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## Indoor radon risk associated to post-tectonic biotite granites from Vila Pouca de Aguiar pluton, northern Portugal



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#### ABSTRACT

At Vila Pouca de Aguiar area, northern Portugal, crops out a post-tectonic Variscan granite pluton, related with the Régua-Vila Real-Verín fault zone, comprising three types of biotite granites. Among these granites, PSG granite yield the highest average contents of U, probably due to its enrichment in accessory U-bearing minerals such as zircon. In the proximity of faults and joints, these granites are often affected by different degrees of hydrothermal alteration, forming reddish altered rocks, commonly known as "episyenites". These altered rocks are probably associated to the occurrence of hydrothermal processes, which led to uranium enrichment in the most advanced stages of episyenitization. In these granites, both average gamma absorbed dose rates in outdoor and indoor air are higher than those of the world average. Furthermore, even in the worst usage scenario, all these granites can be used as a building material, since their annual effective doses are similar to the limit defined by the European Commission. The geometric mean of radon activity of 91 dwellings located at the Vila Pouca de Aguiar pluton is 568 Bq m<sup>-3</sup>, exceeding that of other northern Portuguese granites. Measurements carried out during a winter season, indicate that 62.6% of the analysed dwellings yield higher indoor radon average values than the Portuguese legislation limit (400 Bq m<sup>-3</sup>), and annual effective doses due higher than the world's average value  $(1.2 \text{ mSy y}^{-1})$ .

The interaction of geogenic, architectural and anthropogenic features is crucial to explain the variance in the geometric mean of radon activity of dwellings from Vila Pouca de Aguiar pluton, but the role of geologic faults is probably the most important decisive factor to increase the indoor radon concentration in dwellings. Hence, the development of awareness campaigns in order to inform population about the incurred radiological risks to radon exposure are highly recommended for this specific area.

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#### 1. Introduction

Radon is a noble radioactive gas, resulting from uranium decay and characterised by a high mobility in natural systems. Exposure to this gas can be a serious public health problem, since it is responsible for approximately half of the radiation dose received by the human population (UNSCEAR, 2008). According to the Environmental Protection Agency radon gas is reported as the second leading risk factor of lung cancer after tobacco, which is in accordance to recent epidemiological studies carried out in the European Union, suggesting that radon inside dwellings may cause about 20,000 deaths per year (Dubois, 2005). The risk rises 16% for every 100 Bq m<sup>-3</sup> increase in residential concentrations (Darby et al., 2005). Lung cancer mortality in northern Portugal was assessed by Veloso et al. (2012), using data provided by the North Regional Health Administration and indoor

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http://dx.doi.org/10.1016/j.ecoenv.2016.07.009 0147-6513/© 2016 Elsevier Inc. All rights reserved. radon concentrations from a national survey conducted by the Portuguese Environmental Agency. A sub multiplicative interaction between smoking and indoor radon exposure was considered for the estimation of the number of lung cancer deaths attributed to indoor radon exposure, which ranges from 1565 to 2406, for the period between 1995 and 2004. This study indicated that among the 8514 lung cancer deaths observed, from 18 to 28% could be associated with indoor radon exposure.

The main source of indoor radon are the radionuclides in the underlying soils and rocks, which can diffuse into indoor air, but in some cases building materials can also contribute for radon exposure (Janik et al., 2015). Indoor radon could be controlled by a large number of geogenic, climatic and anthropogenic factors, which must be studied and monitored in order to access the health risk (Cosma et al., 2013, 2015). Among geogenic factors, the local structural geology assumes a primordial role in the prediction of indoor radon concentrations and radiometric anomalies associated with faults, as it is demonstrated by Pereira et al. (2010) in Central Portugal and Drolet and Martel (2016) in the Province of Quebec,

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Canada. However, many other studies put in evidence the relationship between Rn and uranium-rich granitic sources, such as those carried out by Bossew et al. (2008) in Austria, Ielsch et al. (2010) in the region of Bourgogne, Massif Central, France, Kemski et al. (2009) in the Fichtelgebirge and Erzgebirge areas, Germany, Appleton and Miles (2010) in England and Cho et al. (2015) in South Korea.

Several studies proved that the relation between indoor radon and climate is complex and, in many situations, inconclusive (e.g. Kobayashi, 2000; Miles, 2001; Groves-Kirkby et al., 2006). In fact, Groves-Kirkby et al. (2015) indicate that, in the town of Northampton, England, the monthly arithmetic mean radon concentration shows minimal or zero cross-correlation with temperature, atmospheric pressure and wind-speed, but there is a minimal negative correlation with precipitation and possibly also with relative humidity. The same study demonstrates that there are evidences of significant latency response in radon concentrations in respect to some climatic parameters, especially those related with the presence of atmospheric moisture. On the other hand, Perrier et al. (2009) and Perrier and Girault (2013) have demonstrated the sensitivity of soil-gas radon concentration to atmospheric pressure changes. The anthropogenic influences and the architectural characteristics of the structures (building age, floor level, foundation, building material, building type, room type) could also be important to explain the distribution of indoor radon concentrations (e.g. Kemski et al., 2009; Friedmann and Groeller, 2010; Girault and Perrier, 2012; Kropat et al., 2014).

In this context, it is extremely important to understand, in a first stage, the influence of the geology in the indoor radon variation, and, whenever necessary, to inform the population about the incurred radiological risks if no preventive measures were taken (Appleton et al., 2011). The Portuguese legislation regarding the air quality allows radon concentrations up to 400 Bg m<sup>-3</sup> inside of dwellings, and the measurement of this gas is only required in new constructions within granitic areas (Decree-Law nr. 79/2006). In Portugal, indoor radon exposure has been largely ignored, as in many other countries, by government or health authorities. Worldwide annual exposure to natural radiation sources ranges from 1 to 10 mSv  $y^{-1}$ , being its average estimated in 2.4 mSv  $y^{-1}$ . About 1.2 mSv  $y^{-1}$  is due to the exposure of radon inhalation (UNSCEAR, 2000).

The present study systematises radiometric and indoor radon results gathered at the Vila Pouca de Aguiar pluton (northern Portugal) to assess the indoor radon risk in dwellings located in an area with an important regional fault system intersecting different granitic rocks. These ones show different degrees of alteration (episyenitization) and weathering. This work also intended to evaluate the influence of house configuration and indoor human behaviour patterns in the radon gas exposure.

### 2. Geological setting

At Vila Pouca de Aguiar area, northern Portugal, crops out a post-tectonic Variscan granite pluton, with ca. 200 km<sup>2</sup> and an elongated form, constrained by NNE-SSW fractures related with the Régua-Vila Real-Verín fault zone (Fig. 1a and b). The posttectonic granite pluton comprises three types of biotite granites: Telões granite (TLG), Pedras Salgadas granite (PSG) and Souto granite (STG) that are variably porphyritic (Fig. 1b). Scarce rounded microgranular mafic enclaves of granodioritic and, more rarely, tonalitic composition, varying in size from 10 to 20 cm, are also observed especially in the TLG (Gomes, 2008). Souto granite is a coarse grained rock, outcropping in a small area in the south, southwest and east of the pluton. The three granites can be distinguished macroscopically by their grain size and biotite content, but there are no clear-cut contacts between the two dominant granites (TLG and PSG). The Vila Pouca de Aguiar granite pluton cross-cuts two-mica syntectonic granites, related to the third deformation phase (D<sub>3</sub>) of the Variscan event, as well as the Upper Ordovician to Lower Devonian metasedimentary sequence (Ribeiro, 1998; Martins et al., 2009), developing a metamorphic contact aureole. Based on geological, geochemical, isotopic, AMS and gravity data, Sant'Ovaia et al. (2000) and Martins et al. (2009) determined that the Vila Pouca de Aguiar granite pluton is a normally zoned composite laccolith, yielding older, more mafic marginal phases toward younger, more felsic central phases, that has been emplaced at the very end of the Variscan orogeny, fed by magma that upwelled from the fault zone. Its zoning is related to the successive emplacement of two main independent magma batches. The emplacement of this pluton was strongly constrained by the Régua-Vila Real-Verín fault zone, which crosscuts whole northern Portugal. This fault system began its development during the deformation phase D<sub>3</sub> of the Variscan orogeny, being reactivated on the deformation phase D<sub>4</sub> as a sinistral strike-slip fault with transtensional component. After that, with the rotation of the main stress direction to E-W, the Régua-Vila Real-Verín fault zone was reactivated as a thrust fault (Baptista, 1998). At present, this fault zone still has seismic activity (Fig. 1a and b) (Baptista, 1998).

At Vila Pouca de Aguiar pluton, granites are affected by various stages and degrees of weathering, increasing with the proximity of faults and joints. On the other hand, the occurrence of reddish altered rocks, commonly known as "episyenites", is constrained to TLG and PSG granites. These episyenites probably resulted from a hydrothermal alteration process and involved alkali metasomatism, magmatic quartz dissolution and transformation of the granite primary minerals (Genter and Traineau, 1992; Genter et al., 2002; Dezayes et al., 2005; Jaques et al., 2010). Two degrees of alteration can be distinguished: a) pervasive rock mass alteration, affecting large areas of the granite without visible modification of the rock texture and b) vein alteration, where the primary minerals (silicates) of the granite have been partly dissolved.

Due to the abundance of granites in the studied region, these rocks are frequently employed as building material by the locals, namely in the form of blocks in dwelling walls or as small slabs and tiles for lining purposes in external walls. Granites are also commonly used in indoor paving, kitchen counters, fireplaces and as coarse aggregates in the production of concrete.

#### 3. Material and methods

The petrographic study of polished thin sections of non-altered and episyenitized granites was carried out at the Department of Geology of University of Trás-os-Montes e Alto Douro, whereas backscattered electron imaging was performed at the National Laboratory of Energy and Geology (LNEG), S. Mamede de Infesta, Portugal.

A portable GF Instruments  $\gamma$ -ray spectrometer, model Gamma Surveyor compact 2, equipped with a NaI detector and a measuring range of 100 Kev to 3 MeV was employed in the field radiometric survey, covering different granite areas and the associated episyenitized domains, with a total of 84 analysis. Besides the gamma-ray flux, this technique also allows the estimation of K, eU and eTh concentrations. In each site the spectrometer was set close to the rock surface and then three measurements were made during a period of two minutes each. The final value resulted from the average of those three measurements.

Based on the field radiometric survey, 15 representative samples were selected for precise radiometric laboratory measurements, using a high efficiency multichannel gamma-ray spectrometer (ORTEC), equipped with a 3" NaI (Tl) crystal detector and Digibase system. The detection limit for K, U and Th are 0.1%,

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