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Editorial

Radon applications in geosciences – Progress & perspectives

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Abstract. During the last decades, the radioactive noble gas radon has found a variety of geoscientific applications, ranging from its utilization as a potential earthquake precursor and proxy of tectonic stress over its specific role in volcanic environments to a wide range of applications as a tracer in marine and hydrological settings. This topical issue summarizes the current state of research as exemplified by some original research articles covering the aforementioned as well as other closely related aspects and points to some important future directions of radon application in geosciences. This editorial provides a more detailed overview of the contents of this volume, a brief summary of the rationale underlying the diverse applications, and outlines some important perspectives.

1 Introduction

Radon is a naturally occurring colorless, odorless and radioactive noble gas. It is the heaviest of the noble gases and has the highest melting point, boiling point, critical temperature, and critical pressure. Radon is soluble in water, its solubility decreases with increasing temperature [1]. Among the radioactive isotopes ^{222}Rn (radon, hereafter) has the longest half-life of 3.8 days, the other naturally occurring isotopes (^{218}Rn,^{219}Rn,^{220}Rn) having half-lives less than 1 hour.

Thoron (^{220}Rn) was discovered by E. Rutherford in 1899 from ionization experiments with thorium, and at the time was called "emanation". Rutherford was later surprised to find that its radioactivity decreased with time since radioactive decay had not been observed before due to the very long half-lives of the radionuclides known up to that time. In 1900 the German physicist F.E. Dorn discovered what would be radon (^{222}Rn) as a substance emitted by radium. A year later Rutherford and H. Brooks demonstrated that radon is a radioactive gas by measuring its diffusion

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into air and comparing the coefficient of diffusion with that of other gases and vapors. J.J. Thomson discovered in 1902 radon in water using a series of experiments on the electrical conductivity produced in gases when passing through water. Subsequently H.F.R. von Traubenberg demonstrated in 1904 that radon existed in the tap water of the Freiburg city (Germany), which was followed by the discovery of radon in groundwater.

^{222}Rn is generated within solid mineral grains by the radioactive decay of radium (^{226}Ra). Since radium is present in virtually every mineral material, radon is ubiquitous in the natural environment, constantly produced in every rock, soil or aquifer matrix. Radon atoms generated inside mineral grains can escape into the air or water-filled pore space, and further migrate by diffusion and/or advection to the subsurface air or water medium/phase. In the atmosphere radon is a potential health hazard if inhaled, as its short-lived decay products can be deposited on respiratory tract tissues and damage the cells, contributing to an increased risk of lung cancer.

Its widespread occurrence in nature and its unique characteristics make radon easily measurable by nuclear techniques even in very small amounts. Moreover, its half-life of 3.8 days is particularly suitable for tracking time-varying environmental phenomena. Therefore, radon has found a wide range of applications in various fields of geosciences, from seismology to environmental tracing in air, soil and water.

These applications are the focus of this issue, which covers the current progress, main challenges and future perspectives for the use of radon in geoscientific contexts. In the following, we provide a brief overview on its main topics and put them into the context of present-day cross-disciplinary radon research.

2 Indoor radon

While the atmospheric concentration of radon gas is usually low [2], about 10 Bq/m^3 [3], radon tends to build-up indoors, posing a potential health hazard. Lung cancer is the principal health concern associated with exposure to radon [4]. The short-lived decay products of radon can be deposited on respiratory tract tissues and subsequent alpha particle emissions from radioactive decay can disrupt the DNA structure within lung cells, contributing to an increased risk of lung cancer. Radon accounts for about half of the average indoor exposure to non-medical ionizing radiation [3] and is the second most important cause of lung cancer after smoking [6].

The hazardous health effects associated with radon exposure have been known since the 16th century, although radon was only discovered in the turn of the 20th century. Agricola noted as early as 1556 [7] a high frequency occurrence of fatal lung conditions among the miners of the Schneeberg mine in the Ore Mountains between Saxony and Bohemia [8]. It was not until the 1940s that a causal link between lung cancer in miners and radon exposure was established [9].

In non-mining contexts the accumulation of radon in domestic buildings was first observed in 1971, and ascribed to the use of uranium tailings as local landfill [10]. Since then the potential for hazardous exposure due to accumulation of radon in indoor environments became gradually evident, particularly in well-insulated ones [11]. While obvious sources of radiation exposure, such as related to nuclear facilities, are carefully monitored and controlled, radon monitoring in dwellings is scarce and scattered, and the risk of indoor radon exposure is often overlooked.

Radon easily enters indoor environments by diffusive and mainly advective migration from radon-rich subsoil [12]. Considering average soil gas radon concentrations, observed average indoor levels can be accounted for by a soil gas contribution of a few percent to the indoor air [13]. A smaller contribution to indoor radon accumulation can also originate from water (particularly from wells) and also from the building

materials themselves. Building materials produced from rock or soil always contain uranium and radium, usually in low concentration, but some materials such as alum shale concrete or phosphogypsum, can have high concentrations of radium, contributing significantly to indoor radon exposure. In this issue Morelli et al. [14] examine the radon exhalation rate of building and decorating materials commonly used in Sicily (Italy) and show that surface exhalation rates are higher for volcanic materials typically used in buildings in the Etnean area.

3 Tectonic applications

Since the 1970s radon has been repeatedly reported as a potential earthquake precursor (e.g. [15–23]). The main rationale is an expected enhancement in radon exhalation due to stress associated with the preparatory stages of an earthquake. However, the precursory nature of radon anomalies has not yet been convincingly demonstrated nor established, leaving this topic very controversial so far.

Field experiments in which artificial explosions have been generated in order to simulate small-scale seismic events ([24,25]) failed to produce radon anomalies, but the results were inconclusive due to the absence of a progressive stress field influencing the media as in the case of a "non-provoked" earthquake.

While at present the scientific community generally agrees that earthquake prediction is not possible, radon remains one of the strongest candidates as potential earthquake precursor. Woith [26] gives an impressive in-depth account of the recent history of the subject and provides a comprehensive critical review of the current status of radon as an earthquake precursor.

The association between radon concentration anomalies and major tectonic faults could result from mechanical cracks in the rocks or too slow crack growth determined by local strain of the media. However, contradictory results have been reported in the literature concerning the correlation between radon anomalies and tectonic structures (e.g. [27–31]). In this issue, Steinitz et al. [32] report on extensive radon monitoring performed along the western fault of the Dead Sea Transform, NW Dead Sea and show systematic differences at on-fault versus off-fault positions. This result demonstrates the geologic, possibly structural control on radon variation patterns which is a strong argument in favor of the application of radon as a proxy of geodynamic activity.

4 Volcanic applications

Radon is considered a useful tracer of volcanic activity due to its noble gas nature and its ability to be transported from depth (by carrier gases such as CO_2) without being chemically altered. However, volcanic environments are complex and associated co-existing phenomena such as flow systems along faults or in fumaroles are a further complicating factor.

Positive anomalies in radon emissions have been repeatedly associated with changes in volcanic activity (e.g. [33–36]) and related earthquake events (e.g. [37]). A positive correlation was found between the increase in radon and the Volcanic Explosivity Index of four American stratovolcanoes (El Chicon (1982) and Popocatepetl (1994) in Mexico, Poas (1987–1990) in Costa Rica and Cerro Negro (1982) in Nicaragua) [38]. The measurements in the field were performed with solid-state nuclear track detectors and electrets. The ratio between the magnitudes of the radon in soil peaks generated when the eruptive period started and the average radon values corresponding to quiescence periods indicate a dependence on the volcanic eruptive index for each of the eruptive periods. Possible mechanisms for the observed increase

of radon emissions associated with volcanic activity include the transport (up-flow) of subsurface radon due to increased heat flow or the discharge of dry steam.

In the current issue results from volcanic environments in the Macaronesian region are presented. Silva et al. [39] discuss the application of continuous radon monitoring at the Furnas volcano (São Miguel Island, Azores), illustrating the stumbling blocks encountered when trying to resolve radon patterns in complex geological settings. Martin-Luis et al. [40] report on radon and CO_2 monitoring at the Teide volcano (Tenerife, Canary Islands) emphasizing the distinct behavior of the two gases and its association with environmental conditions. Steinitz et al. [41] also discuss results from radon monitoring in Tenerife suggesting an extra-terrestrial (solar) component driving temporal variability.

5 Tracing applications

Radon's omnipresence in natural environments, its noble gas nature, and its half-life of 3.8 days, make it particularly suitable as a natural environmental tracer.

Since radioactive decay is its only significant sink, radon is an ideal tracer for atmospheric transport. Radon has been used in studies of atmospheric vertical advection, residence and transit times of atmospheric molecules, and to trace flows of air masses (e.g. [42–45]).

Applications of radon as a hydrological and marine environmental tracer include its use for investigating water exchange in reservoirs [46], groundwater migration (e.g. [47]), groundwater interaction in marine (e.g. [48]) or freshwater environments (e.g. [49]), submarine groundwater discharge (e.g. [50–52]) or groundwater contamination (e.g. [53]). In this issue Schubert [54] provides a review on the use of radon as an environmental tracer and for the assessment of subsurface non-aqueous phase liquids (NAPL) contamination.

The conventional approach for continuous measurement of radon in water is to bubble air through the water sample and to measure radon in the air circuit by counting the alpha particles emitted by radon and its progeny (e.g. [55]). The radon concentration in water is then calculated from the distribution of radon at equilibrium between the air and water phases. Schubert and Paschke [56] examine the gas specific water/air phase transfer kinetics of radon, CO_2 and CH_4 and show that the dissolved gases exhibit a similar temporal response to aqueous concentration changes. Petermann and Schubert [57] present a model for the correct estimation of short-term fluctuations in radon-in-water concentration from the corresponding radon-in-air measurements taking into account the response delay due to the water/air transfer kinetics of radon and the delayed decay equilibrium between radon and its decay products.

6 Challenges & perspectives

Radon proved to be a useful tool in a wide range of geoscientific applications. However, the complexity of factors influencing radon variability, together with the complexity of the phenomena themselves (e.g. earthquakes, volcanic fluids, surface-groundwater interaction) constrain its practical use.

A key aspect for the diverse radon applications is the identification of the multiple, non-linearly interacting factors affecting radon release from source and transfer in the liquid phase. Meteorological conditions are thought to be a major influence on radon migration, since rainfall, winds, and temperature gradients induce pressure differences and influence the water saturation of porous media. Atmospheric pressure is considered to influence the radon flux at the soil-air interface by drawing radon-rich

air from the host rock during pressure drops [58]. However, although radon sensitivity to pressure has been documented (e.g. [59–62]), a number of studies both in experimental and field settings concluded that atmospheric pressure does not influence radon variability (e.g. [63–67]).

An approach to advance our understanding of the physical mechanisms influencing radon variability is to conduct specific experiments in the laboratory in order to assess radon variability under controlled conditions (e.g. [63,68,69]). Furthermore, investigation of radon variability in underground observatories such as the Bloch observatory in Israel [70], Gran Sasso in Italy [71], or the low-background laboratory in Belgrade [72], allows to go closer to natural conditions while keeping some control on environmental factors, particularly internal temperature. However, even under such idealized conditions, radon time series display complex temporal patterns usually characterized by strongly non-stationary daily, intra-seasonal and seasonal variability. A clear understanding of the origin of such patterns is still lacking. Besides the conventional candidates, meteorological and environmental conditions, a possible influence of solar radiation originating in the deep solar interior was recently proposed [73].

Furthermore, the interpretation of radon measurements is hindered by the strongly non-linear character of radon time series, its marked non-stationary and heteroskedastic character [74] and strongly coupled oscillatory components varying on multiple time scales (e.g. [70,75]). Radon time series can also display long-range dependence, as demonstrated in this issue by Donner et al. [76]. However, unveiling and quantifying these features requires the application of sophisticated nonlinear time series analysis approaches (e.g. [77,78]) in order to decompose the multiple scales of variability and discriminate between the different drivers of radon variability.

Future progress in the use of radon in geoscientific applications will therefore rely on high-resolution and continuous long-term measurements of radon and meteorological parameters, as well as synergistic advances in experimental simulation, data analysis of radon time series and the numerical modeling of radon generation and migration.

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