APPLICATION OF RADON-222, AS A NATURAL TRACER IN ENVIRONMENTAL STUDIES

Ph.D. thesis

Dr. József Hakl

Lajos Kossuth University
Debrecen, 1997

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Ezen értekezést a KLTE **fizika** doktori program *fizikai módszerek alkalmazása interdiszciplináris kutatásokban* alprogramja keretében készítettem 1995 - 1997 között és ezúton benyújtom a KLTE doktori Ph.D. fokozatának elnyerése céljából.

Debrecen, 1997 szeptember

Dr. Hakl József jelölt

Tanúsítom, hogy Dr. Hakl József doktorjelölt 1995 - 1997 között a fent megnevezett doktori alprogram keretében irányításommal végezte munkáját. Az értekezésben foglaltak a jelölt önálló munkáján alapulnak, az eredményekhez önálló alkotó tevékenységével meghatározóan hozzájárult. Az értekezés elfogadását javaslom.

Debrecen, 1997 szeptember

Dr. Hunyadi Ilona témavezető

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Preface

Preface

It happened in October 1985 that Dr. György Somogyi, the late leader of the Track Detector Group of the Institute of Nuclear Research of the Hungarian Academy of Sciences (Debrecen, Hungary) invited me to accompany him on one of his favourite duties. He took me to a cave (Hajnóczy cave, Bükk Mountains, Hungary) not for recreation but to work, to change the etched track type radon concentration measuring devices. It was when I have got my first impression about the application of a nuclear technique in environmental studies. The underground tour lasted 6 hours. I got stiffness for two days and I promised myself never to repeat this action again.

It did not happen though. The things around caves started to interest me and I readily took part in field-work. In the spring of 1987 Dr. György Somogyi suddenly died of heart attack. Dr. Ilona Hunyadi took the lead of the group and under her leadership I got the chance to continue the work in this field. It has became and still it is my main scientific area. I have enjoyed this work very much.

It was Dr. Ilona Hunyadi, who in the early hard times fought with great enthusiasm and success for financial background of radon works; exposing her young colleagues, in the meantime, to the national and the international scientific community. Here I should like to thank for her selfless help, advice and all she had done for the group.

Also, I should like to thank my closest colleagues Dr. István Csige and Attila Vásárhelyi, with whom I had the chance to work together in the laboratory.

During the field and laboratory works my closest colleagues were Gábor Géczy, Dr. András Várhegyi, Dr. János Somlai, Dr. László Lénárt, Dr. István Törőcsik, Dr. J. L. Seidel, Ferenc Szolga and László Rénes, with whom I spent exciting times discovering the secrets of the nature.

Particularly, I should like to thank to Mrs. Enikő Molnár for the excellent technical and administrative work and for all she has done for us in the Radon laboratory of Debrecen.

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1 Introduction

With increasing pollution world-wide the need for understanding of the dynamics of environmental processes has became of primary concern of mankind. The problems are focused on two, in certain sense opposite areas. These are the industrially heavily contaminated areas and yet clean but endangered regions. In the first area questions regarding the propagation of contaminants, in the second one the transport processes themselves are in the centre of public and scientific attention, respectively.

One of the methods suitable to study these processes, which reflect the interaction of atmosphere, hydrosphere and lithosphere, is the use of tracer isotopes. Subsurface natural fluids in the majority of cases carry small amounts of radioactive isotopes. Among them the alpha radioactive ^{222}Rn, as a member of ^{238}U decay series, is ubiquitously present in the ground and in the lower atmosphere as well. Through the measurement of radon concentration in the geological environment, information can be obtained about the transport processes as well as about penetrated geological structure. This approach is applicable also in cases of radioactive contaminants, where the essential question is the retention of transport.

Among the endangered regions in highly permeable areas, as e.g. karst[1] is *per se*, the problems are perhaps the sharpest, as they serve as natural water reservoirs for mankind. In these areas, owing to the set of interconnected fractures and voids, the propagation of external effects is very quick and deep. One of the most appropriate methods to trace transport with natural radon in these systems is the application of etched track detectors allowing desirable large-scale *in situ* measurements. At the Institute of Nuclear Research of the Hungarian Academy of Sciences first underground environmental alpha radioactivity studies, based on the latter technique, were initiated by Dr. G. Somogyi in 1977. I have entered this step by step widening radon field in 1987. My scientific interest focused on studying radon transport, giving emphasis to cave investigations.

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[1] Areas in which the rock has suffered solutional activity with the development of caves and enlarged fractures

Caves occur everywhere on the earth, although mainly in karst areas - as they are formed mostly in limestone environments. They may appear to men as static environment, where not only the continual darkness but also the temperature and high relative humidity is stable. This belief is, however, not justified, because fluid currents can cause measurable changes in the physical parameters. The underground radon measurements supplied us with a great amount of data, both spatially and temporally variable, confirming the awaited diversity of data. The complexity of environmental transport processes manifests itself everywhere. The influence of inflowing waters, the presence of underground air circulation as well as the morphology and structural dependence of transport processes are traceable in our radon records. Based on these investigations, physical model interpretations of the observed fluid motions were developed in collaboration with the Department of Physical Geography, Eötvös Lóránd University (Budapest, Hungary) and Department of Hydro- and Engineering Geology, Miskolc University (Miskolc, Hungary). The so-called vertical geogas microbubble radon transport model was developed in collaboration with Mecsek Ore Mining Ltd (Pécs, Hungary). Its validity conditions were tested in measurements done in a 270 m deep karst well at Miskolc University and on a 12 m high model column at the Laboratory of Hydrogeology, University of Montpellier (Montpellier, France). These collaborations resulted in developing of a microprocessor controlled automatic radon-measuring unit in Hungary, which, since 1991, is routinely used in fieldwork parallel with track etch technique.

The object of this thesis is to summarise results obtained during the last decade. I have included those findings, in obtaining of which my role was decisive. As a main part, I will outline results related to radon concentration measurements in karst caves and in natural (not necessarily karst) waters. Additionally, I will summarise results of methodological developments connected to the solutions of tasks of environmental radon activity concentration measurements. These are the studies of radon transport through different filter and blocking materials, and the developments of new measuring techniques for underwater and continuous radon concentration measurement purposes.

2 Review of literature

2.1 Sources of radon in karst

Radon is a mobile, chemically inert radioactive element. All the three naturally produced isotopes, ^{222}Rn (radon), ^{220}Rn (thoron), and ^{219}Rn (actinon) decay by emitting alpha particles. These noble gas isotopes are produced from radium decay as steps in lengthy sequences which originate from uranium or thorium series – ^{222}Rn from ^{238}U; ^{220}Rn from ^{232}Th; and ^{219}Rn from ^{235}U. Their respective half lives are 3.82 d, 55.6 s, and 3.96 s (mean lives 5.51 d, 80.2 s, and 5.71s). The relative importance of the three isotopes increases with their mean lives and relative abundance. ^{219}Rn is the shortest lived, and is virtually always produced in much smaller amounts than is ^{222}Rn, since the natural ^{235}U/^{238}U ratio of these ultimate progenitors is 0.00719. Hence ^{219}Rn is largely ignored. ^{220}Rn too is short lived relative to ^{222}Rn and consequently moves a much smaller distance from its source than does ^{222}Rn. In air, for diffusion constant, D, of 0.1 $cm^2 \cdot s^{-1}$, the mean distances of diffusive motion are 2.2 m for ^{222}Rn and 0.029 m for ^{220}Rn. Hence in circumstances where signals from relative distant sources or processes in the earth are sought, ^{222}Rn is by far the dominant nuclide, and ^{220}Rn provides only a local background that one want to exclude during detection. Characteristic feature of radon isotopes is their high mobility in comparing them to other members of the radioactive decay series. They are able in a short time to escape into the pore space from the mineral in which they are born. The radon atom that escapes is either released by direct ejection by recoil from alpha emission (Kigoshi, 1971) or by diffusion through damaged channel after chemical solution of it with pore water (Fleischer and Raabe, 1978). From the pore space radon atoms migrate towards microcracks, fractures and cave volumes either by diffusion or forced flow.

The sources of radon in caves are the bedrock and deposits. Radon levels in caves are determined primarily by the uranium content of the rock. Limestone and other sedimentary rocks are found to contain about 1.3 - 2.5 ppm ^{238}U (16 - 31 $Bq \cdot kg^{-1}$) on average. The relatively high values of radon found in caves are due to these minute quantities of parent substances that occur naturally on and within the interior surfaces of the caves. Increased ^{238}U concentrations can be associated either with fluorite mineralizations or hydrocarbons present in the surrounding limestone.

Uranium can be oxidised and mobilised by groundwater flow. Once reducing conditions are encountered, the uranium is readily precipitated from solution. This leaching fixation process leads to the enrichment of uranium in adjacent deposits. This secondary transport and enrichment process is important in cave environment, as fractures in rock can increase the surface area interacting with water. Experimental evidence of this effect in cave environment was found by Navrátil *et al.* (1993), who observed enrichment of uranium on cave walls.

The immediate radon source in caves is the ^{226}Ra content of the rock. Nazaroff *et al.* (1988) reports mean ^{226}Ra content of carbonates to be 25 Bq·kg^{-1} (range 0.4-233), which were found to be distributed lognormally. The results of ^{226}Ra determinations of bedrock and soil samples from the Hungarian caves examined fall in range 0.6-26 Bq·kg^{-1} (Hunyadi *et al.*, 1997). The relatively impermeable soils (deposits), such as clay, do not have sufficient porosity to allow transfer of significant amounts of soil gas, therefore their contribution to radon budget is small (Michel, 1987). Accordingly, Burkett (1993) found radon emitted from the clay to be not sufficient to account for the radon concentrations measured in the cave.

2.2 Radon as a tracer

Subsurface natural fluids in the majority of cases carry small amounts of environmental isotopes. The behaviour of these elements, and the variation of their concentration in time and space, is the result of physical, chemical and biological interactions. These elements, as their physical properties, concentrations, etc., provide information on flow and kinetics of the carrying substances, are called natural tracers.

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 priory*. Therefore, the interpretation of the concentration data is not straightforward: it needs interdisciplinary expertise of hydrogeologists, geologists, physicist, radiogeochemists, etc. Joint efforts have given results in different fields. The observations of subsurface fluid motions traced by natural radon were followed 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. (Fleischer, 1988).

In the speleology, similarly to the previously mentioned fields, these types of measurements have already found their applications, and they give important contributions to the better understanding of the natural regimes of caves. Cunningham and LaRock (1991) delineated six microclimate 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.

The most common and most apparent phenomenon, which was discovered in the majority of the investigated caves throughout the world, was the annual change of radon activity concentration. Wilkening and Watkins (1976) identified temperature gradients favourable 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 $\Delta T/f$, where ΔT is the temperature difference between the cave and outside and f is a friction factor characterising the flow resistance (Wilkening and Watkins, 1976; Atkinson *et al.*, 1983; Quinn, 1988).

2.3 Modelling of Observed Features

The underground radon transport can be described by the following transport equation:

$$\frac{\partial C}{\partial t} = D_{eff} \Delta C - \nabla(\vec{v}C) - \lambda C + \phi,$$

where C $[m^{-3}]$ is the radon concentration in pore space, D_{eff} $[m^2 \cdot s^{-1}]$ is the effective diffusion coefficient of radon, $\vec{v}$ $[m \cdot s^{-1}]$ is the velocity of the carrying substance, λ $[s^{-1}]$ is the decay constant of radon and ϕ $[m^{-3} \cdot s^{-1}]$ is the source term. In the equation first term describes diffusion, second term advection, third term decay and fourth term sources of radon. For the solution of transport equation, first, it is necessary to know the velocity field (e.g. Navier-Stokes equation, which by itself is a sufficiently complicated problem); then taking into account the source term, and the initial and boundary conditions C can be determined. The emerging phenomena are determined by the form, shape and structure of the underground void space. The above equation generally can be solved only numerically.

The realisation of radon transport processes sharply depends on the configuration of the interconnected underground cavities. In the case of blind end systems, atmospheric pressure variation are the main control parameter (Wigley, 1967; Ahlstrand, 1980), 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 (Wilkening, 1979). 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.

The interpretation of the data can be affected by the presence of these unknown (unassumed) ‘entrances’. Yarborough (1980), in a study of nine caves, identified two general types of physical cave configurations that affect airflow patterns and radon concentrations. Type 1 caves have most passages at or above the entrance elevation, Type 2 caves have most passages below the entrance elevation. When the outside temperature exceeded the cave temperature, he found that Type 1 caves exhaled, while stagnation occurred in Type 2 caves; when the outside temperature fell below the cave temperature, both caves inhaled. As Type 1 cave is horizontal "mirroring" of Type 2 cave the seasonal ventilation patterns should be alternatives of each other. The clear asymmetry in this case, however, points to the difference between the air flow-through and blind end system. This indicates, that substantial hidden parts of the upward directed systems remained unrevealed from the descriptive point of view.

The above examples illustrate the problems of interpretation and modelling. In two special cases, however, the emerging processes are analytically easy to survey. These are the cases of idealised two-entrance horizontal flow-through (type A), and one narrow entrance vertical caves (type B). In the case of the horizontal model cave, the second entrance may represent the set of vertical fractures, which connect the main passage to the surface through the overburden.

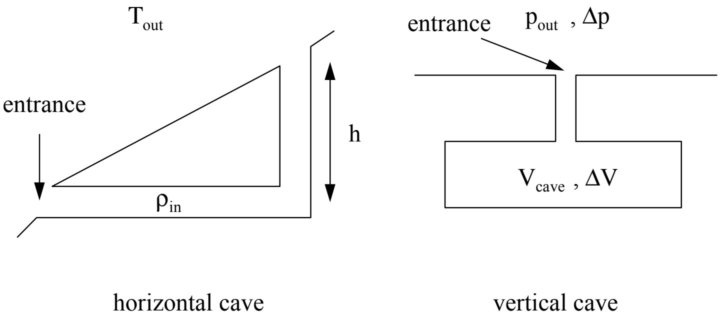

Figure 1: Schematic horizontal (type A) and vertical (type B) model caves; h is the elevation difference between the upper and lower entrances (horizontal cave), ρ_{in} is the air density of inside air, T_{out} is the outside temperature, p_{out} is the ambient outside pressure, V_{cave} is the cave volume and ΔV is the volume of air passing the vertical cave entrance in case of Δp change in the ambient outside pressure.

First, let us consider a schematic horizontal cave (see Fig. 1, left). In this case, owing to the temperature dependence of air density the pressure exerted at the lower end of the cave by the outside air will differ from the inside one. This pressure difference in this case is (Atkinson *et al.*, 1983):

$$\Delta p \approx -gh\rho_{in}\frac{\Delta T}{T_{out}},$$

where ρ_{in} is the density of inside air, g is the gravitational acceleration, h is the height difference between the two entrances, ΔT is the temperature difference between the cave and the outside, and T_{out} is the outside temperature. According to this relation, the direction of airflow through the entrance depends on the season; in warm season air flows out of, and in cold season air flows into, the cave at the lower entrance.

A more realistic model of fracturized karstic overburden above a horizontal cave is the model of parallel vertical voids connecting the main passage to the surface (Scheidegger and Liao, 1972). The variation of ^{222}Rn activity concentrations in this case is interpreted by an air circulation model (Géczy *et al.*, 1988). According to it, in warm season, air will flow into the cave from the direction of the radon-rich fracture system (and will flow out of the cave through the entrance at lower altitude) resulting in high radon levels in cave. In cold season, the direction of the flow is reversed. From the direction of the lower entrance fresh outside air flows into the cave forming considerably lower radon concentration levels than in summer

In the case of narrow-entrance vertical caves, the most effective processes in inducing air motions are the atmospheric pressure changes (Fig. 1, right). Falling ambient atmospheric pressure drains air from the cave; increasing atmospheric pressure presses air into the cave through the entrance. The volume of air passing the cave entrance:

$$\Delta V \approx -\frac{V_{cave}}{p_{out}} \Delta p \,,$$

where V_{cave} is the cave volume, p_{out} and Δp are the ambient atmospheric pressure and the change in the ambient atmospheric pressure, respectively. Airflows induced by atmospheric pressure changes can be rather quick. At the entrances of giant caves their speed can reach several tens of km·h^{-1}(Cunningham and LaRock, 1991).

3 Materials and methods

For the purpose of environmental radon concentration measurements opened diffusion chambers equipped with LR-115 type II alpha sensitive polymer track detector were used. The diffusion chamber consisted of a cup (diameter: 5.5 cm, height 12 cm) with a detector located at the bottom of the cup. The other end of the diffusion cup was opened to the atmosphere and looked downwards during the exposure. The discrimination against thoron (^{220}Rn) was obtained by the delay effect of the diffusion on the basis of the use of a sufficiently long cup. The sensitivity of the detector is 2.3 alpha-tracks$\cdot$cm^{-2}/kBq$\cdot$m$^{-3}\cdot$h at standard etching conditions (2.5 N NaOH, 60 °C, 2.5 hours). Generally, in each cave the track etch detectors were placed along the main passages of caves more or less equidistantly. There were generally 3-20 regular measuring sites per cave. Extra observation points were established at “characteristic” places. Typical exposure time was 1 month. About 10000 observation data were obtained in 31 investigated Hungarian caves during years 1977-1997.

From the beginning of the 1990s, microprocessor controlled automatic field radon monitors (type Dataqua[#]) were gradually introduced into cave studies. The single channel type measured only radon concentration, while the multiparameter version simultaneously registrated the temperature and air pressure. Radon concentration was measured in 1 hour cycles using open type diffusion chamber (delay time ≈ 1000 s) equipped with alpha sensitive Si based semiconductor detectors. The sensitivities of the units are 6.7 cph/kBq$\cdot$m$^{-3}\cdot$h (detector sensitive area: 1 cm^2) and 17.8 cph/kBq$\cdot$m$^{-3}\cdot$h (detector sensitive area: 3 cm^2) with an 0.1 cph initial background (cph stands for count per hour). Generally, in each cave the continuous radon monitors were placed in the main passages at characteristic places. There were 1-5 regular measuring sites per cave. A total number of approximately 500000 hourly readings was obtained with 11 monitors in the investigated five caves during the years of 1991-96.

Measuring sites

The most characteristic results were obtained in the following caves from the 32 investigated ones:

[#] Produced by Dataqua Ltd., Kölcsey F. u. 1, 8220 Balatonalmádi, Hungary

Bükk Mountains, Bükk National Park, Hungary

- The *Létrási-Vizes cave* is a multi-level typical swallet cave. The main passage declines towards the end of the cave, which is located at 85 m depth from the natural entrance. The cave collects waters from the surrounding area, but its streamlets often go dry. The cave can be considered as cave of type B.
- The *Hajnóczy cave* can be considered as cave of type A. Its passages are located along 3 parallel fault zones. The distance between two farthest points of the cave is 150 m.
- The *Szepessy cave* is a cave of type B. The depth of the entrance shaft is 130 m, the maximum depth of the cave is 165 m.
- The *Istvánlápa cave* is a cave of type B. It is one of the deepest caves in Hungary. The depth of the entrance shaft is 210 m, the maximum depth of the cave is 240 m.

Aggtelek Karst, Aggtelek National Park, Hungary

- The *Baradla cave* is nearly a horizontal cave located between Aggtelek and Jósvafő villages. The cave has several entrances located along and at the ends of the main passage, which length is 6 km. The main passage declines towards the Jósvafő entrance. The elevation difference between the entrances at Aggtelek and Jósvafő is 60 m.
- The *Vass Imre cave* is a cave very similar to the model cave of type A. It has one entrance and the main passage is situated horizontally. The length of the cave is 600 m. In the middle part of the cave there is a siphon, which was closed by water two times during the overall observation period.

Pilis Mountains, Hungary

- The *Sátorkő-puszta cave* is a vertical cave of type B. The depth of the cave is 48 m.

Mecsek Mountains, Hungary

- The *Abaliget cave* is very similar to the model cave of type A. It has one entrance and the main passage is situated practically horizontally. The length of the main passage is 500 m.

Buda Mountains, Hungary

- The *Szemlő-hegy* cave is a horizontal cave with the main entrance situated at lower altitude, and several other 'entrances' located at higher altitudes of the hill. The cave passageways are situated in two levels but the cave can be considered as cave of type A with one dominating large opening to the surface. The length of the lower passage is 350 m. The end of the main passage is 65 m depth from the surface, the vertical distance between two levels is 15 m.

Keszthely Mountains, Hungary

- The *Cserszegtomaj well cave* is a horizontal maze cave of type B, which was formed in 50 m depth on the boundary of dolomite and sandstone. It has one 50 m deep artificial vertical entrance. The whole formation is covered by clay. The radon concentration is unusually high due to the sandstone environment of high porosity and bad ventilation.

Lamalou Karst, France

- The *Lamalou cave* is a horizontal water cave with a flowing stream inside the cave. A part of the cave, except floods, is aerated. The aerated part of the cave has one vertical and one horizontal entrance. The aerated part of the cave system can be considered as cave of type A. The length of the vertical entrance shaft is 20 m, the length of the horizontal part of the aerated passage is 90 m.

4 Summary of results

(Papers referred as [A..] containing the new scientific results can be found in the attachment.)

1. Characterisation of airborne ^{222}Rn concentration occurrences in karst caves

1.a Compiling all the globally available radon concentration data from 220 different caves world-wide I have found, that the distribution of ^{222}Rn concentrations is lognormal [A1, A2] (GM=1130 Bq·m^{-3}, GSD=6.3), which is in good accordance with the awaited distribution for a geochemical element. The lower end of the scale is associated either with big cave chamber volumes or high ventilation rates; the upper end is characterised by closed, badly ventilated places and uranium-rich sediments.

1.b For the 31 examined Hungarian caves I have determined the cave average annual mean radon activity concentrations, 0.3-20 kBq·m^{-3}*, the characteristic annual maximum/minimum ratios, 2-50, and the periodicity, which was typically one or half a year* [A3]. In the majority of cases the marked seasonal variations can be characterised with high radon concentration values in summer and low radon concentration values in winter. In few cases, reversibly, summer minima and winter maxima were observed.

1.c I have observed a long-term variation of the annual mean radon activity concentration in all the studied caves [A1, A4]. The phenomenon shows the effect of slowly changing environmental parameters on radon transport processes. Such an environmental parameter may be the annual precipitation. The variation of annual precipitation, which may due to climatic changes, influences water content of the cave embedding rocks, which on the other hand affects radon emanation power of rocks. The long-term variation can be amplified selectively at different sites in a given cave. This latter effect is markedly shown on time series taken at two different depths of the Sátorkő-puszta cave. The ratio of the difference of data pairs to the mean of the same data pairs increases with years (see Fig. 4. in [A1]).

2. Influence of karstification and morphology of caves on airborne ^{222}Rn concentrations

2.a By model calculations I have shown, that the saturation value of radon activity concentration in fractures and the radon exhalation from fractures strongly depend on the size of the fracture. According to calculations, at low airflow velocity to aperture size ratios, the advective ^{222}Rn transport along a fracture is strongly reduced by lateral diffusion inside the fracture [A1, A6]. These theoretical predictions were justified by measurements done in the Vass Imre and Lamalou caves. In the Vass Imre cave, which can be characterised by relatively undeveloped fractures with small size openings, I found no daily variation in the radon records [A1], in spite of the observed daily air flow variation. On the other hand, in the Lamalou cave, which is embedded in a well karstified strata, characterised by big solutional openings and fracturized volumes, strong daily fluctuations of ^{222}Rn activity concentrations were recorded [A7].

2.b I have found that in horizontal caves the number of vertical fracture systems communicating the surface affects the width of the outside temperature interval, which characterises the transition from the low to the high daily average radon concentration values [8]. While in the Vass Imre cave the transition width is around 5 °C, it is around 10 °C in the Abaliget cave. In the Vass Imre cave, only one less developed fracture system, in the Abaliget cave, more than one more-developed vertical fracture systems exist. The step by step widening of the transition interval as a function of the number and size of openings, is further supported by the data obtained in the Szemlő-hegy cave. Here the step function type curve is practically reshaped to a linear function (see Fig. 3. in [A8]).

3. Identification of air circulation pathways and microclimate zones in cave systems based on airborne ^{222}Rn concentration measurements

3.a I have pointed out, that in a consequence of the seasonally directed underground transport, enhanced surface exhalation can be expected on karstic terrains seasonally [A3]. Experimentally the existence of the latter phenomenon was verified in the Bükk Mountains, Hungary, where the radon time series measured inside the Hajnóczy cave and in a slit above the Hajnóczy cave were inversely correlated (see Fig. 6. in [A2]) showing high winter radon concentration values on the surface. Winter maxima were as well observed on several karstic terrains of Hungary [A3]. The above phenomenon can serve as an explanation to other observations published in the literature.

3.b I have identified microclimate zones based on the analysis of temporal variations of radon concentrations in caves. In the Baradla cave these zones are [A5]: 1. the entrance region in the Aggtelek part with summer maxima and winter minima; 2. the middle part from Libanon hill to Vöröstó entrance with constant radon levels; and 3. the region between the entrances of Vöröstó and Jósvafő with winter maxima and summer minima. I have pointed out, that the temporal behaviour of radon activity concentration in the third microclimate zone of the Baradla cave refer to a definite a connection between the main passage of the Baradla cave and the Rövid-Alsó cave situated at a lower altitude. Based on the difference in temporal variation of radon concentrations three microclimate zones were as well identified in the Létrási-Vizes cave [A2, A9].

4. Determination of underground airflow velocity and its relation to cave average annual mean airborne ^{222}Rn level

4.a I have determined the propagation speed of outside fresh air intrusions underground and a volume of a vertical cave from the continuous measurements of radon concentration [A1, A2]. In horizontal caves fresh air intrusion appears when the outside temperature falls below the cave air temperature, while in vertical caves their presence is related to an increase of the ambient outside pressure. Utilising the time differences among radon falls measured at distinct places, their propagation speed can be calculated. This speed is about 50 $m \cdot h^{-1}$ along the main cave passage of the horizontal Vass Imre cave. As the length of the cave is 600 m, the whole known volume of the cave is flushed through with outside air in 12 hours. Contrary, the propagation speed of the dilution effect in the vertical Cserszegtomaj well cave is around only 2 $m \cdot h^{-1}$ in a 10-20 m region from the bottom of the entrance well. This low value shows that the cave is highly unventilated. In average 2 hPa of atmospheric pressure increase flushes out radon gas from the complete volume of the vertical entrance. Taking into account the known volume of the entrance well and the ideal gas law, it indicates, that the volume of the cave is in the order of 10000 m^3, an order of magnitude larger, than it was estimated from the volumes of known passages.

4.b I have found that cave average annual mean airborne ^{222}Rn levels depend on the strength of underground air motions [A2, A8]. In the Vass Imre cave the volume of infiltrated air is naturally controlled by the penetrability of the siphon. When the siphon is open, there is continuous air flow either in or out from the cave through the entrance (winter and summer, respectively). When the siphon is closed, there is no measurable air flow through the entrance. The restriction of the air flows resulted in an overall drop of 30% in the annual mean radon concentration level in the cave, accompanied by the decrease of the amplitude of the seasonal radon concentration variation. This effect can be explained on the basis of the air circulation through the cave covering strata. It shows the increase of mean radon levels due to periodically changing flowing conditions. The absence of seasonal variation in radon levels and the low radon activity concentration values found in the deepest parts of the narrowest vertical caves in Hungary (Szepessy, Istvánlápa) may also be attributed to this phenomenon [A3], as from the point of view of ventilation the deep caves can be considered as the most closed ones.

5. ^{222}Rn transport in water

5.a I have found that water inlets can be significant sources of radon in cave. In deeper parts of the Létrási-Vizes cave the regular seasonal variation of radon levels are considerably disturbed [A1, A2, A4, A9]. Approaching the endpoint of the cave, the periodical character of the readings decreases and the variation of springtime values becomes higher. On the other hand, there is high similarity between temporal variations of the yield of one of the intermittent streams and radon concentration measured in the air. As subsurface waters permeating porous rocks can be significantly enriched in solved radon, this effect is explained on the way that radon is essentially carried to the place by the latter intermittent stream.

5.b Using radon as a tracer I have found that thermal gradient induced convectional mixing is taking place in the water column of a 270 m deep karst well [A4, A10]. I have developed a model to calculate the depth dependence of the vertical transport velocity from the measured vertical ^{222}Rn concentration profiles. The results indicated continuous upward transport of radon in the water column with a mean velocity of about 0.7 $m \cdot h^{-1}$. I have pointed out, that this high value can be attributed to the vertical thermal gradient induced convectional mixing of water, and I rejected the possibility of transport by geogas microbubble theory. The strong effect of vertical mixing on transporting radon was observed as well in model experiments at the Hydrogeological Department, University of Montpellier, France, in an 8.5 m high model column [A11]. As the search for new U ore deposits in Hungary in late 80s was partly based on the interpretation of the vertical radon profiles taken underwater in shallow drills, the recognition of underwater vertical mixing processes along bore holes in this respect played a supplementary role.

6. Methodological developments

6.a Silicon photodiode based Dataqua radon monitoring devices were developed and brought to successful applications with my significant contribution [A12]. The idea of utilising silicon detectors for field radon measurements (by alpha particle detection) came from Dr. M. Monnin, Hydrogeological Department, University of Montpellier, Montpellier, France. The hardware of the device (analog and digital electronics) was developed by the Dataqua Electronic Ltd. The design of the sensitive volume of the radon head of the instrument and the electronic testing were done by me at the Institute of Nuclear Research. I have calibrated the instrument in the radon reference chamber of the Swedish National Institute of Radiation Protection (Stockholm). The obtained calibration coefficient, 6.7 cph/kBq$\cdot$m$^{-3}\cdot$h for the 1 cm^2 surface detector, coincides well with that I have previously calculated.

6.b A novel method to measure radon permeability of thin foils using etched track technique was developed with my significant contribution [A13]. During the measurement the sample foil separates the radon source volume from the measuring volume. I have elaborated an analytical model to describe the non steady state temporal variation of radon activity concentrations in source and in the measuring volumes as a function of the permeation characteristic of the investigated foil. This method allows to determine radon permeability coefficient as well as radon/thoron separation factor of a given foil in few hours. I have tested the reliability of the method in a series of laboratory measurements using polyethylene foils of different thickness. The obtained permeability value, $7.1*10^{-8}$ cm$^2\cdot$s^{-1}, is in good agreement with the literature data. The permeation characteristics of different materials are of interest from the point of view of sealing homes against invasion by radon. As a practical application of the method we have examined six types of floor covering materials, and found, that they can reduce radon entry into dwellings by 2-3 orders of magnitude.

6.c Two experimental techniques for the determination of the effective diffusion coefficients of radon in polymer/silicate gels and clay suspensions were developed with my significant contribution [A16, A17]. Similarly to the previous method, one side of the samples was exposed to the radon source, the other side was closed by a small measuring volume. Track etch type radon monitors were used to measure the radon exposures on both sides of the samples. The diffusion mass transport in the sample was numerically modelled for different exposure times and sample thicknesses. Diffusion constant was a fitting parameter to obtain best fit to the experimentally measured radon exposures. I used Dataqua type continuos radon monitors to test the experimental arrangement against leaks. The procedure was based on testing of the variation of the diffusion constant in subintervals of the full exposure time. The variation in the value of the diffusion constant was the indicator of leakage. We have determined the effective diffusion coefficient of radon in polymer/silicate gel-containing porous media, $3.3*10^{-6}$ $cm^2 \cdot s^{-1}$, and in Montax/clay suspension, $6.0*10^{-6}$ $cm^2 \cdot s^{-1}$. The corresponding values for the bulk phases are comparable to that characteristic in pure aqueous solutions, therefore the sealing technology applying these materials can be very attractive.

6.d A new method for the determination of the radium and dissolved radon content of water samples using track etch type radon monitors was developed with my significant contribution [A14]. During the measurement an immersed small volume radon monitor is sealed together with the water sample into a container. The air filled radon monitor is protected from the water by a thin radon permeable rubber foil. The dissolved radon and the radium content of water samples is determined by two subsequent exposures. In the first exposure the sum of dissolved radon and radium concentrations, in the second exposure the radium concentration itself are measured. I have developed a model, which describes the temporal variation of the radon concentration in the measuring volume of the small radon monitor. For the given arrangement I have determined experimentally the calibration coefficient, 24.1 $tracks \cdot cm^{-2}/(d \cdot kBq \cdot m^{-3})$ [A15], which value coincided well with the calculated one. Characteristic for the method is that it not applies any type of separation procedures, the use of which is often difficult in field circumstances.

5 Tézisek

(Az új tudományos eredményeket a [ ]-ben megadott hivatkozások tartalmazzák.)

1. A karsztbarlangok légterében mért radonkoncentráció értékek jellemzése

1.a Áttekintettem az elmúlt két évtizedben publikált barlangi radonmérési programok eredményeit s megállapítottam, hogy a világ 220 barlangjában mért radonkocentráció adatok eloszlása lognormális [A1, A2] (GM=1130 Bq·m^{-3}, GSD=6,3). Az eloszlás típusa megegyezik más geokémiai elemek szokásos földi eloszlásával. A skála alsó részét nagy térfogatú termek ill. erősen szellőző helyek, míg a felső részt zárt, rosszul szellőző helyek és uránium gazdag mineralizációk jellemzik.

1.b Nyomdetektoros mérések alapján megállapítottam, hogy a 31 vizsgált magyarországi barlangban az éves átlagos radon aktivitáskoncentráció a 0,3-20 kBq·m^{-3} tartományba esik, a mért radon idősorok periodicitása tipikusan egy vagy fél év. Jellemző az éven belüli 2-50 radonkoncentráció maximum/minimum arány [A3]. Az barlangok többségében nyári maximumot és téli minimumot tapasztaltam, ritkábban fordítva.

1.c Minden egyes vizsgált barlangban a mért évi átlagos radon aktivitáskoncentráció hosszú idejű változását tapasztaltam [A1, A4]. Ez a hosszúidejű trend eltérő lehet egy adott barlang különböző pontjain. Az utóbbi jelenséget jól mutatja a Sátorkő-puszta-barlang két különböző mélységében felvett radon idősor (lásd 4. ábra [A1]). A két adatsornak az adatpárok átlagára normált különbsége időben növekvő tendenciát mutat. A megfigyelt jelenség tükrözi a lassan változó környezeti paraméterek hatását a radontranszport folyamatokra. Ilyen paraméter lehet a kőzetek radonkibocsátási tényezőjének a lassú változása a barlangot befoglaló kőzet víztartalmának változása miatt. Ez utóbbi paraméter értéke a klimatikus változások miatti éves csapadékmennyiség változását tükrözheti.

2. A karsztosodás fokának és a barlang morfológiájának hatása a barlangi levegő radonkoncentrációjára

2.a Modellszámításokkal kimutattam, hogy a repedésekben kialakuló telítési radonszintek és a radon exhalációja a repedésekből erősen függ a repedés méretétől. Kis repedésméret/légáramlási sebesség arányoknál, a repedés menti advektív radontranszportot erősen csökkenti a keresztirányú diffúzió hatása [A1, A6] A számítások következtetéseit a Vass Imre- és a Lamalou-barlangokban folytatott radonkoncentráció mérések eredményeivel igazoltam. A Vass Imre-barlang egy gyengén fejlett repedésrendszerrel jellemezhető, a Lamalou karszt, Franciaország, pedig egy erősen karsztosodott terület nagy oldási üregekkel és töredezett rétegekkel. A Vass Imre barlangban felvett radon idősorokra a nagyfokú napi stabilitás a jellemző a megfigyelt erős napi légáramlás változások ellenére. A jelenség a diffúzió erős simító hatását mutatja. Ezzel szemben a Lamalou barlangban, a felszíni hőmérséklet napi változásainak megfelelően, erős napi radonszint változásokat találtunk [A7].

2.b Vízszintes barlangokban a napi átlagos radonkoncentráció ugrásszerűen változik a napi átlagos felszíni hőmérséklet függvényében, egy átmeneti tartománnyal az alacsony téli és magas nyári értékek között. Mérésekkel rámutattam, hogy az átmeneti tartomány szélessége függ a felszínnel kapcsolatban álló függőleges repedésrendszerek számától [A8]. A Vass Imre-barlangban ennek az átmeneti tartománynak a szélessége 5 °C, az Abaligeti-barlangban pedig 10 °C. A Vass Imre-barlangban csak egy, a barlang vége felé elhelyezkedő repedésrendszer található. Ezzel szemben az Abaligeti-barlangban több mint egy, jobban fejlett függőleges töréstzóna található. Az átmeneti tartomány szélesedését a repedések számának és méretének függvényében a Szemlőhegyi-barlangban mért adatok is alátámasztják. Itt a megfigyelt görbe gyakorlatilag lineáris (lásd 3. ábra [A8]).

3. Légáramlási utak és mikroklimatikus zónák azonosítása barlangrendszerekben radonkoncentráció mérések alapján

3.a Rámutattam, hogy az évszakosan váltakozó irányú légáramlás következménye a karsztos felszíneken az évszakosan változó radonexhaláció [A3]. Erre a jelenségre a legmeggyőzőbb kísérleti bizonyítékot a Hajnóczy-barlangban és a barlang feletti felszínen egy repedésben mért radonszintek ellenütemű változása szolgáltatta (lásd 6. ábra [A2]). A felszínen magas téli radonkoncentrációt mértünk, de téli maximumokat találtunk Magyarország számos más karsztos területén is. A fenti jelenség magyarázatul szolgálhat az irodalomban publikált további megfigyelésekre is.

3.b Mikroklimatikus zónák jelenlétét mutattam ki a radon idősorok analízise alapján. A Baradla-barlangban ezek a zónák [A5]: 1. az aggteleki bejárati szakasz magas nyári és alacsony téli radonkoncentráció értékekkel; 2. A Libanon-hegy és a vöröstói bejárat közti rész időben stabil radonszintekkel; valamint a 3. jósvafői és vöröstói bejáratok közötti rész magas téli és alacsony nyári radonszintekkel. A radonkoncentráció mérések alapján rámutattam, hogy légkörzés szempontjából kapcsolat van a Baradla barlang és az alatta elhelyezkedő Rövid-Alsó-barlang között. Mikroklimatikus zónák jelenlétét a tavaszi radonkoncentráció értékek szórásának különbözősége alapján sikerült kimutatnom a Létrási-Vizes-barlangban is [A2, A9].

4. A felszín alatti légáramlási sebesség meghatározása és kapcsolata az éves átlagos radonkoncentrációval

4.a Meghatároztam a friss külszíni levegő barlangba való behatolásának terjedési sebességét és egy vertikális barlang térfogatát a folyamatos radonkoncentráció mérések alapján. Ez a jelenség vízszintes barlangokban a külső hőmérsékletnek a barlangi hőmérséklet alá való csökkenésekor, függőleges barlangokban az atmoszférikus légnyomás meredek növekedésekor tapasztalható. A detektorok közötti távolságot és a radonszint esések közötti időeltolódását figyelembe véve a légáramlás sebessége a Vass Imre barlang főjárata mentén kb. 50 $m \cdot h^{-1}$ [A1, A2]. Mivel a járat hossza 600 m, ezért külszíni hatások 12 óra alatt érik el a barlang végpontját. Ezzel szemben a Cserszegtomaji-kútbarlangban a külszíni hatások terjedési sebessége a kút aljától 10-20 m-re csak kb. 2 $m \cdot h^{-1}$. Ez az alacsony érték mutatja, hogy a barlang szellőzése rossz. A mérések szerint átlagosan 2 hPa légnyomás emelkedés hatására terjed ki a friss levegő radonkoncentráció hígító hatása a bejárati kút teljes térfogatára. Az ideális gáztörvény figyelembevételével a barlang térfogata eszerint kb. 10000 m^3, ami egy nagyságrenddel nagyobb, mint az ismert barlangi járatok térfogatának becsléséből számított érték.

4.b Kimutattam, hogy kapcsolatot van a felszín alatti évi átlagos radonkoncentráció és felszín alatti légáramlások erőssége között [A2, A8]. A Vass Imre-barlangon átáramló levegő mennyiségét a barlangban található szifon természetes módon szabályozza. Ha a szifon zárva van, nincs mérhető légáramlás a barlangban. A légáramlás természetes korlátozása éves szinten mintegy 30%-os átlagos radonkoncentráció csökkenést okoz, egyben csökkentve az évszakos radonkoncentráció változások nagyságát. Ez az jelenség a fedő rétegeken keresztüli légkörzéssel magyarázható s a periodikusan változó áramlási feltételek átlagos radonkoncentrációt növelő hatását mutatja. A legmélyebb magyarországi függőleges barlangok (Szepessy, Istvánlápa) mélyén talált alacsony radonkoncentráció szintek és a radonkoncentráció szezonális változásainak a hiánya [A3] szintén a fenti jelenségnek tulajdonítható, mivel szellőzés szempontjából a mély barlangok általában a legzártabbak közé tartoznak.

5. Radontranszport vizekben

5.a Kimutattam, hogy a szellőzési folyamatokon túl, esetenként, a barlangi vízfolyások is befolyásolják a barlangi levegő radonszintjét [A1, A2, A4, A9]. A Létrási-Vizes-barlang mélyebb részein a radonszintek évszakos periodikus változása jelentősen módosul, a barlang végpontja felé a periodikus jellege csökkenése mellett nő a tavaszi értékek szórása. Ezzel szemben nagy hasonlóság van a barlang végpontján fakadó egyik időszakos forrás vízhozama és a levegőben mérhető radonszint időbeli változásai között. Mivel a felszín alatti vizek oldott radontartalma jelentősen nőhet a porózus kőzeteken való áthatolásuk során, így a hasonlóság azzal magyarázható, hogy a radont ez az időszakos patak szállítja a helyszínre.

5.b Egy 270 m mély fúrt karsztkútban folytatott radonkoncentráció profil mérése alapján keveredési folyamatokat azonosítottam a függőleges vízoszlopban [A4, A10]. Az adatok értelmezéséhez kifejlesztettem egy eljárást, amely kapcsolatot teremt a mért radonkoncentráció profil és a mélységfüggő vertikális transzportsebesség között. Az eredmény a radonnak a függőleges vízoszlop menti 0,7 $m \cdot h^{-1}$ átlagos sebességű felfelé irányuló mozgását jelzi. Megállapítottam, hogy a megfigyelt nagy vertikális sebesség a vertikális termikus gradiensek indukálta konvekciónak tulajdonítható, a geogáz mikrobuborékok által gyorsított radon feláramlás lehetőségét elvetettem. A vertikális keveredés erős hatását egy 8,5 m magas modelltornyon (Montpellieri Egyetem, Franciaország) lefolytatott kísérletsorozat is igazolta [A11]. Mivel a 80-as években Magyarországon az urán kutatás részben a sekélyfúrásokban felvett víz alatti radonkoncentráció profilok értelmezésén alapult, az utóbbi eredmény felhasználást nyert a módszer ipari alkalmazásában.

6. Módszerfejlesztések

6.a Jelentősen hozzájárultam a szilícium fotodiódán alapuló Dataqua radonmérő eszköz kifejlesztéséhez [A12]. A fotodióda alkalmazhatóságának ötlete terepi radonkoncentráció mérésekre (alfa részecskék detektálása útján) Dr. M. Monnin-tól (Montpellieri Egyetem, Hidrogeológiai Tanszék, Montpellier, Franciaország) származik. A műszer hardwerét és szoftverét a Dataqua Elektronikai Kft. fejlesztette ki. A mérőfej érzékeny térfogatának tervezését modellszámításokkal, az elkészült mérőfejet a mérőtérfogat és az elektronikus beállítások változtatásával kísérletileg teszteltem. A Svéd Nemzeti Sugárvédelmi Intézet stockholmi referencia radonkamrában folytatott méréseim alapján meghatároztam a műszer kalibrációs állandóját, 6,7 $\mathrm{cph/kBq \cdot m^{-3} \cdot h}$, mely jó egyezésben volt a számításaimmal. A Dataqua márkanevű műszer prototípusát terepi körülmények között a Mecsekurán kft. (Pécs) munkatársa, Dr. Várhegyi András tesztelte.

6.b. Jelentősen hozzájárultam a radon vékony fóliákon belüli diffúziós állandójának nyomdetektor technikán alapuló mérését szolgáló új eljárás kifejlesztéséhez [A13]. A mérés során a vizsgálandó fóliát egy radonforrás és egy mérőkamra közé helyezzük. Kidolgoztam a módszer hátterét képező modellt, amely leírja a radonkoncentráció időbeli változását a mérőkamrában a mérendő diffúziós állandó függvényében. Az eljárás lehetővé teszi a vizsgálati idők csökkentését néhány órára. A módszer alkalmazhatóságát különböző vastagságú polietilén fóliákkal laboratóriumi körülmények között teszteltem. A kapott érték, $7{,}1 \cdot 10^{-8}\ \mathrm{cm^2 \cdot s^{-1}}$, jól egyezik az irodalomban publikáltakkal. A különböző anyagok radonáteresztő képessége a radont a lakásokból kizáró szigetelő anyagok iránti igény miatt érdekes. A módszer gyakorlati alkalmazásaként megvizsgáltuk néhány padlóburkoló műanyag radonáteresztő képességét. Úgy találtuk, hogy mindegyik anyag 2-3 nagyságrenddel csökkenheti a talajeredetű forráshoz köthető radonszintet a lakásokban [A13].

6.c Jelentősen hozzájárultam a radon polimer/szilikát gélekben és agyag szuszpenziókban való effektív diffúziós állandójának mérését szolgáló két további eljárás kifejlesztéséhez [A16, A17]. Hasonlóan az előbbi eljáráshoz, a vizsgálandó anyagot egy radonforrás és egy mérőkamra közé helyeztük. Nyomdetektorokkal mértük a radonexpozíciót a minták két oldalán. A mintán belüli diffúziós tömegtranszportot numerikus modelleztük különböző mintavastagság és besugárzási idők esetére. A diffúziós állandót, mint paramétert, a mért expozíciókhoz tartozó legjobban illeszkedő modellezés eredménye szolgáltatta. A kísérleti elrendezés zártságát Dataqua típusú folyamatos radonmérőkkel felvett idősorok elemzése alapján ellenőriztem. Az elemzés a besugárzási idő rész-időintervallumaira meghatározott diffúziós paraméter állandóságának vizsgálatán alapult. Úgy találtuk, hogy a vizsgált polimer/szilikát gélt tartalmazó porózus közegben az effektív diffúziós állandó 3,3*10^{-6} $cm^2 \cdot s^{-1}$, míg a Montax/agyag szuszpenzióban 6,0*10^{-6} $cm^2 \cdot s^{-1}$. Az eredmények alapján a tiszta fázishoz tartozó értékek összemérhetők a pusztán vizes oldatokat jellemző adattal, ezért az ezeket az anyagot alkalmazó szigetelési eljárások nagyon vonzóak lehetnek.

6.d. Jelentősen hozzájárultam a vízminták rádium és oldott radon tartalmának mérését szolgáló, nyomdetektor technikán alapuló új módszer kidolgozásához [A14]. A mérés során a vízmintát egy kis térfogatú, gumimembránba csomagolt radon monitorral együtt egy üvegedénybe zárjuk. A gumimembrán átengedi a radont de kizárja a vizet a radonmonitor mérőtérfogatából. A vízminta oldott radon és rádium tartalmát két független méréssel határozzuk meg. Az első méréssel a rádium és oldott radon együttes koncentrációját, a második méréssel a rádium koncentrációt határozzuk meg. Kidolgoztam a kis térfogatú radon monitor mérőtérfogatán belüli a radonkoncentráció időbeli változását leíró modellt. Az általunk használt mérési elrendezésre laboratóriumi mérésekkel meghatároztam modellben szereplő kalibrációs állandó értékét, 24,1 nyom$\cdot cm^{-2}$/nap$\cdot$kBq$\cdot m^{-3}$ [A15], mely jó egyezésben volt számításaimmal. A kidolgozott módszerre jellemző, hogy semmilyen elválasztási technikát nem alkalmaz, melyek használata terepi körülmények között gyakran nehézkes.

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7 Attachment

7.1 List of relevant publications referred in the thesis

A1. **Hakl J.**, Hunyadi I. and Várhegyi A. (1997) Radon in caves. Chapter VI.1 in: *Radon measurements by etched track detectors* (eds: Ilic R., Durrani S.A.), pp. 261-283, Singapore, World Scientific,

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A3. **Hakl J.**, Hunyadi I., Csige I., Géczy G., Lénárt L., Törőcsik I. (1992) Outline of natural radon occurences on karstic terrains of Hungary. *Radiation Protection Dosimetry*, **45**, pp. 183-186

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A5. **Hakl J.**, Hunyadi I. and Törőcsik I. (1991) Radon measurements in the Baradla cave. In: *Conference on the karst and cave research activities of educational and research institutions in Hungary*, (eds. L. Zámbó, M. Veres), pp. 109-115, ISBN 9637173692, Berzsenyi College, Szombathely

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A11. Várhegyi A, **Hakl J.**, Monnin M., Morin J.P. and Seidel J.L. (1992) Experimental study of radon transport in water as test for a transportation microbubble model. *Journal of Applied Geophysics*, **29**, pp. 37-46

A12. Várhegyi A. and **Hakl J.** (1994) A silicon sensor based radon monitoring device and its use in environmental geophysics. *Geophysical Transactions*, **39**, pp. 289-302

A13. **Hakl J.**, Hunyadi I., and Tóth-Szilágyi M. (1991) Radon permeability of foils measured by SSNTD technique (Non-equilibrium approach). *Tracks and Radiation Measurements*, **19**, pp. 319-320

A14. **Hakl J.**, Hunyadi I., Varga K. and Csige I. (1995) Determination of radon content of water samples by SSNTD technique. *Radiation Measurements*, **25**, pp. 657-658

A15. **Hakl J.**, Hunyadi I., Csige I., Vásárhelyi A., Somlai J., Faludi G. and Varga K. (1995) Determination of dissolved radon and radium content of water samples by track etch method. *Environment International*, **22**, pp. S315-S317

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