MEASUREMENT OF RADON EXHALATION RATE FROM THE GROUND SURFACE: CAN THE PARAMETER BE USED FOR A DETERMINATION OF RADON POTENTIAL OF SOILS?

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ABSTRACT

Temporal and spatial variability of radon exhalation rate from the ground surface and of other parameters were followed at four reference areas during a one-year climatic cycle. The analysis of obtained data resulted in the conclusion that the method of radon exhalation rate determination cannot be recommended to be used as a standard supplementary method for radon risk classification of foundation soils.

INTRODUCTION

A uniform method that is used in the Czech Republic for the determination of radon potential of foundation soils is based on the measurement of soil-gas radon concentration and on the determination of soil permeability [1 - 2]. The samples of soil-gas are collected at a depth of 0.8 m below the ground surface. In some special cases - when the thickness of soil cover is very low, or when the soil pores are saturated with water - the sample collection at this depth is rather complicated, or almost impossible. There is a question: Would it be possible to substitute some other parameter for the soil-gas radon concentration?

Radon exhalation rate from the ground surface is one of the parameters characterising radon potential of soils, or radon potential of waste materials contaminated by natural radionuclides [3 - 5]. Different techniques for the determination of radon exhalation rate are available - for example the simple accumulator method [6]. On the other hand it is evident that there is at least one serious disadvantage connected with the method: As the ground surface is strongly affected by changing meteorological conditions, large temporal variations of radon exhalation rate can be expected.

In 2000, a large research project dealing with an improvement of the original classification method was prepared [7]. The research was divided into ten sections and one of them was focused on testing of the usefulness of radon exhalation rate measurements for the determination of radon potential of soils.

Testing was based on repeated measurements of radon exhalation rate from the ground surface at four reference areas during a one-year climatic cycle. Temporal and spatial changes of other important parameters (soil-gas radon concentration, soil permeability, soil moisture) were also measured and evaluated.

METHODS

The determination of radon exhalation rate using the simple accumulator method is based on the measurement of increasing radon concentration in a cylindrical canister placed on the measured surface. The relation between the radon concentration in the canister $\mathbf{c_{Rn}(t)}$ and the radon exhalation rate **J** is described by following equation:

$$c_{Rn}(t) \quad [Bq.m^{-3}] \quad = \quad \frac{J\ [Bq.m^{-2}.s^{-1}]\ .\ S\ [m^2]}{V\ [m^3]\ .\ \alpha\ [s^{-1}]}\ .\ (1 - EXP(-\alpha\ .\ t)) \qquad (1)$$

where,

t = duration of canister exposure;

S = canister base;

V = canister volume; and

α = (k_{Rn} + k_L) = constant describing a loss of radon caused by radioactive decay (k_{Rn} = $2.10^{-6}.s^{-1}$; radon decay constant) and by leakage (k_L).

The radon exhalation rate **J** is then expressed by:

$$J\ [Bq.m^{-2}.s^{-1}] = \frac{c_{Rn}(t)\ [Bq.m^{-3}].\ h\ [m]\ .\ \alpha\ [s^{-1}]}{1 - EXP(-\alpha\ .\ t)} \qquad (2)$$

where **h = V/S** is the canister height.

The role of parameters **α** and **J** is illustrated in Fig. 1. A theoretical increase of radon concentration in the accumulator calculated for different values of both parameters is presented there. As can be seen, the value of **J** corresponds to the „steepness" of the curve, while the value of **α** is related to the time period that is necessary to reach the equilibrium between radon increase and radon losses.

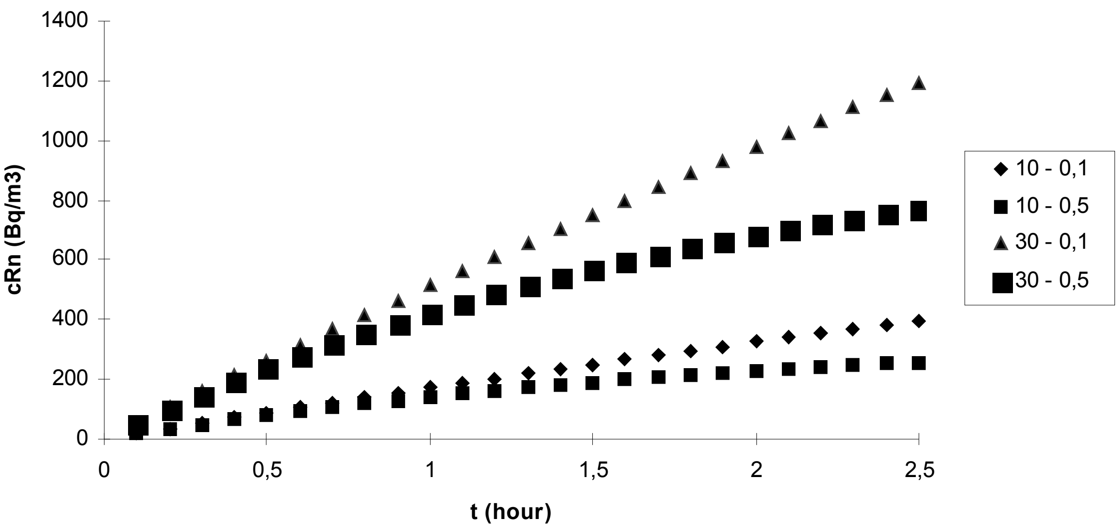


Figure 1 - A theoretical increase of radon concentration in the accumulator calculated for different values of radon exhalation rate **J** and of constant describing a loss of radon **α**. Symbol „10 - 0,1" indicates that the increase of radon concentration was calculated for J = 10 $mBq.m^{-2}.s^{-1}$, and for α = $0,1h^{-1}$.

Parameters **α** and **J** can be determined using a numerical method, or an analytical one, if values of **cRn(t)** for different **t** are available.

Note: The analytical solution of the equation (2) is based on the choice of regular time intervals. If the samples of air from the canister are taken in time **t_1** and **t_2**, where $t_2 = 2\ t_1$ and if we suppose that **J** does not change, i.e. that J (t_1) = J (t_2), then using the equation (2) it is possible to write:

$$\frac{cRn(t_2)}{cRn(t_1)} = \frac{1 - EXP(-\alpha\ .\ t_2)}{1 - EXP(-\alpha\ .\ t_1)} \qquad (3).$$

Due to the fact that:

$$1 - EXP(- 2 . \alpha . t_1) \quad = \quad 1^2 - \{EXP(-\alpha . t_1)\}^2 \qquad (4)$$

we can change the equation (3) to:

$$\frac{cRn(t_2)}{cRn(t_1)} - 1 \quad = \quad - EXP(-\alpha . t_1) \qquad (5).$$

Parameter α is then calculated using the equation:

$$\alpha \quad = \quad -(1/t_1) . \ln ((av(t_2)/ av(t_1)) - 1) \qquad (6).$$

During the field survey, cylindrical canisters having the base of 0.08 m^2 and 0.2 m high were used. One measurement of radon exhalation rate was based on a determination of raising radon concentration in four air samples collected from the accumulator in regular 40-, or 60-minutes intervals. One measurement thus took from 160 to 240 minutes. Air samples were transferred into previously evacuated 125-ml Lucas cells and measured in the laboratory.

At first, values of radon exhalation rate were calculated using the analytical approach. The results were then used as an input for numerical calculations. When the increase of radon concentration in the canister was not regular, a more detailed analysis was made. These cases are described lower.

As for the determination of other parameters, samples of soil-gas were collected from a depth of 0.8 m below the ground surface using small-diameter hollow steel probes and a syringe and also transferred into previously evacuated Lucas cells. Direct in situ measurements of soil permeability were made using the equipment RADON-JOK. The method is based on a soil-gas withdrawal by means of negative pressure. Soil moisture was determined by weighing the original and dried soil samples. Temporal changes of soil moisture were determined using an indirect method based on measurement of the dielectric constant of soil.

During field measurements, meteorological conditions were also registered.

REFERENCE AREAS, FIELD MEASUREMENTS

The majority of field measurements was performed at four reference areas characterised by different geological conditions (reference areas Dubnice, Stráž, Růžená, Žibřidice). The reference area Dubnice can be briefly described as an area with variable upper soil layers, with variable soil permeability ranging from low to medium values and with „medium“ values of soil-gas radon concentrations. The reference area Stráž is situated on the uranium mill tailings. The conditions at the area are therefore relatively homogeneous, characterised by medium, or high soil permeability and by extremely high soil-gas radon concentrations. At the reference area Žibřidice, we expected a high saturation of upper soil layer during the one-year follow-up. This expectation was fulfilled only partly. The fourth reference area, called Růžená, was situated in a region with a low thickness of soil cover, with high soil permeability and with high soil-gas radon concentrations.

During one measuring day, radon exhalation rate, soil permeability, as well as soil gas radon concentration were determined at ten measuring points. Soil moisture was measured in six probes in different depths below the ground surface.

Temporal changes of all parameters were followed at four reference areas from summer 2000 to summer 2001. Measurements were repeated every second month, i.e. they were repeated seven times at each area.

Two different ways of putting the canisters on measured surface were tested at the reference areas Dubnice and Stráž: (a) canister is placed on an undisturbed soil surface and sealed by clay or sandy clay (this way will be marked as „surface“); (b) the upper soil layer is removed and canister is placed about 10 cm below the ground surface (this way will be marked as „-10cm“). Both ways are illustrated in Fig. 2.

In September 2001, supplementary measurements of radon exhalation rate, of soil permeability, and of soil gas radon concentration were made at another reference area Zdiměřice, characterised by an extremely low soil permeability and by water saturation of the upper soil layer.

On the whole, 290 measurements of three main parameters were made.

Figure 2 - Two different ways of putting the canisters on measured surface: (a) canister was placed on an undisturbed soil surface and sealed by sandy clay (marked as „surface"); (b) canister was placed about 10 cm below the ground surface (marked as „-10cm")

a)

b)

RESULTS AND DISCUSSION

Examples of results obtained at two reference areas (Dubnice, Růžená) are summarised in Tab. 1 - 2. Temporal variations of median values of radon exhalation rate and of soil-gas radon concentration at the same areas are illustrated in Fig. 3 - 4.

During the statistical evaluation of data, values corresponding to the detection limit were substituted for any measured values lower than this limit (i.e. the value of 2 $mBq.m^{-2}.s^{-1}$ was substituted for all values lower than 2 $mBq.m^{-2}.s^{-1}$).

As can be seen in Table 1, the spatial variability of radon exhalation rate at the **reference area Dubnice,** expressed as a ratio SD/mean, ranged typically from 30 to 70% („surface" method) and from 15 to 80% („-10 cm" method) respectively. An extremely high variability was observed in March 2001, most probably due to a strong wind during measurement. The spatial variability of soil-gas radon concentration was similar, ranging from 30 to 70%.

Table 1 - Reference area Dubnice: Summary of measurement results (SD = standard deviation of population)

Date	18.7.2000	21.9.2000	3.11.2000	11.1.2001	19.3.2001	7.5.2001	23.7.2001
Radon exhalation rate from the ground surface J (mBq.m^{-2}.s^{-1})							
„surface“ (number of measuring points: 6)							
arith. mean	7.5	10.7	7.6	4.2	4.7	3.5	9..
SD	3.4	4.2	2.8	2.9	6.1	1.2	4.5
SD/mean	0.45	0.40	0.37	0.69	1.31	0.34	0.49
minimum	4.3	6.4	4.8	2.0	2.0	2.0	5.2
maximum	12.4	16.4	11.3	9.8	17.1	4.8	17.6
median	7.0	8.8	7.0	3.5	2.0	3.5	8.3
„-10 cm“ (number of measuring points: 4)							
arith. mean		26.0	14.8	7.4	15.2	16.3	46.9
SD		16.1	2.2	1.3	13.9	13.2	35.1
SD/mean		0.62	0.15	0.18	0.92	0.81	0.75
minimum		8.6	13.1	6.2	2.0	5.6	12.0
maximum		42.2	17.7	9.1	34.6	34.2	88.6
median		26.6	14.1	7.1	12.0	12.7	43.5
Soil-gas radon concentration c_{Rn} (kBq.m^{-3}; number of measuring points: 10)							
arith. mean	18.4	15.8	18.2	18.5	11.8	19.7	19.7
SD	7.5	4.2	4.7	13.2	8.2	12.4	9.8
SD/mean	0.41	0.27	0.26	0.72	0.69	0.63	0.50
minimum	12.0	10.4	10.8	2.9	1.0	1.0	9.1
maximum	37.3	24.4	23.7	46.5	23.1	44.6	38.7
median	15.9	15.0	17.9	14.8	12.5	16.0	15.5
Soil permeability k (m^2; number of measuring points: 10)							
arith. mean	1.9E-12	2.1E-12	3.5E-12	2.7E-12	1.8E-12	1.2E-12	1.6E-12
SD	2.2E-12	2.0E-12	4.1E-12	5.0E-12	3.6E-12	1.9E-12	3.0E-12
SD/mean	1.17	0.98	1.19	1.87	2.02	1.52	1.91
minimum	1.0E-13	5.2E-14	5.2E-14	5.2E-14	5.2E-14	5.2E-14	5.2E-14
maximum	6.3E-12	5.2E-12	1.3E-11	1.6E-11	1.1E-11	4.8E-12	9.9E-12
median	1.2E-12	1.4E-12	1.3E-12	2.7E-13	5.2E-14	5.2E-14	4.1E-13
Soil moisture w (%)							
depth 0.1 m (number of measuring points: 6)							
arith. mean	21.9	22.5	21.4	23.2	26.3	20.7	22.8
SD	3.3	3.4	3.7	4.7	4.8	3.8	3.6
SD/mean	0.15	0.15	0.17	0.20	0.18	0.18	0.16
minimum	16.4	17.8	17.0	18.6	20.2	15.9	17.7
maximum	25.7	27.0	26.8	31.2	32.6	26.4	28.2
median	22.3	23.2	21.3	23.0	26.6	20.6	23.2
depth 0.9 m (number of measuring points: 6)							
arith. mean		12.9	10.0	21.2	27.3	22.2	26.7
SD		4.0	4.0	9.6	10.0	11.1	9.9
SD/mean		0.32	0.41	0.45	0.37	0.50	0.37
minimum		7.7	6.7	12.1	12.8	8.0	9.5
maximum		19.7	17.7	34.4	35.3	31.8	33.4
median		13.1	9.2	19.4	32.5	27.4	29.3

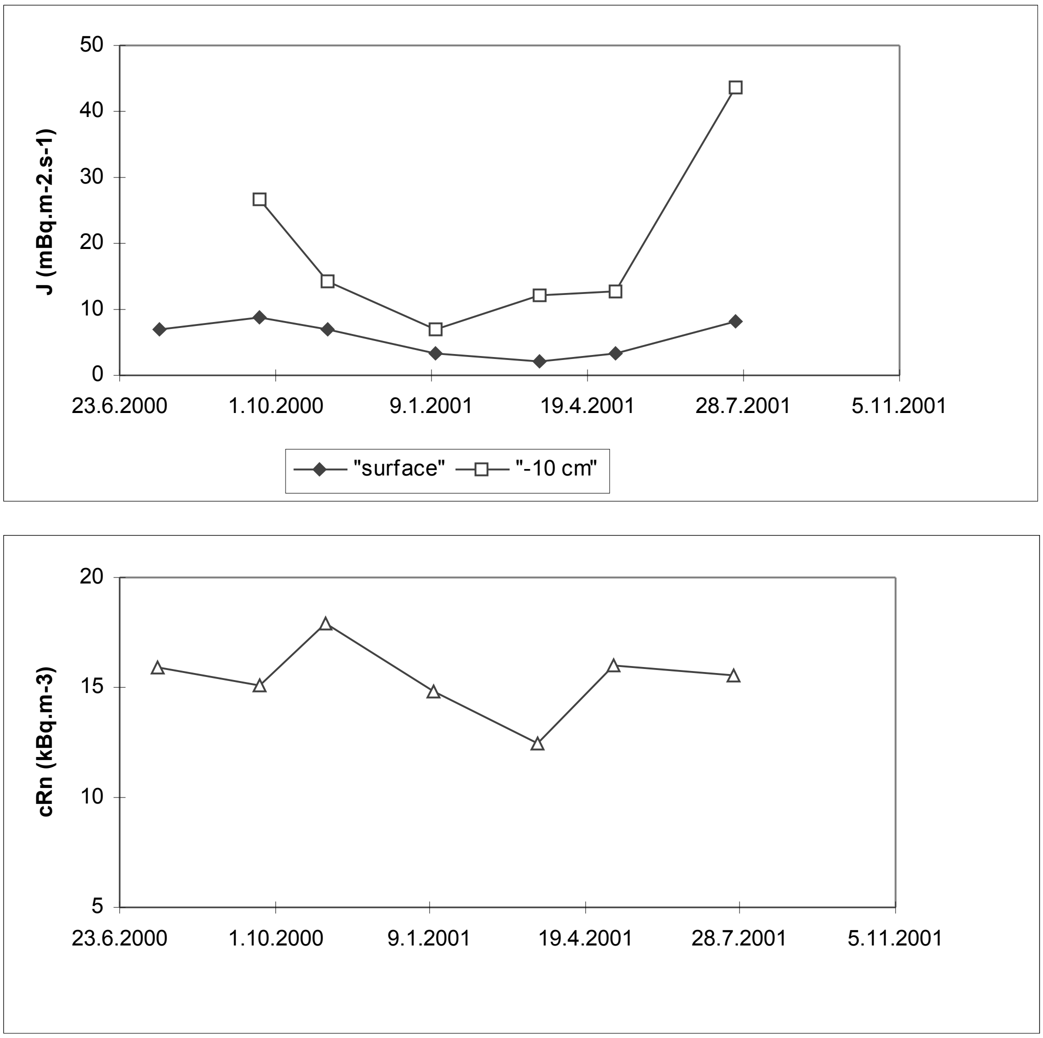


Figure 3 - Temporal variations of median values of radon exhalation rate **J** and of soil-gas radon concentration $\mathbf{c_{Rn}}$ at the area Dubnice

As for the temporal variability of radon exhalation rate using the „surface" method, the highest median value of 8.8 mBq.m^{-2}.s^{-1} was observed in September 2000, while the lowest one of 2.0 mBq.m^{-2}.s^{-1} in March 2001. The ratio of these two values was 4.4. When the „-10 cm" method was used, the same ratio was 6.1. In general, the lowest values of radon exhalation rate were observed under extreme meteorological conditions, i.e. when the soil surface was frozen, or saturated by water, or when strong wind occurred. The temporal variability of median values of soil-gas radon concentration was significantly lower. They ranged from 12.5 to 17.9 kBq.m^{-3}.

A relation between median values of radon exhalation rate determined using two different methods („surface" and „-10 cm") during a one-year cycle is described by a correlation coefficient of 0.76. It is evident that the values obtained using the „-10 cm" method were higher.

A correlation between the radon exhalation rate from the surface and the soil-gas radon concentration, as well as a correlation between the radon exhalation rate and the soil moisture in a depth of 0.1 m was very weak.

Similar temporal and spatial patterns were observed at the **reference areas Stráž and Žibřidice**. Due to homogeneous geological conditions at the area Stráž, the spatial variability of soil-gas radon concentration expressed as a ratio SD/mean was only about 10%. A correlation coefficient between median values of radon exhalation rate measured using two different methods was 0.85 at this reference area.

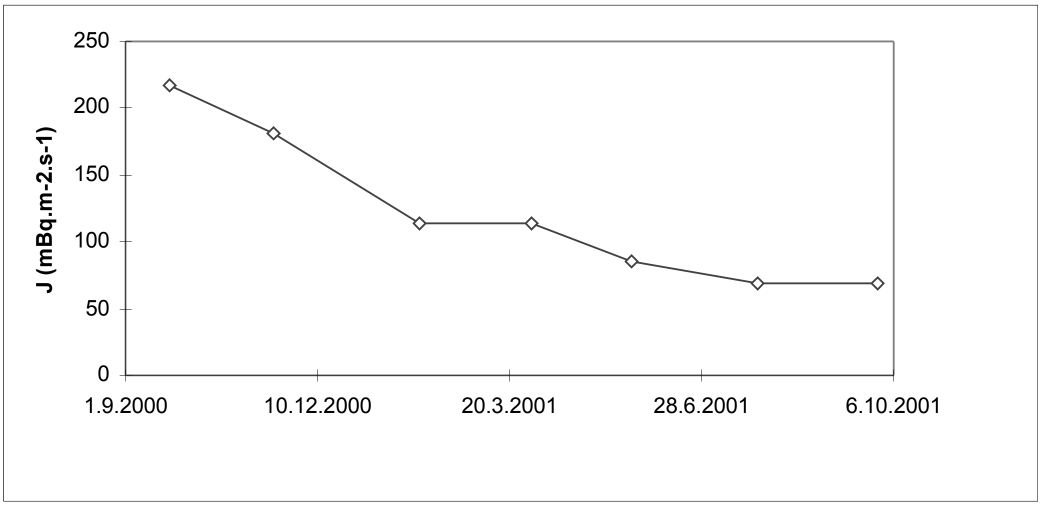
Similar spatial variability of radon exhalation rate and of soil-gas radon concentration as at the area Dubnice was observed also at the fourth **reference area Růžená** (see Tab. 2). Values of the ratio

SD/mean ranged typically from 15 to 75% for radon exhalation rate („surface“ method only) and from 30 to 70% for soil-gas radon concentration respectively. But also temporal variability of both parameters was comparable at this area. The ratio of the highest vs. the lowest median values during a one-year cycle was 3.1 for radon exhalation rate and 4.3 for soil-gas radon concentration.

As can be seen in Fig. 4 and in Tab. 2, an inverse correlation between median values of the radon exhalation rate and of the soil-gas radon concentration (correlation coefficient -0.74), as well as between median values of the radon exhalation rate and of the soil moisture in a depth of 0.1 m (correlation coefficient -0.79) was found.

Table 2 - Reference area Růžená: Summary of measurement results (SD = standard deviation of population)

Date	24.9.2000	17.11.2000	1.2.2001	31.3.2001	23.5.2001	27.7.2001	28.9.2001
Radon exhalation rate from the ground surface J ($mBq.m^{-2}.s^{-1}$)							
„surface“ (number of measuring points: 10)							
arith. mean	208.5	188.7	116.7	119.5	95.3	70.9	84.8
SD	30.8	32.7	40.8	25.0	72.8	52.3	51.9
SD/mean	0.15	0.17	0.35	0.21	0.76	0.74	0.61
minimum	145.0	147.0	55.6	75.0	6.3	6.4	14.0
maximum	241.0	237.0	200.0	161.0	241.0	150.0	167.0
median	217.0	180.5	113.5	114.0	86.0	69.5	69.0
Soil-gas radon concentration c_{Rn} ($kBq.m^{-3}$; number of measuring points: 10)							
arith. mean	68.9	127.1	99.1	124.9	178.2	187.6	224.9
SD	38.6	48.2	40.2	41.1	119.8	123.7	145.7
SD/mean	0.56	0.38	0.41	0.33	0.67	0.66	0.65
minimum	21.6	72.7	45.0	46.3	27.7	42.9	40.2
maximum	125.9	230.5	159.5	198.2	346.7	351.0	411.6
median	61.8	118.9	97.3	125.1	154.0	158.0	264.4
Soil permeability k (m^2; number of measuring points: 10)							
arith. mean	7.7E-12	3.6E-12	7.8E-12	4.8E-12	5.7E-12	4.7E-12	8.5E-12
SD	6.6E-12	3.1E-12	5.1E-12	5.0E-12	4.5E-12	5.4E-12	5.6E-12
SD/mean	0.86	0.84	0.65	1.04	0.80	1.13	0.65
minimum	3.8E-13	6.0E-13	3.4E-13	3.2E-13	1.2E-12	3.5E-14	8.1E-13
maximum	1.6E-11	9.1E-12	1.6E-11	1.4E-11	1.6E-11	1.6E-11	1.6E-11
median	5.0E-12	2.9E-12	9.1E-12	3.0E-12	4.5E-12	2.3E-12	8.4E-12
Soil moisture w (%)							
depth 0.1 m (number of measuring points: 6)							
arith. mean	10.2	9.7	13.3	17.9	16.3	15.3	15.3
SD	1.3	1.8	4.2	2.6	4.7	4.0	2.6
SD/mean	0.13	0.19	0.31	0.14	0.29	0.26	0.17
minimum	8.8	7.7	8.3	14.0	7.3	7.7	12.1
maximum	11.7	12.5	20.8	21.6	21.0	19.6	19.0
median	10.4	9.4	12.4	17.8	17.2	16.1	14.9



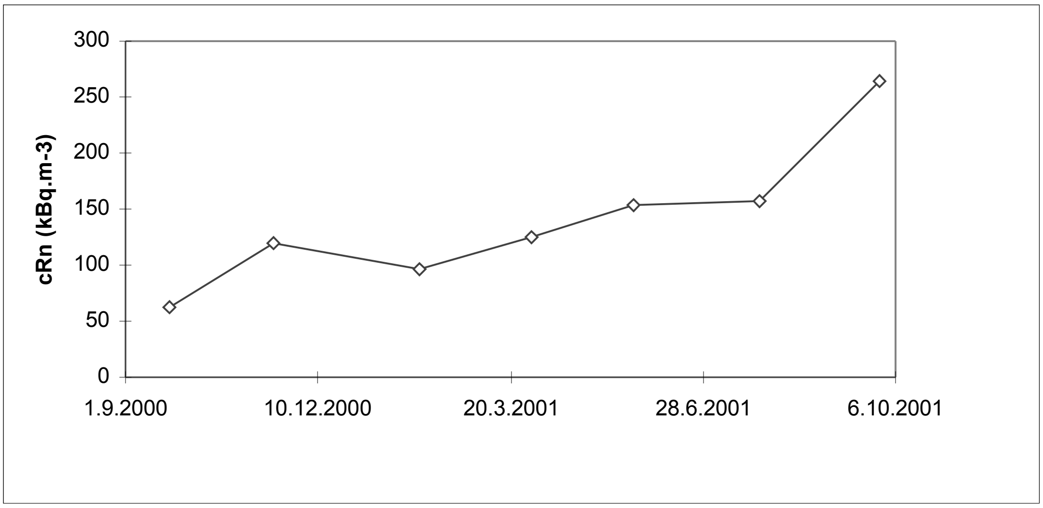


Figure 4 - Temporal variations of median values of radon exhalation rate **J** („surface“ only) and of soil-gas radon concentration $\mathbf{c_{Rn}}$ at the area Růžená

There are two main reasons for different findings at the reference area Růžená: At first, a thickness of the soil cover is low and variable at this area, i.e. in different measuring days, samples of soil-gas were taken from different depths below the ground surface. The average sampling depth was not constant. Temporal as well as spatial variability of soil-gas radon concentration was thus influenced by changes of soil-gas radon concentration with depth [2, 8].

Secondly, the surface of the reference area was significantly changed between March and May 2001. Heavy machines crossed the area during the wood exploitation. The resulting compression caused a substantial decrease of permeability of the upper soil layer at the test site. The decrease of permeability has resulted in a decrease of radon exhalation rate from the surface and in an increase of soil-gas radon concentration.

A possibility to use the results of radon exhalation rate measurements for a determination of radon potential of soils at areas characterised by a very low permeability was tested at **the area Zdiměřice**. Results of a supplementary survey were following: Soil-gas radon concentrations (13 measuring points) ranged from 2.1 to 190 kBq.m^{-3} (arithmetic mean = 107 kBq.m^{-3}, SD = 64.4 kBq.m^{-3}, SD/mean = 0.60, median = 125 kBq.m^{-3}), while the values of radon exhalation rate from the surface were very low (10 measuring points, values ranging from 2 mBq.m^{-2}.s^{-1} to 10 mBq.m^{-2}.s^{-1}, arithmetic mean = 5 mBq.m^{-2}.s^{-1}, SD = 3 mBq.m^{-2}.s^{-1}, median = 6 mBq.m^{-2}.s^{-1}).

Some problems connected with the measuring technique itself were observed during field measurements of radon exhalation rate. Theoretical equations given in „Methods“ are valid only if both basic parameters, i.e. radon exhalation rate **J** and parameter α describing radon losses from the canister, remain constant during the whole measurement. But the shape of a curve that describes increasing radon concentration in the accumulator indicates very often that the above assumption may be false. Changes of radon exhalation rate as well as changes of ventilation of the canister may occur during measurement. Variations of radon exhalation may be initiated for example by the sun. When the sun heats the air in the canister, resulting pressure differences will cause changes of radon exhalation. On the other hand, a ventilation of the accumulator is strongly influenced by the wind. Changes of both parameters during measurement result in increasing errors.

Three types of differences between theoretical curve and measured radon concentrations were observed:

(a) There is a single outsider, i.e. a single value that is lower than expected. This case is not very frequent and most probably it is caused by an error during the sample collection. The outsider value is usually excluded and it is not evaluated.

(b) The increase of radon concentration is very fast and the evaluation results in very low values of parameter α describing radon losses (lower than $\mathbf{k_{Rn}} = 2.10^{-6}.s^{-1}$, or even lower than zero). This effect is caused by a decrease of α and/or by an increase of **J** during measurement. In this case there are two solutions: To suppose that $\alpha = \mathbf{k_{Rn}} = 2.10^{-6}.s^{-1}$, to calculate radon exhalation rate **J** for all single values of radon concentration $\mathbf{c_{Rn}}$, and to determine an arithmetic mean of **J**. Or to use only the last value of $\mathbf{c_{Rn}}$ and $\alpha = 2.10^{-6}.s^{-1}$ for the calculation of **J**.

(c) After an initial increase the radon concentration in the canister raises only slowly, remains almost constant, all even falls down. This case is critical because the evaluation results not only in high values of parameter α (higher than 1 h^{-1}, or even higher than 2 h^{-1}), but unfortunately also in overestimated values of radon exhalation rate **J**. Changes of a ventilation of the canister caused by wind represent the most common reason. A strong wind, or a changeable wind increases the error of measurement and the evaluation of data is then always complicated.

CONCLUSIONS

It is possible to conclude that the spatial variability of radon exhalation rate is comparable or a little higher than the spatial variability of soil-gas radon concentration. A larger variability was observed, when measurements were made under extreme meteorological conditions, i.e. when the soil surface was frozen, or saturated by water, or when strong wind occurred. If the radon exhalation rate were used for the determination of radon potential of soils, the requirements concerning a minimal number of measuring points and scales of measuring points should be at least the same as for soil-gas radon concentration.

The temporal variability of radon exhalation rate is significantly higher than the temporal variability of soil-gas radon concentration if the soil-gas samples are collected from a depth of 0.8 m below the surface. Two different methods of putting the canisters on measured surface („surface“ and „-10 cm“) were tested at two reference areas. We had expected that the second one should be less sensitive to changes of meteorological conditions, but the assumption was not confirmed. The temporal variability of results obtained by using both methods was similar.

In general, a correlation between the radon exhalation rate from the surface and the soil-gas radon concentration, as well as a correlation between the radon exhalation rate and the soil moisture was very weak. This conclusion is valid even for the soil moisture measured at a depth of 0.1 m.

Measured values of radon exhalation rate are substantially affected by conditions on the soil surface. Significantly lower values were observed, when the soil surface was frozen or covered by water. A decrease of radon exhalation rate was found at the reference area Růžená after the changes of soil surface caused by a compression of the upper soil layer.

Supplementary measurements that were realised at the area Zdiměřice have confirmed that the use of radon exhalation rate is not suitable for the determination of radon potential of soils at building areas with low permeable, or saturated soils. If a water saturation of upper soil layers is connected with a low soil permeability, the radon exhalation rate from the surface is very low, even in case when the values of soil-gas radon concentration indicate a high radon potential.

For these reasons the method of radon exhalation rate determination cannot be recommended to be used as a standard supplementary method for radon risk classification of foundation soils.

ACKNOWLEDGMENT

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