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LONG-LIVED NATURAL Ra ISOTOPES IN MINERAL, THERAPEUTHIC AND SPRING WATERS IN CAXAMBU, MINAS GERAIS

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ABSTRACT

In many countries all around the world, there is an increasing tendency to replace surface drinking water by commercially available bottled mineral water from different springs for consumption purposes. These practices involve an increased risk of finding higher activities of natural radionuclides in such waters, predominantly of the uranium-radium decay series. Naturally occurring radionuclides such as ^{238}U, ^{226}Ra, ^{222}Rn, ^{210}Po and ^{228}Ra are found frequently dissolved in water supplies and their concentrations vary over an extremely wide range. However, from the point of view of radiation hygiene, results of many worldwide surveys indicate that only ^{222}Rn, ^{226}Ra and ^{228}Ra have been found in concentrations that may be of health concern. ^{226}Ra and ^{228}Ra are the most radiotoxic isotopes of radium due to their long half-lives, 1,600 and 5.75 years, respectively. Since their chemical behavior is similar to that of calcium, radium isotopes tend to accumulate mostly in the skeleton after ingestion. For the purposes of this study, samples of mineral spring water were taken in Caxambu, a thermal resort located in the extreme south of Minas Gerais, 370 km south of Belo Horizonte. Caxambu is renowned by its waterpark, which has twelve sources of mineral water already being used in treatment of stomach, liver and kidney diseases, between others. ^{226}Ra activity concentrations varied from 83 to 3,599 mBq L^{-1}, the highest value determined at Venancio spring. Activity concentrations of ^{228}Ra ranged from 69 to 4,481 mBq L^{-1}. ^{228}Ra/ ^{226}Ra activity ratios varied from 0.079 to 4.2.

1. INTRODUCTION

Environmental radiation originates from a number of naturally occurring and man-made sources. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has estimated that the exposure to natural sources contributes more than 98% of the radiation dose to the population (excluding medical exposure) [1, 2]. There is only a very small contribution from nuclear power production and nuclear weapons testing. The global average human exposure from natural sources is 2.4 mSv y^{-1}. There are large local variations in this exposure depending on a number of factors, such as height above sea level, the amount and type of radionuclides in the soil, and the amount taken into the body in air, food and water. The contribution of drinking water to the total exposure is very small and is due largely to naturally occurring radionuclides in the uranium and thorium decay series. There are certain areas of the world, such as parts of the Kerala state in India and the Poços de Caldas plateau in Brazil, where levels of background radiation are relatively high. Levels of exposure for the general population in such areas may be up to 10 times higher than the average background level of 2.4 mSv. However, no deleterious health effects associated with this elevated radiation exposure have been detected [1, 2].

The presence and behavior of natural radionuclides in groundwater is of interest to health physicists, geochemists and uranium explorations. The natural radioactive substances in mineral and drinking water originates from the dissolution of gases and rock minerals and as a result of recoil nuclides. The overall quality of these groundwater sources depends largely upon the hydrological, physico-chemical conditions and geological characteristics of the aquifer. The geological setting strongly influences the occurrence of natural radionuclides in water. Their concentrations are variable and depend on the nature of the aquifer rock types and the prevailing lithology. Groundwater drawn from fractured aquifers in basement rocks can present considerable to high natural radioactivity levels [3-8].

Mineral water is a type of groundwater suitable for bottling, i.e. water which contains more than 1 g L^{-1} of total dissolved solids (TDS). Another kind of bottled groundwater may be medicinal water which can be either sweet (TDS < 1 g L^{-1}) or mineral, but it should contain components beneficial to the human body. There are also a lot of bottled groundwaters, which are neither mineral nor medicinal. Such water is also named mineral or just table water. In many countries all around the world, there is an increasing tendency to replace surface drinking water by commercially available bottled mineral water from different springs for consumption purposes. These practices involve an increased risk of finding higher activities of natural radionuclides in such waters, predominantly of the uranium-radium decay series [9]. It has been known that many mineral springs contain relatively high concentrations of radium and radon. Published values of ^{226}Ra in mineral waters ranged to more than 3.7 Bq L^{-1}, several times greater than values normally reported for public water supplies [9,10]. Many spring waters studied world-wide contain ^{226}Ra in amounts that exceed the USEPA maximum contaminant level of 185 mBq L^{-1}. Also several naturally occurring radionuclides such as ^{238}U, ^{226}Ra, ^{222}Rn, ^{210}Po and ^{228}Ra are found frequently dissolved in domestic water supplies and their concentrations vary over an extremely wide range [10]. Measurements of their levels in drinking water are therefore important to reduce the potential exposure of the public. However, from the point of view of radiation hygiene, results of many worldwide surveys indicate that only ^{222}Rn, ^{226}Ra and ^{228}Ra have been found in concentrations that may be of health concern [11-17]. ^{226}Ra and ^{228}Ra are the most radiotoxic isotopes of radium due to their long half-lives, 1,600 and 5.75 years, respectively. Since their chemical behavior is similar to that of calcium, radium isotopes tend to accumulate mostly in the skeleton when ingested. Although there is more ^{232}Th than ^{238}U in nature on an activity basis, there are geochemical factors that cause local concentrations of uranium, which often results in greater amounts of ^{226}Ra relative to ^{228}Ra in natural waters.

The World Health Organization (WHO) produces international requirements on water quality and human health in the form of guidelines that are used as the basis for regulation and standard setting in developing and developed countries world-wide. In the “Guidelines for Drinking Water Quality”, second edition, 1993 [18], the measurement of gross alpha and beta activities was recommended. These values were fixed to 0.1 and 1.0 Bq L^{-1} for alpha and beta activity, respectively. In third (current) edition [19], the guidance levels for the radionuclides originating from natural sources or discharged into the environment were presented and the total alpha activity value was raised to 0.5 Bq L^{-1}. In both cases the correct estimation of the total ingested dose from water for human consumption presupposes the parametric value of 0.1 mSv y^{-1}. This comprises 10% of the intervention exemption level recommended by the ICRP for dominant commodities (e.g., food and drinking water) for prolonged exposure situations, which is most relevant to long-term consumption of drinking water by the public [2, 19]. This value is accepted by most WHO Member States, the European Commission and

FAO. No deleterious radiological health effects are expected from consumption of drinking-water if the concentrations of radionuclides are below the guidance levels (equivalent to a committed effective dose below 0.1 mSv y^{-1}).

As far the total ingested dose evaluation is concerned, it cannot be assessed by a gross direct measurement, but it has to be calculated by summing the dose from each radionuclide present in the water, mainly alpha emitters, considered very dangerous radionuclides when ingested. These guidelines concern only tap and bottled water as "drinking waters"; contrary, there are no official limits for the bottled mineral waters and springs. But, nowadays, a non-negligible part of people consume bottled mineral water and soft drinks made from it [13], and the bottled mineral water has become an important element of the human diet. Consequently, the monitoring of the natural radionuclides concentration in mineral waters is attracting a lot of interest and is becoming more and more important [20, 21]. The Brazilian production of bottled mineral water raised from 1.8 10^9 L per year in 1997 to about 5.0 10^9 L per year in 2009 estimates [22] and the Southeast region is responsible for more than 65% of the total country production.

This paper presents activity concentrations of the long-lived radium isotopes, ^{226}Ra and ^{228}Ra, determined in 13 samples of mineral water from Caxambu Thermal Spa, Minas Gerais state, Brazil. Caxambu is a city that belongs to the "Waters Circuit", province classified according to the hydrogeology of mineral and medicinal waters, which includes also Cambuquira, Lambari and São Lourenço municipalities, where springs have attracted tourists for health treatment since 19th century. The tourism of health is an important segment of the tourism sector and it has been explored in several countries. In spite of Brazil to have a lot of thermal stations, academic studies on this theme almost do not exist, except papers of researchers of the area of health that focus the applied therapeutic techniques in the treatments based on hydrotherapy. In a previous study carried out by Godoy & Godoy [8] on the assessment of natural radioactivity levels in Brazilian groundwater, the highest values of U_{nat}, ^{226}Ra and ^{228}Ra were observed in bottled mineral waters produced in "Waters Circuit" region. In order to evaluate potential health hazards, doses due to ingestion of Caxambu Thermal Spa spring waters were estimated to assess the contribution of these radionuclides to public exposure from natural radioactivity.

2. MATERIAL AND METHODS

For the purposes of this study, samples of mineral spring water were taken in Caxambu, a thermal resort located in the extreme south of Minas Gerais, 370 km south of Belo Horizonte. Situated in an upland region with agreeable temperatures ranging between 15° C and 25° C, it was a resort much favored by the Brazilian imperial family in the 19th century. Caxambu means in Tupi-Guarani (Brazilian indian language) "the water that boils with bubbles". Caxambu is renowned by its waterpark, with an area of about 210 10^3 m^2, which has twelve sources of mineral water already being used in treatment of stomach, liver and kidney diseases, between others. Its population was estimated at 23,482 in 2004. The studied groundwater sources are exploited from sandstones, mudstones and conglomerates of Mesozoic-Cenozoic age. This groundwater represents different types of waters, mainly meteoric but also paleoinfiltrated, methamorphic and diagenetic. They are exploited at different depths and their ages (time of underground flow) vary considerably as does the CO_2 content.

The chemical composition of spring waters from Caxambu Thermal Spa is quite different. Beleza Spring for example, is hypotermal salt water rich in hydrocarbonate and iron, it has been indicated for stomach diseases; Dona Leopoldina Sping is a thermal water rich with calcium, magnesium, hydrocarbonate and sulphur; Conde D'Eau and Princesa Isabel Spring has alkaline sparkling waters, highly enriched in iron; Duque de Saxe Spring is also alkaline, hydrocarbonated and sulphurous; Dom Pedro Spring is lightly radioactive at source, and strongly sparkled; Mairink II and Marink III, despite being used for treatment and drinking purposes by the local residents, are commercially bottled under the brand name Caxambu.

For Ra isotopes, about 10 L of spring water were collected in the following locations using polyethylene containers: Mairink I Spring, Mairink II Spring, Mairink III Spring, Mairink III bathing water, Mairink III swimming pool, Dom Pedro Spring, Viotti Spring, Beleza Spring, Duque de Saxe Spring, Dona Leopoldina Spring, Conde d'Éau and Princesa Isabel Spring, Venâncio Spring and Ernestina Guedes Spring. These waters have been used on a large scale for medical and bathing purposes; for therapy, rehabilitation and recreation, while mineral waters are also used for dinking. All water samples were taken directly from springs or wells. The water samples pH were adjusted by addition of 10 mL of 65 % HNO_3, to prevent losses by sorption of the studied radionuclides onto the vessel walls. All sampling was performed in March 2011.

After the collection, samples were promptly taken to the "Laboratório de Radiometria Ambiental", from "Gerência de Metrologia das Radiações". A volume of 2 L of each water sample was concentrated to 1 L by evaporation and they were analysed in replicates. The radiochemical procedure adopted for ^{226}Ra and ^{228}Ra determination is described in detail by Oliveira *et al*. [21]. For these analyses, carriers of Ba^{2+} (20 mg) and Pb^{2+} (20 mg) were added to the samples as yield tracers, in the presence of 1 M citric acid solution. The radiochemical separation was accomplished by addition of 3 M H_2SO_4, with heating. Precipitate of barium, lead and radium sulphate was dissolved with nitrile triacetic acid/ NaOH, for separation of the decay product ^{210}Pb by complexation at pH 12.5- 13.0. At these conditions, the radium isotopes remain co-precipitated as a barium sulphate and the pH was adjusted to 4.5 – 5.0 with glacial acetic acid. The solution was then separated from the $Ba(Ra)SO_4$ precipitate by centrifugation. In order to purify the $Ba(Ra)SO_4$ precipitate obtained, it was dissolved with ethylene diamine tetracetic acid/ NH_4OH and re-precipitated by adjusting the pH of this solution to 4.5 – 5.0 with glacial acetic acid. The $Ba(Ra)SO_4$ precipitate was then filtered and the chemical yield determined gravimetrically. Considering that 20 mg of Ba^{2+} carrier was added to the water samples at the beginning of the radiochemical procedure, a final total recovery of $Ba(Ra)SO_4$ precipitate would be 34.8 mg. The chemical recovery of $Ba(Ra)SO_4$ precipitate for these set of samples was (90 ± 5)%.

The measurement of ^{226}Ra activity concentrations were carried out by counting the gross-alpha activity of a $Ba(Ra)SO_4$ precipitate using a low-background gas-flow proportional counter model Berthold LB 770, after sufficient time had elapsed for decay of ^{223}Ra and ^{224}Ra (21 days). The self-absorption factor for the four alpha-particles emitted in secular equilibrium with ^{226}Ra (^{226}Ra – 4.8 MeV; ^{222}Rn – 5.5 MeV; ^{218}Po – 6.0 MeV; ^{214}Po – 7.7 MeV) was determined by adding a ^{226}Ra standard solution, with activity concentrations ranging between 8.7 and 20.3 Bq to a deionized water sample. The value of the self-absorption factor obtained was 0.3001 ± 0.0065. The ^{228}Ra activity concentration was determined by gross-beta counting of the same precipitate, measuring the activity of its

daughter product ^{228}Ac, since it emits beta-particles of higher energy (1.2 and 2.1 MeV) in contrast to the lower energy of ^{228}Ra beta particles (40 keV). Lower limits of detection (LLD) for this method are 2.2 mBq L^{-1} for ^{226}Ra, for 100 minutes counting time, and 3.7 mBq L^{-1} for ^{228}Ra, for 400 minutes counting time, at the 95% confidence level [21]. The overall uncertainties of these measurements were below 10%.

The methodology was validated through the participation in a proficiency test organized by the Analytical Quality Control Services (AQCS) of the International Atomic Energy Agency (IAEA), called the "Interlaboratory Study on Determination of Radium and Uranium Radionuclides in Water", in January 2003. A total of six water samples (3 natural and 3 synthetic) were diluted with Milli-Q purified water. These samples were measured for ^{226}Ra using the co-precipitation technique described previously. Uncertainties were lower than 5%. The final evaluation of our results reported in this intercomparison indicated they were in a good agreement with IAEA reference values and were not biased by a systematic error, both with low and high ^{226}Ra activities.

During the period of this investigation, the methodology was checked experimentally by participating in the rotinery Quality Assurance Programs, organized in Brazil by "Instituto de Radioproteção e Dosimetria – IRD" (Brazilian Nuclear Energy Commission).

3. RESULTS AND DISCUSSION

The ^{226}Ra and ^{228}Ra activity concentrations determined in 13 samples of spring waters available at Caxambu Thermal Spa, as well as the ^{228}Ra/ ^{226}Ra activity ratios are presented in Table 1. Arithmetic mean activities ranged from 83 mBq L^{-1} to 3599 mBq L^{-1} and from 69 mBq L^{-1} to 4481 mBq L^{-1} for ^{226}Ra and ^{228}Ra, respectively. The highest ^{226}Ra activity was found in Venancio Spring, while the maximum ^{228}Ra activity value was determined in Ernestina Guedes. ^{228}Ra/ ^{226}Ra activity ratios varied from 0.079 (Conde D'Eau and Princesa Isabel Spring) to 4.2 (Mairink II Spring). In Brazil, there is a higher natural abundance of Th over U in rocks and soils, and where there is no secondary enrichment of U, ^{228}Ra is normally the dominant isotope found in water. ^{228}Ra/ ^{226}Ra activity ratios above unit was found in 7 of the mineral springs studied in Caxambu Thermal Spa and reinforces the importance of ^{228}Ra in ingestion dose assessments. The increased value of ^{228}Ra/^{226}Ra activity ratio observed in Mairink II Spring can indicate the presence of monazite in the aquifer lithology. Besides that, in the natural environment the solubility of radium is much higher than thorium, especially when higher concentrations of carbonate are detected in water, as in the case of those mineral springs from Caxambu.

The activities of ^{226}Ra and ^{228}Ra determined in mineral springs of Caxambu Thermal Spa are much higher than the values usually determined in drinking water supplies of São Paulo State [21]. However, ^{226}Ra values are of the same order of magnitude as those measured in thermal and mineral waters in the Republic of Croatia [24].

The results presented in Table 1 showed that maximum contaminant level of 185 mBq L^{-1} recommended by USEPA [10] for ^{226}Ra in drinking water was exceeded in 5 samples (Beleza, Duque de Saxe, Conde D'Eau and Princesa Isabel, Venancio and Ernestina Guedes). A conservative approach was used to assess the committed effective dose received by the consumers of these waters. Considering the recommended radioactivity standards for

drinking water established by Brazilian authority "Ministério da Saúde" [23], the gross-alpha activity screening level of 0.1 Bq L^{-1} was exceeded by ^{226}Ra in 11 samples (only Mairink III Spring presented ^{226}Ra activities lower than the gross-alpha limit). In the case of gross-beta recommended radioactivity screening level of 1.0 Bq L^{-1}, ^{228}Ra levels determined in Caxambu Thermal Spa spring waters was exceeded in 3 samples (Beleza, Venancio and Ernestina Guedes).

Table 1. ^{226}Ra and ^{228}Ra arithmethic mean concentrations (in mBq L^{-1} ± standard deviation) in mineral spring waters studied in Caxambu Thermal Spa, Minas Gerais, March 2011.

Sample	^{226}Ra	^{228}Ra	$^{228}Ra/^{226}Ra$
Mairink III bathing water	122 ± 5	284 ± 12	2.3
Mairink III swimming pool	92 ± 1	198 ± 21	2.2
Dom Pedro Spring	163 ± 1	111 ± 1	0.68
Viotti Spring	112 ± 10	69 ± 7	0.53
Beleza Spring	2897 ± 121	2361 ± 95	0.81
Duque de Saxe Spring	259 ± 11	406 ± 26	1.6
Dona Leopoldina Spring	175 ± 22	93 ± 26	0.53
Conde D'Eau and Princesa Isabel Spring	975 ± 12	77 ± 12	0.079
Venâncio Spring	3599 ± 147	1860 ± 120	0.52
Mairink I Spring	110 ± 27	280 ± 27	2.6
Mairink II Spring	140 ± 19	593 ± 13	4.2
Mairink III Spring	83 ± 7	232 ± 26	2.8
Ernestina Guedes Spring	3072 ± 115	4481 ± 129	1.5

LLD (lower limit of detection): ^{226}Ra = 2.2 mBq L^{-1}; ^{228}Ra = 3.7 mBq L^{-1}.

Based upon the arithmetic mean concentrations of ^{226}Ra and ^{228}Ra obtained from all 13 spring water samples studied in Caxambu Thermal Spa, the committed effective doses were evaluated. These results are shown in Table 2. Effective doses were estimated by taking into consideration a daily consumption rate of 2 L [18, 19]. The dose coefficients for ingestion of radionuclides by adult members of the public used in these calculations (in mSv Bq^{-1}) were taken from WHO [19]. The corresponding value for ^{226}Ra and ^{228}Ra are $2.8x10^{-4}$ mSv Bq^{-1} and $6.6x10^{-4}$ mSv Bq^{-1}, respectively.

It must be pointed out here that the committed effective doses obtained were compared with the reference data for acceptable drinking water. In estimating the radium dose contributed from the consumed spring waters, it has to be understood that these waters do not constitute a part of the continuous drinking water supply, but are applied as medicinal waters for definite periods and given amounts. Usually, thermal or mineral water drinking therapy may last 1 to

4 weeks. Also, Ra concentrations in springs which are commercially bottled (Mairink II and Mairink III springs) can be of particular interest in this evaluation, because the consumption of this water category has never been controlled in Brazil and its average intake is difficult to be properly estimated.

Table 2. Committed effective doses (mSv y^{-1}) due to the comsumption of ^{226}Ra and ^{228}Ra in mineral spring waters studied in Caxambu Thermal Spa.

Sample	^{226}Ra H_e	^{228}Ra H_e
Mairink III bathing water	2.5×10^{-2}	1.4×10^{-1}
Mairink III swimming pool	1.9×10^{-2}	9.5×10^{-2}
Dom Pedro Spring	3.3×10^{-2}	5.4×10^{-2}
Viotti Spring	2.3×10^{-2}	3.3×10^{-2}
Beleza Spring	5.9×10^{-1}	1.1
Duque de Saxe Spring	5.3×10^{-2}	2.0×10^{-1}
Dona Leopoldina Spring	3.6×10^{-2}	4.5×10^{-2}
Conde D'Eau and Princesa Isabel Spring	2.0×10^{-1}	3.7×10^{-2}
Venâncio Spring	7.4×10^{-1}	9.0×10^{-1}
Mairink I Spring	2.3×10^{-2}	1.4×10^{-1}
Mairink II Spring	2.9×10^{-2}	2.9×10^{-1}
Mairink III Spring	1.7×10^{-2}	1.1×10^{-1}
Ernestina Guedes Spring	6.3×10^{-1}	2.2

Note: to reflect the persistence of radionuclides in the body once ingested, the committed effective dose is a measure of the total effective dose received over a lifetime (70 years) following intake of a radionuclide (internal exposure). The annual ingested volume of water was assumed to be 730 L.

The highest values of the committed effective doses per year obtained in this work were 7.4×10^{-1} mSv y^{-1} for ^{226}Ra (Venancio Spring) and 2.2 mSv y^{-1} for ^{228}Ra (Ernestina Guedes Spring). In the case of ^{226}Ra, the value obtained represents less than 31% of the average effective dose attributable annually from natural background radiation. However, in the case of ^{228}Ra the annual committed effective dose limit recommended by ICRP (considering all sources of radiation exposure for the world population) [2] was exceeded not only in Ernestina Guedes Spring, but also in Beleza Spring.

In such situations, when either of the screening levels or recommended dose values had been exceeded, the WHO guidelines [18, 19] suggest the identification of specific radionuclides and a calculation of committed effective dose for each radionuclide, as it was performed in Table 2 above. Besides that, if the following additive formula is satisfied, no further action is required:

$$\sum_i \frac{C_i}{GL_i} \leq 1 \quad (1)$$

Where:
C_i = the measured activity concentration of radionuclide i in Bq L^{-1}, and
GL_i = the guidance level value [18, 19] of radionuclide I that, at an intake of 2 L day^{-1} for 1 year, will result in a committed effective dose of 0.1 mSv y^{-1}.
The corresponding guidance levels for ^{226}Ra and ^{228}Ra in drinking-water are 1 Bq L^{-1} and 0.1 Bq L^{-1}, respectively.

This sum exceeded the unit for all spring waters studied in Caxambu Thermal Spa, with exception of Viotti Spring. That means where the sum exceeded unit for a single sample, the recommended dose level of 0.1 mSv would be achieved only if the exposure to the same measured concentrations were to continue for a full year. Hence, such a sample does not itself imply that the water is unsuitable for consumption, but should be regarded as an indication that further investigation, including additional sampling is needed.

According to ICRP recommendations [2] the annual committed effective dose limit for public exposure is 1 mSv y^{-1}. Although some values shown in Table 2 were over this recommended limit, it was concluded that doses resulting from the short-term and temporary exposures to ^{226}Ra and ^{228}Ra from Caxambu thermal and mineral waters are not subject to the prescribed standard.

4. CONCLUSIONS

Radiation exposure through drinking-water can result from the presence of naturally occurring radioactive species (e.g., radionuclides of the thorium and uranium decay series in water sources), in particular ^{226}Ra/ ^{228}Ra and few others. The contribution of drinking-water to the total exposure is typically small. However, there is evidence from both human and animal studies in the current literature that radiation exposure at low to moderate doses may increase the long-term incidence of cancer [1,2, 18, 19].

The monitoring of the natural radionuclides concentration in mineral waters is receiving interest world-wide, because there is an increasing trend in to replace surface drinking water by commercially available bottled mineral water from different springs for consumption purposes and these practices involve an increased risk of finding higher activities of natural radionuclides in such waters. Besides that, it is well known that many thermal and mineral spring waters contain relatively high concentrations of ^{226}Ra and in several locations these springs have been exploited for its supposed curative powers. Therefore, the aim of this study was to investigate the natural distribution of long-lived Ra isotopes in thermal and mineral springs of Caxambu Thermal Spa, which have been used for bathing and drinking, as well as recreation.

It was observed increased ^{228}Ra activity concentrations over ^{226}Ra in mineral spring waters of Caxambu Thermal Spa. Higher levels of HCO_3^- and CO_3^{2-} in these waters can result in increased solubility of radium isotopes and may play an important role for the fate of ^{228}Ra and its equilibrium distribution between solid and liquid phases.

The maximum committed effective doses per year obtained in this work were 7.4 x 10^{-1} mSv y^{-1} for ^{226}Ra (Venancio Spring) and 2.2 mSv y^{-1} for ^{228}Ra (Ernestina Guedes Spring). Considering the case of ^{226}Ra intake via mineral water, the dose obtained represents less than 31% of the average effective dose attributable annually from natural background radiation. However, in the case of ^{228}Ra the annual committed effective dose limit recommended by ICRP (considering all sources of radiation exposure for the world population) was exceeded not only in Ernestina Guedes Spring, but also in Beleza Spring.

The present study has also pointed out that further studies are needed in order to better understand the seasonal variation of natural radionuclides in Caxambu spring waters related to geochemical and hydro-dynamical processes.

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