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To the memory of
DR. GYÖRGY SOMOGYI

CONTENTS

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HISTORICAL REVIEW OF ONE DECADE RADON MEASUREMENTS IN HUNGARIAN CAVES PERFORMED BY SOLID STATE NUCLEAR TRACK DETECTION TECHNIQUE

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(HUNGARY)

1. ABSTRACT

Starting from 1978 in as many as 17 caves of Hungary regular radon observations were carried out at 146 different measuring places performed with monthly changed integrating radon detectors in meteorologically less perturbed circumstances. The shortest observation was near two years long, while the longest is still going on from the beginning. The aim of the first observations was to get basic information about underground radon levels. The phenomenon of seasonal variation, the role of radon transporting substances, the dependence on temperature, depth and rock environment resulted in from the numerous longterm measurements.

2. INTRODUCTION

In the last decades the development and the widespread application of subsurface radon measuring techniques were motivated by the effort to locate the exploitable uranium deposits. For the purpose to estimate the most important parameters (depth, extension, enrichment,..) of the deep uranium deposits from near surface radon measurements it has become important to investigate in details the radon transport processes taking place in some hundred metres upper layer of the earth's crust which is in strong interaction with the atmosphere.

The discovery of solid state nuclear track detectors (SSNTDs) and especially the most sensitive polymers gave a new impulse to the subsurface radon mapping experiments in the 1960-70-s. Passive, integrating and in addition simple and inexpensive radon measuring devices equipped with a small piece of alpha sensitive polymer foils proved to be very powerful tools for the performance of large scale, "in situ" environmental radon measurements of geological interest.

On the coarse of evaluation of temporal and spatial distributions of a great number of simultaneous, continuous radon observation data the attention was focused on new, possible practical and interdisciplinary applications. In addition to the search for uranium ores new researches of oil and natural gas deposits, geothermal resources were initiated and completed by subsurface radon monitoring with SSNTDs.

The observations of subsurface fluid motions traced by natural radon was succeeded by new ideas about the basic transport phenomena and later by new interdisciplinary applications as for example mapping of active faults, investigations of volcanic and seismic activities, earthquake prediction, hydrogeological research etc. For more details we refer to the comprehensive survey of the subject and its references published recently by R.L. Fleischer [Ref.1].

In the speleology, similarly to the previously mentioned fields, these types of measurements already have been found their applications and they give important contributions to the better understanding of the nature of caves.

3. RADON MEASUREMENTS IN HUNGARIAN CAVES WITH SSNTDs

In the Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI) in Debrecen the Nuclear Track Detector Group being the most important national track detector laboratory with good international reputation headed by Dr. George Somogyi was deeply engaged in the SSNTDs radon environmental measurements with the aim of both practical and scientific investigations already in the late 70-s. It was 1978 when G. Somogyi together with G. Németh set off the first dozen radon measuring sites in the Hajnóczy cave of the Bükk mountain.

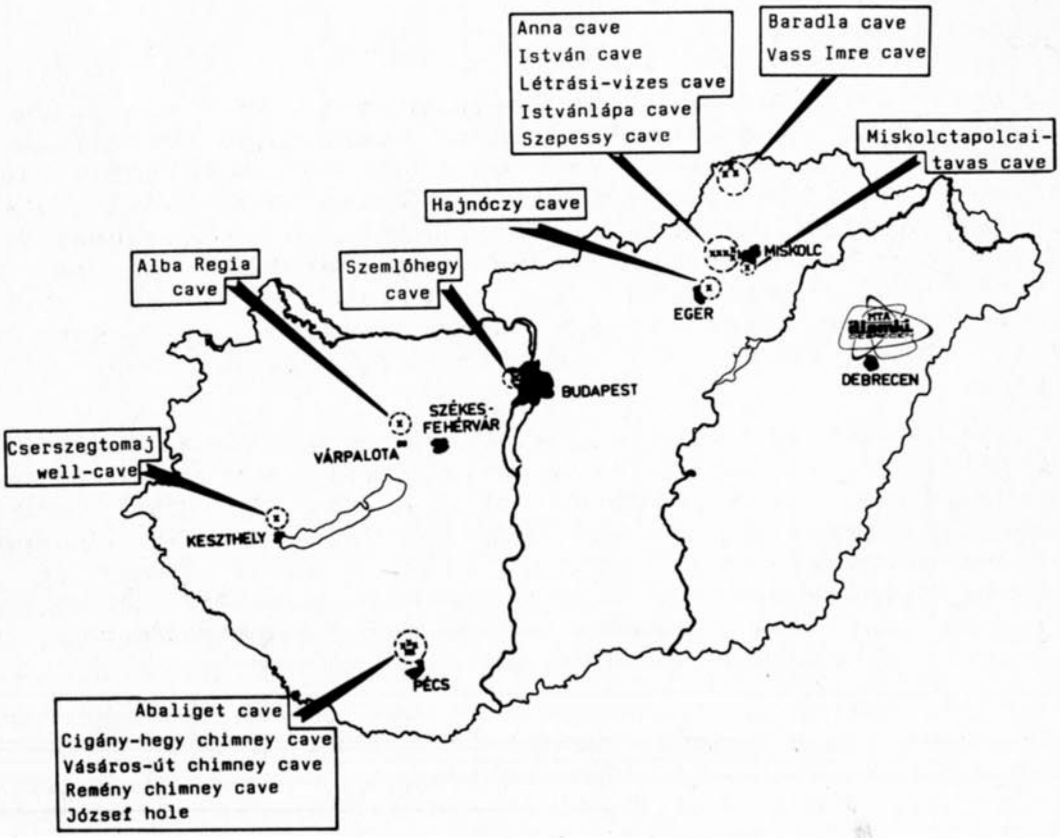

Fig. 1. The location of the caves where regular radon measurements have been performed by solid state nuclear track detectors in the last decade in Hungary.

Later on he has established more fruitful connection with all the interested speleologists and the radon observations in caves were extended to the majority of the Hungarian karstic regions in the last decade (Fig.1 and 2.).

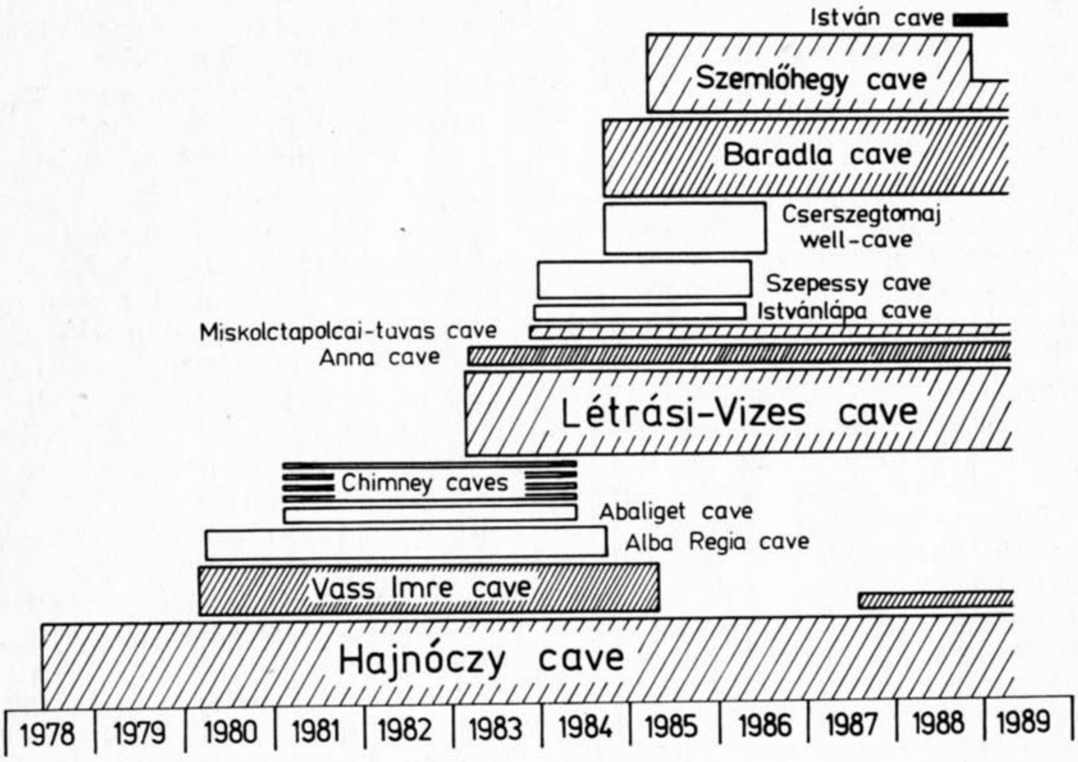

Fig. 2. The date of radon measuring periods for caves with a longer observation time than two years. The vertical extension of the rectangles is proportional to the number of measuring sites established in each cave and it is for example 21 in the Hajnóczy cave .

G. Somogyi was always happy to present experimental results and preliminary conclusions at national symposiums and international conferences [Refs. 2-7] as well as in the Annual Reports of the Hungarian Speleological Society. His interest was permanently on a high level in this subject favoured by him and his energy also seemed endless both in the laboratory and in the cave research until a sudden heart attack took his very successful life in 1987 March at the Hajnóczy cave just after a change of radon detectors. By this way the formulation of the results ad conclusions coming from the observations fell to his colleagues as well as the continuation of the radon measuring program in caves.

The speleologists who took part in the common work for a longer period are G. Géczy, B. Holl, G. Izápi, J. Kárpát, L. Lénárt, L. Rónaki, F. Szolga and I. Törőcsik. We would like to express our gratitude to them also at this occasion for their valuable contributions.

4. RADON IN CAVES AND ITS MEASURING TECHNIQUE BY SSNTD

The radon is a nobel gas and its isotopes (^{222}Rn, ^{220}Rn, ^{219}Rn) as members of the well known natural radioactive decay series are alpha radioactive and present everywhere in the nature as a consequence of that their parent elements(^{238}U, ^{232}Th, ^{235}U, respectively) can be found in more or less concentrations in all constituents of the whole earth's crust.

The characteristics of noble gases mean that radon is difficult to bind in chemical compounds and consequently they can easily separate from the source leaving the place of their origin and freely moving in the underground spaces available for them. It is soluble in water to some extent and can be transported also by water from one place to another.

Because the 0.7% isotopic abundance of the ^{235}U is negligibly low and the 55 seconds half life prevents the ^{220}Rn to move longer distance from its source, the ^{222}Rn isotope of 3.8 days half life is the main constituent of the cave air and waters. The parent element concentrations in limestone environment are 2.2 ppm for ^{238}U and 1.7, 0.016 for ^{232}Th and ^{235}U respectively, according to the world average. (More detailed theoretical considerations can be found in Ref.8. regarding the other characteristic parameters and behaviour of the three radon isotopes in cave environment). As $ppm=10^{-6}*g/g$ is a very low concentration only trace amount of ^{222}Rn, called "radon" in the following, are present in the nature. For the detection of this very low concentration environmental radon the most suitable detectors are the solid state nuclear track detectors which can register their alpha radioactivity.

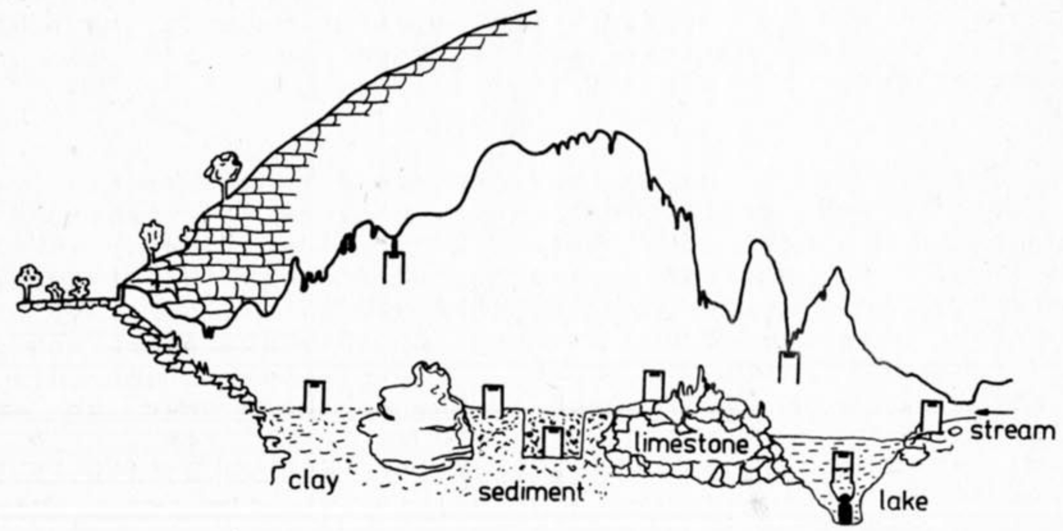

Fig. 3. This schematic draw shows the possibilities how the radon measuring cup equipped with SSNTD can be situated in different media of a cave environment.

Among the numerous advantages of the SSNTDs on one hand their high detection efficiency for alpha particles and on the other a marked insensibility to light, gamma and beta radiation and good resistance to the physical and chemical effects of the geological environment can be mentioned.

The measuring device and its possible arrangements in caves are represented on Fig.3. The 2-4 cm^2, 0.1-1 mm thick alpha sensitive track detectors are attached to the inner bottom of a 10-12 cm long open plastic cup of 7 cm in diameter. The cup protects the detector and keep less disturbed air volume in its vicinity even in that case when it is immersed into water as its open end looks downward.

During the exposition each energetic alpha particle hit the detector foil creates irreversible damage which is conserved in this insulator material for a long time, even for years. The detectors used by us are: i) KODAK LR-115 type II red coloured cellulose nitrate and ii) CR-39 equivalent Hungarian product MA-ND/alpha (allyl-diglycol-carbonate). The damages due to alpha particles can be transformed to microscopic cavities by chemical etching. The number of these etched tracks of 8-10 um in diameter can be counted under optical microscope of 100-200x magnification by manual or automatic scanning. The etching conditions applied are the following: i) 2.5 hours in 10% NaOH solution at 60 °C for LR-115 II and ii) 5-6 hours in 20% NaOH solution at 70 °C for CR-39.

The radon sensitivity of the device was calibrated experimentally and estimated by theoretical calculations, too [Ref.9]. According to our calibration for LR-115 II 1 alpha track/$mm^2 \cdot$30days corresponds to 60 Bq/m^3 radon activity concentration in the air volume of the cup. If the measuring cup opens to the cave air then the measured value can be considered as the radon activity concentration in the cave air and if the measuring cup is immersed into the water the radon activity concentration of the water can be obtained by taking into account the temperature dependent radon distribution coefficient between gas and water phases. This Rn (in gas)/Rn (in water) coefficient goes from 2 to 9 if the temperature changes from 0 to 100 °C. The detectors were changed approximately monthly.

5. GENERAL TENDENCIES AND GLOBAL CONCLUSIONS

Certainly we can state that having so much radon observation series collected continuously in many caves for a long time, we are in the position that by the evaluation of them common tendencies and global conclusions, concerning caves formed in karstic regions of Hungary, can be found. For that caves or region involving more caves where the evaluation of the experimental data are already well-advanced, detailed results and conclusions are presented at this congress in separate contributions [Refs.10-11]. In this paper only a few basic statements and common remarks are given in many cases based on preliminary results.

5.1. Mean radon activity concentration levels

In Table 1. the radon activity concentration levels of the investigated caves are characterized by the mean value which was obtained by averaging all the data observed in the same media of the same cave.

Table 1. Mean radon activity concentrations of subsurface air and waters in Hungarian caves

REGION	CAVE	Radon activity concentration [kBq/m^3] air	water
Aggtelek karst	Baradla	5.3*	0.8 (Styx)
	Vass Imre	2.8	
Bükk mountain	Hajnóczy	3.3	
	Anna	0.7	1.5 (travertine) 3.0 (dolomite)
	Istvánlápa	0.7	0.2
	Miskolctapolcai-tavas	0.5	2.0 (cold) 5.2 (warm)
	Létrási-Vizes	2.2	2.0 (stream) 1.0 (lake) 0.3 (dropping w.)
	Szepessy	0.8	0.2
Buda mountain	Szemlő-hegy	2.7	
Bakony	Alba Regia	1.7	0.3
Keszthely mountain	Cserszegtomaj	14.0	
Mecsek mountain	Abaliget	2.4	
	Cigány-hegy	1.9	
	József-hegy	5.2	
	Remény	1.4	
	Vásáros-út	2.5	

* soil air data also included

The underground cavities investigated by us show a wide variety according to the form, extensions, depth, origin,.. starting from smaller chimney-caves and ending at big complicated systems. However these differences are reflected partly in the measured radon activity concentrations the mean values do not differ one from the other more than a factor of 30. With the exception of the highest value observed in the Cserszegtomaj well-cave they are in accordance with that it can be expected in limestone environment of 2.2 ppm average uranium (radium) content. The enhanced radon activity concentration in the air of the Cserszegtomaj well-cave can be explained by a higher uranium content of the rocks. As it is well known the dolomite and sandstone where this cave is embedded are rich in uranium so this assumption is very likely.

5.2. Short term variations

It is also a general phenomenon that in every caves more or less periodical fluctuations of smaller or larger amplitude can be found around the mean value of the radon activity concentration. The analysis of the time series observed with monthly integration time in the caves in question are still in progress from the point of view of frequency and amplitude distribution of the observed radon data. These parameters are characteristic for the cave and its environment , for the uranium (radium) content of the enclosing rocks and stones and for the extension of that porous surroundings which is in correspondence with the cave air by the intrusion of atmospheric air and radon traced subsurface fluids.

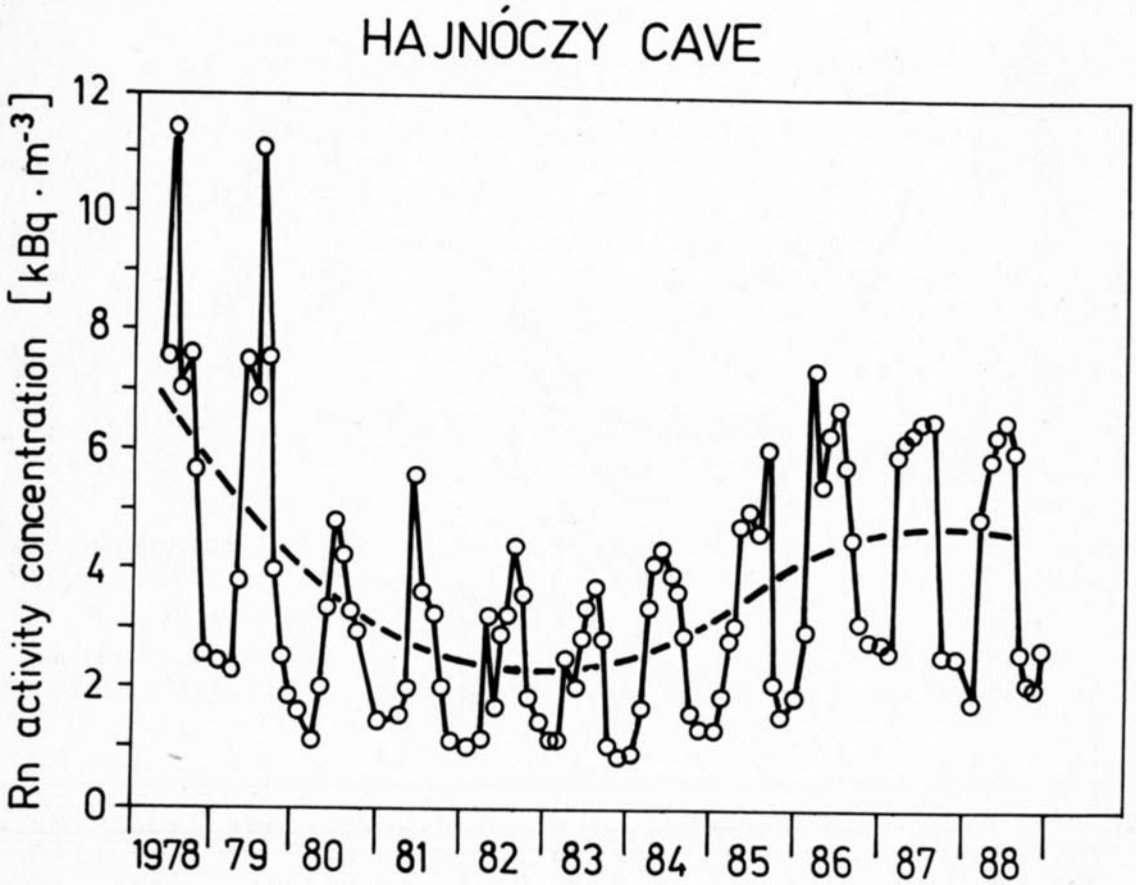

Fig. 4. The seasonal variation with one year periodicity in the radon activity concentration measured with three cups placed on the clay soil in the Great-hall of the Hajnóczy cave. Dashed line resulted in a low pass filter procedure with one year moving average (MA).

It is easy to realize that the direct radon emanation from the wall surface of the known cave labyrinths cannot account either for the maximal values or for the variations occur in the radon activity concentration of the cave air or other substances [Ref.12].

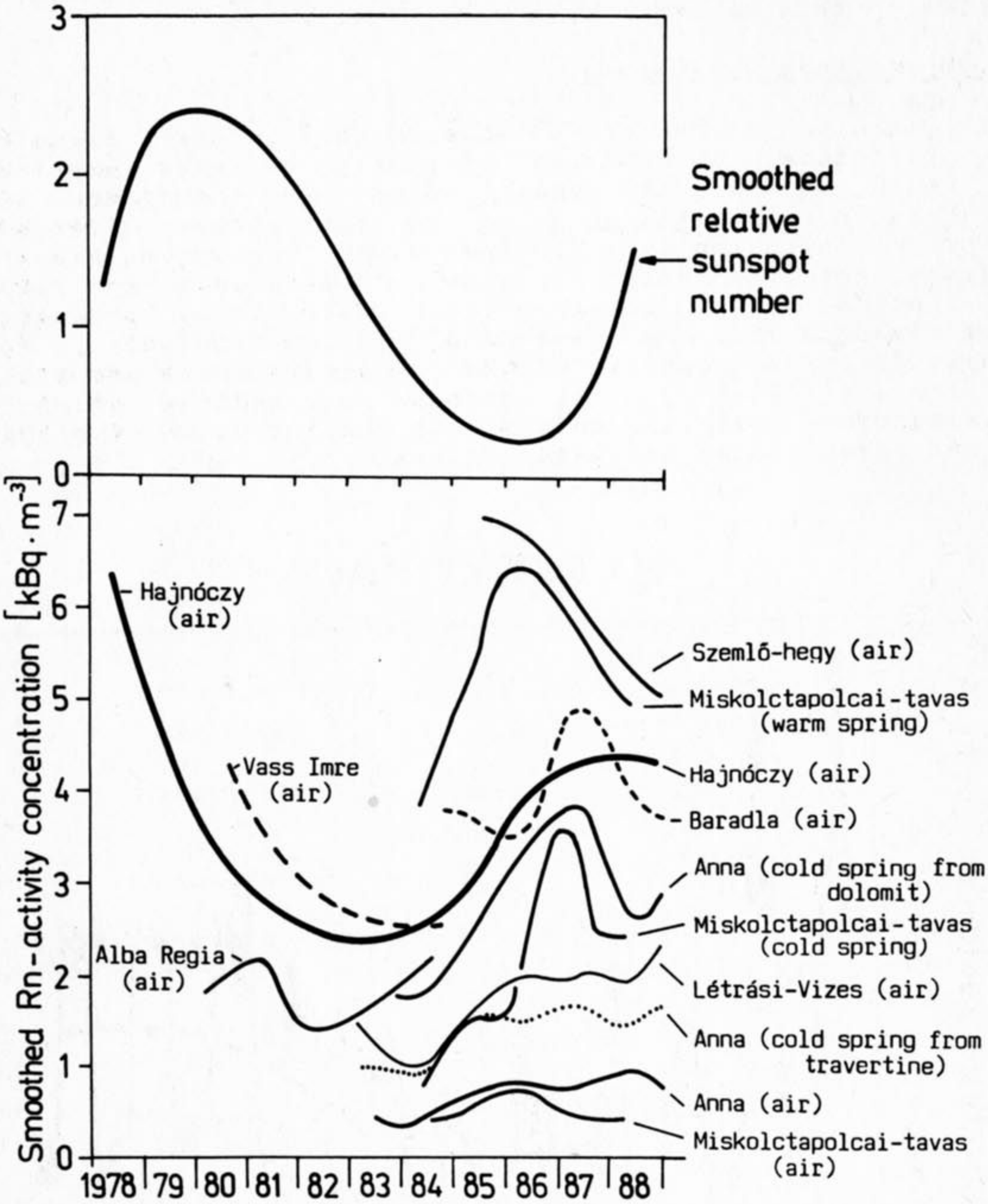

Fig. 5. Long term variation of the subsurface radon activity concentration levels obtained by a one year moving average (MA) procedure from the one month integrated data of the caves involved into the radon studies by SSNTDs. On the upper part of the figure smoothed sunspot numbers are presented in relative units for the corresponding time period.

The actual value of the radon concentration in the cave is mainly influenced by subsurface fluid motions due to 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.

It is demonstrated in our other two contributions [Refs.10-11] that the most common and the most apparent phenomenon which can be discovered from the radon concentration pattern in the majority of the investigated caves is the periodically formed, temperature gradient forced air flow of a seasonally reversed direction. In some cases less regular water inflows are the determining factors in the formation of the radon concentration in caves.

As an example very regular seasonal variations are shown on Fig.4. The longest time series was obtained in the Hajnóczy cave.

5.3. Long term variations

It is clearly seen on Fig.4 that the seasonal variation is superposed on a long term change of the mean radon activity concentration. Therefore we have performed the same averaging procedure for all of radon activity measurements to reveal if any common tendency is present.

It is surprising that in spite of the great variety in the geographical situation of the caves and even for different substances similar basic tendency manifests itself: rapidly decreasing values in the late 70-s are joining through a definite minimum to increasing curves which have a broad maximum around 1986 and decreasing again in the present days. However the measuring period is only eleven years long and this fact does not help to create obvious arguments, the resemblance to the sunspot number figure is quite demonstrative.

Further research are in progress to look for intermediate processes which may help to clarify and understand the correspondence between the two quantities.

ACKNOWLEDGEMENT

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AIR CIRCULATION IN CAVES TRACED BY NATURAL RADON

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1. ABSTRACT

We have been performing continuous radon concentration measurements with solid state nuclear track detector in the Szemlő-hegy Cave since 1985 . The radon concentration shows seasonal variation with summer maxima and winter minimum at each measuring site. The radon concentration increases moving on towards the end point of the cave from the entrance at any time of the year. These changes of the radon concentration can be interpreted with the help of a simple air circulation model of the cave. The possible reasons for the decrease for the annual average radon concentration is also discussed.

2. INTRODUCTION

We have been measuring the radon activity concentration in the cave air of the Szemlo-hegy cave since 1985, joining its complex investigation, by means of solid state nuclear track detectors (SSNTDs) placed at several sections of the cave (Fig. 1.). The cave, similarly to the other large Buda caves, has been formed in eocene nummulitic limestone by the common effect of thermal waters, upflowing along the cracks and of the external waters, infiltrating to the depths. The passages of the cave are nearly horizontal, their total length is 2200 metres. The deepest part of the cave can be found at the end point 44 metres below the surface.

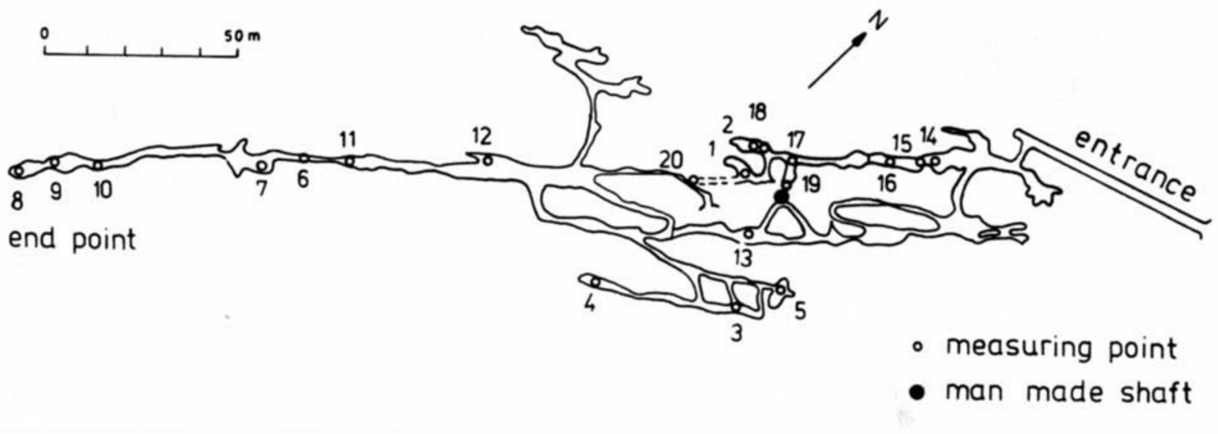

Fig. 1. The map of the Szemlő-hegy cave with radon measuring sites

The ^{222}Rn isotope is a radioactive noble gas which derives in the decay series of ^{238}U which is ubiquitous in the material of the earth's crust. As a consequence of the uranium decay in the rock, involving the cave, an amount of radon gas gets into the cave with the help of the porosity and cracks of the rock. The radon exhalation (assuming a homogeneous stone) is in proportion to the surface of the rock having contact with the air and it is constant in time. The radon concentration of the cave air, formed in this way, is influenced mainly by the air motions in the cave, for the explanation of which we have developed a simple air circulation model.

3. A SIMPLE MODEL OF AIR CIRCULATION IN CAVES

The following model is suitable for describing the air motions in an approximately horizontally situated caves, embedded in a rock with an advanced fracture system.

From the point of view of our model the cave is regarded as a horizontal tubular tunnel whose wall is connected with the external surface by a great number of small cross-sectioned tubes which correspond to the fracture system of the rock. It is assumed that due to the high thermal capacity of the bedrock the temperature inside the cave is independent from the seasons. Their value is equal to the annual mean temperature on the external surface at the given place (apart from the geothermal gradient). The dynamics of the airflow in these caves can be imagined in the following way (Fig. 2):

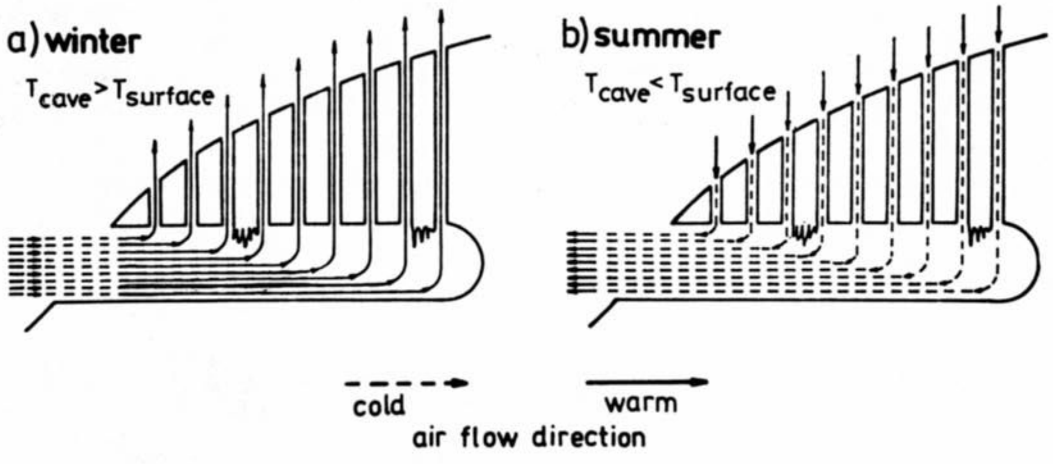

Fig. 2. Air circulation model of a cave with a simple structure

In winter when the temperature in the cave and fracture system is greater than the external one, the so-called "chimney effect" takes place, that is: the warm internal air lifts up to the external surface through the fracture system. For the replacement of the left air, the cold external air flows into the cave through the entrance. The air will be warmed up by the rock, having a much higher heat capacity than the air.

If the external temperature exceeds the internal one then the direction of the airflow becomes the opposite (Fig. 2.a).

In summer the colder air in the cave and in the fracture system is more dense than the external air, thus the cave air flows out of the cave through the entrance (inverse "chimney effect"), (Fig. 2.b).

In both cases the speed of the air motion is determined by the air temperature differences between the cave and outside .

In the following section we look over briefly the basic results and the conclusions.

4. EXPERIMENTAL RESULTS AND CONCLUSIONS

As a result of our four year measuring process the following general tendencies were established in the radon concentration of the cave air:

4. 1. Temporal variation of radon concentration

At any point of the cave an annual periodical variation can be observed in the radon concentration, which shows a good correlation with the variance of the external temperature. The maximum of the radon concentration falls in the summer months, while the lowest values can be observed in winter (Fig. 3).

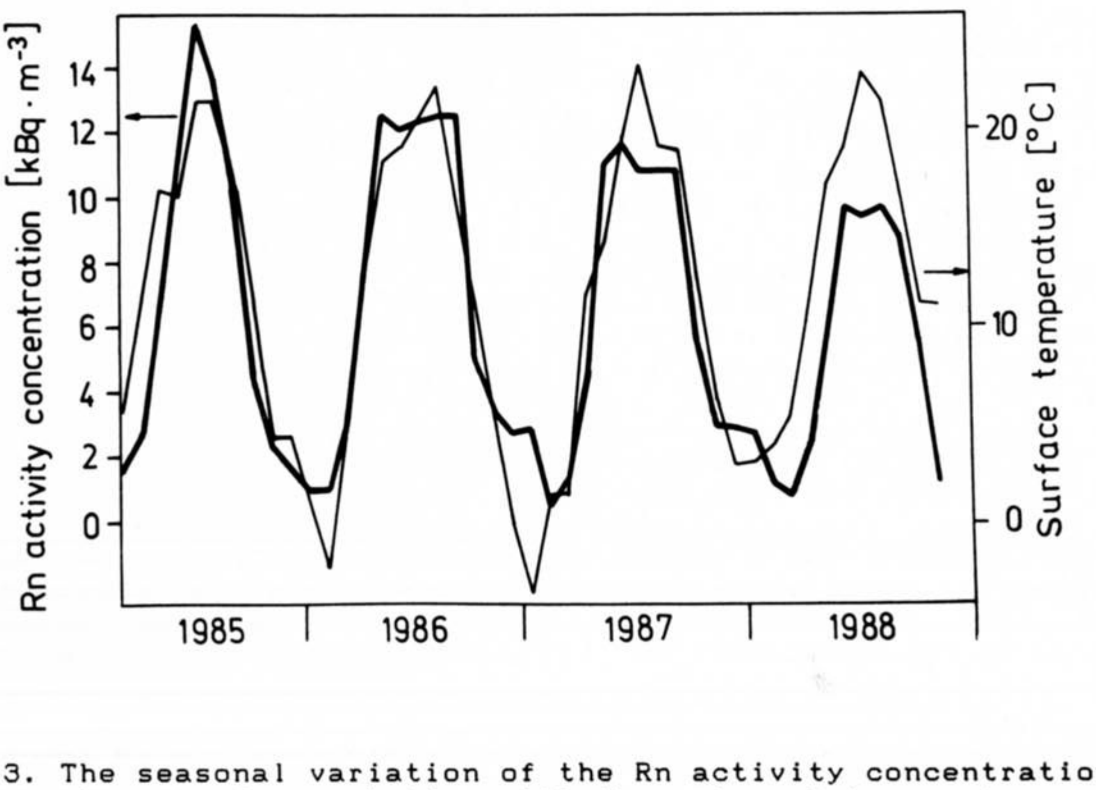

Fig. 3. The seasonal variation of the Rn activity concentration shows good correlation with the external temperature

This phenomenon can be explained in the following way:

Considering a closed subsurface cavity, the radon inflow determined by the cavity surface, and the radon decay depending from the volume included, will together produce a radioactive equilibrium in some half life periods ($T_{1/2}$=3.8 days). In this way the radon concentration in the cavity is determined by the surface/volume ratio of the cavity. If there is no air motion in the cave then a far higher radon concentration is produced in the narrow fracture system than in the roomy cave gallery.

The cave, however, cannot be regarded as a closed cavity; there happens an airflow through the large entrance and the narrow fracture system for the best part of the year. The external air with a negligibly low radon concentration, entering the cave or the fracture system, is enriched by radon as a consequence of the radon flux released from the rock.

According to our air circulation model, in the summer period the external air enters the cave through the fracture system, thus it transports radon continuously from a place of a greater concentration to the measuring point.

Let us first assume that the small tubes, corresponding to the fracture system are of infinite length. Then the radon concentration of the air entering the cave through the fracture system is determined only by the surface/volume ratio of the fracture system, independently from the speed of the inflow. This is the saturation value of the radon concentration in the fracture system. The radon concentration of the cave air increases as a consequence of the radon content transported in, and the greater is the speed of the inflowing air, the better it approaches the value above. In the first approximation, the maximum of the radon concentration falls in the same time as the maximum of the mean external temperature, according to the air circulation model.

In reality, however, the radon concentration of the cave air has two maxima, depending on the location of the measuring point; the first is in May and June, and the second is in August. The maximum of the external temperature falls in this time interval (usually in June) (Fig. 3). The probable reason of this fact is that the tubes, corresponding to the fracture system, cannot be considered to be of infinite length. In that case increasing the speed of the air, flowing through the fracture system, the radon concentration of the air, entering the galery becomes lower and lower comparing to the saturated value.

At a small flowing speed the quantity of the radon, entering the cave through the fracture system can be considered to be in proportion with the flowing speed, at greater speed, however, the effect of the above-mentioned decrease in concentration becomes dominant. As a resultant of these two effects, the radon concentration of the gallery air decreases, above a critical speed. In the case of the Szemlő-hegy cave the external temperature, corresponding to the critical speed is cca. 19 °C.

In the winter period the external air enters the cave through the roomy cave gallery diluting further the low radon concentration, determined by the surface/volume ratio of the gallery. The quicker is the air inflow, the lower concentration is produced at the measuring points.

As a summary, we can state that the temporal variation of the radon concentration of the cave air is the result of the fact that the surface/volume ratio has different values in case of the two possible inflowing way of the external air.

4. 2. Variation of the radon concentration in space

The radon concentration of the cave air shows a variation in space in addition to the temporal changes. At any time of the year, the radon concentration increases advancing to the end point from the entrance (Figure 4.).

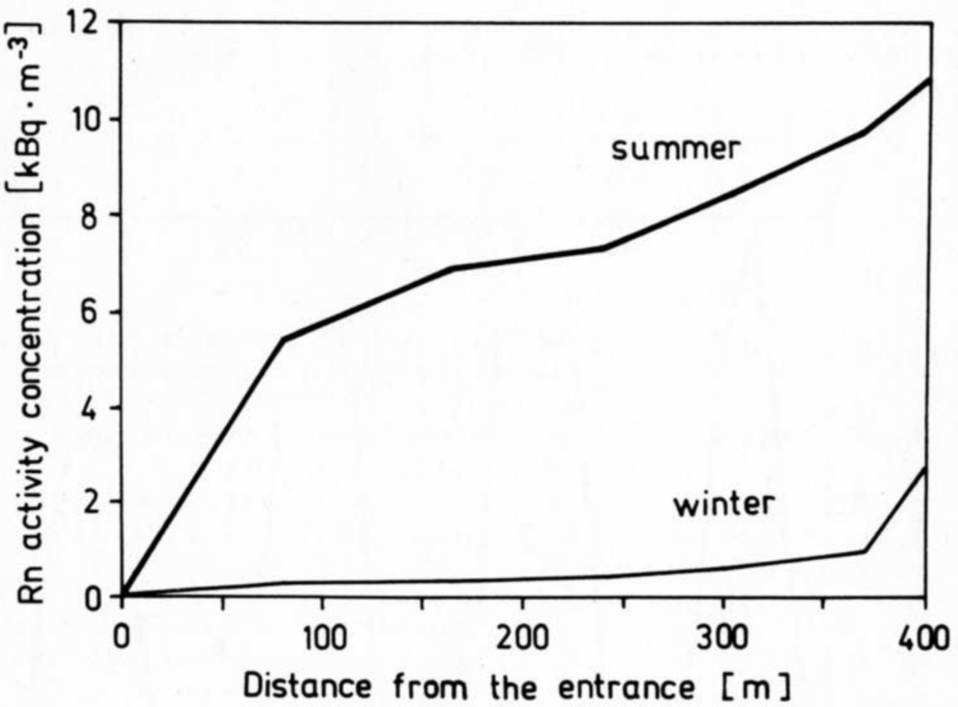

Fig. 4. The radon concentration increases moving on towards the end point at any season of the year

By means of the air circulation model this phenomenon can be explained. In winter the external air, entering the cave through the entrance, receives an amount of radon from the walls of the gallery, therefore the longer way it makes, the grater radon concentration it gets.

In summer the external air enters the cave through longer and longer fracture systems nearing the end point of the cave because of the declivity of the surface. We have already mentioned in Section 4.1. that the tubes, modeling the fracture system cannot be considered to be of infinite length, thus the cave in fact gets the external air with lower radon concentration than the saturated concentration of the fracture system. Assuming equal flowing speed, the shorter tubes will transport less radon into the cave, therefore the air from the end point having a greater radon concentration will be diluted by the air, passing shorter and shorter fracture systems with lower and lower radon concentration, when flowing to the entrance.

Consequently, we can establish that the variation of the cave air radon content in space is the result of the fact that the external air reaches the measuring points after making ways of different lengths.

4.3. The long-term variation of the radon concentration

In addition to the annual variation of the cave air radon content, mentioned in Section 4.1., we can demonstrate a long-term variation as well.

During 4 years the mean radon concentration decreased at each measuring points by cca. 20% (Fig. 5).

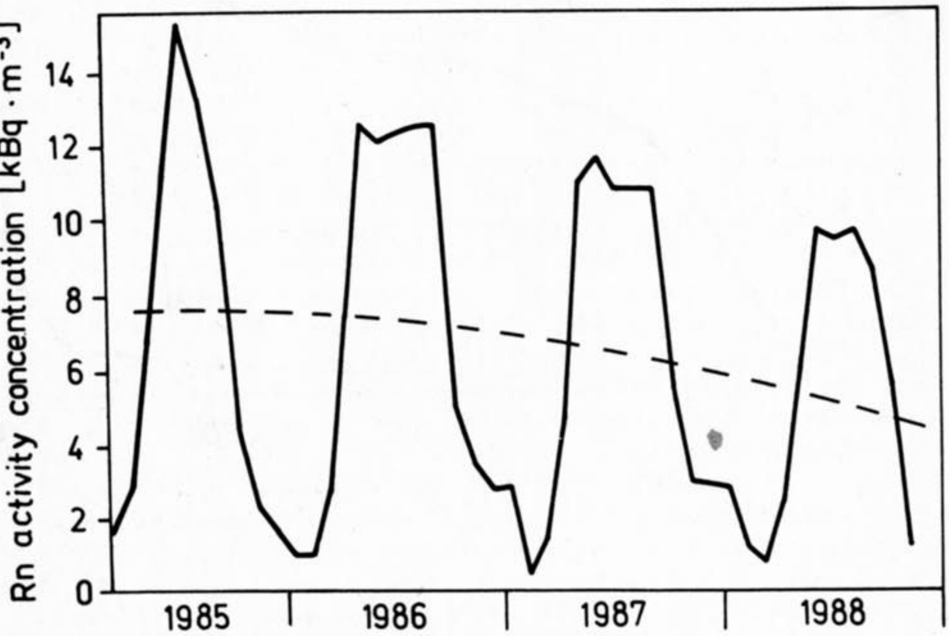

Fig. 5. Decrease of the radon concentration with years

It seems that this decrease is not due to the long-term variation of the external temperature. Depicting the radon concentration of the cave air as the function of the mean monthly external temperature we get curves with hysteresis (Fig. 6). The shape of the curves becomes flatter and flatter during the observation, that is, there entered the cave air less and less radon at the same external temperature.

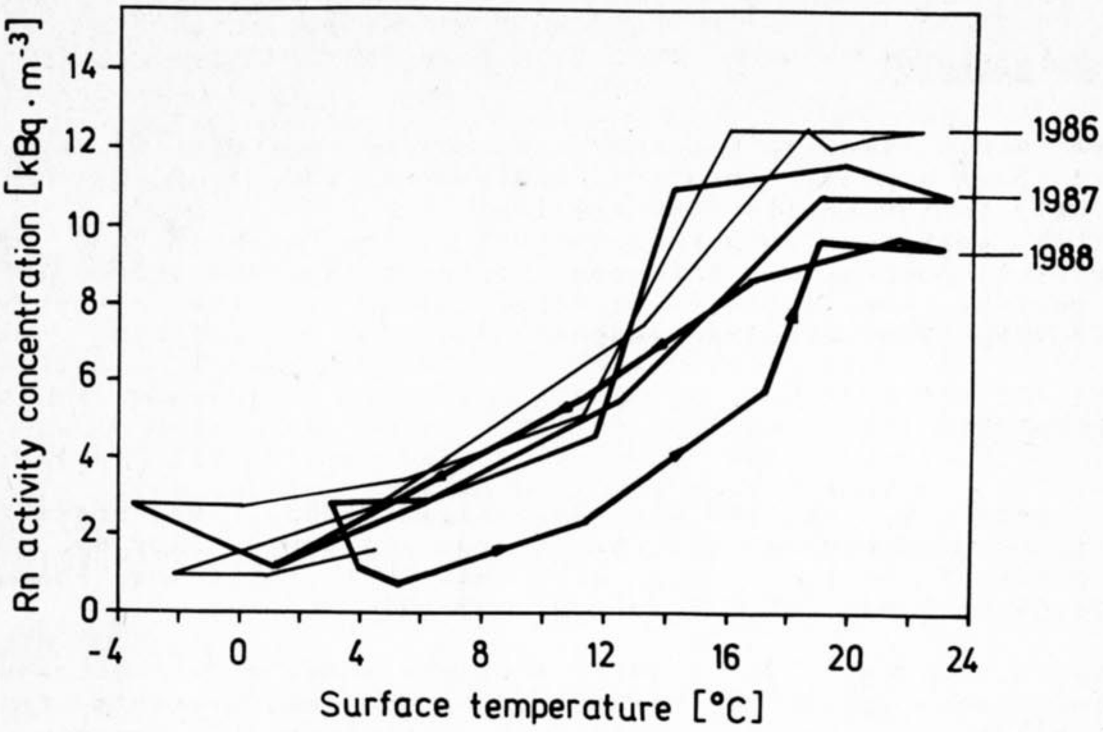

Fig. 6. Relation between the radon concentration and external temperatures changing with years

The variation in the connection between the radon concentration of the cave air and the temperature of the external air indicates that in the cave, regarding it as a physical system, one of the parameters has been changed. Since the mean radon concentration of the cave air, in addition to the quality of the rock, is determined by the porosity of the rock and the surface/volume ratio of the fractures, the reasons of the decrease can be the following:

1. The water content of the rock has changed, which implied a change in the porosity.
2. A part of the micro-fractures has become plugged, therefore the mean surface/volume ratio of the fracture system decreased.
3. The fractures have widened out (in this way their volume increased), thus the surface/volume ratio of the fracture system decreased.

On the basis of the (rather defective) available data there cannot be detected a long-term unidirectional variation neither in the external precipitation, nor in the rock dilation. Therefore we can assume that the radon concentration decreases on the effect of the plugging of the micro-fractures. This hypothesis is also supported by the fact that during the air circulation of the cave, an amount of the pollution of several ten thousand cubic metres air, and the infiltrated external waters are deposited in the cave and in the fracture system.

For the satisfactory clarification of this problem we have begun further measurements.

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ATOMKI Report R3 (1989)

RADON MAPPING IN CAVES OF EASTERN BÜKK REGION

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1. ABSTRACT

Radon mapping has been performed in the Létrási-Vizes, Anna, Istvánlápa, Szepessy and Miskolctapolcai-tavas caves at 37 different places in streaming, dropping and stagnant waters as well as in air and soil. In the majority of cases clear ventilation forced seasonal variation was found, which was strongly affected in some cases by the presence of water. In connection with the Letrasi-Vizes cave it was shown that the inflowing water itself was the most significant radon source.

2. INTRODUCTION

The Institute of Nuclear Research of the Hungarian Academy of Sciences in Debrecen has been performed environmental Rn activity concentration measurements since 1977 with alpha sensitive solid state nuclear track detectors [1]. The first underground Rn measurements started in the middle of 1978 in the Hajnoczy cave situated in the Southern part of Bükk region [2]. These were proceeded by numerous continuous observations in other Hungarian caves. In the Eastern Bükk region we have started measurements at the Létrási-Vizes and Anna caves in February 1983 and at the Miskolctapolcai-tavas cave in October 1983. Also the Szepessy and Istvánlápa caves were investigated for the period slightly longer than one year starting in December 1983 and finally in October 1988 we set off measurement at István cave, which is opened for the public.

2. METHOD

The general description of the Rn measuring technique by solid state nuclear track detector (SSNTD) can be found in Refs. [3,4]. Briefly, the method consist of exposing small strips of solid state nuclear track detector foils by the alpha particles emitted in the radioactive decay of radon, the ubiquitous noble gas. Alpha particles entering the plastic detector create demage zones, which can be revealed as microscopic pits (tracks) by successive etching in NaOH solution. The radon activity concentration is proportional to the observed track density. In our measurements LR 115 type II (Kodak Pathe, France) and MA-ND/alpha (Hungarian Optical Works (MOM), Hungary) track detectors were used in a cylindrical geometry, and the detectors were attached to the bottom inside the cups. In case of underwater measurements a diving cup is used to keep an air gap in the vicinity of the detector foil and the radon activity concentration of water is calculated according to the partition low of radon between water and air phases [5]. Henceforth radon activity concentrations always refer to the indicated phase. The detectors were changed regularly in 30-40 days, and the observed data were corrected to 30 days. These fairly simple measurements afford us an opportunity to study the exhalation of radon gas from the cave walls into the air, and to use the patterns of radon activity to detect currents in cave the air, main direction in the Rn transport processes, to compare Rn concentrations of different substances, to follow seasonal and long term variations, and obviously to find out the average radon concentration. Table 1. summarizes the type and number of different measuring sites.

Table. 1. Measuring sites

Cave	Code	Soil	Air	Spring (Well)	Stream	Lake	Dropping water
Létrási-Vizes	Lt	8	7		3	1	3
Szepessy	S	3	4		1	1	1
Istvánlápa	I	1	2				1
István	I		4				
Mtapolcai- tavas	TT		1	2			
Anna	Am		3	4			

3. RESULTS AND DISCUSSION

3.1. SEASONAL CHANGES AND DEPTH PROFILES

Figs.1. and 2. show typical patterns of radon activity concentrations for the Létrási-Vizes and Anna caves over the six year observation time. One of the curves shows clear seasonal variation with summer maxima and winter minima (Lt7) and similar variation can be found at almost all measuring sites in the Letrasi-Vizes cave with the exception of that part, where more or less continuous water inlets are present (Lt21). In the waters and air of the Anna cave regular changes are still less apparent.Fig.3. shows the vertical cross section of the Létrási-Vizes cave and the corresponding radon activity distribution averaged for the six year long period. In table 2. radon depth distributions for two other caves are presented.

Table 2. Radon concentrations in different depths

Cave name	Code	Depth[m]	Mean Rn activity concentration [Bq/m³]
Szepessy	S1	35	376
	S4	110	830
	S7	140	969
	S10	170	810
Istvánlápa	I4	130	550
	I1	215	700

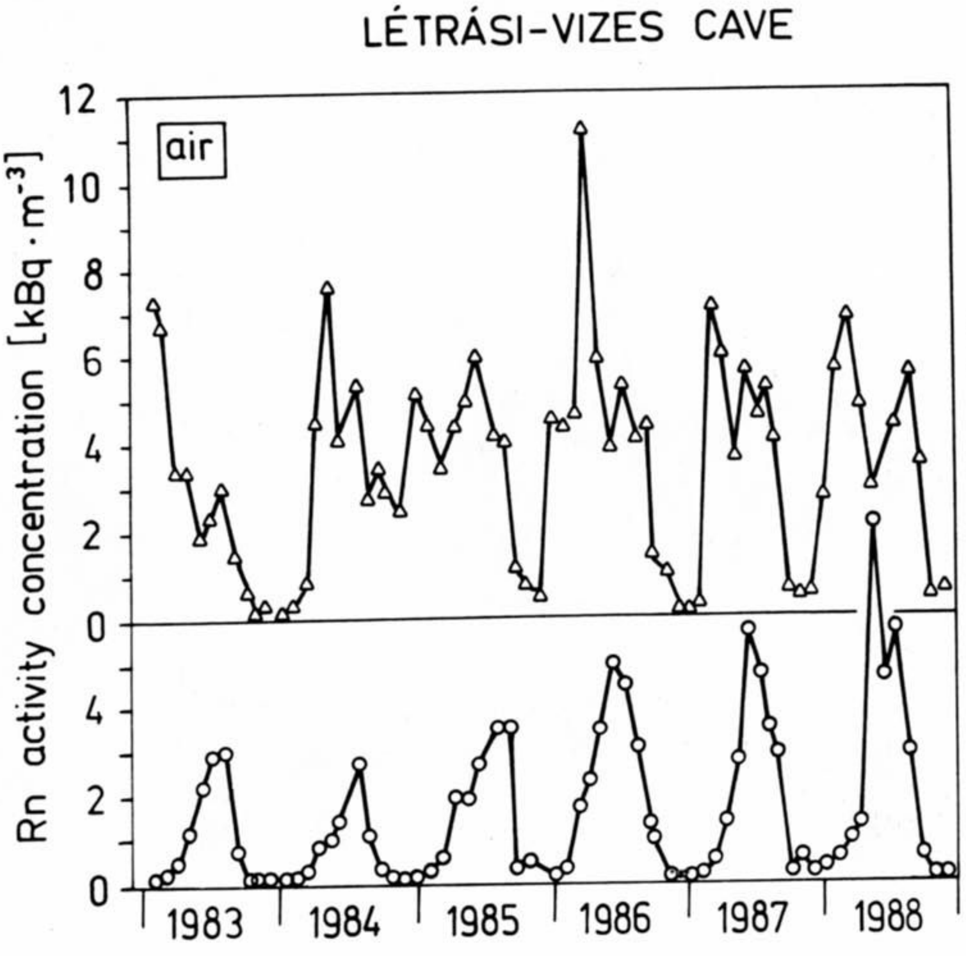

Fig.1. Radon activity concentrations in air of the Létrási-Vizes cave

According to the temporal and spatial variation of the radon activity concentration in the Létrási-Vizes cave we can differentiate three parts (Fig.3.). In the evaluation process of the radon observation series the long term variations of amplitudes were filtered by normalizing the data with one year long moving average (MA). The normalized data are plotted against the elapsed time from the beginning of the actual year and presented on Fig.4.

In the first part the mean radon activity concentration increases from 1 kBq/m³ to 2.2 kBq/m³. The seasonal change of radon activity concentration is symmetrical (Fig.4. Lt7), with a fast increase in spring and fast decrease in autumn. This can be due to the high ventilation of the entrance part.

In the second part the mean radon activity concentration shows a small fluctuation around 2.2 kBq/m³. The seasonal change is more asymmetrical in time (Fig.4. Lt12, Lt18), the variation of spring values is higher.

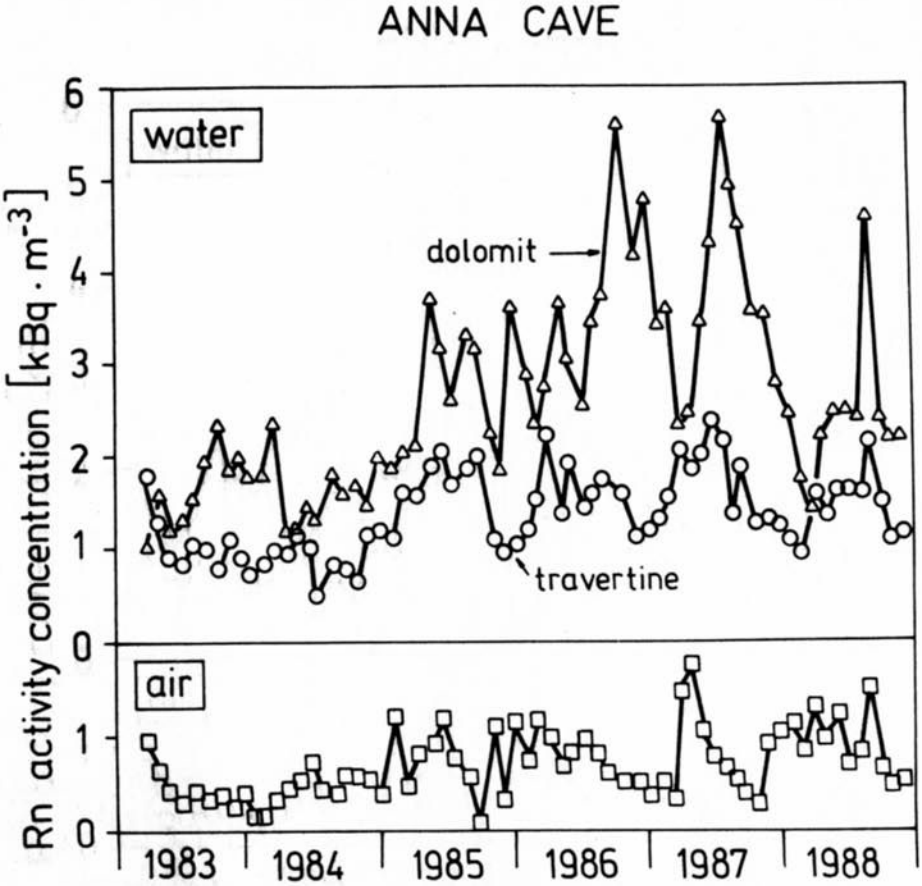

Fig.2. Radon activity concentrations in waters and air of the Anna cave

The change of the radon activity concentrations in the first two parts showed a close connection with the temperature differences between the cave and outside. It follows that in these parts air currents play a dominant role on governing the radon levels [6,7].

The highest mean radon activity concentrations, around 4 kBq/m^3, were measured near the Lake fed by a stream. In this third part the data can be characterized by a high variation in spring and a lower variation in autumn (Fig.4. Lt21). It means that beside the effects of ventilation the inflowing waters have more significant role influencing the radon levels.

3.2. RADON ACTIVITY CONCENTRATIONS IN DIFFERENT SUBSTANCES

Simultaneous measurements of radon activity concentration in different substances were performed at 6 places, 2 in the Létrási-Vizes, 1 in the Istvánlápa and 3 in the Szepessy caves.

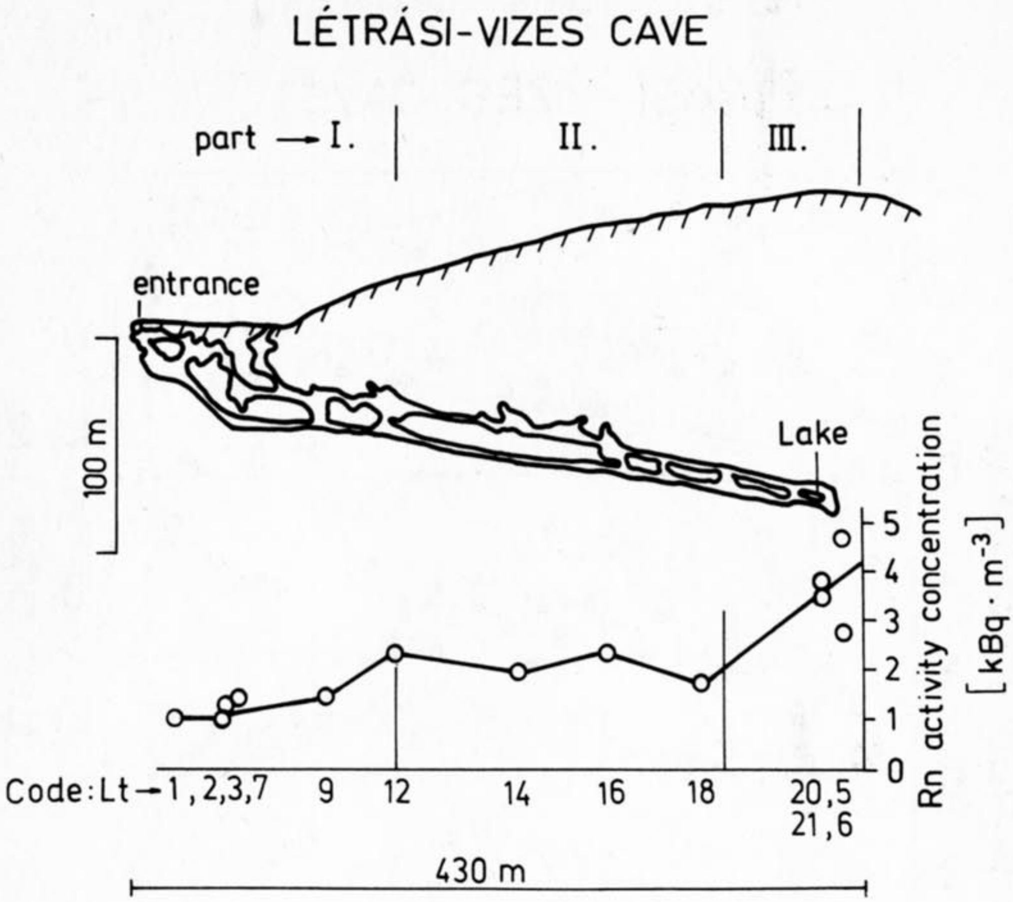

Fig.3. The vertical cross section of the Létrási-Vizes cave and the corresponding average radon activity concentrations at the measuring sites

The longest, therefore the most valuable time series obtained are at the Lake in the Létrási-Vizes cave. Fig. 5. shows radon activity concentration changes at the Lake for four sites over the six year long time period. The observations are the following:

a/ In the Létrási-Vizes cave at the Lake the radon concentration varied strongly with time. The highest average radon activity concentration was found in the stream (2.0 kBq/m³, air equilibrium equivalent = 5.1 kBq/m³), then in decreasing order in the air above the Lake (3.4 kBq/m³), on the surface of a clay hang near the stream (3 kBq/m³) and in the water of the Lake (1.0 kBq/m³, air equilibrium equivalent = 2.6 kBq/m³).

b/ When the stream in the Létrási-Vizes was dry, the radon concentration in the Lake fell to zero, while the concentrations in other substances depending on the season were low in winter and high in summer. In case of active stream, elevated radon levels were always measured in the surrounding of the Lake. The radon concentration found in stream showed a good correlation with the water yield of the stream (see Fig. 5.).

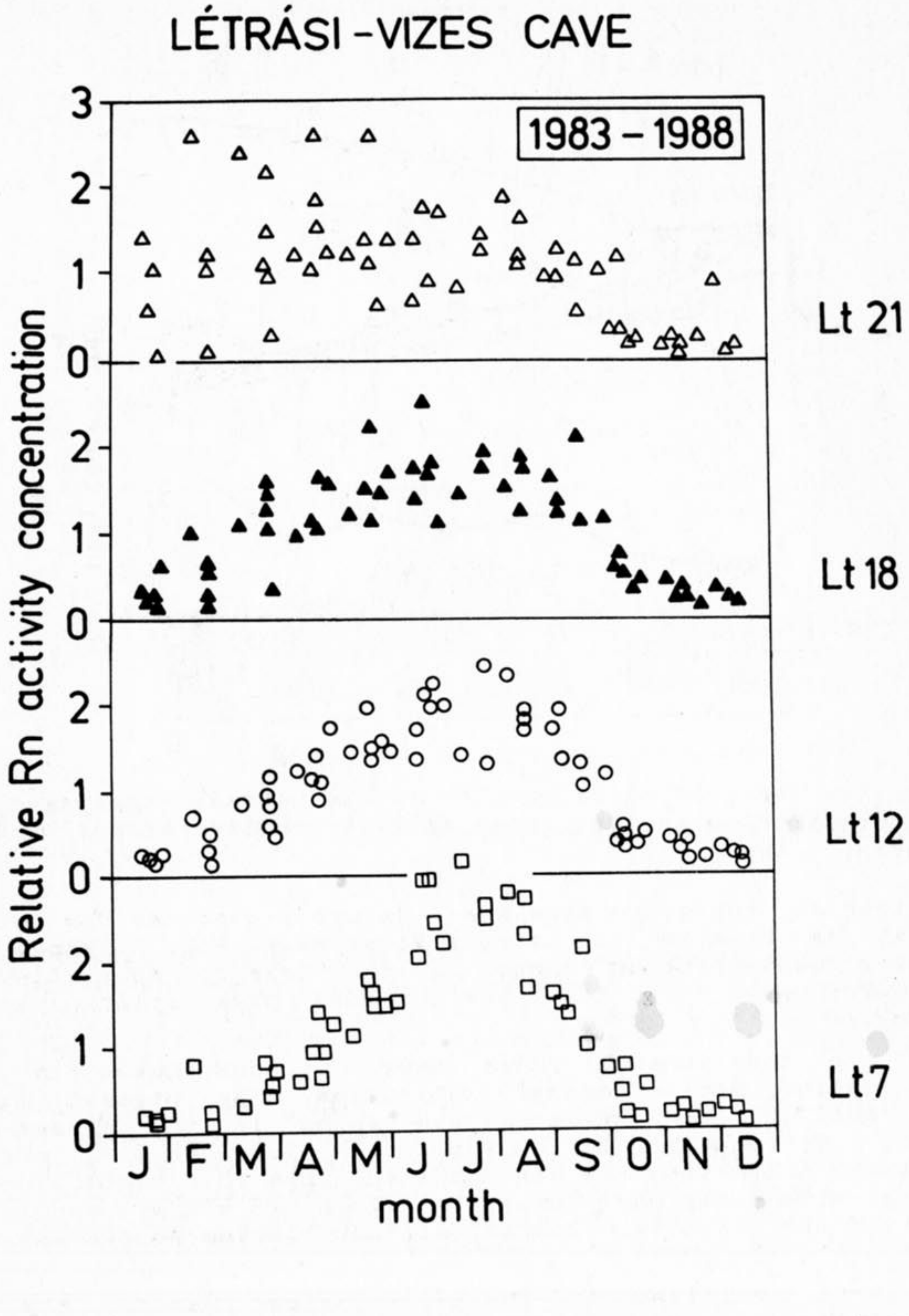

Fig.4. Relative radon activity concentrations in air calculated by normalizing the data by one year long moving averages (MA)

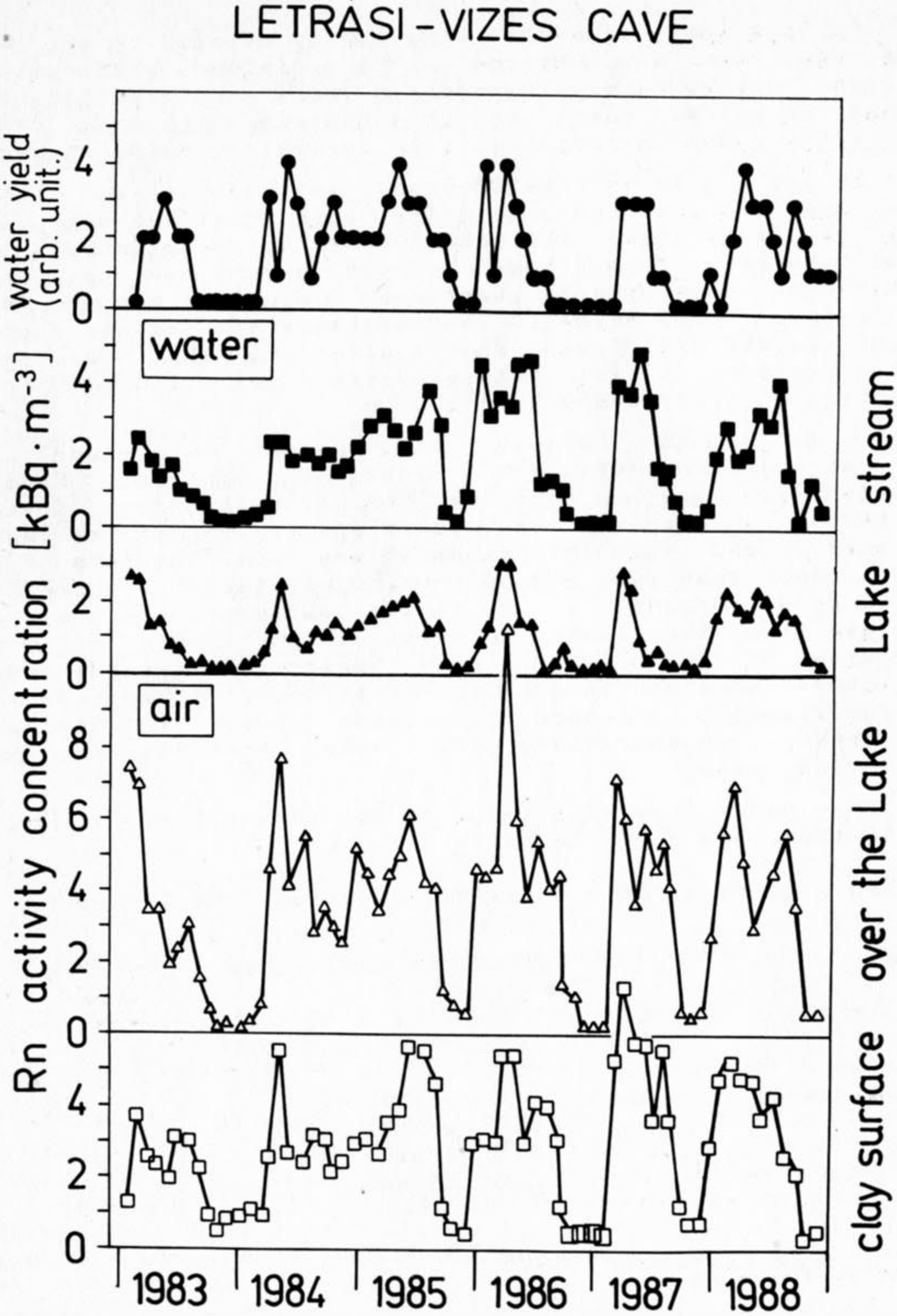

Fig.5. The radon activity concentrations in different substances at the Lake in the Létrási-Vizes cave and water yield of the stream over the six year period of observation

It follows that radon is essentially carried to the Lake by the stream. These observations can be explained by the way, that subsurface waters permeating porous rocks can significantly be enriched in solved radon, so then entering the cave they may increase the radon concentrations by degassing inside the cave.

c/ At each site in the Szepessy and Istvánlápa caves we always measured low radon activity concentrations with only a slight change in time. The sequence of the averaged data showed another character then those observed around the Lake in the Létrási-Vizes cave. The highest mean values were found in soil and air (about 900 Bq/m³). The concentration values found in stagnant waters are lower and similar to that measured in dropping waters of the Létrási-Vizes cave (200 Bq/m³, air equilibrium equivalent=500 Bq/m³).

The difference between the values of mean radon concentrations in different caves is apparent. From the summarized data we can conclude that from the point of view of ventilation the Szepessy and Istvánlápa caves can be regarded much more closed than the Létrási-Vizes cave. The form of these caves (more than 100 m deep narrow vertical entrances) also gives a good support for the latest statement. In these caves there are also the clastic deposits of non karstic origin among the sources of radon, which were transported into the caves by late waters. However in the lack of strong fluid motions, which can significantly influence the radon levels in caves, the formed radon concentrations are lower, than in the nearby Létrási-Vizes cave.

The mean radon levels in these caves, taking into account all the detectors, are given in table 3.:

Table 3. Mean radon activity concentrations [kBq/m³]

Cave	Air	Spring (Well)	Stream	Lake	Dropping w. water
Letrasi-Vizes	2.2		2.0	1.0	0.3
Szepessy	0.8	0.2		0.1	0.2
Istvanlapa	0.7				0.2
Mtapolcai-tavas	0.5	2.0 (cold)			
		5.2 (warm)			
Anna	0.7	1.5 (travertine)			
		3.0 (dolomite)			

3.3. LONG TERM VARIATION

It is apparent on Fig 1. that beside the seasonal changes in the radon activity concentrations at Lt7 measuring site a long term variation is also present effects. This fact can be seen more clearly on the smoothed time series on Fig.6. The mean air radon activity concentration of the Létrási-Vizes cave changed a factor about 2 and the maximum was in 1986-87. The same effect was observable in the cold water spring from dolomite in the Anna cave, where an increase can be found since 1983 till 1987.

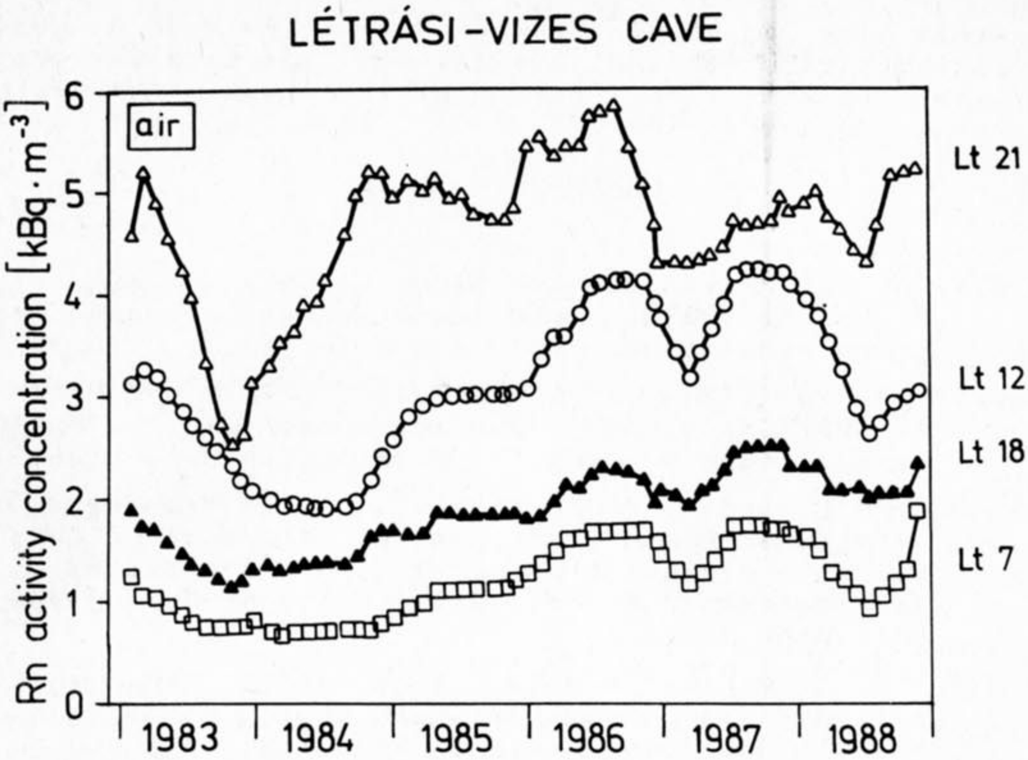

Fig.6. The long time variation of the radon activity concentrations in air at different measuring sites in the Létrási-Vizes cave

In the Miskolctapolcai-tavas cave we observed similar patterns in the warm water spring but only in 1983-85.

4. CONCLUSIONS AND SUMMARY

The primary sources of radon in cave are the bedrock and clay itself. The concentrations of radon in air are formed according to fluid motions, which motions, in addition, were also detectable in the majority of radon records. At the Létrási-Vizes cave when air entered the cave through the entrance in winters, it diluted, decreased radon concentrations, while if through smaller fissures in summers, where it accumulated considerable amount of radon, increased radon concentrations in the cave itself. The presence of radon carrying stream was also traceable in air radon records. In the Szepessy and Istvánlápa caves the lack of strong fluid motions was observable. The similarities found in the long time variation of radon concentrations of different caves of Eastern Bükk region indicate the presence of a general phenomenon, for the better knowledge of which we would like to continue and extend the Rn observations in future.

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ATOMKI Report R4 (1989)

TIME INTEGRATED RADÓN MEASUREMENTS PERFORMED IN A KARSTIC WELL WATER

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1. ABSTRACT

The radon profile of a 270 m deep university well was studied with LR-115 type II detectors. The results indicate a continuous upward transport of radon with a mean velocity of about 0.7 m/h. The phenomenon can be interpreted by the thermal gradient induced convectional mixing of water.

2. INTRODUCTION

Recently the use of plastic track detectors within a defined air volume has become the most reliable procedure for time-integrated, long term measurement of radon activity concentrations under different environmental conditions. Measurements of environmental radon have been performed mostly in connection with geological surveys, health hazard studies and recently for earthquake prediction. However, the dynamics of radon migration is not well understood due to many uncertainties. For a better understanding of the role of external factors, which can significantly influence the transport of radon in water, we have been investigating for three years the change of radon content in a deep well. One of our main purposes was to study the seasonal variation of radon content and its possible correlation with certain environmental parameters (depth, water temperature), and on the other hand, to get information about the transport mechanism and transport velocity of the radon migration itself.

3. DESCRIPTION OF MEASURING SITE AND DEVICES

The well is located at the University Campus of Miskolc, Hungary, in the Northern-Eastern part of the Bukk mountains in a limestonic area. It was drilled in 1971 and was completed with an iron tube casing 270 m in length and 30 cm diameter and screened only at the bottom (Fig. 1.). The water in the well is stagnant and the possibility of horizontal flow or water mixing through the pipe wall is excluded. The temperature of the water at the bottom of the well is 36 °C and it decreases towards the surface with a gradient 0.1 °C/m. Seasonal temperature variations affect only the upper 20 m of the well, the temperature of the water in these parts is in the range 5-15 °C. Starting from 1985 the radon content has been measured simultaneously at 16 different depths in diving cups with a monthly changed LR-115 type II detectors. The exposure times ranged from 30 to 40 days and the data were corrected to 30 days.

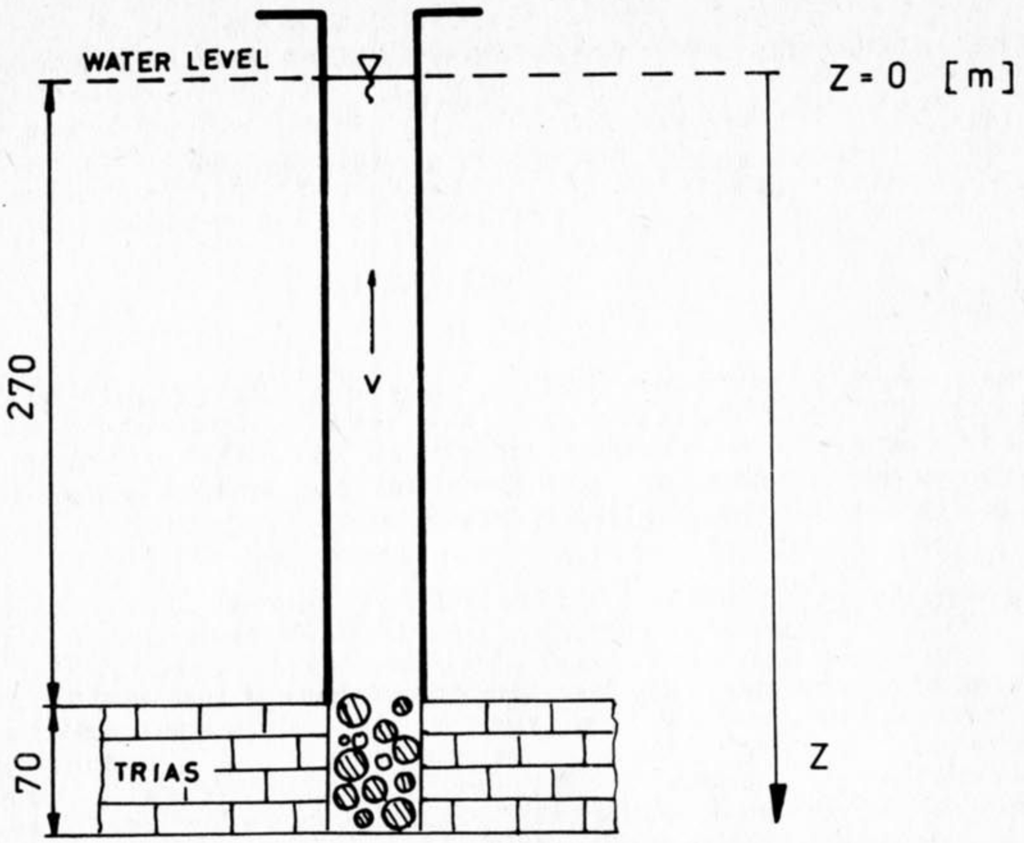

Fig. 1. A schematic view of the Miskolc University well

4. THEORETICAL CONSIDERATIONS

Connection between the measured track densities and transport velocities. Assuming that under steady-state conditions a radon concentration gradient exists only in the direction of depth, the radon transport inside the water column, neglecting local radon production (Krishnaswami and others, 1982) and diffusion, is governed by the differential equation:

$$\frac{d}{dz}\,(v(z)c_w(z)) - \lambda c_w(z) = 0 \qquad \text{for } 0<z<270 \qquad (1)$$

where $c_w(z)$ is the radon concentration at depth z [atom*m^{-3}], $v(z)$ is the upward transport velocity [m*h^{-1}] and λ is the decay constant of radon [h^{-1}]. The integration of (1) yields:

$$c_w(z) = c_w(0)\int_0^z v(z)^{-1}\left(\lambda - \frac{dv(z)}{dz}\right)dz \qquad (2)$$

which describes the radon concentration in the water column. The corresponding concentration of radon, c_A, in the air volume of the cup is formed according to the partition law, which can be described by an empirical formula (Serdjukova and Kapitonov, 1975):

$$c_A = c_w/\alpha = c_w/(0.1057+0.405\exp(-0.05t)) \qquad (3)$$

where α is the radon partition coefficient between air and water and t [°C] is the temperature of the substance. Due to reciprocal density dependence of the range of alpha particles in air, the generated track density would be given by the next formula:

$$\rho(z)=\frac{\alpha(0)}{\alpha(z)}\frac{T(z)}{T(0)}\frac{1}{1+0.1z}\rho(0)\int_0^z v(z)^{-1}(\lambda-\frac{dv(z)}{dz})dz \quad (4)$$

where term ((T(z)/T(0))/(1+0.1*z) describes the efficiency decrease of track registration at depth z, (z) and T(z) are the track density and temperature [°K] at depth z, respectively. Rearranging (4) we get:

$$\ln(F(z)\frac{\rho(z)}{\rho(0)})=\int_0^z v(z)^{-1}(\lambda-\frac{dv(z)}{dz})dz \quad (5)$$

where all the correction factors are included in F(z). The left hand side of (5) is determined only at discrete points through measurements. Fitting these points with a continuous function f(z) we get:

$$f(z)=\int_0^z v(z)^{-1}(\lambda-\frac{dv(z)}{dz})dz \quad (6)$$

The parameters in f(z) can be calculated from the least squares fit:

$$\Sigma(f(z)-\ln[F(z)\frac{\rho(z)}{\rho(0)}])^2 \longrightarrow \min \quad (7)$$

The general solution of Eq. (6) is:

$$v(z)=u(z)^{-1}(\int_0^z \lambda u(z)dz+v_c) \quad (8)$$

where u(z)=exp(f(z)), and v_c is an integration constant (Korn, 1975). Equation (8) describes the connection between the measured track densities and radon transport velocities.

5. RESULTS AND DISCUSSION

For the period of the observations the shapes of the track density distributions obtained were similar to the curve shown in Fig. 2.; they differed only in the mean by a factor 2-3. The alpha-track densities ranged from 50 to 400 tracks/cm^2*30 days and the corresponding monthly mean radon contents of the well water were in the range from 300 Bq/m^3 to 150 Bq/m^3, which were very low compared to other waters in the area. After performing the transformation (5) it is clearly seen that the radon content gradually decreases with increasing distance from the bottom of the well. The main trend of the measured data can be approximated by a straight line f=k*z, which describes

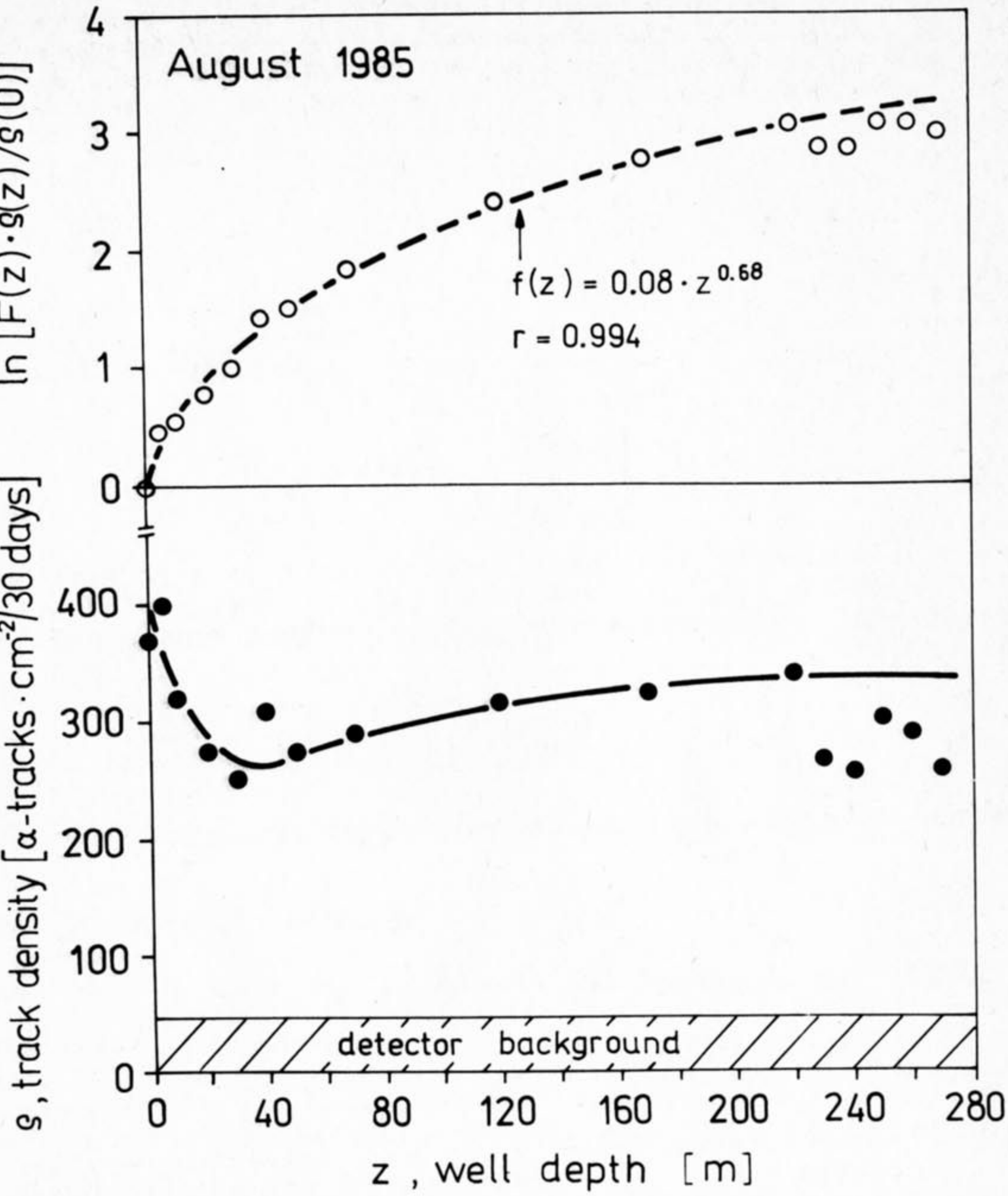

Fig. 2. A typical track density profile and the corresponding transformed curve

one dimensional upward flow of a radioactive gas with a constant velocity $v_{LIN} = \lambda / k$ (Somogyi and co-workers, 1986). The correlation coefficients are in the interval of 0.8-0.9 and the values of v_{LIN} range from 0.58 to 0.83 m/h. The mean of the calculated velocities is $\overline{v}_{LIN} = 0.7$ m/h with a standard deviation of 10%. The good correlation indicates that the approximation is very good; nevertheless it can be improved by fitting the data with a power type function $f = a*z^b$ (see upper part of Fig. 2.). In this case v can be calculated numerically from the equation:

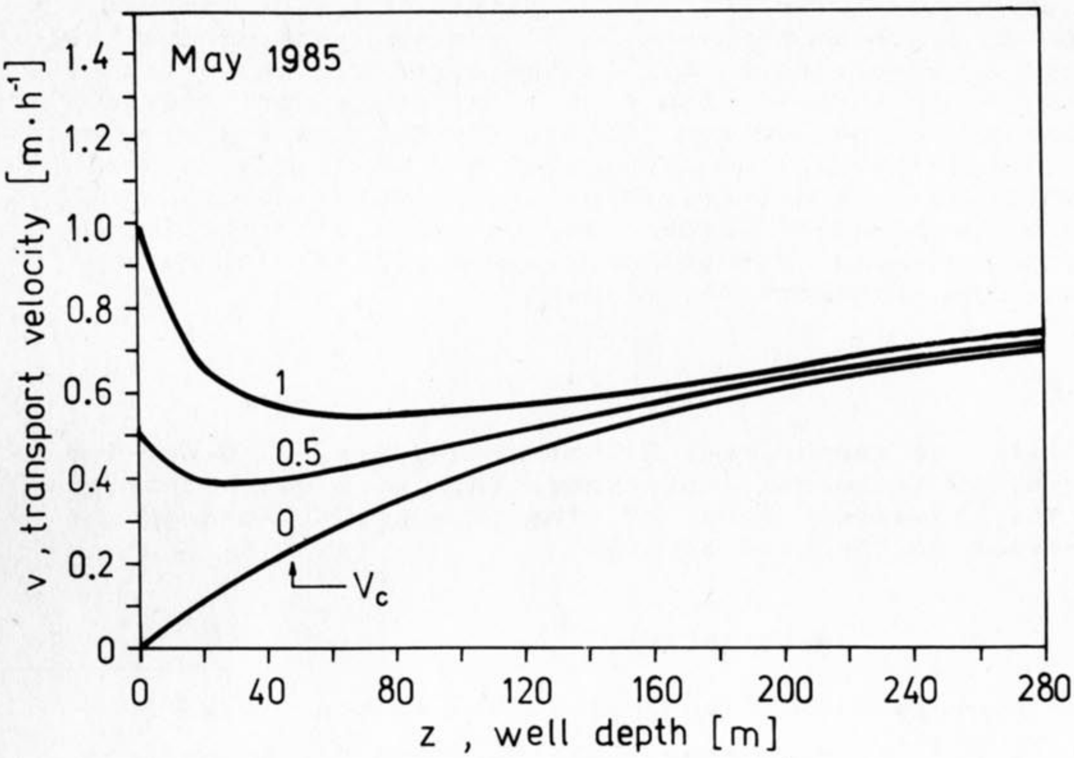

Fig. 3. Upward velocity vs. depth calculated for different v_c-s

$$v(z)=\exp(-a*z^b)\left(\int_0^z \exp(a*z^b)dz+v_c\right)$$

where parameters a and b are determined from Eq.(7). Fig.3. shows the result of such a calculation. The value of v_c can be determined from the boundary condition at the top of the well. In our case it was estimated from the upper three points through constant v approximation. The obtained values of v_c are in the interval 0.05-0.25 m/h.

For the interpretation of the observed high transport velocities, the data were compared to calculations performed according to the microbubble transport theory (Várhegyi and others, 1986). The obtained correlation coefficients are in the interval 0.6-0.7, which are rather poor compared to those obtained above.

On the other hand, in every physical system, where thermal gradients are present, thermal convection takes place, which can induce convectional flows in certain circumstances. From the point of view of hydrodynamic stability such a system is characterized by the product of the Grashof and Prandtl numbers (Landau, 1980):

$$GP = \frac{A*R^4*g*b}{\chi*\nu}$$

where A is the thermal gradient [°C*m⁻¹], R is the radius of the tube [m], g is the gravitational acceleration 9.81 [m*s⁻²], b is the coefficient of thermal expansion [°C⁻¹] and χ and ν are the coefficients of thermal conductivity and kinematic viscosity [m²*s⁻¹], respectively. This number determines the stability of a given physical system against convectional movements. If GP is larger than a critical value, the criteria for the beginning of convection is fulfilled. Knowing all the parameters (Juhász, 1987), we can calculate GP for this well. The values obtained are 2-3 orders of magnitude higher than the critical value. Concluding we can say, that most probably the thermal gradient induced convection plays a significant role in governing the radon transport in this well.

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