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RADON TRANSPORT PHENOMENA STUDIED IN KARST CAVES - INTERNATIONAL EXPERIENCES ON RADON LEVELS AND EXPOSURES

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ABSTRACT

The results of radon measurements in caves obtained by using of nuclear track detectors are summarized. Mean radon concentrations are ranging worldwide from 0.1 to 20 $kBqm^{-3}$ with 2.8 $kBqm^{-3}$ arithmetic average. From long-term extended radon measurements in caves not only a detailed dosimetric picture can be drawn, but using radon gas as a radioactive tracer, the subsurface and near-to-surface transport processes can be studied, too. It will be shown that long-term radon monitoring by nuclear track detectors, in conjunctions with active detectors which enables detection of fast dynamic changes, offers very important information for naturally-occurring transport processes.

KEYWORDS

Radon; cave; natural tracer; karst; etched track;

INTRODUCTION

Limestone contains in average about 1.3 - 2.5 ppm ^{238}U (16 - 31 $Bq \cdot kg^{-1}$), e.g. its daughter product ^{222}Rn can be found naturally in all caves. These minute quantities of parent substance result in relatively high values of radon in caves. Caves occur everywhere on the earth, although mainly in karst areas - as they are formed mostly in limestone environments. Karst strata have a characteristic feature: due to the infiltrating waters a special chemical dissolution process is taking place in the rock matrix. The phenomenon results in forming of an interconnected set of larger openings and fractures; a cave is a part of this system. Due to the formed special morphology the cave is able to communicate through the overburden under the influences of changing atmospheric pressure or temperature. These morphology, meteorology (and surface topology) linked flows play the most significant role in governing the underground radon transport in these areas.

In the last two decades the etched track techniques have found their place also in radon measurement projects aimed to study the cave environment. These measurements have ranged from sporadic observations up to regular long-term studies, and they were motivated by dosimetric and transport study approaches as well. In Hungary the first *in situ* field radon measurements in caves performed by

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etched track techniques started as early as 1977 and were initiated by Dr G. Somogyi, one of the pioneers of the etched track field. Since then, the number of studied caves has increased substantially and we have reached 10000 observation data in 31 Hungarian caves. After recognizing the link between the secondary porosity, fracturization and formed radon levels the ever-pressing interest to understand underground radon transport in karst environment has been increased. During the last several years active electronic devices, automatic multiparameter radon monitors have been included into cave studies. Including a short literature review the following results are excerpts based on this data set.

MATERIALS AND METHODS

For the purpose of cave radon measurement, different types of opened and closed diffusion chambers, equipped with alpha sensitive polymer track detectors, were used. Somogyi *et al.* (1983) described different types of multi-detector devices suitable for measurement in caves. The most significant improvement in the measuring technique was the thermally stabilized double-wall diffusion chamber (filled with water), which avoided the frequent problem of wetting of detector surfaces. Nowadays several commercially available radon monitors developed for indoor radon measurements are used in cave environment, which, owing to the high relative humidity, may be not always optimal. The typical exposure time ranges from one week to several months.

RESULTS AND DISCUSSION

Review of Published Data

Continuous radon measurements covering at least one year long period were performed by the etched track technique in Hungary, Italy, Slovakia and Luxembourg. Seasonally one week long observations were performed in England, and measurements not covering a full year are reported from Mexico and USA.

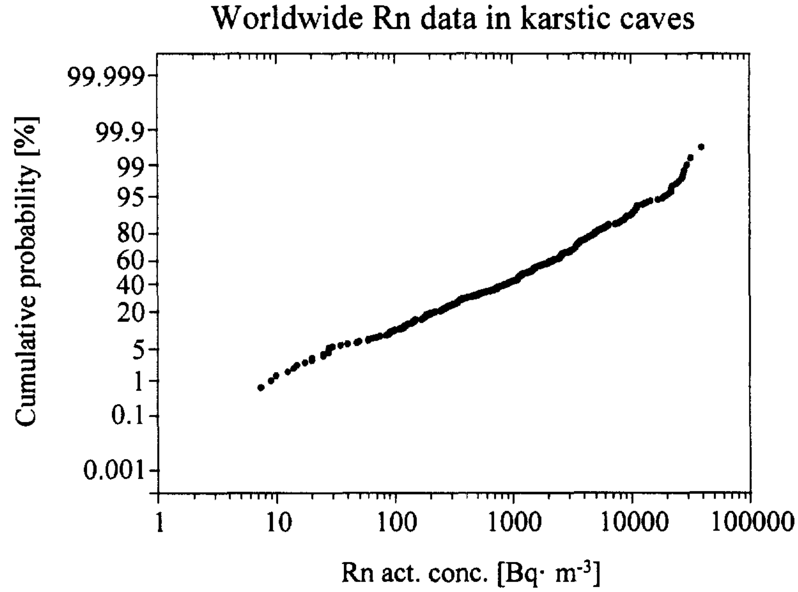

Figure 1. The distribution of radon activity concentrations reported from different caves worldwide. In the graph we compiled radon data available from 220 different caves.

Taking into account these results the annual average radon concentrations in karst caves range from 0.1 to 20 kBqm^{-3} with a 2.8 kBqm^{-3} arithmetic average. The lower end of the scale is associated with big chamber volumes or high ventilation rates; the upper end is characterized by closed, badly ventilated places and uranium-rich sediments. We note however, that based on long-term continuous radon observation the relative variations of radon between monitor locations indicate that the cave atmosphere is not a uniform radon environment over any given time period; so it is very hard to represent one cave with one radon value. The distribution represented on Fig. 1. provides compiled results of cave radon literature. The distribution of the data is close to lognormal.

Radon as a Natural Radioactive Tracer of Subsurface Transport Processes

The application of radon as a natural tracer is not yet common and widespread. Among the natural tracers it would be considered on the one hand as ideal since it is easily detectable even in small quantities, which do not modify the characteristic of the environment. On the other hand, unfortunately its sources appear everywhere and are spread over in a manner unknown *a priori*. The realization of radon transport processes in addition sharply depends on the configuration of the interconnected underground cavities. In the case of blind end systems, atmospheric pressure changes are the main control parameter, which are superimposed by convective air exchange due to temperature differences in cave systems with vertical extension. In the case of relatively large entrances, the convective air exchange due to temperature differences can mostly account for the radon transport process taking place. In the case of two or more entrance systems, where the other ‘entrances’ can be complexes of smaller fissures and fractures, chimney effect winds may dominantly govern the radon transport, or in some cases atmospheric winds may do so. It is obvious, that the interpretation and modeling of the concentration data is not straightforward: it needs interdisciplinary expertise of hydrogeologists, geologists, physicist, radiogeochemists, etc.

In the speleleology these types of applications give important contributions to the better understanding of the natural regimes of caves. Cunningham and LaRock (1991) delineated six microclimatic zones in Lechuguilla cave, Carlsbad caverns, National Park, New Mexico using radon grab sampling in conjunction with observed airflow data. Atkinson *et al.* (1983) from a single set of etched track measurements in the Castleguard cave, Columbia icefields, Alberta, Canada, identified the effect of tributary air currents from larger fissures.

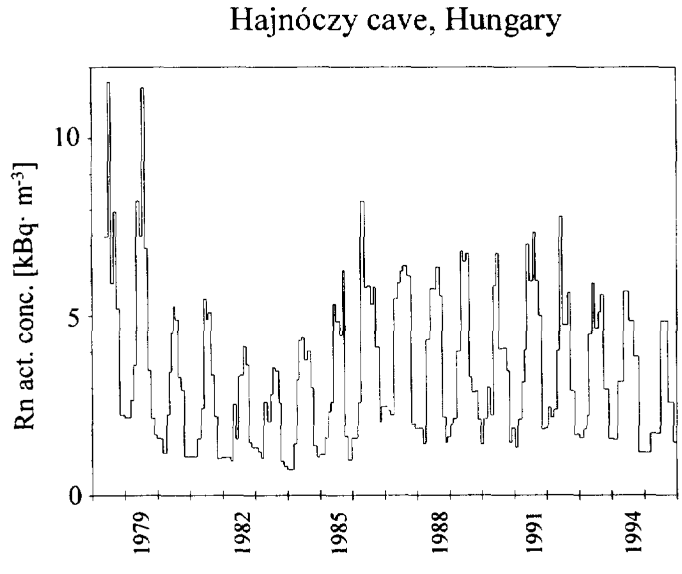

Figure 2: Radon time series observed by using etched track technique in the Nagy-terem (Great Hall) of the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary. Each observed value is the average of three simultaneous readings with one month integration time.

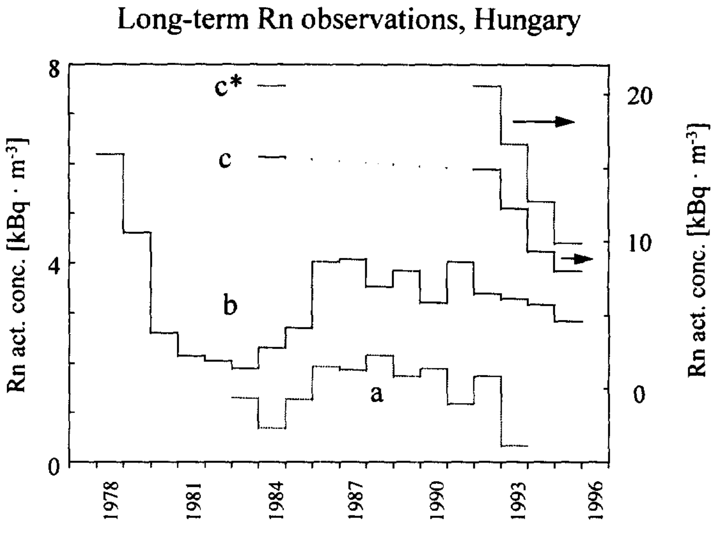

Figure 3: Long-term change observed in three Hungarian caves of different types. Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary, is dominated by chimney effect winds; Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary, is characterized by temperature difference driven convective air motions; and Cserszegtomaj well-cave, Keszthely Mountains, SW Hungary, is influenced mainly by atmospheric pressure changes. The curves represent the middle part of the Létrási-Vizes cave (a); the Nagy-terem (Great Hall) in the Hajnóczy cave (b); and cave average (c) and a remote part (c*) in the Cserszegtomaj well-cave.

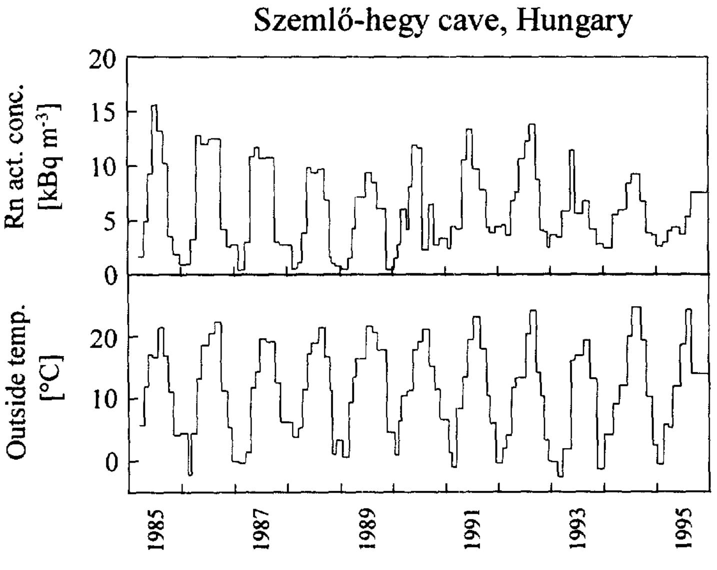

Figure 4: Outside mean temperature and radon time series obtained at the Közgyűlés-terem (General Assemly room) of the Szemlő-hegy cave, Buda Mountains, Central Hungary. The sealing of an artificial shaft in winter 1989/90 influenced ventilation regime and consequently radon level in the cave.

So far the longest time series were obtained in the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary (Fig. 2), which shows very regular seasonal variation. The same effect was found in a few other Hungarian caves, but it is also reported by Lively and Krafthefer (1995). The observed fluctuations of smaller or larger amplitude are a general phenomenon, where the frequency and amplitude distribution of the data is a common characteristic for the cave and its environment.

It is also clearly seen on the Fig. 2, that the seasonal variation is superposed on a long-term change of the mean activity concentration. This long-term change can be found in different types of caves, as it is shown on Fig. 3. They display the effect of slowly changing environmental parameters on radon transport processes. Such external parameter may be the change in radon emanation power due to the slow change of water content of the cave surrounding, which may reflect the variation in annual precipitation and global meteorological situation influenced by sunspot cycling (Hunyadi *et al.*, 1988).

The observed annual change of radon activity reflects two basic processes. Wilkening and Watkins (1976) identified temperature gradients favorable to *vertical convective transport* through relatively large openings. They identified as well transport of radon by *air movement through cracks and fissures* due to pressure gradients (Wilkening, 1980). As karst caves are situated generally in highly fracturized rocks, such a configuration is favourable for the emergence of air circulation through this fracture system. The strength of such air motions is taken to be proportional, to a first approximation, to dT/f, where dT is the temperature difference between the cave and outside and f is a friction factor characterizing the flow resistance (Wilkening and Watkins, 1976; Atkinson *et al.*, 1983). A typical radon time series indicating this type of temperature control is shown on the Fig. 4 (Szemlő-hegy cave, Buda Mountains, Central Hungary). On approaching deeper parts of the karst, the radon levels are mostly stable, as the strength of air motion decreases due to their 1/f dependence; and owing to saturation effects the amplitude of changes also decreases (Hakl *et al.*, 1992). Radon levels in deep, complex caves cannot be simply related to outside atmospheric parameters (Cunningham and LaRock, 1991).

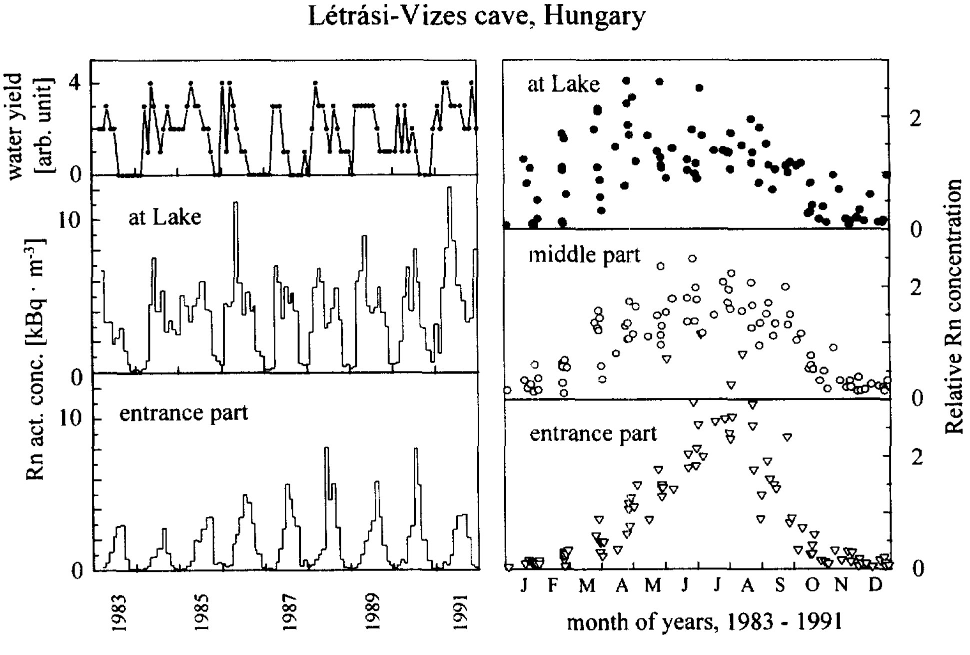

Figure 5: The yield of the stream feeding the Lake and the temporal change of radon activity concentrations in air at the Tó (Lake) and at the entrance part of the Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary (left). The form of the upper curves, in spite of low temporal resolution, shows a connection between the two quantities. The phenomenon is explained by the fact that water is the dominating radon source in cases of floods. The difference in springtime variations of normalized monthly radon distributions (right) reflects the same phenomenon, simultaneously showing radon regimes of the three distinct microclimatic zones.

The actual value of the radon concentration in the cave is influenced by subsurface fluid motions due to the periodically or randomly changing gradients in the environmental parameters (temperature, pressure, humidity, stresses,...), and by the radon concentration saturated in the pore spaces of the surrounding rocks. Using long-term temporal and large scale spatial variation measurements from the analysis of temporal radon variations, the complex interplay of the two traced substances, water and air, can be identified.

Such measurements were performed in the Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary (Lénárt *et al.*, 1988). Typical patterns of temporal changes are characterized by summer maxima and winter minima. Similar variations were found at almost all measuring sites with the exception of that part where more or less continuous water inlets were present. According to the temporal variation, Lénárt *et al.* (1988) differentiated three parts in the cave. In the first part (entrance part) the mean radon activity concentration in air increased from 1 kBqm^{-3} to 2.2 kBqm^{-3}. In the second part, it fluctuated around 2.2 kBqm^{-3}. We note that in these two parts there was a slight asymmetry in the way the radon changes between summer and winter. In the springtime, radon levels in the cave increase gradually, whereas in the fall decline rapidly. (On increasing the time resolution of the measurements, the same effect can be made much pronounced, as was found by Lively and Krafthefer (1995) using active techniques.) The highest air radon values, around 4 kBqm^{-3}, were found in the third part, which was characterized by the presence of several streams and a Lake fed by them. In the third part the dominant role of the periodic stream is evident from the long-term measurement (see Fig. 5); while in the first two parts the change of the radon activity concentration showed close connection with the temperature difference between the cave and outside.

Radon Risk Inside and Above the Caves

It is known, that high radon exposure results in excess lung cancer mortality rate among miners. A majority of experts believe that elevated radon levels in homes induce excess lung cancer mortality rate among the general public, too. High radon levels that occur in caves may rise the question whether any people are at risk. Most concerned groups are, the cavers and tour guides. Moreover, some cold karst caves are used for therapeutic treatment of patients suffering from chronic respiratory diseases. These patients, and the staff members of the therapy are also concerned about being subjected to higher risks. Hungarian data were summarized by Csige *et al.* (1996) (see Table 1).

Table 1. Summary of radon doses received on the course of speleotherapy in Hungary (Csige *et al.*, 1996).

			Annual effective dose, mSv	
Cave name	Period	Sample size	Mean, (Range)	Geometric mean, (Geometric STD)
Abaliget[1]	1994	127 patients	0.54, (0.03-1.26)	0.40, (0.77)
Béke[2]	1994 summer	56 patients	1.91, (1.86-1.97)	1.91, (0.02)
Szemlő-hegy[3]	1990-1992	229 patients	0.85, (0.10-5.00)	0.62, (0.76)
Szent István[2]	1994	360 patients	0.06, (0.01-0.17)	0.04, (0.88)
Tapolca[4]	1994	481 patients	0.87, (0.04-2.19)	0.45, (1.32)
Baradla[2]	1990-1994	12 tour guides	2.66, (0.12-5.55)	2.13, (0.80)

1 - Mecsek Mountains, S Hungary; 2 - Aggtelek Karst, Aggtelek National Park, NE Hungary; 3 - Buda Mountains, Central Hungary; 4 - Balaton Highland, W Hungary; In the case of Szemlő-hegy cave (3) cumulative effective doses are given over 1990-1992, as not all the patients attended each year to the therapeutic courses.

Currently, in Hungary radon is continuously monitored in all the five caves where therapeutic treatment of bronchial asthma and chronic bronchitis is going on. Some caves open to tourists are also monitored. In the case of the best known of these (Baradla cave, Aggtelek National Park, Hungary), the radon doses of tour guides were estimated for the years 1990-1994.

According to Navrátil *et al.* (1994) the range of patients' doses from nine different therapeutic caves in Europe is 0.07-1.32 mSv, which well fits with the above table. Based on internal working time reports, annual effective doses for therapy staff members are about 0.4 mSv in Abaliget cave, 0.12 mSv in Szent István cave and 6 mSv in Béke cave. Annual personnel exposures also cover a wide range. Tour guides in Abaliget cave receive about 12 mSv annually, Cigna and Clemente (1981) cite Yarborough, who has found 0.005-19.9 mSv per year for seven different US caves; whereas Nikodemová (1995) reports effective dose rates 0.17-4.05 mSv per month for personnel in Slovakian caves.Most active cavers may be exposed to even higher doses; so personal radon dosimetry is highly recommended for those people. One of the authors (J. Hakl) has measured his own annual effective dose due to radon inhalation while working in different caves, and found that it was higher than 30 mSv in 1992. Even higher personal doses can be achieved in caves with unusually high radon concentrations. Hyland and Gunn (1994a) estimated that it took 33 hours to reach 15 mSv limit in some caves of North Pennines in the U.K. Two important radiological consequences follow from the periodical behaviour of subsurface air circulation in karst. First is the seasonally varying underground radon level which results in a large difference between summer and winter values, and hence also in the radon exposures received by cave visiting people.

The other consequence of the seasonally directed transport phenomenon is that variation in radon exhalation can also be expected on karst terrains seasonally. A very convincing experimental result of the phenomenon is represented on Fig. 6, where radon time series measured inside the Hajnóczy cave, Hungary (with minima in the cold winter season), and in a slit above the cave (with winter maxima) are shown. Similar winter maxima were found on several karstic terrains of Hungary (Hakl *et al.*, 1992). Elevated radon levels already observed in houses in summertime (Wilson *at al.*, 1991) and wintertime (Gammage *et al.*, 1992) are both attributed to this phenomenon, showing that the season of maxima depend on the position of the house with respect to the underground air circulatory system. The inverse correlation of radon levels inside a cave and in a house above the cave was found directly by Lively and Krafhefer (1995).

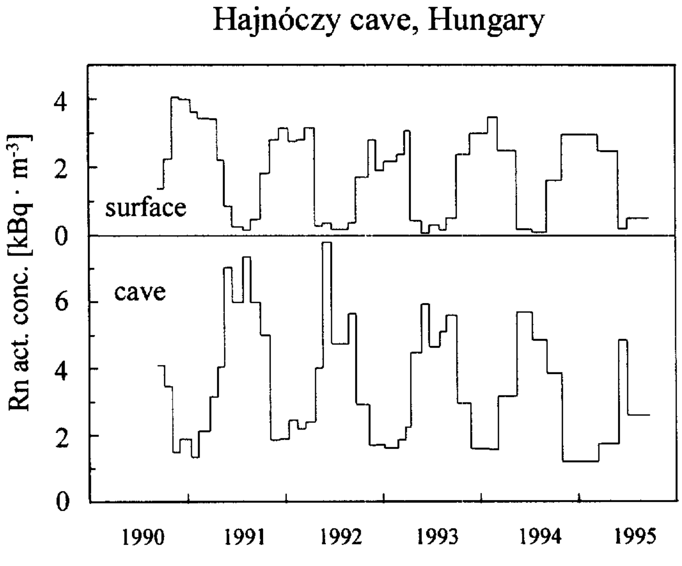

Figure 6. Push-pull type change of seasonal radon activity concentrations observed in the Nagy-terem (Great Hall) of the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary, and in a surface slit above the cave.

Combination of Etched Track and Real-Time Detection Techniques

Combining etched track and real-time detection techniques, spatial and detailed temporal variation of radon concentration can be obtained. Such type of combination of continuous and integrating radon measurements was performed in the Vass Imre cave situated in the Aggtelek Karst, Aggtelek National Park, NE Hungary. It has one entrance and is situated practically horizontally with no visible vertical connection to the surface. In the middle of the cave there is a syphon, which was closed by water several times during the observation period (1987-1993). When the syphon is open, there is continuous air flow either in or out from the cave through the entrance (winter and summer, respectively). When the syphon is closed, there is no measurable air flow through the entrance. The radon concentration is being measured with etched track detectors since 1987 at 12 sites located at equal intervals along the cave passage, and in 1991 three more continuous real-time radon monitors were installed at characteristic points of the cave. Etched track detectors changed monthly showed elevated levels in the summer and lower levels in the winter. The penetrability of the syphon markedly affected the radon levels through controlling the mass of infiltrated air through the cave system. The restriction of the air flows has different effect on the forming of radon levels depending on the place of measurement (in front of *or* behind the syphon) and on the season. In summertime the restricted air flow decreases (8.5 kBqm^{-3} → 4.0 kBqm^{-3} *or* 8.5 kBqm^{-3} → 3.1 kBqm^{-3}), but in wintertime it increases (1.2 kBqm^{-3} → 2.5 kBqm^{-3} *or* 1.2 kBqm^{-3} → 3.5 kBqm^{-3}), the mean radon levels in the cave, resulting in an overall drop (4.8 kBqm^{-3} → 3.3 kBqm^{-3}) of 30% in the annual mean radon concentration . This asymmetric effect can be explained on the basis of the air circulation through covering strata (Géczy *et al.* 1988) and is in agreement with the results of numerical calculations (Holford *et al.* 1993) showing the increase of mean radon levels due to periodically changing flowing conditions. On analyzing temporal changes in radon time series, we found advection as the dominant transport process. This is also shown on the transient part of the radon record from the Vass Imre cave, Aggtelek Karst, Aggtelek National Park, NE Hungary, which corresponds to a summertime syphon opening (see Fig. 7 left). The shape of the curve, on comparing it to the results of numerical calculations on radon exhalation from porous media (Janssens *et al.* 1984), corresponds to the case when a sudden pressure drop results in instantaneous air-flow development. The pressure drop effect was also observed experimentally during a planned syphon closing and opening. On the other hand, the absence of strong daily changes in the radon record, which would correspond to the observed daily air flow fluctuations, shows the strong smoothing effect of diffusion, which can be due to relatively undeveloped fracture system of this cave. (We note that in caves located in more karstified environment, more or less pronounced daily fluctuations in radon records can be observed.) The cave radon concentration 'shut down' is very quick in autumn and is controlled purely by advection. An early cold front of short duration caused a fall in radon records at the first and second measuring sites, but it did not affected the end of the cave. High radon levels were recovered for two days by reversing the air-flow. Finally, permanent low radon values were formed in the cave air after the arrival of another, stronger cold front. The time-difference between the radon falls corresponded to an air transport velocity of about 50 m·h^{-1} along the main cave passage (see Fig. 7, right).

In some case, in caves with small entrances the changes in outside temperature have only a small effect on radon concentration inside the cave, and only on an annual scale. In contrary, the atmospheric pressure have strong influence on radon level, pressure changes play important role in controlling radon transport. The radon time series, observed in the Cserszegtomaj well-cave, Keszthely Mountain, SW Hungary, show this effect markedly (Fig. 8, left). Decreasing pressure increases the radon level and inversely. The hysteresis found in the pressure-radon correlation curve indicates nonlinearity of processes. It corresponds to a time shift in the response of air-flow velocity to the pressure changes (Wigley *et al.* 1967). From the data it is also possible to estimate the cave volume, using simultaneous pressure measurements (and utilising the known volume of the vertical entrance well). We found it to be 8-10 times larger then was estimated from the calculated volumes of passages. On the right part of the figure, for the sake of comparison, also displayed the radon response of a chimney effect controlled cave system.

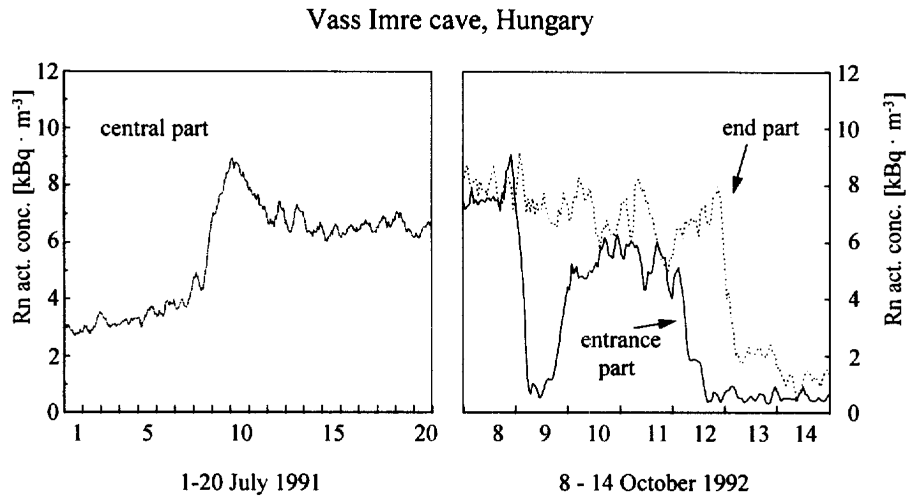

Figure 7: The real time records of radon time series from Vass Imre cave, Aggtelek Karst, Aggtelek National Park, NE Hungary, at the times of syphon opening (left) and arriving of early cold fronts during fall (right).

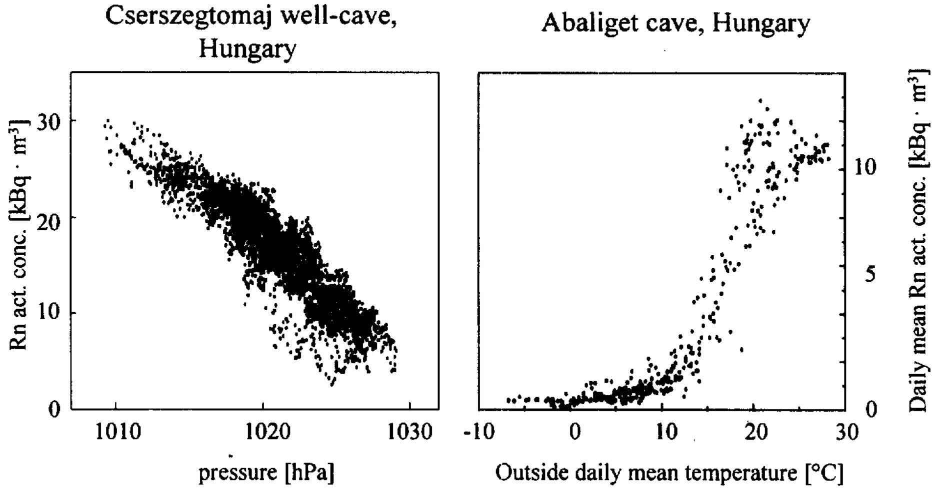

Figure 8. Radon activity concentration vs atmospheric pressure observed at one of the five continuous observation stations in the Cserszegtomaj well-cave, Keszthely mountains, SW Hungary (left). The 'data cloud' corresponds to the 2 month section of the continuous data record. The inverse pressure control on radon levels is well seen in the figure. On the right-hand part of the figure we have displayed radon levels vs outside temperature observed in the Abaliget cave, Mecsek mountains, S Hungary. The S-type dependence of radon concentration on the outside temperature is typical for caves dominated by chimney effect winds.

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