Yaoundé area, with a population of 1.5 million inhabitants were estimated from measurements of mean specific activity using a well calibrated Canberra NaI(Tl) detector system. Water samples were collected from reservoirs and taps during the dry and the rainy seasons respectively in December 2002 and July 2003. The radionuclides observed with regularity belonged to the decay series naturally occurring radionuclides headed by ^{238}U and ^{232}Th as well as the non-series nuclide ^{40}K. The average specific activity values obtained for ^{40}K, ^{226}Ra and ^{228}Ra, respectively were for reservoir water 70 ± 11 BqL^{-1} ; 8.7 ± 3.5 BqL^{-1} ; 0.6 ± 0.2 BqL^{-1} during the dry season, and 50 ± 9 BqL^{-1} ; 8.5 ± 3.7 BqL^{-1} ; 0.6 ± 0.2 BqL^{-1} during the rainy season and for tap water, we have respectively 111 ± 17 BqL^{-1} ; 11.4 ± 3.7 BqL^{-1} ; 1 ± 0.3 BqL^{-1} during the dry season, and 51 ± 10 BqL^{-1} ; 9 ± 3.5 BqL^{-1} ; 0.7 ± 0.2 BqL^{-1} during the rainy season. 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 and 1.052 mSv for the tap water.

***Keywords**: Natural radionuclide, concentration, NaI(Tl) detector, Annual effective dose.*

1. Introduction

There are different forms (isotopes) of uranium but ^{238}U is the predominant contributor to natural radioactivity. The average ^{238}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 (Padam *et al.*, 1996). Meanwhile, the average ^{232}Th content of the earth's crust is about 9.6 mg/kg (Firestone *et al*., 1996). 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}U, ^{235}U and ^{234}U) have a non-negligible radiotoxicity (Who, 1978). Several radionuclides coming from the radioactive decay chain starting from ^{238}U and ^{235}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}Ra, an alpha emitter with a half-life of 1600 years, and ^{228}Ra, a beta emitter with a half-life of 5.8 years (Marovic *et al.*, 1996). 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 (Wreen *et al*., 1985).

The purpose of the present work is 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.

2. Materials and Methods

2.1 Sampling and Sampling Locations

The study was conducted over the Yaoundé area which lies at the latitude of 3°52'N and longitude of 11°31'E, covering a total area of 297 km^2, with an average altitude 740 m. The study area was partitioned into 12 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 which lie respectively at a latitude and a longitude of 3°51'33"N and 11°29'53"E 3°52'6"N and 11°33'20"E; 3°52'50"N and 11°31'33"E. 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 (Olivié-Lauquet *et al*., 2000). 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 (Watson, 1986). After the water samples were collected as mentioned above, they were transfered to 1 litre kegs prior to processing for γ-spectrometry analysis. The water reservoirs, which have been connected to a network of pipes that carry water into the residences, were equaly collected using 1 litre plastic kegs.

All the samples of water were acidified with 11 M of (H_3O^+, 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 (IAEA, 1986). Marinelli beakers of 1 litre volume capacity 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.

2.2 Instrumentation

The Gamma-counting equipment was a Canberra sodium iodide thallium activated NaI(Tl) crystal detector model GC2018-7500, serial number b 87063. The 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 (Knoll, 1988). 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. 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) detector, 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 NaI(Tl) 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. The techniques used are well described elsewhere. The photopeaks observed with regularity in the water samples were identified to belong to the naturally occuring series decay radionuclides headed by ^{238}U and ^{232}Th, and a non-series natural radionuclide, ^{40}K. 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}Ra and ^{228}Ra were obtained indirectly from the γ-rays emitted by their progenies which were in secular equilibrum with them while that of ^{40}K was estimated directly by its γ-line of 1460.8 keV. ^{226}Ra concentration was determined by measuring the 609.3 keV γ-rays from ^{214}Bi. The 583.0 keV γ-rays of ^{208}Tl was used to determine that of ^{228}Ra. The gamma spectroscopy analysis was carried out by a sophisticated spectra-analysis program, SAMPO 90 (Aarnio *et al.*, 1992) which matched γ-energies at various energy levels to a library of possible isotopes. This data analysis routine substracted a linear background distribution from the pulse-height spectra of both the sample 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 γ-ray emission probability (γ-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.

2.3 Determination of Activity Concentrations and the Annual Effective Dose

Each radionuclide concentration C, in each water sample was evaluated using the relation:

$$C = \frac{N\left(E_\gamma\right)}{\varepsilon\left(E_\gamma\right)\times I_\gamma \times V \times t_c} \quad (1)$$

where

$N\left(E_\gamma\right)$: net peak area of the radionuclide of interest

$\varepsilon\left(E_\gamma\right)$: efficiency of the detector for the energy E_γ

I_γ: intensity per decay for the energy E_γ

V: volume of the water sample

t_c: total counting time in second (36000 sec)

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}Ra and ^{228}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}Ra and ^{228}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}Ra and ^{228}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 ($Sv.Bq^{-1}$) 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 (Nour Khalifa, 2004):

$$H = \sum_i I_I \times 365 \times D_i \quad (2)$$

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

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

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

where

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

The ingestion dose coefficient D_i for ^{226}Ra and ^{228}Ra is respectively 2.2 x 10^{-7} and 2.8 × 10^{-7} $Sv.Bq^{-1}$ (ICRP, 1994).

Table 1. Mean specific activity content (BqL^{-1}) in reservoir and tap water in Yaoundé area.

Type of Water sample	Number of Samples	2.4 Radionuclides					
		^{40}K		^{226}Ra		^{228}Ra	
		Range	Mean	Range	Mean	Range	Mean
Reservoir water(dry season)	12	33.8 -114.6	70 ± 11	6.2 -10.1	8.7 ±3.5	0.4 - 0.8	0.6 ± 0.2
Reservoir water(rainy season)	12	23.3 - 83.7	50 ± 9	6.1-11	8.5 ±3.7	0.6 - 0.7	0.6 ± 0.2
Tap water (dry season)	36	74 -138.6	111 ± 17	8.4 -13.8	11.4 ± 3.7	0.3 - 1.6	1 ± 0.3
Tap water(rainy season)	36	23.3 – 85.8	51 ± 10	4.3 -12.2	9 ± 3.5	0.2 - 1.1	0.7 ± 0.2

Table 2. The annual effective doses for the present investigation.

Nuclide	Type of water	I_i ($Bq.d^{-1}$)/ person	H ($mSv\ y^{-1}$)
^{226}Ra	Reservoir	8.7 (dry season) 8.5 (rainy season)	0.877
	Tap	11.4 (dry season) 9 (rainy season)	0.991
^{228}Ra	Reservoir	0.6 (dry season) 0.6 (rainy season)	0.048
	Tap	1 (dry season) 0.7 (rainy season)	0.061

3. Results and Discussions

3.1 The Mean Concentrations of Different Radionuclides

Table 1 shows the summary of the ^{40}K, ^{226}Ra and ^{228}Ra concentrations in sampled reservoir and tap water collected during the dry and the rainy seasons.

The mean concentration of ^{226}Ra agreed with a range of values obtained by many investigators namely Buchli and Burkart (1989): 10 - 43 BqL^{-1} ; Mc Curdy and Russel (1981): 0.08 - 36.4 BqL^{-1}, in imported bottled water and Dana (1987): 1.5 -124 BqL^{-1} in public water supplies in North Carolina. The ^{228}Ra concentrations recorded for this work fell within the wide range of values 0.05- 4.6 BqL^{-1} quoted for the USA's imported bottled water (Mc Curdy and Russel 1981). The concentration values are relatively low during the rainy season; this could be due to the dilution effets 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 this type of water samples is lower than the results (10.4 ± 1.7 BqL^{-1}) of Tchokossa (1998) in the reservoir water of Mukuro, in Nigeria. ^{228}Ra activity is not too different to the result (620 ± 10 $mBqL^{-1}$) obtained by Hakam (2001) in the drinking water from Fez locality in Morocco. While they are higher than 0.20 - 135 $pCiL^{-1}$ equivalent to 0.007 - 0.05 BqL^{-1}, obtained by Nour Khalifa (2004) in tap water from Qena locality in Egypt. They are still within the range of 0.00 - 8.75 BqL^{-1} reported by David *et al.* (1981), quoted by Mc Curdy and Russel (1981) for domestic bottled water marketed and consumed in USA.

As a summary, the higher levels of ^{40}K observed in all water samples are a function of the geological formation of the area (Watson, 1986). ^{40}K is the principal naturally occurring source of internal radiation, despite its low isotopic abundance (AIEA, 1986). However, because it is an essential biologic element which is under close metabolic control, variations in dietary composition have little effect on the body content or on the radiation dose received (NCRP, 1967). The specific activity due to natural thorium is relatively low in all the water samples investigated ; this is because ^{238}U is more mobile than ^{232}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 (ICRP, 1994). The mineralogy of this area is dominated by orthoclase which generates more ^{40}K (Olivié-Lauquet *et al.*, 2000) through the following hydrolysis reactions (Tchokossa, 1998):

$$\underset{\text{orthoclase}}{2KAlSi_3O_8} + 2H^+_{aq} + 9H_2O \rightarrow \underset{\text{kaolinite}}{H_4Al_2Si_2O_{9(aq)}} + 4H_4SiO_{4(aq)} + 2K^+_{(aq)} \quad (1)$$

$$\underset{\text{orthoclase}}{3KAlSi_3O_8} + 2H^+_{aq} + 12H_2O \rightarrow \underset{\text{muscovite}}{KAl_3Si_3O_{10}(OH)_{2aq}} + 6H_4SiO_{4aq} + 2K^+_{(aq)} \quad (2)$$

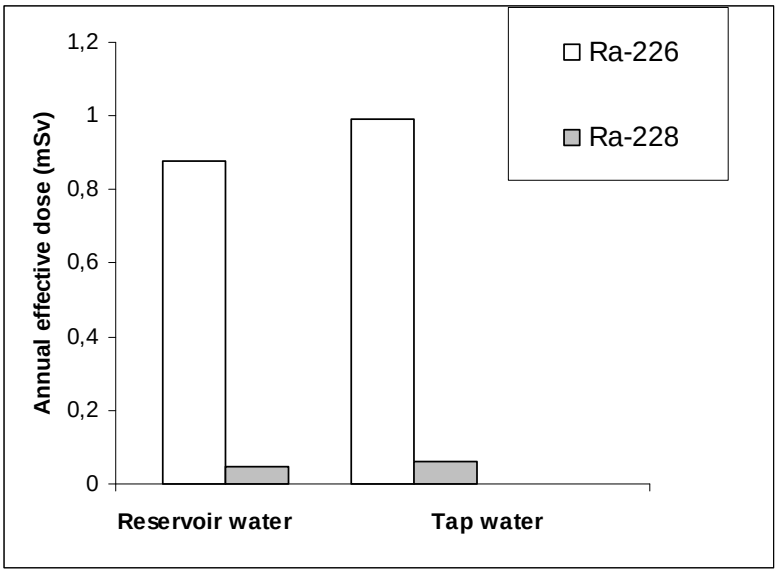

Figure 1. The annual effective dose received by Yaoundé adults as a result of the ^{226}Ra and ^{228}Ra ingestion.

Table 3. Average values of ^{226}Ra and ^{228}Ra concentration in units of BqL^{-1} in water samples of the investigation in comparison with other countries.

Country	Type of water	^{226}Ra	^{228}Ra	Reference
China	ground water	0.001- 1	-	Weihai zhuo et al (2001)
Denmark (Island of Bornholm)	tap water wells	- 0.55	- -	Ulback and klinder (1984)
China	ground water	Up to 4	0.01	Ziquiang et al (1988)
Portugal	mineral water	0.02	-	Bettencourt et al (1988)
Poland (Lodz)	underground water	0.01- 0.05	-	Jankowski et al (2000)
Egypt (Qena) Egypt (Safaga-Qusier)	drinking water ground water drinking water ground water	0.05 0.08 0.06 0.1	0.03 0.04 0.03 0.05	Nour Khalifa (2004)
USA	imported bottled water	-	0.05 - 4.6	Mc Curdy and Russel (1981)
USA	bottled Water marketed	-	0.0 - 8.7	David et al (1981)
Hungary	bottled mineral Water	0.1- 3	-	Somlai et al (2002)
Tunisia	springs	0.03 - 4	-	Labidi et al (2002)
Morocco	springs wells rivers cold springs lakes	0.009 - 4 0.001- 0.025 0.0008 - 0.005 0.002 - 0.01 0.0008 - 0.023	0.002 - 0.6 0.0006 - 0.02 0.0008 - 0.002 0.0007 - 0.003 0.0006 - 0.01	Hakam (2001)
North Carolina	imported bottled water	10 - 43	-	Mc Curdy (1981)
North Carolina	public water supplies	1.6 -124	-	Dana (1987)
Nigeria (Ile Ife)	well tap water dam borehole stream	3 - 13.5 10.5 - 13.6 10.3 - 10.6 7 - 16 6.4 - 7.7	0.34 - 4 2.3 - 2.6 2.3 - 2.7 2.3 - 4 3.4 - 3.7	Tchokossa (1998)
Cameroon (Yaoundé)	reservoir water (dry season) reservoir water (rainy season) tap Water (dry season) tap water (rainy season)	6.2 -10.1 6.1-11 8.4 - 13.8 4.3 -12.2	0.4 - 0.8 0.6 - 0.7 0.3-1.6 0.2 - 1.1	present work

3.2 Daily Intake Of ^{226}Ra And ^{228}Ra From Water Samples

In the area under investigation, the doses received by the ingestion of ^{226}Ra and ^{228}Ra are shown in figure 1. The annual effective dose received by the populations of Yaoundé area as a result of the investigation of both ^{226}Ra and ^{228}Ra in water is estimated to be 0.877 and 0.048 mSv for reservoir water and 0.991 and 0.061 mSv for tap water. According to ICRP recommendations (1991) the limit for public exposure should be expressed as an effective dose of 1 mSv.y^{-1}. The doses obtained in our study are nearer this recommendation.

4. Conclusion and Recommendations

The results of the study has indicated that the average specific activity concentration of ^{40}K , ^{226}Ra and ^{228}Ra in the reservoir and tap water in this area have consistent values with those reported for many other countries in the world (see table 3). 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 uranium and radium isotope activities measured in analyzed water samples are comparable to those reported in previous works throughout the world. The annual effective dose received by Yaoundé adults as a result of ingestion of this drinking water are 0.925 mSv for reservoir water and 1.052 mSv for tap water respectively. Knowing that it is nearer the limit of 1mSv.y^{-1} fixed by ICRP (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 could not be dissolved have been eliminated.

Following this work, the government of Cameroon adopted the ICRP's norms and several measures are in process of being taken to ameliorate the quality of these waters.

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Correspondence to: R. M. Nemba
(rnemba@yahoo.fr)