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Geological Materials as Sources of Rn Emissions †

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Abstract: Geological materials are a potential source of pollutants, among which there is the radioactive isotope ^{222}Rn, which result of radioactive decay of daughter radionuclides of uranium (^{238}U). It is emitted as a gas that it can be released to the air to enter the human body, with the potential to affect internal organs (mostly the lungs) by alpha particles production. While the presence of uranium in the materials is a necessary condition for the production of Rn-222, the amount of gas emitted by the material depends on other characteristics that allow the migration of the gas. The main aim of this communication concerns a statistical analysis of results from diverse types of rocks.

Keywords: radiological hazards; exhalation rates; built environment

1. Introduction

The Geological materials can be a source of radiological pollution in the built environment due to the presence of radioisotopes, both by the emission of gamma radiation and by the production of radon (^{222}Rn from the uranium decay chain) that can contribute to internal radiation by inhalation and ingestion (Markkanen [1]; UNSCEAR [2]). The radon production directly depends on the uranium content of a rock, but besides, there are other factors that control radon release, namely porosity and moisture in the pore space. Thus, the radiological impact in terms of radon release will depend on the characteristics of the building materials that are used and the amounts of those materials and, in the case of radon, with also be affected by the environmental conditions (e.g. atmospheric moisture or whether the windows are open or closed).

2. Analysis of publications

The consulted publications presented results of mass exhalation rates or surface exhalation rates or both (few publications) from diverse types of rocks. Granites are the rock type most frequently considered (Allen et al. [3]; Chauhan [4]; Marocchi et al. [5]; Moura et al. [6]; Pereira et al. [7,8]; Bavarnegin et al. [9]; Pereira et al. [10]; Rafique & Rathore [11]; Guillén et al. [12]; Marques et al. [13]; Andrade et al. [14]), but there are also studies that include other igneous rocks (Chauhan [4]; Marocchi et al. [5]; Moura et al. [6]; Pereira Gómez et al. [7]; Rafique & Rathore [11]; Guillén et al. [12]), with a predominance of pyroclastic rocks (studied in Marocchi et al. [5]]; Andrade et al. [14]; Turhan et al. [15]; Kayakökü et al. [16]. There are some studies with results on metamorphic rocks such as marbles (Bavarnegin et al. [9]; Rafique & Rathore [11]; Andrade et al. [14]) and silicate metamorphic rocks (Chauhan [4]; Marocchi et al. [5]; Pereira et al. [10]; Andrade et al. [14]), and also some studies that include information in sedimentary rocks being (Bavarnegin et al. [9]; Pereira et al. [10]; Andrade et al. [14]). From these publications we collected (when available) minimum, average (mostly mean but also median) and maximum values. Given the general goal of this kind of publication it is expected that they are focused on materials that, at least potentially, can give high values of radon exhalation.

In Figure 1 we present of the results of mass exhalation rates (a,c) and surface exhalation rates (b,d) considering the original values (a,b) and their logarithms (c,d), obtained with the software Statistica 11 (Statsoft). As commonly happens in geochemical data, the distribution of original values is skewed due to the presence of higher values (since the considered parameters cannot have negative values). On the contrary, the logarithms of values show trends towards more balanced histograms, which are nonetheless somehow biased towards higher values (which would be expected given what was referred above).

The vast majority of the collected results for surface area exhalation rates are below the value of 58 Bq m^{-2} h^{-1} referred in UNSCEAR [2] for soil worldwide (but one should recall that the exhalation rate depends on factors such as porosity). The same material that in our review of gamma radiation (Sanjurjo-Sánchez & Alves [17]) was the clear highest value (with a concentration index which was 3 times higher than the second highest value) is also the one that presents the higher deviation from the set of other results: the surface exhalation rate for a local stone (Bavarnegin et al. [9]) ,which seems to be enriched in uranium due to circulation of solutions, achieving ^{226}Ra values that (using the conversion factors indicated in IAEA [18]) correspond to almost 7000 ppm given a Clarke of concentration (as defined by Ferman in 1933, according to Laznicka [19] and using the Clarke values indicated in this last author) around 4000. This uranium content will be around four times higher than the highest content referred in Laznicka [19] for the five biggest uranium deposits. The second highest value in terms of surface exhalation rate (which is around a ⅕ of the highest value) is for a granite studied by Allen et al. (2013). In the set of results of mass exhalation rate, the highest value corresponds to a tuff studied by Turhan et al. (2015), which is closely followed (around 10% lower) by a tuff studied by Marocchi et al. (2011).

The lowest value of surface exhalation rate also corresponds to granite studied by Bavarnegin et al. [9] while in terms of mass exhalation rates the lowest value was found for a marble studied by Andrade et al. [3] followed by a orthogneiss studied by Marocchi et al. [5].

The effect of heterogeneity is illustrated by the study of Allen et al. [3] which found that the highest surface exhalation rate value of a measurement area is more than fifteen times higher than the global value of the slab and around three times higher than the highest value reported (in Bavarnegin et al. [9]).

One can also highlight the results of Rafique & Rathore [11] which showed a significant value for a marble, being located in the central region of the histogram of log values and higher than many values for granites.

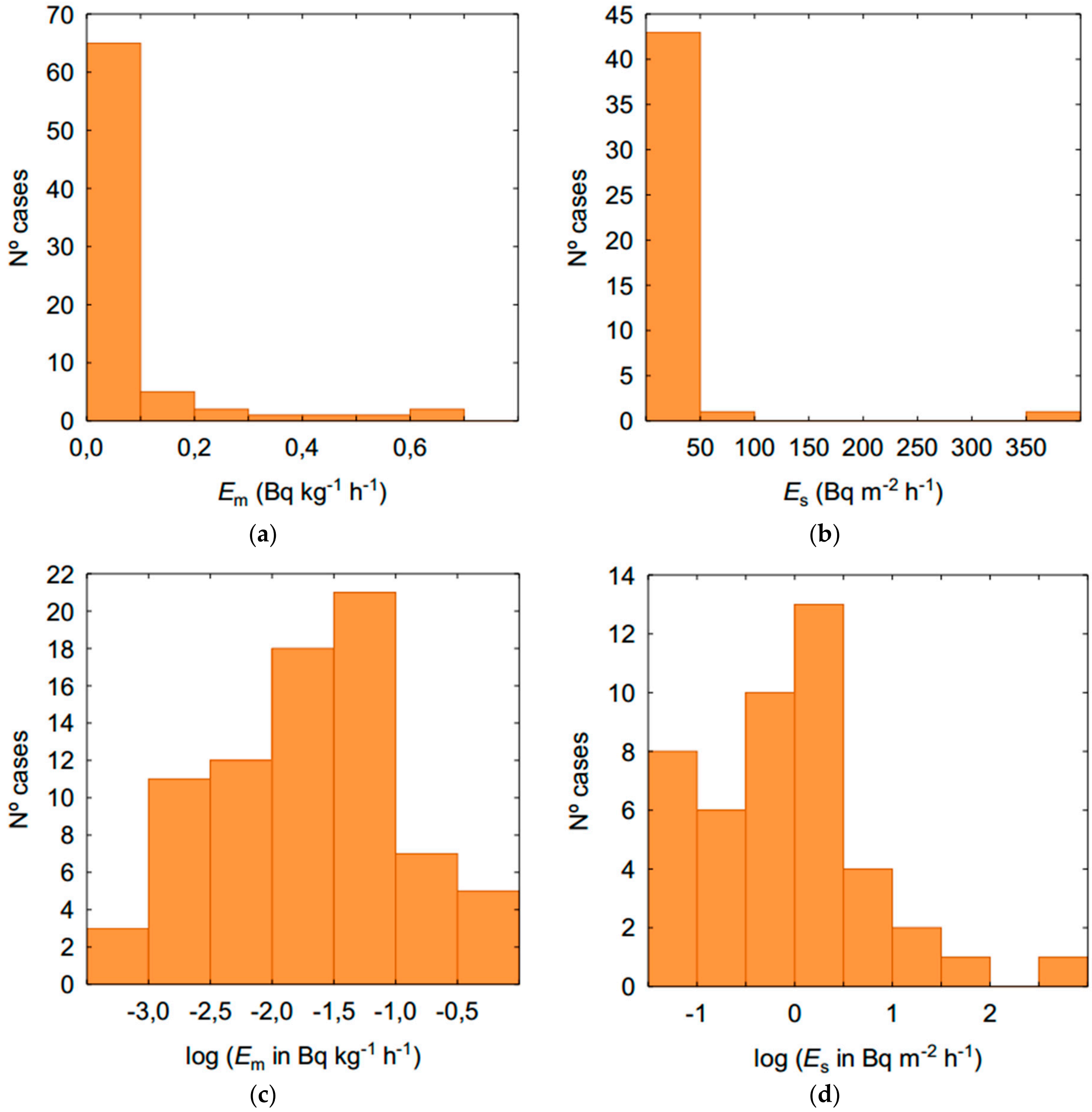

Figure 1. Histograms of values of mass exhalation rates (**a**) and their logarithms (**b**) and of surface exhalation rates (**c**) and their logarithms (**d**).

3. Final Considerations

As radon is the most important gaseous pollutant released by natural stones, it is of great interest to know the exhalation rates from these materials. Radon release depends on the uranium content of the rocks but also on their porosity, being affected by environmental factors as well. Like in many geochemical studies, the collected data on surface and mass exhalation rates of radon from diverse natural stone seem to be better described by a lognormal distribution, namely due to the presence of some cases with high uranium content. In general, the surface area exhalation rates of natural stones are below the soil worldwide average value.

The behavior of uranium and radon exhalation in these materials needs to be studied in more detail in order to assess the risks to human beings, as one of the more noticeable risks of radon is the accumulation indoor in areas where these rocks are used as building materials. Nonetheless, from the studied set one can suggest values of 0.7 Bq kg^{-1} h^{-1} and 80 Bq m^{-2} h^{-1} as conservative upper values for mass exhalation rates and surface exhalation rate, respectively. In the case of surface exhalation rates we found one study with a result around five times above the indicated upper value. However, this specific case can be considered an anomaly in terms of building materials, one which is not usually available internationally (but which, represents a worrying situation for its uses), corresponding to a local situation with a remarkable uranium enrichment that achieves contents at the level of uranium deposits.

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