“BABEŞ-BOLYAI” UNIVERSITY, CLUJ-NAPOCA
FACULTY OF ENVIRONMENTAL SCIENCE
AND ENGINEERING

PhD THESIS - SUMMARY

RADON AND RADON FLUX FROM SOIL. APPLICATIONS IN ENVIRONMENTAL SCIENCE, GEOLOGY AND GEOPHYSICS

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The researches discussed in the present thesis have been carried out in two PhD stage:

- in the past, at the Eötvös Loránd University (ELTE), Faculty of Science (TTK), Budapest, Hungary under the scientific supervision of Prof. Dr. Ádám Kiss and Dr. Ákos Horváth and, recently, at the Babeş-Bolyai University, Faculty of Environmental Sciences and Engineering, Cluj-Napoca under the scientific supervision of Prof. Dr. Constantin Cosma.

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Key words:

- ***natural radioactivity,***
- ***radon in soil,***
- ***radon flux from soil,***
- ***permeability of soil,***
- ***radon risk,***
- ***radon potential,***
- ***radon-prone area,***
- ***faults,***
- ***thoron,***
- ***geophysics***

Abstract

The main aim of this thesis is to applying the methods of measuring radon and radon flux from soil, in environmental science, geology and geophysics. *Chapter I* presents the properties of radon, sources, generation in soil and rocks, and the presence of it in different environmental factors: atmosphere, indoor air, and water, respectivelly. *Chapter II* presents the main processes of radon migration in the soil (by diffusion and advection) and transport to the atmosphere, by describing of the general model of radon transport in soil, in order to determine values for radon flux, based on the results of measurements and calculations. The methods of radon in soil measurements applide in these studies are presented in *chapter III*, by describing a method for radon concentration measurement in soil, and the permeability of soil. *The last chapter IV* describes in details studies of radon in soil with applications in environmental science, which gives results for **radon risk assessment** by determining the radon potential from soil. Applications in geology and geophysics, give results in the **identification of tectonic faults** and for the **determination of geophysical parameters of geological formations**, by the role of radon as „trace element".

1. INTRODUCTION

One of the main members of natural radioactivity of the earth crust is **radon** (^{222}Rn). Being a noble gas and having a relatively long life-time, has a greate mobility to reach considerable distances in different geological environments. Radon is present everywhere, in rocks and soil, in subsurface and deep water, in atmosphere and indoor air, in different concentrations. The concentrations of radon in geological environments depend mainly on the migration processes (by diffusion and advection) and the abundance of it parent nucleus radium, located in minerals in the rocks and soils. Upward migration of radon gas in soil is facilitated by the presence of faults with or without contribution of carrier gas, CO_2 [*Etiope and Martinelli, 2002*]. In the atmosphere reaches by diffusion to the surface, this exhalation forming the radon flux of the earth crust [*Ristoiu et al., 1995*].

Radon and radon flux from soil are used as indicators for some applications such as radon risk assessment by the determination of radon potential of the soil [*Cosma,* ***Papp, et al., 2010***], identification of the faults [***Papp et al., 2010***], in applying migration models in soil and geological environments [*Etiope and Martinelli, 2002*] and for the transport to the atmosphere and inside homes, respectivelly [*Rogers and Nielson, 1991*].

There are at least three different issues of great importance in radon studies. The first issue relates to the presence of radon and radium in ground waters (wells, mineral springs, geothermal waters, etc). In addition to the knowledge of radiation doses received by population in using these water sources (by ingestion, inhalation, spa treatment) [*Szabo, 1978; Cosma et al., 2008; Moldovan et al., 2009*], radon monitoring in groundwaters and geothermal waters is a great interest in geophysical studies [*Cosma et al., 1996 a,b; Horváth et al., 2000; Roba et al., 2010*]. The second aspect is related to the radon potential in soil and the flux from the earth (soil) surface. By this, is important that radon anomalies indicate radioactive accumulation (U, Th) or the presence of tectonic faults. In such areas, radon flux from soil is significantly higher [*Cosma et al., 1996 a,b*]. The third aspect, that is also very important, is related to radon concentrations inside homes. Outdoor air has an average radon concentration of 4-8 $Bq \cdot m^{-3}$ that depends on the geological and meteorological conditions. Inside homes, the radon concentration may produce normal amounts of 20-80 $Bq \cdot m^{-3}$, through accumulation, leading in some cases to values of the order of thousands of $Bq \cdot m^{-3}$ [*Cosma et al., 2009*]. In case of high indoor radon levels the main radon sources are the soil and building materials, which contain radioactive materials or uranium waste in uranium areas. These zones are considered „radon-prone areas" [*Sandor et al., 1999; Saintz et al., 2009*].

Increased radon values can also found in underground uranium mines, or in the preparation of the phosphate materials. Studies performed on different uranium miners revealed a true correlation between radon concentrations and lung cancer risk. Today, there are ongoing epidemiological studies (**SERTIR, 2008**) and Joint European Projects (**POSCCE, 160/2010**), which seek to highlight the connection between lung cancer risk and radon concentrations, even of normal indoor concentrations of 40-300 $Bq \cdot m^{-3}$ [*Cosma et al., 2009*]. This aspect forms the main area of most radon researches.

In addition to these important aspects of radon studies, another research field are the applications in geophysics, where an important aspect is the origin and migration of radon. Radon considered as „*trace element*" or „*monitoring element*", can give information about geophysical properties of geological formations [***Papp et al., 2008***]. Another important application is the use of radon monitoring techniques in the studies of volcanic eruptions [*Gasparini and Mantovani, 1978; Imme et al., 2006*], and of seismic activities, where the monitoring radon concentration variations in bore-holes and groundwater can be applied to earthquakes forecast [*Igarashi et al. 1995; Yang et al. 2005*].

The aim of the researches was to apply the radon and radon flux from soil measurement methods in studies of environmental science, geology and geophysics, which are presented in the thesis. The applied methods give results for the **risk assessment of radon from soil** by determining the radon potential from soil, to **identify tectonic faults** and for the **determination of geophysical parameters of geological formations**. These studies are based on an extensive bibliography about radon, and take into account various aspects of migration and transport to the atmosphere, and the behavior of this element in different environmental factors. These studies were worked out in two doctoral stages. In first stage, at Eötvös Loránd University, Faculty of Science (ELTE-TTK), Budapest (between 2001-2004), I had the opportunity to familiarize myself with nuclear methods in order to study environment and, in the second stage, at the Babes-Bolyai University, Faculty of Environmental Sciences (between 2008-2011), I had the opportunity to apply acquired knowledge in various research activities related to radon.

The thesis is structured in four chapters, as follows:

The first chapter briefly analyzes the properties of radon; it sources in the soil and earth crust, generation in soil and rocks, and the presence of it in different environmental factors: atmosphere, indoor air, water and ground-waters, respectivelly. This chapter shows the different concentrations that can generated in these environmental factors.

The second chapter presents the migration mechanisms of radon in soil and transport to the atmosphere, based on the results of radon and radon flux measurements in soil. The main characteristics of the soil are introduced and the main migration processes, diffusion and advection, are treated. Also, in this chapter the general model of radon transport in soil is presented, by describing the general transport equation. In the end of the chapter different methods for solving the transport equation by approximation, focusing on the values of radon fluxes obtained from measurements and calculations are presented.

Chapter three deals with the details of methods of measuring radon and radon flux from soil, by instantaneous and continuous measurement methods, based on different techniques. This chapter contains also the description of the applied measuring method of radon in soil, and a special method for measuring soil permeability, which was used in the following studies. The results from an international intercomparison exercices of radon in soil measurements are also presented.

Chapter four presents applications of radon studies in environmental science, geology and geophysics. This chapter is divided into three sections, in which the first section presents *applications of radon studies in environmental science*, in the view of radon risk assessment from the soil, by the determination of the radon potential from soil. In the first part of this section are presented methods of radon risk assessment from the soil, in comparison with the Czech method, which is particular for a building site, by measuring radon concentration in soil and the soil permeability. Part two of this section presents an application of radon risk assessment from the soil in the uranium area of Stei-Baita, based on indoor radon surveys. This area was declared as a " radon-prone area". The second section presents *applications of radon studies in geology*, where the upward migration of the soil gases toward the surface is controlled by tectonic faults and therefore radon studies can be used in the *identification and location of the direction of the faults*. In the first part of this section is presented a radon and thoron study in the post-volcanic area of Harghita, conducted in the area of the mofettes and mineral springs in Harghita-Bai (Harghita Mountains), by systematic measurements of radon and thoron in the soil. The second part of this section presents the results of radon measurements at Peceneaga-Camena fault (Dobrogea), based on a current geodynamic research conducted by the Institute of Geodynamics, from Bucharest. The third section presents *applications of radon studies in geophysics*, where radon can play a role of a monitoring element in geological environment. The first part of this section details a method for determining geophysical parameters, measured by the accumulation of radon gas through diffusion in a closed bore-hole of geological formations. The second part presents a mathematical model based on diffusion process, in order to determine diffusion parameter of radon in geological environments. This parameter gives information on the permeability for gases in different geological formations. The last part of this section presents the reproducibility of the method in other bore-holes made in soil environment.

The final part of the thesis contains conclusions based on the results obtained from all presented studies in connection with radon in soil.

2. The applide method of radon concentration measurement in soil

The method for measuring radon concentration in soil was based on the sampling of soil gas and the detection of radon gas. Radon activity concentration from the soil gas was measured using a LUK3C radon and thoron detector. This detector was developed for radon measurements in soil, and determines the concentration of the radon gas relatively quickly (directly from the alpha decay of radon and progenies). The measurement technique used for this detector was based on a scintillation technique with Lucas cells, by volume 145 mL. The scintillation material of the Lucas cell (deposited on the interior wall of the cell) was ZnS. The efficiency of this technique was (2.2 counts/sec) at 1 Bq radon activity, deposited in the Lucas cell, when radon is in equilibrium with its daughters.

For the collection of soil gas, a special sampling probe was used (a steel pipe with the length D = 1m and diameter d = 1cm), which was inserted into the soil at a certain depth. To create an active volume at the end of the probe in soil, it should remove a few cm. For soil gas sampling was used a Janet Syringe, with a volume of 145 mL (equal with the volume of the Lucas cell). The syringe is connected to the upper end of the sampling probe, at soil surface. Before concentration measurement, radon gas from soil must forced by three subsequent extractions, to avoid contamination by atmospheric air. After the third extraction (by the volume of the syringe equal with the volume of the cell) the soil gas probe was introduced into the detector cell with the help of a preliminary vacuum technique. The scheme for the soil gas sampling and its insertion into the detector is shown in fig.2.1.

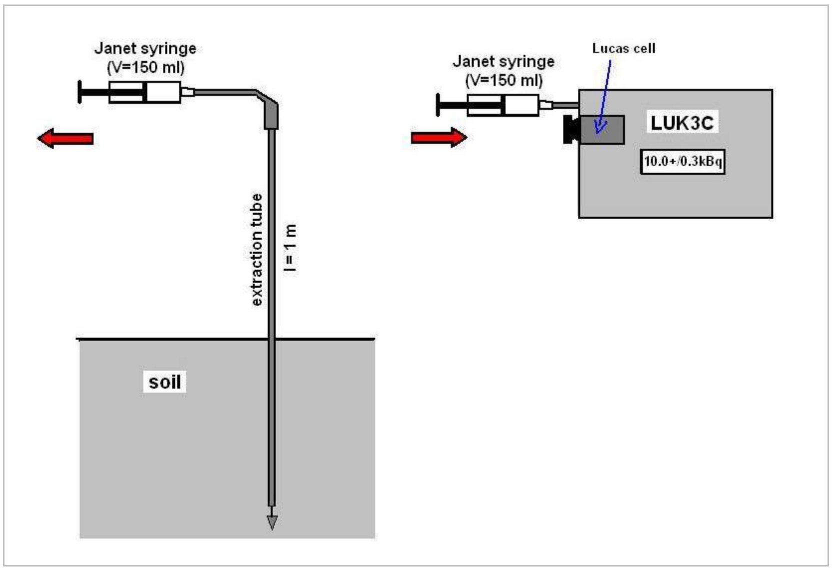

Fig. 2.1. The scheme for soil gas sampling and insertion of it into the Lucas cell of the LUK3C detector, for radon activity concentration measurement.

The principle of the measurement method is in separation of the counts come from the alpha decay of Rn, from the total incoming alpha counts (Rn+Tn). Since the half-life of Tn (55.6 sec) is much shorter than the half-life of Rn (3.82 days), Tn effectively decays in ~ 5 minutes. During this time, the detector does not measure, which called delay time. Following the delay time, the detector performed several countings that comes from the decay of the Rn atoms in the scintillation cell, and it finished when the statistic errors get under 5 %. Finally, the detector determines an average Rn concentration (corrected from the background of the cell) and an estimation for Tn concentration, that is determined from total (Rn+Tn) counts minus the average Rn counts (corrected from the background of the cell). The total time of one measurement is no more than 10 minutes [*Barnet et al., 2008; Neznal et al., 2004*].

2.1. Intercomparison exercise of radon measurements in soil, RIM 2010

The method of radon measurement in soil gas was verified at an Intercomparison Exercise of Radon Measurements in soil (RIM 2010), Prague, 20-21 of September 2010. The results of the intercomparison was included in a certificate issued by the organizers of the exercise, „*Protocol on the evaluation of comparison measurement of radon (^{222}Rn) activity concentration in soil gas at reference sites Cetyne, Bohostice and Buk (Czech Republic)*".

The intercomparison measurements were performed at radon reference sites Cetyne, Bohostice and Buk in the Czech Republic, in the frame of the "*10th International Workshop on the Geological Aspects of Radon Risk Mapping*", 22-25 of September 2010, Prague. Administrator of the three radon reference sites is the Faculty of Science, Charles University, Prague. Each reference site implies 15 stations of measurements in a grid of 5x5 m, and the soil gas was sampled from the depth of 0.8 m. The rock type of the reference sites was very homogenous, the soils were clayish and sand and the permeability covered all the three classes, low, medium and high. About the mean radon concentration estimated by the administrator of the reference sites are: **32 kBq·m^{-3} for Cetyne reference site, 47 kBq·m^{-3} for Bohostice reference site and 146 kBq·m^{-3} for Buk reference site.**

Results of the radon intercomparison measurement, RIM 2010

The mean values of radon concentrations obtained by our group at the three reference site (in all 15 stations of each) are: **37.8 ± 3.8 kBq·m^{-3} for Cetyne, 52.3 ± 4.9 kBq·m^{-3} for Bohostice and 132.8 ± 23.9 kBq·m^{-3} for Buk.**

Tests of radon comparison measurements at reference sites

Evaluation of the results of intercomparison measurements of radon in soil is based on the comparison of individual measurements reported by a participant group with radon data of all other groups, and with radon data of a database of the respective reference site. This evaluation was performed with a computer program that includes three statistical tests.

Tests 1 and 2 compare the results of a participant group with the results of the other groups, which performed measurements in the same day and climatic conditions. *Test 1 makes differences between radon concentrations determined by a participant group and by all groups at single stations* (N=15) *of the three reference site*. *Test 2 makes a linear regression* (y=a+bx) *of datas determined by a tested group* (y) *at single stations of the three reference sites* (N=3x15=45)*, and radon data for relevant stations reported by the administrator and all other groups* (x). ***Test 3*** *makes differences between the means of radon data reported by a participant group and the means of data of all groups in the database of a single radon reference sites*. The database is gradually formed using radon data of the groups which passed the test since 2000. At present, the data of 180 groups form the databases of reference sites.

After test 1, values outside of the confidence interval at the reference sites are: **4/14 for Cetyně, 5/15 for Bohostice, and 6/15 for Buk**. After test 2, the values of the parameters from linear fit for our group are: **a = 0,599** and **b = 0,984**, by the correlation coefficient $\mathbf{R^2}$ **= 0,982**. After test 3, the results of the comparison between the mean of the data of our group and the data of all the participating groups including the database since 2000, for the three reference sites are: **0.96 for Cetyně**, **0.99 for Bohostice, and 1.041 for Buk. The average of these values is 0.997.**

Conclusions

Test 1 and test 2 (orientation tests based on the comparison with participating groups) show a good agreement of the results of our group with other groups in intercomparison exercices. Test 3 (the decisive test based on comparison with the databases of radon reference sites) shows good agreement between our mean values with radon data of all successful groups (N = 180) who measured at radon reference sites since the year 2000 and form a database. After the decisive test 3, the comparisons fulfil the test criteria and the estimation of the radon concentration in soil gas by our group are very well acceptable, with an average of 0.997 between the determination of our group and of the other participants.

3. <u>The applied method for soil permeability measurement</u>

Permeability is a main factor in the transport processes of the gases in soil that greatly affecting the flux of radon gas in soil or the exhalation of it from the soil. The permeability of soil and rocks is one of the most important factors determining possible radon sources of a building site, therefore is one of the main parameters for the final radon risk assessment of building sites. All methods used for radon risk classification is based on determination of radon concentration in soil, and of the permeability of soils and rocks [*Barnet et al., 2008*].

In situ determination of the soil permeability can be performed by measuring the flow rate of a water column from a bottle, which is connected directly to a soil probe, for soil gas extraction. The probe is inserted into the soil at a reference depth of ~ 0.8 m, as in the radon concentration measurements. The principle of the instrument is based on the extraction capacity of the gas from soil by a negative pressure, through a soil probe with constant height. The active area of the probe is created at the end of the probe (in soil) by a lost tip, at a given depth. The scheme of the device for soil permeability measurements is shown in fig.3.1.

The flow rate of water column (*q*) depends on soil permeability (*k*), so that for high permeability of the soil the flow of water is high, and for low permeability, the flow is low, respectivelly. The range of water flow rate is between 0.036 şi 7.9 L·min^{-1}. Thus, the flow rate (*q*) is directly proportional to the soil permeability (*k*).

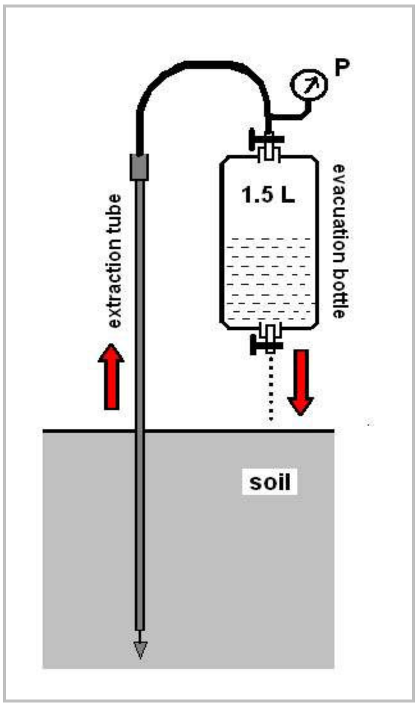

Fig. 3.1. The system for soil permeability measurements. The upper end of the plastic tube is connected directly to the soil probe.

For the permeability measurements, soil gas is considered homogeneous, isotropic and incompressible (pressure differences are slightly small than atmospheric pressure). The relation between the permeability and the flow rate is described by:

$$k = \frac{Q}{\Delta p} \cdot \frac{\mu}{F} \quad (3.1),$$

where, k[m^2] is the soil permeability for gases, Q[m^3·s^{-1}] is the gas flow, μ[Pa·s] is the dinamic viscosity of air (=1,75·10^{-5} Pa·s, la 10°C), Δp [Pa] is the pressure difference between surface and the active area of the probe, F[m] is the shape factor of the probe (depending on its geometry).

The critical point of this method is in the determination of the shape factor *F(L,d,D)* of the probe. Applicable solutions are described in [*Damkjaer and Korsbech, 1992; Mosley et al., 1996; Barnet et al., 2008; Radon-Jok, manual*] and the resultant formula is:

$$F = \frac{2 \cdot \pi \cdot L}{\ln\left(\frac{2 \cdot L}{d} \cdot \sqrt{\frac{4 \cdot D - L}{4 \cdot D + L}}\right)} \quad (3.2),$$

where, L[m] is the length of the area of the probe head, d[m] is the diameter of the active area and D[m] is the depth below the surface, in the approximation of $L >> d$.

3.1. Calibration of the instrument of measuring soil permeability

The laboratory calibration of the instrument for soil permeability measurements consists in studying the dependence of water flow rate and air pressure on the extraction time of the gas from soil. In order to studying the dependence of the instrument parameters in the laboratory, we used the bottle (with a volume of V≈1.5 L) equipped with two valves (in the upper and bottom part of the device), and a sensible control valve to simulate the permeability and a pressure gauge to measure the negative pressure created in the bottle.

Calibration method is based on measuring the following parameters: intermediate times (t_i) at every volumes (V_i), corresponding to the divisions on the bottle, and negative pressures (p_i) which are measured by the pressure gauge. Measurements for 11 different openings of the control valve that simulate the permeability were performed, each set of measurements was repeated three times. Readings of the parameters was performed according to the divisions on the tube, that corresponds to the volumes (V_i), from 300 to 300 mL.

The ratio (q/p) for different openings of the control valve was calculated. Firstly, the flows (q_i) and pressures (p_i) for each division, and then their ratios (q_i/p_i) were calculated. For the selected five divisions an average value $(q/p)_m$ and a standard deviation $\Delta(q/p)_m$ was calculated. The dependence of the $(q/p)_m$ on the flowing out time (t_m) of the water from the bottle, for a volume of water (V) is shown in fig.3.2.

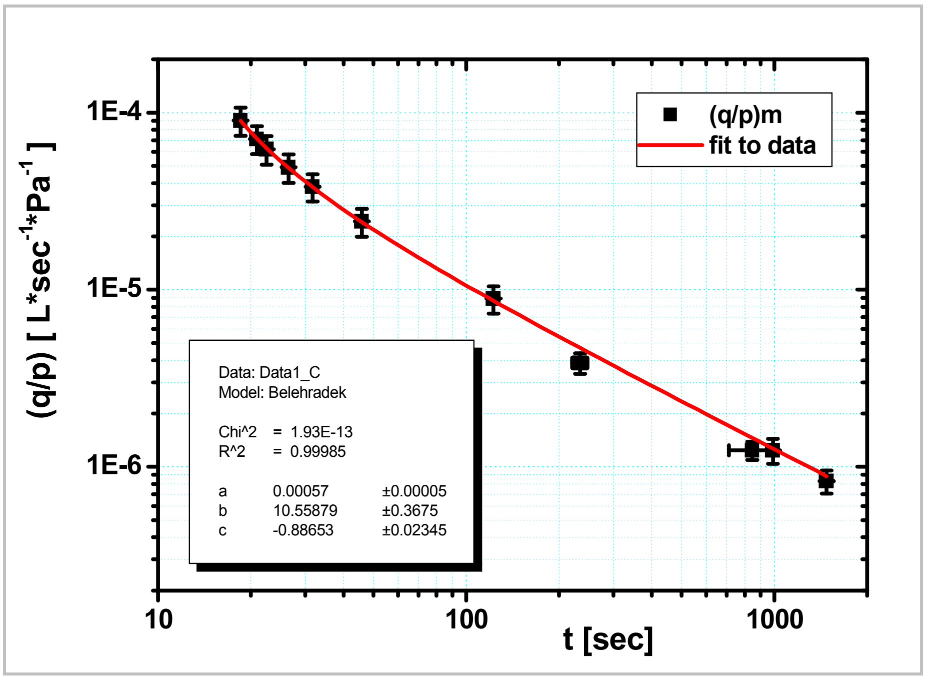

Fig. 3.2. The dependence of the ratio $(q/p)_m$ on the time of the water flow (t_m) from the bottle. (in duble logarithmic scale).

The relationship between the ratio (q/p) and the flow time (t) can be determined by fitting a power function to the measured datas:

$$\boxed{q/p(t) = a \cdot (t-b)^c} \qquad (3.3)$$

where the values of the obtained parameteres from the fit are: a = (5,7±0,5)·10^{-4} L·Pa^{-1}, b = 10,56±0,37 s and c = -0,89±0,02. Here, $b \approx$ 10.6 s is a time parameter representing the detection limit of the device that corresponds to the value of the flow rate of 7.9 L·min^{-1}. The upper limit of the instrument is ~ 40 min that corresponds to a more low value of water flow rate of only 0.04 L·min^{-1}.

Intercomparison of the special method for soil permeability measurements with RADON JOK permeameter

It was performed also a laboratory intercomparison of the "special instrument" with Radon-Jok permeameter (from Radon v.o.s.), which is based on the comparison between the parameters of both instruments. Therefore, the Radon-Jok permeameter was used, in addition with a sensitive control valve to simulate the permeability (the same as for the calibration of the special instrument) and a pressure gauge to measure the negative pressure that was created in the rubber sack of the Radon-Jok. In this configuration, was measured the following parameters: intermediate times (t_i) of filling the air sack of Radon-Jok and the negative pressures (p_i) exercited in the sack, measured by the pressure gauge.

Measurements were performed for seven openings of the control valve, which simulates the permeability. The study of the Radon-Jok permeameter is based on the calculation of the (q/p) for different openings of the control valve. The ratio (q_i/p_i) was calculated from the values of flows (q_i) and pressures (p_i) for each time (t_i). Because during the extraction process, pressure differences (Δp_i) between different times (t_i) are very small (thus filling the sack of the Radon-Jok has a constant rate), from the values of the ratios (q_i/p_i) was determined an average $(q/p)_m$ and a standard deviation $\Delta(q/p)_m$. The dependence $(q/p)_m$ on the filling time of the sack (t_m) is shown in fig.3.3.

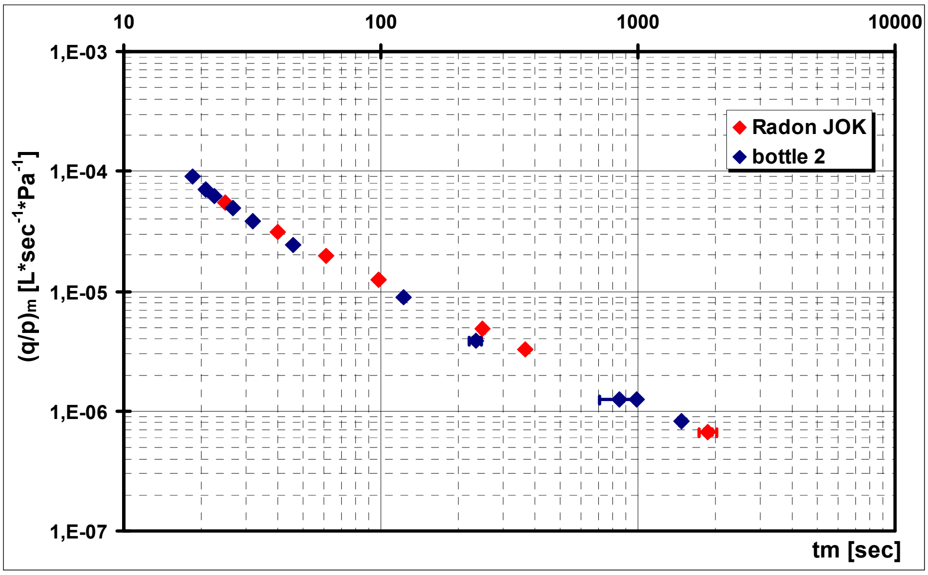

Fig. 3.3. Result of the intercomparison between the „special method" for permeability measurement and the Radon-Jok permeameter. Red points are the ratio $(q/p)_m$ depending on time (t_m) measured by Radon-Jok and the blue points are the same dependence measured by the special method (in double logarithmic scale).

Conclusions

The device developed for in situ soil permeability measurements can be easy to use in the field. The permeability parameter (*k*) depends on the ratio of the water flow rate and the negative pressure created in the bottle (*q/p*), and depends also on the shape factor of the soil probe (*F*). The measuring range by the special method is from the very high permeability value ($5\cdot10^{-8}$ m^2, corresponding to an extraction time of 11 sec) to ultra low permeability value ($7\cdot10^{-14}$ m^2, corresponding to an extraction time of 40 min). Intercomparison of the two instruments for permeability measurements gave good results. The values of the parameters $(q/p)_m$ is in a good correlation within the limits of the measurement errors.

4. APPLICATIONS OF RADON STUDIES IN ENVIRONMENTAL SCIENCE, GEOLOGY AND GEOPHYSICS

In this chapter aspects of the radon in soil measuring method are presented, with specific *applications in environmental science* (by the assessement of radon risk from soil); *in geology* (by the identification of the tectonic faults and location their direction), and *in geophysics* (by the determination of the geophysical parameters of geological formations).

4.1. Radon studies with applications in risk assessment of radon from soil

Researches have shown that radon is the main source of natural radiation for the population, with a contribution of about 57 % to annual dose. In some areas, this can reach contributions over 95 %, increasing 5-10 times the dose, towards to the annual mean exposure of 2.2 mSv. Depending on geological and meteorological conditions, regions can be identify with significant accumulations of radioactive materials, so that the annual dose can be 55-200 times higher than the global average. Comparing the annual collective dose due to radon exposure for the population in Romania (1.77 mSv) and the average annual dose around the world (1.2 mSv), it can be seen that the value for Romania is slightly increased [*Cosma et al., 2009*]. In Romania there were and exist regional programs for indoor radon, leading to a very large range of the results, from few tens to several thousands of $Bq\cdot m^{-3}$. In the past 20 years, the Laboratory of Environmental Radioactivity, in the frame of Environmental Science Faculty, Babes-Bolyai University, conducted regional campaigns to evaluate the indoor radon in Transylvanian counties (Cluj, Bihor, Alba, Bistrita, Sibiu), by performing measurements over 1,800 homes. Based on the results, the estimated average exposure of the indoor radon is 82.5 $Bq\cdot m^{-3}$ [*Cosma et al 2009; Sainz et al, 2009*].

In radon risk areas ("**radon-prone areas**") the concentration of radon gas in atmosphere and indoor can reach high levels, which is due to soil and building materials. The assessment of the radon risk from soil is based on the determination of the radon potential by measuring radon concentration from soil and the permeability of soil. [***Papp et al., 2009***; ***Papp et al., 2010;*** *Cosma,* ***Papp, et al., 2010***].

4.1.1. Radon risk assessment method from soil or building sites

The risk assessment of soil radon is useful in the development of protective measures to radon for new buildings. The soil radon risk assessment method are used also for radon mapping, which results are used for evaluating the geogenic radon potential of specific geological units [*Kemski et al., 2001*]. The used model for risk assessment of soil radon is the model of Neznal [*Neznal et al., 2004*], which is based on the determination of the radon potential by measuring radon concentration in soil and permeability of the soil. The model uses three categories of risk: *low, medium* and *high*. Thus, a high radon concentration and soil permeability provides a high risk and an increased probability of penetration radon in homes.

Tabel 4.1. The evaluation of radon index (radon risk) [from *Neznal et al., 2004*].

Radon risk category RI	Soil gas radon concentration C_{Rn} [$kBq\cdot m^{-3}$]		
Low	$C_{Rn} < 30$	$C_{Rn} < 20$	$C_{Rn} < 10$
Medium	$30 \le C_{Rn} < 100$	$20 \le C_{Rn} < 70$	$10 \le C_{Rn} < 30$
High	$C_{Rn} \ge 100$	$C_{Rn} \ge 70$	$C_{Rn} \ge 30$
permeability	*low*	*medium*	*high*

The **soil gas radon concentration** is one of the main parameter in determining the radon potential of a building site. Usually, the determination of soil gas radon concentrations is performed by instantaneous measurements of soil gas probes, sampled from a standard depth of **0.8 m**, and represented in units of [$\mathbf{kBq\cdot m^{-3}}$]. The detection limit for radon concentrations must be at least **1** $\mathbf{kBq\cdot m^{-3}}$, because the reliability of detection instruments and sampling errors.

Because of the inhomogeneous distribution of radon in soil and the presence of anomalies, the determination of radon potential requires several measurements of radon concentrations. The evaluation of a building site of a single building ≤ 800 m^2, must be performed at least 15 sampling points, within the building's soil and its close vicinity. When a building site > 800 m^2 is evaluated, soil gas samples are collected in a 10x10 m grid, so that must be covered subsoil around the building and its close vicinity. It is recommended to increase the number of sampling points and to use a grid of 5x5 m, in cases of local anomalies higher than three times of third-quartile (3*$C_{Rn,75}$) [*Neznal et al., 2004*].

When classifying a building site ≤ 800 m^2 for a single building (involving at least 15 soil gas radon concentration measurements), the <u>decisive value for radon concentration is the third quartile of the data set</u> (***C_{75}***). Values **less than 1 kBq·m^{-3}** are excluded from the dataset, which are not characteristic for soil radon levels, and values higher than the third quartile are considered local anomalies of radon [*Neznal et al., 2004*].

<u>**Soil permeabiliy**</u> is the second main parameter in determinig the radon potential of a building site. High permeability allows an increased transport of radon from soil and transfer to the building, thus in case of permeable soils can be estimated an increased radon risk. Soil permeability can be determined by in siut measurements, where the ***k*** pemeability is given in [**m^2**]. In situ soil permeability measurements usually is carried out at a depth of **0.8 m** in soil, and the measurement method consist in measuring the flow rate of the soil gas by extraction or by pumping in soil at constant pressure [*Neznal et al., 2004*].

According to radon risk assessment, the categories of soil permeability are the follows: $k < 4{,}0\cdot10^{-13}$ m^2 for low permeability; $4{,}0\cdot10^{-13}$ $m^2 < k < 4{,}0\cdot10^{-12}$ m^2 for medium permeability; and $k > 4{,}0\cdot10^{-12}$ m^2 for high permeability. The number of in situ permeability measurements are the same as for soil radon measurements, at least 15 measurements for a building site (≤ 800 m^2 area), or perform measurements in grides of 10x10 m for sites with area > 800 m^2. Third quartile of the data set reduces the influence of erroneous results, and local anomalies of permeability [*Neznal et al., 2004*].

<u>The decisive value</u> for classifying a building site ≤ 800 m^2 (by at least 15 in situ permeability measurements) <u>is the third quartile of permeability data set (***k_{75}***)</u>. Higher values then the third quartile are excluded from dataset. For building sites > 800 m^2, permeability measurements must be performed in grids of 10x10 m, that depends on the homogeneity of

the place and of dataset. Local anomalies can influence the final classification of permeability [*Neznal et al., 2004*].

Radon potential of the soil

The determination of radon potential of soil is based on the evaluation of soil gas radon concentration and the permeability of soils. High soil gas radon concentration and high permeability of soil, gives high probability of entering radon gas in buildings. **The radon index of a building site (RI)** indicating the level of radon risk release from the bedrock or subsoil, and **the radon potential (RP)** espressing the radon index of the place (RI), for the categories *low, medium and high* [*Neznal et al., 2004*].

The model of radon potential is based on the tabel of radon index classification (table 4.1), by which the model can be represented on a diagram according to fig.4.1. According to this, radon potential **RP** is defined by:

$$\boxed{RP = (C_{Rn} - 1)/(-\log k - 10)} \tag{4.1}$$

where, C_{Rn} [kBq·m^{-3}] is the third quartile of soil gas radon concentration data, and k[m^2] is the third quartile of soil permeability data [*Neznal et al., 2004, Barnet et al, 2008*].

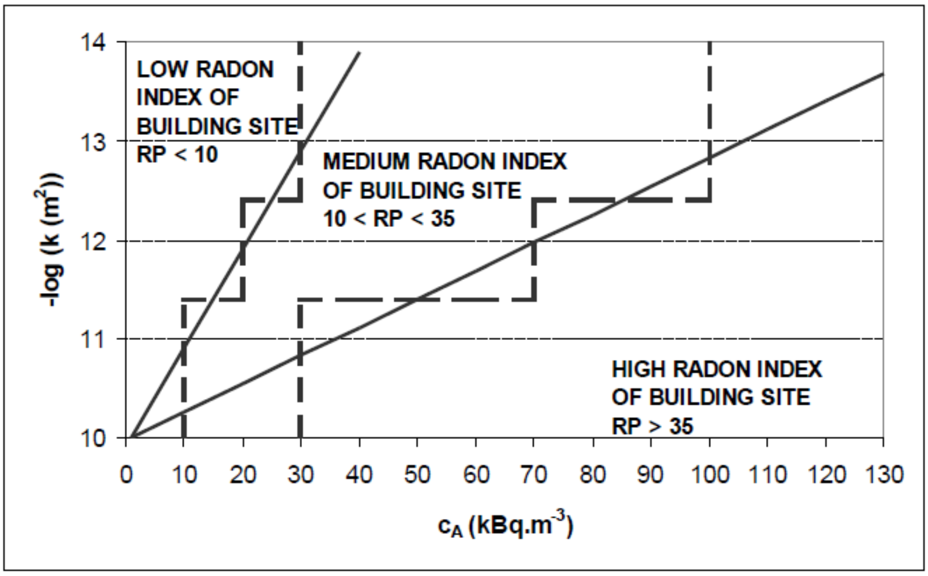

Fig. 4.1. Radon potential of a building site [by *Neznal, 2004; Barnet et al., 2008*]

For risk assessement of a building site by the radon potential model, the decisive values to determine radon potential (RP) are the third quartile of data series of soil gas radon concentrations (***C**$_{Rn75}$*) and the third quartile of data series of soil permeability (***k**$_{75}$*).

Therefore, the radon index (RI) of a building site are the follows: for RP <10, RI is low; if 10 ≤ RP <35, RI is medium, and if RP ≥ 35, RI is high [*Neznal et al., 2004; Barnet et al., 2008*].

4.1.2. Connection between indoor radon and radon in soil and building material in Baita-Stei uranium area

The present study is a part of an extensive research on indoor radon exposure in the region of Transylvania. The most important high background radiation area in Transylvania was located in Băița-Ştei (Bihor County), where have been found indoor radon concentrations higher than 1000 $Bq{\cdot}m^{-3}$ [*Cosma, et al. 2009; Sainz et al, 2009*].

The area of Băița-Ştei is located in the Bihor Mountains (NW part of Romania) in the neighborhood of "Avram Iancu" and "Băița" uranium mines. Geographical coordinates of the zone are: N: 46° 28' and E: 22° 34', and the altitude is 430 m. This area includes the town Ştei and few villages (Băița-Plai, Băița-Sat, Nucet, Fânațe, Cîmpani etc.), with a total of approximately 15.000 inhabitants.

The main reasons for high indoor radon concentrations in this region is the using of uranium tailings as building material (from the uranium mines operating in the period of 1952-1990), and the building subsoils with high permeability. Most of these houses were built using radioactive tailings or local building material (sand, gravel, etc.) from the Băița-Criş river bad, having increased amounts of radium and uranium [*Sandor et al, 1999*].

Indoor radon measurements and results

The starting point of radon in soil measurements is a detailed study of indoor radon in Stei-Baita area, conducted between 2003-2008. The study based on integrated measurements of radon in a large number of 335 selected houses in the zone, whose results are the base of a research project: *IRART, „IMPLEMENTATION OF RADON REMEDIATION TECHNIQUES IN DWELLINGS OF BĂIȚA URANIUM MINE AREA"* [*Cosma, **Papp, et al., 2011**; Cucoş, **Papp et al., 2011***].

The integrated indoor radon measurements are made by using CR-39 track detectors, according to the NRPB Measurements Protocol. In order to evaluate the average concentrations of indoor radon, detectors were exposed in habited rooms of the houses (e.g. bedrooms, at a height of 1.0-1.5 m from the floor) [*Cosma, et al., 2009; Sainz et al, 2009*].

The indoor radon concentrations are in a wide range from **15 to 2000 Bq·m^{-3}**, depending on the structure of the building material and the type of soil under the building. Considering the geological and the seasonal corrections, the mean value of the indoor radon concentration is **343.5 Bq·m^{-3}**, which is 4.16 times higher than the average indoor radon concentration of 82.5 Bq·m^{-3} reported for Transylvania [*Cosma, et al. 2009, Sainz et al, 2009*]. Indoor radon concentrations in the monitored region are significantly higher than the recommended level of 100 Bq·m^{-3} for occupational and residential exposure of population.

The indoor radon concentration data set contains two relevant sub-domains that can indicate the main sources of indoor radon. The first, having the range **bellow 400 Bq·m^{-3}** corresponds to a percentage of ~ **90 %** of the investigated houses and coming from **soil and normal building material, as a main source for indoor radon**. The second, having the range oved than **600 Bq·m^{-3}** correspond to a percentage of ~ **6 %** of the houses and coming from **uranium waste used in building constructions, as a secondary main source of indoor radon** [*Sainz et al., 2009*].

<u>Radon in soil measurements and results</u>

The preliminary radon in soil measurements in Băița-Ştei area were performed in 2010 autumn, when relatively dry conditions prevailed in the area. The aim of the measurements was the determination of the radon potential of soil, in order to make a risk assessement of radon in soil for the area [*Cosma, **Papp, et al., in press 3***]. Therefore, we performed 30 radon in soil and permeability of the soil measurements, at 10 places in the whole area (along the path of Crişul-Băița river). The sampling depth of radon gas from soil for concentration and permeability measurements was between 50-80 cm. The sampling depth depends on the structure of the upper soil layer. It mentioned that in Apuseni Mountains frequently appear solid rocks in the upper soil layer at a depth of only 50 cm.

Method of radon in soil and permability of soil measurements are presented in details in the chapter 2 and 3, respectively. The results of the radon concentration from soil and the permeability of soil measurements in Băița-Ştei area are summarized in the table 4.2, for the 10 investigated places.

For radon risk assessment in the area Băița-Ştei, the presented model of radon potential (paragraph 4.1.1) was used. In a given site, were performed several measurements of radon in soil and permeability of soil, repectivelly. In estimations, radon concentrations higher than 100 kBq·m^{-3} were excluded, because they are local anomalies in the site. Low permeability values, < $4 \cdot 10^{-13}$ m^2 were also excluded, because they are not representative for Băița-Ştei region (geologically, soils in the zone having high permeability). The highest value of radon concentrations and of soil permeability, measured in a site was decisive for radon potential of soil calculations. These values of radon potentials (**RP**) and radon risk classes (i.e. radon index, **RI**) are presented in Table 4.2.

Table 4.2. The values of the measured radon concentrations from soil and errors (CRn ± dCRn) and the values of the determined permeability of soil (k). (The number in below of the name of each site represents the number of measurements at the site, and the parameter D[m] is the depth of sampling soil gas.)

Site	*D* [m]	*CRn ± dCRn* [$kBq \cdot m^{-3}$]	*k* [m^2]	*RP*	*RI*
Ştei (3)	0.8	44.1 ± 2.0	1.7E-12	69.8	HIGH
	0.8	40.2 ± 2.0	2.4E-12		
	0.8	22.3 ± 1.0	2.4E-11		
Lunca Ştei (2)	0.8	22.0 ± 1.2	1.9E-13	11.5	MEDIUM
	0.8	30.1 ± 1.5	3.0E-13		
Câmpani (4)	0.7	53.5 ± 2.6	1.2E-11	101.1	HIGH
	0.7	54.5 ± 2.6	2.4E-11		
	0.7	63.4 ± 2.9	2.4E-11		
	0.8	159.5 ± 7.8	7.1E-12		
Fânațe 68 (2)	0.6	58.1 ± 2.7	6.8E-13	26.8	MEDIUM
	0.6	59.1 ± 2.6	-		
Nucet Popas (2)	0.5	61.3 ± 2.8	9.8E-12	59.8	HIGH
	0.65	46.9 ± 2,1	2.4E-11		
Nucet Criş (1)	0.7	27.1 ± 1.3	5.2E-11	93.2	HIGH
Băița 204 (5)	0.8	11.9 ± 0.7	high	59.1	HIGH
	0.8	7.9 ± 0.5	high		
	0.8	45.4 ± 2.0	high		
	0.8	128.7 ± 5.1	1.8E-11		
	0.8	23.1 ± 1.1	2.8E-13		
Băița 206 (5)	0.8	5.5 ± 0.5	1.3E-11	73.0	HIGH
	0.6	7.6 ± 0.5	high		
	0.8	42.2 ± 1.2	2.7E-11		
	0.8	35.2 ± 1.5	high		
	0.8	16.6 ± 0.8	high		
Băița Plai (4)	0.4	462.9 ± 13.3	3.3E-11	1607.2	HIGH
	0.4	398.5 ± 12.0	-		
	0.4	446.1 ± 13.0	-		
	0.5	512.0 ± 14.0	4.8E-11		

The values of radon potentials of soil covers two categories of radon risk (medium and high) in a range bettween 11.5 and 1607.2, so that for most of the sites the radon risk was high. One of the sites, Băița-Plai shows very high radon risk with RP = 1607.2, which is characteristic for the site, because closed to this is the entrance of the old uranium mine "Baita-Plai", where the soil has very high uranium content.

<u>Radon exhalation measurements from gravel, used as building material and results</u>

The aim of these measurements was to determine the exhalation rate of a common building material used in construction of the houses. The material is a mixture of gravel and stones coming from Criş-Băița river. The river passes near Băița-Plai uranium mines and crosses Ştei-Băița valley. The sample was dried at a temperature of ~70°C, and was sorted into three grains fractions. The first was a sandy-gravel fraction with the diameter of the grains of 1-2 mm, the second was a gravel fraction with the diameter of the grains between 5 mm and 2 cm, and the third was a stone fraction with the diameter larger than 2 cm.

In order to measure radon exhalation rate, the fractions of the sample of building material were placed into a tight vessel, in which was followed radon concentration in time. The concentration measurements were performed by Radim3A radon monitor, in Eman version. In this case, the vessel was mounted and sealed to the detection chamber of the Radim-Eman radon monitor. The results of radon exhalation measurements of the three fractions are shown in fig.4.2.

During the measurement, the radon concentration growth, and tends to an equilibrium value. This equilibrium value depends on the emanation factor of radon atoms from the sample, and on the radium content of the sample. From fig.4.2 it can be seen that in the first part (i.e. for the first 18 points) the rate of rising radon concentration can be considered to be linear, in all three cases. Therefore, the rate of rising can be the slope of the fitted line, **k** [$Bq \cdot m^{-3} \cdot h^{-1}$]. Therefore, the exhalation rate of the sample (**ER** [$Bq \cdot kg^{-1} \cdot h^{-1}$]) was determined from the slope **k** [$Bq \cdot m^{-3} \cdot h^{-1}$] of the linear fit, by the:

$$\boxed{ER = k \cdot \left(V_{air} / M_s\right)} \quad (4.2),$$

where, $V_{air}[m^{-3}]$ is the volume of the vessel that is determined from the total volume of the entire container (radon receiver and source containers, V_{tot} = 4.0 L, reduced by the volume of the sample (V_s), and M_s[kg] is the mass of the sample. The results of the fits and the calculations are shown on the table 4.3.

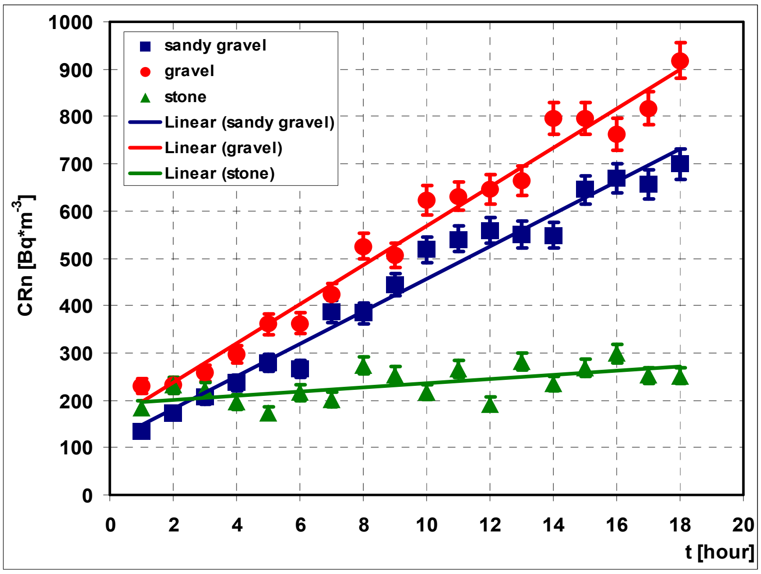

Fig.4.2. Radon concentration (*CRn*) versus time (*t*) for the first section (linear part) of the measurement. The circles are the data from sandy-gravel fraction, the squares are the data from gravel fraction, and the triangles are the data from stones, with errors. The solid lines are linear fits to the three data sets of concentrations (with the correlation coefficients (R^2) : 0.977 for sandy-gravel, 0.970 for gravel and 0.399 for stone.

Table 4.3: Results of the rising slope of the radon concentrations (*k*) from the linear fits, and the radon exhalation rates (*ER*) and errors.

fractions	k ± dk [$Bq·m^{-3}·h^{-1}$]	V_{air} [L]	M [kg]	ER ± dER [$Bq·kg^{-1}·h^{-1}$]
sandy gravel	34.24 ± 1.50	3.44	1.4	0.084 ± 0.004
gravel	41.40 ± 1.60	3.34	1.7	0.081 ± 0.003
stone	4.33 ± 1.33	3.31	1.7	0.008 ± 0.003

Conclusions

This study is based on the results of integrated indoor radon measurements performed in 335 dwellings from Băița-Ştei radon prone area (Bihor county), near Băița uranium mine, during 2000-2010. The results indicate an annual average value of indoor radon concentrations of about 343.5 $Bq·m^{-3}$, which is 4.16 times higher than the average indoor radon concentration of 82.5 $Bq·m^{-3}$estimated for Transylvania.

The distribution of indoor radon values shows that there are two independent radon sources for the houses in the area. The first source coming from soil and the second is coming from uranium waste used in building constructions.

For characterizing soil as main source of indoor radon, were performed radon in soil and permeability of soil measurements at 10 selected sites. The measurement results and estimations of radon potentials of soils provide that the majority (80%) of the investigated places shown high radon risk. These places are inhabited or building areas. One of the places, Băița-Plai shown very high radon risk (RP=1607.2). This is characteristic for the place, because closed to this is the entrance of the old uranium mine "Baita-Plai", where the soil has very high uranium content.

The second main source of the elevated indoor radon in the area is the building materials. A mixture of sandy gravel and stone used as building material, shows highest radon exhalation. From laboratory measurements on the three grain size fractions and calculations can conclude to different exhalation rates values. The first two (sandy-gravel fraction with smallest grain sizes and gravel fraction) had the same radon exhalation, while the third (normal stone with big grain size) have lower radon exhalation. As the radon exhalation of the sample, the sandy-gravel and gravel fractions had ~10 times higher then of the stones. Although, this material has an important contribution to public exposure in houses that use this as building material.

Acknowledgements. This work represents a part of a research supported by the project 586-12487, Contract No. 160/15.06.2010 with the title "*IMPLEMENTATION OF RADON REMEDIATION TECHNIQUES IN DWELLINGS OF BĂIȚA URANIUM MINE AREA/ IRART*" of the Sectoral Operational Programme "Increase of Economic Competitiveness" co-financed by The European Regional Development Fund.

4.2. Radon studies with applications in geology

An important aspect of radon (^{222}Rn) and thoron (^{220}Rn) studies is the origin and migration of these gases with applications in geophysics. Here both, radon and thoron have a role of trace elements, which can indicate accumulation of radioactive material in the crust [*Cosma et al. 1996 a*] or the presence of tectonic faults [*Cosma et al., 1996 b*]. These radioactive gases are created continuosly in the earth crust and migrate together to the surface, mainly through diffusion and advection processes [*Etiope şi Martinelli, 2002*]. Such studies can be applied to the identification of tectonic faults and to locate its direction [*Font et al. 2008; Szakács and Néda 2009; Cosma, **Papp, et al., in press 2***].

Faults serving as pathways for the ascendent migration of these gases towards surface, and can be identified by detecting high radon and thoron activities in the soil, as anomalies. Detection of high thoron activities in soil gas may indicate a fast migration mechanism from a distant source, due to the short half-time of thoron (55 sec) than of radon (3,82 days). This is possible only in presence of a carrier gas (e.g., CO_2) as typically occurs along faults and fractured rocks [*Etiope and Martinelli, 2002*]. Gas-emitting tectonic structures can be mapped as a combination of radon and thoron measurements, supported by CO_2 measurements. Thoron studies combined with other geologic investigations help to precisely determine the source and the origin of radon in the crust [*Szakács and Néda 2009;* ***Papp et al., 2010***].

4.2.1. Radon and toron study and relation by the location of fault zones in postvolcanic area of Harghita-Bai

In the aftermath of post-volcanic activity, high amounts of CO_2 emanations reach the surface, potentially carrying radioactive gases, such as radon (^{222}Rn) and thoron (^{220}Rn). Depending upon local geological conditions, upward flowing gas may appear at the surface either as dry CO_2 emanation (mofette) or as CO_2-rich mineral water. A mofette is mainly low temperature, dry gas, mostly CO_2 emanation, that breaks out from the Earth's deep interior to the surface. Mofette gases typically have a magmatic origin or a mantle component. There are many occurrences of dry CO_2 emanations along the Neogene Harghita volcanic range (East Carpathians, Romania). The upward migration of these gases is facilitated and controlled by tectonic faults [*Neda et al. 2008 a,b; Szakacs and Neda 2009;* ***Papp et al., 2010***].

The study presented here is based on measurements of radon and thoron activity concentrations in soil performed around the mofettes and the mineral springs at Harghita-Bai, Harghita Mts. The aim of the study was to identify and to locate, through the use of systematic measurements of soil radon and thoron activity concentrations, the possible direction of the fault system which controls the occurrence of the two mofettes and the mineral springs in the resort area.

The studied area is part of the North Harghita Mountains which is part of the Neogene volcanic range in the East Carpathians and belongs to the large Varghis volcano. Harghita Bai is a resort village (N 46°23'18", E 25°38'19") with an altitude between 1300-1400 m (a.s.l), and is located at ca. 18 km to the east of Miercurea Ciuc, the capital city of Harghita county. The volcanic rocks in the zone occur within one of the main hydrothermal zones of the Varghis volcano, along a presumed NS striking fault. The CO_2 content of the post-volcanic emanations can be as high as 99.8% of the total gas emitted. The mofettes from Harghita Bai are essentially CO_2 emanations, with trace amounts of sulphurous and other gases.

Radon and thoron measurements in the area of the mofettes and mineral springs at Harghita Bai, and results

The field measurements of radon and thoron, near the mofettes and the mineral springs at Harghita-Bai, were performed in June 2009, when relatively dry conditions prevailed in the area. Measurements were performed in two groups, each one along a well-determined profile line. One of them was located in front of the mofettes (profile A) and the second was located at a distance of ~ 200 m from the mofettes to the south (profile B) (see fig.4.3 and 4.5). In the case of profile A, the profile line was normal to the direction that connects the two mofettes. In the case of profile B, the profile line was normal to the direction that connects the mofettes and the mineral springs in the resort area of Harghita-Bai.

The main selection criteria needed for the two measurement profiles were that they to be normal to the assumed fault directions. First, we attempted to identify the local fault supposed to be represented at the surface by a virtual line connecting the two mofettes. The direction of the supposed local fault strikes 50° to the W from the N direction. Secondly, it was assumed that the major fault system was located along mofettes, and that in Harghita-Bai the mineral springs were aligned, with a strike 5° to the E from N.

Method for measuring radon and thoron concentrations in soil along the selected profiles is presented in details in chapter 2. The depth for sampling the soil gas in case of profile A was of 60 cm, resulted from a local measurement in a vertical profile in soil, from 30 cm to 80 cm depth. Measurement results show that the value of the radon concentration at the depth of 60 cm (4.61 $kBq{\cdot}m^{-3}$) does not change significantly, within errors. In case of profile B, the sampling depth of 40-50 cm was chosen based on soil properties (eg. solid rocks) that prevent penetration of the probe in deeper depths.

Measurements along profile A. In first case (profile A) radon and thoron concentrations were measured in 11 points, in front of the two mofettes (labelled from A1 to A11). The coordinates (latitude and longitude) of these points, recorded by a GPS, are presented in table 4.4, and their locations are shown in fig.4.3. The line connecting the measurement points is normal to the presumed direction of the local fault that connects the two mofettes. The distance between the first point (A1) and the last point (A11) of the profile is 44.3 m, and the average distance between the points is of ~4.4 m.

Table 4.4. The coordinates and altitudes (a.s.l.) of the two mofettes (A and B) and the extremities of the profile A

Points		N	E	Alt.
Mofette A		46°23'19.02"	25°38'20.34"	1298 m
Mofette B		46°23'18.60"	25°38'20.70"	1297 m
Extremty points of the profile A	**A1**	46°23’18.30"	25°38’19.30"	1306 m
	A11	46°23’18.80"	25°38’21.40"	1298 m

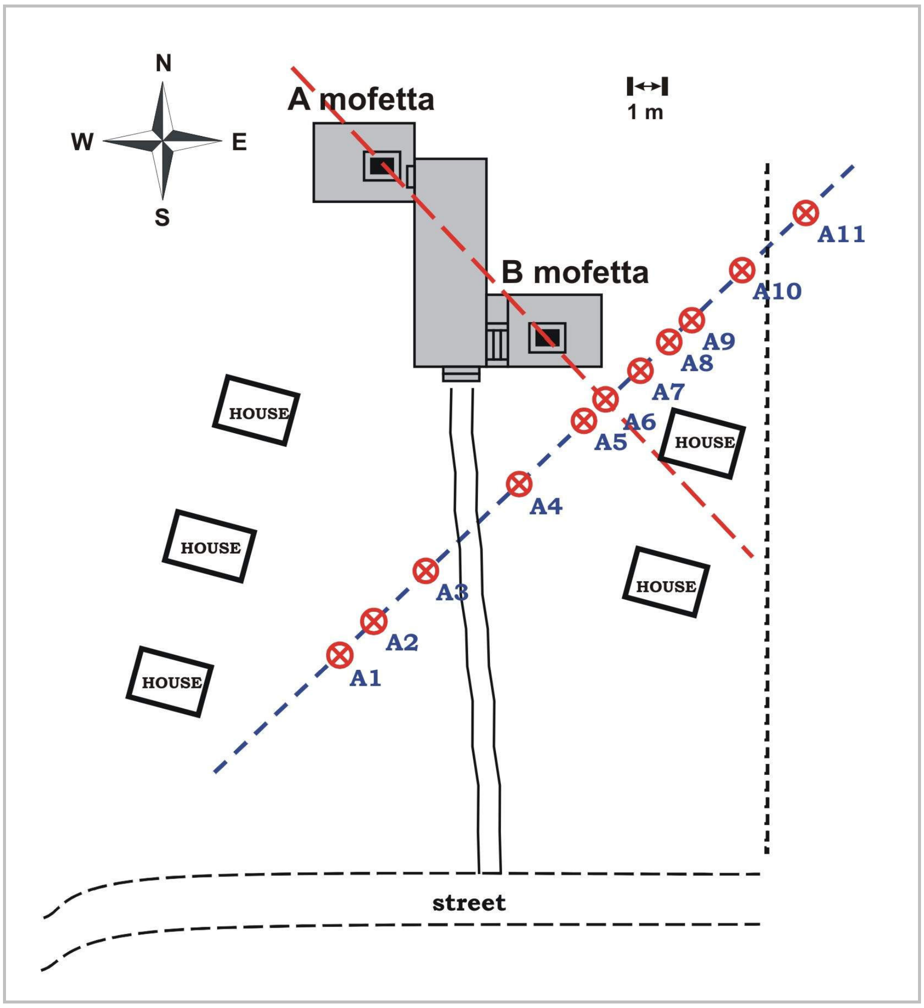

Fig. 4.3. The location of the two mofettes (black rectangles within gray fields) of nearby houses (oblique white rectangles), and the 11 measurement points (circles with interior crosses) aligned normal (dotted line) to the direction connecting the two mofettes (the presumed fault direction, dashed line).

Results of the measurements performed on profile A are presented in fig.4.4 as distribution of the measured radon and thoron concentrations, where the values are in the order of kBq·m^{-3}. The values of radon concentrations have a wide range, between 2.5 and 19.1 kBq·m^{-3}. This range has two extremities (points A1 to A3, and points A10 and A11, respectively) and a middle section (points A4 to A9) showing a distribution with a maximum in point A6 which has a value of 7.0 kBq·m^{-3}. Values of the thoron activity concentration showed a similarly large range, between 0.9 and 18.6 kBq·m^{-3}, those for the radon concentrations, with a maximum in the same point A6, which has a value of 18.6 kBq·m^{-3}.

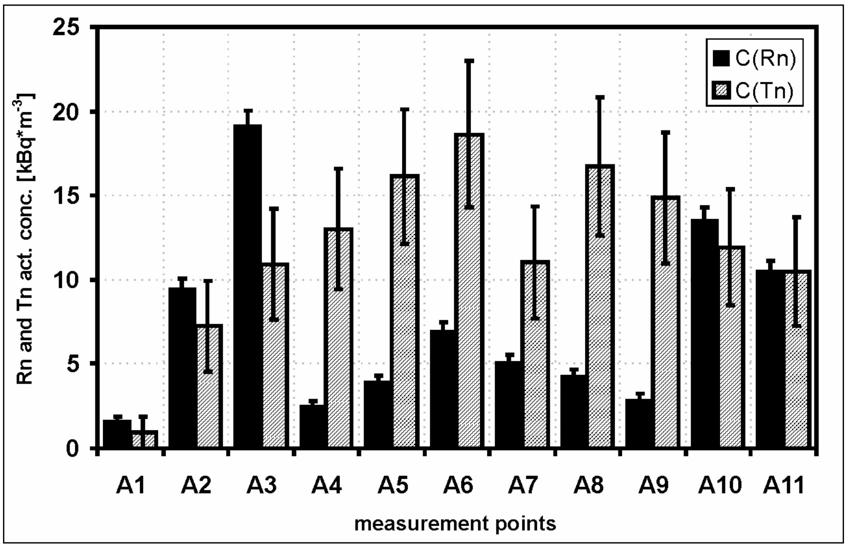

Fig. 4.4. The distribution of the measured radon and thoron concentrations of profile A. Black columns are Rn concentrations, and the patterned columns are the Tn concentrations.

The difference in the order of magnitude between the middle domain and the extremities of the total distribution of profile A is likely caused by the fact that the measurements were performed in different soil horizons, at 60 cm measurement depth from the surface, due to the variable thickness of the upper soil horizon. Therefore, the points A1, A2, A3, A10, and A11 were measured in the same soil horizon (at 62 cm depth of the eluviation horizon), while in points A4 to A9 soil gas was sampled from another soil horizon (the clay-rich or the illuviation horizon). The concentration values of the middle domain relative to the values of the extreme points were influenced by the thickness of the eluviation horizon of the soil. In the case of the middle domain, the thickness of this soil horizon may be much lower than of the extreme points. Thus, the shallower position of the clay-rich horizon and its moisture content agree with the fact that permeability is low and that only a small fraction of the ascending radon gas reached the measurement depth. The 3.84 day half-life of radon may be sufficient to allow it to ascend from below the clay-rich layer.

Since the thoron concentrations had a maximum, it is possible that the moisture content and clay-rich character of the soil horizon in which the measurement was made did not prevent the flow of thoron gas. It is possible also that thoron originated from another (higher) soil level than radon, at all of the measuring points. The reason for this is the much shorter half-life of thoron which results in a shorter migration length compared to radon.

Measurements along profile B. In the case of profile B, radon and thoron concentrations were measured at 5 points (labelled from B1 to B5) that were selected along a line normal to the direction connecting the mofettes, the mineral springs, and spas. The coordinates (latitude and longitude) of the points were recorded by GPS and are presented in table 4.5, and the locations of the points are shown in fig.4.5. The line connecting the points is normal to the direction of the presumed fault zone. The distance between the first point (B1) and the last point (B5) of the profile is 37.2 m, and the average distance between the points is of ~ 9.3 m.

Table 4.5. The coordinates and the elevations (a.s.l.) of the mofette house, the mineral water springs, and the pools in Harghita-Bai, as well as the coordinates of the extremity points of the profile B.

Place		N	E	Alt.
The Mofette house		46°23‘18.30"	25°38‘21.36"	1303 m
Csipike spring		46°23‘10.92"	25°38‘18.78"	1287 m
Vallató spring and spa		46°23‘13.08"	25°38‘18.24"	1293 m
Lobogó spa		46°23‘20.46"	25°38‘21.66"	1288 m
Szemvíz spring		46°23‘22.38"	25°38‘22.44"	1285 m
Extremity points of the profile B	**B1**	25°38‘18.42"	46°23‘16.02"	1291 m
	B5	25°38‘20.22"	46°23‘15.72"	1291 m

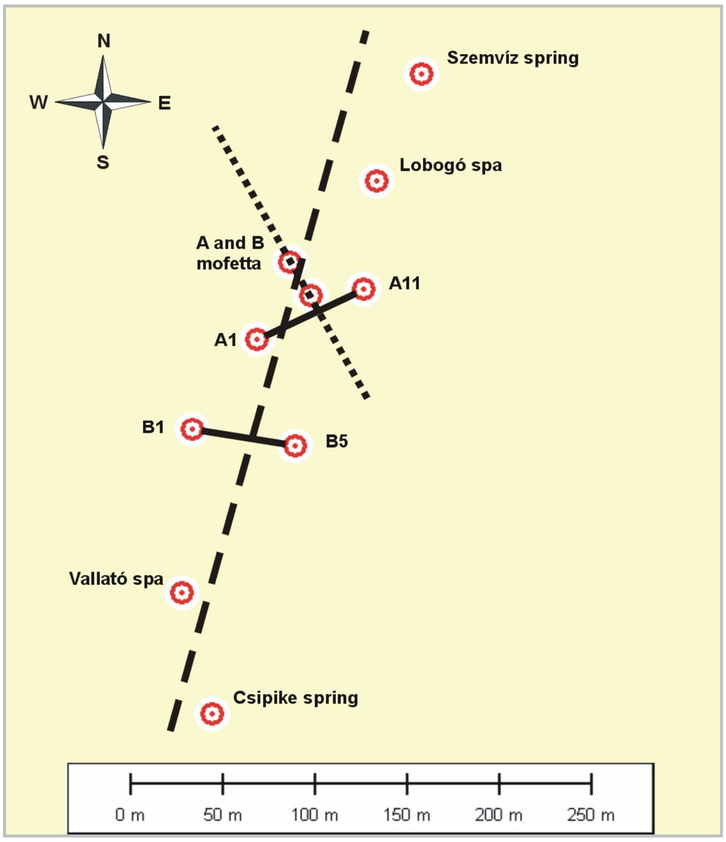

Fig. 4.5. The locations of the two mofettes (A and B), the extremity of the profile A (points A1 and A11), the extremities of the profile B (points B1 and B5), and the locations of the mineral water springs and pools in Harghita-Bai. The dotted line (perpendicular to profile A) is the presumed direction of the local fault, and the broken line (perpendicular to profile B) is the direction of the fault zone.

Results of the measurements performed on profile B are shown in fig.4.6 as distributions of radon and thoron concentrations. It can be seen that the values of the radon concentrations have a wide range, between 1.2 and 30.6 kBq·m^{-3}. The distribution has a maximum in point B2, with a value of 30.6 kBq·m^{-3}. Thoron activity concentrations values have also a wide range, between 7.2 and 38.2 kBq·m^{-3}, as for the radon concentrations. The distribution also contains a maximum in point B3, with a value of 38.2 kBq·m^{-3}.

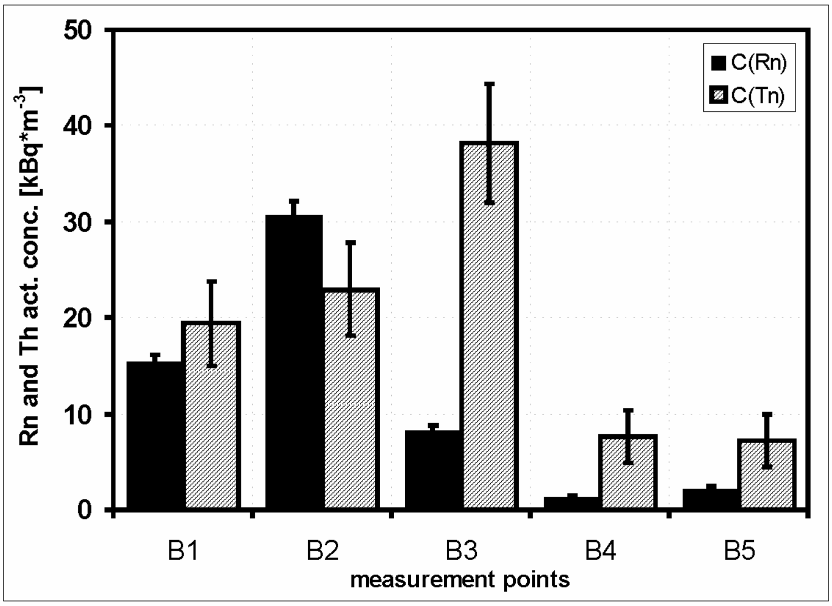

Fig. 4.6. The distribution of the measured radon and thoron concentrations of profile B. The black columns are Rn concentrations, and the pattern columns are Tn concentrations.

In the measurements performed in profile B, the radon and thoron maximum concentrations were found in different measurement points. The radon maximum was found at point B2, while the thoron maximum was measured at point B3. However, none of these points were found at profile extremities. The interpretation is that the fault lies between points B2 and B3, perhaps closer to B3 than to B2 due to the shorter diffusion length of thoron as compared to radon. Another probability is that the fault is inclined, and that the thoron maximum represents its intersection with the surface, while more diffusive radon represents its average location. If so, such measurements may be used to detect fault inclinations as well as their simple presence. In this case, the fault would incline towards the east.

The measurement results along profile B differ notably from those obtained along profile A, since they do not show segmental distribution of radon concentration values. In the case of the profile B the shallower sampling depth (40-50 cm) limited all of the measurements to the same uppermost soil horizon of the, without the influence of the wet and impervious clay-rich soil horizon on radon diffusivity. The more even (roughly horizontal) surface along profile B is associated with a more uniform soil horizon A, while along the inclined topographical surface along profile A the uppermost soil horizon A is of variable thickness.

Acknowledgements. This work was supported by the Institute of Researcher Programmes (KPI) of the Sapientia - Hungarian University of Transylvania (EMTE), through a Project entitled: "*Study of the post volcanic activities in Székely Land, in Transylvania*", 2008-2009.

4.2.2. Radon study on Peceneaga-Camena fault (Dobrogea)

These study present preliminary results of radon measurements at Peceneaga-Camena fault zone (Dobrogea). The aim of the study is to identify and locate the direction of the fault, with radon in soil measurements. Measurements were performed in a village (Fântâna Mare) which stands in the direction of the fault, and where currently is in progress a geodynamics research concerning the displacement of the fault [*Besutiu, Zlagnean, 2009; 2010*].

The Peceneaga-Camena fault (PCF) is one of the most well known regional faults on the Romanian territory, with *direction NW-SE*, and represents the northern limit of the Moesian platform. Several Romanian geophysicists consider *PCF as the plate boundary between the Moesian Microplate and the East European Plate* [*Besutiu, Zlagnean, 2009*]. *PCF crops out along its Dobrogean sector, separating the Upper Proterozoic Green Schist series of Central Dobrogea and the Palaeozoic-Mesozoic deposits of the North Dobrogea folded belt* (fig.4.7) [*Săndulescu, 1994*].

Various geological evidences have been provided to the *PCF active strike-slip activity*. Based on micro-kinematic studies on other faults, a horizontal displacement was estimated of about 3 mm/year [*Săndulescu, 1994*]. In order to clarify the current PCF geodynamic behaviour, the Institute of Geodynamics of the Romanian Academy started a geodetic experiment. On the PCF outcropping segment, at Fântâna Mare village a geodynamic observatory has been installed. In the name of Baspunar Experiment, the observatory has been equipped with two high accuracy Leica-TC-1201 total stations deployed on stable concrete pillars grounded in the Green Schist series of Central Dobrogea, pointing towards two laser reflectors installed on the other flank of PCF, at a distance of 300 m and, 350 m respectively, on the Triassic-Jurassic deposits of North Dobrogea.

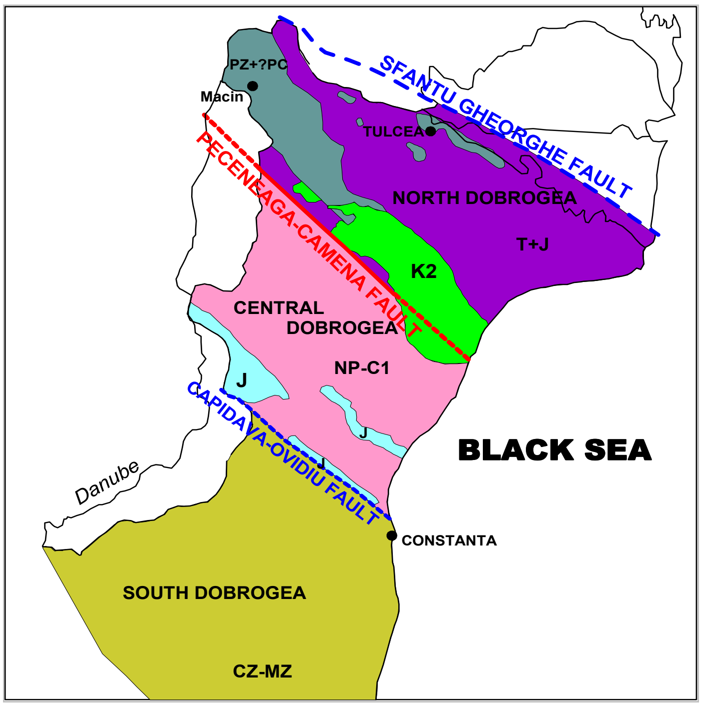

Fig. 4.7. Tectonic sketch of Dobrogea showing Peceneaga-Camena Fault setting. (PZ+?PC - Paleozoic+?Precambrian (Măcin unit); PZ - Paleozoic (Tulcea unit); T+J - Triasic+Jurasic (Tulcea unit); K2 - Upper Cretaceous (Babadag); NP-C1 - Neo-proterozoic- lower Cainozoic (Histria formation)).

Radon measurements on the Peceneaga-Camena fault zone, and results

Radon measurements on the Peceneaga-Camena Fault were performed in 2010 (June and July), in Fântâna-Mare village (Baspunar, Dobrogea), under dry conditions in the zone. The geo-coordinates of the village are: N: 44°51'42", E: 28°29'49", and the altitude between 130-170 m. According to the Baspunar experiment, the Peceneaga-Camena Fault crosses between the location of the mentioned geononamics observatory and the two laser reflectors (mounted on the church and the school of the village).

The applied method of radon in soil measurements is the same as in the case of Harghita-Bai, and is presented in chapter 2. The depth of sampling soil gas was 60 cm. Radon in soil measurements were performed in several places on the one side and other of the fault, by taking as reference points the observatory with the church and the school. The selection criteria of measurement places were that they be within profiles which would be normal to the direction of the fault (NW-SE). In total 50 measurements of radon in soil were performed, grouped in 5 profiles. Because of field conditions, only a part of these measurements are relevant to made representative distributions of radon concentrations. Thus, three sets of data was found, which form three well-defined profiles. For geo-coordinates of the measurement places and of the reference points (observervatory, church and school) we used a GPS.

Coordinates of the three profiles and altitudes are presented in Table 4.6, and are represented on a Google Earth map (see fig.4.8.), with the profiles and the direction of the fault.

Table 4.6. Coordinates and altitude of the reference points (observatory, church and school), and the extremities of the three profiles (A, B and C).

Places		N(gr,min,sec)	E(gr,min,sec)	Alt.(m)
observatory		44° 51' 32.7"	28° 29' 44.5"	154
church		44° 51' 42.9"	28° 29' 37.4"	142
school		44° 51' 41.2"	28° 29' 38.3"	141
Extremties of the profile A	**PA1 (P26)**	44° 51' 42.4"	28° 29' 26.7"	148
	PA5 (P30)	44° 51' 39.2"	28° 29' 25.8"	146
Extremties of the profile B	**PB1 (P9)**	44° 51' 39.8"	28° 29' 35.5"	141
	PB5 (P4)	44° 51' 34.8"	28° 29' 32.9"	159
Extremties of the profile C	**PC1 (P14)**	44° 51' 39.7"	28° 29' 43.4"	132
	PC5 (P15)	44° 51' 34.1"	28° 29' 38.5"	148

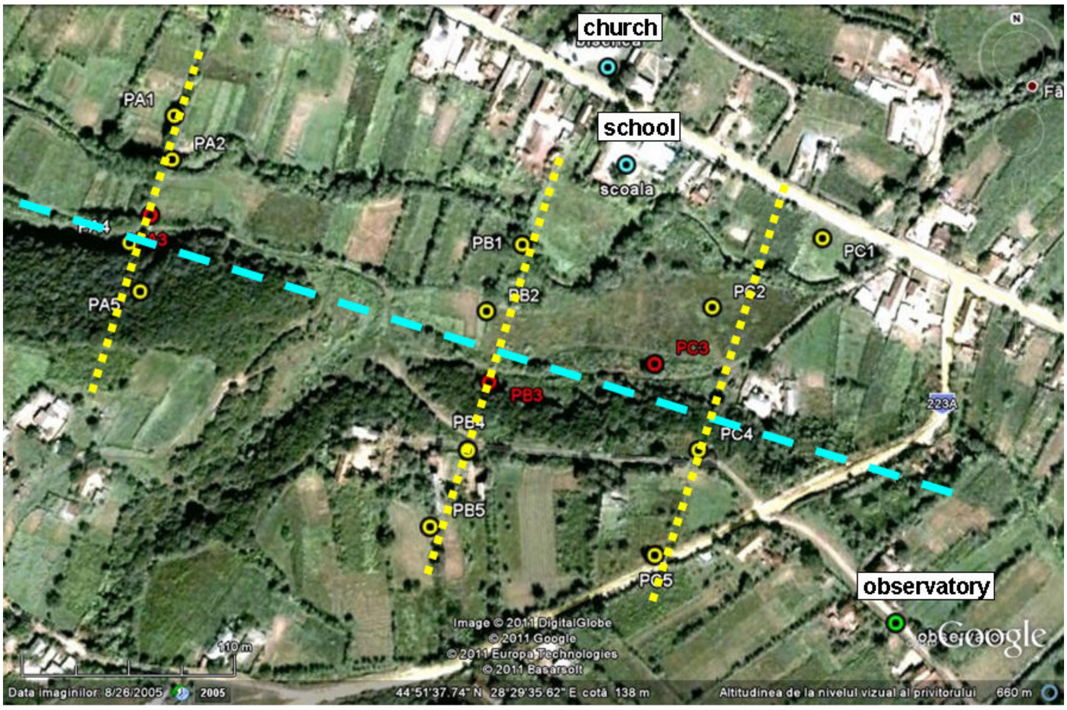

Fig.4.8. Location of measurement places along the three profiles (PA, PB, and PC) with references: observervatory, church and school. The three dotted lines correspond to the measurement profiles, and the normal dashed line corresponds to the direction of the Peceneaga-Camena Fault. This line crosses near the points with maxima of radon concentrations.

Radon concentration values for all the 50 measurements lie within a wide range, between 8.0 and 50.3 kBq·m^{-3}. Values above 50 kBq·m^{-3} are radon anomalies that are representative for fault zones, and radon risk areas. The representations of the measured concentrations are made on the selection of the points that gives distributions with maxima. Thus, the three profiles contain 5 measurement points. The distances between the first and last points of the profile A is 101 m, for profile B is 186 m, and for profile C is 205 m. The distributions of the measured concentrations for the three profiles are shown in fig.4.9.

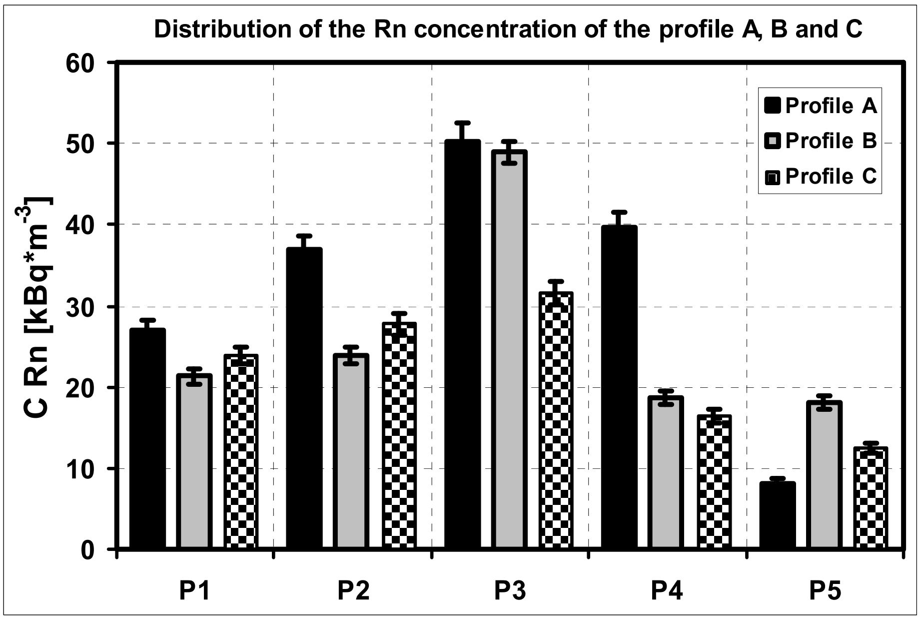

Fig. 4.9. The distributions of the relevant radon concentrations for the three profiles A, B and C, with errors.

Distributions of radon concentrations with a maximum was obtained, for the three profiles. For the profile A, the maximum of concentration is 50.2 kBq m^{-3}, for the profile B the maximum value is 48.9 kBq m^{-3} and for the profile C the maximum is of 31.5 kBq m^{-3}, respectively. These distributions and maxima, shows that for the fault zone the radon concentrations in soil (at a depth of ~ 60cm) are in the range of 20 and of 50 kBq m^{-3}. This seems to be the range for radon concentration values measured in soil on fault zones.

<u>Conclusions</u>

The radon and thoron measurements performed in soil at Harghita-Bai were qualitatively reproducible and have good representations. The measurement results are consistent with both the existence and the presumed position of the NW-SE fault zone that connects the two mofettes. The mofettes and the mineral water springs are indeed connected along a second fault zone that strikes N-NE, as previously stated. The measurement results displayed normal distributions for the activity concentrations of radon and thoron, with one maximum value for both cases. The distributions are consistent with the hypothesis that the main fault line crosses through the point corresponding to maximum values of radon and thoron concentrations. Also it is clear that high precision thoron concentration determinations gave a more accurate location for this type of tectonic feature than radon, as a result of its shorter half-life and its shorter diffusion path of thoron.

Radon measurements carried on Peceneaga-Camena Fault have good representation, which results indicated the existence and position of the fault zones oriented NW-SE. Measurement results shows distributions of the studied profile, with a single maximum of radon concentrations. These distributions are also consistent with the hypothesis that the fault crosses through the points with maximum values of radon concentrations.

4.3. Radon studies with applications in geophysics

The radon (^{222}Rn) presents everywhere in the terrestrial crust, it's concentration in different geological formations depends mainly on geophysical parameters of the environment, like radium (^{226}Ra) content (sometimes uranium), and diffusion parameter. In closed spaces inside geological formations, radon can play a role of *trace monitoring element*, if apply correctly the method to measure it's activity concentration. A measuring method is to follow the accumulation through diffusion, which can be studied in bore-holes, closed and isolated from the atmospheric air exchange. The study of diffusion processes in different geological environments correlated with theoretical models and calculations can give information on the *diffusion length* of radon atoms which is in relation with the *permeability of rock or soil* [***Papp et al., 2004; Papp et al., 2005***].

The study described in this subchapter was my research at Eötvös-Loránd University (ELTE), Budapest, in a previous doctoral stage, between 2001-2004.

4.3.1. The method for determination of geophysical parameters by radon measurements in bore-holes of geological formations

This study proposes a new *method for measuring radon concentration in closed bore holes* of geological formations and to use the results to *estimate the diffusion parameters* of these geological environments [***Papp et al., 2008***].

<u>The aim of the study (work)</u>

The main aim of the present study has been to develop and test a method for an easy estimate of the diffusion and radium content parameters of geological formations using the natural monitoring capacity of radon gas. The basic idea is to follow and determine the accumulation of radon gas, as well as the variation of its concentration in time, in a closed bore-hole created in clay formations or soil. The growth rate of radon concentration in time depends on the diffusion parameter of radon gas in surrounding environments, i.e. on the permeability of the medium.

In order to following the accumulation of radon gas in bore-hole, it is necessary to empty the hole of radon. This can be achieved through a simple cleaning process, by which the air in the bore-hole is changed by a gas with no radon content. Cleaning process can be made by fresh air (with low radon content), or using bottled nitrogen gas (which does not contain radon). However, such cleaning processes influence deeply the radon content inside bore-hole and in the soil layers around the hole. After the cleaning process, the accumulation of radon gas shows a characteristic time dependence, whose growth rate is determined by the diffusion parameter of the surrounding environment [***Papp et al., 2006, Papp et al., 2007***].

<u>Experimental method</u>

To testing the above mentioned method, a site at Mátyáshegy (Budapest) was chosen, where the elevation a.s.l. is relatively high (~150 m), higher than the city itself and the soil is mostly sandy (silty-sand). A bore-hole was made using a geological hand-drill into the ground. The hole had a diameter of of ~ 8.0 cm and ~ 4.5 m depth (~ 23 L volume). The depth of the hole was chosen to be deep enough that the meteorological parameters did not influence the radon concentration in the hole.

<u>Measurements of radon concentrations from bore-hole were made by the Alpha Guard radon monitor</u>. This is a pulse-counting ionization chamber, which monitors continuously radon concentration from bore-hole air. Besides the radon concentration, this device could measure the air pressure, temperature and relative humidity of the environment at the surface. In order to circulate the air and to measure the radon concentration in the bore-hole, we used the Alpha Guard detector with the Alpha Pump device. The draw speed of Alpha Pump was 1 L/min, in flow mode. In these measurements the radon concentrations were measured close to the bottom of the bore-hole, from a depth of ~ 4,5 m. At this depth the values of the concentrations were presumable least influenced by the outside meteorological parameters.

Measurement results

A. The influence of meteorological parameters on the radon concentration

A necessary condition for the radon accumulation measurements through diffusion in bore-holes of geological environments is that the hole was deep, tightly closed and isolated enough that the radon concentration in the hole did not depend on the meteorological factors at the surface. The measurements to determine the dependence of the meteorological factors on the radon concentration in soil air were performed in January and February 2006, where one of the continuous records over a period of 11 days is shown on fig.4.10.

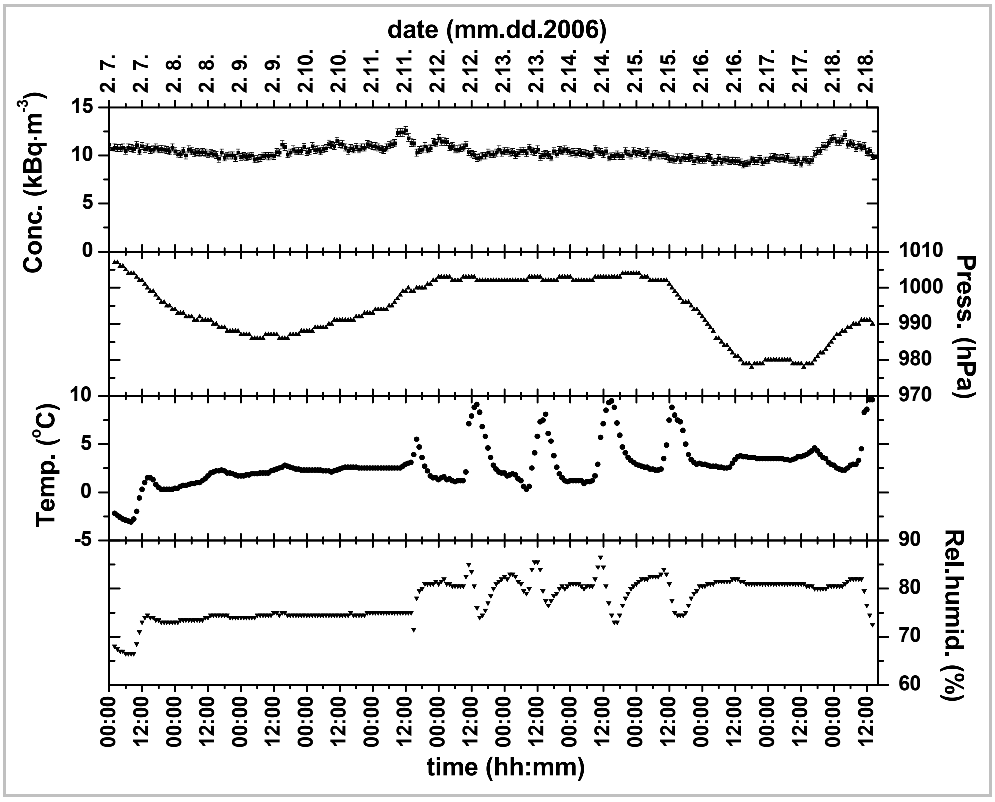

Fig. 4.10. The dependence of the radon concentration (*Conc.*) measured at the bottom point in the bore-hole on the meteorological parameters, air pressure (*p*), temperature (*T*) and relative humidity (*RH*) measured at surface (all measured with Alpha Guard monitor).

According to this period, the average value of the radon concentration close to the bottom of the bore-hole (at the depth of 4.4 m) was 10.4 ± 0.4 kBq·m^{-3}, the minimum value is 9.0 ± 0.3 kBq·m^{-3} and the maximum is 12.6 ± 0.4 kBq·m^{-3}, respectively. During most of the time the radon concentration varied within about 10% of the average value. However, even the values of these periods may be seen as adequate for our purposes, determining of the target parameters, with limited accuracy of 10-20 %.

The radon concentration close to the bottom of the bore-hole and the three meteorological parameters at surface showed only weak correlations. The corresponding correlation coefficients are: $R_{C,P}$ = 0.33 (between concentration and air pressure), $R_{C,RH}$ = -0.21 (between concentration and relative humidity) and $R_{C,T}$ = -0.30 (between concentration and temperature). These results support that the variation of radon concentration is only weekly influenced by the meteorological conditions at the surface.

B. Measurements of the inflow of radon into radon-free bore-hole

In order to study the diffusion phenomena of the radon gas, it is necessary to remove the radon from the bore-hole, through a "cleaning" process of the hole. For this, we used bottled dry nitrogen gas, which did not contain radon. The flow of the nitrogen gas from the bottle was controlled by a pressure regulator. After each cleaning procedure, the time-dependence of the radon concentration was measured for a period of several hours. Measurements were performed with the same Alpha Guard radon monitor. Measurements show that the radon level starts from low level activities and rises to a same saturation equilibrium level in each case. The results of the measurements of characteristic time dependence of radon concentrations are presented in fig.4.11, corresponding to different cleaning processes (by different quanities of nitrogen gas).

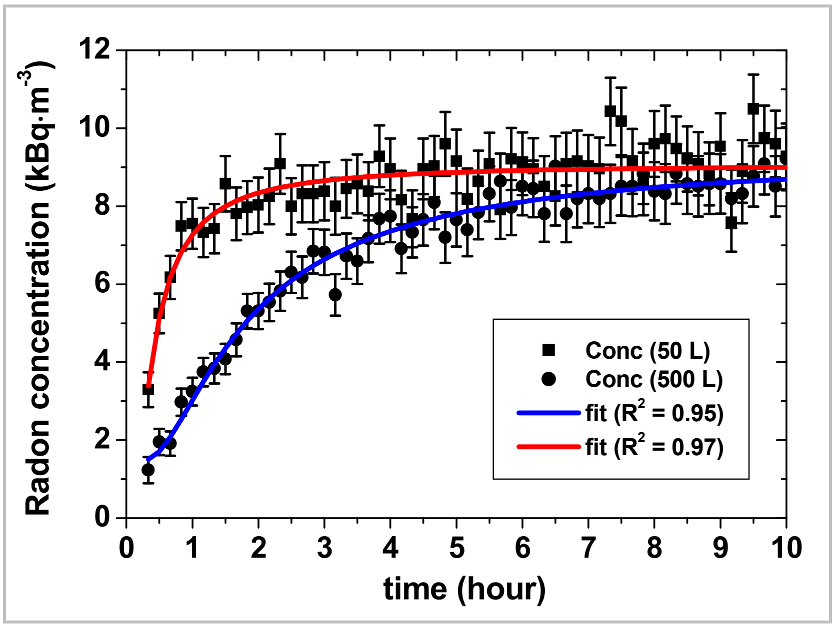

Fig. 4.11. The time-dependence of the radon concentration *C(t)* in the bore-hole (examples). The squares are the measured data corresponding to the 50 L cleaning process and the circles are the measured points for 500 L cleaning process. The continuous solid curves are the fits of the calculated radon concentration to the different measurements (see in text).

To represent the results in a simply way, we parametrized the data by a simple function:

$$C(t) = C_{eq} \cdot [1 - \exp(-k \cdot (t - t_0))] \tag{4.3}$$

where, C_{eq} is the equilibrium radon concentration, k is a parameter defining the rise of the concentration, t is the time, and t_0 is a parameter that takes a possible time-shift in the detector response into account. From the overview of the data, one can see that following the cleaning processes, the k parameter depends on the quantity of the nitrogen gas (V) used at constant pressure (p) (fig.4.12). One can see that the k parameter do not depend on the pressure of the cleaning gas (p), at the same quantities (V), in each case (fig.4.13). The volume-dependence of the k parameter at same pressure shows that larger volumes of nitrogen gas used in the cleaning procedure hardly affect the rate of radon influx any more.

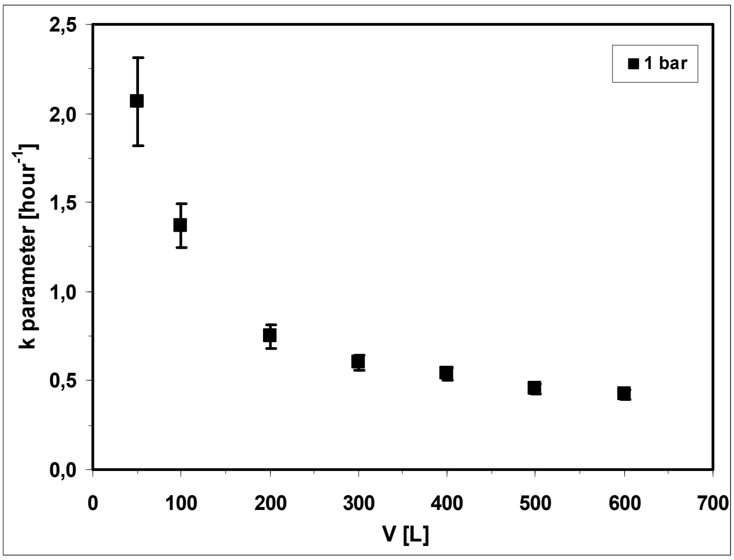

Fig. 4.12. The *k* parameter from fits to a series of radon fill-up curves versus the volume (*V*) of the nitrogen gas used in cleaning procedure (for the same gas pressure of 1 bar).

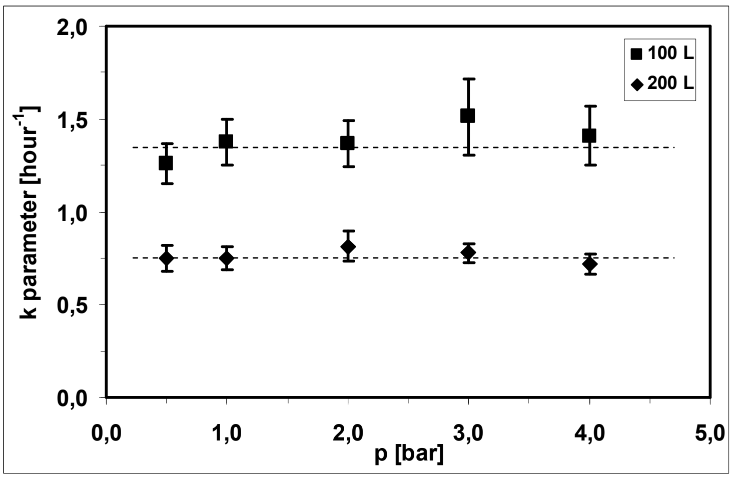

Fig. 4.13. The *k* parameter from fits to a series of radon fill-up curves versus the nitrogen gas pressure (*p*) used in cleaning procedure (for two values of the volume of the gas (*V*)). The square corresponding to 100 L, and the circles corresponding to 200 L of nitrogen gas.

4.3.2. The matematical model for the determination of the geophysical parameters and the evaluation of the results

We suppose that the inflow of radon into the cleaned, radon-free bore-hole is mainly the result of diffusion of this gas from the surrounding geological environment. We suppose that there are basically two types of layers around the bore-hole, so that the cleaning process beside the hole itself has emptied the layer just at the surface of the hole. Beyond that effectively emptied layer, the geological formation contains the homogeneous equilibrium radon concentration.

In order to estimate the level of the radon concentration in the bore-hole in a given time, we have to add the contributions from all parts of the geological environment, i.e. both from the wide surrounding zone that has the equilibrium radon concentration and from the emptied layer.

Therefore, modeling the diffusion of radon gas in a given surrounding geological environment, we use the diffusion equation of radon, taking into account the decay and production of radon [*Andersen, 2000; Cosma et al., 2001*]:

$$\frac{\partial C(\vec{r},t)}{\partial t} = D \cdot \frac{\partial^2 C(\vec{r},t)}{\partial \vec{r}^{\,2}} - \lambda \cdot C(\vec{r},t) + G \qquad (4.4)$$

where, $C(\vec{r},t)$ is the radon concentration depending on $\vec{r}(x,y,z)$ vector and time t, D is the effective diffusion constant, $\lambda = 2.1 \cdot 10^{-6}\ s^{-1}$ is the decay constant of ^{222}Rn, $G = \lambda \cdot C_{eq}$ is the production rate of radon in the pore-air and C_{eq} is the equilibrium radon concentration in soil.

The $C(\vec{r},t)$ solution of this equation gives the contribution of a source of the diffusing radon gas on a chosen small volume with the time. We suppose that each small region in the surrounding environment gives contribution to the bore-hole by diffusion. In order to estimate the radon level in the bore-hole, we add the contributions from each point $\vec{r}(x,y,z)$ of the surrounding geological environment as function of the time [*Koshlyakov et al., 1964*]:

$$p(t) = \frac{1}{(4 \cdot \pi \cdot D \cdot t)^{3/2}} \cdot \int_0^r exp\left[-\left(\frac{x^2 + y^2 + z^2}{4 \cdot D \cdot t} \right) \right] \cdot dx \cdot dy \cdot dz\,. \qquad (4.5)$$

The integration should be done for all points from the edge of the bore-hole, considering the hole in a cylindrical geometry. After calculations, the probability of the contribution *p(t)* will be the following:

$$p(t) = 0,5 \cdot \left[1 - \exp\left(-\frac{u^2}{2} \right) \right] \cdot s(v) \tag{4.6}$$

where the parameters $u = \frac{r}{\sqrt{2 \cdot D \cdot t}}$ and $v = \frac{h}{\sqrt{4 \cdot D \cdot t}}$ depends on the thickness of the emptied layer in cylindrical geometry (in which r is the radius and h is the height of the cylinder) and considering $r \approx h$. The parameter $s(v)$ is approximated by an <u>*error function*</u> in the form: $s(v) = erf(v) = 1 - \left(1 + a_1 \cdot v^1 + a_2 \cdot v^2 + a_3 \cdot v^3 + a_4 \cdot v^4\right)^{-4}$ where, a_1 = 0.278393; a_2 = 0.230389; a_3 = 0.000972 and a_4 = 0.078108 [*Abramowitz and Stegun, 1964*].

It turns out that the result of the integration depends basically on the ratio $q_i = r_i / \sqrt{D}$, where r_i is the sum of the radius of the bore-hole (r_0) and effective thickness of the emptied layer (Δr_i) in the i-th cleaning process ($r_i = r_0 + \Delta r_i$), and D is the diffusion parameter. In addition, *p(t)* is the parameter which scales the absolute value of the C_{eq} radon concentration to the measured value $C(\vec{r}, t)$:

$$\boxed{C(\vec{r}, t) = \left(C_{eq} - C_0\right) \cdot \left[1 - 2 \cdot p(t) \cdot e^{-\lambda \cdot t}\right] + C_0} \tag{4.7}$$

where, C_{eq} is the equilibrium radon concentration and C_0 is the initial level of radon, which incidentally was not emptied from the bore-hole in the beginning of the measurements.

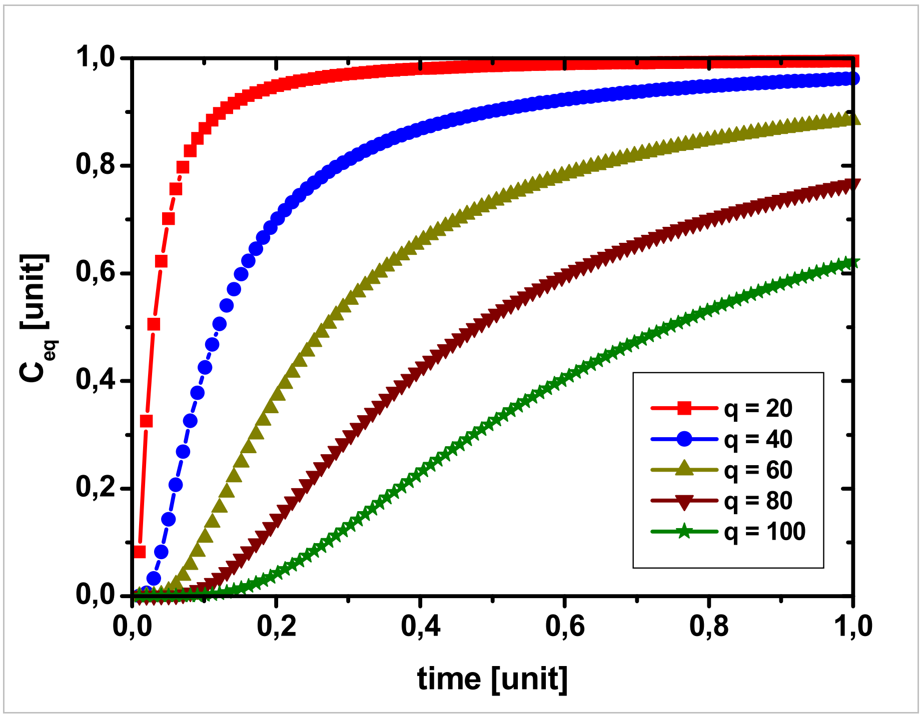

Fig. 4.14. The distribution of the concentrations (*C(t)*) from the diffusion model of radon gas. Different curves corresponds to different values of the parameter q_i determined from the effective thickness of the emptied layer (*Δr_i*), where the initial concentrations C_0=0.

The results of the calculations following the fitting of the final equation (4.7) to the measured radon concentrations series are shown in fig.4.11, for the two examples described above. In each case of the cleaning process we determined the parameter $q_i = r_i / \sqrt{D}$ that practically depends on the thickness of the cleaned layer (Δr_i) and on the diffusion coefficient (D). The dependence of the parameter (q_i) on the volume of the nitrogen gas used in cleaning processes (V_i) at constant pressure is shown in fig.4.15.

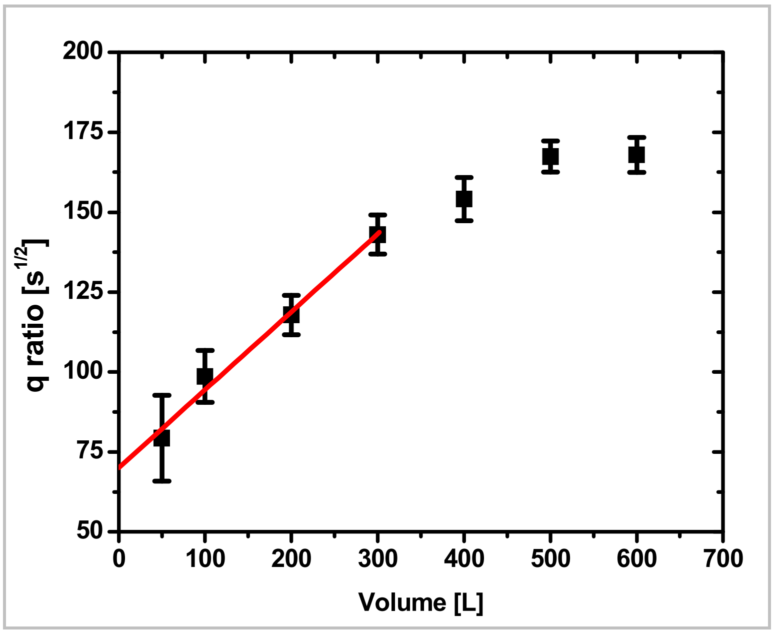

Fig. 4.15. The dependence of the $q = r/\sqrt{D}$ parameter on the volume of the cleaning process (V). The solid line represents the extrapolation of the data for a volume 0.

In order to determine the D diffusion parameter, we should definitively know the r_i value at least in a single case. Of course, none of the measured cases is identical with the ideal case (in which the air in the hole does not contain any radon while the walls of the hole have equilibrium radon content), which could tell the real value of D. However, if we could find the value of the $q_i = r_i / \sqrt{D}$ parameter with no emptied layer (where $r_i = r_0$ the radius of the bore-hole), we could determine D. Such a case is experimentally not achievable because it would mean a sudden change of the air in the bore-hole with radon-free air without pumping anything. Nevertheless, we could make an extrapolation of the results in fig.4.15 for volume $V \to 0$ of the cleaning gas, therefore for the case $r_i \to r_0$. The smooth change of the $q = r/\sqrt{D}$ suggests that the real value may not be far from the extrapolated curve around the volume $V = 0$, i.e. $q_0 = \left(r/\sqrt{D}\right)_{(0)} = (70{,}0 \pm 3{,}6)\sqrt{\text{s}}$.

As the nominal value of the bore-head was 4.0 cm in radius, we think that the effective value of the radius was a bit more than the nominal value, close to **4.4 cm**. Thus, the corresponding value of the diffusion parameter is: $\boldsymbol{D = (3,9 \pm 0,4)\cdot 10^{-7}\ m^2\cdot s^{-1}}$. This value of diffusion parameter agrees well with values of other methods applied for the case of *fine silty-sandy soil type* [*Yu et al., 1993; Nazaroff and Nero, 1988*].

Conclusions

We have shown that in a closed bore-hole of several meters depth the radon concentration measurements are stable and reproducible up to ~10% under different meteorological conditions. This fact allows the introduction of a simple method for the estimation of the diffusion parameter of the surrounding geological environment. By cleaning the bore-hole from radon and measure the influx of radon, the rising of the concentration is determined mainly by the diffusion process of radon gas from the surrounding geological environment.

The experimental data were compared with a model that took the general laws of diffusion into account. The calculations have described the experimental time-dependence functions in the framework of a consistent picture rather well and gave a quick estimation for the diffusion parameter of the geological environment, by an uncertainty of 20 %.

In so far as the measurements and theoretical calculations are *reproductible* in geological environments by different permeability, this method may have practical application in the *developement of the final geological depository of radioactive materials* by determining the gas permeability of rocks.

Acknowledgements. This work was supported by the *Agency for Research Fund Management and Research Exploitation* (KPI) of the Hungarian State (contract GVOP-3.2.1. 2004-04-0233/3.0).

5. SUMMARY AND CONCLUSIONS

The thesis includes three aspects of radon in soil measurements. Mathematical treatment of radon migration in the soil and the transport to the atmosphere show that diffusion process is the dominant mechanism of radon transport from soil to the surface and to the atmosphere. For radon transport from building subsoil to indoor, advection process is the dominant mechanism.

The applied method for measuring radon in soil is simple, efficient and easy to use, which is based on the sampling of soil gas and measuring radon concentration by LUK3C scintillation detector (with Lucas cells). Measurement method was tested at an International Intercomparison Exercise of Radon Measurements in soil (RIM 2010), at three reference sites in Czech Republic (RIM, 2010). The results and statistical tests shows good agreements of the data measured by our group, with the data of other participant groups, and of the average values of the database of reference sites. Comparisons fulfil the test criteria and the values estimated by our group are well acceptable, with a relative ratio of 0.997 between the determination of our group and of the other participants.

Soil permeability is an important parameter in transport processes of radon in the soil, which greatly influences the radon flux or exhalation from soil. In situ permeability measurements can be made with a special method, by measuring the flow rate of a quantity of water from a plastic bottle, connected directly to the soil probe. According to the method, the permeability parameter depends on the ratio of water flow rate relative to the negative pressure created in the bottle ($k \sim q/p$), and depends on the shape factor of the soil probe. The measuring range of the soil permeability by this method is from very high permeability value ($5{\cdot}10^{-8}$ m^2), to ultra low permeability value ($7{\cdot}10^{-14}$ m^2). As a result of comparison of the special instument with Radon-Jok permeameter, the tested parameters (q/p) are in good correlation within errors, thus measurements performed by this instrument are reliable.

One of the use of **applications of radon in soil studies in environmental science** is the risk assessment of radon in soil. The area of interest is Baita-Stei (near Baita uranium mine) that was classified as a "**radon-prone area**", where radon concentrations in air and inside the houses reaches high levels (over 13% of the investigated houses have concentrations higher than 400 Bq·m^{-3}).

- This study is based on the results of integrated indoor radon measurements performed in 335 dwellings from Băița-Ştei, which results indicate an annual average value of indoor radon concentrations of about **343.5 Bq·m^{-3}**, which is 4.16 times higher than the average indoor radon concentration of 82.5 Bq·m^{-3}estimated for Transylvania. The distribution of indoor radon values shows that there are two independent radon sources for the houses in the area. The first source coming from soil and normal building materials (90 % of the houses with radon levels less than 400 Bq·m^{-3}) and the second is coming from uranium waste used in building constructions (6 % of the houses with radon levels higher than 400 Bq·m^{-3}).
- For characterizing soil as main source of indoor radon, were performed radon in soil and permeability of soil measurements at 10 selected sites in Stei-Baita area. The risk assessment of radon from soil is based on the determination of radon potential, by measuring soil gas radon concentrations and the permeability of soil [***Papp et al., 2009****;* ***Papp et al., 2010****; Cosma,* ***Papp, et al., 2010***]. Measured values of soil gas radon concentrations are in a wide range between **5.5 and 512.0 kBq·m^{-3}** and the values of soil permeability are from **5·10^{-8} m^{2}** (low permeability) and **2.8·10^{-13} m^{2}** (high permeability)**.** The measurement results and determination of radon potentials of soils provide that the majority (80%) of the investigated places shown high radon risk. These places are grasslands or building subsoils.
- For characterizing building material as the second main source of the elevated indoor radon in the area, radon exhalation measurements on a sample of sandy gravel and stone mixture, coming from Criş-Băița river were performed. Measurement results show that exhalation from the fraction of sand and gravel is ten times higher than exhalation from stone. Although, this material has an important contribution to public exposure in houses that use this as building material.

Applications of radon in soil studies in geology leading to identify tectonic faults and to locate its direction, by the role of trace element of radon and thoron. Faults serving as pathways for the ascendent migration of the gases towards surface, and can be identified by detecting high radon and thoron activities in the soil. Detection of high thoron activities may indicate fast migration mechanism considering the short half-life of thoron (55 sec) than of radon (3,82 days), which is possible only in presence of a carrier gas (CO_2) as typically occurs in fault zones. Gas-emitting tectonic structures can be mapped as a combination of radon and thoron measurements, supported by CO_2 measurements [***Papp et al., 2010***].

- The radon and thoron measurements performed in soil at Harghita-Bai are consistent on the one hand with the existence and the presumed position of the NW-SE fault zone that connects the two mofettes in the zone, and on the other hand the measurement shows that the mofettes and the mineral springs aligns along a direction oriented to N-NE, aross the major fault zone. The measurement results show distributions for the activity concentrations of radon and thoron, with one maximum value for both cases. The distributions are consistent with the hypothesis that the main fault line crosses through the point corresponding to maximum values of radon and thoron concentrations. Also it is clear that high precision thoron concentration determinations gave a more accurate location for this type of tectonic feature than radon, as a result of shorter half-life and shorter diffusion path of thoron [***Papp et al., 2010***].
- Radon measurements carried on Peceneaga-Camena Fault (Dobrogea) where currently a geodynamics research concerning the displacement of the fault is in progress, have good representation. Measurement results show distributions of radon concentrations with a single maximum, for the studied profiles. Therefore, the measurement results show the existence and the position of a fault zone directed to NW-SE. These distributions are also consistent with the hypothesis that the fault crosses through the points with maximum values of radon concentrations [*Cosma,* ***Papp, et al., in press 2***].

Radon studies with applications in geophysics can be use for the determination of geophysical parameters of geological environments, by radon monitoring in bore-holes of geological formations. In closed spaces inside geological formations, radon can play a role of *monitoring element*, if apply correctly the method to measure it's activity concentration. A measuring method is to follow the diffusion through accumulation, which can be studied in bore-holes, closed and isolated from the atmospheric air exchange. The study of diffusion processes in different geological environments correlated with theoretical model calculations can give information on the *diffusion parameter* of radon atoms which is in relation with the *permeability of rock or soil* [***Papp et al., 2004; Papp et al., 2005; Papp et al., 2008***].

- In a closed bore-hole of several meters depth the radon concentration measurements are stable and reproducible up to ~10% under different meteorological conditions.

This fact allows *the introduction of a simple method for the estimation of the diffusion parameter of the surrounding geological environment*. By cleaning the bore-hole from radon and measure the influx of radon, the rising of the concentration is determined mainly by the diffusion process of radon gas in the geological environment.

- The experimental data were compared with a model that took into account the diffusion, the decay and the generation of radon. The calculations show that the model of the cleaned bore-hole followed by the diffusion model of radon gas in the surrounding environment is valid and describe well the accumulation of radon gas in the bore-hole. The calculations give estimation for the diffusion parameter of the geological environment, with an error of ~ 20%. In other hand, the equilibrium radon concentration gives estimation for the radium content of the geological environment, with a precision of ~ 15 %.
- The applied method was verified in other two bore-holes, where results are consistent by the measurements from the first tested bore-hole. In so far as the measurements and theoretical calculations are *reproducible* in geological environments by different permeability, *this method may have practical application in determining the gas permeability of rocks. This application can have an importance in the development of the final geological depository of radioactive materials.*

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