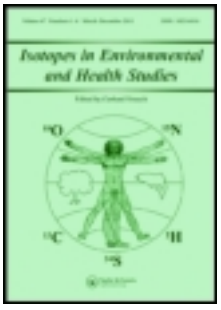

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Time variations of ^{222}Rn concentration and air exchange rates in a Hungarian cave†

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A long-term radon concentration monitoring was carried out in the Pál-völgy cave, Budapest, Hungary, for 1.5 years. Our major goal was to determine the time dependence of the radon concentration in the cave to characterise the air exchange and define the most important environmental parameters that influence the radon concentration inside the cave. The radon concentration in the cave air was measured continuously by an AlphaGuard radon monitor, and meteorological parameters outside the cave were collected simultaneously. The air's radon concentration in the cave varied between 104 and 7776 Bq m^{-3}, the annual average value was 1884 ± 85 Bq m^{-3}. The summer to winter radon concentration ratio was as high as 21.8. The outside air temperature showed the strongest correlation with the radon concentration in the cave, the correlation coefficient (R) was 0.76.

Keywords: cave; Hungary; natural radioactivity; radon-222; seasonal periodicity; time dependence

1. Introduction

For decades, it has been widely known that elevated radon activity concentrations can be measured in underground places such as karst caves [1–8]. The actual values depend on (1) the radon exhalation rate from the inner surfaces of the cave, (2) the volume and shape (morphology) of the cave, and (3) the inflow of outside air and its mixing ratio with the cave air [9]. The influence of meteorological conditions on the radon levels and their temporal variations depends mostly on the shape of the cave and the number and direction of cracks and fissures connecting the cave chambers to the outdoor air [10]. In karst areas, radon can easily migrate below the surface [11].

Radon-222 is an inert radioactive nuclide with a half-life of 3.8 days. Being the decay product of ^{226}Ra, it belongs to the ^{238}U series. As ^{226}Ra are present in soils, bedrock and building materials in various activities [12], radon is often observed from these materials in the environment. Being a noble gas, radon easily diffuses into the surrounding media. In soil gas, it can reach

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concentrations of up to hundreds of kBq m^{-3} [13,14]. Once emitted to the atmosphere, radon gas is normally diluted to concentrations below about 50 Bq m^{-3}. However, in poorly ventilated, enclosed spaces such as caves, radon appears by diffusion and advection through the capillaries and cracks of minerals and rocks [15], and with its short-lived daughter products, it can reach harmful concentrations [16].

When radon gas and its decay products, namely polonium, bismuth and lead isotopes, are inhaled, densely ionising alpha particles can interact with biological tissues in the lungs. Since even a single alpha particle can cause major genetic damage to a cell, a threshold radon concentration is unlikely to exist below which there is no risk of lung cancer [17]. Health effects of radon, most notably lung cancer, have been investigated for several decades. The first epidemiological studies focused on underground miners exposed to high concentrations of radon in their occupational environment [18]. They all emphasised the importance of radon studies and the need for knowledge of the behaviour of this gaseous radioactive isotope in closed and underground places.

Our major goal is to determine the time dependence of radon concentration in the Pál-völgy show cave in Budapest, Hungary, to understand the exchange pattern of the cave air with the outdoor air based on radon concentrations and to determine the factors that basically affect the radon concentration in the cave air. These results contribute to a better understanding of the nature of radon gas and provide new data on radon studies in caves.

2. Geological background of the studied cave

Hungary, located in the centre of the Pannonian Basin, where the geothermal flux is one of the highest in Europe, is known for its hot water springs and numerous caves. The Pál-völgy cave is situated in the Buda Hills in Budapest, which is the north-eastern part of the Transdanubian Central Range (Figure 1).

The explored length of the cave is around 19 km, of which 500 m is paved, lit and open to the public (Figure 2) [19]. The dominant wall rock of the cave is the Eocene Szépvölgy Limestone Formation. Eocene Buda Marl and Oligocene Tard Clay are deposited onto the limestone. A large multiphase hydrothermal cave system developed in the Szépvölgy Limestone Formation and

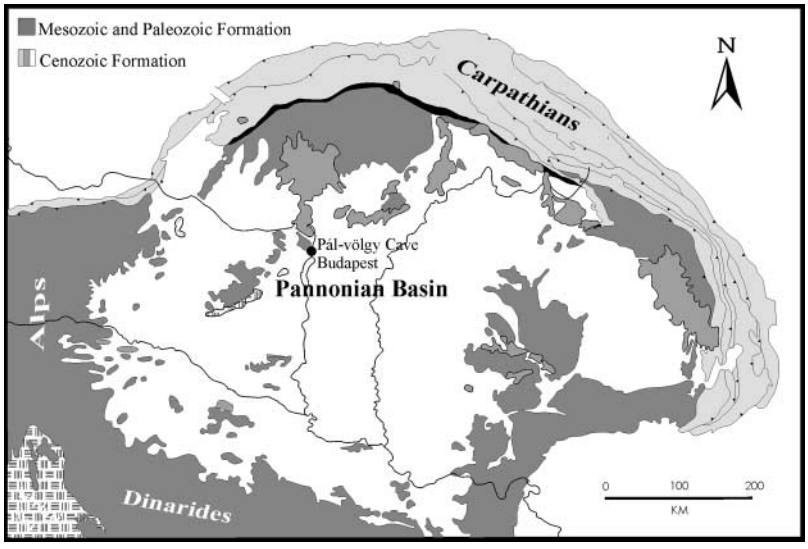

Figure 1. Simplified map of the Carpathian-Pannonian region showing the location of Budapest where the studied Pál-völgy cave is situated.

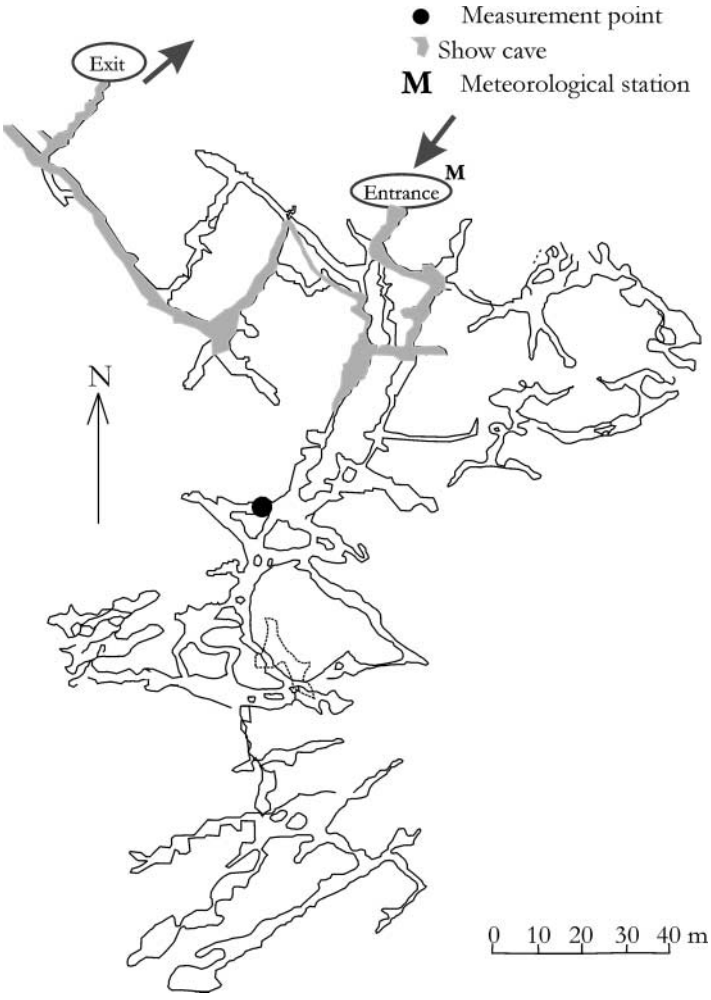

Figure 2. Map of the studied cave section. The grey area indicates the show cave, whereas the black dot marks the position of the AlphaGuard instrument during the measurement. M, meteorological station outside the cave.

partially in the Buda Marl resulting in a long-term complex paleokarstic evolution from the Late Eocene to the Quaternary [20]. The galleries of the maze system are decorated with characteristic dissolution forms and mineral precipitations as well as with dripstones at some places. The mineral assemblage of the cave contains 22 types of speleothems, some of them are rare or even unique to Hungary [19].

3. Methods

In order to define the time dependence of the radon concentration in the Pál-völgy cave, we measured the radon concentration constantly for 1.5 years (27 October 2009–22 February 2011) using an AlphaGuard radon monitor with an integration time of 1 h. This equipment is a portable ionisation chamber enabling the continuous monitoring of the radon concentration (Bq m^{-3}) and other meteorological parameters such as indoor air temperature (°C), indoor pressure (mbar) and indoor relative humidity (%). The same meteorological parameters were measured simultaneously

Table 1. The summer and winter periods used to calculate the summer and winter radon concentrations.

Seasonal period	Start date of season during measurement	End date of season during measurement
1st winter period	27 October 2009	18 March 2010
Summer period	8 June 2010	28 August 2010
2nd winter period	3 December 2010	22 February 2011

Notes: The division is based on visual examination of the time sequence plot of Rn measurements (Figure 3). Similar time series partitioning was carried out by Perrier *et al.* [21] (15 November to 15 March, 15 July to 15 September and 15 December to 15 March).

outside the cave by an FWS 20 meteorological station approximately 6 m away from the cave entrance.

The radon monitor was set at the same point during the whole measurement cycle: on the ground at a junction of five paths, and approximately 1 m away from the cave wall. In order to ensure undisturbed natural conditions, the sampling point was about 200 m from the show part of the cave (limited by access to electric power). The instruments were checked every month and the data were downloaded three times during the measurement period. The instrument was complemented with an AlphaPump used at an air flow rate of $1\,\mathrm{l\,min^{-1}}$.

The summer to winter concentration ratio and the air exchange rate [1,21] are very important for cave research. For example, the ventilation rate of the chamber can imply the presence of new cave sections. To estimate them, we applied a calculation based on the work of Perrier *et al.* [21]. A constant radon flux at the rock surface and negligible atmospheric radon concentration and summer ventilation rate were assumed:

$$\text{air exchange rate in winter} = \boldsymbol{\lambda} \cdot \left(\frac{A_{\text{summer}}}{A_{\text{winter}}} - 1 \right), \tag{1}$$

where λ ($\mathrm{s^{-1}}$) is the decay constant of radon, and A_{summer} and A_{winter} are the mean values of radon concentration in the summer and winter periods, respectively, as defined in Table 1. The air exchange rate gives the portion of the chamber's air volume that can renew in 1 s and has the unit ($\mathrm{s^{-1}}$).

4. Results and discussion

The data presented in Figure 3 show an obvious seasonal variation. In summertime, the radon concentration reached a maximum value of $7000\,\mathrm{Bq\,m^{-3}}$, whereas in wintertime, a significant decrease of concentration was observed (200–300 $\mathrm{Bq\,m^{-3}}$). As a result, the summer to winter concentration ratio is high (21.8, calculation based on Table 2). Usually, the summer to winter radon concentration ratio in caves varies from 1.1 to 10 [9,11,21,22]. The reason for the observed high summer to winter radon concentration ratio is probably the elevated ventilation rate in the winter period at the measurement point in the studied cave. In spring and in autumn, the radon levels fluctuated between the winter and summer values (Figure 3).

Using Equation (1), the air exchange rate in winter in the studied cave during the measurement cycle was calculated to be $4.38 \times 10^{-5}\,\mathrm{s^{-1}}$. This value is between one and two orders of magnitude higher than the air exchange rates in an underground limestone quarry near Paris, where the values varied between $0.05–0.24 \times 10^{-5}\,\mathrm{s^{-1}}$ at four different measurement points [21].

In a chamber with a well-defined shape, it is possible to calculate the volume of air that can be renewed each second, or how much time is needed to exchange the chamber air. However, in our case, it is very difficult to estimate the volume of the chamber without very high uncertainty.

The seasonal variation of the radon level in natural caves is characteristic of karst caves under moderate climatic conditions [1,15,23]. A 1-year-long continuous radon level monitoring inside

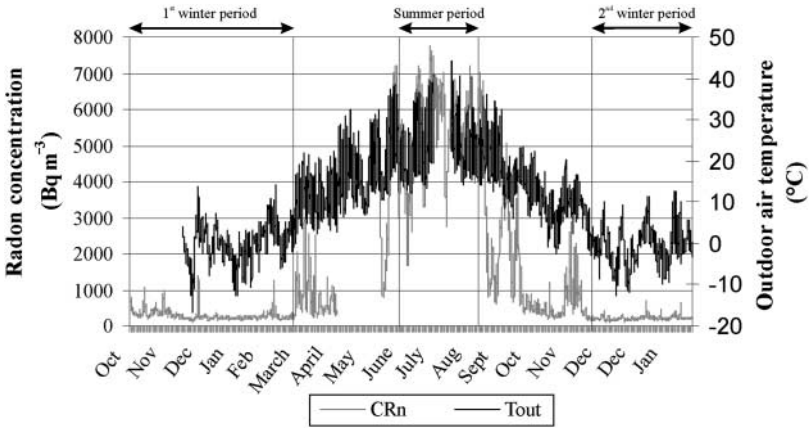

Figure 3. The radon concentration (CRn, grey line) and the outdoor air temperature (Tout, black line) during the long-term radon monitoring (27 October 2009–22 February 2011). Note that the values of radon concentration from 23 April to 30 May and the outdoor air temperature values from 15 to 30 June are not available.

Table 2. Statistical parameters of radon measurements.

	Radon concentration ($Bq\,m^{-3}$)				
Seasonal period	AM	GM	STD	min	max
1st winter	285	267	124	104	1408
summer	5504	5359	1139	1672	7776
2nd winter	218	211	59	94	720

Notes: AM, arithmetic mean; GM, geometric mean; STD, standard deviation; min, minimum value; and max, maximum value.

the Altamira cave in northern Spain showed radon concentration values ranging from 186 to 7120 $Bq\,m^{-3}$ with an annual average of 3562 $Bq\,m^{-3}$ [24]. The seasonal periodicity of radon concentration in the Altamira cave shows an opposite feature as the maximum concentration values were detected in autumn and in winter and the lowest in spring and in summer. The arithmetic average of the worldwide mean radon concentration in caves is 2.8 $kBq\,m^{-3}$ [25]. In the studied Pál-völgy Cave, the radon concentration varied between 104 and 7776 $Bq\,m^{-3}$, hence in the same order of magnitude as in the Altamira cave. However, the determined annual average during our measurement circle, 1.9 $kBq\,m^{-3}$, is lower than that in the Altamira cave and the worldwide average. This lower average characteristic of this cave is probably due to the high air exchange rate. Our result agrees within an order of magnitude with the value measured in 1993 in the same cave by the use of etched track detectors, 2.01 $kBq\,m^{-3}$ [25].

There are similar studies carried out in underground structures (soil pores, tunnels, caves and dwellings), where temporal radon variations influenced by the temperature were observed [26–28]. In our study, the radon concentration was also correlated with meteorological parameters such as temperature (°C), relative humidity (%) and air pressure (mbar). All of these quantities were measured hourly both inside and outside the cave. This allowed us to get enough data to find the possible correlations between different parameters and the actual radon concentration. The strongest linear relationship was found between the radon concentration and the air temperature. The correlation coefficients between radon concentration and indoor and outdoor air temperatures are 0.58 and 0.76, respectively (Table 3). A weak and negative correlation was observed for both indoor and outdoor pressures, as well as for outdoor humidity. Similar values were obtained by

Table 3. Pierson's linear correlation coefficient values between the radon concentration and the temperatures, humidity and air pressures inside and outside the cave.

Parameter	$Temp_{out}$	Hum_{out}	$Press_{out}$	$Temp_{in}$	Hum_{in}	$Press_{in}$
Correlation coefficient	0.76	−0.24	−0.06	0.58	0.2	−0.2

Duenas *et al.* [27] studying the seasonal variations of radon and some meteorological factors affecting variations in radon concentration in the Nerja cave in Spain, except that they found a positive correlation with the indoor humidity. In three different cave chambers, the correlation coefficient between radon concentration and indoor humidity varied from 0.68 to 0.84 in the Nerja cave. In comparison, our results show a strong relation. However, the humidity usually arises with the change of temperature. For example, in the Nerja cave, the indoor humidity varies between 63 and 92%, but in the studied cave, it is around 100% all the year. It does not follow the temperature changes, which are in strong relation to the radon concentration.

The temperature gradient of the outdoor and cave air temperatures appears to be an important factor in controlling ventilation. The direction of radon flow between the cave air and the outdoor air is a density-driven flow. As air density depends on the temperature, the air density differences closely follow the temperature differences between the cave and the outdoor air [29]. Based on our measurements, the average cave air temperature is about 12 °C. If the outdoor air temperature is lower than the cave air temperature, especially in autumn and winter, the cold, dense air flows from outside into the cave, and the radon concentration decreases in the cave. However, if the outdoor air temperature is higher than the cave air temperature, the denser cave air stays where it is, thus, minimising the ventilation. Therefore, new air comes through the cracks, fractures and cavities of the rocks into the cave and the radon concentration increases. Similar results were found worldwide in many caves in Spain, the United Kingdom, Slovenia and Hungary [15,22,27,30,31].

Schubert and Schulz [28] detected a significant diurnal variation in the soil gas radon concentration which is associated with the diurnal inversion of the soil/air temperature gradient. Taking into consideration their results, we looked for a diurnal variation of the radon concentration in the studied cave. For this aim, periodicities were studied by means of Fourier analysis and autocorrelation analysis. In order to decrease the significant noise present in the time series (Figure 4(a) and (b)), the data were smoothed with the robust median-based 5RSSH smoothing kernel. The studied time series is characterised by a strong cycle corresponding to the seasonal variation; differences were used to achieve stationarity in the mean prior to autocorrelation and periodogram calculations. Thus, the treated time series revealed the dominant diurnal 24-h periodicity in the measured radon concentrations, as shown by the autocorrelation and periodogram diagrams in Figure 4(a) and (b).

It is also expected that the actual radon concentration depends on the content of the radon parent isotopes in the rocks surrounding the cave and the emanation coefficient of rocks and clayish cave sediments. According to some studies [21], as well as our ongoing research [32], the major reservoir of trace ^{226}Ra and the main radon source are most probably the clay minerals. The studied cave was formed in limestone and in marl and the latter one can contain even 50% clay minerals modally. To validate this assumption, six clayish cave sediment samples from the cave were taken from the upper layer of the clayish cave sediment. Their specific activity of ^{226}Ra, ^{232}Th and the radon exhalation rates were determined on the samples in the laboratory [32]. The ^{226}Ra activity varies between 25 and 35 Bq kg^{-1}, the ^{232}Th between 21 and 30 Bq kg^{-1}. We observed no difference between the samples collected from the cave section located in the limestone and in the marl. However, the highest radon exhalation rate (12 s^{-1} kg^{-1}) measured was found in clay sediment deposited on marl. The radon exhalation rate of the other samples varied between 1.6 and 5.4 s^{-1} kg^{-1}. Considering that marl has a higher modal content of clay

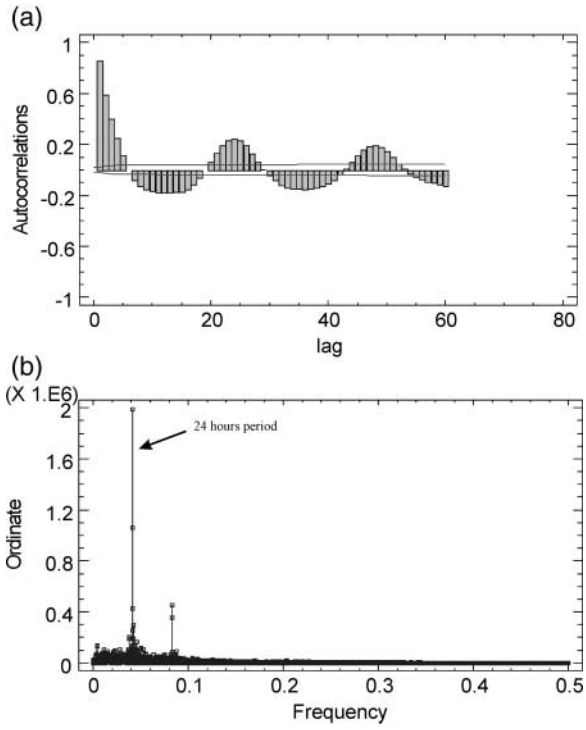

Figure 4. (a) The autocorrelation diagram for the five RSSH smoothed and differentiated radon data, and (b) the periodogram for the five RSSH smoothed and differentiated radon data. Note the strong 24-h periodicity.

minerals than the limestone, and the specific surface of the grains is larger in the marl, the higher clay content in the marl might explain the elevated radon emanation coefficient.

5. Conclusions

Based on approximately 1.5 years of continuous measurements of radon concentration in the Pál-völgy cave, the arithmetic mean of the annual radon concentration was 1.9 kBq m^{-3} and the radon concentration varied between 104 and 7776 Bq m^{-3}. In addition, the results indicate a clear seasonal variability of radon concentration in the cave air: in winter, the radon concentration fluctuates around a low mean value of 253 Bq m^{-3}, in summer, it oscillates around a high mean value of 5504 Bq m^{-3}, while in spring and autumn, the radon level varies between the winter and summer values. According to the results of radon monitoring and the calculated summer to winter radon concentration ratio and air exchange rate, there is a strong relation between the cave and the cave environment. Compared to other caves, the ventilation is very fast in the studied cave section, probably due to the location of the measurement point at the junction of five paths and in relative proximity to the cave entrance. A strong relationship was found between the radon concentration and the outdoor and cave air temperatures. In winter, cold, dense air flows into the cave, and the radon concentration decreases. However, in summer, the hotter and lighter air does not ventilate the colder cave air. Therefore, the radon concentration increases in these periods. Beside the yearly cycle, a significant diurnal variation of the radon concentration was detected, also caused by the temperature gradient changes of the outdoor and cave air temperatures. We assume that the major radon sources in the cave are the clay minerals of Buda Marl.

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