

# Radon Flux Characteristics

Subjects: Environmental Sciences

Contributor: Igor Čeliković, Gordana Pantelić, Ivana Vukanac, Jelena Krneta Nikolić, Miloš Živanović, Giorgia Cinelli, Valeria Gruber, Sebastian Baumann, Giancarlo Ciotoli, Luis Santiago Quindos Poncela, Daniel Rábago

Radon is a noble radioactive gas, and almost half of the effective doses from all ionising radiation comes from exposure to radon and its short-lived decay products. Radon flux measurements provide information about how much radon rises from the ground toward the atmosphere, thus, they could serve as good predictors of indoor radon concentrations. Although there are many different mapping methods with many different input data, radon flux data are generally missing and are not included for the delineation of radon priority areas (RPA).

Keywords: radon flux ; radon priority area ; variations ; meteorological factors

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## 1. Introduction

Radon is a noble radioactive gas, and almost half of the effective doses from all ionising radiation comes from exposure to radon and its short-lived decay products <sup>[1]</sup>. Radon was first recognised as a health problem among miners in the United States of America <sup>[2]</sup> and Czechoslovakia <sup>[3]</sup>; thus, it was identified as a human carcinogenic <sup>[4]</sup>. Afterwards, radon in indoor environments started to be monitored more systematically through national radon programmes <sup>[5][6][7]</sup>. Finally, radon is identified as one of the major causes of lung cancer, accounting for between 3% to 14% of all lung cancers <sup>[8]</sup>; consequently, it was included in the Basic Safety Standards (BSS) for protection against ionizing radiation <sup>[9]</sup>. BSS requires that EU member states define radon reference levels, establish a radon action plan, delineate Radon Priority Areas (RPA, i.e., areas where the annual average radon concentration is expected to be higher than the reference level in a significant number of dwellings), and inform the public about their radon levels <sup>[9]</sup>.

Being chemically inert, radon has been used in many environmental studies as well. It is used as a tracer gas for the individuation of hidden active and dormant faults <sup>[10][11][12][13][14][15]</sup>, monitoring of volcanic activity and groundwater movement, and earthquake prediction <sup>[16][17][18][19][20]</sup>; to improve Atmospheric Transport Models (ATM); to investigate atmospheric mixing processes; and to estimate fluxes of greenhouse gases (GHG) using the Radon Tracer Method <sup>[21][22][23][24][25][26][27][28][29][30][31][32][33][34]</sup>.

To increase the accuracy of both radiation protection measurements and those used for GHG modelling, traceability to SI units is needed for radon release rates from soil, radon concentration in the atmosphere, and valid models for radon dispersal. Both radiation protection and climate research need to improve the accuracy of low-level radon concentration measurements and to ensure traceability to SI units. In this regard, the metrological project EMPIR 19ENV01 “Radon metrology for use in climate change observation and radiation protection at the environmental level” (traceRadon) <sup>[35]</sup> aims to develop traceable methods for the measurement of outdoor low-level radon activity concentrations in the range of 1 to 100 Bq m<sup>-3</sup> with uncertainties of 10% (k = 1), and to improve the accuracy of radon flux measurements. Both the climate and radiation protection communities will benefit from these data <sup>[35][36]</sup>. Improving accuracy and providing measurements capable of resolving diurnal variations will help climate change research by providing necessary inputs for the Radon Tracer Method, which is used to better estimate greenhouse gases emission <sup>[22][34]</sup>.

## 2. Radon Priority Area—RPA

Maps of environmental radioactivity are important on a local, regional, national and even worldwide scale as they assist in the planning and decision making of both authorities and citizens.

The identification of RPAs is a sensitive task as it raises some obligations for the region under consideration <sup>[9]</sup>. In such areas, radon measurements in workplaces are required, and awareness programs and a strategy for reducing radon exposure in dwellings should be established. In some countries, radon measurements in workplaces are required outside of the RPA also <sup>[37]</sup>. A definition of RPA was not given precisely in BSS; instead, it was left to member states to provide

their own definition of the RPA, which as a consequence led to different RPA maps. Therefore, many efforts have been made to discuss a concept of the RPA [38][39][40][41][42].

According to the questionnaire on indoor radon surveys issued in European countries, 11 out of 19 countries have reported  $300 \text{ Bqm}^{-3}$  as the Reference Level (RL), while some have chosen 100 or  $200 \text{ Bqm}^{-3}$  as a RL [43]. Most of the countries participating in the questionnaire have defined RPA on the basis of the percentage of dwellings having indoor radon concentration above RL, i.e., an area is defined as an RPA if the probability of the indoor radon concentration is above certain RL. This probability (threshold) ranges from 1% in the case of Malta and the United Kingdom up to 30% in the case of the Czech Republic [43]. Norway defined all of its territory as RPA. Two other definitions of RPA were also used: an area is defined as RPA (i) if the mean indoor radon concentration of the area is above RL, or (ii) if certain conditions are fulfilled (e.g., the gamma dose rate is above a certain level) [41].

As reported in the BSS, the identification of RPA needs a relatively simple statistical procedure that involves a few types of input data. The most common approach uses data that are expressed in the same method of measure, e.g., indoor radon concentration and the Reference Level. In that case, in order to get a reliable estimation of the RPA, it is important to carry out representative indoor radon surveys [6][7][44][45].

Other approaches involve the use of qualitative (non-measured) input data, such as the type of geology and/or lithostratigraphic data, and quantitative data with different unit measures (e.g., geological, geochemical, morphological, and meteorological data) that in general represent proxy variables related to the geogenic radon potentially delivered by the Earth's surface (e.g., Geogenic Radon Potential or Geogenic Radon Hazard Index), while indoor radon concentration is used for the determination of the RPA [46][47][48][49][50]. The GRHI could be conceptualized in terms of a measure of the "Rn proneness" of an area due to geogenic factors, i.e., a tool to decide whether an area is as RPA as the one used for the definition of the RPA. GRHI incorporates available geological data [49]. On the other hand, GRP is mainly defined through the "Czech method" [46], combining the radon in soil and permeability measurements, which depends on soil properties, hydrology and geology and, therefore, is limited by the availability of the required data [49].

The use of different available input data related to radon production and migration from the Earth's subsurface to the shallow environment, and the ability to enter buildings (e.g., measured quantities, geological data, meteorological data, and anthropogenic factors) led to the development of different mapping methods by using spatial regression techniques (Geographical Weighted Regression, Empirical Bayesian Regression Kriging, Machine Learning, Forest Regression) [48][51][52][53][54][55][56].

In Germany, Petermann et al. (2022) have shown that reducing high radon concentrations only in RPA will affect only a small proportion of residents as less than 1% of lung cancer fatalities are attributable to radon located in the RPA [40]. Therefore, when the aim is to reduce collective risk rather than individual, a map of lung cancer incidences would be more suitable [40]. A decision matrix on the choice of the appropriate radon map that would best serve its purpose is proposed in Hughes et al. in [57].

One of the tasks within the metrological research project 16ENV10 "Metrology for radon monitoring" (MetroRADON—<http://metroradon.eu>, accessed on 25 November 2020) was to evaluate different mapping methods for the delineation of the RPAs and to develop a strategy to harmonise possible inconsistencies of radon levels across the border [58]. In the framework of the MetroRADON project, an intercomparison exercise of different mapping methods was performed to construct geogenic radon potential maps, and it was concluded that different methods predict similar classifications of RPAs, while problems emerge only when the thresholds for the classification of RPAs are close to mean indoor radon concentrations [59].

Since there are different criteria for the definition and identification of RPAs, it is difficult to compare maps created by different national authorities. Bossew et al. demonstrated that the identification of RPAs across country borders is affected by the type of data aggregation (e.g., the geometry of the data support) and the scale of the used support [41]. This problem is defined as the Modifiable Areal Unit Problem (MAUP): the higher the support used to aggregate data, the more difficult the harmonisation of the results at the boundaries [41][60][61][62].

Although in the abovementioned mapping methods, many different input data were used, it could be noticed that data on radon flux were generally missing and were not included for delineation of the RPA. Radon flux measurements take a long time and, therefore, are not cheap, while soil gas measurement is simpler and faster, even when not a dynamic measurement. Radon flux gives an idea of how much radon comes out of the ground towards the atmosphere and, therefore, represents a good predictor of the radon that could enter buildings. Radon in the soil gas only gives information on the level of radon concentration in the ground, but this radon may never escape into the atmosphere. Therefore, Rn

flux measurements should be very useful to identify the RPA. Consequently, the aim here is to investigate to what extent radon flux was used, or could be used, for delineation of RPA.

### 3. Radon Flux Applications

Radon concentrations in soil gas, ground water, outdoor and indoor air, and radon flux are used to study many different phenomena and problems, from radiation protection to identification of hydrocarbon contamination in the soil. Although all these quantities describe parts of the same process—radon transport from  $^{226}\text{Ra}$  decaying to different surrounding media—different quantities are more suitable for various phenomena, and in some cases, several of these quantities can be used together.

In the reviewed literature, the amount of radon activity that exhales each second per square meter is called “radon flux” or “radon exhalation rate”. Herein, the term “radon flux” is used. Radon flux is most commonly expressed in  $\text{mBqm}^{-2}\text{s}^{-1}$  or  $\text{Bqm}^{-2}\text{h}^{-1}$ . However, when radon flux was expressed in  $\text{atoms cm}^{-2}\text{s}^{-1}$ , conversion to  $\text{mBqm}^{-2}\text{s}^{-1}$  was used ( $1 \text{ atom cm}^{-2}\text{s}^{-1} = 21.0 \text{ mBqm}^{-2}\text{s}^{-1}$ ).

Several examples regarding the use of radon flux are listed with a short explanation.

- Radon flux is used in the radon tracer method to monitor emissions of different gases, and it is most often used for greenhouse gases. In this method, radon flux is often considered constant over time and uniform, especially when performing large-scale studies [22][28][32][63][64][65][66][67]. More recent radon flux models revealed large spatial and temporal variability [68][69][70]. Grossi et al. (2014) improved the radon tracer method by using the model developed at the University of Huelva to calculate time-dependent values of radon flux [23]. In this model, it is considered that the only sink for radon is radioactive decay, and that dilution by atmospheric mixing is the same for radon and for other trace gases. Therefore, the surface emissions for any trace gas can be determined if the mixing ratios for these gases and radon are known, provided that radon flux is also known [64][65]. The TraceRadon project aims to build a metrological chain to ensure high-quality flux measurements that could provide increase data input for the radon flux method [36]. Some of the gases investigated in different studies include  $\text{CO}_2$  [32][63][71],  $\text{CH}_4$  and  $\text{N}_2\text{O}$  [27][64][72],  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ , and CFCs [28],  $\text{CH}_4$  [22][23][65],  $\text{CO}_2$  and  $\text{CH}_4$  [66],  $\text{N}_2\text{O}$  [67] and  $\text{H}_2$  [73].
- As there is large enrichment of  $^{222}\text{Rn}$  concentration in groundwaters compared to surface waters,  $^{222}\text{Rn}$  is used as a tracer to identify areas with groundwater discharge [74][75]. From continuous measurements of  $^{222}\text{Rn}$  in water, radon fluxes can be calculated for further used to estimate water fluxes that are otherwise difficult to measure [74][75].
- Radon flux is also used to plan, monitor, and evaluate the remediation of uranium mine and mill sites [76]. Ota et al., have investigated the effectiveness of clay-covered soil surfaces in radon flux suppression. From 80 days of continuous radon flux measurements of clay-covered soil surface and bare soil surface, it was found that suppression of the mean radon flux was 80% [77]. In the USA, regulations require that radon flux from active uranium mill tailing should not exceed  $740 \text{ mBqm}^{-2}\text{s}^{-1}$  [78]; however, although remediation is designed to fulfil requirements for radon flux, monitoring is not mandatory [79][80]. In Argentina, radon flux measurements are performed periodically in mining and processing sites [81]. Radon flux is also measured in India [82]. Radon flux was measured to evaluate the effectiveness of earthen cover after 20 years of service [80], as well as the effects of vegetation, clay cap, and environmental variables [83]. Due to high radium content, radon flux measurement is also of interest for phosphogypsum piles and their remediation [84].
- Very high values of radon flux can be found over active faults, so radon flux measurements can be used to identify faults [85][86]. Various researchers have also performed measurements along and perpendicular to the fault [87][88]. A study by Richon et al. found that superficial soil can mask faults, causing radon fluxes to be indistinguishable from background levels [89]. On the other hand, radon flux measurements, especially over faults, can be used to predict earthquakes [90]. Steinitz et al. found a statistically significant relation between radon flux and weak earthquakes during a multiyear study [91]. A study by Yakovleva and Karataev showed that using radon flux in combination with radon concentration in soil gas may improve the sensitivity of predictions, compared to using only one of these parameters [92].
- Radon concentrations in caves can be calculated based on radon flux [93].
- Radon flux can be used for radiation protection purposes. For example, Lucchetti et al. integrated gamma dose rate measurements with radon and thoron flux measurements to assess people's exposure to natural radiation and potential indoor radon [94]. However, radon exhalation was measured from soil samples, not in situ. Ramola et al. (2011) used radon flux to estimate indoor radon concentrations [95]. However, radon flux was also calculated based on the radon

concentration in soil gas and not measured. Stavitskaya et al. considered radon flux to carry out assessments of the radon risks of building plots [96]. Baeza et al. measured radon flux together with other parameters and indoor radon concentrations, and showed that radon flux alone is sometimes a good predictor of indoor radon concentrations, but in other cases, the correlation is much weaker [97]. Even negative correlations have been found in some cases [98]. Leshukov et al. investigated correlations between coal mine locations and radon flux, as well as indoor radon concentrations, to identify areas where radon preventative measures need to be taken during construction [99].

- Another important topic related to radiation protection purposes concerns building materials (commonly referred to as exhalation rate), which are the second most important source of indoor radon after soil. Building materials rich in  $^{226}\text{Ra}$ , such as granites or tuffs, represent a significant source of indoor radon [100][101]. The contribution to the indoor radon concentration from building materials is even more pronounced in energy-efficient buildings because an increase in the airtightness of homes increases the accumulation of radon indoors [102].

In the reviewed literature, the amount of radon activity that exhales each second per square meter is called “radon flux” or “radon exhalation rate”. Herein, the term “radon flux” is used. Radon flux is most commonly expressed in  $\text{mBqm}^{-2}\text{s}^{-1}$  or  $\text{Bqm}^{-2}\text{h}^{-1}$ . However, when radon flux was expressed in  $\text{atoms cm}^{-2}\text{s}^{-1}$ , conversion to  $\text{mBqm}^{-2}\text{s}^{-1}$  was used ( $1 \text{ atom cm}^{-2}\text{s}^{-1} = 21.0 \text{ mBqm}^{-2}\text{s}^{-1}$ ).

## 4. Factors Influencing Radon Flux

Radon is formed in rocks and soil from the alpha decay of  $^{226}\text{Ra}$ , contained in mineral grains, with the recoil energy of 86 keV. If placed close to the surface of the grain, radon atoms could leave the grain and reach interstitial space (pore), and if they have enough energy left, they could enter surrounding soil grains and become trapped. Although radon atoms can diffuse through the grains into the pores, the diffusion coefficient of radon in solids is very small; hence, a diffusion length is very small too ( $10^{-32}$ – $10^{-13}$  m). This process could be neglected compared to the recoil [103][104][105]. The amount of radon that escapes the rock/soil grain into the pore space with respect to the number of radon atoms that are produced in the grain is defined as the emanation factor  $\epsilon$ . There are numerous physical factors affecting radon emanation, such as the radium distribution in grains; the size, shape, and crystal structure of the grain; moisture content; grain (soil) temperature; atmospheric pressure; and radiation damage [106].

### 4.1. Soil Moisture

Soil moisture will reduce the speed of emanated atoms due to the much greater stopping power of water, which reduces the probability that emanating radon will embed in surrounding grain. This effect increases with the water content, up to the saturation level. The saturation is achieved faster for materials with smaller grain size [105][107][108][109]. However, it was also shown that with an increase of moisture content, the radon emanation factor decreases after reaching a peak. For different types of soil, the emanation coefficient reaches maximum at different water content: for general types of soil, emanation reaches maximum for water content of 5%; for gravel at 1–2%; and for clay at 10–15% [110].

### 4.2. Emanation Coefficient

Radium distribution in solid grains also influences radon emanation. In many primary minerals, radium is distributed uniformly in the grain. In this case, the emanation coefficient is inversely proportional to the grain size until it reaches a saturated value [111]. In the case of secondary minerals, radium is often distributed mostly on the grain surface, causing an almost constant emanation coefficient with grain size [112]. It has been observed that radionuclides with a larger radius tend to accumulate on the grain surface, unlike smaller particles, such as potassium, which tends to be in crystal lattice [113]. The emanation coefficient increases monotonously in the range of temperatures from  $-20^\circ\text{C}$  to  $45^\circ\text{C}$  [114].

In addition to “objective” physical factors, there are also experimental factors that could influence the results of emanation factors, such as the sample preparation, sample instrument, environment, and methods. Emanation factors reach values up to 0.25 for minerals, 0.40 for rocks, and 0.83 for soil, with an average of 0.03, 0.13, and 0.20, respectively [106][115].

### 4.3. Diffusion-Advection

Once the radon atom emanates from the grain, it can move from the soil pores toward the shallow environment and escape into the atmosphere. The dominant radon transport mechanisms are diffusion, advection and exhalation [103][116]. In general, diffusion can be described by the Ficks law: the diffusion coefficient in porous materials depends on the pore structure (tortuosity), pore fluids (water), adsorption properties of the solid matrix, temperature, etc. Radon diffusion will decrease with both a decrease in grain size and an increase in water content [105][117]. Mainly radon produced in shallow

soil layers contributes to surface radioactivity due to a short diffusion length that for most soil ranges between 0.5 m and 1.5 m [117][118].

Radon in the soil can also be transported to the surface by advection processes due to pressure differences. The influence of meteorological conditions occurs near to the surface, while at greater depths, advection is only due to pressure changes (i.e., faults and fractures) and the presence of gas carriers, such as CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub> [7][18][119][120][121][122][123].

Radon transfer from the parent material to the atmosphere is influenced by several physical factors, such as porosity, temperature, pressure gradients, and moisture [124]. A good correlation between radon flux and the porosity of the material was found [125][126].

Bedrocks, superficial deposits, and faults (fracturing) also affect radon flux. Faults increase radon flux, and the influence of superficial deposits depends on the physico-chemical characteristics of the deposits [87][127][128]. The condition of the soil surface also significantly influences radon flux. For example, if the soil surface is frozen or covered by water, the exhalation rate will be drastically reduced. Yamazawa et al. investigated the effect of snow and frozen soil on radon diffusion from the soil to the atmosphere. They found that frozen soil reduces diffusion by a factor of 2, while a 1.2 m thick layer of snow reduces radon flux by a few dozen [129]. Upper horizons formed by weathering crust can have similar effects [46].

When the radium rich layer is covered by a material with low radium content, exhalation is reduced. The thickness of the soil cover and other soil properties—such as compactness, water retention, and geochemistry—will affect radon flux as well [117][130]. It is very important to consider the perturbed ground, which can completely change all of the parameters related to radon exhalation, especially in urban areas [131]. More importantly, radon flux can differ drastically even among adjacent building plots, therefore causing high local spatial variations. Seismic activity [90][132][133], and also traffic and other urban vibrations [131], can cause large temporal variations in radon flux also.

#### 4.4. Moisture and Temperature

The influence of water content is opposite for emanation and exhalation. The combined effect on radon exhalation is such that exhalation increases with the water content until “saturation” is achieved. At this point, radon emanation does not increase with water content, but the diffusion through the water is much slower than diffusion through the air, causing the exhalation to decrease with increased water content. Therefore, radon exhalation shows maximum value at some level of water content, which depends on the soil properties. For many materials, maximum exhalation is achieved for water content of several percent to 10%, and a significant drop in exhalation is observed for water content above 25% [76][117][134][135][136][137].

The temperature also affects different processes occurring in the soil. Increasing soil temperature reduces adsorption and increases the emanation coefficient, thereby increasing the concentration of radon in soil gas [116]. The diffusion coefficient also increases with the temperature; consequently, overall, radon exhalation increases with the temperature. Ambient temperature weakly affects exhalation when it is lower than the soil temperature. However, if the ambient temperature is higher than the soil temperature, it will further increase radon exhalation [105][116]. The variability of radon flux from the same experiment in one period mirrored the changes in the air temperature, while in another period no regular changes of radon flux in tandem with air temperature were observed [138].

Radon exhalation is also affected by other metrological parameters. Light rainfall does not affect exhalation a lot, but heavy rainfall can saturate the soil, greatly reducing the exhalation process. Average yearly radon flux can be significantly different for rainy and dry years [79][105][116][139]. Measured radon exhalation from soil constantly decreased with the precipitation, starting from  $19.6 \pm 0.7 \text{ mBqm}^{-2}\text{s}^{-1}$  for dry soil, over  $12.7 \pm 0.3 \text{ mBqm}^{-2}\text{s}^{-1}$  after 10 mm of rain per day,  $10.3 \pm 0.2 \text{ mBqm}^{-2}\text{s}^{-1}$  (additional 14.6 mm of precipitation) to  $8.7 \pm 0.2 \text{ mBqm}^{-2}\text{s}^{-1}$  (for another 25.9 mm 3 days after) [140].

#### 4.5. Pressure

In general, radon flux is expected to decrease with increasing ambient pressure [116]. The correlation is difficult to demonstrate because usually many different parameters are changing at the same time [79]. Furthermore, the changes in pressure are cyclic; thus, long-term average fluxes are not greatly dependent on the pressure [117]. Additionally, it was found that an increase in wind speed increases radon flux [79][116] and the correlation between air humidity and radon flux is weak [141].

#### 4.6. Other

Relief and vegetation can also have influence on radon flux. Vegetation has direct influence because radon transports through plants. Plants can take up radon from the soil and release it into the atmosphere, effectively serving as radon pumps. Indirect influence is the influence on the soil water content, microclimate, and gas permeability. The influence of relief is indirect because soil development and water content depend on the topography [131].

A survey of literature shows that although most authors find similar trends in the influence of different parameters on radon flux, there is no consensus. For example, Yang et al. found a negative correlation between soil temperature and radon exhalation [141]. Kropat et al. found almost no correlation between secondary permeability, as well as faults and fractures, and radon exhalation. Furthermore, Zmazek et al. found a negative correlation between air pressure and radon flux at some measurement sites before earthquakes [133][142]. In recent research, no strong correlation between porosity and radon flux has been observed [137]. All of these studies show that radon flux is a very complex process; moreover, it is not possible to construct models that are universally applicable, even in rough approximation.

Yang et al. have performed 2 years of continuous measurements of radon flux using numerous environmental parameters: soil water content, soil and air temperature, air humidity, precipitation, wind speed, and directions [143]. A multiple regression analysis was performed by using air pressure, air temperature, soil temperature at 20 cm depth, and the soil water content. Obtained coefficients indicate that air pressure is not an important parameter, which is confirmed with similar long-term continuous measurements by Mazur and Kozak in [138]. Positive and negative effects of air and soil temperature on radon flux were obtained, respectively. The negative effect contradicts other experimental studies and should be further investigated [114]. As radon flux showed strong autocorrelations, including them in forecasting should increase performance significantly. This model can explain about 61% of the variation when autocorrelations are included, compared to only 28% when correlations are excluded [143].

#### 4.7. Diurnal and Seasonal Variations of Radon Flux

Daily outdoor radon variations show typical behaviour, namely, high concentration in the early morning due to an inverse atmosphere mixing. The increase of temperature during sunrise increases atmosphere mixing, causing an increase of radon in the upper atmosphere [26][144][145]. As the major source of outdoor radon is the radon flux from soil, it could be expected that daily variations of radon flux follow a similar pattern as outdoor radon. However, radon flux exhibits the opposite behaviour of outdoor radon concentration. The key factor leading to diurnal variation of radon flux is soil temperature, as variation of soil temperature causes variation of absolute humidity. Thus, radon flux is highest in the afternoon when the temperature is highest and, consequently, the humidity in the soil is lowest, while during the night, radon flux is lowest due to the lowest temperature of the soil (i.e., highest soil humidity). Daily variation of radon flux measured at the campus of Munich-Neuherberg ranged from 2.5 to 50.7 Bqm<sup>-2</sup> h<sup>-1</sup> with an average of 25.3 Bqm<sup>-2</sup>h<sup>-1</sup>. Regarding seasonal variations, the highest radon fluxes were in March and the lowest in October [141].

Seasonal variations of radon flux were investigated by many researchers [68][69][141][145][146][147][148][149][150]. In summary, all these papers agree and found high exhalation rates in seasons characterised by lower precipitation, while lower exhalation rates occurred in months with higher precipitation, higher moisture content, or for frozen soils and/or snow coverage. The summer-to-winter ratio of measured radon flux ranged from almost 1 up to 2.54.

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