Influence of some climatic elements on radon concentration in Saeva Dupka Cave, Bulgaria

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Abstract: This study reveals the influence of some climatic elements on radon concentration in Saeva Dupka Cave, Bulgaria. The research is based mainly on statistical methods. Radon concentration in the cave is determined by two main mechanisms. The first one is through penetration of radon from soil and rocks around the cave (present all year round, but has leading role during the warm half of the year). The second one is through thermodynamic exchange of air between inside of the cave and outside atmosphere (cold half of the year). Climatic factors that affect radon concentration in the cave are temperatures (air, surface, soil) and amount of water, expressed by precipitation or by volumetric water content in the soil. In the cold half of the year, an important factor is the duration and depth of snow cover. Statistical models and projections show that by the year 2070 CE radon concentration in the cave will increase. This increase will occur primarily in the warm half of the year due to the increase in outside air temperatures that will decrease thermodynamic exchange of air between the cave and the atmosphere. By the middle of the 21st century, it is expected that the studied area will no longer have a permanent snow cover and the temperature will remain the only factor for the increase of radon concentration in the cold half of the year. Water content in the soil and precipitation are important factors for radon concentration in the cave, but they have no significant trend and they are not expected to exert any influence in the next 50 years. The projected increase of radon concentration in Saeva Dupka Cave creates a serious risk for the health of the tour guides working there. That is why it is necessary to develop a plan to protect the health of the cave staff. On the other hand, the projected increase of radon concentration does not pose any significant health risk for the tourists visiting the cave.

Keywords: radon concentration, climate, statistical model, projection, Saeva Dupka Cave

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INTRODUCTION

In recent decades, the issue of climate change has become increasingly important. These changes are mainly expressed through increase of air temperature of the lowest atmospheric layer, the troposphere. This increase in the second half of the 20th century and the beginning of the 21st century (1951–2012) was 0.72°C according to the Fifth Assessment Report of the IPCC (Hartmann et al., 2013). Similar trends in air temperature are observed in Bulgaria (Nojarov, 2014, 2019). Climate change also leads to changes in atmospheric circulation both globally and regionally. In turn, these changes lead to changes in precipitation, cloud cover, snow cover, sunshine duration, wind direction and velocity, and other climatic elements. There has been a significant increase in greenhouse

gases and a decrease in the amount of aerosols in the atmosphere over Bulgaria in recent decades (Nojarov, 2016). This largely explains the significant positive trend in air temperatures. In Bulgaria there is also a serious redistribution in intra-annual course of precipitation (Drenovski & Stoyanov, 2009, 2010), which is due to changes in atmospheric circulation (Nojarov, 2017a, b).

Long lasting studies of Rn-222 (hereafter radon) concentration in caves (e.g., Cigna, 2005; Dumitru et al., 2015) usually show a typical regular periodic course, but with irregular year-to-year variations of concentration values (Kowalczyk & Froelich, 2010; Perrier & Richon, 2010). Their correlation with year-to-year variations of climatic elements has not yet been tested in detail in multiparametric studies for a relatively long period (>8 years) of continuous radon monitoring.

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No systematic radiation studies focused on radon in the caves have been conducted for the territory of Bulgaria, although there are over 6000 known caves. There is only one study focused on the concentration of radon in nine caves (Varbanov et al., 1975), but it does not include the Saeva Dupka Cave. In 2007 a team of karstologists within the academic cooperation between Bulgarian Academy of Science (BAS) and Czech Academy of Sciences (CAS) initiated systematic radiological cave research. During the first years of investigation, the methodology and experimental techniques were tested and adapted, including the design of a measuring chamber for passive detectors and exposure time to optimize the measurement of radon concentration, in accordance with morphological and microclimatic features and seasonal dynamics in selected caves (Turek et al., 2012, 2015). More in-depth speleological monitoring of radon began in 2011 in connection with the experimentally developed model for integrated karst monitoring within the ProKARSTerra project. Saeva Dupka Cave is a major object in the development and testing of this model.

The aim of the study is to reveal the influence of some climatic elements on radon concentration in Saeva Dupka Cave, Bulgaria. The inspection of various climatic elements (different temperatures, precipitation, soil water content, snow cover, wind speed and direction) has showed that those that have a significant influence are air and soil temperatures, precipitation, volumetric soil water content and snow cover during the cold half of the year. Accordingly, a correlation was made between these climate elements and radon concentration in order to reveal the relationships. Based on these correlations,

statistical models were created that describe the complex influence of the studied climatic elements on radon concentration in the cave and project this concentration in future periods.

OBJECT OF THE STUDY

Saeva Dupka Cave is situated in the transition zone between the Danube Plain and the Balkan Mountains in northern Bulgaria. The cave is developed in gray-white intraclastic, micro-grained or organogenic (often bio-morphic) limestone with a thickness of over 450 m and very high carbonate content, between 97-99% (Kovachev, 1959). In the lithostratigraphic division of Bulgaria these limestones are referred to as "Brestnitsa Formation" (Tsankov et al., 1994).

The cave entrance (4.5 m height/4.2 m width) is in a small rock wreath at 500 m above sea level in the fault slope of the ridge Lednishki Rit, which is the southern limit of the Brestnitsa karst field, the largest in Bulgaria (Popov, 1962, 1969). It is a sub-horizontal cave, 210 m long (170 m along the axis, Fig. 1). The denivelation between the highest part (Cosmos Hall) and the lowest accessible part (Rockfall Hall) is 20 m (denivelation to the cave entrance: +3 m and -17 m). There are four cave halls (Fig. 1), the total cave area is approx. 4,500 m² and the volume is 50,000 m³. There is no permanent water flow in the cave, but an active infiltration of precipitation water takes place through numerous stalactites and rock cracks in the ceiling, which is 8 to 32 m thick. Saeva Dupka Cave is part of the Brestnitsa karst geosystem, which drains into the large karst spring Glava Panega (type Vauclose).

Fig. 1. Saeva Dupka Cave plan and longitudinal profile with radon monitoring sites (1-5) and cave halls: A - Stack Hall, B - Rockfall Hall, C - Concert Hall, and D - Cosmos Hall (Turek et al., 2020).

On the surface above Saeva Dupka Cave there is a typical karst landscape with karren (clints, grikes) and dolines (sinkholes). The soils are humus-carbonate (Rendzic Leptosols, LPk) and are 0.2–0.5 m to 1–1.5 m thick locally in the grooves of the subsoil karren. Bush and tree vegetation with predominance of the formation of eastern hornbeam (*Carpineta orientalis*) grow over the cave. The climate in the area of the cave is of low mountainous type and belongs to the transition zone between subtropical and temperate climates (Nojarov, 2017c). The average annual temperature for the period 1979–2018 is 10.3°C and the average annual amount of precipitation is 845 mm (Nojarov, 2020).

Saeva Dupka is very rich in secondary calcite speleothems and is one of the most beautiful caves in Bulgaria. In 1962 it was declared a protected area (Natural landmark with 20 ha adjacent area). Since 1964 it has been managed by the Bulgarian Tourist Union, and since 1967 it has been a show cave and offers tourist visits (Popov, 1969). This is one of the most visited caves in Bulgaria, with ~50,000 visitors in 2019. The entire tourist route is about 450 m long, and the guided tour of the cave lasts an average 30–40 minutes.

In this study Saeva Dupka was chosen as a representative cave, because the Experimental Laboratory of Karstology at NIGGG-BAS has developed for the cave a model for long term Integrated Monitoring of Karst System (Speleo-MIKS “Saeva Dupka”, <http://www.prokarstterra.bas.bg/lab/EN/base.html>), which includes radon measurements since 2011 (Stefanov et al., 2014; Turek et al., 2020). Saeva Dupka is a major site in the specialized scientific network BGSpeleo-RadNet (for monitoring of radon in Bulgarian caves). The network is based at the NIGGG-BAS (<http://www.prokarstterra.bas.bg/lab/EN/sci-networks.html>) and is maintained in cooperation with the Department of Radiation Dosimetry of the Institute of Nuclear Physics of the Czech Academy of Sciences, Prague. The choice of points for radiological monitoring in the cave is consistent with its morphological features and microclimatic zoning, based on regular profile microclimatic measurement (2009–2014). In general, in Saeva Dupka Cave, three speleoclimatic zones can be recognized: Entrance (perturbation) zone (with outer part at the entrance and inner part (antechamber), a Transition zone (the Stack and Rockfall Halls) and Internal (stable) zone (the Concert Hall) with a high warm part (Cosmos Hall). As should be expected, the Entrance (perturbation) zone shows highest annual amplitudes of microindicators. Temperature fluctuations in outer and inner parts are from 0 to 14.8°C and from 1.9 to 11.7°C, respectively. In the whole cave system, the lowest mean temperature has been observed in the bottom of Rockfall Hall, from 6.2°C (winter) to 10°C (summer). The highest temperature is measured in the Cosmos Hall – from 10.3 to 11.5°C. The relative humidity of the cave is constantly high (98%), and active condensation on the cave walls and ceiling is observed during the warm period from May/June to September/October.

DATA AND METHODS

Monitoring of the concentration of radon in Saeva Dupka Cave (Fig. 1) began in November 2011. This determines the main study period, which is from 08.11.2011 to 02.02.2020 (dates are in DD.MM.YYYY format). The air temperature in the cave was measured manually at 15 points (at irregular intervals from 15 to 45 days) since the beginning of 2010, and since January 2019 there has been continuous (at 10 minutes interval) instrumental monitoring with two weather stations.

In the Saeva Dupka Cave, the monitoring of radon is at four measurement sites (from No. 2 to 5) and one control point (No. 1) above the entrance to the cave (Fig. 1). The passive method of measuring radon concentration is used. Track etch detectors (Durrani & Bull, 1987) were utilized for radon concentration measurement. These passive detectors are commonly used for monitoring (Vaupotić et al., 2001; Lario et al., 2005; Sainz et al., 2007; Paar et al., 2013) because they show a long-term resistance to practically 100% humidity typical for the cave microclimate. Because of the simple design and low price of the detector, simultaneous measurements can be carried out across a large number of measurement points. During exposure, detectors were placed inside plastic chambers. Based on previous experience, small (~20 ml) cylindrical plastic vials with a thin window were used. The window area of 2 cm² is closed hermetically by thin (6 µm) polyethylene foil. This foil allows the penetration of radon into the measuring space by diffusion, but it is impermeable for the radon daughter products. Several α-particle sensitive CR-39 plastic discs (Page Mouldings Ltd, later Track Analysis Systems Ltd, both UK) 16 mm in diameter and 0.5–0.7 mm thick are utilized as track detectors. Exposure intervals vary mostly between 1 week and 4–5 months (on average, approximately 46 days). Processing of the exposed detectors was performed using combined chemical and electrochemical etching, enlarging the primary alpha-particle tracks (Tommasino, 1970).

The described above measurement arrangement was calibrated in the radon chamber at the National Institute for Nuclear, Chemical and Biological Protection, Kamenná near Příbram, Czech Republic (<http://www.sujchbo.cz/>), at different values of the time integral of radon activity within 8–35 kBq m⁻³ hour. The response R is 2.29 ± 0.28 tracks cm⁻² per 1 kBq m⁻³ hour.

Exposed and etched detectors were scanned using a high resolution (4,800 dpi) scanner Epson Perfection 4990. Photo and α-track densities ρ, tracks per cm², were evaluated from binary pictures using the software ImageJ (<http://rsbweb.nih.gov/ij/download.html>). In the case of track densities >5,000 cm⁻², the effectiveness of track counting by the SW decreases due to the track overlapping, therefore an experimentally determined correcting function was used:

$$\rho = \rho_{sw} \left[\left(1 + (9.920 \times 10^{-6}) \rho_{sw} + (1 \times 10^{-9}) \rho_{sw}^2 \right) \right] \quad (1)$$

where ρ_{sw} is track density obtained by Image J.

The mean value of radon activity concentration over the period of exposure t_{exp} (days), here mentioned as $A_{V,exp}$ (kBq m⁻³), is then calculated according to the formula

$$A_{V,exp} = \frac{\rho}{Rt_{exp}} \quad (2)$$

For the entire radon monitoring period (08.11.2011 – 02.02.2020) more than 400 detectors has been processed and evaluated.

In 2018–2019, in the Concert Hall, an automatic measurement (15 minutes interval) of the concentration of radon with the device AlphaE (Saphymo GmbH) has been conducted. Results of the electronic device AlphaE (continual) and track etch detectors in vials (passive, integral) were compared in parallel for three exposure periods with good agreement, $AV(TED) \approx AV(\text{AlphaE})$, $\Delta \leq 11\%$ (Turek et al., 2020).

The manual measurement of the air temperature in the cave is performed every 1–2 months with Asman aspiration psychrometer (measurement accuracy 0.1–0.2°C). The automatic measurement within the monitoring network Speleo-MIKS “Saeva Dupka” is performed with two automatic weather stations VAISALA WTW530 Weather Transmitter Series (Output resolution 0.1°C, accuracy for sensor element $\pm 0.3^\circ\text{C}$). Their location is in accordance with the microclimatic zoning of the cave: the first station was installed in Stack Hall (transitional speleoclimatic zone), and the second one, in Concert Hall (inner zone). At the same time, the outside air temperature (in the cave office) is measured with an automatic weather station VAISALA WTW520 Weather Transmitter Series. The automatic stations are set to measure at 10-minute intervals.

The main climatic elements data source is The European Center for Medium-Range Weather Forecasts (ECMWF), ERA5 reanalysis (Copernicus Climate Change Service (C3S), 2017). The resolution of these data is $0.25^\circ \times 0.25^\circ$ (30×30 km). The study area at this resolution is covered by two grid cells – $42.45\text{--}43^\circ\text{N}$, $24\text{--}24.15^\circ\text{E}$ and $42.45\text{--}43^\circ\text{N}$, $24.15\text{--}24.30^\circ\text{E}$. Accordingly, the data for these two grid cells were averaged. The climatic elements that fall within the scope of this study are as follows: air temperature, precipitation, skin (surface) temperature, soil temperature layer 1 (0–7 cm below surface), soil temperature layer 2 (7–28 cm), soil temperature layer 3 (28–100 cm), soil temperature layer 4 (100–289 cm), volumetric soil water layer 1 (0–7 cm), volumetric soil water layer 2 (7–28 cm), volumetric soil water layer 3 (28–100 cm), volumetric soil water layer 4 (100–289 cm), and snow cover depth in the cold half of the year. It should be noted that soil layers, as defined in ERA5 reanalysis database, in the area of the cave also include bedrock, because the soil layer there is rarely over 100 cm thick. In order to comply with radon measurement periods, climatic elements data were collected from the database “ERA5 hourly data on single levels from 1979 to present”, which allows for acquisition of average values between specific dates that correspond to radon measurements. In this way, a total of 69 comparable subperiods were obtained with measurements of both radon concentration and studied climatic elements. The study period set

up this way is too short to identify any significant trends so data for the studied climatic elements from the database “ERA5 monthly averaged data on single levels from 1979 to present” were also used. The period of this database is from 1979 to 2019 (41 years total) and reveals statistically significant trends in the studied climatic elements. These trends were then used as input data for statistical models that project future radon concentrations in the cave.

Depending on the radon concentration, the year was conditionally divided (according to outside air temperature) into two subperiods; one with low concentration, cold subperiod (November–April) and one with high concentration, warm subperiod (May–October). This makes it possible to avoid, to some extent, the seasonality of the various studied elements, which has a significant impact on the correlation coefficients and, accordingly, on the statistical models used to project future radon concentrations in the cave. When analyzing radon concentration in the different subperiods, a total of eight subperiods were identified, which are of a transient type, i.e., in which radon concentration increases from low to high values (March, April, and May) or decreases from high to low values (October, November). Accordingly, these subperiods are not typical for either the cold half or the warm half of the year and, therefore were excluded from the study. Thus, the subperiods that belong to the cold half of the year are 21 and those that belong to the warm half of the year are 40. Further statistical analyzes and projections are performed for the respective half of the year, warm or cold.

This study uses mainly statistical methods (Wilks, 2006). The level of statistical significance for all calculations is $p < 0.05$. The trend analysis was done by means of linear regression. The linear regression represents the relationship between the independent variable (time) and the dependent variable (different variables – radon concentration, air temperature, precipitation, etc.) through a linear equation based on the observed values. The trends were calculated separately for the warm and cold halves of the year. Spearman’s rank-order correlation coefficient r_s was used in order to establish the relationships between the various climatic elements and radon concentration in the cave. The advantage of this correlation is that it does not depend on the type of statistical distribution of the studied variables. Statistical distributions of the various variables were also checked and the results showed that they are close to normal distribution. This makes it possible to use multiple linear regression models to show the complex influence of climatic elements on radon concentration in Saeva Dupka Cave. The different climatic elements serve as potential predictor variables in the created statistical models, and radon concentration is predicted variable. Which variables will be included as potential predictor variables is determined largely based on Spearman’s rank-order correlation coefficient. The statistical models are created separately for the cold and warm halves of the year. The criterion that was used as to which predictor (climatic variable) would eventually be included in the statistical model was the

adjusted R^2 criterion. These multiple linear regression models are then used to calculate the projected values of radon concentration in the cave for the next 50 years (2021–2070). These calculations use statistically significant trends of the predictors for the period 1979–2019, which is long enough, and not for the period 2011–2019, which is too short for revealing any stable trends. Those variables, which do not have a statistically significant trend for the period 1979–2019, are included in the model with their average values for this period.

RESULTS AND DISCUSSION

Figure 2 shows the course of radon concentration in the Concert Hall for the period 08.11.2011 to 02.02.2020. This is the hall with the highest radon concentration. It can be seen that the course has a clear seasonal cycle with a maximum in summer and a minimum in winter. This cycle coincides with the course of outside atmospheric air temperature, which means that it has a significant impact on radon concentration in the cave. This influence could be exercised in two ways. Firstly, by penetration of radon from the soil and rocks above the cave, which

penetration is accelerated at higher temperatures. As the temperature rises, the soil dries out (cracks and capillaries remain open) and becomes more permeable to radon. Higher temperatures and soil moisture during the warm period lead to higher organic activity, and respectively to higher CO_2 amount in the soil. Acidic soil solutions are formed, which accelerate karst processes favoring the release of radon from the rocks. The influence of precipitation events is also important: during the warm half of the year they are usually intense but of short duration and penetrate only into the upper soil layers, blocking soil aeration and helping to increase the concentration of radon in the soil, and thus radon penetrates downwards. Secondly, through air circulation in the cave, which largely depends on the difference in temperatures inside and outside the cave. Given that the temperature amplitude in the cave is relatively small, the main factor is the temperature of the air outside the cave. The morphology of the cave also contributes to this (Fig. 1), facilitating the intrusion of colder (and denser) air into the cave cavities lower than the entrance. This second mechanism determines the significant difference in radon concentration in the cold and warm half of the year.

Fig. 2. Evolution of radon concentration in the Concert Hall, Saeva Dupka Cave for the period 08.11.2011 - 02.02.2020.

The course of air temperature in the transition zone of the cave (Stack Hall) and outside for the period with continuous automatic measurements in the cave, which includes 2019 and early 2020, as well as the course of radon concentration in Stack Hall, are shown in Figure 3. It can be seen that the maximum temperature of the air in the cave is in November. This is largely due to the specifics of the course of air temperature outside the cave in 2019, October and November showing a retention in temperature decrease, which means that during these months the air temperature was higher than normal. The average annual air temperature in the Stack Hall is 9°C . The morphology of the cave is such that when the air temperature outside the cave drops below 9°C , it descends into the cave because it is colder and heavier and ventilates the cave, thus reducing radon concentration to low values typical for the cold half of the year. When air temperature outside the cave is above 9°C , the warmer outside air cannot penetrate into the cave and radon in the cave starts to accumulate, reaching high values typical for the

warm half of the year. This mechanism of circulation in the cave can be traced very clearly in Figures 2 and 3. Figure 3 shows that only when air temperature outside the cave drops permanently below 9°C , which occurs in the second half of November, a decrease in air temperature in the cave and a corresponding decrease in radon concentration begin (Fig. 2 and 3).

Seasonal variations of air temperatures and radon concentrations along the longitudinal profile of the cave are shown in Figure 4. It is obvious that temperature variations are insignificant in the internal zone of the cave (Concert Hall and especially Cosmos Hall). In this area, the concentration of radon increases sharply, and in the warm half of the year it shows large fluctuations (from 3,150 to 6,850 Bq m^{-3} in the Concert Hall), which may be associated with the climatic features of the respective year. Figure 5 illustrates another established dependence, a delay of more than a month of the annual peak in average radon concentration in the cave (e.g., Concert Hall) compared to the peak of outside air temperature.

Fig. 3. Changes of the air temperature in the transition zone of Saeva Dupka Cave (Stack Hall), outside air temperature and radon concentration for the period 24.03.2019 – 02.02.2020.

Fig. 4. Seasonal variations (cold–warm periods) of the temperatures and average concentrations of radon along the longitudinal profile of the Saeva Dupka Cave (Turek et al., 2020).

Fig. 5. Seasonal radon concentrations (maximum and average) in the Concert Hall and outside air temperature T_{out} (monthly average) during the period 2011–2020 (Turek et al., 2020).

Once this second mechanism of influence of outside air temperature, leading to a pronounced seasonal course of radon concentration in the cave, has been clarified, it should be removed from the primary data in order to see the influence of outside air temperature through the first mechanism. This is done by the described above division of the data into data for the cold half of the year and data for the warm half of the year, with some transition subperiods being removed from the study. Usually these transition subperiods include April when a smooth increase in radon concentration starts, and October or November when a sharp drop in radon concentration occurs. In the subsequent presentation of the results, the processes and relationships between the various studied elements will be presented by the respective half years, thus clarifying, to some extent, the seasonality caused by air circulation in the cave.

Figure 6 shows average annual air temperatures in the cave (Concert Hall) for the period 2010–2019. The figure also depicts the course of the average air temperatures outside the cave in the warm and cold halves of the year. It is clear that air temperature outside the cave in the cold half of the year largely determines the average annual air temperature in the cave. This is due to the revealed mechanism of air

circulation in the cave, which is active only in the cold half of the year. Air temperatures outside the cave in the warm half of the year have practically no direct connection with air temperature in the cave. Some connection could exist through heat exchange with the soil and rock above the cave, but this process is slow and therefore could only have a long term influence. To see if there are any trends, Table 1 shows the corresponding trends in air temperatures for the period 2010–2019. In all four halls where monitoring is carried out, there is a statistically significant positive trend in air temperature. This is largely due to the first three years (2010–2012), which are cold in the cave (Fig. 6). There is no significant trend in air temperatures outside the cave, but it should be noted that only air temperatures in the cold half of the year have positive trend and this coincides with the trend in air temperatures in the cave. This further confirms the conclusion that it is the air temperature outside the cave in the cold half of the year that has the strongest influence on air temperature in the cave. Due to the short-time period, the revealed trends in the cave should be accepted with some caution. Air temperature in the cave follows approximately the trends in air temperature outside the cave, but with some delay.

Fig. 6. Average annual air temperatures in Saeva Dupka Cave (Concert Hall) and average outside air temperature in the warm and cold half of the year for the period 2010–2019.

Table 1. Trends in average annual (or half annual) air temperatures in the different halls of the cave and outside the cave for the period 2010–2019. Statistically significant values are shown in bold font.

Location	T (°C)
Cosmos Hall	0.081
Concert Hall	0.099
Rockfall Hall	0.093
Stack Hall	0.122
Outside the cave – warm half year	-0.032
Outside the cave – cold half year	0.062

The trends in the other parameters for which there are continuous measurements in the period 08.11.2011 – 02.02.2020 for the warm half of the year are shown in Table 2. The third column of Table 2 shows the trends of the parameters for which there are data for the period 1979–2019. The table shows that there is a statistically significant increase in

radon concentration only in the Concert Hall. In the other halls the trends are not significant. There are no statistically significant trends in the other studied elements of the environment external to the cave. This is explained by the short data period. For this reason, the period for these elements was extended back in time to 1979. These data show that all studied temperatures (air, skin and different soil layers) increase significantly at approximately the same rate. Amongst the other elements, only volumetric soil water content layer 4 (the deepest) shows a significant decreasing trend in the warm half of the year. The table also shows that the trends in the second and third columns differ as a sign, which shows the importance of the selected period. However, the trends in the third column should be considered more reliable and they are used in the subsequent projecting of radon concentrations in the cave.

Table 2. Trends in different parameters in the warm and cold half of the year for which there are continuous measurements in the period 08.11.2011 – 02.02.2020 and for which there are data for the period 1979–2019. Statistically significant values are shown in bold font.

Parameters	2011 – 2020 warm	1979 – 2019 warm	2011 – 2020 cold	1979 – 2019 cold
Cosmos Hall – radon (Bq m ⁻³)	1.139		1.4419	
Concert Hall – radon (Bq m ⁻³)	36.701		0.5342	
Rockfall Hall – radon (Bq m ⁻³)	-14.638		1.5595	
Stack Hall – radon (Bq m ⁻³)	3.982		0.3066	
Air temperature outside (°C)	-0.0163	0.0335	0.0113	0.0492
Skin temperature (°C)	-0.0146	0.0398	0.0116	0.0467
Soil temperature layer 1 (°C)	-0.0208	0.0332	0.0021	0.0298
Soil temperature layer 2 (°C)	-0.0150	0.0381	0.0028	0.0251
Soil temperature layer 3 (°C)	-0.0111	0.0384	0.0074	0.0237
Soil temperature layer 4 (°C)	-0.0007	0.0388	0.0160	0.0226
Precipitation (mm)	-0.012514	0.00244	-0.003098	0.00372
Volumetric soil water layer 1 (m ³ m ⁻³)	0.000123	-0.000441	0.000015	-0.000393
Volumetric soil water layer 2 (m ³ m ⁻³)	0.000237	-0.000355	-0.000032	-0.000396
Volumetric soil water layer 3 (m ³ m ⁻³)	0.000027	-0.000240	-0.000372	-0.000417
Volumetric soil water layer 4 (m ³ m ⁻³)	0.000082	-0.000658	0.000041	-0.000488
Snow depth (mm)			-0.063207	-0.1402

Table 2 also shows trends in the studied parameters for the cold half of the year, for the periods 08.11.2011 – 02.02.2020 and 1979–2019, respectively. Here, the snow depth is included as an additional element. It can be seen that in the second column (shorter period) there are no statistically significant trends. In the third column (longer period) there are statistically significant trends in all types of temperatures (air, skin and different soil layers). Soil temperature trends are approximately one half those of the air and skin temperatures. This is largely due to the snow cover, which acts as a thermal insulator. However, its depth decreases significantly during the studied period, due to the constantly rising air temperature. At some point, this will lead to its predominant non-permanent existence, and then it is likely that the trends in different temperatures will equalize, as is the situation in the warm half of the year. Amongst the other elements, only volumetric soil water content in layer 1 shows a statistically significant negative trend. It should also be noted that in the studied area precipitation does not show any significant trend either in the cold or in the warm halves of the year.

Table 3 shows the Spearman rank-order correlation coefficients of the relationship between radon concentration in the four cave halls and other parameters in the warm half of the year for the period 08.11.2011 – 02.02.2020. In the Cosmos Hall radon has statistically significant positive correlation coefficients with almost all studied temperatures, except the air temperature outside the cave. The highest correlation coefficient is observed with soil temperature of layer 3. The correlation coefficients with precipitation and volumetric water content in the different soil layers are also significant, and the relationship is inversely proportional (i.e., higher amounts of radon are associated with less precipitation and volumetric water content in the soil layers and vice versa). In the Concert Hall the type of relationships is the same as in the Cosmos Hall, but statistically significant coefficients are less. The highest correlation with radon concentration in this hall has the temperature of soil layer 4 (positive correlation) and volumetric water content in soil layer 1 (negative correlation). In the Rockfall Hall, the only statistically significant correlation coefficients are with temperature of soil

layer 4 (positive correlation) and volumetric water content in soil layer 4 (negative correlation). In the Stack Hall, all correlation coefficients are statistically significant. The table shows that to a large extent the correlation coefficients depend on the location of the different halls in the cave. Cosmos and Stack halls are located higher and closer to the surface (arch thickness above the cave is 10.5–11.5 m and 5–12 m, respectively), and show a better correlation with all the studied elements that reflect the climatic conditions

outside the cave. Stack Hall is also just below the cave entrance and reacts quickly to external climatic fluctuations. It has already been clarified that cave morphology directly affects the ventilation regime in the cave. It should also be noted that during the warm half of the year the temperature (regardless of air, skin, or soil) is positively correlated with radon concentration in the cave. Water (whether as precipitation or volumetric water content in the soil) is negatively correlated with radon concentration in the cave.

Table 3. Spearman rank-order correlation coefficients between radon concentration in the four cave halls and other parameters in the warm and cold half of the year for the period 08.11.2011 - 02.02.2020. Statistically significant coefficients are shown in bold font.

Parameters	Cosmos Hall-radon (rs)		Concert Hall-radon (rs)		Rockfall Hall-radon (rs)		Stack Hall-radon (rs)	
	warm	cold	warm	cold	warm	cold	warm	cold
Air temperature outside (°C)	0.30	0.44	0.26	0.65	0.16	0.17	0.41	0.63
Skin temperature	0.34	0.47	0.27	0.67	0.14	0.18	0.43	0.62
Soil temperature layer 1 (°C)	0.33	0.39	0.26	0.61	0.15	0.27	0.41	0.53
Soil temperature layer 2 (°C)	0.40	0.36	0.37	0.61	0.24	0.30	0.51	0.50
Soil temperature layer 3 (°C)	0.52	0.17	0.53	0.35	0.34	0.19	0.63	0.17
Soil temperature layer 4 (°C)	0.47	-0.15	0.63	-0.05	0.46	0.08	0.61	-0.12
Precipitation (mm)	-0.38	0.12	-0.30	0.12	0.00	0.26	-0.45	-0.01
Volumetric soil water layer 1 (m ³ m ⁻³)	-0.59	0.15	-0.48	0.29	-0.33	0.19	-0.64	0.03
Volumetric soil water layer 2 (m ³ m ⁻³)	-0.59	0.21	-0.48	0.32	-0.30	0.16	-0.60	0.06
Volumetric soil water layer 3 (m ³ m ⁻³)	-0.52	0.22	-0.36	0.31	-0.21	0.08	-0.50	0.09
Volumetric soil water layer 4 (m ³ m ⁻³)	-0.42	0.34	-0.37	0.41	-0.54	0.41	-0.46	0.26
Snow depth (mm)		-0.16		-0.34		0.13		-0.30

Table 3 also shows the Spearman rank-order correlation coefficients of the relationship between radon concentration in the four cave halls and other climatic parameters in the cold half of the year for the period 08.11.2011 - 02.02.2020. In the Cosmos Hall the only statistically significant positive correlation coefficient is between radon concentration and skin temperature. In the Concert Hall, statistically significant positive relationships are observed with air, skin, and the first two soil layers temperatures. There are no statistically significant correlation coefficients in the Rockfall Hall (the deepest in the cave). In the Stack Hall, the significant correlation coefficients are the same as in the Concert Hall. In general, it can be summarized that in the cold half of the year only the temperature (of air, skin, and soil) has any significant influence on radon concentration in the cave and only in its higher parts. During this period of the year air circulation in the cave (inflow of colder, but also with low radon concentration air from outside to inside the cave) is almost constant and the temperature of the air outside the cave is essential for radon concentration inside. Based on these functional relationships, it can be concluded that in the warm half of the year the inflow of radon from soil and rocks is essential for its concentration in the cave, while in the cold half of the year the thermodynamic air exchange between the cave and the atmosphere is more important.

Spearman rank-order correlation coefficients shown in Table 3 were used to determine which predictors will be used initially in multiple linear regression models. There are two groups of similar elements in the studied climatic elements. The first group includes skin temperature and temperatures of the four soil

layers. From this group, the element that has the best correlation with radon concentration according to Table 3 was selected. The second group includes volumetric water content in the four soil layers. The element that shows the best correlation with radon concentration was selected from this group. Air temperature outside the cave, precipitation and snow depth only in the cold half of the year were added to the selected from the above two groups' potential predictors (listed in the next two paragraphs). Based on the adjusted R^2 criterion, only those predictors that have a significant contribution to the variations of the dependent variable (radon concentration) were selected to be included in the final multiple linear regression model.

In the warm half of the year in the Concert Hall, the statistical model describes 33.3% of the variations in radon concentration. It includes as predictors the temperature of soil layer 4 (in the model it has a positive sign and describes 76.6% of the variations of the dependent variable) and volumetric water content in soil layer 1 (in the model it has a negative sign and describes 23.4% of the variations of the dependent variable). Figure 7a shows the course of measured and modeled, by the statistical model, radon concentrations during the warm half of the year for the period 08.11.2011 - 02.02.2020 in the Concert Hall. In general, the model describes well measured course of radon concentration except for some peak values in 2017 and 2019. In the Cosmos Hall, the statistical model describes 33.4% of the variations in radon concentration in the warm half of the year. The model includes as predictors the volumetric water content in soil layer 1 (in the model it has a negative

sign and describes 64.8% of the variations of the dependent variable) and the temperature of soil layer 3 (in the model it has a positive sign and describes 35.2% of the variations of the dependent variable). The statistical model for the Rockfall Hall describes 37% of the variations in radon concentration in the warm half of the year. The predictors that are included in this model are volumetric water content in soil layer 4 (in the model it has a negative sign and describes 49.5% of the variations of the dependent variable), temperature of soil layer 4 (in the model it has a positive sign and describes 24.4% of the variations of the dependent variable), precipitation (in the model it has a positive sign and describes 16% of the variations of the dependent variable) and air temperature (in the model it has a positive sign and describes 10.1% of the variations of the dependent variable). The statistical model for the Stack Hall describes 42.9% of the variations in radon concentration in the warm half of the year. It includes as predictors the temperature of soil layer 3 (in the model it has a positive sign and describes 68.9% of the variations of the dependent variable) and precipitation (in the model it has a negative sign and describes 31.1% of the variations of the dependent variable). The course of the measured and modeled radon concentrations in this hall during the warm half of the year for the period 08.11.2011 - 02.02.2020 is shown in Figure 7b, which document a good agreement, and only some extremes in 2016 and 2018 are less well modeled.

In the cold half of the year in the Concert Hall, the statistical model describes 65.6% of the variations in radon concentration. It includes as predictors skin temperature (in the model it has a positive sign and describes 66% of the variations of the dependent variable), snow depth (in the model it has a positive sign and describes 21.7% of the variations of the dependent variable), precipitation (in the model it has a negative sign and describes 7.4% of the variations of the dependent variable) and volumetric water content in soil layer 4 (in the model it has a positive sign and describes 4.9% of the variations of the dependent variable). Figure 8a shows the course of measured and modeled, by this model, radon concentrations during the cold half of the year for the period 08.11.2011 - 02.02.2020 in the Concert Hall. It can be seen that there is a very good agreement between the two graphs except for some peak values. The statistical model for the Cosmos Hall describes 42.3% of the variations in radon concentration in the cold half of the year. It includes as predictors skin temperature (in the model it has a positive sign and describes 55% of the variations of the dependent variable) and snow depth (in the model it has a positive sign and describes 45% of the variations of the dependent variable). In the Rockfall Hall, the statistical model describes 48.6% of the variations in radon concentration in the cold half of the year. The predictors in this model are snow depth (in the model it has a positive sign and describes 50.7% of the variations of the dependent variable)

Fig. 7. Observed and modeled (by the statistical model) radon concentrations in Saeva Dupka Cave during the warm half of the year for the period 08.11.2011 - 02.02.2020 in the Concert Hall (a) and Stack Hall (b).

and temperature of soil layer 2 (in the model it has a positive sign and describes 49.3% of the variations of the dependent variable). In the Stack Hall, the statistical model describes 65.8% of radon concentration variations in the cold half of the year. The predictors that are included are air temperature (in the model it has a positive sign and describes 59% of the variations of the dependent variable), snow depth (in the model it has a positive sign and describes 33.7% of the variations of the dependent variable) and precipitation (in the model it has a negative sign and describes 7.2% of the variations of the dependent variable). Figure 8b shows the course of measured and modeled radon concentrations in the Stack Hall in the cold half of the year for the period 08.11.2011 - 02.02.2020. It can be seen that there is a good agreement between the two graphs except for 2014. In conclusion, based on the predictors that are included in the statistical models, the very important role of the snow cover becomes obvious. This role is not so clear if only the results of Table 3 are taken into account. In general, the snow cover seals the soil and rock above the cave and radon formed there can only flow downwards towards the cave. This explains the positive correlation between snow depth and radon concentration in the cave.

The projected values regarding radon concentration in the cave in the warm and cold half of the year for the period 2021–2070 are shown in Figure 9. Once again, it should

Fig. 8. Observed and modeled (using statistical model) radon concentrations in Saeva Dupka Cave during the cold half of the year for the period 08.11.2011 - 02.02.2020 in the Concert Hall (a) and Stack Hall (b).

be stressed that the projections are made based on predictor trends in the respective statistical models for the period 1979–2019. Those of the trends that are statistically significant in this past period are extrapolated to the future period on the basis of linear regression. For those predictors that do not have a statistically significant trend in the period 1979–2019, the average values for this past period are used also in the future period. Figure 9a shows that in the warm half of the year radon concentration will increase continuously until 2070 in all four studied halls of the cave. These expected trends are mainly due to the positive correlation between the different temperatures (air, skin and soil) and radon concentration in the cave. These temperatures are expected to rise steadily in the coming decades. This will cause an increase in radon concentration in the cave, and respectively, a higher risk for the guides working in the cave (Turek et al., 2020). During the cold half of the year (Fig. 9b) the trend in radon concentration is again increasing, but there is specificity in this trend. By about 2047, radon concentration will remain at approximately current levels. This is due to the decreasing snow depth, which will act as a counterbalance to the increasing air temperature. After 2047, air temperature is expected to remain the only factor. However, given the winter ventilation regime of the cave, which is controlled by the fall of the outside air temperature permanently below 9°C, no significant increase in radon concentration in the cave is expected during the cold half of the

year. The increase in air temperatures inside and outside the cave is approximately synchronous, as shown in Tables 1 and 2. This means that in the future the temperature defining whether or not there is thermodynamic ventilation in the cave will increase above 9°C, but outside air temperature will also increase. Accordingly, this is not leading to a change in the intensity or direction of air exchange between the cave and outside atmosphere. In general, the time frames of the period with accumulation of radon in the cave (warm half year) and the period with low concentration of radon in the cave (cold half year) will remain the same.

CONCLUSION

Radon concentration in Saeva Dupka Cave is determined by two main mechanisms. The first mechanism is through penetration of radon from the soil and rocks above the cave. This mechanism has a leading role during the warm half of the year, when the thermodynamic exchange of air between the cave and outside atmosphere is difficult. Accordingly, during this period of the year radon concentration in the cave is high (accumulation of heavier gas in cave reservoirs). The second mechanism is through thermodynamic air exchange between the inside of the cave and outside atmosphere. This mechanism has a leading role during the cold half of the year, when

Fig. 9. Projected values of radon concentration in the four studied halls of Saeva Dupka Cave during the warm (a) and cold (b) half of the year for the period 2021–2070.

the colder air from outside penetrates the cave and keeps radon concentration at low levels. External environmental (atmospheric) conditions are essential for both mechanisms. Climatic factors that affect radon concentration in the cave are temperatures (air, skin, and soil) and the amount of water, expressed by precipitation or by volumetric water content in the soil. Water content in the soil, which depends on annual distribution and intensity of precipitation, as well as on structure and composition of the soil-vegetation cover and temperature regime, determines the speed and amount of radon penetrating into the cave cavities. During the cold half of the year, an important factor is the duration and depth of snow cover, which isolates the exhalation of radon to the external atmosphere.

Statistical models and projections based on them show that by 2070 radon concentration in the cave will increase. This increase will occur primarily to the warm half of the year, because it is determined mainly by the directly proportional relationship between temperatures and radon concentration. By the middle of the 21st century, it is expected that the studied area will no longer have a permanent snow cover and the temperature will remain the only factor for increasing of radon concentration during the cold half of the year. However, during this period, the ventilation regime of the cave, which is controlled by a permanent drop of the outside air temperature below 9°C, will continue to limit the potential increase of radon concentration in the cave air. The projections regarding the future values of radon concentration in Saeva Dupka Cave are based on the trends in air temperature in the studied area during the last four decades. This trend is positive. In the future, it is possible that there could be some other natural or anthropogenically caused events that may change this course of air temperature and, accordingly, of the radon concentration in the cave. At this stage, however, they cannot be predicted and included in the modeling.

Water content in the soil and precipitation are significant factors for radon concentration in the cave, but they have no significant trend and accordingly they will not have a direct influence in any particular direction in the next 50 years.

The projected increase of radon concentration in Saeva Dupka, which is one of the most visited tourist caves in Bulgaria (open seven days a week), creates a serious risk for the health of the tour guides working in the cave. Based on the data from radon monitoring and fixed stay time in the cave, it is possible to calculate the actual annual effective dose of radon radiation for the service personnel (Thinova & Rovenska, 2011). According to the Recommendation of the Czech State Office for Nuclear Safety (SUJB) (2007) and ICRP Publication 115 (Tirmarche et al., 2010), the personal effective dose reference level is 6 mSv y⁻¹ (2,000 working hours). The real annual effective dose E_a [mSv] for the cave guides in Saeva Dupka for the period 2012–2020, is 6.5 mSv (Turek et al., 2020). This high value and the expected increase in radon concentration in the most active tourist

season requires a plan that necessarily includes measures that will protect human health of the cave staff (e.g., to increase the number of guides seasonally and thus reduce the number of entries for each guide). On the other hand, it does not pose any significant health risk for ordinary tourists visiting the cave all year round (Turek et al., 2015, 2020; Stefanov et al., 2014, 2020).

From the point of view of health (radiation) safety in Saeva Dupka, the working conditions of the cave staff are most suitable in winter and transitional seasons, when radon and CO₂ concentrations that are hazardous to human health in the cave air are below the permissible norms and do not pose risks to the health of cave guides. This optimal period, which is outside the active tourist season, coincides with the academic year in schools and universities, and, due to the accessibility and educational resources of the cave, it has the potential to become an educational center for innovative training using karst (Stefanov & Stefanova, 2020).

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