COMPARISON OF CALCULATED AND MEASURED SOIL-GAS RADON CONCENTRATION AND RADON EXHALATION RATE

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ABSTRACT

The computer model RADON2D for WINDOWS, that enables to estimate the radon exhalation rate from the ground surface and the distribution of soil-gas radon concentration, was tested using a large set of experimental data coming from four reference areas located in regions with different geological structure. A good agreement between calculated and experimental data was observed. In a majority of cases, a correct description of the real situation was obtained using non-modified experimental input data.

INTRODUCTION

To increase the effectiveness of preventive and remedial measures is one of the main goals of the radon program in the Czech Republic. A good knowledge of the radon entry into buildings is required, when optimal preventive or remedial actions are chosen and proposed. First, it is necessary to know the radon exhalation rate from the ground surface and the distribution of soil-gas radon concentrations in the upper soil layers. As the experimental determination of radiological and geotechnical characteristics is very often complicated, especially in case of remedial actions in existing buildings, the importance of numerical modelling is growing.

A large field research was proposed and realized to evaluate the applicability of the available numerical model.

METHODS

Numerical Modelling

The computer model RADON2D for WINDOWS (Jiránek and Svoboda 1997a, 1997b), that was used for the determination of soil-gas radon concentration and of radon exhalation rate from the ground surface, is based on the general equation for the two-dimensional steady-state radon transport in a porous medium (Nazaroff and Nero, 1988):

$$D_e \nabla^2 C + \frac{k}{\varepsilon.\mu} \vec{\nabla} p \vec{\nabla} C + G - \lambda C = 0 \qquad \text{(a)}$$

where C is the radon concentration in the soil-gas [Bq.m^{-3}], D_e is the effective radon diffusion coefficient [m^2.s^{-1}], that depends on the soil-moisture, k is the permeability [m^2], μ is the dynamic viscosity of air in soil pores [17.4 .10^{-6} kg.m^{-1}.s^{-1}], p is the pressure difference [Pa], ε is the soil porosity [-] and λ is the radon decay constant [2.1 .10^{-6} s^{-1}]. The first term on the left side of the equation (a) represents the radon transport due to diffusion, the second one represents the radon transport due to convection. The third term expresses the increase of radon concentration caused by the radon generation in soil pores and the last term represents radon losses due to the radioactive decay.

Equation (a) is solved numerically using a finite element method. The model is based on following assumptions: each element is homogeneous, the soil-gas is incompressible, the pressure distribution is governed by Laplace equation and the flow of soil-gas is linear according to Darcy's Law.

The model uses following input parameters: soil porosity, permeability, effective radon diffusion coefficient and radon generation rate. The soil porosity and the permeability were measured, values of the effective radon diffusion coefficient and of the radon generation rate were derived using other measured parameters.

The determination of the effective radon diffusion coefficient was based on a following equation (Rogers and Nielson, 1991):

$$D_e = D_0 \,.\, \varepsilon \,.\, \exp(-6m\varepsilon - 6m^{14\varepsilon}) \,, \qquad \text{(b)}$$

where D_0 is the radon diffusion coefficient in air [$m^2.s^{-1}$], and m is the water saturation (= the percentage of pore space in soils that are filled with water [-]).

The radon generation rate G was calculated using the equation:

$$G = 1/\varepsilon \,.\, a_{m,226Ra} \,.\, \lambda \,.\, f \,.\, \rho_0 = 1/\varepsilon \,.\, y_m \,.\, \rho_s \,.\, (1 - \varepsilon) \,, \qquad \text{(c)}$$

where $a_{m,226Ra}$ is the mass activity of ^{226}Ra [$Bq.m^{-3}$], f is the radon emanation coefficient, y_m is the mass radon exhalation rate [$Bq.kg^{-1}.s^{-1}$], ρ_0 is the bulk density of soil [$kq.m^{-3}$] and ρ_s is the apparent density of solid particles of soil (the specific gravity of soil) [$kq.m^{-3}$],

$$\rho_0 = \rho_s \,.\, (1 - \varepsilon) \,. \qquad \text{(d)}$$

Changes of radon concentrations with depth and values of radon exhalation rate from the ground surface represent the output of the model.

Field Survey

Four reference areas with different geological structure (Dubnice, Rùžená, Bøezinìves, Èelákovice) were chosen for experimental testing of the model. The main goal of field measurements was to get as detailed information on geological and radiological characteristics as possible.

At each reference area (20 x 20 m, or 20 x 10 m), the survey consisted of soil-gas radon concentration and permeability measurements at nine measuring points, at each measuring point at five different depths below the ground surface (10, 30, 50, 80, and 150 cm); of the measurement of the radon exhalation rate at six measuring points; of the description of the vertical soil profile using six bores; of the collection of soil samples from different depths for the determination of the soil moisture, of the ^{226}Ra activity, of the radon emanation coefficient, of the bulk density of soil, and of the apparent density of solid particles.

The survey was repeated at one reference area (Dubnice) to estimate the temporal variability of measured parameters.

All field measurements were realized in summer and in autumn 1999 - reference area Dubnice: 21-07-1999, and 08-10-1999, respectively; reference area Rùžená: 04-09-1999, reference area Bøezinìves: 22-09-1999, reference area Èelákovice: 30-09-1999.

Measuring Techniques

Soil-gas samples for the determination of soil-gas radon concentrations were collected using a syringe and a small-diameter hollow steel probe and introduced into previously evacuated Lucas cells (Neznal et al., 1996a). The permeability of soil was determined by direct in-situ measurements using the equipment RADON-JOK. The method is based on the soil-gas withdrawal by means of negative pressure. The determination of the radon exhalation rate from the ground surface was based on the measurement of increasing radon concentration in a cylindrical canister placed on the ground (Neznal et al., 1996b). Similarly, the mass radon exhalation rate was derived from the measurement of radon concentration in a glass air-tight container, in which the soil sample had been closed for two weeks. The ^{226}Ra concentration in soil samples was measured using a gamma spectrometer.

Six hand-bored drills were made at each reference area to determine the geological structure and chosen geotechnical parameters. The soil samples for the determination of soil moisture were dried at the temperature of 105-110 C (Czech National Standard No. 721012). The apparent density of solid particles was determined using a 100 ml pycnometer (Czech National Standard No. 721011). Values of the bulk density of soil were estimated using available data obtained at areas with similar geological structure. To get all input data for numerical modelling, the soil porosity and the water saturation were calculated using the above mentioned results of laboratory analyses.

RESULTS AND DISCUSION

Geological Structures

The four reference areas were chosen with respect to the various geological factors influencing the radon potential of soils.

At the reference site Rùžená the bedrock is formed by granites (Central Bohemian Pluton). The bedrock weathering is extensive and irregular, the weathering profile is formed by clayey sands and sandy clays. The bedrock at three other areas is represented by Cretaceous sediments. In case of the area Dubnice, there were described Upper Turonian claystones and marlites. Bøezinìves and Èelákovice areas are formed by Lower Turonian marlites and claystones.

The Quarternary cover is represented by eluvial and deluvial deposits at the reference areas Dubnice and Rùžená, by loess eolian deposits at the reference area Bøezinìves and by fluvial sands and sandy gravels at the area Èelákovice.

Surface conditions were different from one reference area to another one. This fact may be important when the radon exhalation rate from the ground surface is evaluated. Whilst the reference areas Dubnice and Rùžená were situated in meadows, characterized by a rich vegetation, the reference areas Bøezinìves and Èelákovice were situated in agricultural cultivated fields after the harvest, i.e. almost without vegetation. The ground surface was wet at the areas Dubnice, Rùžená and Èelákovice, but dry at the area Bøezinìves.

Soil-gas Radon Concentration

As for the spatial variability of soil-gas radon concentration, the most heterogeneous situation was observed at the area Dubnice. The ratios SD/mean (standard deviation of the population vs. arithmetic mean) of data sets corresponding to different sampling depths ranged from 0.40 to 0.84 during the first measurements in July 1999 and from 0.42 to 0.92 during the second measurements in October 1999. More homogeneous results were obtained at other reference areas. The ratios SD/mean ranged from 0.20 to 0.50 at the area Rùžená, from 0.07 to 0.33 at the area Bøezinìves, from 0.16 to 0.35 at the area Èelákovice.

The spatial variability of soil-gas radon concentration decreased with depth only at the areas Rùžená and Bøezinìves. This finding corresponds with a relative homogenity of geological conditions at these two reference areas.

Examples of observed changes of soil-gas radon concentrations with depth are given in Fig. 1 and Fig. 2. As can be seen, the changes can be significant (Bøezinìves area), as well as very small (Èelákovice area). This conclusion is in a good agreement with the results of a previous research (Neznal et al., 1994).

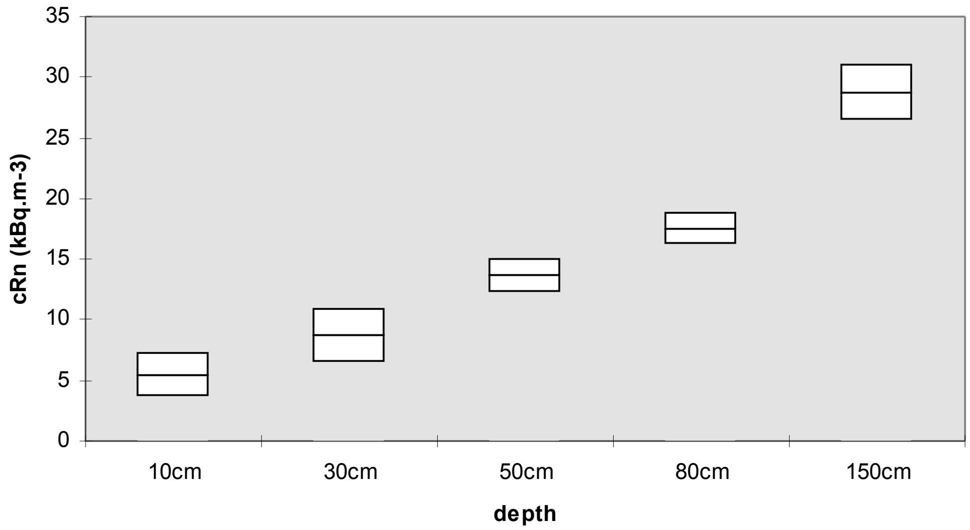

Fig. 1: Changes of soil-gas radon concentration (cRn) with depth (arithmetic mean ± standard deviation), Březiněves area

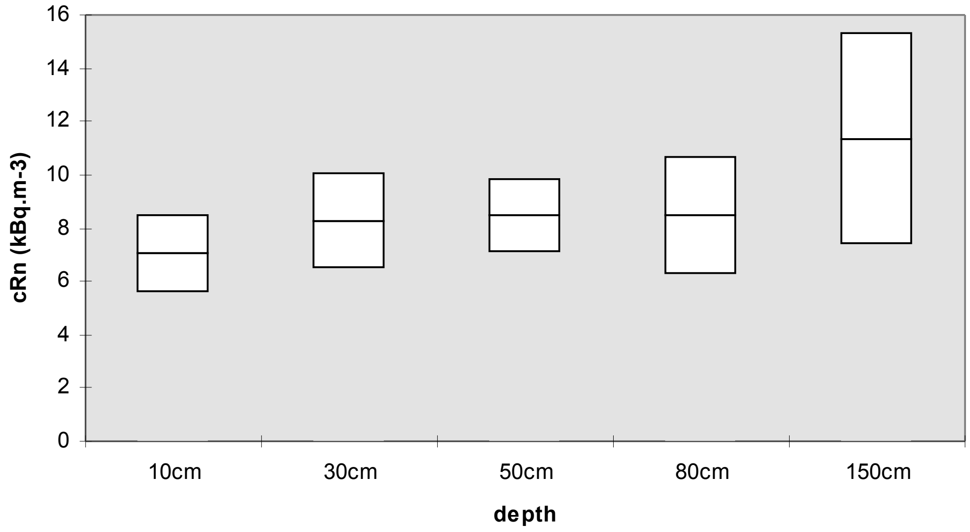

Fig. 2: Changes of soil-gas radon concentration (cRn) with depth (arithmetic mean ± standard deviation), Čelákovice area

Radon Exhalation Rate from the Ground Surface

As expected, the highest average value of radon exhalation rate from the ground (108 $mBq.m^{-2}.s^{-1}$) was observed at the area Rùžená, characterized by a high radon potential. The spatial variability expressed as the ratio SD/mean ranged from 0.16 (Èelákovice area) to 0.51 (Rùžená area). The spatial variability of radon exhalation rate from the ground is thus comparable with the spatial variability of soil-gas radon concentration.

The influence of the humidity of the upper soil layer on the radon exhalation rate can be illustrated by the values of numerical ratio between the average radon exhalation rate and the average soil-gas radon concentration in the upper soil layer (depth of 10 cm). This ratio ranges from 1.3 to 2.4 at the areas Dubnice, Rùžená and Èelákovice, where the ground surface was moist during measurements, while it equals 6 at the area Bøezinìves, where the surface was dry.

Permeability of Soil

The variability of soil permeability was high in vertical as well as in horizontal directions. The ratios SD/mean of data sets corresponding to different sampling depths ranged from 0.66 to 1.70 (July 1999) and from 0.62 to 1.57 (October 1999) at the area Dubnice, from 0.83 to 1.71 at the area Rùžená, from 0.72 to 1.11 at the area Bøezinìves, from 0.64 to 1.08 at the area Èelákovice.

It should be noted that a typical distribution of permeability data is not normal. The use of Gaussian parameters for the evaluation of the results of permeability measurements is thus not correct. This question requires a more detailed statistical analysis.

Two examples of the relation between the soil-gas radon concentration and the permeability are given in Fig. 3 and Fig. 4.

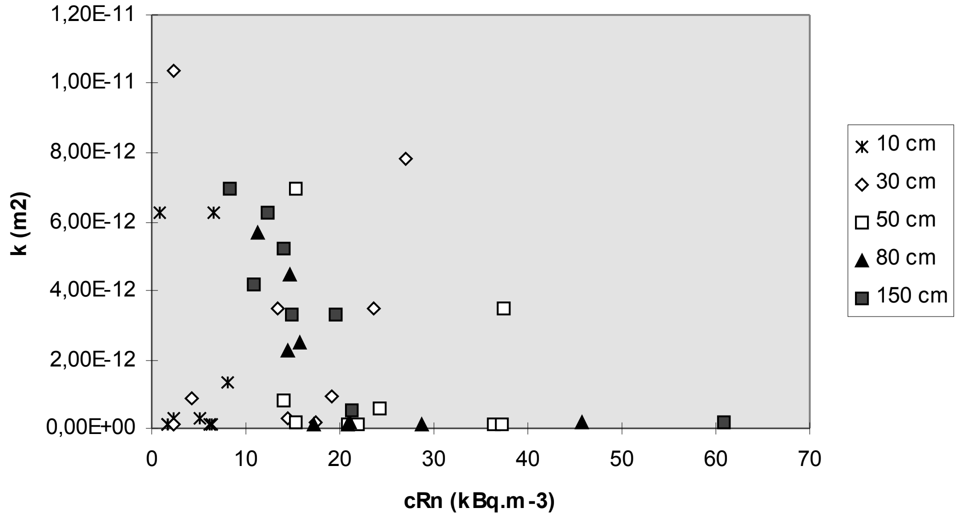

Fig. 3: Permeability (k) vs. soil-gas radon concentration (cRn), Dubnice area (July 1999)

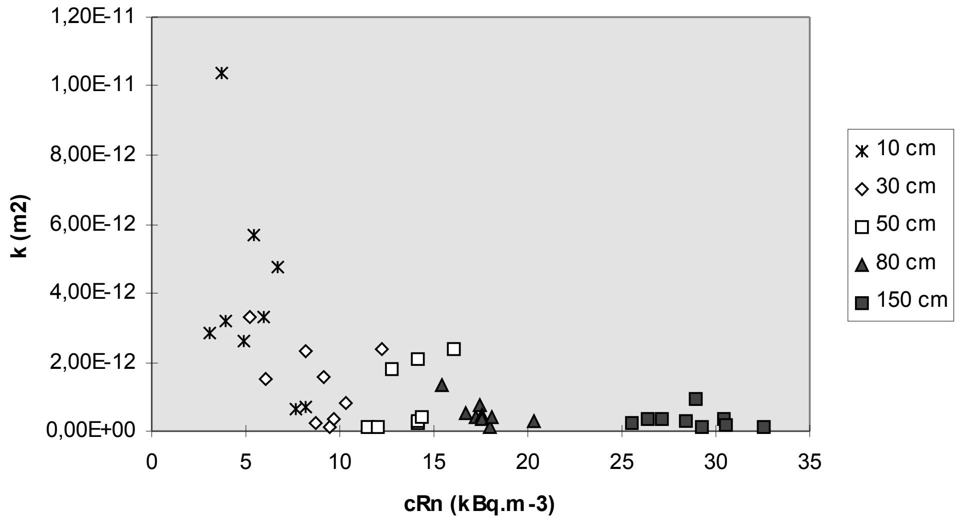

Fig. 4: Permeability (k) vs. soil-gas radon concentration (cRn), Březiněves area

Temporal Variability

A comparison of results of repeated measurements at the area Dubnice enables to evaluate temporal changes of measured parameters. Values of soil-gas radon concentration, of radon exhalation rate from the ground, of soil permeability and of soil moisture in July and in October 1999 were compared.

The soil-gas radon concentration was the most stable from the four above mentioned parameters. Relative changes of the average soil-gas radon concentration were -2.3% in the depth of 10 cm, +5.1% in the depth of 30 cm, -31.2% in the depth of 50 cm, -9.0% in the depth of 80 cm, and -1.5% in the depth of 150 cm, respectively. With the exception of the depth of 50 cm, the changes were not greater than measuring errors.

The average radon exhalation rate decreased from 10.4 to 7.9 $mBq.m^{-2}.s^{-1}$ (-24.4%).

As for the permeability, more significant changes were observed. Relative changes of the average permeability corresponding to different sampling depths ranged from -62.6% (50 cm) to -89.3% (30 cm).

The maximal difference of the average soil moisture was observed in the depth of 50 cm - from 15,6% to 10,0%, the minimal one in the depth of 30 cm - from 14.5% to 13.1%.

Parameters Used as Input Data for Numerical Modelling

Geological situation at each reference area was represented by a sector of soil 1 m wide and 1.5 m deep. This depth was the maximal sampling and measuring depth during field measurements. The sector was divided into five or six horizontal layers to describe changes of geological characteristics.

Following boundary conditions were used. As for the radon concentration in the maximal depth of 150 cm, two different input parameters were tested. The average measured soil-gas radon concentration, or the soil-gas radon concentration calculated as a ratio of the radon generation rate and the radon decay constant using equation (c). The second estimate was thus based on the results of the determination of the mass radon exhalation rate and of the bulk density of soil. The soil surface was under a fixed depressurization of -1 Pa. The soil-gas radon concentration of 10 $Bq.m^{-3}$ on the surface, and the thickness of the surface boundary layer of 4 mm were expected (the thickness of the boundary layer is identical with the distance on which the soil-gas radon concentration becomes equal to the radon concentration in the outdoor air).

Average values of permeability corresponding to different sampling depths were used as the input data of soil permeability.

As for the other geotechnical and radiological parameters (porosity, water saturation, mass ^{226}Ra activity, etc.), the calculations were made for several different combinations of obtained experimental values.

The results of numerical modelling of the situation at the reference area Èelákovice will be used to illustrate the applicability of the model. The modelled sector of soil is given in Fig. 5.

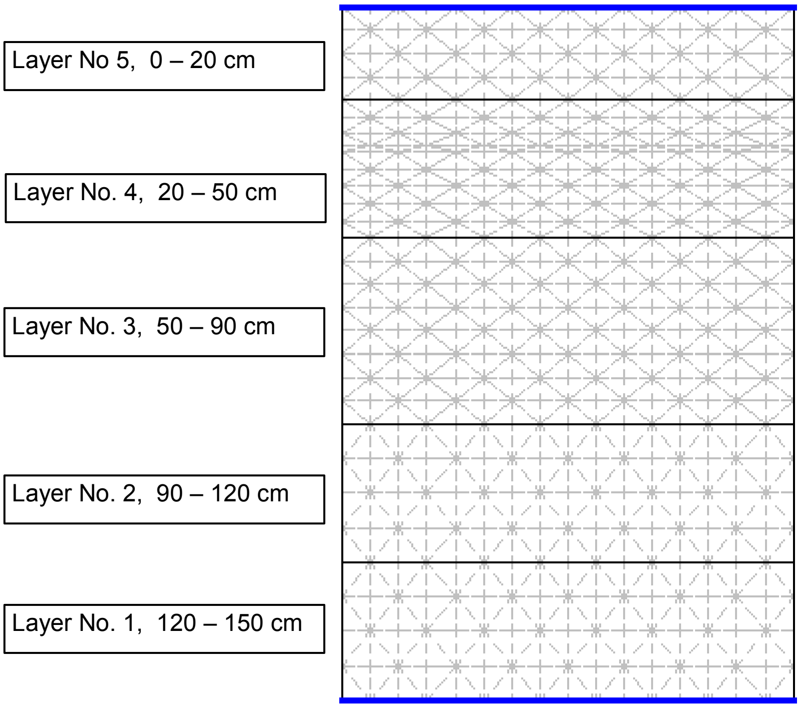

Fig. 5: Modelled sector of soil, Čelákovice area

The characteristics of soil were obtained from the analyses of samples taken from bores V1, V2 (effective radon diffusion coefficient - D_e), and V4, V6 (radon generation rate - G), respectively. They are summarized in Table 1; k is the permeability, ε is the soil porosity.

Tab. 1: Soil characteristics used as input data, Čelákovice area

Layer	**k**	**ε bore V1**	**ε bore V2**	**G bore V4**	**G bore V6**	**D_e bore V1**	**D_e bore V2**
No	(m^2)	(1)		($Bq.m^{-3}.s^{-1}$)		($m^2.s^{-1}$)	
5	3.1E-12	0.44	0.43	4.8E-5	7.5E-5	8.9E-7	1.2E-6
4	9.9E-13	0.38	0.35	5.9E-5	9.9E-5	1.3E-6	1.5E-6
3	4.8E-13	0.32	0.32	2.1E-5	3.2E-5	2.4E-6	2.5E-6
2	6.4E-13	0.32	0.32	2.1E-5	3.2E-5	2.4E-6	2.4E-6
1	1.9E-12	0.29	0.30	4.8E-5	4.1E-5	2.2E-6	1.3E-6

At the reference area Èelákovice, three combinations of input parameters were tested: (A) D_e and ε correspond to the sample taken from bore V1 and G correspond to the bore V4; (B) D_e and ε correspond to the bore V2 and G correspond to the bore V4; (C) D_e and ε correspond to bore V2 and G correspond to the bore V6.

Output of Numerical Modelling - Comparison of Calculated and Measured Soil-Gas Radon Concentration and Radon Exhalation Rate

The output of the model is represented by the distribution of radon concentrations in different depths and by the estimate of radon exhalation rate from the ground surface. An example is given in Table 2 and in Table 3. Calculated values corresponding to three above mentioned combinations of input parameters (A, B, C) are compared with measured values obtained at the reference area Èelákovice.

The agreement between calculated and measured data is relatively good for all tested combinations of input data. As can be seen in Table 1, larger differences of input parameters were observed in the upper soil layers (No 5 and 4). The use of a higher value of the radon generation rate (bore V6) resulted in higher radon concentrations over the whole soil profile and in a higher radon exhalation rate (C compared to A, B). The use of a lower value of the effective diffusion coefficient (bore V1) resulted in a lower radon exhalation rate and in higher soil-gas radon concentrations in the upper soil layers (A compared to B).

The analysis of results obtained at all reference areas has shown a good applicability of the numerical model. The model was sensitive especially to changes of the water saturation. Even very small changes of this parameter caused multiple changes of the diffusion coefficient and significant changes of calculated soil-gas radon concentration and radon exhalation rate. It is evident, that the sensitivity of the model to the changes of input parameters should be studied in more details.

CONCLUSIONS

The field survey was made at four reference areas situated in regions with different geological structure. It resulted in a detailed description of radiological, geological and geotechnical characteristics. Experimental data were used for testing the applicability of the computer model RADON2D for WINDOWS, that enables to estimate the radon exhalation rate from the ground surface and the distribution of soil-gas radon concentrations.

The conclusions can be summarized as follows:

Findings that concern the spatial variability of soil-gas radon concentration and the variability of soil-gas radon concentration with depth correspond to previous experience. The spatial variability of radon exhalation rate from the ground is comparable with the spatial variability of soil-gas radon concentration. The radon exhalation rate from the ground surface is strongly influenced by the humidity of the upper soil layer.

The variability of soil permeability is relatively high in vertical as well as in horizontal directions. A typical distribution of permeability data is not normal. The use of Gaussian parameters for the evaluation of the results of permeability measurements is thus not correct.

The agreement between calculated and measured data of soil-gas radon concentration and of radon exhalation rate is good. In a majority of cases, a correct description of the real situation was obtained using non-modified experimental input data. This conclusion confirm a general applicability of the numerical model. A higher sensitivity of the model to changes of water saturation was observed.

Tab. 2: Comparison of calculated and measured soil-gas radon concentration, Čelákovice area

Depth below surface (cm)	calculated soil-gas radon concentration (kBq.m^{-3})			measured soil-gas radon concentration (kBq.m^{-3})
	A	B	C	
0	1,1	1,2	1,7	
5	2,6	2,4	3,4	
10	4,0	3,5	4,9	7,1 ± 1,4
15	5,2	4,5	6,3	
20	6,4	5,4	7,6	
24	6,9	5,9	8,3	
28	7,4	6,4	8,9	
30	7,6	6,7	9,3	8,3 ± 1,8
32	7,8	6,9	9,5	
35	8,2	7,3	10,0	
39	8,5	7,6	10,3	
42	8,8	7,9	10,7	
46	9,0	8,2	11,0	
50	9,2	8,4	11,1	8,5 ± 1,4
55	9,4	8,6	11,2	
60	9,5	8,8	11,3	
65	9,6	8,9	11,4	
70	9,7	9,1	11,5	
75	9,9	9,2	11,6	
80	10,0	9,4	11,6	8,5 ± 2,2
85	10,1	9,5	11,7	
90	10,2	9,7	11,8	
97	10,4	9,9	11,8	
105	10,6	10,1	11,9	
112	10,8	10,4	11,9	
120	11,0	10,6	11,9	
127	11,2	11,0	11,9	
135	11,3	11,2	11,8	
142	11,4	11,4	11,6	
150	11,4	11,4	11,4	11,4 ± 3,9

Tab. 3: Comparison of calculated and measured radon exhalation rate from the ground surface, Čelákovice area

calculated radon exhalation rate (mBq.m^{-2}.s^{-1})			measured radon exhalation rate (mBq.m^{-2}.s^{-1})
A	B	C	
11,4	12,5	17,7	16,0 ± 2,6

A more detailed study of the model sensitivity to changes of different input parameters is recommended.

The large set of experimental data, that had been collected, could be used to another studies, or analyses, for example to propose a concept of the radon availability, i. e. a single parameter describing the radon potential of foundation soils.

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