

PRELIMINARY RESULTS OF NATURAL RADIOACTIVITY EVALUATION IN UNDERGROUND WATERS OF THE CITY OF CAMPOS DO JORDAO, SP, BRAZIL

Gabriel B. Affa and Brigitte R. S. Pecequilo

Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN)

Av. Professor Lineu Prestes 2242

05508-000 São Paulo, SP

gabriel.beraldo@usp.br and brigitte@ipen.br

ABSTRACT

Twenty-eight 5-liter samples were collected during the year of 2018 on four different campaigns (January, April, August and December) for seven selected sources (SP-123, Britania, Amizade, Dom Bosco, Volta Fria, Simao and Renato) of easy and free access to the population on the city of Campos do Jordão, SP, Brazil. Samples were concentrated from 1L to 50 mL of which 4 mL pipetted on 60 mm diameter stainless steel planchet were analyzed with a Berthold's PC-controlled 10-channel Low-level Alpha-Beta Counter LB 770 Win in three 120-minute counting cycles. Preliminary results have shown low alpha and beta activity concentrations for all sources with values ranging between $0.0288 \text{ Bq/L} \pm 0.0036 \text{ Bq/L}$ and $0.1059 \text{ Bq/L} \pm 0.0118 \text{ Bq/L}$ for alpha activity and between $0.0371 \text{ Bq/L} \pm 0.005 \text{ Bq/L}$ and $0.1352 \text{ Bq/L} \pm 0.0148 \text{ Bq/L}$ for beta activities. Results showed a tendency of higher concentrations during periods of drier weather. All obtained values are below the limits of 0.5 Bq/L and 1 Bq/L, respectively for alpha and beta activity concentrations, recommended by the 2017 WHO Guideline on Drinking Water Management.

1. INTRODUCTION

1.1. Radioactivity in Water

The radioactive elements are present in soil, waters, rocks, plants and in the living beings. Atoms owe this broad coverage to both the geochemical interaction between inorganic elements and their subsequent introduction into the food chain [1]. The presence of the radioactive elements in the water facilitates living beings being exposed to radiation in several ways such as: external exposure - when the individual is only exposed to radiation without incorporating the radionuclide, inhalation of particulates, soil ingestion, groundwater ingestion, food intake and dermal absorption [2].

Regarding radiation doses, an average dose per person of 3.0 mSv/year [3] is estimated, with 80% of this value (2.4 mSv/year) coming from natural sources. Doses of the order of 100 mSv/year are considered of great carcinogenic risk. The recommended value of cancer probability due to water intake is only 5.5×10^{-6} under normal environmental radiation conditions [4].

The main radionuclides possibly present in potable water are the ^{238}U , ^{226}Ra and ^{210}Po alpha emitters and the ^{40}K , ^{228}Ra and ^{210}Pb beta emitters [5]. As identification and concentration of a single radionuclide in drinking water requires expensive time-consuming analyses many times unnecessary, traditionally, gross alpha and beta analysis, a simple radioanalytical procedure, is recommended by WHO (World Health Organization) [6] as the first step as a *screening method*, for being a fast, safe and low cost one. The World Health Organization recommends that the total alpha and beta radioactivity levels which must be observed in potable water, in a way new measures are not necessary, are 0.5 Bq/L for alpha and 1 Bq/L for beta and; if the established limits are crossed it must be carried out an analysis for specific radionuclide [6].

Assessment of natural radioactivity in surface and underground waters have been and are being conducted in several regions of Brazil. The waters of Cambuquira and Marimbeiro [7] and Caxambu [8] were analyzed in the state of Minas Gerais; in Sao Paulo, in the region of Aguas de Lindoia [9], Alto Vale do Ribeira [10] and Águas do Prata [11]; in Bahia, in Caetité (located in the Uranium Province of Lagoa Real) [12].

The work reported in the states of Minas Gerais and Sao Paulo showed, for the most part, doses within the desired standards, with some exceptions caused by high concentrations of radon in specific sources, such as in the city of Caxambu. The study of the correlation between the intake of these waters and the health risks of the population was also made, as in the works of Mazzili [13], Jacomino [14] and Oliveira [15], which associate high doses with the incidence of cancer. Most of the referenced work done in Brazil includes the border region between the states of Sao Paulo and Minas Gerais, a region known as the Circuit of Waters, which includes sources that are used to supply the most populous region of the country.

Finally, with the previous studies as motivators, the city of Campos do Jordão, also located in this area of great importance for the Southeast region of Brazil, was chosen for the evaluation of water sources widely used by regular people.

1.2. Campos do Jordão

Campos do Jordão (SP) is the highest city in Brazil with 1,628 meters above mean sea level. The city is located on the Serra da Mantiqueira, a rocky massif that extends through three states of the Southeast (São Paulo, Minas Gerais and Rio de Janeiro) and reaches altitudes near 3,000 meters at its highest peaks [16].

Founded in 1874, the city was classified for decades by the São Paulo State Government as a climatic and hydro-mineral resort. Nowadays, Campos do Jordão is considered a sightseeing pole, with hundreds of daily visitors, notably in the winter months.

As for hydrography, Campos do Jordao is located on a fractured aquifer in the first unit of Water Resources Management – Mantiqueira (UGRHI - Unidade de Gerenciamento de Recursos Hídricos, in portuguese) among 22 established in the State of Sao Paulo, being defined by the hydrographic basins of the Sapucaí-Guaçu and Sapucaí-Mirim rivers and their tributaries, in the area of Serra da Mantiqueira.

The Campos do Jordão Plateau region is home to a fractured aquifer system. The fractured aquifers are composed of crystalline rocks, igneous or metamorphic, which are compact and do not have voids between the minerals that constitute them. For this reason, the water circulates between the empty spaces generated by the fractures. These fractures are breaks, flat or straight, that form after the rock has already been cooled and consolidated [16].

Groundwater resources are the source of the basic flow of rivers and represent rich water reserves that do not require expensive treatment plants. As groundwater is a component of the hydrological cycle, its availability in the aquifer is related to the basic flow of the drainage basin installed over the area of occurrence. The ratio less than 30% between consumption and water flow in Mantiqueira is among the lowest among the management units and allied to the humid climate with precipitations above 1,700 mm annually [17], ensures abundance for residents and tourists.

The present work aims to evaluate the radioactive activity eventually present in waters of non-commercial sources consumed by the population in the city of Campos do Jordão. Samples from selected sources of the city, collected with seasonal periodicity, were analyzed with a proportional detector, by the screening method for gross alpha and beta activities determination. Finally, potential correlations between activity concentration variation through the seasons and climatic variables were studied. Rainfall was selected as the most significant meteorological variable that could possibly influence the concentration of radionuclides in the underground waters, as, if there is more water available to fill up the aquifers, it would be expected to have more diluted samples of water.

2. MATERIALS AND METHODS

2.1. Selection of underground water sources

The city of Campos do Jordão, with an area of 290.52 km² and a population of 50.540 inhabitants [18] is located on the eastside of the State of Sao Paulo, Brazil. Figure 1 shows the location of Campos do Jordão inserted in the South America continent.



Figure 1: Campos do Jordão, Brazil, and the South America continent.

A more detailed characterization of the lithology of the municipality was made by Szikszay [19], along with physicochemical and radioactive analyzes of water sources located in the surroundings and rural area of the municipality, often difficult to access. On the tangent to this work, the selected sources are located in the districts of the city, in the urban area, where the residences and commerce are concentrated and where there is the largest flow of people who potentially make use of the water.

Some of the selected sources were channeled by the public power, still in the time of the foundation of the city, while others arrive at the surface naturally between the fractures of the rocks. The selected sources are presented in Table 1 and in the map of Figure 2:

Table 1: Assessed underground water sources and their coordinates.

SOURCE	COORDINATES
BRITANIA	22°44'34.2"S 45°35'16.0"W
AMIZADE	22°44'09.3"S 45°35'23.7"W
SIMAO	22°43'41.0"S 45°34'06.2"W
DOM BOSCO	22°44'42.8"S 45°36'15.1"W
RENATO	22°43'06.0"S 45°34'28.8"W
VOLTA FRIA	22°43'59.4"S 45°35'13.1"W
SP-123	22°47'29.5"S 45°37'16.3"W



Figure 2: Map of Campos do Jordão (SP) showing the location of the selected sources. (Google Earth Pro. 01-07-2019)

2.2. Experimental procedure

In 2018, five liters of each of the twenty-eight samples were collected following the year seasons, namely in January, April, August and December. The water samples were conditioned in polyethylene bottles, mixed with nitric acid, with pH around 2 or lower, for protection of microorganisms proliferation, kept away from direct sunlight and stored in an air-conditioned room.

For the total alpha and beta activity determination by the screening method, an initial sample volume of 1 L was concentrated to 50 mL on a heating plate. After, a 4 mL aliquot was pipetted on a clean 60 mm diameter stainless steel planchet and placed under an infrared lamp until total evaporation [12]. Three planchets of every sample were measured with a flow gas proportional detector LB 770 10-Channel alpha-beta Low-Level Counter, during three cycles of 120 min each.

2.3. Total Alpha and Beta Activities Concentrations Calculation

The total alpha and beta activity concentrations were calculated as [12] in equation 1:

$$C = \frac{Am - Bg}{Ef \times V \times Fc \times 60} \quad (1)$$

Where C is the activity concentration (Bq/L), Am the mean sample count (cpm), Bg the average count of detector background radiation (cpm), Ef the detector efficiency, V the sample volume on the planchet, Fc the sample concentration factor and 60 the conversion factor of minutes to seconds.

The detector efficiency Ef was calculated as:

$$Ef = \frac{Rn - Bg}{A \times 60} \quad (2)$$

Where: Ef is the detector counting efficiency, Rn the average count (cpm), Bg the detector background radiation (cpm), A the standard activity and 60 the conversion factor of minutes to seconds.

The uncertainty of the activity concentrations and detectors efficiencies were calculated by error propagation.

3. RESULTS

3.1. Alpha and Beta Activity Concentrations

The activity concentrations results in the 2018 collected water samples are presented in Table 2 for total alpha and in Table 3 for total beta.

Table 2: Total ALPHA activity (Bq/L) in the water samples collected in 2018.

SOURCE	Jan/18 (Bq/L)	Apr/18 (Bq/L)	Aug/18 (Bq/L)	Dec/18 (Bq/L)
SP-123	0.0545 ± 0.0074	0.0716 ± 0.0091	0.0633 ± 0.0084	0.0437 ± 0.0067
BRITANIA	0.0288 ± 0.0036	0.0468 ± 0.0054	0.0324 ± 0.0040	0.0334 ± 0.0054
AMIZADE	0.0302 ± 0.0037	0.0723 ± 0.0078	0.0597 ± 0.0066	0.0322 ± 0.0045
DOM BOSCO	0.0305 ± 0.0041	0.0327 ± 0.0056	0.0320 ± 0.0049	0.0308 ± 0.0040
SIMAO	0.0869 ± 0.0098	0.1059 ± 0.0118	0.0592 ± 0.0072	0.0634 ± 0.0076
RENATO	0.0692 ± 0.0087	0.0414 ± 0.0063	0.0644 ± 0.0081	0.0399 ± 0.0060
VOLTA FRIA	0.0798 ± 0.0093	0.0566 ± 0.0030	0.0415 ± 0.0055	0.0563 ± 0.0070

Table 3: Total BETA activity (Bq/L) in the water samples collected in 2018.

SOURCE	Jan/18 (Bq/L)	Apr/18 (Bq/L)	Aug/18 (Bq/L)	Dec/18 (Bq/L)
SP-123	0.0890 ± 0.0114	0.1032 ± 0.0130	0.0731 ± 0.0100	0.0714 ± 0.0096
BRITANIA	0.0536 ± 0.0066	0.0545 ± 0.0060	0.0371 ± 0.0050	0.0429 ± 0.0075
AMIZADE	0.0907 ± 0.0100	0.0951 ± 0.0118	0.1215 ± 0.0125	0.0781 ± 0.0100
DOM BOSCO	0.1098 ± 0.0126	0.1035 ± 0.0121	0.1152 ± 0.0130	0.1030 ± 0.0117
SIMAO	0.1143 ± 0.0126	0.1352 ± 0.0149	0.1202 ± 0.0138	0.1352 ± 0.0148
RENATO	0.1073 ± 0.0130	0.0920 ± 0.0115	0.1191 ± 0.0140	0.1056 ± 0.0124
VOLTA FRIA	0.1218 ± 0.0138	0.0922 ± 0.0075	0.0909 ± 0.0114	0.0959 ± 0.0120

3.2. Activity concentrations and rainfall

Possible relationships between rainfall and the activity concentrations summarized from Tables 2 and 3 are presented in Figures 2 and 3, respectively for total alpha and beta. The accumulated precipitation data was taken from the National Institute of Meteorology (INMET) database [17]. Campaigns were associated with their respective season: January – Summer; April – Fall; August – Winter; December – Spring.

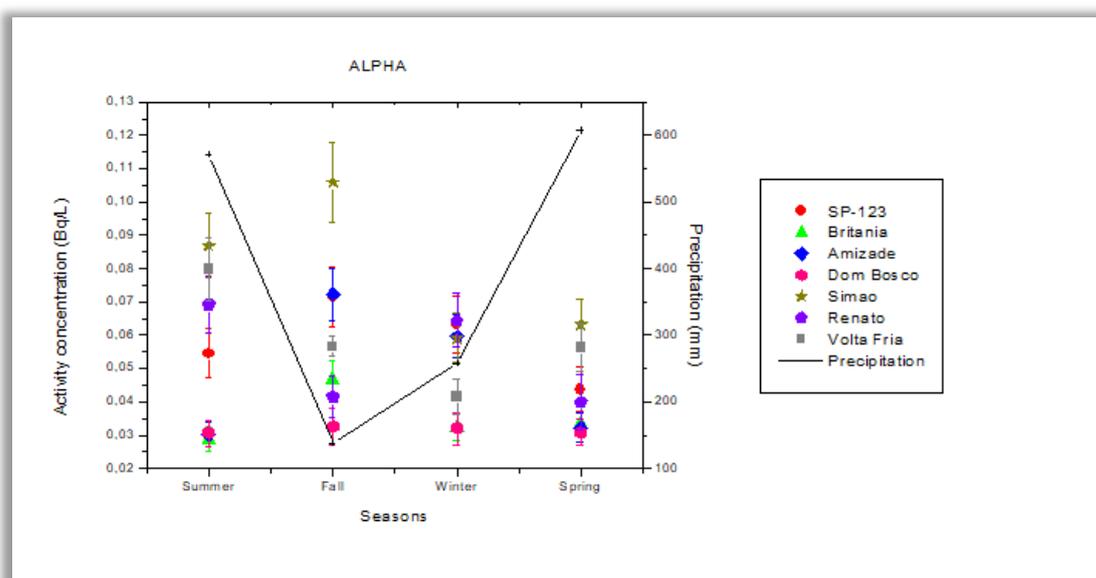


Figure 3: Alpha activity concentrations in Campos do Jordão water samples and accumulated rainfall.

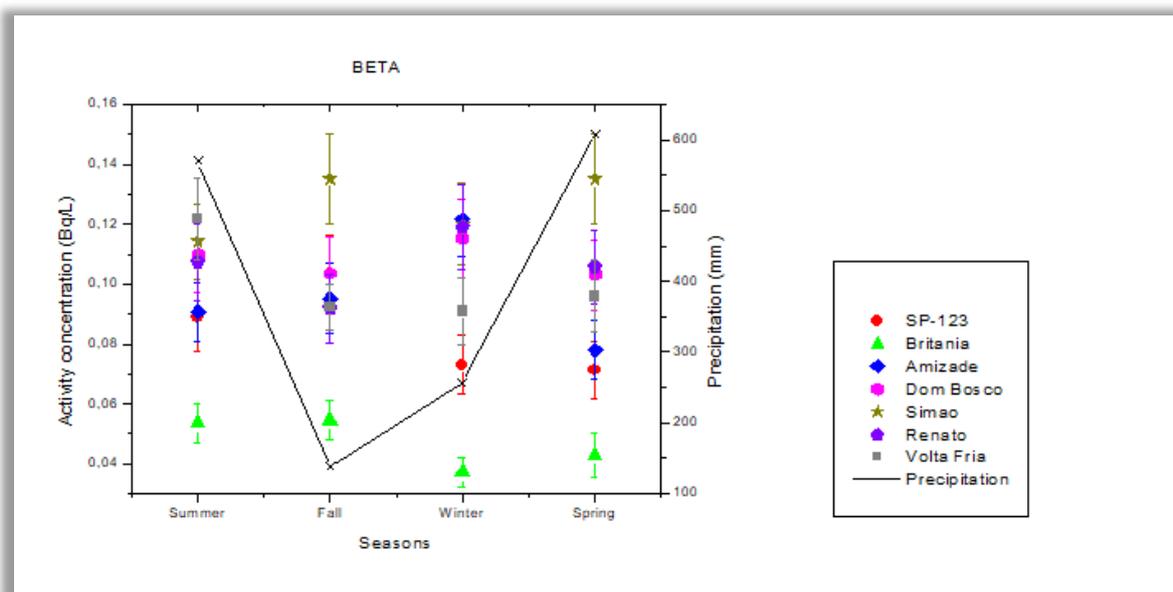


Figure 4: Beta activity concentrations in Campos do Jordão water samples and accumulated rainfall.

4. CONCLUSIONS

The seven sources studied present different levels of concentration of radioactivity; the average values were the lowest for the Vila Britania source and the highest for the Simao source. All activity concentrations of the seven underground water sources are below the recommended limits determined by the WHO Guideline on Drinking Water Management [6] of 0.5 Bq/L for alpha radiation and 1 Bq/L for beta radiation showing that there is no radiological health risk derived from the consumption of the water by population.

The variation of the activity concentrations throughout the year of 2018 was different for each source. However, apparently there is a tendency of higher concentrations during periods of dry weather such as Fall and Winter, particularly for gross alpha for five out of the seven sources (SP-123, Britania, Amizade, Dom Bosco and Simao) and for gross beta for six out of seven (except for Volta Fria).

Further, collection of water samples will continue during 2019 and also, ingestion doses will be calculated.

5. ACKNOWLEDGMENTS

The authors would like to thank Instituto de Pesquisas Energéticas e Nucleares (IPEN) for the use of the laboratory facilities. Gabriel B. Affa would like to thank CAPES for the scholarship grant.

6. REFERENCES

1. MADRUGA, M. J.; SILVA, L.; GOMES, A. R.; LIBÂNIO, A.; REIS, M. The influence of particle size on radionuclide activity concentrations in Tejo River sediments. *Journal of Environmental Radioactivity*, 132, 2014. 65-72.
2. BONOTTO, D. M. Natural radionuclides in major aquifer systems of the Paraná sedimentary basin, Brazil. *Applied Radiation and Isotopes*, 69, 2011. 1572-1584.
3. UNSCEAR – UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION. Sources and Effects of Ionizing Radiation. UNSCEAR 2008, Report to the General Assembly with Scientific Annexes. Volume II. New York, 2008.
4. ICRP - INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION. ICRP, 2008. The 2007 Recommendations of the International Commission on Radiological Protection. ICRP Publication 103. *Ann. ICRP* 37 (2–4).
5. JOBBÁGY, U. WATJEN, J. MERESOVA. Current status of gross alpha/beta activity analysis in water samples: a short overview of methods, *J. Radioanal Nucl Chem*, 2010. 286:393-399.
6. WHO – WORLD HEALTH ORGANIZATION. Guidelines for drinking-water quality. 4a ed. <http://www.who.int/water_sanitation_health/publications/drinking-water-quality-guidelines-4-including-1st-addendum/en/>, 2017. Accessed in 29/08/2017.
7. SANTOS, L. A. B. ; DAMATTO, S. R. ; OLIVEIRA, J. ; CARMO, A. P. . Preliminary results of ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb concentration in mineral waters from Parque das Águas de Cambuquira and Marimbeiro (MG). In: NORM 8 - Naturally Occurring Radioactive Material, 2016, Rio de Janeiro. NORM 8 - Naturally Occurring Radioactive Material, 2016.
8. MENEGHINI, A. A. ; DAMATTO, S. R. ; OLIVEIRA, J. ; CARMO, A. P. . Preliminary results of seasonal variation of ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb concentrations in mineral waters from Caxambu, Brazil.. In: NORM 8 - Naturally Occurring Radioactive Material, 2016, Rio de Janeiro. NORM 8 - Naturally Occurring Radioactive Material, 2016.
9. NEGRÃO, S.G. Exposição à baixas doses de radiações ionizantes decorrente dos radionuclídeos naturais Ra-226, Ra-228 e Rn-222 em Águas de Lindóia. São Paulo: Tese (Mestrado). Instituto de Pesquisas Energéticas e Nucleares. 2012.
10. DE JESUS, S.C. Levantamento dos níveis de radioatividade natural em águas do Alto Vale do Ribeira à Planície Costeira do litoral Sul do Estado de São Paulo. São Paulo: Tese (Mestrado). Instituto de Pesquisas Energéticas e Nucleares. 2009.
11. OLIVEIRA, J.; MAZZILLI, B. P. ; SAMPA, M. H. O. ; SILVA, B. . Seasonal variations of Ra-226 and Rn-222 in mineral spring waters of Aguas da Prata-Brazil. *Applied Radiation and Isotopes*, Oxford, v. 49, n.4, p. 423-427, 1998.
12. SILVA, L. S.; Avaliação da Radioatividade Natural em águas potáveis de superfície e subterrâneas da região de Caetité, BA. São Paulo: Tese (Mestrado). Instituto de Pesquisas Energéticas e Nucleares. 2011.
13. MAZZILLI, B. ; CAMARGO, I. M. C. ; OLIVEIRA, J. ; NIERI NETO, A. ; SAMPA, M. H. O. ; SILVA, B. . Dose evaluation due to ingestion of natural radionuclides of the uranium series in spring waters. *Radiation Research*, Oak Brook, v. 150. n.2, p. 250-252, 1998.
14. JACOMINO, V. M. F. ; BELLINTANI, S. A. ; OLIVEIRA, J. ; FIELDS, D. ; MAZZILLI, B. ; SAMPA, M. H. O. ; SILVA, B. . Estimates of cancer mortality due to

- ingestion of mineral spring waters from a highly natural radioactive region of Brazil. *Journal of Environmental Radioactivity*, Oxford, v. 33, n.3, p. 319-329, 1996.
15. OLIVEIRA, J.; MOREIRA, S. R. D. ; MAZZILLI, B. P. . Natural radioactivity in mineral spring waters of a highly radioactive region of Brazil and consequent population doses. *Radiation Protection Dosimetry*, Ashford, v. 55, n.1, p. 57-59, 1994.
 16. DAEE; IG; IPT; CPRM. Mapa de Águas Subterrâneas do Estado de São Paulo: Nota Explicativa. Governo do Estado de São Paulo. Conselho Estadual de Recursos Hídricos. 2005.
 17. INMET – INSTITUTO NACIONAL DE METEOROLOGIA. http://www.inmet.gov.br/portal/index.php?r=home/page&page=rede_estacoes_auto_graf. Accessed in 06/2019.
 18. IBGE – INSTITUTO BRASILEIRO DE GEOGRAFIA E ESTATÍSTICA. Censo Demográfico de 2010. Ministério do Planejamento, Orçamento e Gestão. 2010.
 19. SZIKSZAY, M.; TEISSEDRE, J. M. Fontes de Campos do Jordão. *Boletim IG*. Instituto de Geociências. Universidade de São Paulo. São Paulo, 10; 1-10. 1979.