2021 International Nuclear Atlantic Conference – INAC 2021
Virtual meeting, Brazil, November 29 – December 2, 2021

ISBN: 978-65-594-1256-3

Determination of 238-U and 232-Th in soil samples collected near the Amazon Tall Tower Observatory, Brazilian Amazon

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1. Introduction

Uranium is an element present in all types of rocks, water and soil. Uranium is the 51st most abundant element in the Earth's crust, with an average concentration about 2 to 4 parts per million [1]. The most abundant radioisotope of U (^{238}U) forms in its decay series the radionuclides ^{226}Ra and ^{222}Rn.
The ^{222}Rn gas has a half-life of 3.82 days, which is comparable to the lifespan of certain atmospheric species (e.g., O3, NOx, CO). In addition, ^{222}Rn is an inert gas and it is relatively easy to measure. These properties make it an interesting tracer for atmospheric studies. Thus, ^{222}Rn is used in global climate models, chemical transport models and also to estimate regional emissions of CO_2 and CH_4 [2, 3]. Finally, in the context of public health knowledge about the ^{222}Rn flux is important for assessing the exposition of human to this radioactive gas [4].
Since ^{238}U is the primary radionuclide in the natural series that produces the ^{222}Rn, the activity concentration of ^{238}U is important for some ^{222}Rn radon flux map modeling methodologies [2, 5].
This study aims to determine the ^{238}U and ^{232}Th activity concentrations in soil samples collected in the Uatumã Sustainable Development Reserve (SDR), an area of dense tropical rainforest where the Amazon Tall Tower Observatory (ATTO tower) is located. The X-ray diffraction technique was applied to assess the homogeneity of soil composition.

2. Methodology

The Uatumã SDR is located in the São Sebastião do Uatumã city, State of Amazonas. The samples were collected in the terrace site of the SDR (the region near the Uatumã river). Five holes were dug: central, north, south, east and west. The distance between the central hole and the others was approximately 10 meters. In each hole, samples were collected from three depths: 5 cm, 30 cm and 50 cm, totaling 15 samples. The samples were transported to the State of São Paulo and prepared to be analyzed using the Instrumental Neutron Activation Analysis (INAA) technique. The sample preparation procedure consisted of leaving them in an oven at 343 K to eliminate the interstitial water, with subsequent grinding in an agate mortar and pestle. The ground soil samples were sieved, homogenized and stored in decontaminated flasks.

After proper preparation, approximately 120 to 150 mg of samples and certified reference materials (CRM) were weighted into 1.8 x 1.8 cm polyethylene bags. The U and Th content was determined by the INAA by the comparative method. The mass fraction of these actinides was converted into activity concentrations of ^{238}U and ^{232}Th.

Both pipetted standards prepared from standard solutions (Spex CertiPrep) and the SRM 1646a Estuarine Sediment and USG STM-2 Syenite (Table Mountain) were used as comparators.

They were irradiated in the IEA-R1 nuclear reactor of the Nuclear and Energy Research Institute (IPEN/CNEN). The irradiation batch were left in the water-cooled irradiation element by approximately 8 hours under a thermal neutron flux between $2 - 5\ 10^{12}$ cm^{-2} s^{-1}.

After irradiation, the content of U and Th elements were determined by gamma spectrometry. To quantify the U and Th mass fractions, ^{239}Np (228,18 and 277,60 keV) and ^{233}Pa (312,18 keV) radionuclides were used, respectively [6].

3. Results and Discussion

The reference materials analyzed under the same conditions as the samples were used for quality control of the analysis. From the obtained and certified values of the CRM, it was possible to apply the z-score from the modified Horwitz function [7]. The mathematical model of the modified Horwitz function does not depend on the uncertainty of the CRM, which is unavailable for the informative values of the CRM used. The Table I presents the quality control values.

Table I: Mass fraction obtained by INAA (mean values and expanded uncertainties) and the certified values of reference materials

Analysis of certified reference materials		Certified Reference Materials			
		USGS STM-2 (n = 3)		SRM 1646a (n = 2)	
		U[i]	Th	U[i]	Th[i]
Obtained value (Certified value)	(mg kg^{-1})	7.23 ± 0.28	28.1 ± 2.3	1.87 ± 0.09	5.81 ± 0.37
		(7.6)	(27 ± 5)	(2)	(5.8)
Z-score		-0.43	0.42	-0.47	0.01

[i]informative value.

The criterion for considering the z-score value being satisfactory is $|z| \leq 2$ [8]. Despite the small number of CRM analyzed, all z-scores values obtained in this study satisfy this condition and therefore, the methodology used for the analysis of these actinides can be considered adequate.

The Table II presents the results of ^{238}U, ^{232}Th activity concentration and the ^{238}U/^{232}Th ratio of the samples collected in the holes: center (CH), east (EH), west (WH), north (NH) and south (SH). Samples collected in the depths of 5, 30 and 50 cm are indicated by 5, 30 and 50 after the hole identification.

Several factors can influence the ^{222}Rn exhalation rate from the land: soil type, moisture content, bulk density and the ^{238}U content [2 ,5]. The soil samples analyzed in this work were predominantly sandy.

Figure 1 shows the results obtained using the X-ray diffraction technique. In the qualitative analysis, the presence of quartz (ICSD #152), kaolinite and goethite were identified. The obtained diffractograms show that the analyzed soil samples are homogenous, which indicates that the exhalation of ^{222}Rn from the soil should not be affected by a heterogeneity of the soil composition.

The reported ^{238}U and ^{232}Th activity concentrations in this work are intended be used as a proxy for ^{222}Rn exhalation rate in Uatumã SDR, where the Amazon Tall Tower Observatory is located.

Table II: ^{238}U and ^{232}Th activity concentration in the soil samples collected at Uatumã SDR.

Soil sample identification	^{238}U, Bq kg^{-1}	^{232}Th, Bq kg^{-1}	$^{238}U/^{232}Th$
Center hole – 50	69.3 ± 3.0	90 ± 17	0.77
Center hole – 30	74.9 ± 2.4	76 ± 4	0.98
Center hole – 5	67.4 ± 2.9	81 ± 15	0.83
North hole – 50	85.2 ± 3.2	101 ± 19	0.84
North hole – 30	50.2 ± 1.5	77 ± 4	0.65
North hole – 5	64.4 ± 1.7	79 ± 15	0.82
South hole – 50	75.3 ± 2.0	93 ± 17	0.81
South hole – 30	74.6 ± 2.4	110 ± 6	0.68
South hole – 5	71.1 ± 4.9	100 ± 8	0.71
East hole – 50	87.3 ± 5.8	113 ± 10	0.77
East hole – 30	87.1 ± 2.3	114 ± 6	0.77
East hole – 5	64.8 ± 2.5	93 ± 8	0.69
West hole – 50	73.2 ± 2.7	108 ± 9	0.68
West hole – 30	69.7 ± 2.2	100 ± 5	0.69
West hole – 5	65.6 ± 2.5	102 ± 8	0.64

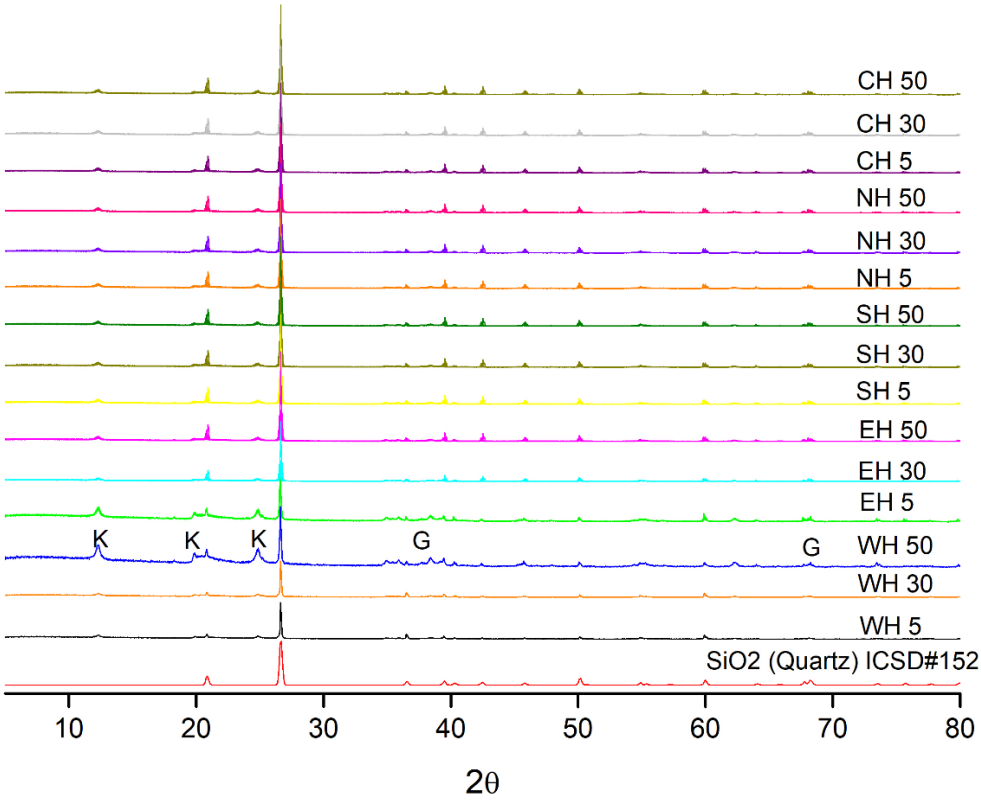

Figure 1: Diffractograms obtained for the soil samples analyzed in this study, where (K) means kaolinite and (G) means goethite.

4. Conclusions

The activity concentrations of ^{238}U and ^{232}Th in soil samples collected near the ATTO tower were estimated from the U and Th contents determined by the INAA comparative method. The z-scores values obtained indicate that the methodology used for the determination of U and Th is adequate. The activity concentrations of determined radionuclides can be used as proxies of the ^{222}Rn that exhales from soil. X-ray diffraction analysis showed that the soil sample composition is homogenous. In the future, more soil samples collected in other regions of SDR will be analyzed, thus enabling a better understand of the distribution of these radioisotopes in the soil of this region.

Acknowledgements

Authors acknowledge the financial support provided by the Brazilian National Nuclear Energy Commission (CNEN), the National Institute for Amazonian Research (INPA), the Max Planck Institutes, the Coordination of Superior Level Staff Improvement (CAPES) for the doctoral scholarship (process number 88887.577746/2020-00) and the Brazilian National Council for Scientific and Technological Development for the doctoral scholarship (process number: 161797/2021-0).

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