2013 International Nuclear Atlantic Conference - INAC 2013
Recife, PE, Brazil, November 24-29, 2013
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-05-2**

DETERMINATION OF ^{226}Ra, ^{228}Ra, AND ^{210}Pb IN MUSHROOM FROM A NATURALLY HIGH RADIOACTIVE REGION

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ABSTRACT

Many studies have shown that mushrooms are organisms which efficiently accumulate radionuclides and can be used as indicators of environmental contamination and ecosystem quality. The Poços de Caldas plateau, in Minas Gerais, is a region that has elevated natural radioactivity due to the presence of radiological anomalies of volcanic origin. Seventy areas of radioactive anomalies have been identified in this region. From the radiological point of view the determination of ^{226}Ra, ^{228}Ra, and ^{210}Pb is relevant because they are decay products of the natural series of ^{238}U and ^{232}Th, mainly responsible for natural radioactive exposures of man. The present paper is part of a broader study conducted in the Poços de Caldas plateau, in which the concentration activities of ^{226}Ra, ^{228}Ra, and ^{210}Pb in mushroom samples were determined. The mushrooms were collected at different points of the plateau under the influence of radioactive anomalies and away from the influence of anomalies. From statistical studies a correlation between the accumulation of radionuclides in mushrooms and anomalies was established and it was possible to confirm the efficiency that the mushrooms present as environmental contamination indicators.

1. INTRODUCTION

The biological environment monitoring has demonstrated that many living organisms are particularly useful in the evaluation of pollution levels and the quality of ecosystems [1]. Of these organisms, you can highlight the crustaceans, fish and mushrooms that are useful to assess the levels of pollution and the quality of the ecosystem [2, 3]

Within the animal Kingdom, such species are considered "indicators", because their presence can indicate substances or elements present in large amounts in the environment, in other words, can be considered as bioindicators of occurrence of a particular element [4].

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The measurement of radioactivity in the environment and in food is extremely important to monitor radiation levels, that man can be exposed directly or indirectly. In particular, the mushrooms have the ability to retain important radionuclides of radiological and toxicological point of view, such as ^{137}Cs, ^{238}U, ^{232}Th, and ^{210}Pb, on the environment [5, 6].

Studies of high natural radioactivity regions have aroused a lot of interest from the point of view of radiation protection. In nature there are natural radioactive elements of high atomic number, which perform successive disintegrations and give rise to another also radioactive isotope and so on, until they reach a stable isotope. Before becoming stable, during the successive disintegrations, they tend to reach the radioactive equilibrium. In each of these successive decays the nuclei emit alpha or beta particles and gamma radiation. These nuclei are called sequences Radioactive Series, noted the existence of 3 of these series, which are: Uranium series, series of Thorium and Actinium series, starting with the ^{238}U, ^{235}U and ^{232}Th respectively. From a radiological point of view the natural radioactive series of ^{238}U and ^{232}Th are the main responsible for the exhibitions natural radioactive man on Earth, who contribute the most to the dose rate from natural sources [7].

For studies that assess the human exposure dose of natural radionuclides determination ^{226}Ra, ^{228}Ra, and ^{210}Pb is of great importance, since they are natural series decay products of ^{238}U and ^{232}Th [8].

The Poços de Caldas has the aspect of a boiler felled in a volcano, caused by tectonic events, processes of weathering and erosion. This plateau has several radioactive anomalies known worldwide, one of which, the Morro do Ferro, presents some of the highest rates of natural external dose in the world [9].

This work is part of a broader study conducted in the Highlands of Poços de Caldas, in which the concentration activity of ^{228}Ra, ^{226}Ra, and and ^{210}Pb were determined in samples of mushrooms, aiming to evaluate the behavior of mushrooms as indicators of radioactive contamination in the environmental in areas under the influence of radioactive anomalies and away from the influence of anomalies.

2. METHOD AND MATERIALS

2.1. Sampling

Twenty four wild mushroom samples were collected in different points on the plateau of Poços de Caldas (MG), a region with high natural radioactivity. The samples were collected in humid areas under trees or in open fields directly from the soil. The samples were between 1 to 15 cm in size. Fig. 1 shows the sites of collections in the Plateau and points of radioactive anomalies.

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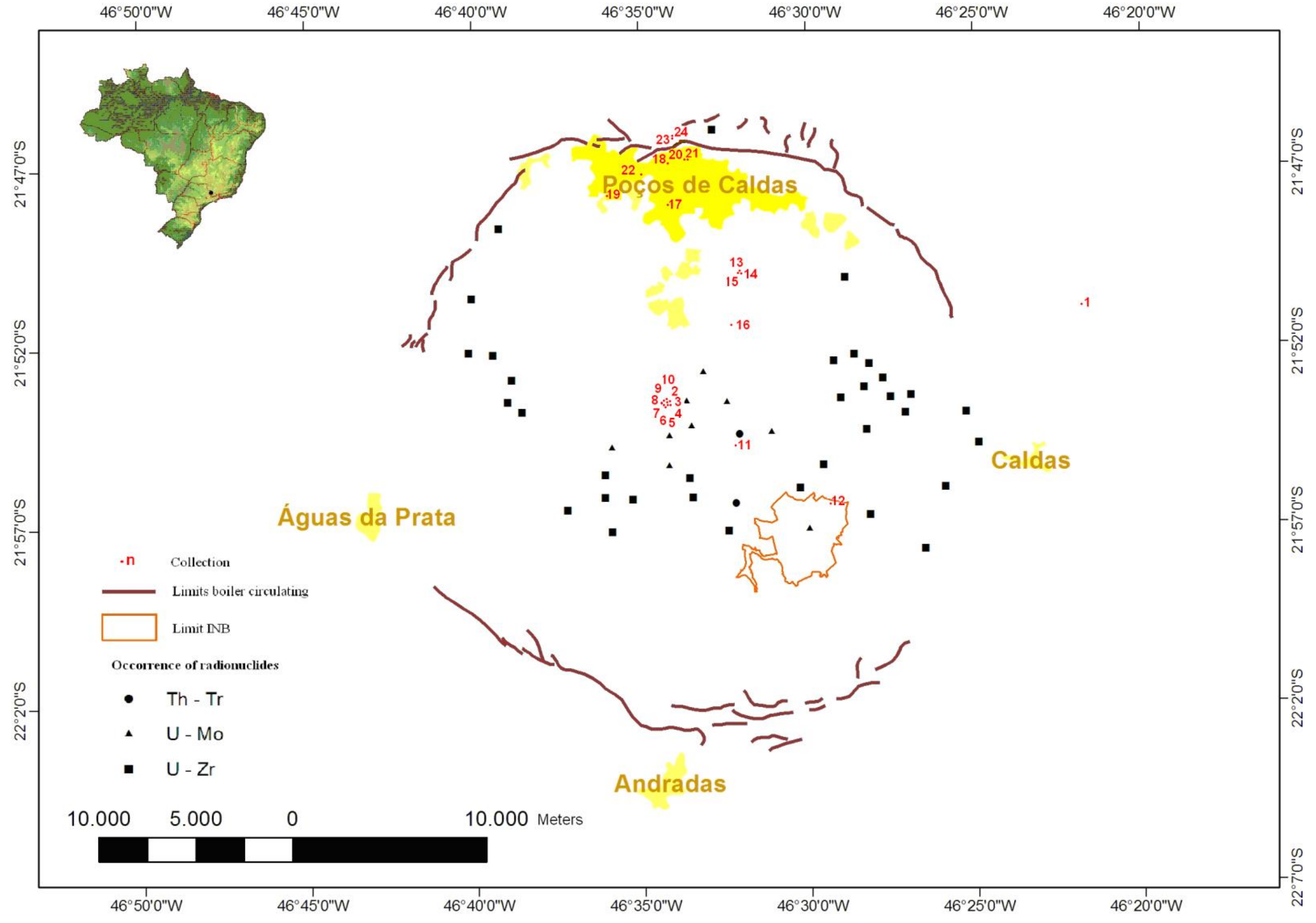

Figure 1: Collection sites in Poços de Caldas Plateau and points of radioactive anomalies

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In Tab. 1 sampling points are subdivided into groups.

Table 1: Subdivision of sampling points

Group	Samples	Place Approaching
I	1	Sítio Coroado – Caldas
II	2, 3, 4, 5, 6, 7, 8, 9 e 10	Campos do Agostinho
III	11	Morro do Ferro
IV	12	Indústria Nucleares do Brasil (INB)
V	13, 14, 15 e 16	Santuário Mariano Schoenstatt
VI	17, 18, 19, 20, 21 e 22	Urban zone Poços de Caldas
VII	23 e 24	Pedra Balão - Poços de Caldas

It is worth mentioning that the groups II and III were sampling points located next to regions that have natural radioactive anomalies of U and Th, known as Campo do Agostinho and Morro do Ferro, respectively.

2.2. Sample Preparation

The mushroom samples were washed with tap water and then with deionized water in order to eliminate impurities from the soil. Then, the samples were dried in an oven at 85 ^{0}C for 24 hours. After drying the samples were ground and sieved through a 100 mesh sieve.

Then approximately 5 g of each sample was burnt to ash at 450 °C for 24 hours in a muffle furnace. Then, the ash samples were dissolved with three acids (nitric, perchloric, and hydrofluoric). The solution was evaporated and reconstituted with nitric acid.

2.3. Radioisotopes Separation

The ^{228}Ra radioisotope was obtained by radiochemical separation, with Pb^{2+} (20 mg.mL^{-1}) e Ba^{2+} (20 mg.mL^{-1}) used as a carrier to determine loss during the analysis. The radionuclide was coprecipitated as $Ba(Ra)SO_4$ by pH change using 0.3M sulfuric acid. After that, alkaline

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redissolution was carried out and EDTA was added to the precipitate. The resulting solution was coprecipitated with another pH change using 5 mL of 25 mg/mL ammonium sulphate and concentrated glacial acetic acid and filtered in cellulose acetate membran. ^{226}Ra and ^{228}Ra in the precipitate was quantified in proportional low level counting using simultaneous alpha and beta channel.

The supernatant containing the ^{210}Pb, 1 M Na_2S was added to precipitate in the form of sulfide. The precipitate formed was dissolved with 5 M HNO_3 at where the formation of elemental sulphur and lead remains in solution. Then the solution is evaporated to dryness and the lead is finally precipitated in the form of $PbCrO_4$ with addition of purified water, 40% ammonium acetate and solution of 30% Na_2CrO_4.

The micro-precipitate formed, were filtered through a 0.45 µm Milipore membrane. The samples were stored for 30 days until counting to wait the secular equilibrium between ^{210}Pb and ^{210}Bi.

2.4. Radioisotopes Quantification

A Canberra S5-XLB Tennelec Total Alpha and Beta device with proportional gas flow was employed to quantify the samples of ^{226}Ra, ^{228}Ra, and ^{210}Pb.

3. RESULTS AND DISCUSSION

The process validation and quantification of radiochemical separation of ^{210}Pb, ^{226}Ra, and ^{228}Ra was validated through the analysis of reference Material IAEA-Soil-327 of the International Atomic Energy Agency. The results obtained are presented in Tab. 2.

Table 2: Results of ^{210}Pb, ^{226}Ra, and ^{228}Ra in IAEA-Soil-327

Radionuclide	Activity ± Uncertainty ($Bq.kg^{-1}$)	Certified Value ($Bq.kg^{-1}$)	Relative Error (%)
^{210}Pb	49,0 ± 7,0	58,8	16,7
^{226}Ra	36,5 ± 1,3	34,1	7,0
^{228}Ra	35,0 ± 4,0	38,7	9,6

The results obtained for analysis of reference material IAEA-Soil 327, show that the method applied to radionuclides analysis of ^{228}Ra, ^{226}Ra, and ^{210}Pb for radiochemical separation and counting of Total alpha and Beta emitters is reliable and accurate, relative error of less than 17%.

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The results obtained with the values of the standard uncertainties propagated in determinations of ^{226}Ra, ^{228}Ra, and ^{210}Pb radionuclides in samples of mushrooms are shown in Tab. 3.

Table 3: Radiometric Determination

Sample	^{210}Pb ± Uncertainty (Bq.kg^{-1})	^{226}Ra ± Uncertainty (Bq.kg^{-1})	^{228}Ra ± Uncertainty (Bq.kg^{-1})
1	20 ± 2	21 ± 2	33 ± 4
2	41 ± 5	43 ± 9	43 ± 6
3	11 ± 1	12 ± 2	12 ± 2
4	19 ± 2	24 ± 3	27 ± 6
5	22 ± 2	27 ± 4	27 ± 8
6	10 ± 1	30 ± 1	34 ± 8
7	34 ± 3	33 ± 6	30 ± 4
8	65 ± 6	27 ± 1	19 ± 4
9	64 ± 7	102 ± 9	120 ± 10
10	13 ± 1	8,0 ± 0,3	7,0 ± 0,2
11	11 ± 1	92 ± 6	112 ± 10
12	160 ± 10	170 ± 20	120 ± 10
13	40 ± 4	40 ± 10	48 ± 8
14	< 10	< 4	< 4
15	< 10	4,3 ± 0,2	< 4
16	230 ± 3	9 ± 1	11 ± 3
17	40 ± 4	35 ± 6	34 ± 4
18	36 ± 3	8 ± 1	40 ± 8
19	< 10	4,2 ± 0,6	< 4
20	25 ± 2	16 ± 1	13 ± 3
21	12 ± 1	6,0 ± 0,4	7,0 ± 0,2
22	13 ± 1	< 4	< 4
23	16 ± 2	8,0 ± 0,2	7 ± 2
24	< 10	< 4	< 4

The chemical recovery for samples of Ra was 73 to 99%, and for samples of Pb between 61 to 84%.

Evaluating the results obtained it was possible to observe a wide range of activities between the same radionuclides in different sampling locations, with most of the greater activity of samples ar isotopes relative to the including ^{210}Pb. This can be explained by the transport of including ^{210}Pb in the environment, which is mainly through the atmosphere ("fallout"), being transported in solution virtually negligible. While the Ra is primarily transported in solution, dispersing in the middle by water bodies [10].

Studies evaluated ^{210}Pb activities compared with those of ^{226}Ra in fungi collected in three regions of France, and including ^{210}Pb activities were higher than those of ^{226}Ra. This can be

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understood as ^{210}Pb present in the samples may not be just the result of soil to transfer the mushroom, but also from direct deposition on the mushroom of the atmosphere [11]. Similar behavior was also observed in some sampling points in this work, as in nearly all specimens of Group VI, collected in the urban area of the plateau of Poços de Caldas.

3. CONCLUSIONS

With the studies of this study it was possible to emphasize the important role of the mushroom as an indicator for environmental monitoring. As well as the importance of determination of radionuclides in an environment that has high natural radioactivity, as part of the natural radioactive series.

ACKNOWLEDGMENTS

The authors would like thank the National Commission of Nuclear Energy – Laboratory of Poços de Caldas (CNEN/LAPOC), the National Development Council Scientific and Technological (CNPq) and the Technological Service Network of Radiation Protection and Dosimetry (SIBRATEC) by financial support.

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