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# A method for evaluating $^{228}\text{Ra}$ in environmental matrices and its use at Poços de Caldas plateau, Brazil

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## Abstract

This work describes a methodology developed for performing the extraction, detection and quantification of  $^{228}\text{Ra}$  in waters, suspended solids and sediments. The technique proved to be useful for analyzing samples from the hydrological environment of Morro do Ferro, Poços de Caldas plateau, Brazil. The  $^{228}\text{Ra}$  activity in underground waters of 5 boreholes drilled in the area varied from 0.02 up to 14.5 Bq/l, whereas for the surficial waters the variation was from 0.04 to 0.51 Bq/l; for the suspended solids, the values ranged from 1.5 up to 419 Bq/g, whereas it was possible to find a value of 2.04 Bq/g for the sediments. These results show the applicability of the method for characterizing different matrices of environmental interest. © 2002 Elsevier Science Ltd. All rights reserved.

*Keywords:*  $^{228}\text{Ra}$ ; Waters; Sediments; Suspended solids

## 1. Introduction

Radium has about 25 isotopes with mass numbers between 206 and 230. All are unstable and only four occur in the natural series of radioactive decay:  $^{226}\text{Ra}$  (half-life = 1622 years) in the uranium series,  $^{223}\text{Ra}$  (half-life = 11.1 days) in the actinium series,  $^{228}\text{Ra}$  (half-life = 6.7 years) and  $^{224}\text{Ra}$  (half-life = 3.64 days) in the thorium series. Among these radium isotopes, only  $^{228}\text{Ra}$  is a beta emitter, the others being alpha emitters (Ku and Broecker, 1976).

Radium in the environment may be released due to the interaction of waters with rocks, soils or mineralized bodies, where the mining and processing of phosphate minerals, apatite, copper, gold, lignite, coal and bauxite also can contribute to the enrichment of Ra in the superficial and underground waters, since it is present in the U–Th decay

series (Iyengar, 1984, 1990; Jaworowski, 1990; Dickson, 1990).

The geochemical behavior of radium including its mobilization into water is little known in Brazil, partly because of the analytical difficulties for its quantification. This work describes a method developed to quantify  $^{228}\text{Ra}$  in water, suspended solids and sediments, phases that need to be analyzed in order to estimate the Ra transport in the environment. The technique was implemented at LABIDRO (Isotopes and Hydrochemistry Laboratory), Department of Petrology and Metallogeny, Institute of Geosciences and Extracts Sciences, University of the State of São Paulo (UNESP), having been adapted from Hancock and Martin (1991), and consisting of the following steps: Ra coprecipitation, Ra separation in anion exchange column, and gamma spectrometry for evaluating the  $^{228}\text{Ra}$  activity (Mancini, 1997). In addition, the principle of isotopic dilution was also used, as described by Gomes (1978) and Bonotto (1982) for U analysis, where in the present case,  $^{133}\text{Ba}$  was added to each sample as a radioactive tracer for determining the chemical yield due to the similarities between barium and radium.

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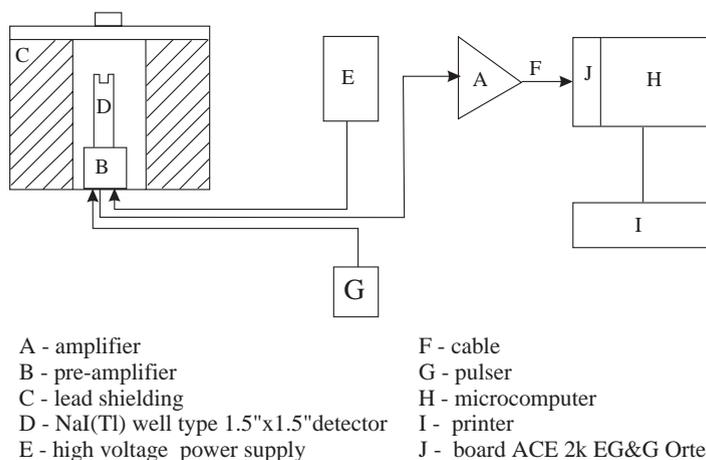


Fig. 1. Outline of the gamma spectrometric system used for the detection of  $^{228}\text{Ra}$  from  $^{228}\text{Ac}$ .

## 2. Laboratorial procedures

### 2.1. Calibration of the gamma spectrometer

The gamma spectrometry was used in this work because it is a method with easy handling and fast response. The system is composed of an NaI(Tl) well-type detector, a high-voltage power supply, amplifiers and a computer which possess a board ACE 2k EG&G Ortec, and a multichannel simulator software (1024 channels) (Fig. 1). The count rate for each photopeak of interest was calculated using the procedure described in the manual of MAESTRO software, version A65-BI, EG&G Ortec (Ortec, 1996). The calibration of the system was carried out through different single-element radioactive sources, including  $^{133}\text{Ba}$  (Table 1). The quantification of  $^{228}\text{Ra}$  was performed taking into account the condition of secular radioactive equilibrium between  $^{228}\text{Ra}$  and its direct descendant,  $^{228}\text{Ac}$ , which is reached in approximately 45 h (Mancini and Bonotto, 1998). Table 2 shows the gamma emissions used for the quantification of  $^{228}\text{Ac}$ . Due to the overlap of the  $^{133}\text{Ba}$  peaks with the  $^{228}\text{Ac}$  low-energy peak (Fig. 2), it was necessary to analyze the samples in duplicate.

### 2.2. Matrices preparation

Water samples are filtered through a  $0.45\ \mu\text{m}$  Millipore membrane. Sediments are dried, disaggregated and sieved to separate the grain size fraction between 0.053 and 0.177 mm. After filtering, each water sample is divided into two aliquots of equal volume, with the  $^{133}\text{Ba}$  radioactive tracer being added to one aliquot, with an activity corresponding to 49 Bq. Then, both aliquots are evaporated to a final volume of 100 ml.

The membrane used for filtering is dried and weighed for determining the amount of suspended solids, a

Table 1

Energy (Chukreev, 1994; Lederer et al., 1967) of the gamma emissions of the radioactive sources used for the calibration of the spectrometric system and channel of occurrence of the photopeaks

Nuclide	Energy (keV)	Channel
$^{133}\text{Ba}$	356	276
	389	298
$^{137}\text{Cs}$	661	522
$^{60}\text{Co}$	1170	899
	1330	1020

Table 2

Gamma emissions (Michel et al., 1982) for evaluating  $^{228}\text{Ac}$

Nuclide	Energy (keV)	Probability of emission (%)
$^{228}\text{Ac}$	338	12.3
$^{228}\text{Ac}$	911	29.0
$^{228}\text{Ac}$	964–969	23.0

parameter necessary for the evaluation of  $^{228}\text{Ra}$  content in this phase. The membrane is digested with  $\text{HNO}_3$  and the solution is brought to dryness. The dry residue is dissolved with 0.1 M HCl to a volume of 200 ml; then the solution is divided into two aliquots of equal volume, with the  $^{133}\text{Ba}$  radioactive tracer (activity = 49 Bq) being added to one of them.

The sediments need to be dissolved/fully digested in order to evaluate the  $^{228}\text{Ra}$  activity. About 1 g of the material is generally enough, which is inserted into a Parr-type digestion system (Bonotto, 1996), along with 5 ml of HF, 5 ml of  $\text{HNO}_3$  and 5 ml of HCl. After dissolution/digestion, the sample is diluted with 0.1 M HCl to a volume of 200 ml, and the solution is divided

into two equal aliquots, with the  $^{133}\text{Ba}$  radioactive tracer (activity = 49 Bq) being added to one of them. After obtaining all aliquots in acid (0.1 M HCl), the same procedures are used for water, suspended solids and sediments.

### 2.3. Radium coprecipitation

Ra precipitation is performed by adding to each aliquot (volume of 100 ml) 1 ml of concentrated  $\text{H}_2\text{SO}_4$ , 2 g of  $\text{K}_2\text{SO}_4$  and 1 ml (drop to drop) of  $\text{Pb}(\text{NO}_3)_2$ . This causes the Ra to precipitate as  $\text{Pb}(\text{Ra})\text{SO}_4$  which is an insoluble, slightly acid salt. After allowing the precipitate to settle, the supernatant phase is eliminated. The precipitate is washed with 20 ml of 1:1 0.1 M  $\text{K}_2\text{SO}_4$ –0.2 M of  $\text{H}_2\text{SO}_4$  solution, and the precipitation and separation repeated. Then, 5 ml of 0.1 M ammoniacal EDTA (pH 10) and two drops of ammonia are added to the  $\text{Pb}(\text{Ra})\text{SO}_4$  precipitate, and the beaker warmed to aid dissolution. If necessary, 1 ml portions EDTA

solution are added until the precipitate has dissolved (Hancock and Martin, 1991).

### 2.4. Radium separation in anion exchange resin

To separate radium and barium from actinium and thorium, an anion exchange resin (AG1-X8, 100–200 mesh, chloride form) is used, which retains Ac and the Th whereas Ra and Ba complexed with the EDTA passes through it. The beaker containing the solution is rinsed 3 times with 1 ml of EDTA, which is passed through the column. After collecting the effluent containing Ra and Ba (about 8 ml), the date and hour are recorded. It is important to wait a time of at least 45 h before gamma counting the sample. The solution is stored in an appropriate cylindrical bottle for the reading. For processing another aliquot, the resin is rinsed with 0.01 M EDTA.

### 2.5. $^{228}\text{Ra}$ activity

Fig. 3 shows the typical gamma spectrum for the aliquot without (Fig. 3(a)) and with (Fig. 3(b))  $^{133}\text{Ba}$  yield tracer. Both the gamma spectra are needed to provide the parameters necessary for determining the  $^{228}\text{Ra}$  activity in the sample.

Initially, it is necessary to calculate the activity ratio (AR) between the  $^{228}\text{Ac}$  lower energy peak (1) and the  $^{228}\text{Ac}$  higher energy peak (2) for the spectrum without  $^{133}\text{Ba}$  yield tracer (Fig. 3(a)). Because the count rate in the first peak may be represented by  $\text{Tc}_1$ , whereas in the second by  $\text{Tc}_2$ , the AR between these two peaks ( $\text{AR}_{1/2}$ ) may be written as

$$\text{AR}_{1/2} = \frac{\text{Tc}_1}{\text{Tc}_2}. \quad (1)$$

For the spectrum containing  $^{133}\text{Ba}$ , it is necessary to calculate the ratio between the overlapping  $^{133}\text{Ba}$  and  $^{228}\text{Ac}$  peaks (3) and the  $^{228}\text{Ac}$  higher energy peak (4)

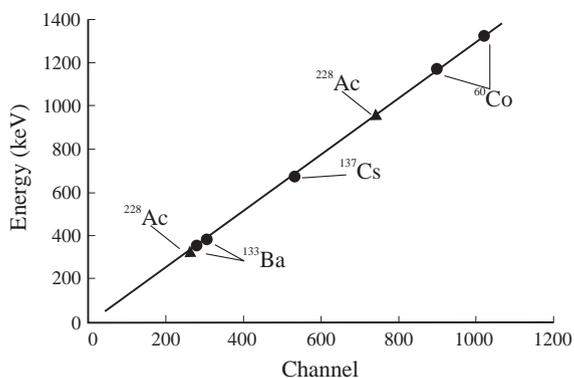


Fig. 2. Calibration curve of the gamma spectrometer for the detection of  $^{228}\text{Ac}$ .

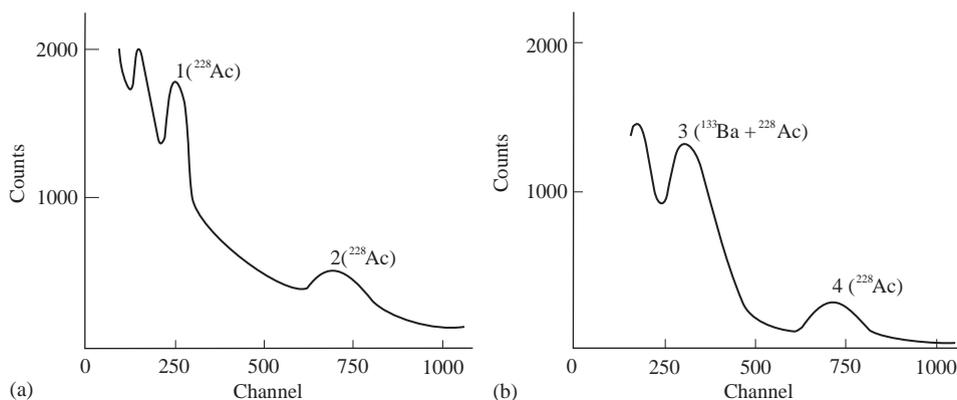


Fig. 3. Gamma spectra showing (a) the  $^{228}\text{Ac}$  and (b)  $^{228}\text{Ac} + ^{133}\text{Ba}$  peaks.

(Fig. 3(b)), taking into account their respective count rates, i.e.  $T_{C_3}$  and  $T_{C_4}$ . Thus, the AR between these two other peaks ( $AR_{3/4}$ ) may be expressed by

$$AR_{3/4} = \frac{T_{C_3}}{T_{C_4}}. \quad (2)$$

Because  $T_{C_3}$  is the sum of the count rate of the two overlapped peaks, it can be written as

$$T_{C_3} = T_{C_{Ba}} + T_{C_{Ac}}, \quad (3)$$

where  $T_{C_{Ba}}$  is the count rate for barium and  $T_{C_{Ac}}$  is the count rate for actinium.

Substituting  $T_{C_3}$  into Eq. (2), we have

$$AR_{3/4} = \frac{T_{C_{Ba}} + T_{C_{Ac}}}{T_{C_4}}. \quad (4)$$

Rewriting Eq. (4)

$$T_{C_{Ba}} = \left( AR_{3/4} - \frac{T_{C_{Ac}}}{T_{C_4}} \right) T_{C_4}. \quad (5)$$

Since both aliquots belong to the same sample, the AR between the  $^{228}\text{Ac}$  lower energy peak and the  $^{228}\text{Ac}$  higher energy peak is the same at the gamma spectra without and with  $^{133}\text{Ba}$  yield tracer. Thus, it is possible to write

$$AR_{1/2} = \frac{T_{C_1}}{T_{C_2}} = \frac{T_{C_{Ac}}}{T_{C_4}}. \quad (6)$$

Substituting Eq. (6) into Eq. (5), the count rate for  $^{133}\text{Ba}$  may be expressed as

$$T_{C_{Ba}} = (AR_{3/4} - AR_{1/2})T_{C_4}. \quad (7)$$

Finally, the measured count rate for  $^{133}\text{Ba}$  ( $T_{C_{Ba}}$ ) is compared with the tracer activity added to each aliquot

(49 Bq), which allows to evaluate the  $^{228}\text{Ra}$  activity ( $At_{Ra}$ ) in the sample (in Bq) according to the following equation:

$$At_{Ra} = \frac{49T_{C_4}}{T_{C_{Ba}}}. \quad (8)$$

For standardizing the data, the  $^{228}\text{Ra}$  activity ( $At_{Ra}$ ) must be divided by the analyzed volume (for water aliquots) or weight (for aliquots of suspended solids and sediments).

## 2.6. Performance of the method

The New Brunswick Laboratory (NBL) Certified Reference Materials from the US Department of Energy and consisting of monazite sand-silica mixture (thorium standards in the range  $2.5\text{--}10^4 \mu\text{g/g}$ , i.e. NBL 110-A, NBL 109-A, NBL 108-A, NBL 107-A, NBL 106-A) were used to calibrate a gamma spectrometer with a planar NaI(Tl) scintillation crystal for performing measurements of  $^{228}\text{Ra}$  activity based on the  $^{228}\text{Ac}$  higher energy peak (911–969 keV). The condition of radioactive equilibrium is assured throughout the  $^{232}\text{Th}$  decay series in the standards, and the obtained data fit the straight line (Fig. 4)

$$\text{Log}(At) = 1.96 + 1.02\text{Log}(EI), \quad (9)$$

where “At” is the  $^{228}\text{Ra}$  activity (Bq/g) and “EI” is the effective intensity, i.e. count rate per mass (cps/g).

Such calibration curve was utilized to evaluate the  $^{228}\text{Ra}$  activity in 20 g bottom sediments enriched in Th and REE which were collected on 25 June 1996 at the drain of the area investigated in this work (Fig. 6). The

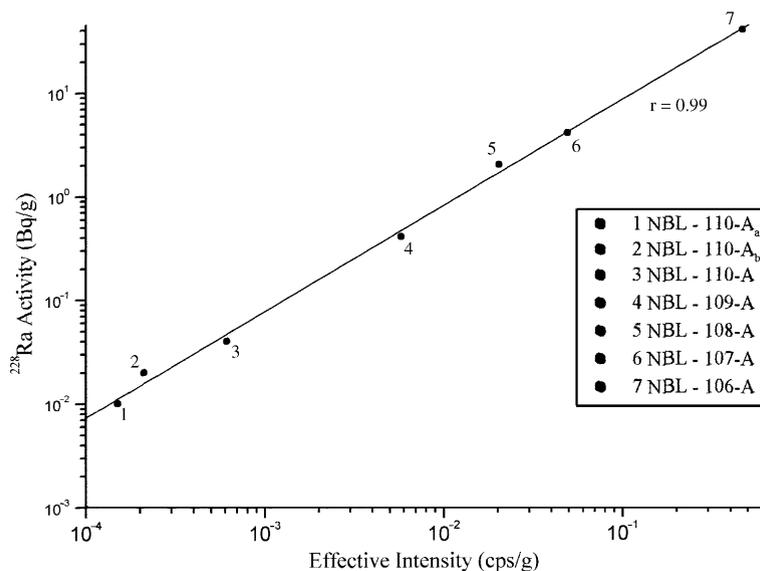


Fig. 4. The logarithm of the  $^{228}\text{Ra}$  activity in NBL standards plotted against the logarithm of the effective intensity (count rate per mass)

collection of the sediments was carried out with a 1 m long PVC tube (diameter of 50 mm), which was introduced in the place chosen for sampling; the sample was collected and stored in a plastic bag. After preparation at the laboratory (drying, disaggregation and sieving) the gamma counting was performed, which yielded an  $^{228}\text{Ra}$  activity corresponding to  $2.2 \pm 0.2$  Bq/g.  $^{228}\text{Ra}$  activity in bottom sediments from the same area determined by Campos et al. (1986) ranged from 0.14 up to 3 Bq/g, including the obtained value within this interval.

Because the method described in this paper for the analysis of sediments is destructive, it was avoided to dissolve/digest the certified reference materials. Thus, the performance of the technique was checked on dissolving 0.5 g of the same sediments collected at the drain, whose  $^{228}\text{Ra}$  activity had been measured by the non-destructive gamma-ray assay. The values obtained for  $\text{AR}_{1/2}$  and  $\text{AR}_{3/4}$  correspond, respectively, to 0.73 and 48.33, which allowed to find a  $^{228}\text{Ra}$  activity of  $2.04 \pm 0.07$  Bq/g. Therefore, within experimental errors, the non-destructive and destructive techniques supplied similar results, implying that despite using a yield tracer ( $^{133}\text{Ba}$ ) that is not an isotope of the analyzed element (Ra), its similar chemical behavior during Ra coprecipitation is enough to provide confident data. A good aspect involving the data acquisition by the proposed method is that the count rates (cps) for the equations in Sections 2.5 do not need to be corrected for the differing emission probabilities of the various peaks, which is a great advantage over the existing gamma-ray spectroscopic methods (Adams and Gasparini, 1970; Ivanovich and Harmon, 1982).

The efficiency of the NaI(Tl) well-type detector was evaluated for  $^{133}\text{Ba}$  gamma rays (356–389 keV) generated by a solution containing only the amount of tracer added (49 Bq). The count rate corresponded to 10.1 cps, a value that allowed to estimate a detector efficiency of 20.6%. Considering that the total yield found during analysis of the sediments by dissolution/digestion was 5.8%, and taking into account that the chemical yield may be calculated by dividing the total yield by the detector efficiency, it is possible to estimate 28.3% for the chemical yield of the process. Such value is very reasonable when compared with others reported for methods utilizing yield tracers during chemical steps, for instance,  $^{236}\text{U}$  or  $^{232}\text{U}$  for  $^{238}\text{U}$  analysis by alpha spectrometry (0.5–35%) (Osmond and Cowart, 1976; Gomes, 1978; Bonotto, 1982, 1990; Rosholt, 1984; Sánchez and Tomé, 1990).

The chemical phase of radium separation in anion exchange resin was also carefully investigated to ensure that the existing (coprecipitated)  $^{228}\text{Ac}$  had in fact been retained on the column. In order to do this, the effluents containing Ra were immediately recovered after passing through the resin and submitted to gamma counting,

with the obtained spectra only indicating radioactive background (aliquot without  $^{133}\text{Ba}$  yield tracer) or  $^{133}\text{Ba}$  (aliquot with tracer). Another reading for the same aliquots was performed after 1 week of the first measurement, it being possible to identify the presence of  $^{228}\text{Ac}$  peaks in the new gamma spectra, suggesting  $^{228}\text{Ac}$  accumulation from  $^{228}\text{Ra}$  at least within the 45 h waiting time. The resin utilized in the process was also subjected to gamma counting being possible to identify a doublet  $^{212}\text{Pb}$ – $^{224}\text{Ra}$  peak (around 240 keV) generated by  $^{228}\text{Ac}$ ,  $^{228}\text{Th}$  and Pb retained on the column.

The efficiency of the cleaning of the resin with 0.01 M EDTA was checked after running the same sediments sample by performing the gamma counting of the cleaning solutions. The spectrum of the cleaning solution related to the aliquot without tracer indicated no presence of  $^{228}\text{Ac}$ , implying on the effectiveness of the cleaning procedure. Furthermore, the spectrum of the cleaning solution related to the aliquot with  $^{133}\text{Ba}$  yield tracer exhibited the presence of a small amount of  $^{133}\text{Ba}$ , which, very importantly, had been retained on the resin as also evidenced by its gamma counting. So, such finding confirms again the efficiency of the cleaning step with 0.01 M EDTA.

### 3. Application of the method

The developed technique was used for analyzing samples from the hydrological environment of Morro do Ferro, situated at the Poços de Caldas plateau, state of Minas Gerais, Brazil. The Morro do Ferro is located near the center of the plateau (Fig. 5), which presents a circular format (average diameter of 33 km), area of about 800 km<sup>2</sup>, altitude from 1300 to 1600 m, and topography consisting mainly of valleys, soft mounts and hills. The place is well known to possess a mineralization of thorium and rare earths in the presence of magnetite dikes intercalated by argillaceous layers. In the regional geologic context, it is part of a mass of tinguaita located in the edge of an area where the rock suffered intense hydrothermal alteration (Almeida, 1977).

The Morro do Ferro contributes to the formation of a small basin of surficial draining, where there are depressions, streams and marshy areas. The surficial hydrology in the drainage basin is simple, consisting mainly of two streams, one north and another south of the mount. The north stream is of secondary importance and drains the face north of the mount. The south stream has origin in its base, receives most of the draining from the hill, its tributaries due to runoff are ephemeral, and the discharge is significant only during the wet periods (in the dry period, the water supply is performed by emergent underground waters) (Lei, 1984).

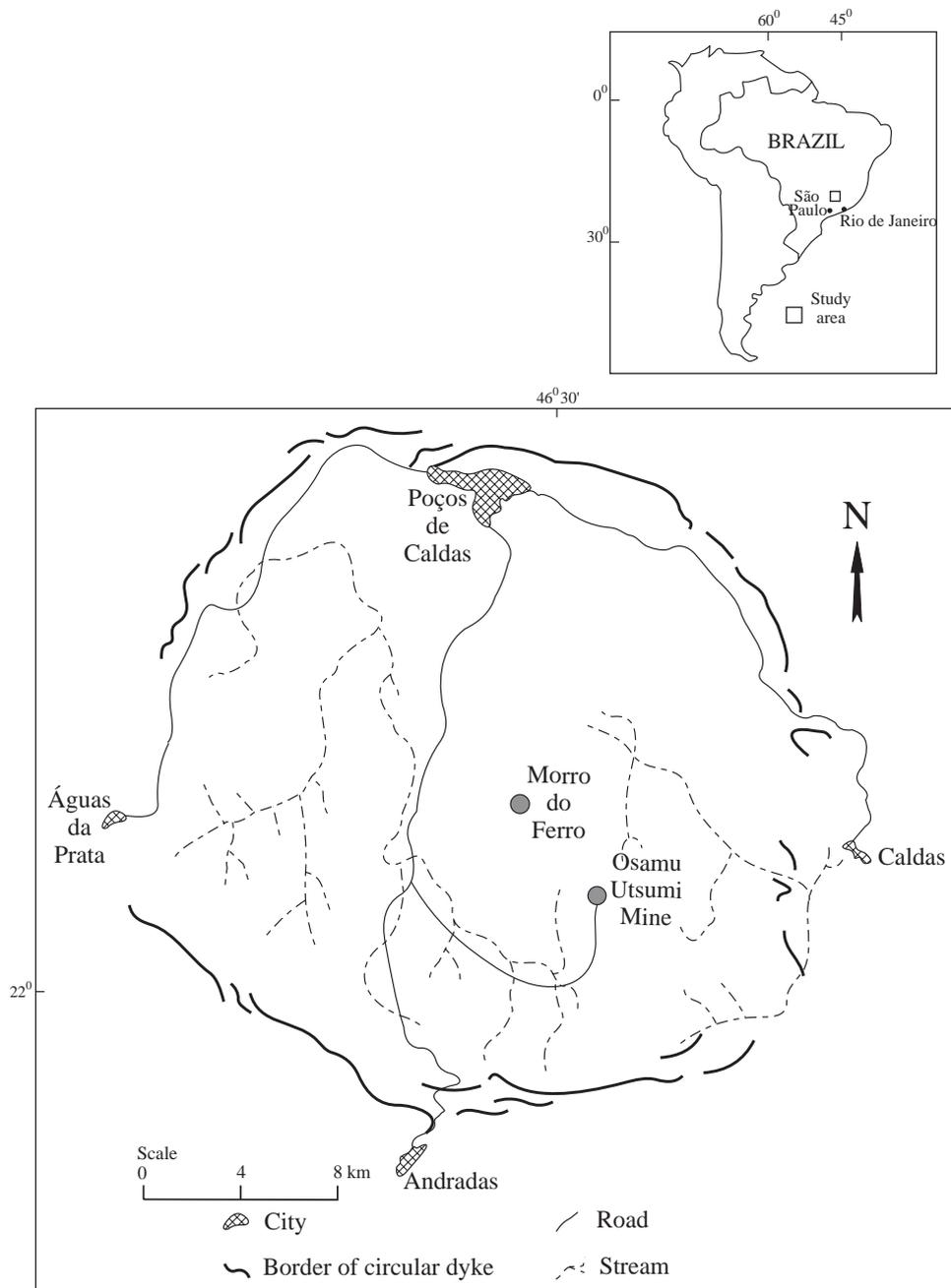


Fig. 5. Location of Morro do Ferro in the Poços de Caldas plateau. Adapted from Holmes et al. (1991).

IPT (1982) drilled nine boreholes (SR-1 up to SR-9) in the place for installing piezometers, with the aim of characterizing the subsurface flow. The depths varied from 13 to 65 m, where three drills are located at the mineralized area (SR-5, SR-7 and SR-8), two distant from this area (SR-1 and SR-3), and four around it (SR-2, SR-4, SR-5 and SR-9) (Fig. 6). Another four holes were drilled in the 1980s, two in the mineralized

area (MF-10 and MF-11), one in the neighborhoods of this area (MF-13) and another next to south stream (MF-12) (Holmes et al., 1992) (Fig. 6).

Two sampling campaigns were realized at the Morro do Ferro, one in the dry period (June 1996) and another in the wet period (January 1997), where underground water samples were taken from the following boreholes: SR-4, SR-5, SR-6, SR-9, and MF-12. Two surficial

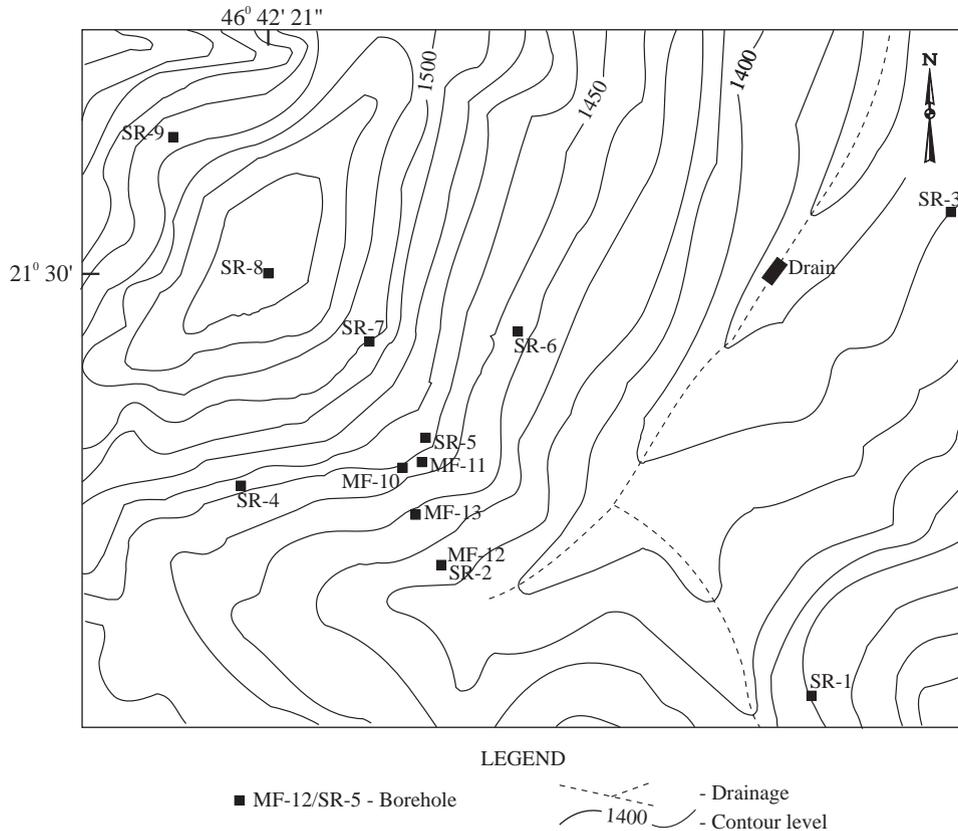


Fig. 6. Location of the piezometric wells drilled at the Morro do Ferro. Adapted from IPT (1982) and Holmes et al. (1992).

water samples were also collected: Vert., collected in a drain constructed to measure the south stream flow (Lei, 1984) and Pr. Vert., collected approximately 100 m upstream of the drain (Fig. 6).

A volume of 15–20 l for each underground and surficial water sample was stored in polyethylene bottle. Manual devices were used to collect the samples, and care was taken to avoid the introduction of bottom sediments into the surficial water samples.

## 4. Results

### 4.1. The critical level of detection

Because low count rates were found in some peaks of interest, it was necessary to decide if the obtained signals actually corresponded to a peak, i.e. if they were “true” or “false”. For this, the critical level (Lc) established by Currie (1968) was evaluated. The Lc takes into account the count rate of the background ( $T_{cB}$ ) in the energy region corresponding to the peak of interest, as described by Bonotto (1986). The minimum detectable

activity ( $A_m$ ) was evaluated by the relationship

$$A_m = \frac{49Lc}{T_{cBa}} \quad (10)$$

Two background readings were carried out as reported in Table 3 that allowed to evaluate critical levels corresponding to  $0.0088 \pm 0.0005$  cps (region of energy = 356–389 keV), and  $0.0072 \pm 0.0004$  cps (region of energy = 911–967 keV).

### 4.2. $^{228}\text{Ra}$ in waters

Figs. 7(a) and (b) illustrate the gamma spectra obtained for the sample collected at SR-9 in 24 June 1996, which well demonstrate the presence of the peaks of interest. The  $^{228}\text{Ra}$  activity for underground and surficial waters in the two sampling campaigns is reported in Table 4, as well the minimum detectable  $^{228}\text{Ra}$  activity calculated for several samples. The obtained values are quite variable, ranging from  $<0.02$  Bq/l for ground waters of SR-6 up to 14.5 Bq/l for ground waters of SR-5. The chemical yield was variable (0.4–26%), with an average of 12%. A higher amount of sample is needed to be analyzed for

Table 3  
Count rate of the background ( $T_{cB}$ ) in the spectrometric system

Date of reading	Nuclide	Region of energy (keV)	Count time (s)	Number of counts	$T_{cB}$ (cps) <sup>a</sup>
October/96	$^{228}\text{Ac} + ^{133}\text{Ba}$	356–389	59987.2	37,429	0.6239
	$^{228}\text{Ac}$	911–967		24,200	0.4034
January/97	$^{228}\text{Ac} + ^{133}\text{Ba}$	356–389	59987.3	38,185	0.6366
	$^{228}\text{Ac}$	911–967		25,359	0.4227

<sup>a</sup> cps—counts per second.

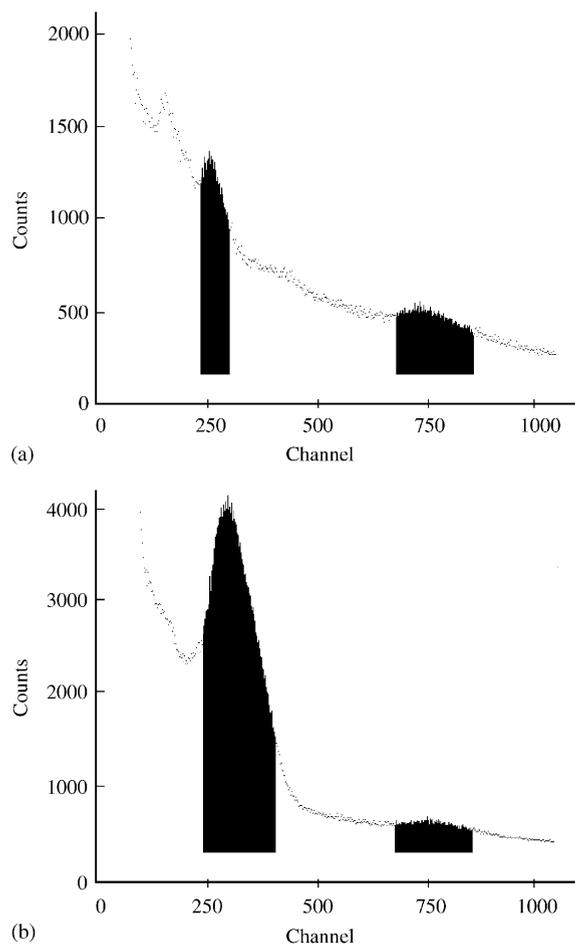


Fig. 7. Gamma spectra obtained for the groundwater sample of SR-9 collected in 24 June 1996 which was analyzed (a) without radioactive tracer and (b) with radioactive tracer ( $^{133}\text{Ba}$ ).

generating signals in the  $^{228}\text{Ac}$  photopeaks of low activity. Franca (1983) also evaluated the presence of  $^{228}\text{Ra}$  in underground and surficial waters from the same area, and the obtained values are also reported in Table 4. Similarities and differences are found between the values determined by Franca (1983) and those obtained in this paper.

The water table in the area is a subdued replica of the topography, with recharge on the elevations from precipitation and discharge locally to streams (IPT, 1982; Holmes et al., 1992). The hydrographs of the boreholes is variable, exhibiting very sudden rises and declines or gradual rises, depending on the silty clay thickness above water table, presence of magnetite dikes intersected, and preferential paths of circulation such as fractures and fissures (IPT, 1982). Bonotto (1989) found in the area a relation between dissolved U content increase and water table rise due to rainwater infiltration and, like for uranium, seasonal variations in dissolved  $^{228}\text{Ra}$  concentrations were observed for most of the samples, with the higher values for samples of SR-5, SR-6 and SR-9 being obtained when collection was performed in the wet period (January 1997). The main discrepancy in this behavior was observed for sample of MF-12, which may be explained by the location of this borehole adjacent to the valley bottom, where it suffers influence of the stream as indicated by an upward vertical groundwaters flow (Holmes et al., 1991). Such process is more significative in the wet period, as a consequence, the  $^{228}\text{Ra}$  concentration in the stagnant water becomes affected by infiltrating dilute waters containing a lower content of dissolved radium.

#### 4.3. $^{228}\text{Ra}$ in suspended solids

Table 5 reports the results for  $^{228}\text{Ra}$  activity in suspended solids from Morro do Ferro area, where it is possible to see that most of the data represent the minimum activity, which can probably be attributed to the small amount of material recovered by the use of Millipore membrane when filtering the water sample. As expected, the highest value was found for the sample of SR-5, varying between 242 and 419 Bq/g. Campos et al. (1986) also determined the  $^{228}\text{Ra}$  activity in suspended solids collected at several places of Morro do Ferro, amongst them at drain (Vert). Although the values reported in Table 5 for Vert are below the minimum detectable  $^{228}\text{Ra}$  activities, it is possible that they may represent the average activity of 10.5 Bq/g found by Campos et al. (1986).

Table 4  
<sup>228</sup>Ra activity in underground and surficial water samples

Sample	Sampling 06/96			Sampling 01/97			Franca (1983)
	Volume (l)	Activity (Bq)	Act./vol. (Bq/l)	Volume (l)	Activity (Bq)	Act./vol. (Bq/l)	Act./vol (Bq/l)
MF-12	8.00	16.8	2.10	8.50	1.9	0.22	
SR-4	9.50	<0.2	<0.02	7.75	<0.2	<0.02	0.16–0.34
SR-5	6.50	40.5	6.23	7.25	105.0	14.48	5.18–21.2
SR-6	7.25	<0.1	<0.01	7.50	1.3	0.17	0.05–2.07
SR-9	7.50	2.3	0.31	8.50	45.4	5.34	0.14–0.84
Pr. Vert.	8.30	<0.4	<0.05	9.00	4.6	0.51	
Vert.	8.00	<0.3	<0.04	8.60	<0.3	<0.03	0.01–0.12

Table 5  
<sup>228</sup>Ra activity in samples of suspended solids

Sample	Sampling 06/96			Sampling 01/97		
	Weight (g)	Activity (Bq)	Act./weight (Bq/g)	Weight (g)	Activity (Bq)	Act./weight (Bq/g)
MF-12	0.009	0.30	32.92	0.013	0.95	73.22
SR-4	0.014	<0.26	<18.52	0.126	<0.19	<1.53
SR-5	0.007	1.70	242.33	0.262	109.70	418.70
SR-6	0.008	<0.50	<62.96	0.076	<0.16	<2.10
SR-9	0.029	0.18	6.26	0.032	<0.20	<6.25
Pr. Vert.	0.014	<0.23	<16.14	0.025	<0.23	<9.18
Vert.	0.019	<0.09	<4.87	0.028	<0.34	<12.04

## 5. Conclusion

With the development of this work, it was possible to customize and implement a methodology for measuring <sup>228</sup>Ra in samples of water, suspended solids and sediments, that are very important matrices for environmental studies. The chemical yield obtained when analyzing real samples is compatible with others reported for methods utilizing yield tracers during chemical steps. So, the method proved to be useful and reliable, since not much chemical separation steps were necessary to provide the data. Under this aspect, the technique is still promising to be utilized because there is no need to correct the different gamma emission probabilities in the spectra. Another advantage consists on the possibility of using low-cost instrumentation for acquiring the data. The technique was successfully used in the hydrological environment of Morro do Ferro, Poços de Caldas plateau, Brazil, where most of the obtained results corresponded to values found by other researchers. The determination of the critical level of detection of <sup>228</sup>Ra assisted the identification of signals close to the background, assuring reliability for the implemented method. However, it was observed that for samples having low radium content, it is necessary to conduct the chemical treatment with a higher amount of material, a limitation imposed by the low-cost instrumentation utilized.

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