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## Preface

# “Radon, health and natural hazards II”

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## 1 Introduction

The radioactive noble gas radon-222, characterised by a half-life of approximately 3.8 days, is produced by the alpha disintegration of radium-226 in the uranium-238 decay chain. Radon, released from rocks and soils to the atmosphere, is an important health hazard (Darby et al., 2004), and an important tracer of geophysical processes (Tanner, 1964), able to reveal meaningful information to reduce natural hazards, including volcanic eruptions and earthquakes (Laiolo et al., 2012; Neri et al., 2011; Plastino et al., 2011; Gillmore et al., 2010).

The session NH8.3 *Radon, Health and Natural Hazards* at the 2010 European Geosciences Union (EGU) General Assembly provided a vivid illustration that understanding radon in natural and man-made environments remains the subject of applied and fundamental research. This special issue is dedicated to results presented at this EGU session and marks the second year of the UNESCO IGCP Project 571 “Radon, Health and Natural Hazards”.

## 2 Overview of radon research presented at the EGU NH8.3 sessions

Current research on radon in the environment necessarily comprises overlapping groups of activities. The research presented at the EGU NH8.3 sessions can be usefully categorised into four such groups, namely: radon hazard, properties of radon sources, radon as a tracer of air and groundwater, and data monitoring and analysis. These groups and the individual contributions are discussed in the following. Unless stated otherwise, radon refers to radon-222 and radium to radium-226.

## 2.1 Assessment of radon hazard in homes and underground settings

The evaluation of radon risk in dwellings, for the general population, and underground settings, mainly for workers, remains a major activity. Mapping of radon in homes is in progress in various parts of Europe (Dubois et al., 2010), India (Ramachandan and Sathish, 2011) and Brazil (Anjos et al., 2011); and its feasibility is being evaluated in places where no consistent map is available, such as Poland (Kozak et al., 2011). In addition to organisational issues, radon mapping also raises methodological issues associated with the evaluation of exposure, dose and risk, such as seasonal correction factors.

In addition to the exposure of the general population, especially in relation to the geological context, significant exposure can take place in underground sites, for visitors and more significantly for underground workers. For example, radon concentration has been studied in some abandoned mines of Cumbria (UK), an area which up to now was considered a low radon risk (Gillmore et al., 2010). However, in one lead mine (Hudgill Burn), a concentration in excess of  $25\,000\text{ Bq m}^{-3}$  was observed and values larger than  $1000\text{ Bq m}^{-3}$  were observed in other former copper and lead mines. Proper time restrictions thus need to be implemented for volunteers and industrial archaeological workers and, when necessary, personal radon monitors need to be provided. This case study shows that there remains still a long way between knowledge of potential, theoretical radon risk and proper assessment and implementation in practice.

## 2.2 Fundamental properties of radon sources

The radon source concentration is defined by the product of radium concentration  $C_{Ra}$  and the fraction of radon atoms that can reach the pore space, a quantity known as the emanation coefficient  $E$  (Sakoda et al., 2011; Nazaroff, 1992; Tanner, 1964). The product  $EC_{Ra}$ , referred to as the effective radium concentration, expressed in  $Bq\ kg^{-1}$  (Stoulos et al., 2004), is therefore the relevant quantity defining the ability of a given medium to produce mobile radon. While research during the last decade focused on the theoretical understanding of the emanation coefficient  $E$ , some recent efforts have been dedicated to experimental measurements. For example, the roles of humidity and absorption were studied in an analogue of Martian regolith (Meslin et al., 2011), and the effect of temperature was studied in one rock sample using liquid scintillation (Lee et al., 2010).

Radon emanation experiments provide an interesting and subtle characterisation tool for building stones. While such measurements are performed, in the first instance, to screen out potentially hazardous construction material, some fundamental issues can also be raised. For example, a study of the bluish granites from Extremadura (Spain), which do not present significant radiological risk, revealed contrasting radon emanation behaviours, with exhalation rates varying by more than a factor of 10, from 0.01 to  $0.24\ Bq\ kg^{-1}\ h^{-1}$ , among facies having similar uranium content, of the order of 6 to  $7\ Bq\ kg^{-1}$  (Pereira et al., 2012). Such differences in radon emanation must be due to widely different values of the emanation coefficient, most likely resulting from varying radium mineralogical distributions. Thus, emanation experiments can give access to important properties of the structure of rocks, a fact that justifies the current renewal of interest (Sakoda et al., 2011).

While the emanation coefficient was known to increase with temperature, an apparent effect attributed mostly to adsorption (Meslin et al., 2011), only few data have been available so far, with only limited numbers of samples. In this issue, the effect of temperature is studied using 12 rock and 12 soil samples (Girault and Perrier, 2011). A systematic increase of  $E$  with temperature is observed, with an average value of about  $0.7\ \% \ ^\circ C^{-1}$ , but this temperature sensitivity shows large variations from sample to sample, covering a range from 0.1 to  $2\ \% \ ^\circ C^{-1}$ . Large values of temperature sensitivity can be of consequence in practice. This study illustrates that basic physical properties of radon in natural samples remain poorly known and that further theoretical and experimental studies need to be carried out. One topic which has not been addressed at all by the current research is the experimental study of radon diffusion in rocks. To take into account the variation of the radon diffusion coefficient with porosity or water content, one has to rely on empirical laws derived mostly from concrete samples (Nazaroff, 1992), which raises some concern when modelling radon transport in rocks in natural conditions.

Effective radium concentration, which is relatively easy to measure, beyond just being a parameter in the modelling of radon transport, can be a useful quantity to characterise soil and rock samples. For example,  $EC_{Ra}$  has been studied together with low-field magnetic susceptibility in sets of soil samples from Nepal (Girault et al., 2011). In some cases, large spatial variations of  $EC_{Ra}$  are observed while the magnetic susceptibility remains constant. In other cases, the opposite situation is observed. The contrasting patterns could be controlled by the selective adsorption of radium on iron oxides, which also contribute to the magnetic susceptibility, by contrast to adsorption on clay minerals or organic matter, occurring independently of the production of magnetic minerals.

## 2.3 Radon as a tracer of air currents and groundwater circulation

After the pioneering discovery of the seasonal variation of radon concentration in the Carlsbad Caverns, US (Wilkening and Watkins, 1976), there has been an increase in the continuous monitoring of radon concentrations, facilitated by easier availability of reliable and cost-effective radon monitors (Papastefanou, 2002). Indeed, studying the time-dependence of radon concentration is now a method of choice for the systematic characterisation of the ventilation regime of underground settings. For example, radon concentration was monitored from July 2005 to October 2009 in Postojna Cave (Slovenia), and was observed to range from  $200\ Bq\ m^{-3}$  in winter to  $3000\ Bq\ m^{-3}$  in summer (Gregorič et al., 2011). This seasonal variation indicates the presence of significant air currents in winter, but rather more stationary air in summer. In this case, a model could be developed, relating radon concentration to temperature, which could be used to forecast radon concentration and potential radon exposure to visitors. This study is also an example where radon measurements are used now for actual quantitative predictions on the conditions of a natural system.

The dynamics of radon concentration are also useful to study in the presence of both natural and mechanical ventilation. For example, short-term variations of radon concentration were studied in one natural cave, Niedzwiedzia Cave, and a former uranium mine, Fluorite Adit, located in Kletno in Poland (Fijałkowska-Lichwa and Przylibski, 2011). At both sites, convective air motions are observed over daily time scales, and sometimes problematic radon levels are reached despite the fact that mechanical ventilation is operated. This study shows that the presence of mechanical ventilation does not offer a guarantee of reduced exposure and that radon should be systematically monitored wherever and whenever a potential risk exists in underground spaces.

Radon measurements are not only useful to assess the presence of air motion, but also to characterise groundwater circulation. After some decades of observations, collected in various parts of the world, patterns and general features now

emerge. In this context, a comprehensive database of measurements of radon concentration in springs in the Sudetes (Poland) has been used to draw lessons of general interest (Przylibski, 2011). In this study, radon concentration in groundwater ranges from values lower than  $16 \text{ Bq l}^{-1}$  to exceptional values larger than  $1000 \text{ Bq l}^{-1}$ . These data confirm the generally observed fact that radium concentration in water, usually smaller than  $1 \text{ Bq l}^{-1}$ , cannot account for the radon concentration in water. In addition, the highest values of radon concentration correspond to poorly mineralised waters. Such shallow water sources are, unfortunately, used for private water supplies for human and animal consumption. This and other studies open new perspectives on the use of radon concentration to assess radiation risk and also the circulation pattern and vulnerability of shallow groundwater.

#### 2.4 Radon monitoring and analysis of time-series

Pluri-annual time-series of radon concentration in various conditions with a sampling time of one hour has led, over the last few years, to new interpretations and increasing concern regarding proper data analysis (Gillmore et al., 2010). Recently, an innovative monitoring scheme was tested at a quarry located in the uraniumiferous Beiras granite in Portugal, with 7 stations using radiation detectors installed at a depth of a few meters (Pereira et al., 2011). Preliminary results, with a sampling time of one minute, are available over periods of a few months. Significant differences are observed between nearby sites, separated by less than 100 m, both in the mean concentration, which ranges from 100 to  $3000 \text{ Bq m}^{-3}$ , and the daily variation, but classes of similarity also emerge for some sites. Temporal variations of radon concentration, thus, may reflect the heterogeneity of the local transport properties, which opens new possibilities for the characterisation of fracture zones. Such multiple networks of stations should therefore be deployed systematically at other sites, for example industrial mining sites or  $\text{CO}_2$  storage facilities.

Time-series of radon concentration have been used to study potential relationships with geodynamical effects such as earthquakes. In general, the correlation of any time-series with earthquakes is a delicate issue, to handle with utmost caution, and it must always be remembered that correlation does not imply causality. An example is shown with a revisited study of radon time-series at the time of UK earthquakes which occurred in 2002 (Crockett and Holt, 2011). In order to define anomalies in a statistically meaningful manner, a Standardised Radon Index (SRI) is defined from the radon time-series, analogous to the Standardised Precipitation Index (SPI) in meteorology. This quantity has the property of equalising different non-linear responses so that dissimilar anomalies can actually be revealed as being equiprobabilistic. Such research will help in defining anomalies in a more convincing manner and, ultimately, may reveal signals associated with earthquakes and other geophysical processes.

More generally, radon time-series offer an example of the interplay of signals of different origin with largely non-linear couplings, a situation often encountered in natural systems which makes the discovery and interpretation of unknown sources a difficult task. While more involved data analysis techniques are being developed, the lack of very long time-series of radon concentration appear as a clear limitation. Indeed, experiments carried out over the last decade in various parts of the world were punctual investigations and experimental conditions have been rarely maintained for sufficiently long periods of time (20 yr or more). In order to develop such work, radon observatories would now appear to be necessary, rather than one-off radon monitoring experiments.

### 3 Conclusion and perspectives

The various contributions to the radon sessions and this special journal issue reflect the diversity of researches carried out with radon in the environment. Whether applied or fundamental, from the instrumentation of industrial sites to the monitoring of active faults, all situations reveal common aspects. In addition, developments achieved in the area of radiological protection, for example, often have direct implications in theoretical developments, and vice-versa. While such joint developments characterise most researches in the geosciences, radon appears particularly fertile ground for a number of reasons. First, the unique alpha-particle detection of radon-222 (or radon-220) is totally unambiguous. This is not the case, for example, for magnetic signals, affected by the presence of underground cables, or electrical signals, which can be dominated by anthropogenic sources. Secondly, radon is a noble gas and its interaction processes are relatively simple to describe compared with compounds affected by various types of chemical reactions. Finally, radon-222 transport is essentially controlled by its half-life of 3.8 days, which is particularly well suited to study the processes relevant for systems at human scales, environmental problems such as contaminant plumes or the processes associated with volcanic eruptions or the nucleation phase of earthquakes.

Thus, while radon is important to study in its own right as a hazard, because of its radiological impact on the population, it also appears as one powerful tool to study our environment. As such, it is a tool which still has not been investigated sufficiently and where numerous important discoveries are within reach, provided sufficient efforts and dedication are pursued. These are goals of this project and other programmes putting radon as a major subject, and these papers further these goals in disseminating information, research results and good practice.

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