Abstract. The natural radioactivity in commercial granite samples of 6 quarries of Espírito Santo State, southeast Brazil, was determined from the $^{226}$Ra, $^{232}$Th and $^{40}$K contents. The assessed quarries were localized in regions of municipality Ecoporanga, Nova Venêcia, Colatina, Afonso Cláudio, Castelo and Mimoso do Sul. Three samples of each beach 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 $31 \pm 10$ Bq.kg$^{-1}$ to $219 \pm 29$ Bq.kg$^{-1}$ for $^{232}$Th, from $17 \pm 2$ Bq.kg$^{-1}$ to $270 \pm 20$ Bq.kg$^{-1}$ for $^{226}$Ra and from $498 \pm 21$ Bq.kg$^{-1}$ to $1481 \pm 60$ Bq.kg$^{-1}$ for $^{40}$K. The southern region of Espírito Santo State 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 region. Further, more samples will be collected from other quarries, allowing an evaluation of the health hazard indexes.

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].

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 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 Cláudio, Castelo and Mimoso do Sul municipality covering the south region of the Espírito Santo.

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, Espírito Santo State is responsible for more than 60% of the improvement, production and export of the brazilian granites and the geological characteristics favor the appearance of natural radioactivity [2].

So, it is very important to know the radioactivity content of these commercial granites, in order to evaluate the radiation hazard in these areas.

The objective of this work is to determine the concentration of natural radionuclides in commercial granites 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 it was considered the locations throughout the mountain chain of Espírito Santo state, having as choice criterion the activity of commercial granite extraction for the exportation and application in the civil construction. The samples had been yielded by the responsible companies for the extraction in these localities as are resold. The samples had been homogenised by spraying about 270 meshes in tungsten carbet ring mill. 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. 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 52.5mm 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 $^{238}\text{U}$ and $^{232}\text{Th}$ series. For each location, the samples were prepared in triplicate[3].
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 an 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer. The measured resolution for the $^{60}$Co 1332.5keV is 1.9keV. The spectra were analyzed with the WinnerGamma software[4]. All nuclides activities are given with uncertainty statistics at ± 1σ confidence level. Detections limits are given at ± 2σ 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 beach samples. The software output represents already the radionuclide concentration.

The $^{232}$Th concentration was determined as the weighted mean from the average concentrations of $^{228}$Ac (gamma transitions and intensities: 911.07keV (27.8%) and 968.9keV (16.7%)), $^{210}$Pb(gamma transitions and intensities: 238.63keV (43.5%) and 300.9keV (3.25%)) and $^{212}$Bi (gamma transition and intensity: 727.33keV (6.6%)),$^{226}$Ra concentration was determined as the weighted mean from the average concentrations of $^{214}$Pb(gamma transitions and intensities: 295.21keV (18.7%) and 351.92keV) and $^{40}$K the concentration of $^{40}$K is determined directly by its gamma transition of 1460.83keV [4]. All the activities had been corrected by a self-attenuation factor. That was necessary because the apparent densities (around 2 g.cm$^{-1}$) and the composition of the sample cause an modification in the self-absorption for the proper sample [5].

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 150000s.

3. RESULTS AND DISCUSSION

3.1. Activity Concentration in granites in Espírito Santo State.

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

Table - Comparatives values for $^{40}$K, $^{226}$Ra and $^{232}$Th for the studies localities.

<table>
<thead>
<tr>
<th>Commercial name</th>
<th>Municipality</th>
<th>ID</th>
<th>$^{40}$K Bq.kg$^{-1}$</th>
<th>$^{226}$Ra Bq.kg$^{-1}$</th>
<th>$^{232}$Th Bq.kg$^{-1}$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sta. Cecília</td>
<td>Ecoporanga</td>
<td>1</td>
<td>1185 ± 50</td>
<td>17 ± 2</td>
<td>73 ± 9</td>
<td>*</td>
</tr>
<tr>
<td>Branco Primata</td>
<td>Nova Venêcia</td>
<td>2</td>
<td>944 ± 39</td>
<td>18 ± 2</td>
<td>68 ± 9</td>
<td>*</td>
</tr>
<tr>
<td>Preto São Gabriel</td>
<td>Colatina</td>
<td>3</td>
<td>989 ± 41</td>
<td>19 ± 2</td>
<td>71 ± 9</td>
<td>*</td>
</tr>
<tr>
<td>Iberê Crema Bordeaux</td>
<td>Afonso Claudio</td>
<td>4</td>
<td>523 ± 22</td>
<td>273 ± 14</td>
<td>37 ± 7</td>
<td>*</td>
</tr>
<tr>
<td>Cinza Corumbá</td>
<td>Castelo</td>
<td>5</td>
<td>1405 ± 57</td>
<td>44 ± 3</td>
<td>224 ± 18</td>
<td>*</td>
</tr>
<tr>
<td>Cinza Andorinha</td>
<td>Mimoso do Sul</td>
<td>6</td>
<td>972 ± 41</td>
<td>41 ± 3</td>
<td>148 ± 13</td>
<td>*</td>
</tr>
<tr>
<td><strong>Average Values</strong></td>
<td></td>
<td></td>
<td><strong>1003 ± 42</strong></td>
<td><strong>69 ± 4</strong></td>
<td><strong>103 ± 11</strong></td>
<td>*</td>
</tr>
</tbody>
</table>

|                        | Espírito Santo      | 1003 ± 42 | 69 ± 4 | 103 ± 11 | * |
|                        | China               | 672       | 112    | 71.5     | [6] |
|                        | Egypt               | 852 ± 297 | 187 ± 90 | 118 ± 14 | [7] |
|                        | Italy               | 1600 ± 100 | 153 ± 13 | 360 ± 30 | [8] |
|                        | Worldwide           | 420       | 32     | 45       | [1] |

* Present Work
Measurements with 68% (± 1σ) confidence level, k=1
The results of our work are summarized in Fig. 2, for easier contemplation.

![Bar chart showing concentrations of K-40, Ra-226, and Th-232 in granite samples.]

4. CONCLUSIONS

The concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K in granites samples from 6 quarries of Espírito Santo state, southwest of Brazil, were investigated by high resolution gamma-ray spectrometry. The activities ranged from $31 \pm 10$ Bq.kg$^{-1}$ to $219 \pm 29$ Bq.kg$^{-1}$ for $^{232}$Th, from $17 \pm 2$ Bq.kg$^{-1}$ to $270 \pm 20$ Bq.kg$^{-1}$ for $^{226}$Ra and from $498 \pm 21$ Bq.kg$^{-1}$ to $1481 \pm 60$ Bq.kg$^{-1}$ for $^{40}$K. In all the samples the $^{40}$K showed to the biggest source of gamma emission.

The next step is the assess of the radium equivalent activity ($\text{Ra}_{eq}$) and the external hazard index due to the natural radioactivity in those granites.

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

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REFERENCES


