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Uranium distribution and radon exhalation from Brazilian dimension stones

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ABSTRACT

This paper provides evaluations of the radiometric behavior and exhalation patterns of radon gas in decorative and dimension stones explored in the Brazilian states of Minas Gerais and Espírito Santo, given the importance of determining radon gas concentrations in human-inhabited environments. A total of 10 silicate rock types were studied, featuring different petrographic/petrophysical characteristics given by seven magmatic rocks (three of which are granitic pegmatites) and three metamorphic rocks. The study, comprising radiometric data of *U* and monitoring of ^{222}Rn gas exhalation, shows a strong correlation between petrographic parameters and the physical properties of rocks. *U* levels ranged between 2.9 and 37 ppm, revealing a good coherence between the presence and the absence of radioactive element-bearing accessory minerals for each rock type. The rate of radon exhalation from the stones is related to the petrographic/petrophysical features of each material. By comparing the ^{222}Rn level generated by a rock to the amount effectively emanated by it, the rate of emanated gas proves to be insignificant; also, a rock that produces more Rn will not always emanate more. Simulations performed to estimate the radon levels inside residences or any given indoor environment showed that nine samples attained values below the 4 pCi/L EPA limit, whereas one was above that limit.

1. Introduction

The distribution of uranium in the Earth's crust is related to magmatic activities during the Earth formation processes. The mean abundance of uranium in the continental crust is 1.3 ppm, with higher concentrations in the upper continental crust (2.7 ppm) and inferior in the lower crust (0.2 ppm) (Ulbrich and Gomes, 1981). Uranium occurrence is more pronounced in granitic rocks, granitic pegmatites and syenites, appearing in a wide variety of minerals and found in most rock-forming minerals, secondary and accessory minerals, or forming their own minerals. In this case, it is concentrated in few less-abundant species, such as uraninite (UO_2).

The distribution of uranium, present in trace amounts in igneous rock-forming minerals such as quartz and feldspar, may be linked to processes of isomorphous replacement, concentration and adsorption in imperfect minerals, inclusion in the crystal lattice and in micro-crystals, as suggested by Pertlik et al. (1974). Biotite retains 19% to 22% of total uranium, and heavy minerals such as zircon, monazite, apatite, magnetite, ilmenite and riebeckite contain 61% to 65% of the uranium present in a given rock type (Moreira-Nordemann, 1977).

Radon gas (^{222}Rn), a product of uranium (^{238}U) decay series, represents the second leading cause of lung cancer in the United States after cigarette smoking, resulting in approximately 20 thousand deaths a year, according to studies by the US Environmental Protection Agency (EPA, 2003).

The exhalation rate of radon gas from a given stone is related to *U* levels, reflecting the origin of the rock as well as the influence of textural and structural features of the material. Whenever ^{226}Ra decays, a radon atom and alpha particle are formed and simultaneously ejected in opposite directions. This mechanism can result in expulsion of the radon atom from the crystal or molecular lattice in which the radon atom was formed. The petrophysical characteristics of rocks (microfissure condition, grain size, arrangement, alteration degree and contact surfaces between constituents) have direct influence on permeability, a parameter that can contribute to radon gas exhalation by facilitating the internal transport and flow to the outside environment of rocks.

In 2008, US news outlets reported that increased radon levels found inside homes could be linked to countertops made from dimension stones (ABIROCHAS, 2008; Murphy, 2008). The same article mentioned that similar claims had been made periodically over the last decade by manufacturers and distributors of materials competing with dimension stones (Murphy, 2008).

Because Brazil has taken an important role in the decorative and dimension stone business due to the vast lithological variety

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found in its territory, with the United States as its main client (ABIROCHAS, 2011), there is a clear need to study this theme, focusing not only on the commercial aspects, but also on human health.

This work features assessments of ^{222}Rn emanates from silicate-based dimension stones sourced from different geological environments in the states of Minas Gerais and Espírito Santo, southeastern Brazil.

2. Sampling and physical characterization

The analyzed samples include magmatic rocks of different lithologic types used for decorative purposes that are commercially mined in Brazil and distributed overseas (diorite, syenite, charnockite, monzogranite and three granitic pegmatites), as well as three metamorphic rocks. The locations of all rocks can be found in the geological maps in Figs. 1 and 2, while Table 1 shows the petrographic/petrophysical characteristics of the analyzed samples.

A total of 10 test specimens were built for each sample, in a roughly cubic shape, each side measuring about 5 cm and weighing between 300 and 400 g. The specimens were rinsed in running water and oven-dried for 24 h at 110 °C. The specimens were then removed from the oven, cooled at room temperature and weighed individually to obtain the dry weight (mass *C*). Next, the test specimens were immersed and saturated in a desiccator for 24 h, using a vacuum pump for the first three hours. After 24 h, the specimens were removed from the water, superficially dried and air-weighed, thereby obtaining their saturated weight (mass *B*). They were then weighed again on a hydrostatic scale, suspended in a recipient containing water, to obtain the submerged weight (mass *A*).

The values obtained in this trial result from the following equations: dry apparent specific mass (kg/m^3)$=C/(B-A)$; saturated apparent specific mass (kg/m^3)$=B/(B-A)$; apparent porosity (%)$=100\times(B-C)/(B-A)$; apparent water absorption (%)$=100\times(B-C)/C$. Lastly, the arithmetic mean of the physical indices was calculated according to NBR norm 12766 (ABNT, 1992) for each lithologic type. Table 2 contains the obtained results.

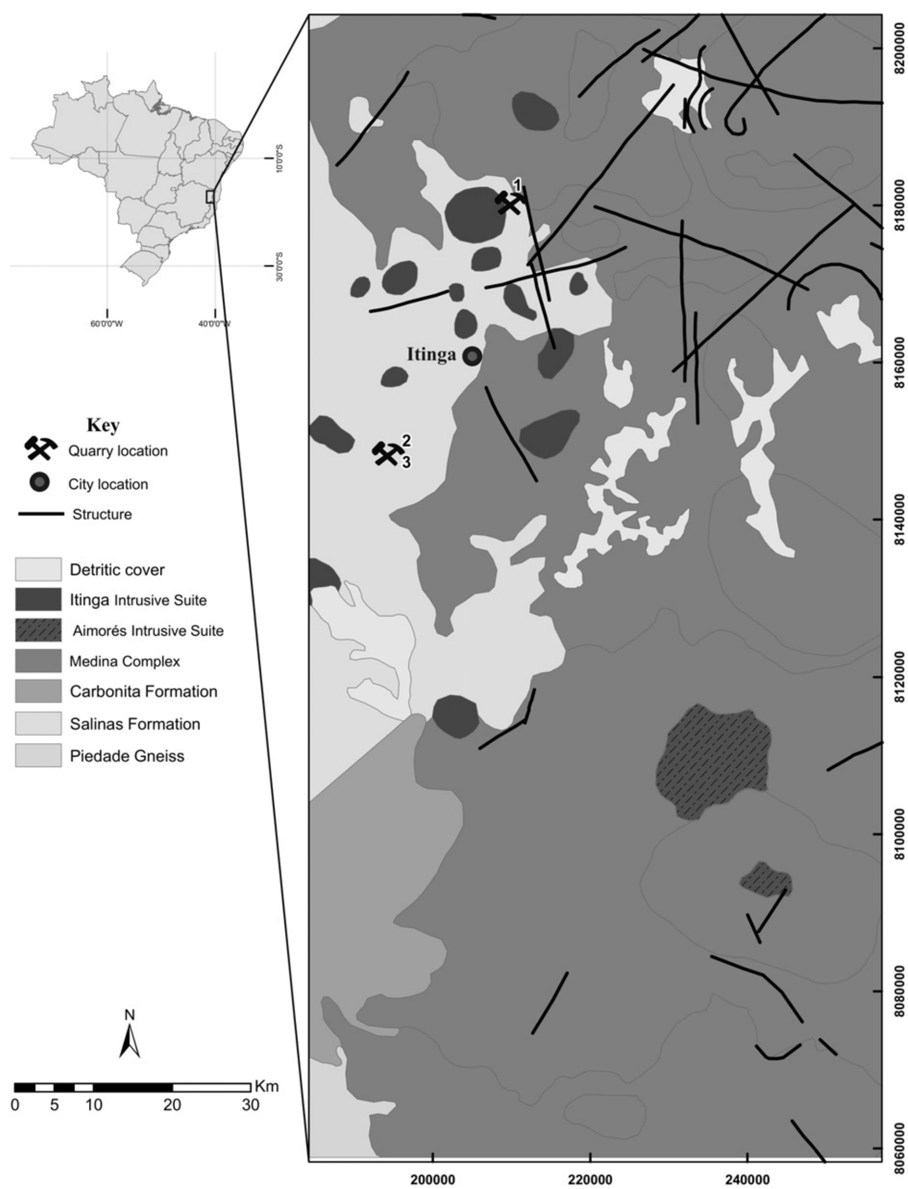

Fig. 1. Simplified geological map with the location of the rock specimens belonging to Minas Gerais State (CPRM, 2003). 1—Rock I (GC); 2—Rock F (MS); 3—Rock E (TI).