^{222}Rn determination in mineral waters from the Poços de Caldas Plateau in Brazil

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Abstract. It is estimated that 50% of the radioactive effective dose equivalent to man comes from radon and its radioactivity daughters. The main ^{222}Rn sources are the soils, building materials and potable waters. There is an especial interest in spas waters with high natural radioactivity. It's considered that the use of these waters as drinking waters is a significant radiation exposure factor, and it is worthwhile to assess the consequent dose. Such estimation has been made for the Poços de Caldas Plateau, which is a region of high natural radioactivity, from volcanic origin, containing several spas (Águas da Prata, Caldas, Poços de Caldas and Andradas). The ^{222}Rn content was determined in 23 spring waters in Poços de Caldas and neighboring cities. In water sampling, modified Marinelli flasks were used. The determinations were effected with a high resolution gamma ray spectrometer. High concentration variations were observed in the collected mineral waters, the highest values having been found in Fonte Villela's waters, in Águas da Prata town (926 Bq/l); Grande Hotel's in Caldas Town (420 Bq/l) and COLAB's in Poços de Caldas region (289 Bq/l). The annual whole body effective dose equivalent estimate for adult due to water ingestion, using the Crowford – Brown's biokinetic model's adult dose conversion factors, was higher than 1 mSv/year in 61% of the analyzed waters. Key words: ^{222}Rn, spring waters, Poços de Caldas plateau, radioactive dose, natural radioactivity.

Introduction

The Poços de Caldas Plateau is situated in the southwest region of Minas Gerais State, Brazil. It is formed by a volcanic caldera with a diameter of approximately 35 km, with a central depression surrounded by a 300 meters high borderline.

The Plateau has a high natural radioactivity with many radioactive anomalies containing uranium and thorium in association with rare earth, zirconium and molybdenum minerals.

In this region (figure 1) are located the spas of Águas da Prata, Caldas, Poços de Caldas and Andradas, which have many mineral waters springs amply used by local people and tourists. Two of these springs are commercially exploited.

These spring's waters come from prolonged contact with mineral rocks and so it's expected to find radioactive elements dissolved and diffused. Lauria and Godoy (1988) have found low concentration of ^{238}U, ^{234}U, ^{226}Ra, ^{228}Ra, ^{232}Th, ^{230}Th, ^{228}Th in these spring's waters, except for one spring Fonte Villella, with high ^{226}Ra and ^{228}Ra values.

In this paper, ^{222}Rn concentration was determined in 23 mineral water springs in the Poços de Caldas Plateau cities. The sampled springs were the most widely used by the population as drinking water. These waters were collected just at the emergence point.

An estimate for annual effective dose equivalent of adult individual of the public consuming waters has been made.

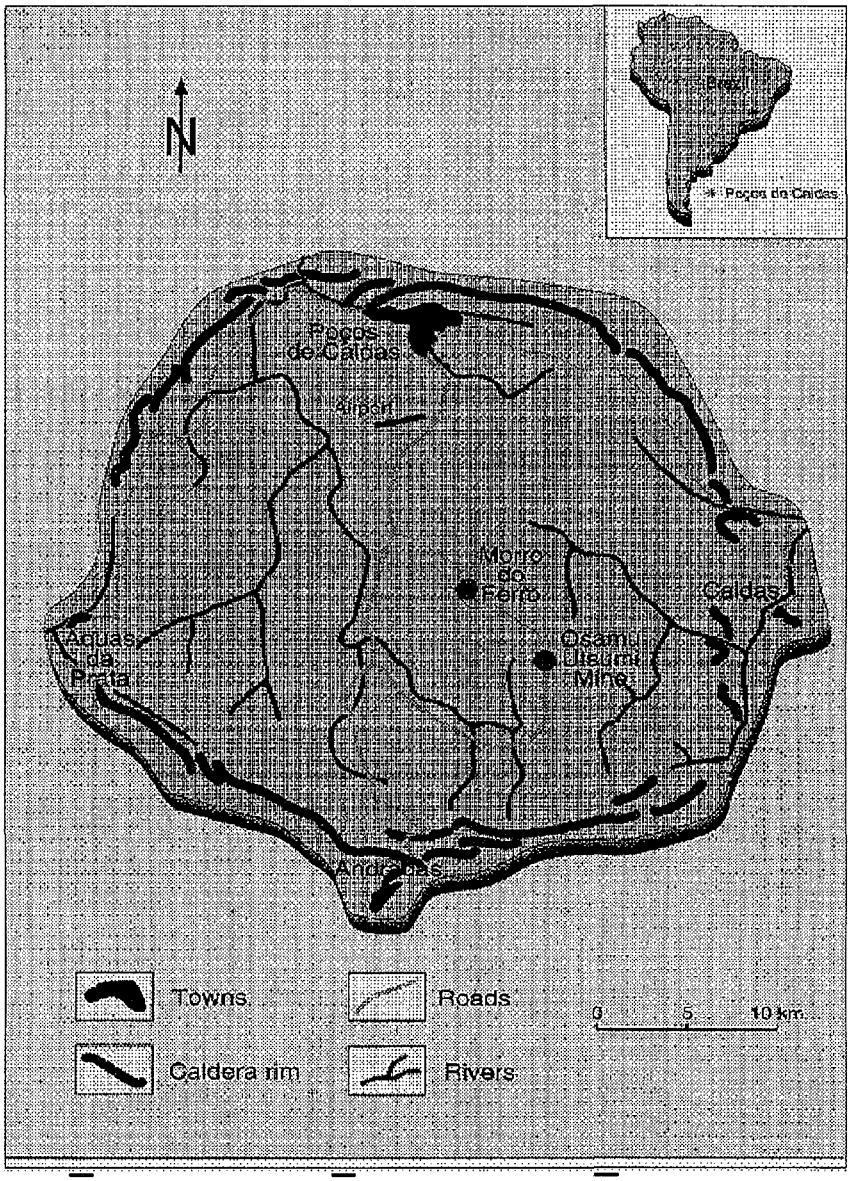

Figure 1. Poços de Caldas Plateau (NAGRA, 1993).

Materials and methods

Sampling

The water samples were collected using modified Marinelli flasks (Figure 2), like those described by Belloni, Cavaioli, Ingrao, Mancini, Notaro, Santaroni, et al (1995) but without the lock valve. In substitution of the lock, a flexible plastic cover with thread and a long staff was made to close the opening. For the other opening a stopper with a long staff, so that when closing the flasks after filling it, there is a small internal pressure, to avoid bubble formation. The sampling was made using a tube placed at the bottom of the flasks, letting the water overflow for a couple minutes. Three samples were taken from each spring.

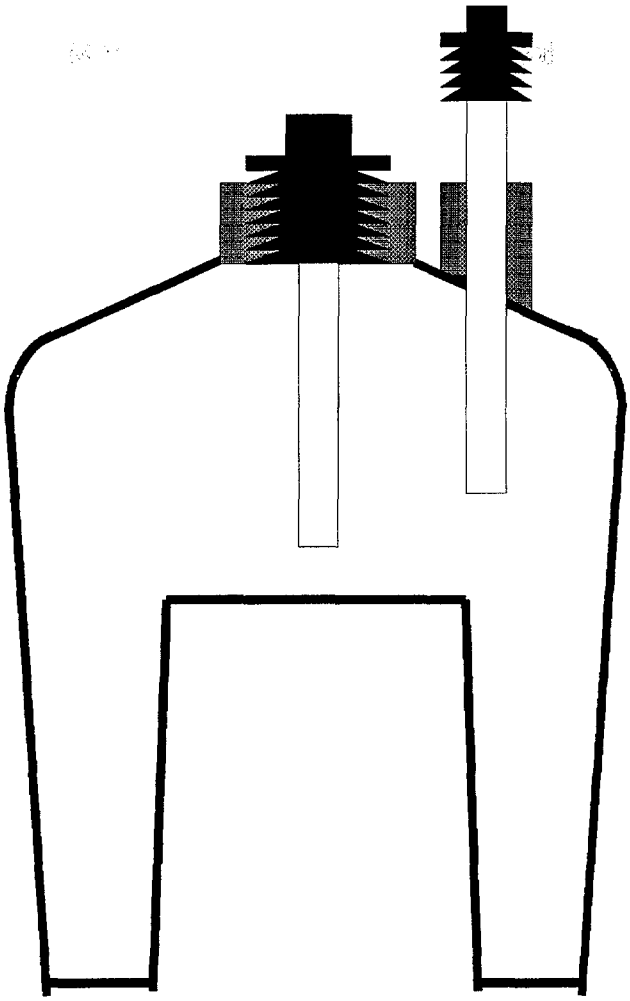

Figure 2. Modified Marinelli flask.

The samples were taken to the laboratory in thermal boxes, to maintain the temperature.

Gamma ray spectrometry

After four hours, to allow radioactive equilibrium of ^{222}Rn and it daughters' ^{214}Pb and ^{214}Bi, the samples were measured at 295, 352 and 609 KeV, in a gamma ray spectrometer model GMX — 4519 from CANBERRA, with relative efficiency of 45%, for 15 minutes.
For the system calibration, an Amershan ^{226}Ra standard solution, diluted to a final activity at 1467.88 Bq/l was used. This solution was placed into a counting flask, sealed and after 30 days (to reach radioactive equilibrium of ^{226}Ra and its daughters) the ^{214}Pb and ^{214}Bi photo peaks were measured.
Using the same geometry, the samples were counted under identical conditions, the results being obtained by direct comparison with the standard solution above mentioned.
The algorithm used to calculate the activity is:
$A_a = A_p \times CPS_a/CPS_p$
Where:
A_a = sample activity;
A_p= standard sample;
CPS_a= counts per second at the photo peak of the sample;

CPS_p = counts per second at the photo peak of the standard;

Dose estimate

The annual whole body effective dose equivalent due to radon ingestion was estimated using Crawford–Brown biokinetics model's adult dose conversion factors (Crawford–Brown, 1990), assuming an individual ingests 2 l of water per day and 1,5 day elapsed between water collection and consumption.

Table: ^{222}Rn concentration in spring waters and estimated of annual effective equivalent dose due radon ingestion

Fountains	**Rn-222** (Bq/l)	**Dose**[a] (mSv/year)
Poços de Caldas		
Águas Minerais Poços de Caldas 1	**60,50 ± 2,50**	**0,81**
Águas Minerais Poços de Caldas 2	**27,33 ± 6,44**	**0,36**
Bianuchi	**150,50 ± 2,00**	**2,01**
Cibel	**54,67 ± 3,11**	**0,73**
COLAB	**289,00 ± 10,0**	**3,86**
Cristo	**39,67 ± 1,11**	**0,53**
Fraya	**64,50 ± 0,50**	**0,86**
Monjolinho	**122,67 ± 4,22**	**1,64**
Morro das Cabras	**152,33 ± 4,89**	**2,03**
Primavera	**170,50 ± 1,50**	**2,27**
Quati 1	**180,00 ± 17,3**	**2,40**
Quati 2	**214,67 ± 0,80**	**2,87**
Sant'Ana	**86,33 ± 1,11**	**1,15**
Sinhazinha	**84,00 ± 4,00**	**1,12**
Águas da Prata		
Antiga Prata	**53,00 ± 4,67**	**0,71**
Paiol	**6,00 ± 1,33**	**0,08**
Radioativa	**88,33 ± 15,11**	**1,18**
Vilella	**925,67 ± 38,22**	**12,36**
Caldas		
Grande Hotel	**417,67 ± 9,11**	**5,58**
Sebastião Cândido	**79,67 ± 1,78**	**1,06**
Andradas		
Rua Cedro	**88,50 ± 1,50**	**1,18**
Serra	**35,67 ± 1,11**	**0,48**
Vila Mosconi	**13,67 ± 0,44**	**0,18**

a – annual whole body effective dose equivalent for adult

Results

The ^{222}Rn activity concentrations in water samples and the annual effective equivalent dose to an adult are presented in table 1. The values are an average of three measurements of three samples collected in the same spring.

The highest value for ^{222}Rn was found in the Villela spring, in Águas da Prata town, in agreement with the highest ^{226}Ra and ^{228}Ra values cited by Lauria and Godoy (1988).
All ^{222}Rn values are higher than 3,7 Bq/l, the limit recommended by US-EPA (Crawford–Brown, 1991), which corresponds to a cancer incidence probability of 10^{-4}. All the same, the values surpass the Brazilian norm reference value of the 0,1 Bq/l (Brazil, 1990).
If the US-EPA recommendation is to be adopted, all spring waters should be treated, or the population be instructed on ^{222}Rn concentration decreasing techniques.
In 14 of these 23 analyzed samples (61%), the estimated effective equivalent dose is higher than 1mSv/year, which is the maximum annual dose for an individual of the public in Brazil.
These results suggest that this region's mineral waters are an important factor in radioactive dose due to natural exposure and so collective dose assessment and cancer risk should be determined.
Another sampling campaign will be conducted in the rainy period, in order to check the ^{222}Rn content variation in the spring waters analyzed.

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