Radon Levels in Manita Peć Cave (Croatian NP Paklenica) and Assessment of Effective Dose Received by Visitors and Tourist Guides

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Abstract

The Manita peć cave is located within the Croatian National Park Paklenica and it is the only cave in the park among 70 others speleological objects that is open for visitors. With its length of 175 meters it belongs to shorter speleological objects. It is of great interest among tourists because of numerous cave ornaments and the proximity to popular tourist destinations on the Adriatic coast. The temperature and humidity are stable during a year with values about 10 °C and 100 %, respectively.

In June 2010, the 15 months monitoring of natural radioactivity from radon and its short lived progeny started. Integrated radon measurements by means of solid state nuclear track etched detectors were conducted during each season of the year. Additional measurements were performed during summer period because of intense tourist visits. The daily variations of radon and its progeny were measured by the AlphaGUARD measuring system.

The average radon concentration in summer 2010, as well as in 2011, was 1.1 kBq m⁻³, and it was much higher than in other seasons (0.03 kBq m⁻³ in winter 2010, for example). The obtained values categorize Manita peć cave among caves with lower radon concentration than the world average (2.8 kBq m⁻³). The daily variations in radon and its progeny concentrations as an influence of tourist activities during cave tours were not observed. Statistical t-test shows that radon concentration is uniformly distributed within the cave in each season.

The highest average effective dose of radon and its short lived progeny that was received by visitors during their 30 minutes tour was during summer period and equalled $1.86 \,\mu$ Sv. On the other hand, the maximum effective dose received by the tourist guide was $0.247 \,\text{mSv}$ in 2010 and $0.215 \,\text{mSv}$ in 2011.

Key Words: radon, cave, effective dose, equilibrium factor

Introduction

Radon is produced by the radioactive decay of radium (²²⁶Ra) along with the emission of α particle inside mineral grains that contains uranium. Although most of radon never leaves mineral crystal lattice where it is created until its own radioactive decay (half-life of radon is $\tau_{1/2} = 3.825$ days), smaller amounts of radon manage to escape and travel through Earth's crust guided by various transport mechanisms (by diffusion, advection, convection or trapped by geogases) [Nazzarof et al, 1988].

Caves in karst areas are mostly made of limestone. Since limestone contains 1.3 - 2.5 ppm of uranium (²³⁸U) in average and radon is one of the uranium progeny, one could expect elevated radon concentrations in some caves. In general, radon concentration in caves (that are complicated systems to be mathematically described) depend on different parameters and the dominant ones are: radon exhalation from the cave surfaces, shape and size of the cave, air flow and air exchange rate from inside to outside and vice versa, mixing of the outside air with the air inside the cave (related with meteorological parameters inside and outside the cave). The temperature difference between the air inside of the cave is the principal mechanism of air exchange in horizontally structured caves, while in vertical caves the same mechanism is governed by the pressure difference between the upper and lower part of the cave [Hakl et al., 1996]. According to this, in the horizontal caves maximum radon concentrations are expected during summer period because of the constant air

temperature in the cave during the year; during winter the air temperature in the cave is higher than the outside temperature and due to this temperature gradient the air from the cave is extracted and the fresh air flows into the cave reducing the radon concentration.

During the last three decades, radon concentrations were measured in the caves throughout the world [Wilkening and Watkins, 1976; Fernández et al., 1984; Papastefanou et al., 1986; Pinza-Molina et al., 1999; Przylibski, 1999; Sperrin et al., 2000; Dueñas et al., 2005; Lario et al., 2005; Espinosa et al., 2008], as well as in our neighboring countries [Kobal et al., 1986, 1987; Csige et al., 1996; Jovanovič, 1996; Szerbin, 1996; Bahtijari et. al., 2008]. Hakl et al. [1997] systematized radon concentrations in caves in Hungary, Italy, Slovakia, Luxembourg, England, Mexico and the United States of America. They concluded that the distribution of analyzed data is log-normal, with arithmetic mean of 2.8 kBq m⁻³ and values were in range of 0.1 and 20 kBq m⁻³. A similar result was obtained by Cigna [2005] when he analyzed newly published radon concentration in caves throughout the world, and calculated mean radon concentration of 2.5 kBq m⁻³.

In the Republic of Croatia, radon in pits and caves is being extensively measured since 2004. Detectors are set up during speleological researches with the purpose of exploring physical and chemical properties of the karst underground. Until today, radon concentrations were measured in about twenty caves and pits in the Velebit and Žumberak mountains. In the deep pits on Velebit mountain the highest radon concentration was measured in Lubuška pit (3.8 kBq m⁻³) [Paar et al., 2008]. In Žumberak mountain the highest radon concentration was measured in the Dolača cave (21.8 kBq m⁻³), 250 meters from the entrance during summer period [Paar et al., 2009]. Furthermore, a radon monitoring in the Đurovića cave was performed. The Đurovića cave is located near the control tower of Dubrovnik airport, which makes it very interesting for tourists. Detectors were exposed in late autumn 2008 and during spring and summer 2009, and the obtained average radon concentrations were 9.5, 17.9 and 25.0 kBq m⁻³, respectively [Radolić et al., 2009].

It is well known that radon and its short lived progeny have the largest contribution to the annual effective dose received by general population from natural radioactive sources [UNSCEAR, 2000.]. Radon does not present a health hazard in an open environment, due to natural air circulation, but it is the major health hazard in closed and/or poorly ventilated spaces like offices and public places beneath the ground level, and especially in caves due to high radon concentration in some of them. That potential health hazard was recognized by the international agencies related to the subject: International Commission on Radiological Protection (ICRP) and International Atomic Energy Agency (IAEA), that gave the recommendation [ICRP, 1994] and safety standard [IAEA, 2003] for the radon and its short lived progeny according which the effective dose for general public should not exceed 3 mSv per year, and for the occupationally exposed groups (maintenance workers, tourist guides that spend most of their working hours in the caves) the recommended value is 20 mSv in one year (or 50 mSv in 5 years!). In spite of these recommendations, many workers in caves can easily receive, due to high radon concentration, doses that are higher than the recommended ones [Papastefanou et al., 1986; Jovanovič, 1996; Pinza-Molina et al., 1999; Sperrin et al., 2000, Radolić et al., 2009]. Because of that specific nature of the profession, one of the most common ways in reducing the effective dose is to limit the number of working hours inside the cave [Vaupotič et al., 2001].

The effective dose assessment from radon and its short lived progeny is based on the epidemiological model described in the ICRP-65 publication [ICRP, 1994], which gives the conversion coefficient between the effective dose and exposure (effective dose of 1.1 mSv for general public and 1.425 mSv for professionally exposed groups is equivalent to exposure from radon and its short lived progeny of 1 mJ h m⁻³). The ratio between exposure to radon progenies and equivalent equilibrium radon concentration is $5.56 \cdot 10^{-6}$ mJ h m⁻³. By measuring the equilibrium factor, *F* (ratio between equilibrium equivalent radon concentration and actual radon concentration), radon concentration, *c* and time of exposure, *t* it is possible to determine an effective dose according to the following expression:

$$E[mSv] = 7.923 \cdot 10^{-6} \cdot F \cdot c[Bqm^{-3}] \cdot t[h].$$
⁽¹⁾

Material and methods

The measurement of radon concentration is performed by a passive method that includes series of solid state nuclear track-etched detectors LR 115 type II (manufacturer Kodak-Pathé, France). A cylindrical plastic vessel (cup with 11 cm in diameter and height of 7 cm) is covered on the top with filter paper (surface density 0.078 kg/m²). Inside the vessel, on the bottom, a LR 115 detector (with dimensions $2x3 \text{ cm}^2$), that is called *diffusion detector*, is placed. The diffusion detector records only tracks left by alpha particles emitted from radon, because radon progeny cannot penetrate through the filter paper. The other detector is placed on the outside surface of the plastic vessel and it is the so-called *open detector* (Figure 1.). The open detector records tracks of alpha particles emitted from the radon as well as its short lived alpha progeny (²¹⁸Po and ²¹⁴Po). This method with two detectors enables determination of radon equilibrium factor *F* between radon and its short lived progeny, in order to estimate the effective dose, according to equation (1) as better as possible [Planinić et al., 1997].



Figure 1. Plastic vessel with placed diffusion (on the right) and open (to the left) LR 115 type II detectors.

Continuous measurements of radon and its short lived progeny concentrations, as well as certain meteorological parameters (air temperature, barometric pressure, relative humidity) is performed by AlphaGUARD measuring unit (manufacturer: Genitron Instruments GmbH, Germany). The central part of this measurements system is AlphaGUARD PQ 2000 PRO whose detector is a radon pulse ionization chamber with active volume 0.56 dm³. The instrument was working in the diffusion mode with the measurement time interval of 60 minutes [AlphaGUARD User manual, 1998]. Determination of equilibrium equivalent concentration of radon progenies as well as equilibrium factor is performed by Radon WL Meter TN-WL-02 (manufacturer: Thomson Nielsen, Canada) connected to the AlphaGUARD measuring system and is using its internal memory to store recorded data.

Radon monitoring in the Manita peć cave started on 2nd of June 2010 by positioning 17 detectors inside the cave (Figure 2.) – eleven of them were along the tourist route (locations M4, 5, 7, 9, 11 - 17), and six were exposed inside the area of restricted movements (M1, 2, 3, 6, 8, 10) which is defined by the National Park's Action plan [2007, p 123] for the Manita peć cave. According to the researching plan, detectors were exposed in the following climatic seasons: summer 2010 (June – August), autumn (September – November), winter (December 2010 – February 2011), spring (March - May), and summer 2011 (June – August) with shorter monitoring periods during summer 2010 and 2011 that coincides with the intensive tourist activity.

After being exposed, LR 115 type II detectors were chemically etched in 10% aqueous solution of NaOH at a temperature of 60 °C for 120 minutes. Tracks were counted visually with the optical microscope Olympus BX51 with the magnification of 10x10. Radon concentrations are determined as the product of track densities on diffuse detector and sensitivity coefficient (30.0 ± 2.0 Bq m⁻³ / tr cm⁻² d⁻¹) which was determined during the calibration process at the NRPB radon chamber

(National Radiological Protection Board; Chilton, Didcot, Oxfordshire, Great Britain), as well as at the former radon chamber of Physics Department at Faculty of Education in Osijek, Croatia.



Figure 2. Ground plan of Manita peć cave with marked measuring locations of radon detectors (picture is given by courtesy of National Park Paklenica).

Results and Discussion

Radon levels in Manita peć cave

The results of all measurements performed by series of solid state nuclear track etched detectors LR 115 type II in the Manita peć cave for the appropriate climatic season are summarized in Table 1. The entrance of the cave is different than the rest of the cave in geomorphological sense and the radon concentration on this location is under the strong influence of air circulation as the consequence of two entrances that are 10 meters apart from each other. According to this, these measured values from M17 location will not be included in further consideration.

Table 1. The average radon concentrations with associated standard deviations ($c \pm \sigma_c / \text{Bq m}^{-3}$) in Manita peć in different climatic seasons and for two different location sets of detectors. Parameters of applied statistical t-test (*t* is empirical and t_{crit} is theoretical values of t-distribution while *df* is degree of freedom).

	$c \pm \sigma_c / Bq m^{-3}$				
	Summer 2010	Autumn 2010	Winter 2010	Spring 2011	Summer 2011
Tourist route	1065 ± 135	252 ± 54	31 ± 7	132 ± 32	1056 ± 150
Restricted area	1153 ± 249	297 ± 29	34 ± 8	144 ± 25	1043 ± 115
t	1.131	1.863	0.619	0.831	0.083
t _{crit} ; df	2.074 ; 22	2.160;13	2.131 ; 15	2.145;14	2.048;28

Our assumption that these differences between average radon concentrations are not statistically significant was confirmed by an appropriate statistical t-test on the significance level of 0.05 for all seasons (Table 1). To conclude, radon concentration is uniformly distributed inside the cave and its spatial distributions during the monitoring period are shown on the Figure 3. Visualization of these radon concentrations distributions was performed by ArcView software using geospatial methods of nearest neighbours. Figure 3 shows that there is a different pattern in radon concentration distribution at the beginning and at the end of both climatic summers (2010 and 2011). The average radon concentration during the first measuring period (summer_1: 2^{nd} of June – 14^{th} of July 2010) was 922 Bq m⁻³, with standard deviation of 101 Bq m⁻³. At the second measuring period (summer_2: 14^{th} of July – 01^{st} of September 2010) the average radon concentration was 1236 Bq m⁻³ with standard deviation of 124 Bq m⁻³. This statistically significant increase of radon concentration in the second

half of climatic summer 2010 (t = $6.024 > t_{crit} = 2.110$ for 17 degrees of freedom and at the significance level of 0.05) compared with the first half, showed usefulness of the detectors exposure in both periods, in order to determine effective doses received by the visitors and tourist guides as much accurate as possible. In 2011, the same test showed, that there was a statistically significant difference in radon concentration between first and second part of summer period (t= $4.257 > t_{crit} = 2.045$ for 29 degrees of freedom), too.



Figure 3. Spatial distributions of radon concentration in the Manita peć cave in each climatic season during monitoring period (June 2010-August 2011).

Comparing average values of radon concentration on all measuring locations (M1 - M16)during two successive summers 2010 ($c_{2010} = 1135 \pm 135$ Bq m⁻³) and 2011 ($c_{2011} = 1061 \pm 71$ Bq m⁻³), it is evident that the radon concentration for the same measuring locations in the same period of the year is almost identical. Together with differences between maxima and minima in radon concentration in summer and winter, respectively, it points out that the temperature differences between the air inside and outside the cave is the main mechanism for the radon concentration regulation, which is a principal characteristic of horizontally structured caves [Kobal et al., 1988, Dueñas et al., 2005, Bahtijari et al., 2008, Espinoza et al., 2008]. When the average daily temperature of the air outside the cave becomes higher than the air temperature inside the cave (which is more or less constant), the radon concentration starts to rise. Our previous measurements in Đurovića cave gave the results that radon concentration value can vary up to 45% due to climatic season influences. Compared to average yearly value – that means 2.75 time higher radon concentration in summer than in autumn period. [Radolić et al., 2009]. In comparison with similar karst caves in the neighbouring countries [Hakl et al., 1997, Cigna, 2005], Manita peć cave with its average summer radon concentration of 1 kBq m⁻³ belongs to a group of caves with radon concentration below the average (approximately 2 times lower than the average radon concentration in caves throughout the world).

Daily variations of radon concentration as well as of equilibrium factor are monitored on measuring location M3 (Figure 2) by the previously described AlphaGUARD measuring system that enables recording data every 60 minutes. Location M3 is positioned in restricted areas but not far from the end of touristic routes. The measurements were performed at the beginning and at the end of each climatic season (winter, spring and summer and autumn) in order to check possible microclimatic influence on radon levels in caves or on the concentration of its short lived progeny. In Figure 4, daily radon variation time series are shown for summers 2010 and 2011, which is the time period of intense tourist exploitation of Manita peć cave. By performing measurements we have not seen any daily variation in radon concentration as a consequence of enhanced human activity (increase/decrease radon and its short lived progenies during the cave tour).



Figure 4. Time series of radon concentration (•; $c / Bq m^{-3}$) at measuring location M3 in Manita peć cave in summer 2010 and 2011 during enhanced human (mainly tourist) activity.

Equilibrium factors

Equilibrium factor *F* is determined according to described method with two LR 115 II detectors [Planinić et al., 1997] and average equilibrium factors for each monitoring season: $F_{SUMMER-2010} = 0.42 \pm 0.18$, $F_{AUTUMN-2010} = 0.53 \pm 0.17$, $F_{WINTER-2010} = 0.61 \pm 0.15$, $F_{SPRING-2011} = 0.58 \pm 0.15$ and $F_{SUMMER-2011} = 0.41 \pm 0.15$ (Figure 5). The average equilibrium factor during the monitoring period of fifteen months was 0.48 with standard deviation of 0.18. The results of our measurements are in good agreement with measurements of equilibrium factors in other karst caves [Solomon et al., 1992, Szerbin, 1996, Vaupotič et al., 2008], although many authors use the supposed value of 0.5 for the average yearly equilibrium factors for the caves [Hyland and Gunn, 1994, Duffy et al., 1996, Gillmore et al., 2000]. The two-day measurements of equilibrium factor of $F_{AlphaGUARD} = 0.50 \pm 0.18$ for the period autumn 2010 - summer 2011.



Figure 5. The average values of equilibrium factors *F* in monitored climatic seasons with its standard deviations.

If we consider the detector position for the each measurement location along the tourist route, as well as in the restricted area, the differences between the obtained equilibrium factors in these two sets are not statistically significant (at significance level of 5%) in every monitoring season (the calculated probabilities of applied t-distributions for summer 2010, autumn 2010, winter 2010, spring 2011 and summer 2011 are: 0.430, 0.097, 0.722, 0.286 and 0.192, respectively). This means that the equilibrium factors are uniformly distributed inside the cave as well.

Effective dose assessment for the visitors and tourist guides in the Manita peć cave

The Manita peć cave can be visited only under the escort of a tourist guide, and the group usually counts 25-30 persons. The sightseeing tour lasts approximately 30 minutes. The most intensive tourist activity is during the summer period (June – August) when almost $\frac{3}{4}$ of total tourist number visit the cave [Action plan, 2007]. In 2010, 11,500 visitors visited the Manita peć cave (Table 2), out of which 8500 came during the summer period. A similar distribution of visitors was in 2011, with almost the same number of visitors (11,700).

Months Working hours		N in 2010	N in 2011
APRIL	Saturday from 10.00 a.m 1.00 p.m.	173	477
MAY	Wednesday and Saturday from 10.00 a.m 1.00 p.m.	737	683
JUNE, OCTOBER	Monday, Wednesday and Saturday from 10.00 a.m 1.00 p.m.	1384	1493
JULY, AUGUST, SEPTEMBER	Every day from 10.00 a.m 1.00 p.m.	9225	9039

Table 2. Working days of the Manita peć cave in NP Paklenica and number of visitors per months, Nduring tourist seasons (from April to October) in 2010 and 2011.

By using equation (1) the effective dose rate for the cave visitors in summer 2010, with the average radon concentration ($c_{SUMMER} = 1113$ Bq m⁻³) and average equilibrium factor ($F_{SUMMER} = 0.42$), was 3.7 µSv/h. Thus, cave visitors during their 30 minutes visit in period from June – August received the effective dose of 1.85 µSv. For the same months in 2011, a visitor received the effective dose of 1.7 µSv.

This is, of course, the simplest model for assessing the effective dose for visitors, which can be improved by taking in account the difference in radon concentration as well as equilibrium factor in the first and second part of the summer, and the difference in radon concentration on every measuring location along the tourist route. The dose rate in the Manita peć cave during the first part of summer 2010 was 2.6 μ Sv/h, and in the second part of summer 4.1 μ Sv/h. This means that cave visitors in June and the first half of July received a smaller dose than those who visited the cave in the second part of July and August ($E_{June-July} = 1.3 \mu$ Sv compared to $E_{July-August} = 2.05 \mu$ Sv). In 2011, the dose rate in the first part of summer was 3.2 μ Sv/h and in the second part 3.7 μ Sv/h, which makes a smaller difference in the received doses as well: $E_{June-July} = 1.6 \mu$ Sv compared to $E_{July-August} = 1.85 \mu$ Sv.

If we take in account the radon concentration and equilibrium factors on the locations along the tourist route (3, 4, 5, 7, 9, 11, 13, 14, 15, 16) and the approximate time spent in the vicinity of these locations (expressed in minutes as follows: 3, 3, 5, 3, 5, 2, 5, 2, 1, 1), than equation (1) becomes:

$$E = 7,923 \cdot 10^{-6} \begin{pmatrix} \overline{c_3} \cdot \overline{F_3} \cdot \frac{1}{20} + \overline{c_4} \cdot \overline{F_4} \cdot \frac{1}{20} + \overline{c_5} \cdot \overline{F_5} \cdot \frac{1}{12} + \overline{c_7} \cdot \overline{F_7} \cdot \frac{1}{20} + \overline{c_9} \cdot \overline{F_9} \cdot \frac{1}{12} + \overline{c_{11}} \cdot \overline{F_{11}} \cdot \frac{1}{30} + \overline{c_{15}} \cdot \overline{F_{15}} \cdot \frac{1}{60} + \overline{c_{16}} \cdot \overline{F_{16}} \cdot \frac{1}{60} \end{pmatrix}$$

The calculated average effective dose, that was received by the visitors in 30 minutes of their visit of the Manita peć cave in the first part of summer 2010 was 1.6 μ Sv and 2.2 μ Sv for the visitors in the second part of summer 2010. Similar values obtained for summer 2011 when the calculated average effective dose received by the visitor was 1.6 μ Sv for the first part and 1.5 μ Sv for the second part of summer. Although the average radon concentrations and average equilibrium factors were more or less the same in both years (2010 and 2011), small fluctuations on some locations along the tourist route gave an interesting and surprising result. The higher average value of radon concentrations inside the cave in the second part of summer 2011, in comparison to the first part with the same equilibrium factors, gave smaller effective doses to the visitors in late July and August. Of course, these differences are small and lie inside the errors of radon measurements and dose assessments.

In 2010, six tourist guides spent 426 hours inside the cave in total. Seven of them were working for 408 hours in 2011. The highest effective dose in both years received a guide who spent the most time in the cave and the received doses were 0.247 mSv in 2010 and 0.215 mSv in 2011. If we compare these values with the ICRP recommendations [1994] or EU [CEU, 1996] about the maximum dose from radon and its short lived progeny for the occupational exposures (maximum effective dose of 20 mSv per year and cumulative dose in 5 years should be lower than 50 mSv - which means 10 mSv per year in average!), it is clear that effective doses received by tourist guides in the Manita peć cave inside National Park Paklenica are 50 times lower than the maximum doses allowed.

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