



**RADON AND THORON VARIATIONS IN SOIL AIR – THREE YEARS STUDY IN
BRATISLAVA**

**VARIÁCIE RADÓNU A TORÓNU V PÔDNOM VZDUCHU – TROJROČNÉ ŠTÚDIUM V
BRATISLAVE**

Andrej Mojzeš¹

Abstract

The article is devoted to the evaluation of the dependence between the measured values of the ^{222}Rn and ^{220}Rn activity concentration in soil air and some meteorological parameters (temperature, humidity and pressure of atmospheric air and precipitation) in 3 annual cycles from 21.1.2003 to 23.6.2005. The graphs show the annual course of the observed variables, and the tables provide a basic statistical evaluation and overview of the dependences based on linear regression. The results confirmed the main dependence of the ^{222}Rn and ^{220}Rn content in the pore space of the soil cover on the atmospheric temperature and humidity in the annual cycle. The values of the linear regression coefficients are not significant due to the time delay of the course of the ^{222}Rn and ^{220}Rn activity concentrations in the soil behind the course of the atmospheric temperature. The time shift is 2 to 3 months. In accordance with literary sources, it documents the existence of predominantly diffusive transport of soil gases in a quasi-homogeneous geological environment without tectonics.

Abstrakt

Článok je venovaný vyhodnoteniu závislostí medzi nameranými hodnotami koncentrácie aktivity ^{222}Rn a ^{220}Rn v pôdnom vzduchu a niektorými meteorologickými parametrami (teplota, vlhkosť a tlak atmosférického vzduchu a úhrn zrážok) v 3 ročných cykloch od 21.1.2003 do 23.6.2005. Prezentované grafy znázorňujú ročný priebeh pozorovaných premenných a tabuľky poskytujú základné štatistické vyhodnotenie a prehľad závislostí na základe lineárnej regresie. Výsledky potvrdili hlavnú závislosť obsahu radónu a torónu v pórovom priestore pôdneho krytu od teploty a vlhkosti atmosférického prostredia v ročnom cykle. Hodnoty koeficientov lineárnej regresie nie sú významné z dôvodu časového oneskorenia priebehu koncentrácií aktivity ^{222}Rn a ^{220}Rn v pôde za priebehom najmä teploty atmosférického

vzduchu. Časový posun je 2 až 3 mesiace. V súlade s literárnymi zdrojmi dokumentuje existenciu prevažne difúzneho transportu pôdnych plynov v kvázihomogénnom geologickom prostredí bez tektoniky.

Key words

Activity concentration of ^{222}Rn and ^{220}Rn , soil air, temperature, humidity and pressure of atmospheric air, precipitation deposit, seasonal variations

Kľúčové slová

Koncentrácia aktivity ^{222}Rn a ^{220}Rn , pôdný vzduch, teplota, vlhkosť a tlak atmosférického vzduchu, úhrn zrážok, sezónne variácie

1. Introduction

Radon ($_{86}\text{Rn}$) is natural colorless, odorless and tasteless inert radioactive gas, one of the heaviest gases in nature. Three isotopes of radon are common in the natural decay series: ^{222}Rn (radon sensu stricto) originates in the ^{238}U decay series and has a half-life of 3.82 days, ^{220}Rn (thoron) is in the ^{232}Th chain with a half-life of 55.6 s and, ^{219}Rn (actinon) is in the ^{235}U series with its half-life of 4.0 s. All are alpha particle emitters. Actinon ^{219}Rn , due to its short half-life and the comparative scarcity of its long-lived parent ^{235}U , can usually be omitted from any radiological considerations (Wilkening, 1990). The common isotopes of radon are present in the environment because they are produced continuously by the decay of longer-lived nuclides found in minerals containing uranium, thorium, or actinium. Once formed in the Earth's crust the radon is free to diffuse into soil air or pore-water filling and then by pressure driven flow or further diffusion to the atmosphere or empty underground spaces and human houses. Soil gas infiltration is recognized as the most important source of residential radon. Other sources, including building materials and water extracted from wells, are of less importance in most circumstances. Radon is a major contributor to the ionizing radiation dose received by the general population. Recent studies on indoor radon and lung cancer in Europe, North America and Asia provide strong evidence that radon causes a substantial number of lung cancers in the general population. Radon is the second cause of lung cancer after smoking (WHO, 2009).

The chief source of radon in indoor air is from the soil under and around the basement and/or foundation. Soil gas transport to the interior far exceeds the combined contribution from building materials, drinking water, natural gas, and other sources (Wilkening, 1990). Elevated radon activity concentrations indoors are often associated with particular geological formations; however, the only way to accurately determine the concentration of radon in a particular building is to measure it inside (IAEA, 2019).

However, another approach to suppose indoor radon level in new buildings is the radon risk assessment of building sites in the form of the radon index (RI). A building site's radon index indicates the level of risk of radon release from bedrock and can be expressed numerically as the radon potential (RP) of a building site, which depends on the soil characteristics and the building's foundation type. Determining the radon index (RI) of a building site is based on the assessment of radon (^{222}Rn) activity concentration in the soil gas and of the permeability of the underlying soils (Neznal et al., 2004).

The rate of radon release depends on many factors including radium (Ra) concentration, soil grain size, soil porosity and permeability, soil moisture content, atmosphere pressure and precipitation conditions. Thanks to meteorological parameters' changes the soil radon activity concentration shows the short-time variations during a day and the long-time seasonal variations in a year cycle. It seems that the seasonal variations of soil radon activity concentration depend on climate conditions as some authors report the higher values in wet cold autumn and winter months, and the lower values in dry warm spring and summer ones (Titov et al., 1985; Matolín & Prokop, 1992; Matolín, 1994; Holý et al., 1995, 1997; Winkler et al., 2001; Mojzeš, 2004, 2005, 2007; Mojzeš & Putiška, 2006, 2012; Petersell et al., 2017; Szabó et al., 2013) while the others oppositely (Zmazek et al., 2002; Al-Shereideh et al., 2006; Choubey et al., 2011; Inan et al., 2012; Kamra, 2015). More complicated seasonal soil radon course could be developed under the conditions of faulty site (Font et al., 2008; Moreno et al., 2016; Miklyaev et al., 2021), permeable glacial sediments (Sundal et al., 2008), volcanic areas (Cigolini et al., 2009), groundwater saturated soils (Perrier et al., 2009) especially in hilly regions.

The annual variations of the radon (^{222}Rn) and thoron (^{220}Rn) activity concentration in soil air belong to the long-range changes. The measurements are carried out inside the shallow near-surface layer up to a depth of maximally 1 meter for the purposes of applied geophysical, geochemical and geological practice. The reason for the long-range radon and thoron variations lies on strong influencing by intensive changes of meteorological parameters of close atmospheric environment in a year sun cycle. The knowledge of these time changes character in connection with the information about the geological environment properties is the key to both the right assessment of the radon and thoron activity concentration values and to the correct interpretation of the results for studied geological situation anytime during a year cycle.

2. Object of study

The target is to evaluate the results of repeated measurements of the radon and thoron activity concentration in soil air of studied geological environment within the years 2003 – 2005, to give their statistical assessment and to find out the most important dependencies between the values of ^{222}Rn , resp. ^{220}Rn activity concentration in soil air and single meteorological parameters (temperature, humidity, pressure and precipitation).

3. Methods used and area of study

Measurements of ^{222}Rn and ^{220}Rn activity concentration in soil air of 0.8 m depth were carried out manually in a single place once per week during the period from 21.1.2003 to 23.6.2005 when these measurements were stopped. This kind of weekly, resp. daily data set is mostly represented by 5 values (quintuplet) acquired by the immediate measurements of soil air samples taken from the same probe in 10 – 13 minutes' intervals. This way, one week's, resp. one day's measurements were covering an approx. 1.5-hours' long day period and usually from 8:00 to 10:00 in the morning. Several interruptions of radon and thoron measurements (from 20.3.2003 to 5.6.2003, from 24.6.2003 to 14.8.2003 excluding 16.7.2003, from 19.12.2003 to 12.1.2004 and from 10.8.2004 to 6.9.2004) were caused by personal absence of the

author for another project works. This is the risk of one-man manual measurement. Even though the lack of 2003 data is considerable, the attempt of its regular evaluation within the 2003 – 2005 data set overview is undertaken.

The portable radon detector LUK-3R (producer Special Measurement Methods Ltd., Prague, Czech Republic) based on scintillation Lucas cells detection was used.

All radon and thoron values were taken from one measurement station – a probe installed inside the Faculty of Natural Sciences (Comenius University in Bratislava) campus (Fig. 1, site Rn). The area lies on the southward oriented slope of the Malé Karpaty Mts. with easy gradient to the Danube River terrace in the SW part of Bratislava. The basement is built by the mixture of slope loams and sandy-gravel Quaternary terrace sediments. The uppermost layer where the soil radon and thoron measurements were performed is a heterogeneous building made-up ground. Its clay particles content is approx. 49 % what means a middle permeable environment to radon and thoron gas movement (Regulation of Ministry of Environment of Slovak Republic No.1/2000-3).

All meteorological data were provided by close meteorological station managed by the Department of Astronomy, Physics of the Earth and Meteorology of the Faculty of Mathematics, Physics and Informatics located approx. 300 m away from the radon and thoron measurement station in the same university campus (Fig. 1, site Meteo). The atmospheric temperature, humidity, pressure and precipitation data cover the period from 1.1.2003 to 30.6.2005 with hourly frequency.

4. Results of study and discussion

There were 519 measurements carried out for 104 days (104 weeks) in the period from 21.1.2003 to 23.6.2005. Time courses of measured content of ^{222}Rn , resp. ^{220}Rn isotopes in soil air are presented in Figs. 2 and 3 in the bottom part (the ^{222}Rn , resp. ^{220}Rn activity concentration in $\text{kBq}\cdot\text{m}^{-3}$). The polynomial curve (bold line) fitted through measured values underlines the seasonal course of both isotopes' changes (variations).

For the possibility to compare the time courses of ^{222}Rn and ^{220}Rn activity concentration with their probable reasons there are also presented the time courses of observed atmospheric air parameters, i.e. temperature, humidity and pressure in Figs. 2 and 3, and also the precipitation deposit in Fig. 4b,d. Their dependence is well known from former works, e.g. Matolín and Prokop, 1992; Holý et al., 1995; Mojzeš, 2004; Mojzeš and Putiška, 2006 and Mojzeš, 2007.

Basic statistical characteristics for all studied variables are presented in Table 1.

For completeness the dependence of the ^{222}Rn and ^{220}Rn activity concentration on single meteorological parameters by the coefficients of linear correlation is presented in Table 2.

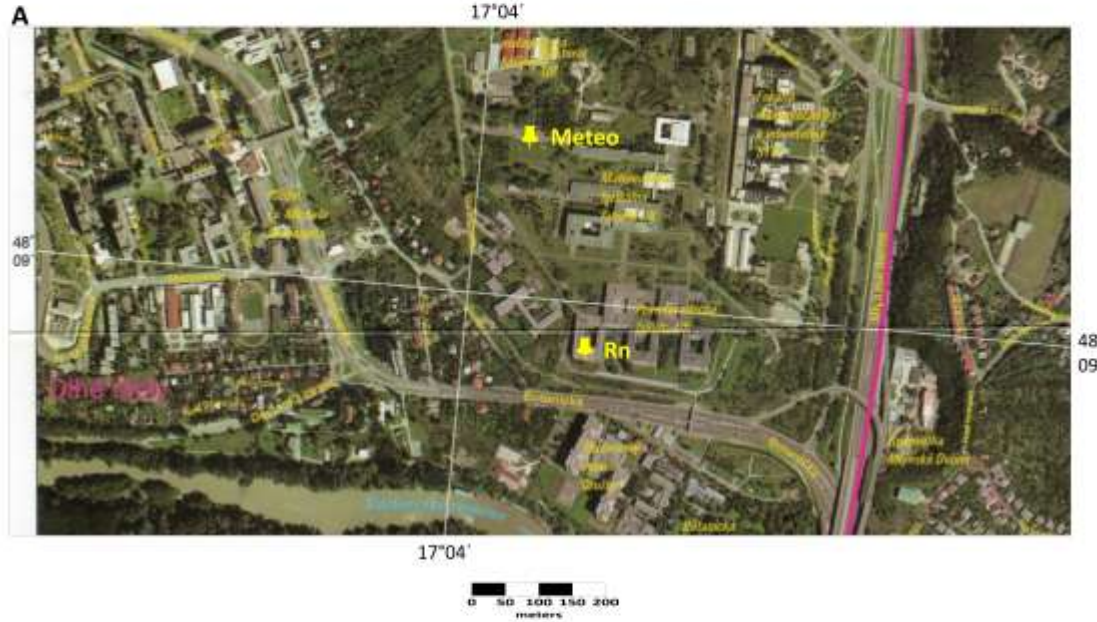


Fig. 1 Localization of the measurements' stations on the background of A) Bratislava – Orthophotomap 1:10 000 (VKÚ & GEODIS SLOVAKIA, 2002) and B) Regional geological maps of Slovakia 1:50 000 (Polák et al., 2011) (Explanations: Rn – site of soil air radon measurements (N 48°08'56.07", E 17°04'10.07"), Meteo – site of meteorological measurements; geological legend: 1 – anthropogeneous deposits: embankments, waste and mine dumps (subrecent – recent), 2 – fluvial, lithofacially undivided loams, sandy loams, loamy sands to gravels of the lower flood-plain level (Late Holocene), 8 – fluvial, lithofacially undivided loams, sandy loams, loamy sands to gravels of valley alluvial plains of rivers and streams, 15 – deluvial-fluvial outwash (rainwash) loams with gravels, 16 - deluvial-fluvial outwash (rainwash) loams with rock fragments, 21 – deluvial loams to sandy loams, 24 – deluvial loamy and sandy-loamy gravels and rock fragments, 31 – eolian silty, subsidiary fine-sandy loams – loess (Würm), 35 – fluvial sandy gravels of lower middle terraces (Late Riss) with cover of loess (Late Würm), 36 – fluvial sandy gravels of lower middle terraces (Late Riss), 39 – fluvial sandy gravels and gravels of the upper middle terraces (Early Riss) with loess cover (Late Würm), 40 – fluvial sandy gravels and gravels of upper middle terraces (Early Riss), 44 – fluvial sandy gravels of undivided two levels of upper terraces (Mindel undivided) with loess cover (Late Würm), 45 – fluvial sandy gravels of undivided two levels of upper terraces (Mindel undivided)).

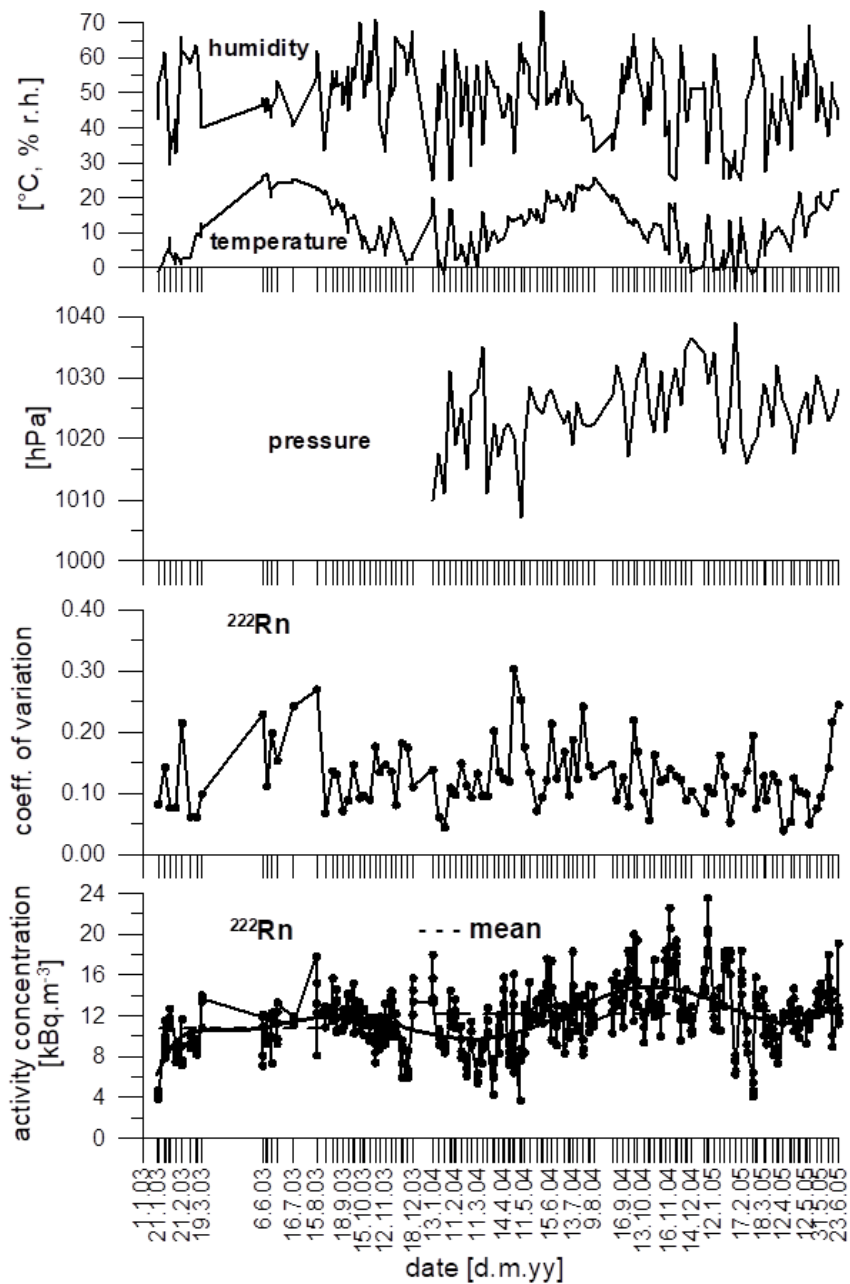


Fig. 2 Time courses of single measured and calculated variables of ^{222}Rn (radon)

The ^{222}Rn curve (Fig. 2, bottom) presents repeated maxima in autumn and winter months while minima appear in spring months. The most expressive maximum (approx. $15 \text{ kBq}\cdot\text{m}^{-3}$ with single peaks up to $24 \text{ kBq}\cdot\text{m}^{-3}$) was measured in October 2004 – January 2005. The second less expressive maximum (approx. $12 \text{ kBq}\cdot\text{m}^{-3}$ with single peaks up to $16 \text{ kBq}\cdot\text{m}^{-3}$) was recorded in September – October 2003. There is possible to see three minima on the curve: the first one in January – March 2003 (approx. $9 \text{ kBq}\cdot\text{m}^{-3}$ with single drops up to around $4 \text{ kBq}\cdot\text{m}^{-3}$), the second one in January – April 2004 (approx. $10 \text{ kBq}\cdot\text{m}^{-3}$ with single drops up to around $4 \text{ kBq}\cdot\text{m}^{-3}$) and the third one in February – April 2005 (approx. $11 \text{ kBq}\cdot\text{m}^{-3}$ with single drops up to around $4 \text{ kBq}\cdot\text{m}^{-3}$). It can be seen slight increase in average values of both minimum and maximum in time. The average minimum starts from approx. $9 \text{ kBq}\cdot\text{m}^{-3}$ in 2003 to $10 \text{ kBq}\cdot\text{m}^{-3}$ in 2004 and $11 \text{ kBq}\cdot\text{m}^{-3}$ in 2005. The average maximum starts from $12 \text{ kBq}\cdot\text{m}^{-3}$ in 2004 to $15 \text{ kBq}\cdot\text{m}^{-3}$ in 2005. The baseline (mean) drawn in ^{222}Rn , resp. ^{220}Rn activity concentration graphs in Figs. 2 and 3 at the average annual levels (Table 1: $11.8 \text{ kBq}\cdot\text{m}^{-3}$ for ^{222}Rn , resp. $36.2 \text{ kBq}\cdot\text{m}^{-3}$ for ^{220}Rn) divides the fitted course of both variables into two parts in each year: the periods from November-January to May-July are usually below the mean baseline level and the periods from May-July to November-January are usually above the mean baseline level. It seems that the rainy season which is more or less typical in May – July period is the beginning of soil radon increase during the year cycle. It could follow from previously that the ^{222}Rn activity concentration in soil air should have the strongest dependence on measured humidity of atmospheric air. But the values of coefficients of linear correlation presented in Table 2 are not very significant. The ^{222}Rn activity concentration shows the most stable positive correlation to temperature of atmospheric air (from 0.23 to 0.40 in single periods), less stable negative correlation to humidity of atmospheric air (from -0.05 to -0.27 in single periods) and variable one to pressure of atmospheric air.

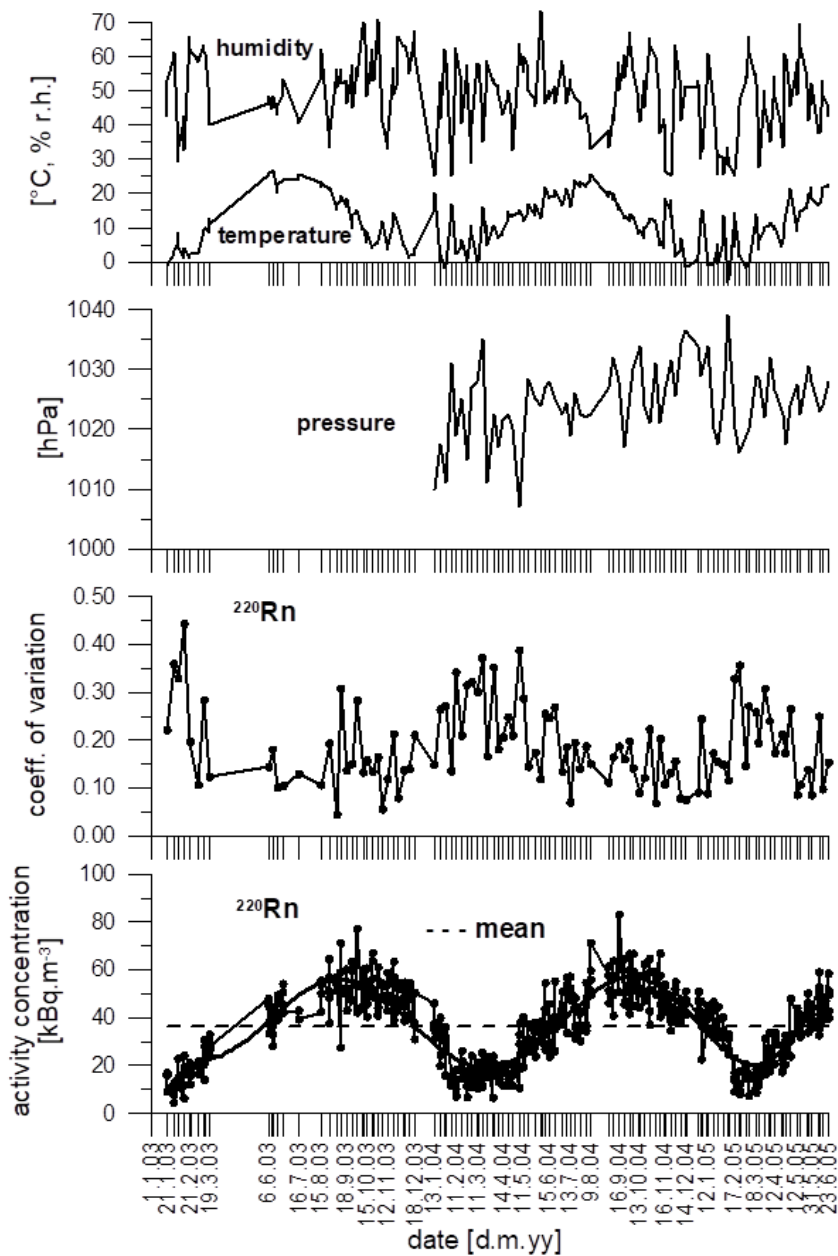


Fig. 3 Time courses of single measured and calculated variables of ^{220}Rn (thoron).

In the case of ^{220}Rn measurement with LUK-3R, the radon detector gives only estimated value of activity concentration in soil air sample with average assessment error of about 25 % (Plch, 1994). The ^{220}Rn course (Fig. 3, bottom) shows that the values have higher range (Table 1), the curve is more uniform and forms a wave line more expressive than the ^{222}Rn one. The same intensive maxima were measured in August – October 2003 and September – November 2004 accompanied by three intensive minima in January – March 2003 and February – April 2004 and 2005. They correlate very good with ^{222}Rn minima and maxima, resp. they are slightly preceding in time. It follows from Table 2 that the ^{220}Rn activity concentration has the most stable positive correlation to temperature of atmospheric air (from 0.29 to 0.40 in single periods) which is a little stronger than in ^{222}Rn case. Next stable positive correlation is to atmospheric pressure (from 0.27 to 0.29). No significant correlation exists to the humidity of atmospheric air.

The strongest correlation for both ^{222}Rn and ^{220}Rn activity concentration is the positive dependence on temperature of atmospheric air and it is a little stronger for ^{220}Rn . At the same time their phase delay behind the curve of atmospheric air temperature is evident in Fig. 2 and 3 (Mojzeš, 2004; Mojzeš and Putiška, 2006; Mojzeš, 2007). Polynomial fits of the ^{222}Rn and ^{220}Rn activity concentration and air temperature courses in Fig. 4a,c allow easily estimation of time shift approximately between two - three months (Mojzeš and Putiška, 2012). The delay is simultaneously the reason for low linear correlation values.

Tab. 1 Basic statistical parameters for all measured variables for whole measured period 21.1.2003 – 23.6.2005 and single years

	whole period 21.1.2003 – 23.6.2005						year 2003 21.1. – 18.12.2003					
	Rn	Tn	T	H	P	PD	Rn	Tn	T	H	P	PD
No.	519	519	421	421	421	413	151	151	144	144	144	144
Min	3.7	4.5	-12.2	32.0	728.1	0.0	3.9	4.5	-8.4	41.0	730.0	0.0
Max	23.6	83.1	31.8	100.0	761.3	35.9	17.8	77.2	31.8	100.0	759.8	29.4
Range	19.9	78.6	44.0	68.0	33.2	35.9	13.9	72.7	40.2	59.0	29.8	29.4
Mean	11.8	36.2	10.3	74.4	745.1	3.2	10.8	40.8	11.3	73.1	745.2	3.0
SD	3.1	15.8	9.3	15.0	6.0	4.8	2.5	16.1	10.1	15.4	5.9	4.3
Var	9.9	248.0	86.3	225.2	35.4	23.4	6.2	257.8	101.4	237.5	34.8	18.0
CoV	0.27	0.44	0.90	0.20	0.01	1.50	0.23	0.39	0.89	0.21	0.01	1.42

	year 2004 13.1. – 31.12.2004						year 2005 4.1. – 23.6.2005					
	Rn	Tn	T	H	P	PD	Rn	Tn	T	H	P	PD
No.	243	243	192	192	192	184	125	125	85	85	85	85
Min	3.7	6.6	-12.2	32.0	728.1	0	4.1	7.3	-7.1	40.0	730.3	0
Max	22.6	83.1	26.6	97.0	758.5	35.9	23.6	59.0	27.4	97.0	761.3	16.2
Range	18.9	76.5	38.8	65.0	30.4	35.9	19.5	51.7	34.5	57.0	31.0	16.2
Mean	12.2	36.2	10.7	75.6	745.2	3.6	12.4	30.6	7.7	74.2	744.7	2.9
SD	3.2	16.2	8.8	14.6	5.9	5.6	3.4	12.4	8.6	15.1	6.1	3.9
Var	10.2	263.7	76.9	213.9	35.2	31.4	11.7	152.7	74.7	228.7	37.4	15.2
CoV	0.26	0.45	0.82	0.19	0.01	1.58	0.28	0.40	1.13	0.20	0.01	1.33

Legend:

- Rn - activity concentration of ^{222}Rn in soil air [$\text{kBq}\cdot\text{m}^{-3}$],
- Tn - activity concentration of ^{220}Rn in soil air [$\text{kBq}\cdot\text{m}^{-3}$],
- T - temperature of atmospheric air [$^{\circ}\text{C}$],
- H - humidity of atmospheric air [% r.h.],
- P - pressure of atmospheric air [mm Hg],
- PD - precipitation deposit [mm],
- No. - number of measurements,
- SD - standard deviation (σ),
- Var - variance,
- CoV - coefficient of variation (V_k).

The detailed view on the ^{222}Rn , resp. ^{220}Rn activity concentration curve (Figs. 2, 3) shows that it consists of a chain of 5-points' clusters of values measured within a single day (week) which are separated from each other by approx. one week. The clusters of daily measurements (quintuplets) have different consistence – the points in some quintuplets are near to each other but in another ones, they are more scattered. For the evaluation of variability of the radon activity concentration inside 104 quintuplets of daily measurements the coefficient of variation V_k was used

$$V_k = \sigma / \varnothing, \quad (1)$$

where σ – standard deviation, \varnothing – mean.

Time course of the coefficient of variation is presented as the second graph from bottom up in Fig. 2 for the ^{222}Rn activity concentration and in Fig. 3 for the ^{220}Rn activity concentration. Their polynomial fits are presented in Fig. 4b,d together with the precipitation deposit amount.

Higher importance in evaluation is given to the coefficient of variation of the ^{222}Rn activity concentration because these values are determined with higher reliability. The year course of the coefficient of variation of the ^{222}Rn activity concentration shows three usually 1 – 2 weeks long periods with higher values. The first period is in June – July 2003, the second one in April/ May 2004 and the third one in June 2005 (Fig. 4b). The higher variability of daily measurements' values here could be caused exactly as the result of starting rainy season (higher precipitation).

In the case of time course of the coefficient of variation of the ^{220}Rn activity concentration (Fig. 3, 2nd graph from bottom) there is evident negative correlation to ^{220}Rn measured quantity – higher and unstable values of the coefficient of variation are connected with low ^{220}Rn content in soil air but lower and more balanced ones are in periods with sufficient ^{220}Rn production in soil air which shows expressive dependence on environment temperature probably through the coefficient of emanation (Fig. 4d).

Tab. 2 *Coefficients of linear correlation between measured variables for whole measured period 21.1.2003 – 23.6.2005, single years and smaller periods*

whole period 21.1.2003 – 23.6.2005	^{222}Rn activity concentration in soil air	^{220}Rn activity concentration in soil air
Temperature of atm. air	0.19	0.40
Humidity of atm. air	-0.07	-0.00
Pressure of atm. air	0.07	0.21
Precipitation deposit	0.01	-0.05
Number of correlated pairs	104	

year 2003 21.1. – 18.12.2003	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.48	0.49
Humidity of atm. air	-0.38	-0.17
Pressure of atm. air	0.10	0.28
Precipitation deposit	-0.03	0.17
Number of correlated pairs	31	
year 2004 13.1. – 31.12.2004	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.16	0.31
Humidity of atm. air	-0.01	0.11
Pressure of atm. air	0.18	0.19
Precipitation deposit	-0.04	-0.13
Number of correlated pairs	48	
year 2005 4.1. – 23.6.2005	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.03	0.42
Humidity of atm. air	0.14	-0.14
Pressure of atm. air	-0.14	0.18
Precipitation deposit	0.11	0.13
Number of correlated pairs	25	
1st half of year 2003 21.1. – 30.6.2003	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.41	0.92
Humidity of atm. air	-0.58	-0.75
Pressure of atm. air	0.01	0.28
Precipitation deposit	0.08	0.29
Number of correlated pairs	12	
2nd half of year 2003 1.7. – 31.12.2003	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air

Temperature of atm. air	0.58	0.22
Humidity of atm. air	-0.42	-0.23
Pressure of atm. air	-0.01	-0.04
Precipitation deposit	-0.11	0.39
Number of correlated pairs	19	
1st half of year 2004 1.1. – 30.6.2004		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.45	0.47
Humidity of atm. air	-0.26	0.15
Pressure of atm. air	0.11	-0.10
Precipitation deposit	0.00	0.09
Number of correlated pairs	24	
2nd half of year 2004 1.7. – 31.12.2004		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	-0.33	0.03
Humidity of atm. air	0.18	0.01
Pressure of atm. air	-0.05	-0.02
Precipitation deposit	0.24	0.09
Number of correlated pairs	24	
winter 2003 21.1. – 20.3.2003		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.51	0.66
Humidity of atm. air	-0.59	-0.44
Pressure of atm. air	-0.02	0.56
Precipitation deposit	-0.09	-0.26
Number of correlated pairs	8	
spring 2003 20.3. – 21.6.2003		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
no measured data		
summer 2003		
	²²²Rn activity concentration	²²⁰Rn activity concentration

21.6. – 23.9.2003	in soil air	in soil air
Temperature of atm. air	-0.15	-0.74
Humidity of atm. air	0.28	0.42
Pressure of atm. air	-0.32	0.45
Precipitation deposit	0.34	-0.58
Number of correlated pairs	7	
autumn 2003 23.9. – 21.12.2003		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.55	0.40
Humidity of atm. air	-0.42	-0.53
Pressure of atm. air	0.12	-0.12
Precipitation deposit	-0.08	0.57
Number of correlated pairs	13	
winter 2003/2004 21.12.2003 – 20.3.2004		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.38	-0.17
Humidity of atm. air	-0.34	0.14
Pressure of atm. air	-0.30	-0.58
Precipitation deposit	0.03	-0.03
Number of correlated pairs	10	
spring 2004 20.3. – 21.6.2004		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.39	0.74
Humidity of atm. air	-0.14	0.34
Pressure of atm. air	0.52	0.28
Precipitation deposit	-0.07	0.13
Number of correlated pairs	13	
summer 2004 21.6. – 23.9.2004		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	-0.68	-0.29
Humidity of atm. air	0.49	-0.07

Pressure of atm. air	-0.15	0.25
Precipitation deposit	0.66	0.16
Number of correlated pairs	12	
autumn 2004 23.9. – 21.12.2004		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	-0.38	0.68
Humidity of atm. air	0.14	0.17
Pressure of atm. air	-0.30	0.23
Precipitation deposit	0.19	0.11
Number of correlated pairs	8	
winter 2004/2005 21.12.2004 – 20.3.2005		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.47	-0.01
Humidity of atm. air	-0.28	-0.01
Pressure of atm. air	-0.03	0.49
Precipitation deposit	0.08	0.06
Number of correlated pairs	16	
spring 2005 20.3. – 23.6.2005		
	²²²Rn activity concentration in soil air	²²⁰Rn activity concentration in soil air
Temperature of atm. air	0.21	0.75
Humidity of atm. air	0.29	0.29
Pressure of atm. air	-0.35	0.04
Precipitation deposit	-0.05	-0.01
Number of correlated pairs	14	

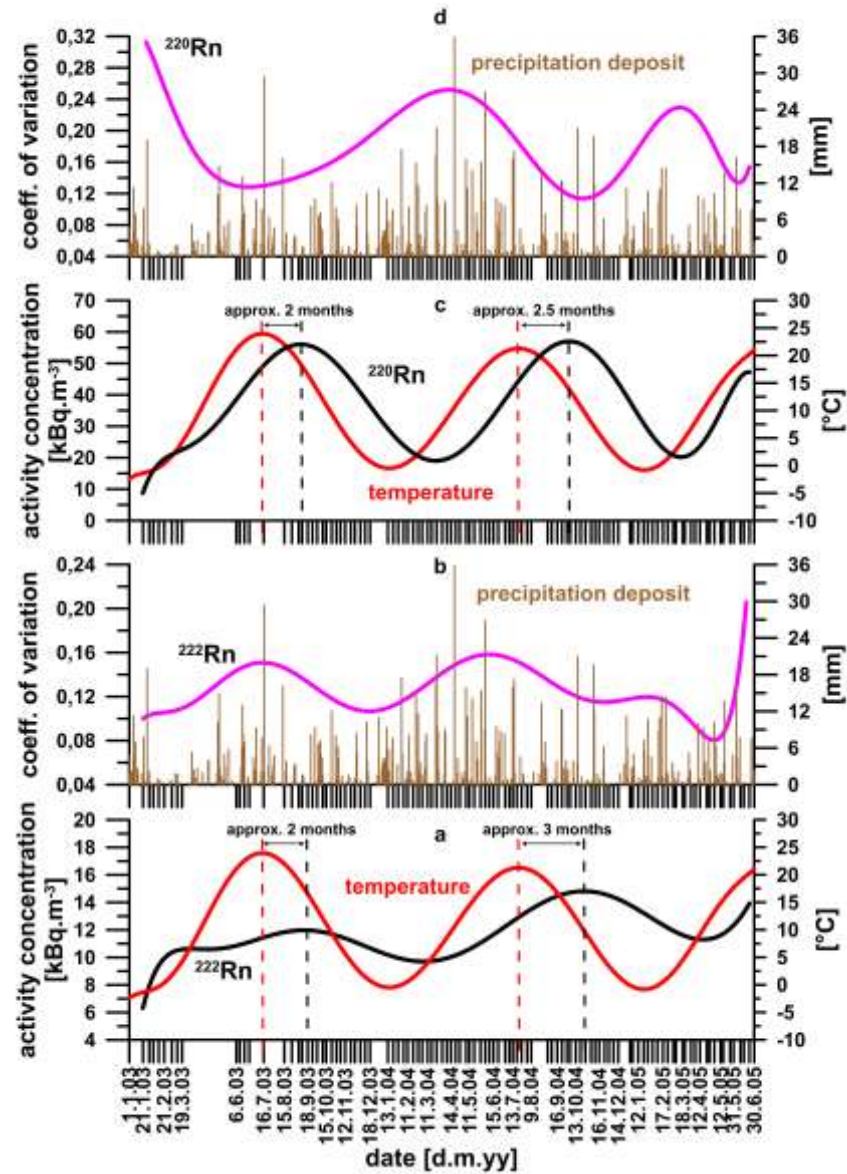


Fig. 4. Polynomial smoothed time courses of single measured and calculated variables of ^{222}Rn (radon), ^{220}Rn (thoron), air temperature and precipitation deposit with marked time shifts

5. Conclusions

The results document a close connection between radon content in pore space of soil cover and some meteorological parameters of aerial atmospheric environment in dependence on weather changes in a year cycle. The known wave character of the ^{222}Rn and ^{220}Rn activity concentration courses during whole year-around cycle was confirmed: the highest values during autumn and the lowest ones during spring months what reflect also the highest values of linear correlation to the temperature of atmospheric air.

The monitoring site is located in the Quaternary slope deposits on the left bank of the Danube River and, as can be seen from the geological situation (Fig. 1), is not affected by any tectonic fault. This allows us to conclude that the predominant way of radon release and movement in the porous soil environment is diffusion. This assumption is also confirmed by the variability of the measured values of the activity concentration of ^{222}Rn and ^{220}Rn in the soil air, which is significantly lower compared to tectonically based localities (Mojzeš, 2004).

An attempt to express the dependence of the measured values of soil radon activity concentration on temperature, humidity and atmospheric air pressure by means of linear correlation was not successful (Tab. 2) since most of the coefficients were not significant. The reason is the time shift of the course of changes between the measured physical parameters of the atmospheric and soil environment. This is most obvious when comparing the concentration of soil radon activity with atmospheric temperature. As can be seen from Fig. 4a,c, the time lag of the ^{222}Rn and ^{220}Rn activity concentration curves behind the atmospheric air temperature curve is between 2 and 3 months. This finding agrees very well with the results of monitoring and modeling of heat diffusion in groundwater (Hodasová et al., 2020): “The measurements manifest a seasonal change in temperature (sinusoidal course) with a phase shift (maximum) of approximately 3 months (maximum air temperature is reached in August and groundwater in November). The phase shift and magnitude of the amplitude were the same in all three wells, indicating that the groundwater heat comes from the same source. In our case, it can be said that the main source of heat for groundwater was heat transferring from the surface.”

An attempt at a more detailed analysis of the causes of the variability of the daily measurement sets (Fig. 4b,d) may indicate that, in addition to the contribution of the fluctuation of radioactive alpha decay itself, both temperature and soil moisture may participate in the variability, in our case represented by the temperature of the atmospheric air and the amount of precipitation deposit entering the soil. However, these dependencies are not entirely clear from our results and require further study. For example, Fig. 4b,d could indicate a positive correlation of the coefficient of variation of ^{222}Rn activity concentration with both temperature and precipitation, and in the case of ^{220}Rn there could be a negative correlation with temperature. Variability of daily measurements (represented in the form of coefficient of variation of radon and thoron activity concentrations) is the highest exactly during transitional period from cold to warm part of the year, in the period from April to July when also the rainy season appears.

The results show the need to compare the measured ^{222}Rn and ^{220}Rn activity concentrations in soil air with temperature and humidity not only of atmospheric air but also in soil layer down to the depth of soil air sampling. Then this dependence could be presented even more strongly through the comparison with the temperature of the soil environment that is delayed after atmosphere.

In conclusion, it is necessary to state what was already mentioned in the introduction of the contribution, that the observed variations and their course are typical for similar geological environments and especially for areas with similar one's climatic conditions, and in other environments and in other climatic conditions may be different. For example, the existence of a wet rainy period in May to July is mentioned for Bratislava region (the works of Holý and Mojžeš), which are not mentioned by the authors of measurements from other regions (for example, the works of Matolín, Titov and others).

Acknowledgements

This contribution was worked out within the ambit of the project VEGA No. 1/0468/10 „Spatial and time demonstration of the unsaturated zone in geophysical fields“. The additional processing was worked out thanks to the projects APVV-21-0159, VEGA No. 1/0107/23 and VEGA No. 1/0587/24.

References:

- AL-SHEREIDEH, S.A., BATAINA, B.A. & ERSHAIDAT, N.M. Seasonal variations and depth dependence of soil radon concentration levels in different geological formations in Deir Abu-Said District, Irbid – Jordan. *Radiation Measurements*, 2006, 41, p. 703-707
- CHOUHEY, V.M., ARORA, B.R., BARBOSA, S.M., KUMAR, N. & KAMRA, L. Seasonal and daily variation of radon at 10 m depth in borehole, Garhwal Lesser Himalaya, India. *Applied Radiation and Isotopes*, 2011, 69, p. 1070-1078
- CIGOLINI, C., POGGI, P., RIPEPE, M., LAIOLO, M., CIAMBERLINI, C., DELLE DONNE, D., ULIVIERI, G., COPPOLA, D., LACANNA, G., MARCHETTI, E., PISCOPO, D. & GENCO, R. Radon surveys and real-time monitoring at Stromboli volcano: influence of soil temperature, atmospheric pressure and tidal forces on ²²²Rn degassing. *Journal of Volcanology and Geothermal Research*, 2009, 184, p. 381-388
- FONT, LI., BAIXERAS, C., MORENO, V. & BACH, J. Soil radon levels across the Amer fault. *Radiation Measurements*, 2008, 43, p. 319-323
- HODASOVÁ, K., KRČMÁŘ, D. & ZATLAKOVIČ, M. Assessment of the influence of a building upon groundwater temperature pattern using numerical modelling. *Acta Geologica Slovaca*, 2020, 12(2), p. 161-170
- HOLÝ, K., BÖHM, R., POLÁŠKOVÁ, A. & ŠTELINA, J. Variations of ²²²Rn concentration in outdoor atmosphere and in soil air. *In the Conference Proceedings of 19th Radiation Hygiene Days, Jasná pod Chopkom*, 1995, p. 129-131
- HOLÝ, K., BÖHM, R., MATOŠ, M., POLÁŠKOVÁ, A. & HOLÁ, O., 1997: The results of three-years continual monitoring of ²²²Rn volume activity in soil air and the approaches to its variations' interpretation (Výsledky trojročného kontinuálneho monitorovania objemovej aktivity ²²²Rn v pôdnom vzduchu a prístupy k interpretácii jej variácií). *In the Conference Proceedings of "Radioactivity in Environment"*, Spišská Nová Ves, 1997, p. 49-51 (in Slovak)

- INAN, S., KOP, A., CETIN, H., KULAK, F., PABUÇCU, Z., SEYIS, C., ERGINTAV, S., TAN, O., SAATÇILAR, R. & BODUR, M.N. Seasonal variations in soil radon emanation: long-term continuous monitoring in light of seismicity. *Nat Hazards*, 2012, 62, p. 575-591
- INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA) *Design and Conduct of Indoor Radon Surveys*. Safety Reports Series No. 98, Vienna, 2019, 128 p.
- KAMRA, L. Seasonal emanation of radon at Ghuttu, northwest Himalaya: differentiation of atmospheric temperature and pressure influences. *Applied Radiation and Isotopes*, 2015, 105, p. 170-175
- MATOLÍN, M. *Assessment of radon risk from geological basement (technical texts)*. (*Stanovení radonového rizika geologického podloží (technické texty)*). Charles University, Faculty of Sciences, Prague, 1994 (in Czech)
- MATOLÍN, M. & PROKOP, P. Variation of radon volume activity in soil air in a year climatic cycle. *Radon investigations in Czechoslovakia III.*, Geological Survey, Prague, 1992, p. 1-5
- MIKLYAEV, P.S., PETROVA, T.B., SHCHITOV, D.V., SIDYAKIN, P.A., MURZABEKOV, M.A., MARENYY, A.M., NEFEDOV, N.A. & SAPOZHNIKOV, Y.A. The results of long-term simultaneous measurements of radon exhalation rate, radon concentrations in soil gas and groundwater in the fault zone. *Applied Radiation and Isotopes*, 2021, 167, 109460
- MOJZEŠ, A. Time courses of soil radon volume activity in selected areas of Bratislava. *Contr. Geophys. Geod.*, 2004, 34, 4, p. 405-412
- MOJZEŠ, A. Time courses of soil radon volume activity in selected areas. *Transactions of the VŠB – Technical University of Ostrava, Civil Engineering Series*, 2005, Vol. 5, No. 2, p. 97-103 (in Slovak)
- MOJZEŠ, A. Contribution to analysis of time courses of soil radon concentration (years 1997 - 1998). *Contr. Geophys. Geod.*, 2007, 37, 1, p. 29-42
- MOJZEŠ, A. & PUTIŠKA, R. Contribution to analysis of time courses of soil radon concentration (year 2004). *Contr. Geophys. Geod.*, 2006, 36, 1, p. 49-62
- MOJZEŠ, A. & PUTIŠKA, R. Contribution to analysis of time courses of radon volume activity in soil air. *Acta Geologica Slovaca*, 2012, 4, 1, p. 1-5 (in Slovak)
- MORENO, V., BACH, J., FONT, LI., BAIXERAS, C., ZAROCCA, M., LINARES, R. & ROQUÉ, C. Soil radon dynamics in the Amer fault zone: an example of very high seasonal variations. *Journal of Environmental Radioactivity*, 2016, 151, p. 293-303
- NEZNAL, M., NEZNAL, M., MATOLÍN, M., BARNET, I. & MIKŠOVÁ, J. *The new method for assessing the radon risk of building sites*. Czech Geological Survey Special Papers 16, Prague, 2004, 48 p. Available from: <https://www.radon-vos.cz/pdf/metodika.pdf>
- PERRIER, F., RICHON, P. & SABROUX, J.-C. Temporal variations of radon concentration in the saturated soil of Alpine grassland: The role of groundwater flow. *Science of the Total Environment*, 2009, 407, p. 2361-2371
- PETERSELL, V., TÄHT-KOK, K., KARIMOV, M., MILVEK, H., NIRGI, S., RAHA, M. & SAARIK, K. Radon in the soil air of Estonia. *Journal of Environmental Radioactivity*, 2017, 166, p. 235-241
- PLCH, J. *LUK-3R. Integrated system for radon and thoron volume activity measurements*. Manuscript. SMM, Prague, 1994 (in Czech)

- POLÁK, M., PLAŠIENKA, D., KOHÚT, M., PUTIŠ, M., BEZÁK, V., FILO, I., OLŠAVSKÝ, M., HAVRILA, M., BUČEK, S., MAGLAY, J., ELEČKO, M., FORDINÁL, K., NAGY, A., HRAŠKO, Ľ., NÉMETH, Z., IVANIČKA, J. & BROSKA, I. *Geological map of Malé Karpaty Mts. Regional geological maps of Slovakia 1:50 000 (Geologická mapa Malých Karpát. Regionálne geologické mapy Slovenska 1:50 000)*. Ministry of Environment SR & State Geological Institute of Dionýz Štúr, Bratislava, 2011 (in Slovak)
Regulation of the Ministry of Environment of the Slovak Republic No.1/2000-3 for construction and editing of natural and artificial radioactivity maps in scale 1:50 000
- SUNDAL, A.V., VALEN, V., SOLDAL, O. & STRAND, T. The influence of meteorological parameters on soil radon levels in permeable glacial sediments. *Science of the Total Environment*, 2008, 389, p. 418-428
- SZABÓ, K.Z., JORDAN, G., HORVÁTH, A. & SZABÓ, C. Dynamics of soil radon concentration in a highly permeable soil based on a long-term high temporal resolution observation series. *Journal of Environmental Radioactivity*, 2013, 124, p. 74-83
- TITOV, V.K., VENKOV, V.A., AVDEJEVA, T.L. & KUVŠINNIKOVA, E.I. *Exposure emanation methods of mineral deposits exploration (Ekspozicionnyje emanacijonnyje metody poiskov mestoroždenij poleznykh iskopaemych)*. Nedra, Leningrad, 1985, 132 p. (in Russian)
- VKÚ & GEODIS SLOVAKIA Bratislava – *Orthophotomap 1:10 000*. Harmanec, 2002 (in Slovak)
- WILKENING, M. *Radon in the Environment*. Studies in Environmental Science 40, Elsevier, 1990, 137 p.
- WINKLER, R., RUCKERBAUER, F. & BUNZL, K. Radon concentration in soil gas: a comparison of the variability resulting from different methods, spatial heterogeneity and seasonal fluctuations. *The Science of the Total Environment*, 2001, 272, p. 273-282
- WORLD HEALTH ORGANIZATION (WHO) *WHO Handbook on Indoor Radon. A Public Health Perspective*. 2009, 110 p.
- ZMAZEK, B., ŽIVČIČ, M., VAUPOTIČ, J., BIDOVEC, M., POLJAK, M. & KOBAL, I. Soil radon monitoring in the Krško Basin, Slovenia. *Applied Radiation and Isotopes*, 2002, 56, p. 649-657

Author:

¹Assoc. Prof. RNDr. Andrej Mojzeš, PhD., Department of Engineering Geology, Hydrogeology and Applied Geophysics, Faculty of Natural Sciences, Comenius University in Bratislava, Ilkovičova 6, SK-842 15 Bratislava, Slovak Republic, andrej.mojzes@uniba.sk