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Activity Concentrations of Natural Radionuclides in Commercial Granite Samples from Espírito Santo State, Brazil

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Abstract
Natural radioactivity, namely ^{226}Ra, ^{232}Th and ^{40}K, was determined in commercial granite samples of 6 quarries of Espírito Santo State, southeast Brazil, located in regions of municipalities Ecoporanga, Nova Venécia, Colatína, Afonso Cláudio, Castelo and Mimoso do Sul. All samples were measured by high resolution gamma spectrometry after a 30-days ingrowth period. The results show concentrations varying from 40±5 Bq·kg^{-1} to 225±20 Bq·kg^{-1} for ^{232}Th, from 20±5 Bq·kg^{-1} to 275±15 Bq·kg^{-1} for ^{226}Ra and from 520±20 Bq·kg^{-1} to 1405±60 Bq·kg^{-1} for ^{40}K. The radium equivalent index, considering the activity concentrations, ranged from 190±60 Bq·kg^{-1} to 475±70 Bq·kg^{-1}. The southern region of Espírito Santo State (Afonso Claudio, Castelo and Mimoso do Sul) shows the highest values concentrations for ^{226}Ra, ^{232}Th and ^{40}K, while the lowest values were observed for the central and north region (Colatina, Nova Venécia and Ecoporanga).

Keywords
Espírito Santo Granites, Gamma Spectrometry; ^{226}Ra, ^{232}Th, ^{40}K, Ra_{eq}

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1.0 Introduction

The main external source of ionizing radiation exposure to the human body are the naturally occurring radioactive elements in the soils and rocks, namely ^{40}K and the radionuclides from the ^{238}U and ^{232}Th series originated in the earth's crust, present everywhere in the environment [1].

Granites used as finishing material for civil construction are well known for their high natural radioactivity content, depending on the geological and geographical conditions of the quarries locations. Granites are formed from igneous rocks or silicate metamorphic rocks. In Brazil, the geological characteristics of several Espírito Santo State quarries make feasible the assumption of significant levels of natural radioactivity in their granite products. As Espírito Santo State manufacturers are responsible for more than 60% of the improvement, production and export of all Brazilian granites [2], in order to evaluate radiation hazards to mankind using those products, it is very important to determine the radioactivity content in those granites.

The objective of this work is to determine the concentration of natural radionuclides in commercial granites from Espírito Santo state and evaluate the applicability of those materials. The natural radioactivity in commercial granite samples of 6 quarries from State of Espírito Santo, southeast Brazil, was determined by measuring the ^{226}Ra (from the ^{238}U series), ^{232}Th

(from the ^{232}Th series) and ^{40}K concentration activities. The assessed quarries are located in the pre-Cambrian bulk, corresponding to the Ecoporanga and Nova Venêcia municipality, located in the north region, Colatina municipality, located in the central region and Afonso Claudio, Castelo and Mimoso do Sul municipality covering the south region of the Espírito Santo.

2.0 Materials and Methodology

2.1. Sampling Collection and Preparation

In the samples collection we considered the locations throughout the mountain chain of Espírito Santo state, southeast of Brazil, using as choice criterion the activity of commercial granite extraction for the exportation and application in the civil construction. The samples, as are resold, had been provided by the responsible companies for the extraction in these localities. The selected locations are show in Fig.1.

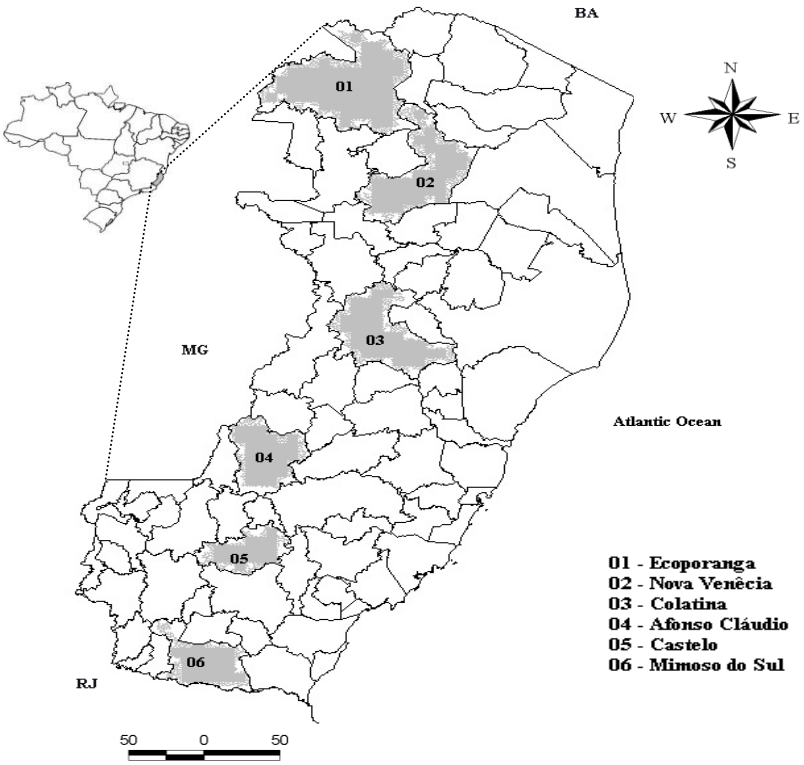

Fig. 1 - Map of geographic localization of the regions throughout mountainous chain of the Espírito Santo state, Brazil. ID locations of the collected samples are numbered from 1 to 6 (see Table 1).

All granites samples had been homogenized by spraying about 270 meshes in a tungsten carbide ring mill.

Each sample was sealed in a standard 100-mL HDPE flat-bottom cylindrical flask with 52.5mm plan screw cap and bubble spigot and stored for approximately 4 weeks before counting, in order to allow the reaching of secular equilibrium in the ^{238}U and ^{232}Th series [3]. For each location, three samples were prepared.

2.2. Measurements

All samples were measured during 150000s by high resolution gamma spectrometry with a coaxial high-purity germanium detector (HPGe) of 15% relative efficiency with conventional electronics and a 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer. The detector is 10-cm thickness lead shielded. The measured resolution for the ^{60}Co 1332.5 keV is 2.8 keV. All spectra were analyzed with the InterWinner 6.0 software [4]. The detector efficiency curve was determined with a multielement gamma standard solution, for the same sample geometry. The background radiation was determined by measuring a sample of high pure water in the same sample geometry.

^{226}Ra activity concentration was determined as the weighted mean from the average concentrations of the ^{214}Pb gamma transitions of 295.21 keV and 351.92 keV and ^{214}Bi gamma transition of 609.32keV. ^{232}Th activity concentrations was determined as the weighted mean from the average concentrations of the ^{228}Ac gamma transitions of 911.07 keV and 968.9 keV, ^{212}Pb gamma transitions of 238.63 keV and 300.9 keV and ^{212}Bi gamma transition of 727.33 keV. The concentration of ^{40}K was determined by its single gamma transition of 1460.83keV [4].

All InterWinner activities results had been corrected by a self-attenuation factor, as the samples apparent densities (around 2 g.cm^{-3}) were higher than the apparent density of the radioactive standard solution used for the efficiency curve [5]. Also, the samples composition can eventually modify the self-attenuation factors [6-7].

2.3. Radium Equivalent Activity

As the distribution of radioactivity in natural samples is not uniform, in order to assess the health effects from the natural radioactivity, the activity of ^{226}Ra, ^{232}Th and ^{40}K are converted into a single quantity termed Ra_{eq} (Radium equivalent), assuming that 370 Bq·kg^{-1} of ^{226}Ra, 259 Bq·kg^{-1} of ^{232}Th and 4810 Bq·kg^{-1} of ^{40}K produce an equal gamma ray dose rate [8].
The Ra_{eq} index is calculated by the expression:

$$Raeq = 370\left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations in Bq·kg^{-1}, respectively for ^{226}Ra, ^{232}Th and ^{40}K.

The recommended value for residential applications of building materials should not exceed the limit of 370 Bq·kg^{-1} [9-10]. Other limits and their respective recommendations are shown in Table 1 [11].

Table 1 – Suggested limits of radium equivalent for building materials [11]

Ra_{eq} ($Bq \cdot kg^{-1}$)	Class	Recommendation
<370	1	Residential
370-740	2	Industry
740-2220	3	Roads and bridges
2220-3700	4	Non-residential or foundations
>3700	5	No possible applications.

3.0 Results and Discussion

3.1. Activity Concentrations in Granites of Espírito Santo State

The average concentrations values of ^{40}K, ^{232}Th and ^{226}Ra are presented in Table 2 and summarized in Fig. 2, for easier contemplation.

Table 2 – Activity concentrations for the studied locations in Espírito Santo State and literature values

Commercial name	Municipality	ID	^{226}Ra ($Bq.kg^{-1}$)	^{232}Th ($Bq.kg^{-1}$)	^{40}K ($Bq.kg^{-1}$)	Ra_{eq} ($Bq.kg^{-1}$)	Ref.
Sta. Cecília	Ecoporanga	1	20 ± 5	70 ± 10	1185 ± 50	210 ± 60	*
Branco Primata	Nova Venêcia	2	20 ± 5	70 ± 10	945 ± 40	190 ± 60	*
Preto São Gabriel	Colatina	3	20 ± 5	70 ± 10	990 ± 40	195 ± 60	*
Iberê Crema Bordeaux	Afonso Claudio	4	275 ± 15	40 ± 5	520 ± 20	370 ± 50	*
Cinza Corumbá	Castelo	5	45 ± 5	225 ± 20	1405 ± 60	**475 ± 70**	*
Cinza Andorinha	Mimoso do Sul	6	40 ± 5	150 ± 10	970 ± 40	330 ± 50	*
Average Values							
	Espírito Santo		69 ± 4	103 ± 11	1003 ± 42	296 ± 58	*
	Jordania		41.52 ± 3.23	58.42 ± 0.44	897 ± 43	194.13	[7]
	China		112	71.5	672		[12]
	Egypt		187 ± 90	118 ± 14	852 ± 297		[13]
	Italy		153 ± 13	360 ± 30	1600 ± 100		[14]
	Worldwide		50	50	500	370	[1]

* Present Work
Uncertainties are given with ±1σ (k=1)
Highlighted: Radium equivalent largest value.

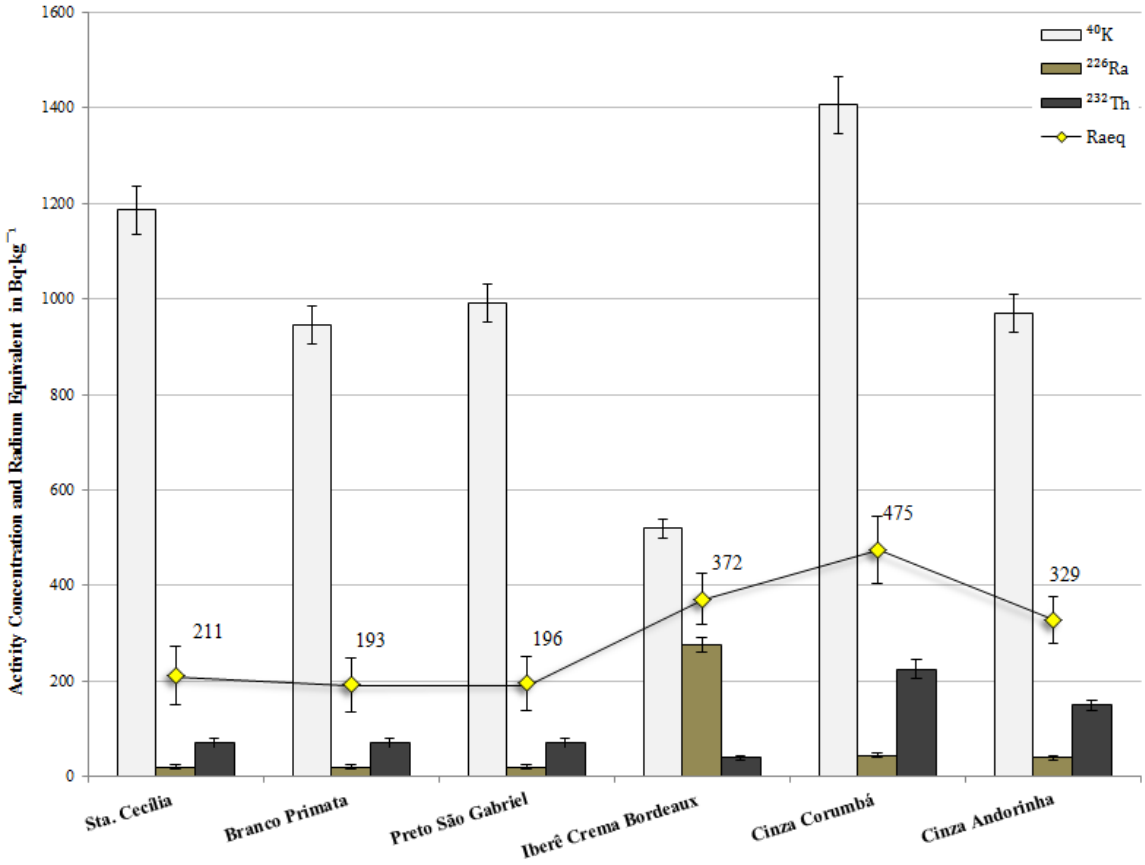

Fig. 2 - ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations and Ra_{eq} values for the studied locations

The southern region of Espírito Santo State (ID: 4, 5 and 6) shows the highest values for ^{226}Ra, ^{232}Th and ^{40}K. The lowest values of concentration for the same radionuclides were observed for north and central regions (ID:1, 2 and 3) of the Espírito Santo State. The radium equivalent index, considering the activity concentrations, ranged from 190±60 Bq·kg^{-1} to 475±70 Bq·kg^{-1}.

4.0 Conclusions

The Cinza Corumbá granite (ID 5) exceeded the proposed limit for a residential application however, can be used for industrial applications in according with OECD proposed limits.[9] Due to proximity to the lower limit for class 2, applications can be tolerated in cases where the use is designed for a low human permanence area, as squares, lounges, decorative boards and monuments, and others with a same purpose.

The average values for the granites assessed for the Espírito Santo State are in accordance with other values found in the literature for other places [7,12-14] and the World average [1]. (see Table 2)

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