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Preliminary data of potentially hazardous radon concentrations in Modrič Cave (Croatia)

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Abstract: Instigated by relatively high cave-air CO_2 concentrations in Modrič Cave (Croatia) recorded for the purpose of speleothem-based paleoclimate research, we established preliminary monitoring of radon (^{222}Rn) concentrations within the cave for a 4.5-year period (2018–2022). As radioactive geogenic gas, radon, which often correlates with cave-air CO_2 concentrations, presents a potential health hazard in cases of longer exposure time in high concentration conditions. Since the Modrič Cave is open to tourists and long-term scientific research has been performed within, a safety assessment for radon concentrations was essential. The integrated measurements of radon concentrations were performed by passive LR115 detectors that were exposed from three to six months at eight sites within the cave. Preliminary results showed seasonal variations of radon concentrations (0.08–13.6 kBq/m^3) governed by the cave ventilation patterns, but superimposed on this, cave morphology and bedrock architecture control the radon variations on finer spatial scale. The 3-months average ^{222}Rn concentration of up to 13.6 kBq/m^3 during summer in one of the cave passages is among the highest measured seasonal averaged radon concentrations in Croatian caves, but maximum concentrations were even higher. Based on obtained results and calculations, potential negative health effects of radon exposure for cave visitors, guides and scientists were assessed and the results showed values of exposure to be below recommended levels. Calculated worst-case scenario for cave guides (most affected by radon and its progeny) revealed that they would receive dose slightly below the occupational dose limit of 20 mSv/y (19.0 ± 5.2 mSv/y) in the touristic part of the cave and significantly higher doses (34.1 ± 9.2 mSv/y) in the non-touristic part of the cave. To detect precise spatio-temporal radon concentration variations (up to diurnal scale) we recently established continuous radon measurements. This will enable detection of possibly health threatening short-term peaks in radon concentration and consequently further improve cave management.

Keywords: radon, cave ventilation, geogenic hazard, Modrič Cave, Croatia

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INTRODUCTION

Radon is a naturally occurring heavy, radioactive, geogenic gas with no colour, odour, taste or flammability that occurs in trace amounts in atmosphere. It is an intermediate product of radioactive decay of ^{238}U (^{222}Rn, radon, hereafter), ^{235}U (^{219}Rn, actinon), and ^{232}Th (^{220}Rn, thoron), within the Earth's crust. However, in environmental studies, ^{220}Rn and ^{219}Rn are usually neglected due to their extremely short half-life, and only radon is taken into consideration due to its half-life of 3.8235 days (Nazaroff, 1992, Cigna 2005). Such a half-life prevents transportation on larger distances, so detected radon usually has local origins. In karst regions, the main sources of radon are miniscule amounts of uranium present within the limestone (1.3–2.5 ppm, Hakl et al., 1997), but even those amounts result in relatively high radon concentration in caves (Hakl et al., 1997; Cigna, 2005; Gregorič et al., 2011). Upon the release from minerals, radon atoms migrate into the air or interstitial water and move further through the rocks and soil by diffusion and/or advection (Etiope & Martinelli, 2002; Barbosa et al., 2015). Subsequently, radon reaches the atmosphere where is quickly diluted, but in confined areas, such as basements,

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mines and caves, it tends to accumulate due to the poor ventilation (Nazaroff, 1992; Gregorič et al., 2011) and if inhaled, may become a health risk (Gillmore et al., 1999; Cigna, 2005; Ferreira et al., 2016).

Radon is one of the main sources of natural radiation and is the main contributor to the total dose of radiation received by human population (UNEP, 2016). Any amount of radiation, no matter how small, is potentially harmful and radon carcinogenic properties have been well documented (Cigna, 2005). Moreover, radon has been recognized as a main cause of lung cancer outside of smoking (Craven & Smit, 2006; ICRP, 2010). Namely, some of the radon decay products (RDP) have very short half-lives that increases the probability of radioactive decay within the lungs, where the cells are exposed to the α-radiation emitted by decaying RDPs (Waring et al., 2021). Cave visitors are usually exposed to elevated radon concentrations with no significant consequences, but for cave guides, cavers and cave researchers, this presents more serious health hazard (particularly for those who are smokers as well) (Cigna, 2005; Craven & Smit, 2006; ICRP, 2010).

Ever since the pioneering work of Wilkening and Watkins (1976), radon behaviour in cave environments has been studied worldwide, including use as a trace gas, an indicator of tectonic/seismic activity and as a health hazard. In the Dinaric karst, the most extensive research has been performed in Slovenia (Kobal et al., 1986, 1987; Jovanovič, 1996), mostly in Postojna Cave (Kobal et al., 1988; Vaupotič et al., 2001; Vaupotič, 2008; Šebela et al., 2010; Gregorič & Vaupotič, 2011; Gregorič et al., 2011, 2013, 2014). In Croatia, measurements of radon concentrations in cave air commenced in 2004 (Paar, 2009; Paar et al., 2005, 2008; Radolić et al., 2009, 2011, 2012). A study of radon concentration in 20 caves and pits in coastal Dinaric karst and continental isolated karst patches revealed radon concentrations of up to 3.8 kBq/m^3 in the Velebit Mt. in Lubuška jama Pit, while in the Žumberak Mt. values reached 12.4 kBq/m^3 and 21.8 kBq/m^3 in Provala and Dolača Cave, respectively (Paar et al., 2009). The highest radon concentrations >30 kBq/m^3 are recorded in Šparožna pećina Cave (Šumonja et al., 2022). As for the show caves, a preliminary study in Manita peć Cave (Paklenica National Park, Velebit Mt.) revealed a relatively low average level of radon concentration with a summer peak of only 1.1 kBq/m^3; acceptable for the touristic management of the cave (Radolić et al., 2012). Meanwhile, in the south of Croatia, in the Đurovića Cave near Dubrovnik, radon concentrations up to 25 kBq/m^3 (Radolić et al., 2009) call into question the tourist use of the cave.

Here, we present the preliminary results from radon concentration monitoring in the show cave Modrič Cave (Croatia) based on data collected over a 4.5-year period, encompassing various seasons, in order to obtain insight into spatial and temporal variations of radon concentrations throughout the cave. The monitoring programme has been set-up to estimate the potential risk of radon overexposure to the visitors (tourists, guides and scientists) and it followed a Modrič Cave air CO_2 monitoring project; the latter showed a strong spatial and seasonal variations of CO_2 concentration with exceptionally high values in the innermost parts of the right passage of the cave (Surić et al., 2021a). Bearing in mind that CO_2, as a carrier gas, significantly controls transport and distribution of radon towards the Earth's surface, the findings of Surić et al. (2021a) necessitated a study of radon variations in order to assess whether it follows similar spatio-temporal patterns, and whether radon should justifiably be considered a geohazard.

STUDY SITE

Modrič Cave is located on the Adriatic coast, 120 m from the sea on the slopes of the Velebit Mountain (44°15' N, 15°32' E). It is a morphologically simple, mostly horizontal cave consisting of two passages with total length of 829 m and with only one entrance (Fig. 1). The cave is formed in well-bedded and tectonically fractured Upper Cretaceous limestone (Miko et al., 2002) with up to 27 m thick overburden. The area surrounding the cave is characterized by submerged springs (vruljas), coastal springs and smaller caves (Kuhta et al., 1999), which are firm evidence of intensive faulting and thorough karstification. The surface above the cave is covered with bushes, patches of grass and isolated trees. During the last century, the surface has undergone significant changes in vegetation cover; from an almost barren landscape to advancing, mostly anthropogenic, reforestation (Surić et al., 2021a). The climate of the area is temperate humid with hot summers (Cfa in Köppen-Geiger classification; Peel et al., 2007) with MAAT of 16.2°C, and average annual precipitation of 1218 mm (1993–2018 by Croatian Meteorological and Hydrological Service). Although the Cfa climate type is characterized by the even seasonal distribution of precipitation, on average 47% (556 mm) of precipitation occurs between September and December with peak (155 mm) in November. The driest period is from June to August with the minimum in July (57 mm). The location on the seaward slopes of the Velebit Mt. makes the surface area of the cave exposed to the bora wind, which often reaches gale force, particularly during the winter, thus affecting the cave air circulation patterns, and slightly cave air temperature and relative humidity.

Modrič Cave was discovered in 1985 and it is opened for visitors since 2004, but since it is a site of the adventure tourism (guided small-group off-road tours), the annual number of visitors is low, up to 700 individuals. Moreover, since the discovery, the cave has been a site of numerous scientific studies (i.e., palaeontology, archaeology, palaeoclimatology) (see the references in Surić et al., 2021a).

METHODOLOGY

The integrated measurements of radon concentrations were performed by a passive method that includes a series of solid-state nuclear track-etched detectors LR115 type II (manufacturer Dosirad, France). A cylindrical plastic vessel (cup shape, 9 cm in diameter and 5 cm in height) that was covered on the top with filter paper (surface density 0.078 kg/m^2). Inside the vessel, on the bottom, a LR115 detector with dimensions 2×3 cm^2 was placed. These detectors record only tracks left by alpha particles emitted from radon, because radon progeny cannot penetrate through the filter paper (Planinić et al., 1997).

Radon measurements in the Modrič Cave commenced in mid-July 2018 with the positioning of eight detectors inside the cave (Fig. 1) – one placed 20 m from the entrance of cave (site M0), four along the tourist route in the left passage (sites ML1–ML4), and three (MD1–MD3) were exposed in the right passage, which is the area of limited/restricted entry, visited only by scientists. Following the 18 months of irregular detector exchanges, from March 2020 to December 2022, the detectors were exposed in three-month periods during climatic seasons: spring (March–May), summer (June–August), autumn (September–November) and winter (December–February).

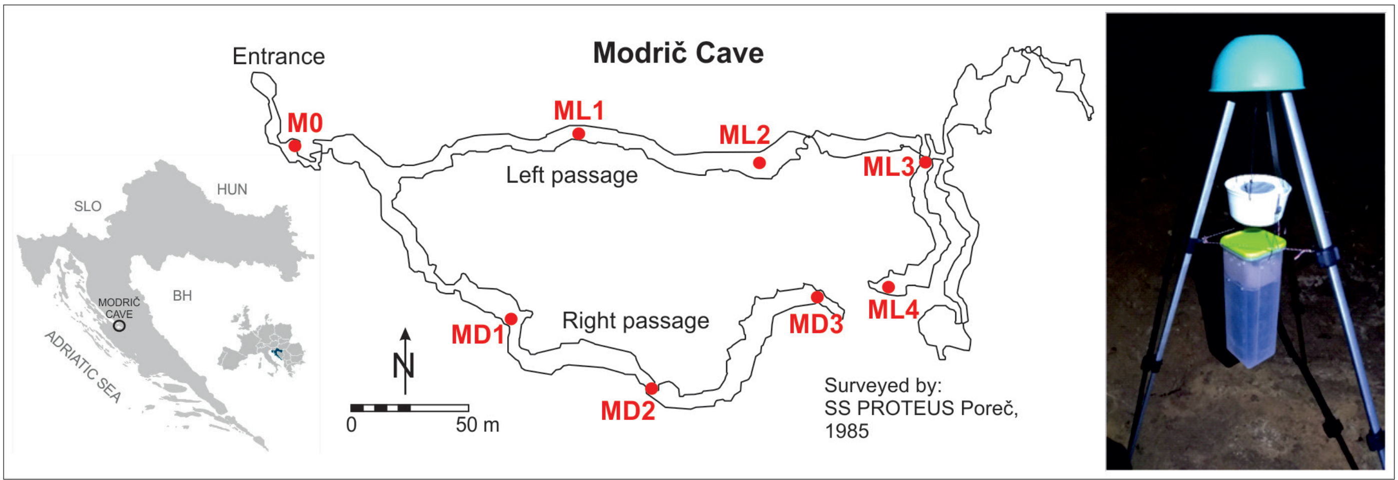

Fig. 1. Location and plan of the Modrič Cave with marked ^{222}Rn measurement sites and plastic vessel with LR115 type II detector placed inside the cave.

After being exposed, the LR115 type II detectors were chemically etched for 120 mins in 10% aqueous solution of NaOH at a temperature of 60°C. Tracks were counted automatically using an optical microscope Olympus BX51, digital camera Olympus c5050 and software for image analysis. Radon concentrations are determined as the product of track densities on diffuse detector and sensitivity coefficient (31.7 ± 3.2 Bq/m per tr/cm^2·d) which was determined during the calibration process at the PTP radon chamber (Physikalisch-technischer Prüfdienst, Vienna, Austria).

The effective doses received by visitors from radon and its progeny were calculated from exposure (radon concentration multiplied by time) and using dose conversion factors (DCF) for tourist caves of 1.5·10^{-5} mSv / Bqh/m^3 recommended by International Commission on Radiological Protection (ICRP) (ICRP, 2017), assuming an average breathing rate of 1.2 m^3/h and equilibrium factor of 0.4. This breathing rate could be applied due to physiography of the Modrič Cave (i.e., its horizontality and shallowness), which requires no intense physical activity for the visitors. Namely, breathing depth and rate impact the residence time of the radionuclides in the lung, so more difficult sections with deeper breathing during the caving increase the health risk (Gillmore et al., 2000), which is not the case here. The equilibrium factor was directly measured onsite by AlphaGUARD measuring system and Thomson Nielsen Radon WL Meter TN-WL-02. Two-days measurements were performed several times during different seasons and upon obtaining quite consistent average values of equilibrium factor (F = 0.43) so for the calculation of the effective dose received, we use the common value of equilibrium factor (0.4).

Time-related particularities of radon concentrations should be complemented by precise determination of cave geometry, in order to detect the spatial determinants (major crevices and joints) that may control different ventilation intensities along the cave passages. For that purpose, we used a GeoSLAM ZEB Revo RT handheld laser scanner that features a rotating LiDAR sensor (30 m range, 43,000 scanner points per second) to build a 3D model of the cave. In spite of the generally simple morphology, detailed scanning of the demanding low and narrow passes required two 2-hours lasting scanning sessions.

RESULTS

Results of radon concentration measurements conducted from July 2018 to December 2022 in the Modrič Cave are summarized in Table 1. The lowest concentration of only 0.08 kBq/m^3 was recorded at the M0 measurement site near the entrance of the cave during the winter 2020/21, while the highest radon concentration of 13.6 ± 1.4 kBq/m^3 was measured in summer 2022 in the right passage. In the right passage of Modrič Cave (MD1–MD3), radon concentrations were usually higher than in the left one (ML1–ML4) and this difference was statistically significant ($p = 4.9\times10^{-12}$, df = 97) at the significance level of 0.05.

The digital 3D model built upon the point cloud of >500 mil. points (Supplementary Fig. S1) gave substantially better insight into finer architecture of the cave that could not be accomplished by standard cave surveying techniques. It also provided basic morphometric data – total surface of the cave of 4,343 m^2 and volume of ca. 20,000 m^3. GeoSLAM point cloud animation is available at sketchfab.com.

Table 1. Concentration of ^{222}Rn in the Modrič Cave (in kBq/m^3) recorded in various periods from July 2018 to December 2022. M0 – near-entrance chamber; ML1-ML4 – left passage; MD1-MD3 – right passage.

Season	Measurement period	M0	ML1	ML2	ML3	ML4	MD1	MD2	MD3
Summer	19 Jul 2018 2 Oct 2018	4.8 ± 0.5	3.7 ± 0.4	3.4 ± 0.4	4.1 ± 0.4	5.7 ± 0.6	8.8 ±0.9	10.6 ± 1.1	10.1 ±1.0
Autumn-winter	2 Oct 2018 5 Mar 2019	1.5 ± 0.2	1.2 ± 0.1	1.2 ± 0.1	1.5 ± 0.2	1.7 ± 0.2	3.5 ± 0.4	4.4 ± 0.4	3.4 ± 0.4
Spring	5 Mar 2019 2 Jul 2019	1.9 ± 0.6	2.1 ±0.2	2.2 ± 0.2	2.3 ± 0.2	3.3 ± 0.3	6.0 ± 0.6	6.1 ± 0.6	5.2 ± 0.5
Summer	2 Jul 2019 18 Oct 2019	3.9 ± 0.4	4.6 ± 0.5	4.0 ± 0.4	4.1 ± 0.4	6.0 ± 0.6	8.1 ± 0.8	9.5 ± 1.0	8.8 ± 0.9
Autumn-winter	18 Oct 2019 3 Mar 2020	0.41 ± 0.04	0.78 ± 0.08	0.60 ± 0.06	0.9 ± 0.2	1.2 ± 0.1	2.6 ± 0.5	2.6 ± 0.5	2.6 ± 0.3
Spring	3 Mar 2020 5 Jun 2020	1.3 ± 0.1	1.4 ± 0.2	1.5 ± 0.2	1.7 ± 0.2	1.9 ± 0.2	4.1 ± 0.8	5.0 ± 0.5	3.9 ± 0.4
Summer	5 Jun 2020 1 Sep 2020	3.4 ± 0.4	3.2 ± 0.3	3.1 ± 0.3	3.1 ± 0.3	4.3 ± 0.5	8.5 ± 0.9	8.9 ± 0.9	8.8 ± 0.9
Autumn	1 Sep 2020 8 Dec 2020	1.3 ± 0.1	1.8 ± 0.2	1.6 ± 0.2	2.1 ± 0.2	2.7 ± 0.3	5.1 ± 0.5	4.5 ± 0.5	5.3 ± 0.5
Winter	8 Dec 2020 3 Mar 2021	0.08 ± 0.01	0.13 ± 0.01	0.19 ± 0.02	0.22 ± 0.02	0.75 ± 0.09	2.1 ± 0.2	2.3 ± 0.2	2.1 ± 0.2
Spring	3 Mar 2021 31 May 2021	0.8 ± 0.1	1.1 ± 0.1	1.1 ± 0.1	1.3 ± 0.1	1.6 ± 0.2	3.6 ± 0.4	3.9 ± 0.4	3.8 ± 0.4
Summer	31 May 2021 6 Sep 2021	3.6 ± 0.4	4.1 ± 0.4	3.8 ± 0.4	3.3 ± 0.3	6.7 ± 0.7	11.1 ± 1.1	11.5 ± 1.2	11.3 ± 1.2
Autumn	6 Sep 2021 6 Dec 2021	1.2 ± 0.1	1.9 ± 0.2	2.1 ± 0.2	3.1 ± 0.3	2.9 ± 0.3	4.3 ± 0.5	4.5 ± 0.5	5.0 ± 0.5
Winter	6 Dec 2021 28 Feb 2022	0.11 ± 0.01	0.18 ± 0.02	0.24 ± 0.03	0.33 ± 0.04	0.8 ± 0.1	2.7 ± 0.3	2.6 ± 0.3	2.2 ± 0.2
Spring	28 Feb 2022 28 May 2022	1.0 ± 0.1	1.4 ± 0.2	1.1 ± 0.1	1.4 ± 0.2	2.3 ± 0.2	4.3 ± 0.4	4.7 ± 0.5	4.4 ± 0.5
Summer	28 May 2022 1 Sep 2022	4.7 ± 0.5	4.4 ± 0.5	4.0 ± 0.4	4.1 ± 0.4	6.3 ± 0.6	13.6 ± 1.4	10.4 ± 1.1	11.2 ± 1.1
Autumn	1 Sep 2022 5 Dec 2022	2.0 ± 0.2	2.4 ± 0.3	2.4 ± 0.3	2.5 ± 0.3	4.1 ± 0.4	5.1 ± 0.5	5.8 ± 0.6	6.5 ± 0.7

DISCUSSION

Spatial and temporal dynamics of radon

The natural behaviour of radon, being a noble gas with low chemical activity, is affected more by physics than by chemistry and its concentration in the caves is governed generally by radon emanation and transport paths (Trique et al., 1999). In fact, this is a complex interaction among a number of features, such as radon source (lithology), weathering rate, airflow, subterranean streams, carrier gasses behaviour, cave morphology, faults and meteorological settings (i.e., temperature, precipitation, air pressure, wind) (e.g., Gillmore et al., 2000; Ciotoli et al., 2017 and references therein). Some of these features, potentially involved in radon distribution in Modrič Cave, are discussed below.

The first and the foremost is the radon source, namely, the uranium content of the bedrock or more precisely, rock and soil capacity for radon gas production (Ferreira et al., 2016). Due to the local geological setting (Kuhta et al., 1999), we can exclude a deep magmatic origin of the radon, so the radon must be from a local subsurface origin, emanated during the karstification processes within the fractured epikarst. According to Hakl et al. (1997), uranium concentrations in limestones (considered to be among the least uraniferous off all rocks) is typically 1.3–2.5 ppm, which is significantly higher than in Modrič Cave limestone bedrock where the radon concentration was measured at 0.5 ppm (Kuhta et al., 1999). Topsoil above the cave had uranium concentrations of 3.2–3.6 ppm, while in cave sediment it varied between 5.8 and 7.4 ppm (Kuhta et al., 1999). In a speleothem core from Modrič Cave, uranium concentration was <0.1 ppm (Kuhta et al., 1999), while the 6 measurements for the purpose of stalagmite dating revealed ^{238}U concentrations range from 30 to 70 ppb (Rudzka et al., 2012). Similarly, in 26 samples of spelean carbonate from adjacent Manita peć Cave, ^{238}U concentrations ranged from 17 to 127 ppb with an average value of 63 ppb (Surić et al., 2021b). Although these measured concentrations of solid rock are relatively low, fractured limestone is prone to weathering much more than other rocks with even higher uranium concentration (e.g., igneous), so by its decomposition during the karstification processes, radon release into the soil is more effective. In addition, radon concentrations are generally elevated in crushed bedrock of faulted zones with higher permeability (Šebela et al., 2010; Waring, 2021), such as in the Modrič area (Kuhta et al., 1999).

Upon the radon release from the mineral lattice, its distribution is controlled by transport paths and patterns, which are governed by environmental settings of the surface and underground. Since the Modrič Cave is hydrologically inactive, transport by an underground stream is excluded, so it appears that in-situ accumulation derives from the subsurface karstified horizon where radon is transported by organic CO_2 produced in the soil horizon or/and by dripwater. Batiot-Guilhe et al. (2007) suggested biogenic transport of radon from deeper parts of the aquifer towards the surface zone, but since the CO_2 concentration increase coincides with peak of vegetation activity (Surić et al., 2021a), we assume that main gas-related dynamics is directed from the upper part of the aquifer toward the cave. We expect that rain events, which enhance both bioactivity and limestone dissolution, will be followed by increased concentrations of both radon and its carrier gas CO_2.

After the transport and accumulation, the cave ventilation becomes the leading mechanism behind spatial and temporal variations in radon concentration (Kowalczk & Froelich 2010; Gregorič et al., 2014; Sainz et al, 2018; Wang et al., 2019). Cave ventilation patterns are influenced by the number of cave entrances, their relative position, the morphology of the cave passages and the predominant weather types outside the caves (Cigna, 2005). Intensive inflow of the outside air into the cave will cause the reduction of the radon concentration as the constant ventilation of the atmospheric air dilutes it, while the absence of ventilation will induce the build-up of the radon concentrations (Gilmore et al., 2000; Cigna, 2005; Smith et al., 2018). In the morphologically simple caves, such as Modrič Cave (only one entrance and two almost horizontal passages, Supplementary Fig. S1), the ventilation is controlled by the differences between the cave (T_{cave}) and the outside temperature (T_{out}) (Milanolo & Gabrovšek, 2009). During the autumn/winter season with $T_{cave} > T_{out}$, the outside air is cooler and denser and starts to flow into the cave pushing the cave air out, while in the spring/summer season with $T_{cave} < T_{out}$ the cave air is denser, thus, ceasing the ventilation (Kukuljan et al., 2021). In the Modrič Cave, outside temperature increases above the cave air temperature during April–May (Surić et al. 2021a) and after that transition period, the concentrations of radon start to increase, reaching maximum in the late summer/early autumn (Table 1). Whereas, during October and November the outside temperature drops below the cave air temperature, which induces significant cave ventilation (Surić et al. 2021a), reducing the radon concentration to minimum values during the late winter and early spring. By comparing measured radon concentrations in these two periods of increased (summer–autumn) and decreased (winter–spring) radon concentrations in Modrič Cave, the ratio was 2.45 ± 0.33 (1 standard deviation) for the time interval from spring 2020 until autumn 2022.

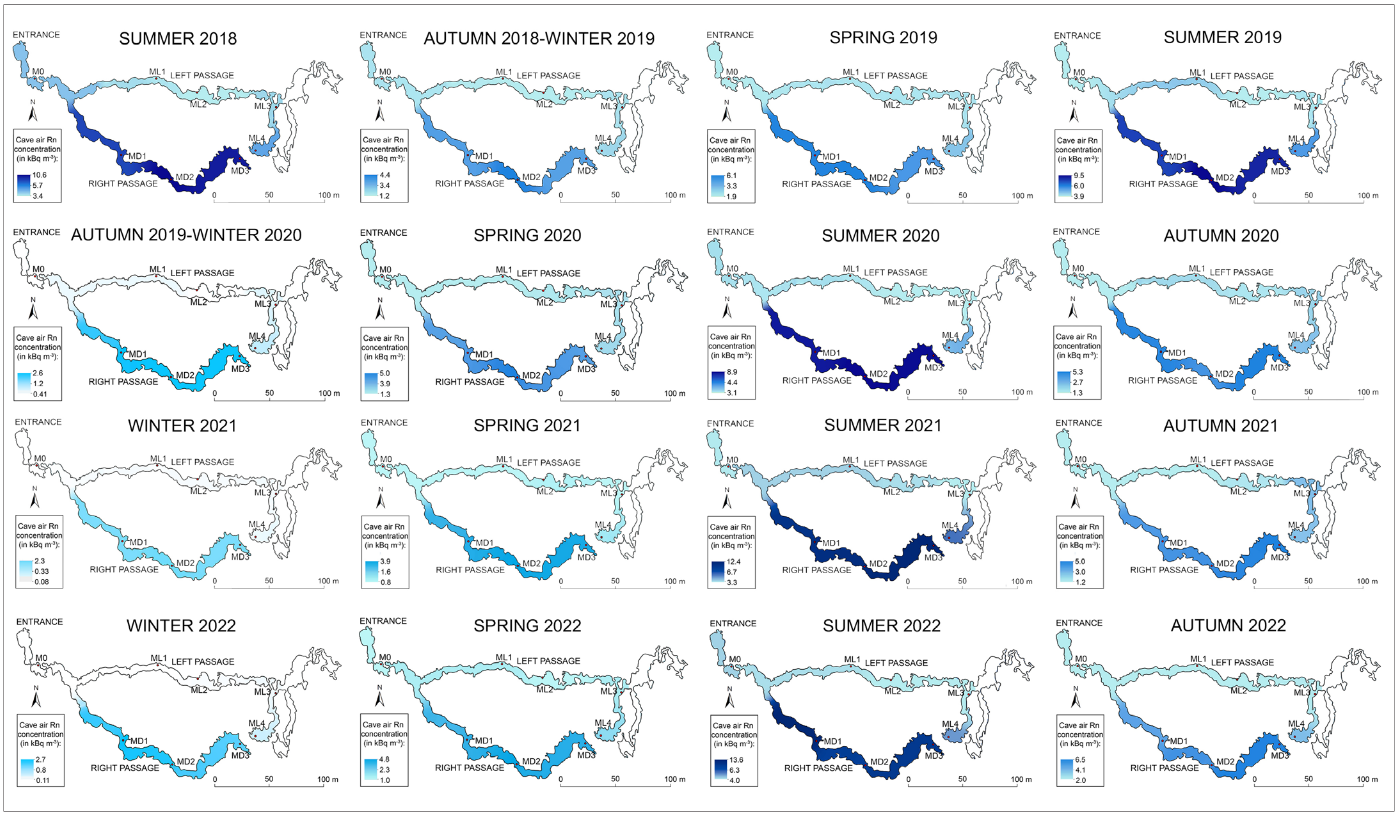

Fig. 2. Spatio-temporal variations of radon concentrations in Modrič Cave from July 2018 to December 2022.

Spatio-temporal distribution of radon concentrations within the Modrič Cave from July 2018 to December 2022 showed significant variability, which is, at this coarse temporal resolution, generally characterized by seasonal patterns and substantial differences between the left and right passages (Fig. 2). In the right passage, the multi-month average values recorded by the passive detectors never fall below 2 kBq/m^3. However, finer time resolution is expected to reveal short-term variations, such as diurnal, governed by the differences between day and night air temperatures both inside and outside the cave (Gregorič & Vaupotič, 2011), as well as wind-driven ventilation (Kukuljan et al., 2021). Given the shallowness of the Modrič Cave and its position where the bora wind reaches gale force (maximum 10-minutes average wind speed

40.9 m/s with maximal gust of 62.7 m/s recorded 2.5 km SW from the cave; Bajić, 2014), these events could substantially modify seasonal density-driven circulation. For both tourists and scientists, these cognitions must be a guide for the planning of visits in order to minimize the exposure, for example, by avoiding the late summer period and preferring post-bora episodes.

Seasonal density-driven variations of the Modrič Cave CO_2 concentration have been discussed in Surić et al. (2021a); in short, the main source is plant- and microbially-derived CO_2 that diffuses downward, in addition to in-cave CO_2 degassing of the dripwater. There is neither an underground stream nor significant CO_2 production by cave biota. Dilution by outside air due to the density driven cave ventilation governed by different air temperature is the main sink of the cave CO_2. During the 2017–2021 campaign, CO_2 concentration in the left passage varied between 480 ppm and 7,228 ppm, while in the right passage its range was from 994 ppm to >10,000 ppm (exceeding the measurement limit of 7755 AZ device) (Surić et al., 2021a). When compared to the radon record, covariance of radon and CO_2 concentration on the seasonal scale is present in the whole cave, regardless the absolute concentrations (Fig. 3), pointing to the common source (subsurface karstified zone) of both gases and their mutual dynamics.

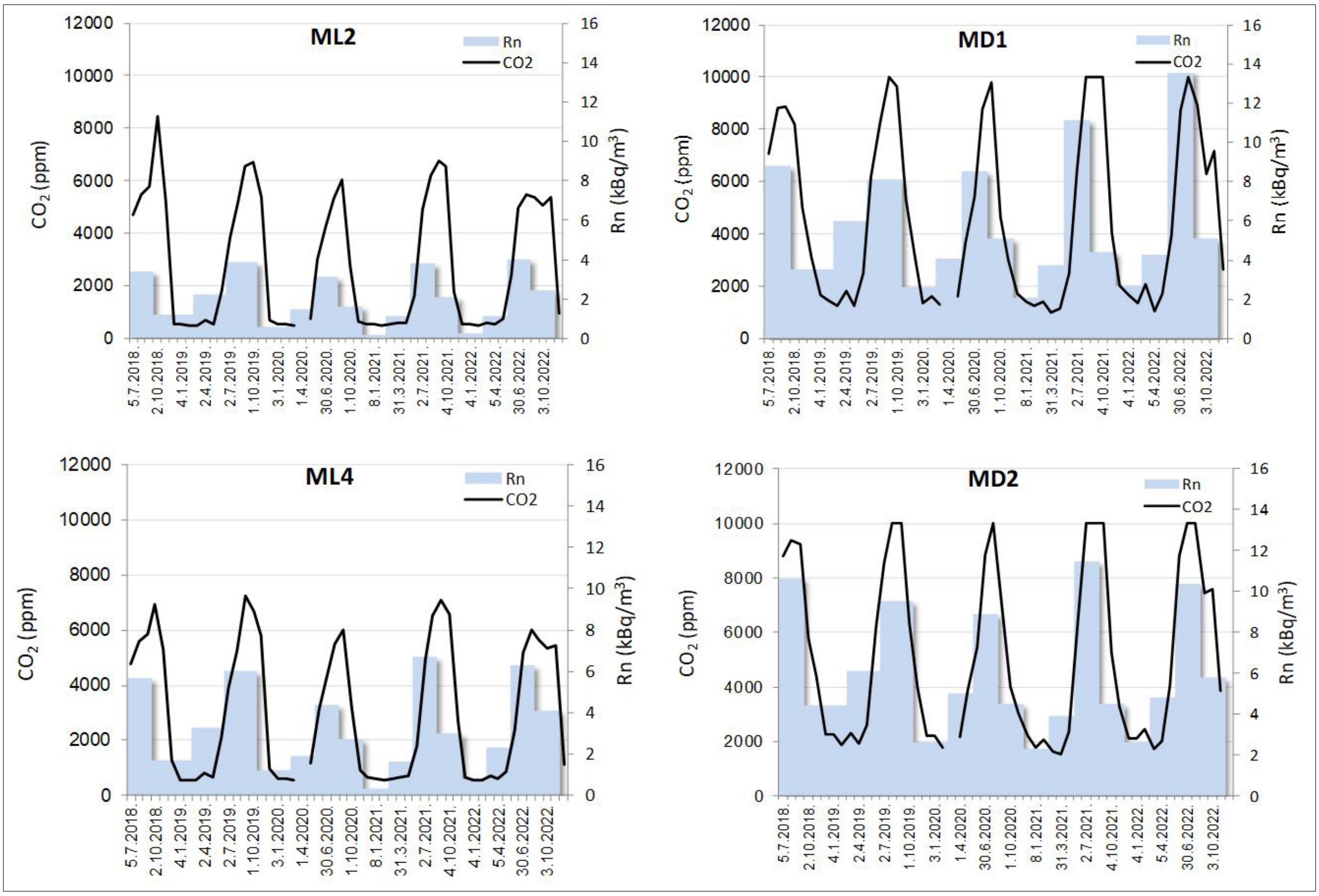

Fig. 3. Radon and CO_2 concentrations at two representative locations in the left passage (ML2 and ML4) and two in right passage (MD1 and MD2) during the 2018–2022 period. Radon concentrations are given as an average value of multi-month measurements by passive detectors (Table 1), and CO_2 concentrations are from Surić et al. (2021a), measured monthly by 7755 AZ handheld device with CO_2 range of 0–9,999 ppm. Note that summer 10,000 ppm values from right passage were in fact higher, above the device measurement limit.

From the spatial standpoint, the main characteristic of the Modrič Cave gas properties is significantly higher radon concentrations in the right passage throughout all the seasons (Fig. 2). Given the relatively small area, with the same lithology, depth and hydroclimate setting of both passages, the only plausible cause of their different gas dynamics can be diverse architecture of overlying subsurface bedrock, which consequently impacts cave ventilation patterns. Knowing that karst massif is a complex system of interconnected fractures and fissures, it is apparent that the total volume of air that is involved in the control of in-cave atmosphere is much larger than that of the principle underground cavity (Bourges et al., 2006; Wang et al., 2019). As revealed by precise laser scanning mapping (Supplementary Fig. S1b), the left passage is more spacious and the overlying rocks are more fractured, which enables more effective ventilation, thus, reducing the radon concentrations. The maximum heights of the left passage (ca. 8 m) are found in its mid-part, close to the site ML2 (Supplementary Fig. S1), where the fractures reach the surface through 15 m of karstified overburden. On the other hand, the right passage is narrower with occasional tight spots that impair the air circulation and allows the build-up of radon concentrations, particularly in the innermost part of the right passage after these tight spots. Another intriguing part is the proximity of the end of the left passage (ML4) and the end of the right passage (MD3), which are separated by relatively thin bedrock portion

(Fig. 2). The linear correlation between measured values of radon concentrations at these two locations is very strong (Pearsons correlation coefficient, r = 0.994) (Fig. 4). By applying a simple linear regression model, the ratio between the radon concentrations remains constant (1.73) regardless their seasonal variations. The presented linear best fit is statistically significant for the experimental data ($p = 1.3 \cdot 10^{-15}$, df = 15) at the 95% confidence level and it could be used for prediction purposes. Such divergences of radon concentrations, within just a few tens of meters, are due to the different internal micro-ventilation patterns (Waring et al., 2021). Crucially, such site-specific properties of the cave environment can be revealed only by detailed field surveying and continuous multi-seasonal monitoring, even in relatively simple caves such as Modrič Cave.

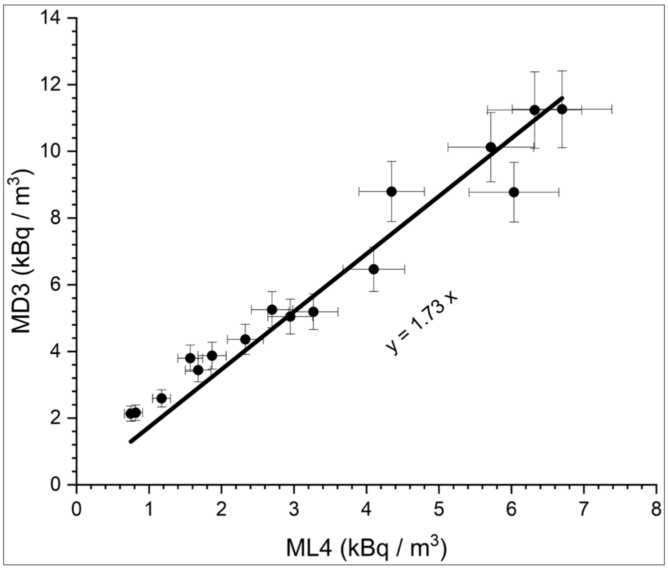

Fig. 4. Correlation of radon concentration over a 4.5-year period at two cave dead-ends (left passage's ML4 and right passage's MD3) within a few meters of one another.

Health precautions

Average annual radon concentration in Modrič Cave based on 4.5-years measurement by passive detectors was 3.86 kBq/m^3 and falls between global estimations for the cave environment of 2.80 kBq/m^3 (Hakl et al., 1997) and 6.16 kBq/m^3 (Waring et al., 2021). However, radon concentration values given as annual averages mask extreme variations that can vary up to two orders of magnitude between minimum and maximum (Waring et al., 2021) and should be avoided as a governing factor for the safety recommendation and regulation. Specifically, in Modrič Cave, the measured 3-months maximum at location MD1 is 170 times higher than measured minimum at the entrance – location M0 (Fig. 1, Table 1). As described earlier, Modrič Cave is usually visited with trained guides exclusively along the left passage. These organized tours occur irregularly (not every day) mainly from mid-April to mid-October and the tours last about 2 hours. The effective doses of radiation that a single tourist received from radon and its progeny during cave visit were 75 ± 20 μSv in 2021, 68 ± 19 μSv in 2020 and 91 ± 25 μSv in 2022. The single received dose calculated for each year was presented as an average of estimated received doses in three seasons (spring, summer, autumn).

The scientific visits to Modrič Cave are organized regularly several times (at least four visits) per season and include collecting data and equipment maintenance, with scientists staying in cave for ca. 2 hours (1 hour in each passage). The effective doses received during these scientific visits to Modrič Cave were 1.44 ± 0.39 mSv, 1.58 ± 0.43 mSv and 1.70 ± 0.46 mSv in 2020, 2021 and 2022, respectively. These obtained values are comparable to annual effective dose for Croatian population received in their homes from indoor radon and its progeny (Radolić et al., 2006). However, cave guides are expected to be the most exposed to the radiation. Due to pandemic circumstances, in 2020 and 2021, the Modrič Cave guide had only 11 (2020) and 12 (2021) tours during summer period and received the effective dose of only 1.05 ± 0.25 mSv in 2020 and 1.41 ± 0.45 mSv in 2021, while in non-pandemic 2022 guides received 15.2 ± 4.1 mSv for 120 visits from April until October 2022. If we apply the worst-case scenario for dose calculation which assumes a daily 2-hour visiting tour from the mid-April until the mid-October (45 days in spring, 92 days in summer and 45 days in autumn), based on 2021–2022 radon concentrations, the guide would receive a maximum effective dose of 19.0 ± 5.2 mSv/y, which is only slightly lower than the recommended dose limit for occupationally exposed workers of 20 mSv/y (averaged over a defined period of 5 years, with no single year exceeding 50 mSv (ICRP, 2007)). Furthermore, due to significant differences between radon concentrations in the left and right passages, if the Modrič Cave visiting route would be organized in the right passage instead of the left one, by applying both, the worst-case scenario and real visits, the tourist guides in 2022 would have received 34.1 ± 9.2 mSv and 27.2 ± 7.3 mSv, respectively, which is up to 79% higher doses than in the left passage.

Bearing all aforementioned in mind, we initiated continuous measurement of both radon and CO_2 concentration at 1-hour resolution by Tesla TSR 3W and Vaisala MI70 + GMP 252 devices, respectively. In order to establish a robust model of cave gases dynamics, all the specificities that were overlooked by this screening study will be taken into account, such as short-term diurnal variations related to differences between day and night cave and outside air temperatures, ventilation induced by short wind episodes, and anticipated build-up of radon concentration generated by rain events. Eventually, recommendation, as well as possible restrictions, for all type of cave visits could be prescribed.

CONCLUSIONS

The preliminary study of the radon concentrations in Modrič Cave revealed spatio-temporal patterns similar to those found in the previously conducted study of the CO_2 concentrations. Radon, emanated from low-uranium (0.5 ppm), but intensively karstified limestone is transferred by biogenic CO_2 as a carrier gas towards the relatively shallow and horizontal cave. There, the main mechanism governing the temporal distribution of the radon concentration is density-driven cave ventilation controlled by the difference between the cave and outside air temperature. Thus, the highest 3-month average values of radon concentration were 13.6 kBq/m^3 during the summer with practically no cave ventilation as the cave air temperature is lower than that outside. In addition, increased soil production of CO_2 enhances transport of radon, as well as its release due to the karstification. The lowest concentrations are related to the winter cooler outside air flow that dilutes underground radon accumulation. Superimposed on the temporal dynamics of cave ventilation is the spatial aspect (i.e., radon concentrations controlled by cave passage morphology and fissure architecture). Distinguishable differences are apparent between the more spacious left passage with heavily fractured overlying bedrock, and the more constrained right passage with poor ventilation and corresponding higher radon concentrations, with minimum multi-month average >2 kBq/m^3.

Based on the obtained data, the effective dose of radiation for tourists, guides and scientists were calculated to determine potential health hazards from radon exposure. The data showed the typical exposure levels were well below the recommended dose limit of 20 mSv/y (ICRP, 2007), particularly for the tourists who received only 68 to 91 µSv. Worst-case scenario for radon exposure of cave guides calculated separately for the left (touristic) and right (non-touristic) passages showed that in the left passage the radon exposure (19.0 ± 5.2 mSv/y) would still be, but only just, below recommended levels, while in the right passage the values would be significantly higher (34.1 ± 9.2 mSv/y) than those recommended by the ICRP (ICRP, 2007).

Obtained data confirm the uniqueness of micro-ventilation patterns as well as the importance and necessity of individual approach to cave investigation on which sustainable and responsible cave management should rely. Commenced continuous radon measurements will provide higher resolution data and better insight into diurnal and meteorologically induced short-term variations of radon concentration.

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