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A new method for the determination of geophysical parameters by radon concentration measurements in bore-hole

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ABSTRACT

We propose a new method to measure the ^{222}Rn concentration in a closed bore-hole and to use the results for estimation of the diffusion parameter and the average radium content of the surrounding geological formations. In a closed bore-hole, only several meters from the surface, the radon concentration is rather constant (in the ±15% range) under different meteorological conditions. The inflow of radon gas, after removing the radon from the bore-hole by dry nitrogen, shows characteristic time-dependence, which is determined by the diffusion parameter for radon in the surrounding environment. The experimental data were well described by a straightforward model calculation. From the results estimate can be given for the diffusion parameter and for the average radium content of the surrounding geological formation.

1. Introduction

Radon (^{222}Rn) is one of the prominent members of the uranium (^{238}U) radioactive decay chain with half-life of 3.8 days. As is uranium, radon is present everywhere in the terrestrial crust. Its concentration in the geological formations is determined by the abundance of its mother nuclide ^{226}Ra, by the migration (by diffusion or any kind of convection) of the noble gas radon and by its decay rate. The physical and chemical properties of radon enable the gas to migrate varying distances in the porous media. Of course, close to the surface, the radon may escape into the atmosphere (Nero, 1989).

The processes that influence the migration of radon to the surface relate to the configuration and size of free spaces in the soil or rock. The diffusion constant in crystallized solid materials is around $\sim 10^{-12}$ m^2 s^{-1} (Keller et al., 2001). This value is so small that the radon practically never gets out from the grains. When the medium is porous, the value of effective diffusion-coefficient varies between about 10^{-8} m^2 s^{-1} and 10^{-6} m^2 s^{-1} for the different media. Therefore, the radon atoms tend to migrate from some cm to some tenths of cm distances (between ~5 cm and ~70 cm) during their life-times (Duenas et al., 1996). If there is an adequate pressure difference around the given location, then deeper layers may contribute to the radon concentrations (Fujiyoshi et al., 2006; Iakovleva and Ryzhakova, 2003).

The migration or transport of radon atoms from the formation site into the atmosphere depends mainly on the following factors: the medium porosity, the moisture content of soil or rock, the pressure difference of soil air and atmospheric air, as well as the temperature. Secondary factors, which can also play roles are wind speed, flux caused by inversion of temperature, and rapid variation of atmospheric pressure. A better understanding of these processes may help to estimate the indoor radon levels in homes (Hámori et al., 2006; Montero et al., 2005), to create a potential radon map of different regions (Kemski et al., 2001), to study the potential source minerals of radon anomaly (Burján et al., 2002), and to try to predict earthquakes (Zmazek et al., 2003; Richon et al., 2007). The common difficulty of all these investigations is that the measurements are mostly difficult to reproduce.

This paper suggests radon concentration measurements of different types. We intend to use the property of radon that it is always and everywhere present and its concentration inside geological formations depends mainly on geophysical parameters of the surrounding, like radium (sometimes uranium) content and the diffusion parameter. In a closed hole inside a geological formation, the ^{222}Rn concentration could monitor the ^{226}Ra content of rocks. Furthermore, if one creates such environments where the diffusion of radon gas is forced by concentration difference, the rate of diffusion may monitor the migration process in deeper caverns or bore-holes closed from environmental air exchange (Shweikani and Hushari, 2005). In this way, by the comparison of the time-dependence of the radon levels in geological formations, simple

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model calculations can provide information on the two geophysical parameters as mentioned above: (1) on the diffusion parameter of ^{222}Rn atoms which is suggested to be in strong correlation with the permeability of rock or soil (Surbeck, 1993) and (2) on the average radium content of soil or rocks (Przylibski, 2004).

The regular observation of the permeability is a prominent task in environmental monitoring practice as well as in hydrogeology. As this parameter influences the flows occurring in the soil or rock to a great extent, it is important to know its characteristics when planning a depository for hazardous chemical or radioactive waste. In hydrogeology, the permeability is one of the important parameters while searching for potential drinking water sources, where pollution from the surface should be avoided (Lafhaj and Shahrour, 2007). In addition, monitoring the radon level in a hole and using a proper transport model of this radioactive noble gas for diffusion and convection could make it possible to study volcanic eruptions (Gasparini and Mantovani, 1978) and to predict earthquake and seismic crisis (King et al., 1993; Monnin and Seidel, 1998). The main aim of the present work has been to develop and test a method for an easy estimate of the diffusion and radium content parameters using the natural monitoring capacity of radon gas.

2. Description of the method

2.1. The aim of the work

The basic idea is that we can 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. In cases where the formation is clayish, the diffusion is presumable small, while in cases where the medium is coarser-grained sand, the diffusion is greater. In both cases, the radon concentration in the bore-hole obviously tends to reach an equilibrium level in time. The change 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 study the inflow of radon into the bore-hole it is necessary to remove the radon from it. This can be done by a “cleaning” process: one has to change the air in the bore-hole for some gas having no radon content. This cleaning can be performed on several ways, such as using nitrogen. However, it is clear that such a cleaning process may influence the radon content of the surface layers of the bore-hole in some depth, as well.

2.2. The experimental procedure

We prepared a bore-hole in soil at Mátyáshegy (a suburb area of Budapest), where the elevation over sea level is relatively high (~150 m), higher than the city itself. At the chosen place the soil consists mostly of sand. A bore-hole was made using a geological hand-drill. The hole had a diameter of ~8.0 cm and ~4.5 m depth (~23 L volume). The depth of the hole was chosen in to be deep enough that the meteorological parameters did not influence the radon concentration in the hole more than the required overall precision of the measurement (see later). The drill core consisted of seven soil layers from gravel rubble and sand to clay.

For the collection and measurement of the radon gas inside the hole we used a tube system (see Fig. 1). The system consisted of three tubes, where the #1 and #2 tubes were the bore-hole air sampling tubes. These had the same length (440 cm). The #3 tube was the cleaning tube (10-cm length) and it was used to empty the radon gas from the bore-hole. In order to avoid distortions of the system close to the surface, an aluminum cylinder of 8 cm diameter and 100 cm length surrounded the pipe-system. This cylinder was pushed into the soil to 90 cm and on top it was closed with an aluminum cap. This cap fixed the tube system and prevented the rapid change of the air in the hole. Each of three pipes had its own valve. The pipes collected and recirculated the air from the 4.5-m depth.

We measured the radon concentration of air from the bore-hole with an Alpha Guard Radon Monitor. This is a pulse-counting ionization chamber, which is suitable for continuous radon concentration monitoring. This device determines with high efficiency the radon concentrations directly from the alpha decay of radon. The detector had high sensitivity and reasonable response in time to the concentration. In contrast to other types of active radon-detectors, temperature and relative humidity do not influence the sensitivity of this detector (Vargas and Ortega, 2006). Besides the radon concentration, this device could measure the air pressure, temperature and relative humidity of the environment at the surface (Imme et al., 2006).

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. Plastic tubes connected pipes #1 and #2 of the tube system to the detector (Fig. 1). The draw speed of Alpha Pump inflow mode was 1 L/min. In these experiments the radon concentration values were determined close to the bottom of the bore-hole, as at this point the values were presumable least influenced by the outside meteorological parameters.

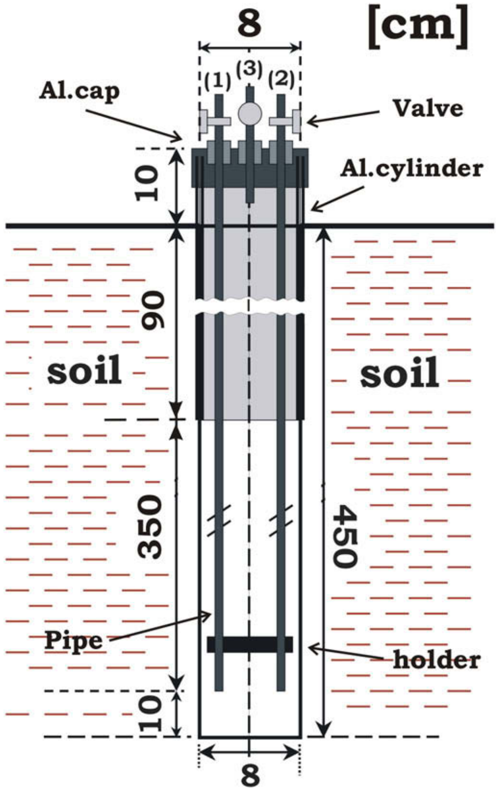

Fig. 1. The schematic view of the bore-hole with the pipes used in the measurements.

2.3. The experimental results

2.3.1. The influence of meteorological parameters on the radon concentration

First, we had to show that the hole was deep and insulated enough that the radon concentration in the hole did not depend essentially on the meteorological parameters at the surface. The measurements for the radon concentration in soil–air and the investigation of its dependence on the meteorological parameters were performed in January and February 2006. In one of the runs we took data continuously for 11 days. The measured parameters are shown in Fig. 2 for the whole period of this experiment. The radon concentration at the location of the measurement (i.e. close to the bottom of the bore-hole (Fig. 1)) 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 in time is only weakly influenced by the meteorological conditions at the surface.

According to these measurements, the average value of the radon concentration close to the bottom of the bore-hole (4.4 m) was (10.4 ± 0.4) kBq m^{-3}. The minimum and the maximum levels – taking the whole period into account – were (9.0 ± 0.3) kBq m^{-3} and (12.6 ± 0.4) kBq m^{-3}, respectively. During most of the time the radon concentrations varied within about 10% of the average value. There were two shorter periods, when the radon levels were about 20% higher than average. However, even the values of these periods may be seen as adequate for our purposes: determination of the targeted parameters with limited (10–20%) accuracy.

2.3.2. 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 (i.e. “cleaning” the bore-hole from radon) before starting the measurements. For the cleaning procedure, we used bottled dry nitrogen gas, which did not contain any radon gas.

The time-dependence of the concentration of the radon gas in the bore-hole after most of the radon was removed from it was measured by the same Alpha Guard Radon Monitor. A regulator controlled the flow-pressure of the nitrogen gas from the bottle. The regulator outlet was connected to both the #1 and the #2 sampling tubes (Fig. 1). Through these tubes the nitrogen gas could get to the bottom of the hole. During the flushes the cleaning valve (#3) was open. In the process, the

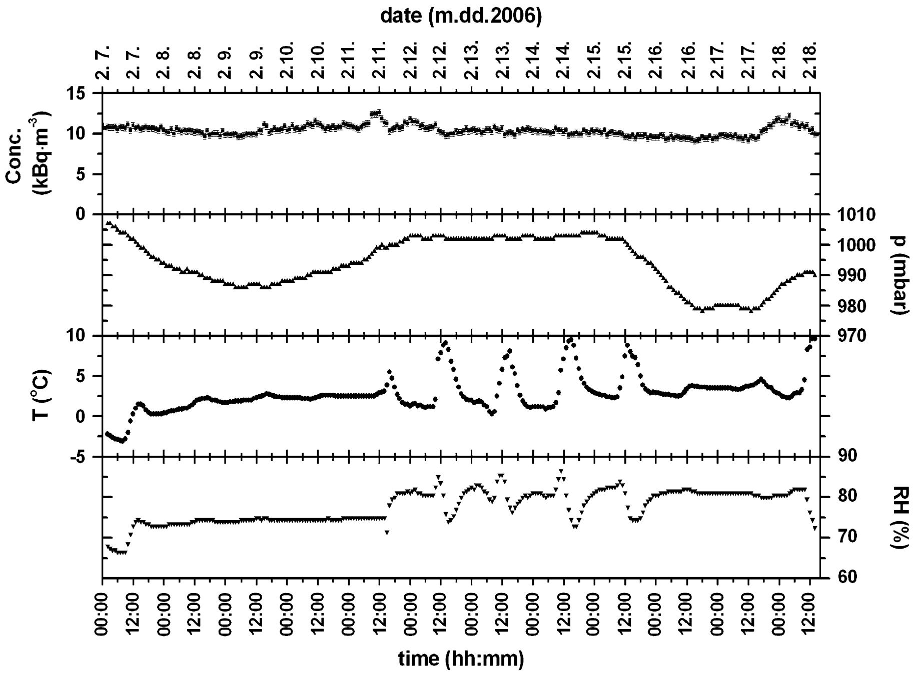

Fig. 2. The dependence of the radon concentration on the time (Conc.) measured at the bottom point in the bore-hole and the air pressure (*p*), temperature (*T*) and relative humidity (RH) measured at the surface (all, measured with Alpha Guard).

nitrogen gas pushed the radon-filled air out of the hole. The air leaves the hole through the #3 cleaning tube (Fig. 1).

After each cleaning procedure, the time-dependence of the radon concentration was measured for several hours. The radon gets in the hole from the surrounding environment, mainly through the wall surfaces of the hole. We took data in 10-min intervals. The measurements show that the radon level starts, as it should, from low-level activities and rises to a similar saturation equilibrium level in each case. These levels were inside of ~20% scatter range, similar to the long-run observations (Fig. 2).

Typical measurements for time-dependence of the radon concentration are shown in Fig. 3 corresponding to two different cleaning processes with different quantities of nitrogen gas. It can be seen that the time needed to fill the emptied hole with radon is in the range of several hours. Therefore, the sampling in each 10-min interval may be a good choice.

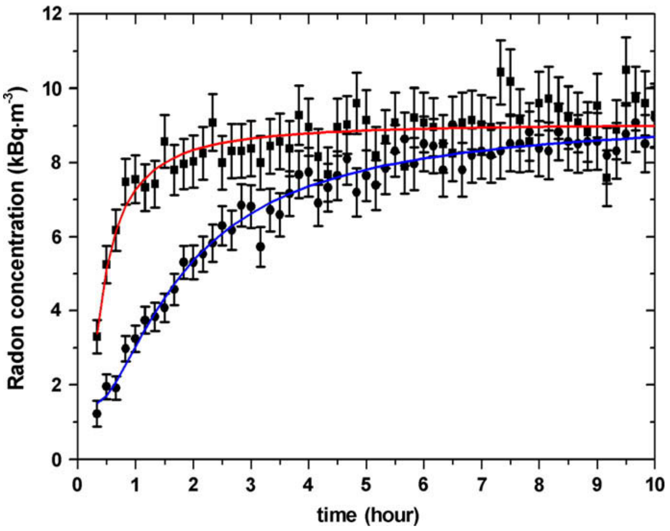

Fig. 3. Two examples for the time-dependence of the radon concentration in the bore-hole. The squares are the measured data corresponding to the 50 L cleaning process and the circles are the measured points for 500 L cleaning process. The continuous solid curves are the fits of the calculated radon concentration to the different measurements (see text). The error bars show the one-sigma level uncertainties.

Just only to present the results in a simple way, we parameterized the data by a simple function:

$$C(t) = C_{\mathrm{eq}}\left[1 - \mathrm{e}^{(-k(t+t_0))}\right].$$

Here C_{eq} is the equilibrium radon concentration, k is a parameter defining the rise, t is the time, and t_0 is a parameter that takes a possible time-shift in the detector response into account. This form described the data well in all cases. The curves in Fig. 3 are the results of the calculations, which are explained later. However, they are indistinguishable from the fits by this simple function.

From the overview of the data, one can see that the k parameter depends on the quantity of the nitrogen gas used at constant pressure for removing the radon from the bore-hole (Fig. 4). The volume-dependence at the same pressure of the k parameter in Fig. 4 suggests that larger volumes of nitrogen used in the cleaning procedure may tend to a saturation value in the sense that even larger cleaning volumes hardly affect the rate of radon influx any more.

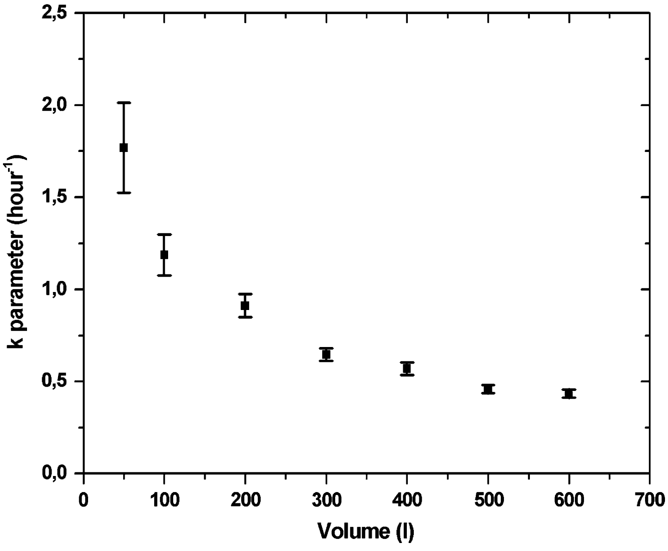

Fig. 4. The *k* parameters from fits to a series of radon fill-up curves versus the volume (quantity) of the nitrogen gas used in cleaning procedure.

3. Discussion

We suppose that the influx of radon into the cleaned, radon-free bore-hole is mainly the result of diffusion of this noble gas from the surrounding geological formation. We suppose that there are basically two types of layers around the bore-hole. The cleaning process itself has emptied the soil layer just around 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 formation, i.e. both from the wide surrounding zone that has the equilibrium radon concentration and from the emptied layer. Calculating the time-dependence of radon concentration in the bore-hole, we shall not take one fact into account. We suppose that the radon, which reached the hole, will stay there. This assumption may have effects much smaller than the precision of the method.

Therefore, modeling the diffusion of radon gas in a given surrounding geological formation, we use the diffusion equation of radon, and in addition we take decay and production of radon into account (Cosma et al., 2001; Andersen, 2000):

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

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

The $C(\vec{r},t)$ solution of this equation gives the contribution of a point 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 p from each point $\vec{r}(x,y,z)$ of the surrounding geological environment as function of the time (Koshlyakov et al., 1964):

$$p(t) = \frac{1}{(4\pi Dt)^{3/2}}\int_0^r \exp\left[-\left(\frac{x^2+y^2+z^2}{4Dt}\right)\right] \mathrm{d}x\,\mathrm{d}y\,\mathrm{d}z.$$

The integration should be done for all points from the edge of the bore-hole. This is a straightforward procedure (Abramowitz and Stegun, 1964), and it does not contain any kind of new physical idea. It turns out that the result of the integration depends basically on the ratio of $q_i = r_i/\sqrt{D}$, where r_i is the sum of the radius of the bore-hole and effective thickness of the emptied layer in the ith cleaning process. In addition, $p(t)$ is the parameter which scales the absolute value of the C_{eq} radon concentration to the measured value $C(\vec{r},t)$:

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

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. This transparent model leads to a good description of the data. The curves in Fig. 3 show the results of the calculations of the same two examples of data as described above.

In each case of the cleaning procedures, we determined the $r_i/\sqrt{D}$ parameter, which was basically defined by the sum of the radius of the hole and the thickness of the emptied layer and the D diffusion. Fig. 5 shows its dependence on the volume of nitrogen gas used in the cleaning process at constant flow. The comparisons of the data with the calculated values show consistency in all cases.

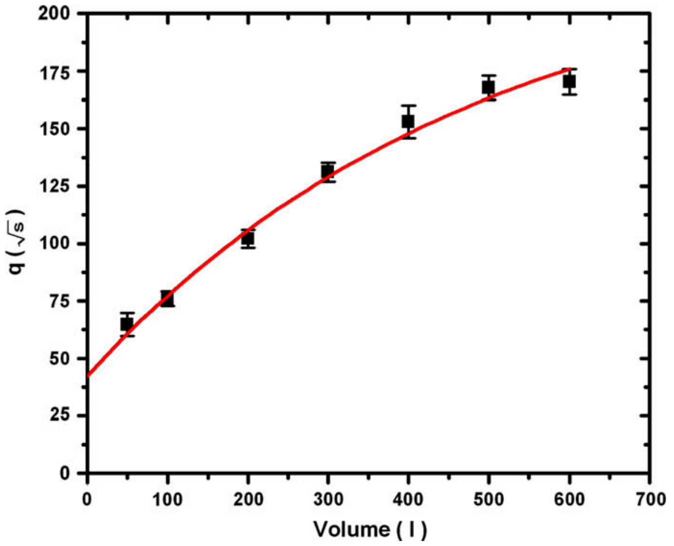

Fig. 5. The dependence of the $r/\sqrt{D}$ parameter on the volume of the cleaning process. The solid line is a curve through the data.

This fact suggests that the model of the emptied bore-hole with following diffusion of radon through the walls is in its main features valid.

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 D value. However, if we could find the value of the $r_i/\sqrt{D}$ parameter with no emptied layer, where r_i is equal with 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. 5 for volume zero. The smooth change of $r/\sqrt{D}$ suggests that the real value may not be far from the extrapolated curve around 0 volume, i.e. $(r/\sqrt{D})_{(0)} = (42.3 \pm 6.4)\sqrt{\mathrm{s}}$.

As the nominal value of the bore-head was 4 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 $D = (8.9 \pm 2.7) \times 10^{-7}\ \mathrm{m}^2\,\mathrm{s}^{-1}$. Allowing an uncertainty of some mm in the radius of the bore-hole, the diffusion parameter should have an uncertainty in the range of about 20%. The variation of the diffusion parameter on the radius of the bore-hole is $\Delta D/\Delta r_0 = (4.7 \pm 0.1) \times 10^{-7}\ \mathrm{m}^2\,\mathrm{s}^{-1}\,\mathrm{cm}^{-1}$. 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).

The average radium content of the medium can be obtained from the value of the equilibrium radon concentration level, C_{eq}. We suppose that in the surrounding soils or rocks, radioactive equilibrium between ^{222}Rn and ^{226}Ra was achieved. Taking the density, porosity and emanation of the medium into account, the radium content of the medium is (Cosma et al., 2001; Andersen, 2000)

$$A_{226\mathrm{Ra}} = \frac{C_{\mathrm{eq}}\varepsilon}{f\rho_{\mathrm{s}}(1-\varepsilon)}.$$

Here, ρ_{s} is the density, ε is the porosity of the medium and f is the emanation factor. The average value of the density of our soil type is $\rho_{\mathrm{s}} = (1.45 \pm 0.06)\ \mathrm{g\,cm}^{-3}$. The average value of the porosity is $\varepsilon = (0.44 \pm 0.03)$. The values of these two parameters we determined from laboratory investigations of the core samples that we took out while drilling the bore-hole. As the value of the f emanation factor may not be greater than 1, it is easy to make a lower estimation to the ^{226}Ra content of the soil. This value is $A_{226\mathrm{Ra}} = (5.7 \pm 0.8)\ \mathrm{Bq\,kg^{-1}}$. This is in the same order of magnitude as results from other methods applied for the determination of radium concentration for soils or geological formations (Yu et al., 1993; Nazaroff and Nero, 1988).

4. Conclusions

We have shown that in a bore-hole of several meters depth, the radon concentration measurements are reproducible up to about 20% under different meteorological conditions. This fact allows the introduction of a simple method for the estimation of the diffusion parameter of the surrounding geological formation. If we clean the bore-hole from radon and measure the influx of radon, the time-dependence is determined by the diffusion parameter.

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 formation. The equilibrium radon concentration gives estimation to a lower limit of the radium content of the geological formation, in a precision of about 15%.

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