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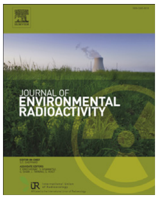

Estimation of the radon production rate in granite rocks and evaluation of the implications for geogenic radon potential maps: A case study in Central Portugal

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Abstract

The goal of this study was to estimate radon gas production rate in granitic rocks and identify the factors responsible for the observed variability. For this purpose, 180 samples were collected from pre-Hercynian and Hercynian rocks in north and central Portugal and analysed for a) ^{226}Ra activity, b) radon (^{222}Rn) per unit mass activity, and c) radon gas emanation coefficient. On a subset of representative samples from the same rock types were also measured d) apparent porosity and e) apparent density. For each of these variables, the values ranged as follows: a) 15 to 587 Bq kg^{-1}, b) 2 to 73 Bq kg^{-1}, c) 0.01 to 0.80, d) 0.3 to 11.4 % and e) 2530 to 2850 kg m^{-3}. Radon production rate varied between 40 to 1386 Bq m^{-3} h^{-1}. The variability observed was associated with geologically late processes of low and high temperature which led to the alteration of the granitic rock with mobilization of U and increase in radon ^{222}Rn gas emanation. It is suggested that, when developing geogenic radon potential maps, data on uranium concentration in soils/altered rock should be used, rather than data obtained from unaltered rock.

1. Introduction

Risk maps are tools that can be used to minimize the impact of environmental factors, including radon gas. For this last case, geogenic radon potential maps have some advantages over those based only on indoor radon concentrations; this is because the former are based on criteria that are less influenced by climatic and anthropogenic variables.

Gruber et al. (2013) discusses in detail the criteria that are the basis of the development of radiogenic radon potential maps in Europe. One of those criteria is the use of uranium concentrations or ^{226}Ra activity in rock/soil, assuming a correlation between these elements and the production/exhalation of radon gas.

However, this correlation has been called into question in some studies (*e.g.*, Amaral et al., 2012; Sakoda et al., 2011), in part due to the high mobility of uranium in the surface or in the near subsurface induced from the alteration processes. Particularly when incorporated in primary sources more susceptible to alteration (as in the case of uraninite), uranium can easily migrate from the structure of the host material and precipitate in cracks, on the surface of the minerals or even be mobilized by circulating groundwater (Pereira et al., 2010) under appropriated pH conditions. In this case, precipitation may occur a large distance away from the primary source in fault materials or in rocks with a matrix of clay, which can originate mineral deposits of economic interest (Pereira and Neves, 2012). These processes may be intrinsic to the geologic unit, related with the mineralogical composition, or have a purely local character, inducing intra-unit variability in the latter.

One way to assess this variability involves using the emanation coefficient which can be obtained from the relationship between ^{226}Ra activity and radon exhaled. This data has been published for several rock types and soils (Girault et al., 2011; Hassan et al., 2011; Przylibski, 2000; Sakoda et al., 2010, 2011; Sroor et al., 2013; Stoulos et al., 2003). The existence of a significant variability in this parameter has been acknowledged by several authors. For example, in a review study, Sakoda et al. (2011) indicated values for granites that varied between 0.04 and 0.40. Girault et al. (2011) showed that part of this variability was associated with the degree of alteration

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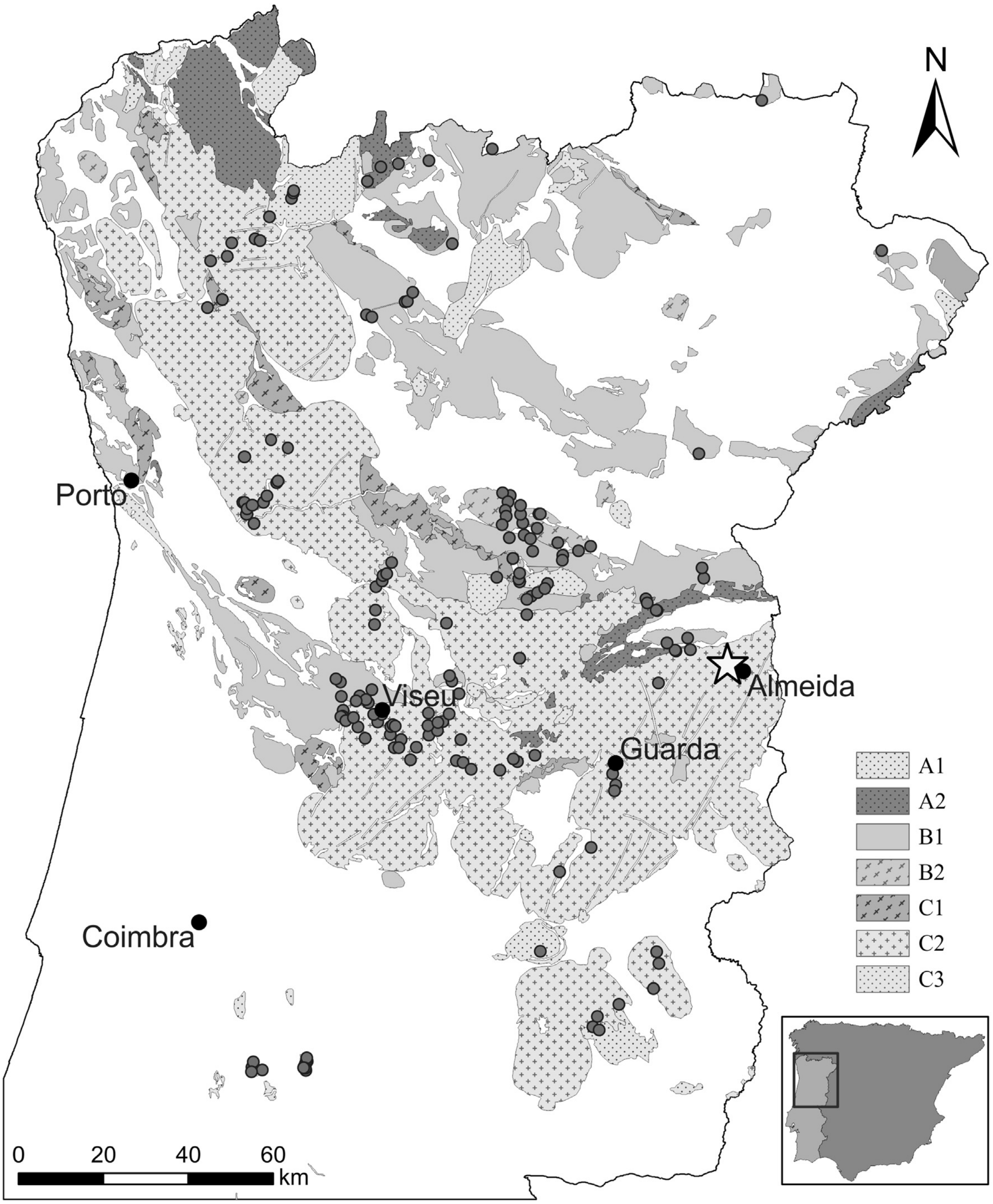

Fig. 1. Simplified geological map of the study area with the location of the samples set.

of the rock; this process enhanced the quantity of radon exhaled. Of course, porosity is an important control factor of the emanation factor, even though this parameter was not often measured (Banerjee et al., 2011).

Knowing the relationship between the ^{222}Rn and ^{226}Ra isotopes (expressed by the emanation coefficient) and the control factors is therefore an essential key in the assessment of the geogenic radon potential as well as in other problems involving the modeling of migration and transport of radon in geologic materials. However, it is also clear from the literature that there are few data available about the emanation coefficient, in particular about the factors, primary or secondary, that control its variability. .

The present work aimed to measure the activities of ^{226}Ra and ^{222}Rn and use these parameters to calculate the emanation coefficient in granitic rocks, which are the main lithology in the Northern and Central regions of Portugal. It also assessed the influence of control factors, particularly the effective porosity and density. Finally, this work assessed the quantity of radon exhaled from those geologic materials and for different degrees of alteration.

1.1. Geological setting

The granitic rocks that outcrop in large areas in Northern and Central Portugal are mostly of Hercynian age. They display high

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