NATURAL RADIONUCLIDES IN MUD AND SPRING WATER USED IN THERMAL THERAPY IN A BRAZILIAN SPA

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ABSTRACT

This paper reports a study concerning the determination of natural radionuclides (^{40}K, ^{232}Th, ^{226}Ra, ^{228}Ra and ^{222}Rn) in samples of radioactive spring water, sulphurous mineral water, mud and indoor air collected at Termas de Araxá (MG), Brazil. This spa is located in one of the highest natural radioactive regions of Brazil.

The ^{226}Ra and ^{228}Ra activities in spring waters were measured by gross alpha and beta counting of a $Ba(Ra)SO_4$ precipitate, respectively, 21 days after the radiochemical separation. Both measurements were carried out in a low-background gas-flow proportional counter. The ^{222}Rn concentrations in spring waters were determined by liquid scintillation method. The indoor levels of ^{222}Rn were determined using a portable detector. Activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in mud samples were determined by gamma spectrometry with a 15% efficiency hyperpure germanium detector.

In the spring waters used in thermal treatments, high levels of dissolved radon up to 657 Bq/L were found. The activity concentrations of ^{226}Ra and ^{228}Ra in the same waters varied from 54 to 98 mBq/L and from 87 to 148 mBq/L, respectively. Bathing mud measurements showed activity concentrations from 0.7 to 1.0 Bq/kg for ^{232}Th, from < 0.3 to 1.0 Bq/kg for ^{226}Ra and from < 2.3 to 11 Bq/kg for ^{40}K, respectively. ^{222}Rn indoor airborne activities up to 3,153 Bq/m^3 were observed at the swimming pool room.

The results obtained were used to assess the natural radiation exposure to patients and personnel. Doses up to $2.3x10^{-1}$ mSv/y were obtained for internal and external irradiation for the patients, while doses up to 0.9 mSv/y were determined for the personnel occupationally exposed.

INTRODUCTION

Several surveys on natural radioactivity occurrence in spring waters and thermal establishments around the world have been performed with the aim of evaluate the relative importance of these radionuclides in the exposure of the local patients and the medical staff to radiation (Andrews and Wood, 1974; Kobal *et al.*, 1979; Uzunov *et al.*, 1981; Morinaga *et al.*, 1984; Kobal and Fedina, 1987; Doretti *et al.*, 1992; Trabidou *et al.*, 1995; Sanchez *et al.*, 1995; Marinovic *et al.*, 1996). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has estimated that exposure to natural sources contributes to more than 98% of the radiation dose to population and the global average human exposure from natural sources is 2.4 mSv/year (UNSCEAR, 1993).

This paper is concerned with the determination of natural radionuclides in a thermal spa named Termas de Araxá, in the State of Minas Gerais, Brazil. Araxá, located about 680 km from São Paulo City, is one of the highest natural radioactive regions of Brazil. This region belongs to an alkaline carbonatic massif named Barreiro Complex, which has roughly a circular shape with 4.5 km of diameter. In the Barreiro Complex, several minerals rich in uranium and thorium are found; weathered mineral deposits of economic interest are barium-pirochlore, phosphate (apatite) and barite (CBMM, 1984).

The mineral waters from Termas de Araxá have been used on a large scale for therapy, rehabilitation and recreation. Besides thermal bathing treatments for several diseases of the locomotory apparatus and dermathologic problems, these waters have been employed in treatments based on the ingestion of mineral waters. In the case of ingestion practices and bathing, two springs called Dona Beja and Andrade Júnior are used by the local residents and by the tourists for therapeutic purposes. Dona Beja's spring water is classified as radioactive regarding the ^{222}Rn levels according to the Brazilian standards (DNPM, 1945) and its chemical composition is oligomineral, although

high barium contents has been observed in this water due to the presence of barite minerals in the aquifer. Andrade Júnior's spring water is also radioactive following ^{222}Rn activity concentration and its chemical composition is alkaline, presenting high levels of H_2S and HCO_3^- dissolved species.

The radiation dose to the patients depends on the types of thermal treatment used in Termas de Araxá. Usually, the therapeutic cycle comprises 21 days, including mineral water bathing (Perola bath), mud bathing, drinking water and physiotherapy in a radioactive swimming pool, all those practices supervised by the medical staff. Sometimes, the collective swimming pool work as an emanatorium. The two spring waters used in the practices of drinking and bathing are the same as described above (Dona Beja and Andrade Júnior). These springs present high levels of dissolved ^{222}Rn, which could become a source of irradiation of the personnel working inside the confined rooms such as thermal baths.

This study aims, therefore, to determine the activity concentrations of ^{40}K, ^{232}Th, ^{226}Ra, ^{228}Ra and ^{222}Rn in radioactive spring waters, sulphurous mineral water, mud and air samples collected at Termas de Araxá and thus evaluate the internal and external exposure of the patients, local residents and the permanent personnel.

MATERIAL AND METHODS

For ^{226}Ra and ^{228}Ra determinations in radioactive and sulphurous mineral waters used in bath treatments, a volume of 10 L of water sample were collected directly from the thermal bath named "Perola", in polyethylene bottles. In the case of drinking water practices, 10 L of mineral water were collected at Dona Beja and at Andrade Júnior springs, using the same bottles. These water samples were acidified with 20 mL of HNO_3 65% in order to get a final pH lower than 1.5 and prevent losses by sorption of the radionuclides in the vessels. The samples were pre-concentrated by heating to reduce the volume from 3L to 1L, each analysis was carried out in duplicate. For the radiochemical separation of ^{226}Ra and ^{228}Ra, carriers of stable barium (20mg) and lead (20 mg) were added to the water sample in the presence of 1M citric acid and nitric acid solutions. The water sample was neutralised and radium was co-precipitated as $Ba,Pb(Ra)SO_4$ by adding 3M H_2SO_4. The precipitate was dissolved with alkaline ethilenediaminetetraacetic acid (EDTA). When the pH is adjusted to 4.5 with glacial acetic acid, $Ba(Ra)SO_4$ is reprecipitated, while interfering elements remain in the solution. The $Ba(Ra)SO_4$ precipitate was filtered and the chemical yield determined gravimetrically. The ^{226}Ra and ^{228}Ra were determined by gross alpha and beta measurement of $Ba(Ra)SO_4$ precipitate, respectively, after 21 days. Both measurements were carried out in a low-background gas-flow proportional counter (Berthold LB 770). The typical lower limits of detection for this method were 2.2 mBq/L for ^{226}Ra and 3.7 mBq/L for ^{228}Ra (Oliveira, 1993).

The ^{222}Rn activity concentration in water samples was determined by liquid scintillation method. For this analysis, a volume of 10 mL was collected in duplicate directly in the counting glass vials, in which the same volume of the universal LSC-cocktail Instagel XF has been previously added (Szikszay and Sampa, 1983). The samples were measured in a liquid scintillation analyser (model TRI-CARB 2100TR, Packard Instrument Co.). The energy spectrum obtained for the water samples were compared with a standard solution of ^{226}Ra measured previously and the energetic spectrum of ^{222}Rn and daughter products were determined. Typical lower limit of detection for ^{222}Rn determination was 1.9×10^{-1} Bq/L.

Bathing mud samples were directly collected in triplicate from a bathtub. For this analysis, a volume of 850 mL of the mixture solution used in mud bathing (sulphurous mineral water plus raw mud) were added to a Marinelli 850 polyethylene beaker and the sample was sealed. After a equilibrium period of 30 days, specific activities of ^{232}Th, ^{226}Ra and ^{40}K in these samples were measured by gamma spectrometry with a hyperpure germanium detector, GEM-15200, from EG&G for 150,000 seconds. The detector was calibrated using standard solutions having radionuclide activities certified by Amersham. The ^{226}Ra activities were determined by taking the mean activity of 3 separate photopeaks of its daughter nuclides: ^{214}Pb at 295.2 keV and 351.9 keV, and ^{214}Bi at 609.3 keV. The ^{232}Th content of the smiles was determined from the intensity of the 911 keV and 968 keV gamma-ray peaks of ^{228}Ac. The potassium content was determined from the 1460 keV gamma-ray peak following decay of ^{40}K.

The portable detector RDA-200 from SCINTREX, was used to measure alpha particle activity originating from radon at 4 different balneological facilities at Termas de Araxá (EPA, 1989). A volume of 20 L of air was sampled through a calibrated scintillator cell connected to one filter holder assembly using a pump. The scintillatior cells were measured 3 hours after the end of sampling in order to determine the ^{222}Rn airborne concentrations.

RESULTS AND CONCLUSIONS

All the samples studied were collected at Termas de Araxá, during the period from August/1998 to May/1999, comprising three sampling runs.

The activity concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn determined in thermal water and spring water used in Termas de Araxá treatments are presented in Table 1. The concentrations of ^{226}Ra and ^{228}Ra varied from 54 to 98 mBq/L and from 87 to 148 mBq/L, respectively. The ^{222}Rn activity concentration, on the other hand, ranged from 107 to 657 Bq/L. The highest ^{222}Rn and ^{226}Ra activity concentrations were observed in Dona Beja spring water. For ^{228}Ra activity, the maximum value was found in Andrade Júnior sulphurous spring water. In all the drinking water samples used by patients and local residents, ^{226}Ra activities did not exceed the maximum permissible concentration established for drinking water supplies of 0.1 Bq/L (WHO, 1993), although there is no regulation controlling ^{226}Ra occurrence in thermal or mineral waters.

The ^{226}Ra activities obtained for spring waters of Araxá are smaller than those obtained by the same authors (Oliveira *et.al.*, 1994) for other springs located at Poços de Caldas Plateau, where many health resorts are found.

The radon levels observed in spring water of Araxá are the same order of magnitude as those observed by Doretti *et al.* (1992) in waters used in thermal therapy in Abano Terme, Italy, and spa waters from Ikaria Island, Greece (Trabidou *et.al.*, 1996).

The activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K measured in bathing mud samples in the same period of study are presented in Table 2. Bathing mud measurements showed activity concentrations for ^{232}Th from 0.7 to 1.0 Bq/kg, for ^{226}Ra from < 0.3 to 1.0 Bq/kg and for ^{40}K from < 2.3 to 11 Bq/kg, respectively. These concentrations are lower than those observed by Doretti *et al.* (1992) in some mud samples collected in the Abano Terme thermal region. The dose rate to the patients due to bathing mud was determined by using the dose conversion factors calculated by Beck *et al.* (1972) for the gamma-ray dose rates in air over radioactive rocks and soils:

$$D = 0.048C_K + 0.49C_{Ra} + 0.76C_{Th}$$

where C is the concentration of the different natural radionuclides in Bq/kg and the dose rate D is expressed in nSv/h. Based in these data, the dose to the patients was evaluated, giving a value of 0.2 μSv/y, which is in agreement with those observed by Doretti *et al.* (1992), in Abano Terme, Italy.

The ^{222}Rn indoor concentrations measured at 4 different balneological facilities at Termas de Araxá and corresponding doses are presented in Table 3. The radon concentration in air vary widely, depending on the specific sites. According to Lettner *et al.* (1996), the radon concentration in radon spas in Austria vary from 10 to 3,300 Bq/m^3. Sciocchetti *et al.* (1990) observed similar situation in Italy with radon concentrations ranging from 0.44 to 514 kBq/m^3. Kobal and Renier (1987) found radon concentrations up to 190 Bq/m^3 at Podcetrtek, Slovenia. In the present paper the maximum ^{222}Rn indoor concentrations were observed at the collective swimming pool, which use radioactive water from Dona Beja spring, in agreement with the values observed in Austria.

The personnel working in thermal spas can be subjected to increased levels of radon and radon daughters exposure in the course of their duties. The results obtained in Table 3 for the occupational dose for the workers at Termas de Araxá did not exceed the ICRP recommended limit of 50 mSv/y for the radon inhalation.

Based upon arithmetic mean concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn determined in Dona Beja and Andrade Júnior springs, committed doses to the critical organs and committed effective doses due to the ingestion of these natural radionuclides were evaluated for the patients considering a daily consumption of 2L/day of mineral water during the treatment period of 21 days at Termas de Araxá. Doses due to ingestion of Dona Beja and Andrade Júnior spring waters were also estimated for the local residents, taking into account the same daily water intake. The annual dose per unit of activity ingested for ^{226}Ra and ^{228}Ra used in these calculations (in Sv/Bq) were taken from ICRP 67. For the ingestion of radon, there are no internationally accepted dose factors. The application of a modified ICRP model to the ingestion of radon in water (Kendall *et al.*, 1988) leads to a value of 10^{-8} Sv/Bq for the committed effective dose per unit intake, with all the dose coming from the gas rather than the decay products (UNSCEAR, 1993). The estimates for the dose to stomach per unit activity of ingested radon vary between $5x10^{-8}$ and $2x10^{-7}$ Sv/Bq (UNSCEAR, 1982; 1984), an average value of 10^{-7} Sv/Bq was adopted here. These results are presented in Table 4. Doses up to $5.2x10^{-2}$ mSv/y, $1.5x10^{-1}$ mSv/y and 2.3 mSv/y were estimated for the critical organs considering the ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn by patients, respectively; while the corresponding committed effective doses reached values of $1.1x10^{-3}$ mSv/y, $3.9x10^{-3}$ mSv/y and $2.3x10^{-1}$ mSv/y for the same radionuclides. Considering the consumption of these waters by the local residents of Araxá, doses up to $9.1x10^{-1}$ mSv/y, 2.6 mSv/y and 38.7 mSv/y were estimated for the critical organs considering the ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn.

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Table 1. Radioactivity of thermal water and springs at Termas de Araxá.

Sample	^{222}Rn (Bq/L)	^{226}Ra (mBq/L)	^{228}Ra (mBq/L)
Dona Beja Spring water	(7)	(3)	(3)
Concentration range	433 - 657	90 – 99	87 - 93
Arithmetic Mean	530 ± 95	96 ± 4	90 ± 4
Andrade Júnior Spring water	(6)	(4)	(3)
Concentration range	107 - 124	54 – 71	135 – 148
Arithmetic Mean	116 ± 7	62 ± 7	142 ± 9
Perola Bath Radioactive water	(6)	(4)	(4)
Concentration range	470 - 612	82 – 96	94 – 116
Arithmetic Mean	534 ± 59	90 ± 6	104 ± 11
Perola Bath Sulphurous water	(6)	(4)	(4)
Concentration range	32 - 39	56 – 72	136 – 173
Arithmetic Mean	36 ± 3	66 ± 7	156 ± 18
Swimming pool Radioactive water	(6)	(4)	(4)
Concentration range	148 - 505	75 – 88	89 – 136
Arithmetic Mean	331 ± 153	79 ± 6	117 ± 21

(n) = number of measured samples

Table 2. Radionuclide concentrations in mud bath determined by gamma spectrometry.

	^{40}K(8) (Bq/kg)	^{226}Ra(8) (Bq/kg)	^{232}Th(8) (Bq/kg)
Concentration Range	< 2.3 - 11	< 0.3 – 1.0	< 0.6 – 1.0
Arithmetic Mean	4.3 ± 3.2	0.5 ± 0.2	0.8 ± 0.1

(n) = number of measured samples

Table 3. Indoor concentrations of ^{222}Rn and committed effective doses due to inhalation at Termas de Araxá.

Sampling place	^{222}Rn (Bq/m^3)	$H_E{}^{222}Rn$ (mSv/y)	
		Patients	Personnel
Radioactive water Perola bath room	686	2.1×10^{-2}	0.3
Sulphurous water Perola bath room	811 100	1.4×10^{-2}	0.2
Radioactive water swimming pool room	3,153 1,861	7.6×10^{-2}	0.9
Mud bath room	557 238	1.2×10^{-3}	0.1

Table 4. Expected doses to the patients and local residents from the consumption of ^{226}Ra, ^{228}Ra and ^{222}Rn present in Dona Beja and Andrade Junior spring waters.

Sample	Patients		Local Residents	
	Dose to the critical organ (mSv/y)	Committed effective dose (mSv/y)	Dose to the critical organ (mSv/y)	Committed effective dose (mSv/y)
Dona Beja spring water				
^{226}Ra	5.2×10^{-2}(a)	1.1×10^{-3}	9.1×10^{-1}(a)	2.0×10^{-2}
^{228}Ra	9.5×10^{-2}(a)	2.5×10^{-3}	1.6(a)	4.4×10^{-2}
^{222}Rn	2.3(b)	2.3×10^{-1}	3.9×10^{1}(b)	3.9
Andrade Júnior spring water				
^{226}Ra	3.4×10^{-2}(a)	7.3×10^{-4}	5.9×10^{-1}(a)	1.3×10^{-2}
^{228}Ra	1.5×10^{-1}(a)	3.9×10^{-3}	2.6(a)	6.8×10^{-2}
^{222}Rn	4.9×10^{-1}(b)	4.9×10^{-2}	8.5(b)	8.5×10^{-1}

(a) H_{bone} = committed dose to the bone.
(b) $H_{stomach}$ = committed dose to stomach.

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