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DETERMINATION OF ^{228}Th, ^{232}Th, and ^{228}Ra IN WILD MUSHROOM FROM A NATURALLY HIGH RADIOACTIVE REGION IN BRAZIL

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ABSTRACT

Mushrooms are fungi which efficiently accumulate radionuclides, as verified by radiochemistry analyses of specimens collected in contaminated areas, specifically after the Chernobyl nuclear accident. Many studies have demonstrated that mushrooms can be used in monitoring of ecosystem contamination and quality. The present paper is part of a broader study conducted in the Poços de Caldas plateau region in Minas Gerais, Brazil, investigating assimilation of natural Uranium and Thorium radionuclide series by mushrooms. This region has elevated natural radioactivity due to the presence of radiological anomalies of volcanic origin. These anomalies are ore bodies containing Uranium and Thorium, the later being highly predominant. Many researchers have been conducted concerning radionuclide incorporation by agricultural products on the plateau. The present paper aims to determine ^{232}Th, ^{228Th}, and ^{228}Ra radionuclides in wild mushrooms collected at different locations in the plateau region. ^{228}Ra was determined by radiochemical separation using sulphate coprecipitation followed by beta radiometry. ^{232}Th and ^{228}Th were determined using anion exchange resin purification followed by alpha spectrometry. Higher values were obtained to ^{228}Th than to ^{232}Th. This is due to higher ^{228}Ra mobility, which decays to ^{228}Th. The accuracy of the analytical methods employed was evaluated using the reference sample IAEA Soil 327. These methods had high chemical recovery and high sensitivity. It was possible to confirm that mushrooms accumulate radionuclide and so can be used in environmental contamination and quality assessment.

Keywords: mushroom, naturally radioactive, Thorium-228, Thorium-232, and Radium-228.

1. INTRODUCTION

The measurement of the radioactivity in the environment and in foods is extremely important to monitor levels of radiation to which man can be directly or indirectly exposed. In particular, different species of fungi, including mushrooms, have the capacity to retain radionuclides from the environment, which are relevant regarding doses received by human beings, such as ^{137}Cs, ^{238}U, ^{232}Th, and ^{210}Pb [1; 2; 3; 4].

The transfer of radionuclides from the soil to organic materials determines the extent of radioactive contamination of food and plants and thus the risk of radiation exposure of the population due to food intake. Radiological studies have shown that some ecosystems have conditions that favor transfer of radionuclides from the soil to organic material [5; 6].

The Poços de Caldas plateau, located in Minas Gerais state, Brazil, is an example in which deep and almost complete decomposition of veins and rocks in subtropical conditions, along with weathering, has produced around 70 areas with radioactive anomalies with naturally high radiation levels [7].

There are many areas in the world that have the characteristic of naturally high radioactivity, which are called Naturally Occurring Radioactive Materials, NORM, caused either by geological and geochemical structures in the soil or by the level of radioactivity in the water that flows from hot water springs, or even due to cosmic rays [8].

Thorium is a naturally occurring radioactive element that is widely distributed in the crust of the Earth. This element is very common in mineral formations in regions with high levels of radioactivity [9].

Due to the naturally high level of radioactivity in the Poços de Caldas plateau region, the objective of the present study was to determine radionuclides of the Throrium series, that is, ^{232}Th, ^{228}Th and ^{228}Ra, in wild mushroom samples collected from several different points on the plateau and to verify the behavior of the mushrooms as indicators of radioactive contamination in the environment.

2. MATERIALS AND METHOD

2.1. Wild mushroom sampling

Wild mushroom samples were collected at several different locations on the Poços de Caldas plateau (state of Minas Gerais, Brazil), a region with naturally high radioactivity. Samples were collected from humid areas under trees or in fields, directly from the soil. The mushrooms varied from 1 to 15 cm in length.

Natural radioactivity was also measured in the sampling areas using a radiation detector and the coordinates were determined using a GPS device. A radioactive anomaly was found at one of the sample points; the sample was identified as sample number 3.

2.2. Preparation of the sample for analysis

Wild mushroom samples were washed with tap water and then deionized water in order to eliminate impurities collected along with the samples. Samples were then dried in an oven at 60 °C for 24 hours. After drying, samples were ground and sieved with a 100 mesh. Then approximately 5 g of each sample was burnt to ash at 450 °C for 24 hours in a muffle furnace.

For radiochemical separation of the radionuclides a ^{229}Th tracer was initially added to determine thorium isotopes and a ^{133}Ba carrier was added to determine radium-228. Then, the ash samples were dissolved with three acids (nitric, perchloric, and hydrofluoric). The solution was evaporated and reconstituted with 8M nitric acid. The solution was divided into two parts in order to determine Th isotopes and ^{228}Ra.

2.3. Separation of Th isotopes

Radiochemical Th separation was carried out using Dowex 1x2 anionic exchange resin pre-treated with 8M nitric acid. The nitric solution was percolated in Dowex 1x2 resin and Th was eluted with concentrated hydrochloric acid. The eluted solution was dried on a hot plate and reconstituted with 0.3 mL of concentrated sulfuric acid, 1 mL of 0.3M sodium sulfate and 4 mL of deionized water.

2.4. Quantification of Th isotopes

Thorium radioisotopes were electrodeposited on polished silver plates with a 1.0 A electrical current for 60 minutes in the final solution. For quantification of thorium isotopes in the samples, an Alpha Spectrometer from Canberra Inc., Alpha Analyst Model, with surface barrier detectors with active area of 450 mm^2 was used. The efficiency of the detector was 0.186 ± 0.005 (counts per second.Bq^{-1}), tested using SRS 63997-121 standard mixed alpha emitter source. The count time was 200.000 seconds.

2.5. ^{228}Ra radioisotope separation

The ^{228}Ra radioisotope was obtained by radiochemical separation, with ^{133}Ba 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 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. The micro-precipitate formed, $Ba(Ra)SO_4$, was filtered through a 0.45 µm Milipore membrane. The samples were stored for 20 days until counting.

2.5. Quantification of ^{228}Ra

A Canberra S5-XLB Tennelec Total Alpha and Beta device with proportional gas flow was employed to quantify the samples.

3. RESULTS AND DISCUSSION

Results obtained for ^{228}Th, ^{232}Th and ^{228}Ra determination are presented in Table 1. To quantify thorium, the samples were counted for 200.000 seconds at 5.43 MeV for the ^{228}Th , 4.84 MeV for the tracer ^{229}Th and 4.01 MeV for ^{232}Th. Chemical recovery of the analyses varied from 58 to 100%. For quantification of ^{228}Ra, samples were counted for 120 minutes through ^{228}Ac coincident alpha and beta emissions at 911 and 969 keV. Chemical recovery of this analysis for the samples varied from 76 to 99%.

From the results, it can be observed that values obtained to ^{228}Th were higher than those to ^{232}Th. This imbalance could be explained by the fact that ^{228}Th originates from ^{228}Ra decay and this is the result of faster and more preferential absorption by mushrooms than ^{232}Th.

Table 1. Results obtained for ^{228}Th, ^{232}Th, and ^{228}Ra determination in wild mushrooms

Sample	^{228}Th (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	^{228}Ra (Bq.kg^{-1})
1	42 ± 2	40 ± 2	33 ± 4
2	63 ± 3	51 ± 2	43 ± 6
3	124 ± 13	97 ± 7	112 ± 10
4	4.1 ± 0.4	3.3 ± 0.3	1.2 ± 0.2
5	28 ± 1	28 ± 1	34 ± 4
6	160 ± 8	142 ± 7	120 ± 10
7	24 ± 5	22 ± 3	27 ± 6
8	28 ± 1	23 ± 4	27 ± 8
9	3.1 ± 1.0	2.6 ± 0.4	40 ± 8
10	43 ± 2	36 ± 2	30 ± 4
11	52 ± 3	49 ± 2	48 ± 8
12	1.7 ± 0.5	0.6 ± 0.1	1.7 ± 0.5
13	11 ± 1	9 ± 1	13 ± 3
14	127 ± 4	113 ± 4	120 ± 10

The methods described in this study were applied to a reference soil sample from the International Atomic Energy Agency (IAEA Reference Soil 327; 31/12/1994 reference date). Results obtained for the reference material are shown on Table 2. The results obtained are in agreement with the certified values. It was thus possible to confirm the validity of both radiochemical separations employed.

Table 2. Analysis of IAEA reference material: Reference Soil 327

Radionuclide	Reference (Bq.kg^{-1})	Analyzed (Bq.g^{-1})
^{228}Th	38.2 ± 1.0	37.0 ± 1.7
^{232}Th	38.7 ± 1.5	37.6 ± 1.7
^{228}Ra	38.7 ± 0.9	34.8 ± 3.5

Figs. 1 and 2 show the alpha spectra of Thorium of the one sample collected in the Poços de Caldas region and the reference material after radiochemical separation, respectively. The radiochemical separation efficiency for Thorium isotopes can be seen by the Figures, since Th photopeaks are only observed.

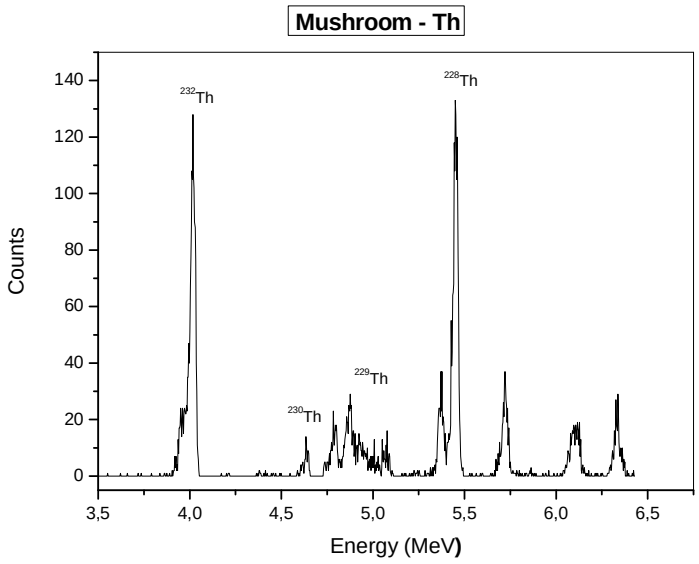

Figure 1. Alpha spectrum for a mushroom sample.

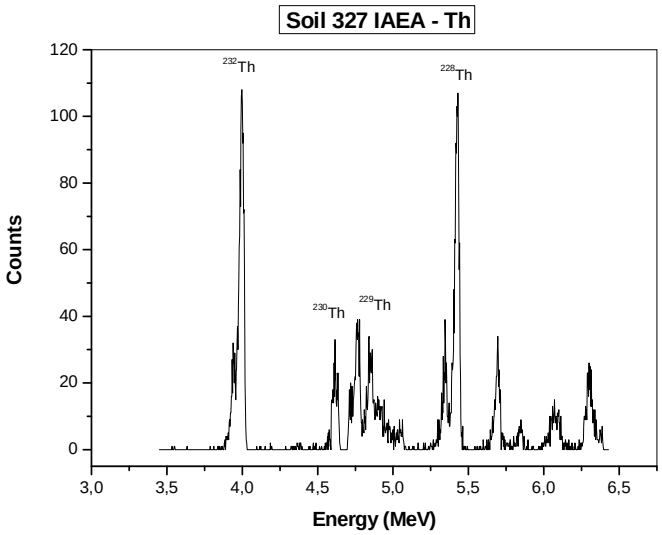

Figure 2. Alpha spectrum for reference material.

Baeza and col. [10] determined Cs, K, Pu, Ca, Th, U, and Ra in different mushroom parts from the *Pleurotus eryngii* species and evaluated that the radioactive disequilibrium between isotopes ^{228}Th and ^{232}Th is directly associated with ^{228}Th, which originate from the decay of ^{228}Ra, as observed by this study.

Taddei and col. [9] determined uranium and thorium isotopes in plants and feces from the population of the Poços de Caldas plateau using coprecipitation and ion exchange separation resins and proved the sensitivity of the method and applicability of plants as environmental monitors.

Linsalata and. Col. [11] evaluated the ingestion of thorium and rare earth elements in the population of Poços de Caldas by means of feces samples and observed that ingestion of these elements is within the value that is set by the Brazilian Radiological Protection and Analysis, which is 0.9 µg /day [12].

The results obtained in this study show that the Poços de Caldas plateau can be considered a NORM region with some points presenting very high levels of naturally occurring radioactivity, which were found in several mushroom samples analyzed.

4. CONCLUSIONS

The study was able to determine radionuclides from the Thorium series, ^{232}Th, ^{228}Th, and ^{228}Ra in wild mushroom samples collected from several different locations on the Poços de Caldas plateau, a region with naturally high levels of radioactivity. From the results obtained, mushrooms could be used as indicators of radioactive contamination in the environment.

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