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NATURAL RADIOACTIVITY ANALYSIS IN COMMERCIAL MARBLE SAMPLES OF SOUTHEAST REGION IN ESPÍRITO SANTO STATE: PRELIMINARY RESULTS

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ABSTRACT

The natural radioactivity in commercial marble samples of 6 quarries in “Cachoeiro do Itapemirim” and “Castelo” municipalities of the south region of Espírito Santo State, southeast Brazil, was determined from the ^{226}Ra, ^{232}Th and ^{40}K contents. In “Cachoeiro do Itapemirim” municipality were assessed the localities of “Alto Moledo”, “Baleeira”, “Alto Gironda” and “Itaóca” and, in “Castelo” municipality, was assessed the locality of “São Cristovão”. Three samples of each quarry were sealed in standard 100-mL HDPE polyethylene flasks and stored in order to obtain secular equilibrium in the ^{238}U and ^{232}Th series. All samples were measured by high resolution gamma spectrometry after a 30-days ingrowth period. Preliminary results show concentrations varying from 1,0 ± 0,2 Bq.kg^{-1} to 6,5 ±0,8 Bq.kg^{-1} for ^{226}Ra, from 1,5±1,2 Bq.kg^{-1} to 3,2±1,7 Bq.kg^{-1} for ^{232}Th and from 5 ± 4 Bq.kg^{-1} to 27 ± 5 Bq.kg^{-1} for ^{40}K.

1. INTRODUCTION

The main external source irradiation 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].

Marbles 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. The Espírito Santo State is responsible for more than 60% of the improvement, production and export of the ornamental rocks. Previous measurements in granites showed significances values of activity concentration for ^{226}Ra, ^{232}Th and ^{40}K [2]. So, it is very important to know the radioactivity content of these commercial marbles, in order to evaluate the radiation hazard in these areas.

The natural radioactivity in commercial marble 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 and ^{40}K concentration activities.

The sampling region is formed in its majority for magmatic rocks of composition felsic and mafic (classification PN3) and arenaceous sediments at argil-carbonatic of metamorphic degree weak at medium (classification PN1) and, in short regions for gneiss rocks of magmatic origin and/or sedimentary of average metamorphic degree and granitic rocks developed during the tectonic period (classification PP1). The classification follows the Brazilian Institute of Geography and Statistics - IBGE, [3].

Marbles are formed from metamorphic rocks originated from calcareous submitted the high temperatures and pressures, founded in regions of calcareous matrix-rocks and volcanic activities. In Espírito Santo State, the marble extractions are localized in the south central region due to geological characteristics of this place.

The objective of this work is to determine the concentration of natural radionuclides in commercial marbles the Espírito Santo state, neglected in earlier studies by other researchers.

2. MATERIALS AND METHODOLOGY

2.1. Sampling Collection and Preparation

In the samples collection was considered the locations throughout the mountain chain of Espírito Santo state, having as choice criterion the activity of commercial marble extraction for the exportation and application in the civil construction. The selected locations are show in Fig. 1. The samples had been granted as resold by the companies responsible for the extraction in these localities. The samples had been homogenized by spraying about 270 meshes in a tungsten carbet ring mil.

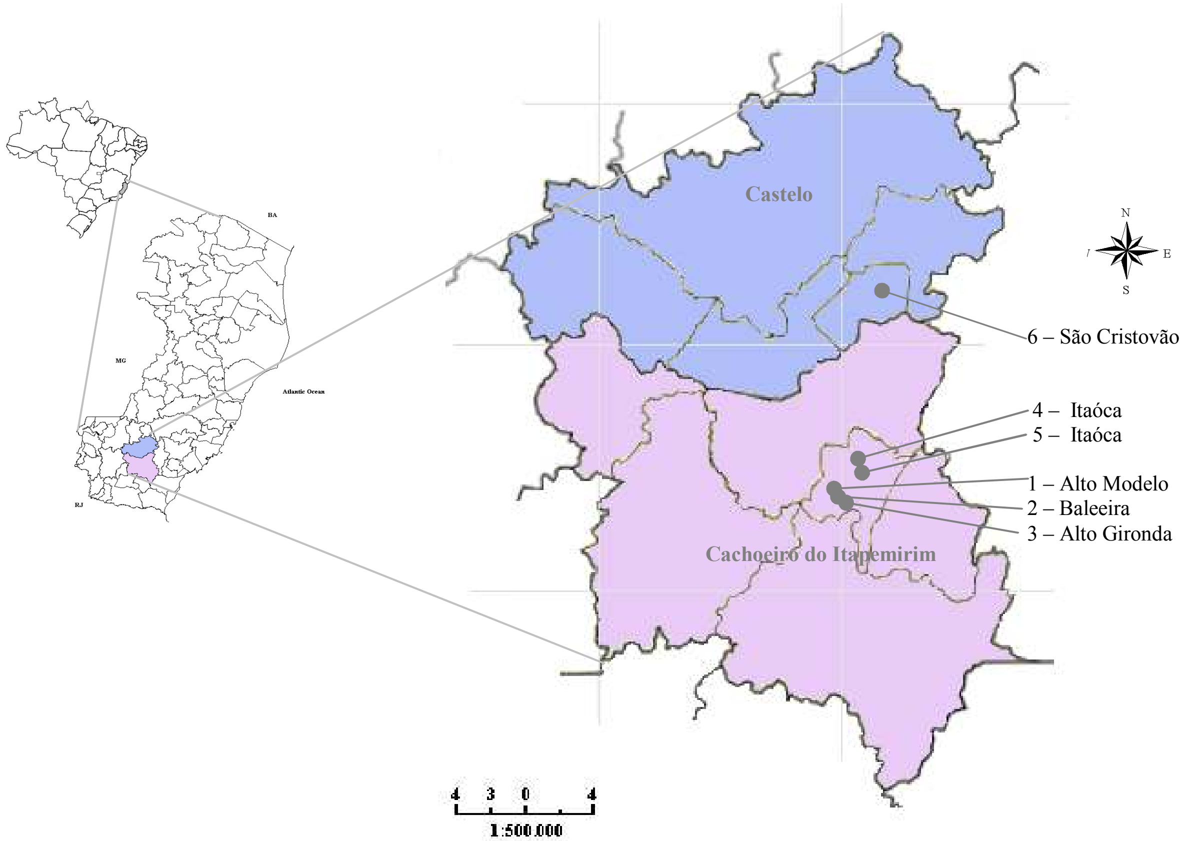

Figure 1. Map of the geographic localization of the marble sampling regions in central south region of Espírito Santo state, Brazil. The numbers (1-6) represent the ID location of the collected samples (see Table 1).

Each sample was sealed in a standard 100 mL HDPE flat-bottom cylindrical flask with 52.5 mm plan screw cap and bubble spigot polyethylene flask 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 [4]. For each location, the samples were prepared in triplicate.

2.2. Measurements

All samples were measured in triplicate 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 measured resolution for the ^{60}Co 1332.5 keV is 1.9 keV. The spectra were analyzed with the WinnerGamma software [5]. All nuclides activities are given with uncertainty statistics at $\pm 1\sigma$ confidence level. Detections limits are given at $\pm 2\sigma$ confidence level with the GTN5 formulae. The detector efficiency curve was determined with a multielement gamma standard solution, for the same geometry as the sample. The background radiation was determined by measuring a high pure water sample in the same geometry as the marble samples.

The ^{232}Th concentration was determined as the weighted mean from the average concentrations of ^{228}Ac (gamma transitions and intensities: 911.07 keV (27.8%) and 968.9 keV (16.7%)), ^{212}Pb(gamma transitions and intensities: 238.63 keV (43.5%) and 300.9 keV (3.25%)) and ^{212}Bi(gamma transition and intensity: 727.33 keV (6.6%)). ^{226}Ra concentration was determined as the weighted mean from the average concentrations of ^{214}Pb(gamma transitions and intensities: 295.21 keV (18.7%) and 351.92 keV(35.8%)) and ^{214}Bi(gamma transition and intensity: 609.32 keV (45%)). The concentration of ^{40}K is determined directly by its gamma transition of 1460.83 keV with 10.7% of gamma intensity[4].

The HPGe detector and the samples were placed inside a conventional lead shield, with 10 cm of thickness. In order to establish the counting time, a fast screening was performed for each sample. All samples were measured during 200000 s.

3. RESULTS AND DISCUSSION

3.1. Activity Concentration in marbles granites in Espírito Santo State.

The average concentrations values of ^{226}Ra, ^{232}Th and ^{40}K are shown in Tab.1.

Table 1. Comparative values ^{226}Ra, ^{232}Th and ^{40}K for the studied localities.

Municipality	Locality	Commercial name	ID	^{226}Ra $Bq.kg^{-1}$	^{232}Th $Bq.kg^{-1}$	^{40}K $Bq.kg^{-1}$
Cachoeiro do Itapemirim	Alto Moledo	Cintilante	1	2,0 ± 0,7	1,6 ± 1,2	5 ± 4
	Baleeira	Branco Extra	2	2,4 ± 0,7	3,2 ± 1,7	21 ± 7
	Alto Gironda	Branco Rajado	3	2,7 ± 0,7	< 2,9	16 ± 4
	Itaóca	Branco Azulado	4	1,1 ± 0,7	< 1,2	16 ± 7
		Branco Carrarinha	5	6,5 ± 0,8	1,5 ± 1,2	27 ± 5
Castelo	São Cristovão	São Cristovão	6	1,0 ± 0,2	< 2,9	17 ± 4

Measurements with 68% ($\pm 1\sigma$) confidence level, k=1

The results of our work are summarized in Fig. 3, for easier contemplation.

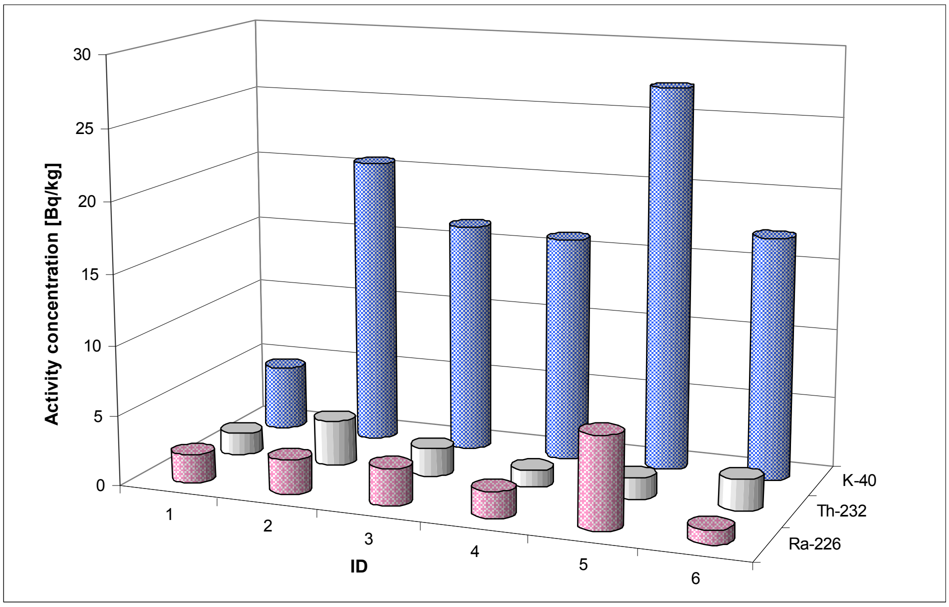

Figure 3. Activity comparative data for the studies localities.

For great extensions, the distribution of radionuclides in soil or rock is not a same, varying in intensity radiation and quantity of radionuclides presents and mean values are significant only for the analyzed area. However, for a simple comparison, the weighted mean values are a good indication. The weighted mean values of ^{226}Ra, ^{232}Th and ^{40}K, obtained from Tab. 1 for central south region of Espírito Santo State are compared with average concentrations from around the world and the 2008 UNSCEAR world wide average value [1] in Tab.2.

Table 2. Comparative average values for ^{226}Ra, ^{232}Th and ^{40}K.

Locality	^{226}Ra Bq.kg^{-1}	^{232}Th Bq.kg^{-1}	^{40}K Bq.kg^{-1}	Ref.
Mean values of Espírito Santo	3,0 ± 0,3	2,2 ± 0,6	18,0 ± 0,1	*
India, Gujarat	12 ± 3	3 ± 2	10 ± 3	[6]
Egypt	40 ± 8	20± 3	157 ± 7	[7]
Algeria	23 ± 2	18 ± 2	310 ± 3	[8]
Cameroon	8 ± 2	0,35	19 ± 2	[9]
Kuwait	3,9 ± 0,5	0,22 ± 0,08	3,7 ± 0,5	[10]
Jordan(Azraq)	20,1	11,4	85	[11]
Turkey	23	15	149	[12]
Nigeria	2	1	7	[13]
China	8 - 157	6 -166	44 - 1353	[14]
Italy(Traventino Marble)	79,0 ± 2,6	35,3 ± 0,8	15,1 ± 1,1	[15]
World Wide	77	84	1200	[1]

* Present Work

4. CONCLUSIONS

The concentration of ^{226}Ra, ^{232}Th and ^{40}K in granites samples from 6 quarries of Espírito Santo state, southeast of Brazil, were investigated by high resolution gamma-ray spectrometry.

The activities ranged from $1,0 \pm 0,2$ Bq.kg^{-1} to $6,5 \pm 0,8$ Bq.kg^{-1} for ^{226}Ra, from $1,5 \pm 1,2$ Bq.kg^{-1} to $3,2 \pm 1,7$ Bq.kg^{-1} for ^{232}Th and from 5 ± 4 Bq.kg^{-1} to 27 ± 5 Bq.kg^{-1} for ^{40}K. In all the samples the major contributions for the gamma irradiation become of the ^{40}K.

The measured values in the present wok show a very low activity concentration for ^{226}Ra, ^{232}Th and ^{40}K for all marble samples analyzed, in contrast with other results founded in literature. The lowest values of concentration for ^{226}Ra were observed in São Cristovão marble in São Cristovão region, the lowest activity concentration for the ^{232}Th was observed for Branco Azulado in "Itaóca" region and the lowest value for de ^{40}K activity concentration was observed for Cintilante marble in Alto Moledo region.

Regarding the natural radioactivity content, the values shows that the marble of Espírito Santo state is an excellent product for application in the civil construction.

The next step is to assess the radium equivalent activity (Ra_{eq}) and the external hazard index due to the natural radioactivity in those granites.

The high-resolution gamma-ray spectrometry is a powerfull tool for natural radioactivity studies and elemental concentrations determination in sand samples.

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