Carpathian Journal of Earth and Environmental Sciences, August 2013, Vol. 8, No. 3, p. 67 - 74

MONITORING OF RADON LEVELS IN SOME TOURISTIC UNDERGROUND ENVIRONMENTS FROM ROMANIA

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Abstract: The purpose of this research is to provide the distribution of radon levels in three underground environments of tourist interest from Romania ("Urşilor" Cave, "Muierilor" Cave and Turda Salt Mine). This study is of great interest since it identifies the values that could present a potential long-term health risk for the full-time staff (guides) spending extended periods conducting tours or carrying out maintenance within these underground environments and less for tourists. Furthermore, a possible relationship between the radon values and the local geology was disscused. Indoor radon concentrations were measured by using solid state CR-39 type RSKS nuclear track-etch detectors that were exposed from 3 to 6 months. The results reveal low radon levels in salt mine with the annual average concentration below the detection limit (around 8 Bq m^{-3}), related to the salt plastic rock without fissures, fractures and consequently, without circulation pathways for radon into the salt mine chambers. This type of environment is proper to be used for speleotheraphy and spa tourism. "Muierilor cave" has relatively low radon concentration varying between 63 and 172 Bq m^{-3}, with only one value of 1184 Bq m^{-3}, as compared with "Urșilor"Cave, which values are in the range of 783-1795 Bq m^{-3} indicating the need of further long term monitoring by using both the passive and the active methods. Our results are comparable with radon concentration in different underground environments reported from other European surveys, lower than many of them. Geological background of these areas could sustain the measured values, on the one side due to the presence of granitic plutons and even the uraniferous mineralizations proximity, and on the other side due to the presence of limestone and its gneiss and mica-schist rocks basement that causes the low diffusion coefficient of radon.

Keywords: indoor radon, CR-39 nuclear track detector, underground environments, geology.

1. INTRODUCTION

Radon is a naturally occurring radioactive gas continually generated from rocks and soils, all over the world, which can diffuse for several meters before decaying into its short-lived decay products ^{214}Po, ^{218}Po, ^{214}Pb, and ^{214}Bi. Radon is a known human carcinogen, representing the major source of ionizing radiation exposure for population. Since prolonged exposure to elevated radon concentrations causes an increased risk of lung cancer, it is important to measure the radon levels in air in all environments and to apply the mitigation action when they exceed the action levels established by the ICRP (Cosma et al., 2009; ICRP, 2010).

As a naturally occurring radioactive element in the ^{238}U decay series, radon can reach very high levels in certain geological formations such as caves or mines. Once accumulated inside the underground environments (caves and mines) radon may induce a potentially health risk for the exposed employees (UNSCEAR, 2008; ICRP, 2010). A health risk linked to radon due to the short period of exposure has not been demonstrated to visitor's members of the public.

The levels of radon in underground spaces could be varying significantly depending on the several factors like geological structure, radium concentration in the rock, porosity, air and water flow, atmospheric pressure and ventilation of undergrounde site (UNSCEAR, 2008). A low level of ventilation could lead to the accumulation of radon and thereby to a high concentration. In general, a seasonal variation of radon levels, with summer maximums and winter minimums is typical for cold karstic caves.

The geology directly influences the concentrations of radon in underground spaces, considering that the main radon source is represented by the radioactive mineral content of rocks within the earth's crust.

In this study, results of the distribution of radon levels in three underground environments of tourist interest from Romania ("Urşilor" Cave, "Muierilor" Cave and Turda Salt Mine – Fig. 1) are presented, in order to evaluate the exposure to radon in underground air and correlate this level with the local geology.

Figure 1. Location of underground environments studied

This work represents the first experience with radon measurements in Romanian touristic caves, and the results may be considered the input for the design of a more extensive radon monitoring campaigns in all touristic underground environments from Romania, taking into account the geology.

2. GEOLOGICAL BACKGROUND

2.1. "Urşilor" Cave

Urşilor Cave is located in the north-western part of Romania, in Bihor Mountains, West Carpathians. It is carved in Upper Jurassic (Tithonic) limestone and consists of 1500 m of galleries situated along two levels, at 100 m and 200 m beneath the surface. The cave presents a major scientific importance due to well preserved paleontological remains of Cave Bear (*Ursus spelaeus)* and due to speleothems diversity. Most of the Cave Bear remains are hosted by the lower level, still active and preserved as a scientific reserve. The upper level is no longer hydrologically active; it is highly decorated and open for tourist access (Onac et al., 2007).

Structurally, the limestone is part of Vălani Nappe, the lowermost structural unit of the Codru Nappe System. This system is part of Apuseni Mountains and consist of many overthrust nappes, most of them being cover nappes (Balintoni, 1997). Lithologically, mainly clastic Permian and carbonaceous Jurassic deposits formed these nappes. Biharia Nappe System is the second nappe system that together with the previous one were thrusted during the Turonian, over the Bihor Autochthonous Unit which forms the most part of northernmost Apuseni Mountains.

Bihor Autochthonous Unit consists of medium grade metamorphic rocks basement (micaschist and quartzite to ortho-gneiss, amphibolite and other metabasic rocks). This unit is being distinguished by a high frequency of pegmatites, important migmatisation phenomena and presence of great sized granitic intrusions such as Codru granites (Balintoni, 1997).

The Paleozoic and Mesozoic sequences are crossed by a series of banatitic (late Cretaceous) magmatic intrusions and covered by volcanics lithologically consisting especially of dacites and rhyolithes. Moreover, ryolithic permian flows appear as interlayers in the Permian deposit sequences (Stoici, 1983). The limestones from our interest area, were more or less affected by the banatitic intrusions from close proximity (contact aureola). At about 30 km south from our interest area is located Băiţa uranium mineralized zone. The uranium mineralization is related to the postmagmatic solutions released by the Băiţa granitoid pluton (Niţu, 2000). The metal assemblage is U-(Co-Ni) of hydrothermal origin, being associated with enrichment in some other elements such as Fe, Pb, Zn, Cu and Mo (Savu, 2011).

2.2. "Muierilor" Cave

"Muierilor" Cave is located in the central part of Romania, on the south-facing slope of Parâng Mountains (South Carpathians). "Galbenului" river carved the Tithonic-Neocomian limestone pile from this area and build-up "Galbenului" Gorges whose right slope hosts the cave. This limestone pile appears on 1-1.5km in width and 12 km in length, on the south-facing slope of Parang Mountains and also in Capatanii Mountains divided from the last ones by the Oltet River.

"Muierilor" Cave consists of an underground gallery system disposed on four levels, the upper level (2) only being exploited by tourists. Lower level (3) has scientific value and the other two only have speleogenetic value. Recent studies (Diaconu et al., 2008) report that the total length of the galleries is more than 7000 m. The general development of the whole system is NNV-SSE, following the direction of one important fault line. The speleogenetic evolution of "Muierilor" Cave is closely linked by water infiltration from the "Galbenul"river along the fault line and formation of the gorges by the main water stream (Bleahu et al., 1976). Moreover, this evolution ends up very close to the impermeable granitic bed body from the basement, where the river left the underground path. Further on, the karstification processes were carried on by percolation water from the surface.

Structurally, the studied cave area is overlapping to the Danubian domain, the alpine tectono-stratigraphical unit from the lowermost part of the South Carpathians nappe systems (Getic-Supragetic nappe system, Severin nappe system and Danubian nappe system) (Balintoni et al., 2011).

The limestone pile that host "Muierilor" Cave is situated at the eastern end of the tectonic window "Parâng-Retezat-Almăj" that reveals the Danubian domain under the Getic crystaline covering it (Mutihac & Ionesi, 1973). The limestone represents the sedimentary cover of the pre-Alpine basement consisting mainly of metamorphic rocks and granitoids. In our study area the metamorphic rocks sequence in the Danubian basement belongs to the Lainici-Păiuș terrane. This consists of metasedimentary rocks (marble, different types of gneiss and amphibolite) characterised by low pressure/high temperature conditions. In its northern part, the Danubian basement is formed by a metavolcanic complex belonging to the Drăgșan terrane characterised by medium pressure/ temperature metamorphism.

In different times, the Danubian domain was affected by highly magmatic activity that resulted in numerous granitoid plutons which intrude the Lainici- Păiuș terrane basement and also the Drăgșan terrane basement. A such intrusive body in the Lainici Păiuș terrane basement is the Cadomian Olteț granitoid pluton situated under the Thitonic-Neocomian limestones.

The recent U/Pb geochronology of the zircons determined by LA-ICP-MS (Balintoni et al., 2011), highlighted that granitoids intruded in the southern part of the Lainici-Păiuș terrane are older (600-590 Ma) than the ones intruding the Drăgșan basement which are predominantly Variscan in age.

2.3. Turda Salt Mine

Situated in the Turda city, on the western border of Transylvanian Depression, Turda Salt Mine is one of the well-known targets of interest for the history of salt exploitation from Romania. It was exploited between 1690-1932. Although in 1932 the mining activity ceased, the existing premises referring especially to the tourism and therapeutic potential that it holds, have kept Turda Salt Mine in the area of national interest. Thus, in 1992 the Turda Salt mine was open to visitors, tourist route including: the gallery used for salt transportation (Franz Joseph Gallery), the exploited salt mines: the bell shape chambers (Joseph and Terezia) and the trapezoidal shape chambers (Rudolf and Ghizela). Due to the really impressive proof of the mining heritage and the perspective of a proper touristic and therapeutic exploitation, the Salt Mine was rehabilitated between 2008 and 2010, the Salt Mine. The new design of the mine adds more value to the objective by the quality of well preserved heritage mining elements, on the one hand and by the modern design and mine chambers' functionality, on the other hand (Bican-Brișan et al., 2011). Since 2010, the Turda Salt Mine is visited by over 350 000 visitors/year.

Salt mine microclimate is one particular aspect used as a therapeutic method. The curative effects of the salt exploitation chambers are given by an ensemble of physical, chemical and biological conditions, which interact and have a complex effect on the human organism, especially upon respiratory dysfunctions.

The salt belongs to an evaporitic horizon that extends in the whole sub-surface of the Transylvanian Basin. From litostratigraphical perspective this belongs to Ocna Dej formation (Mészáros, 1991).

The basement of the area is formed by crystalline rocks, ophiolites, banatites and Jurassic, Cretaceous and Paleogene sedimentary deposits. Moreover, from the tectonic point of view, the region belongs to the western area of Transylvania that shows a specific structure consisting of a closely-grouped succession of symmetrically arranged anticlines and synclines usually having N–S development. Turda salt massif crosses out as diapir associated to the Măhăceni-Ploscoş anticline. It has an elongated shape about 4 km long and 700 m to 200 m in width; sometimes the diapirism was so intense that the salt took the shape of an almost vertical "diapiric blade". In the anticline axis the salt width varies from 750 m to over 1000 m (Petrescu & Bican-Brişan, 1997).

3. MATERIALS AND METHODS

3.1. Measurement sites

3.1.1. "Urşilor" Cave

"Urşilor" Cave is a great cave in Romania, visited by about half a million tourists a year. There is no forced ventilation in the cave and the air is exchanged by natural air draught only through numerous cracks, corridors and shafts connecting the cave with the outdoor atmosphere.

In order to evaluate the distribution of radon concentrations 10 CR-39 radon detectors were placed through the main galleries of "Urşilor" Cave during July - October 2011 (Fig. 2). Other 2 detectors were used for background measurements.

3.1.2. "Muierilor" Cave

5 radon track detectors were placed along the horizontal gallery on the 573 m long, in different parts of the cave between May and August 2012 in order to evaluate the distribution of radon concentration (Fig. 3).

The gallery is located on the upper level of the karstic system which due to the aspect of underground halls and well-decorated wall by calcareous formations is touristic exploited.

3.1.3. Turda Salt Mine

For evaluating the distribution of radon concentration 6 CR-39 radon detectors were placed in different locations of Turda Salt Mine (Ghizela, Rudolf, Josef and Terezia Chamber Mines, as well as to the Josef Mine Balcoon situated at the level of Frantz Josef transport gallery and Managing Director Office, at the surface level between January and May 2012 (Fig. 4).

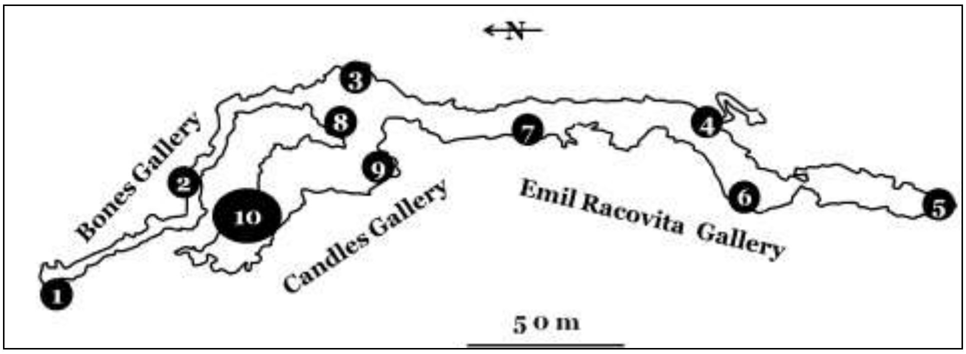

Figure 2. Location of radon detectors in "Urşilor" Cave.

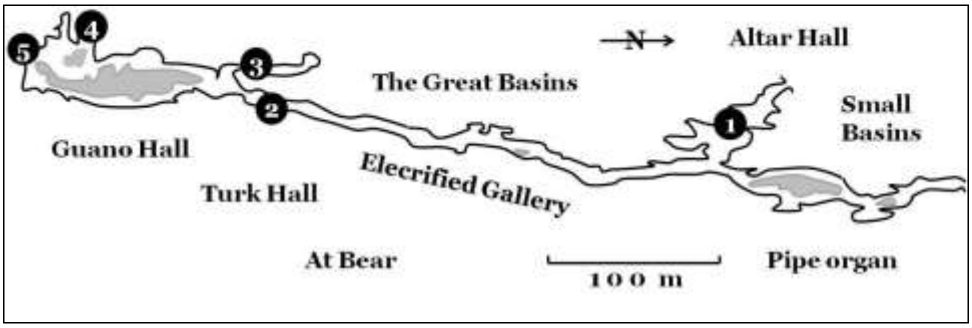

Figure 3. Location of radon detectors in "Muierilor" Cave.

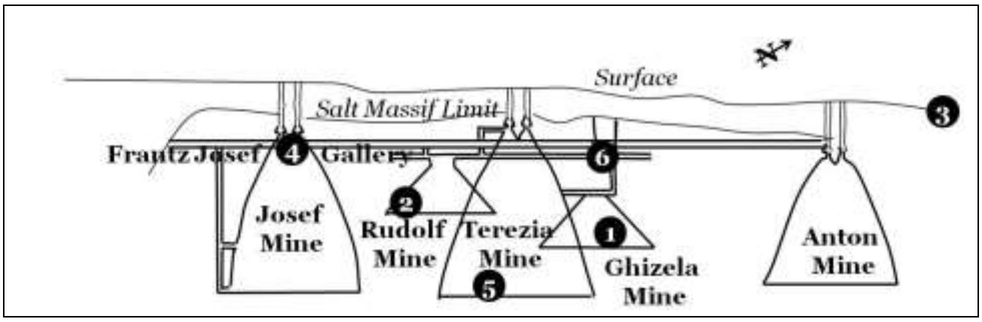

Figure 4. Location of radon detectors in Turda Salt Mine

3.2. Radon measuring techniques

Radon measurements in underground air were carried out by using passive devices employing CR-39 solid state nuclear track detectors, commercially known as type RSKS of RadoSys. This type of detector is based on a CR-39 chip placed under the cap of a cylindrical diffusion chamber sensitive to radon activity. The measurement protocol was made in compliance with quality assurance and control programs proposed by HPA-NRPB or other European authorities (Miles & Howarth, 2008). Several details concerning the experimental procedure has already been described elsewhere (Cosma et al., 2009; Sainz et al., 2009; Cucoş (Dinu) et al., 2012).

The detectors were placed in the main galleries of the studied underground environments for a minimum 4 months in the period 2011-2012. At the end of exposure, the detectors are collected and processed in our laboratory with the aid of RadoSys equipment (produced by from Hungary).

Radon concentration (C_{Rn}) were calculated by the following relation:

$$C_{Rn} = \frac{(\rho \cdot F)}{t} \qquad (1),$$

where $\boldsymbol{C_{Rn}}$ is radon concentration expressed in Bq m^{-3}, $\boldsymbol{\rho}$ represents the track density expressed in tracks mm^{-2}, $\boldsymbol{F}$ is the calibration factor, calculated from manufacturer, expressed in (kBq h m^{-3})/ (tracks mm^{-2}) and $\boldsymbol{t}$ is exposure time expressed in h.

The average measured radon concentration value (C_{Rn}) was corrected for seasonal variations, depending on the time when detectors were exposed during the course of the year (Cosma et al., 2009):

$$C_a = C_m \left[\left(\frac{n_{sp,A}}{n} \right) + r \left(\frac{n_{s,w}}{n} \right) \right] \qquad (2),$$

where $\boldsymbol{C_a}$ is annual mean of radon concentration expressed in Bq m^{-3}, $\boldsymbol{C_m}$ represents measured radon concentration value expressed in Bq m^{-3}, $\boldsymbol{n_{sp,A,s,w}}$ is the number of months from a measurements season (spring, autumn, summer or winter) and **r** is the seasonal correction factor (**1.35 for summer or 0.67 for winter period**) calculated as reference for Romania (Cosma et al., 2009).

The reliability and the accuracy of measurement system were regularly certified in our laboratory by the successful participation to several international intercomparisons, including several calibrations of the detectors at our laboratory using a radon reference chambers (Cucoş (Dinu) et al., 2012).

At the most recent intercomparison exercises organized by NIRS, our results were categorized into the best category with the relative variations less than 4 % (Cosma et al., 2013).

4. RESULTS AND DISCUSSION

Descriptive statistics of annual radon concentration in studied underground environments of tourist interest (***"Urşilor" Cave, "Muierilor" Cave*** and ***Turda Salt Mine***) from Romania are given in table 1. The arithmetic means were used for description and comparisons of results.

As can be seen from table 1, the radon concentration values vary very wide from the limit of detection (8 Bq m^{-3}) to 1795 Bq m^{-3} depending on several factors, such as the geology of surrounding area and ventilation.

The highest values (between 783 and 1795 Bq m^{-3}) were found inside "Urşilor" Cave. However, these values could be considered relatively low compared to values found in other limestone cave in the world (e.g. Tapolca, Hungary up to 11.5 kBqm^{-3}, Petralona, Greece up to 88 kBqm^{-3}). Inside "Muierilor" Cave radon values were low, ranging between 63 and 172 Bqm^{-3}, with only one value of 1184 Bq m^{-3}.

There are some aspects to be discussed concerning the results in this study. The first aspect is related to the potential source of radon. Due the fact that limestone is a bad radon source, this source has to be searched outside.

However, the clay intercalations from limestone can be taken into consideration from this point of view. As a consequence, limestone due to fissures from rock mass contributed more to the radon circulation and less to radon accumulation. The limestone basement consists of gneiss and mica-schist. Mica participates in a significant proportion in these rocks. It is known that flaky minerals like mica have a shape factor that causes the diffusion coefficient to be one-half to one-third the theoretical value (Cigna, 2005). Consequently, the rocks that contain significant proportions of flaky minerals usually oriented so as to impede vertical movement.

Moreover, this type of rocks has not radiogen significance in relation with highly spread granites. It seems that old granitoids (600-590 Ma), disposed on the two southern alignments of the Lainici-Păiuș terrane including Oltet granitic pluton from limestone bed layer, have relatively low radon potential if compared to younger varistic granitoids from Dragsan terrane in the northern part of Parang Mountains.

The second aspect that is considered to be significant in radon accumulation in "Muierilor" Cave is related to air circulation inside the cave.

Table 1. Summarized statistics for the distribution of radon levels in studied underground environments of tourist interest from Romania ("Urşilor" Cave, "Muierilor" Cave and Turda Salt Mine), performed during 2011-2012.

Investigated underground environment	**No. of detectors**	**A.M. ± S.D. *(Bq m^{-3})***	**Annual radon conc. *(Bq m^{-3})***	**Range of radon conc. *(Bq m^{-3})***	**Investigated season**
"Urşilor" Cave	10	1477 ± 270	2216	783-1795	Summer-Autumn
"Muierilor" Cave	6	339 ± 475	509	63-1184	Summer
Turda Salt Mine	5	below the limit of detection 8 ± 6	below the limit of detection	below the limit of detection -15	Winter-Spring
Background measurements	2	41 ± 1	62	40-42	Summer-Autumn

There is a high air circulation along the main gallery where the detectors were placed, considering that there are two entrances with metal bars doors. There was only one exception to this, the value of 1184 Bq $m^{-3,}$ determined in an isolated area of the cave that allowed it to accumulate. It is known that in a cave with strong ventilation, radon can be diluted with outside air, resulting in a lower concentration. This is an important factor when the air exchange time is less than the radon half-life of 3.8 days (Whittlestone et al., 2003)

We also have to consider that radon concentration is influenced by temperatures variations inside and outside the cave so it fluctuates with seasonal change. Generally, a seasonal fluctuation is found with a maximum in summer and a minimum in winter (Cigna, 2005). Our measurements were performed during high temperatures (may-august) so during important temperature variations between the inside and outside atmosphere. This is an aspect that facilitated pressure differences that stopped cold air from going outside trough limestone fractures and stopped hot air from entering in the cave trough cave's access points.

Therefore, it is considered that values are even lower on winter, fact that will be highlighted trough further research.

In "Urşilor" Cave there is a whole different situation. There values are higher than the ones earlier presented. On the one hand, there we are talking about Permian sandstones, with ryolithic layers that conferred to Europe and also Romania a radiogenic potential. Moreover, an important radon source in the area could be the uraniferous mineralisation from Băiţa Bihor given by the magmatic differentiation highlighted trough banatitic intrusion in the Codru Nappe System's basement. Therefore, in this context even higher radon values would be expected in this cave. Moreover, further research on important radon seasonal variations is to be performed. It is normally expected in workplaces (as we could consider this environment from guide perspective) with radon levels above 1000 Bq m^{-3} that active measures be taken to reduce the radon concentration, usually by improving the ventilation. Increasing the ventilation in caves is not possible since it affects the delicate balance of carbon dioxide, partial pressure, temperature and humidity, with the risk of damage to cave decoration.

A particular situation is represented by the underground salt environment of Turda Salt Mine where radon values found are very low. The results showed in Table 1 point out low levels of indoor radon concentration, between the limit of detection (8 Bq m^{-3}) and 15 Bq m^{-3}. Our results reports similar values of radon in Turda Salt Mine previously obtained by active measurement method, in range between 7 – 12 Bq m^{-3} (Călin et al., 2011).

These low values are related to the impossibility of radon to break inside the salt chambers. This is due to salt plastic properties that stop rock fracturing and, as a result, stops radon from entering the mine. Moreover, there is the salt diapir structure that gives a considerable thickness to the salt. In this situation we can also take into consideration clay interlayer but these are less quantitatively important and the potential source represented by this type of impurities in the salt mass is highly isolated. Furthermore, air movement inside the salt mine is vertically and totally insignificant in comparison to air movement representative for cave environment.

In addition, we compare our results with radon concentration in different underground environments reported from other European surveys, shown in table 2.

5. CONCLUSIONS

Our results provide the first data of radon concentration in two types of underground environments from Romania ("Urşilor" and "Muierilor" *caves* and Turda *salt mine*) by using CR-39 nuclear tracks detectors. The work presents the first experience regarding radon measurements

Table 2. Radon levels in different underground environments from other European surveys

Cave/ Salt mine	Location	Average radon conc. (Bq m^{-3})	Reference
Tapolca	Hungary	660 - 11500	*Somlai et al., 2007*
Sannur	Egypt	701 - 1274	*Amin & Eissa, 2008*
Perama	Greece	197 - 1929	*Papachristodoulou et al., 2004*
Petralona	Greece	190-88060	*Papastefanou et al., 2003*
Creswell Crags	UK	27-7800	*Gillmore et al., 2002*
Niedzwiedzia	Poland	100-4180	*Przylibski, 1999*
Altamira	Spain	186-7120	*Lario et al., 2005*
Nerja	Spain	5-488	*Duenas et al., 1999*
Postojna	Slovenia	2300-4200	*Vaupotic et al., 2001*
Ailwee	Ireland	500-4200	*Duffy et al., 1996*
Mitchelstown	Ireland	3100-9200	*Duffy et al., 1996*
Guacharo	Venezuela	80-3200	*Sajo-Bohus et al., 1997*
Ocna Dej	Romania	9.14-31.7	*Călin & Călin, 2010*
Cacica	Romania	20-96.5	*Călin et al., 2011*
Turda	Romania	6.9-12	*Călin et al., 2012*
“Urşilor” Cave	**Romania**	**1477**	***Present work***
“Muierilor” Cave	**Romania**	**339**	***Present work***
Turda Salt Mine	**Romania**	**8**	***Present work***

in Romanian touristic caves, and the results may be considered the input for the design of a more extensive radon monitoring campaigns in all touristic underground environments from Romania, taking into account the geology.

High levels of radon concentration in the range of 783 and 1795 Bq m^{-3} were found in “Urşilor” Cave which indicates the need for further long term monitoring by using both the passive and the active methods. Lowest levels of indoor radon concentration between the limit of detection (8 Bq m^{-3}) and 15 Bq m^{-3} were found in Turda Salt Mine

The results were analyzed in geological context of the area but no clearly statistically significant correlations were found at this stage of the research.

The information achieved from present survey and other studies that will be further performed will be used to identify those karst areas from Romania where radon evaluation of touristic caves would be required. In addition, the main factors affecting radon concentration in each studied underground environment, such as the local geology in relation to ventilation and meteorological conditions, were also examined. Influence of forced ventilation caused by frequently door and window opening during working hours with typical dawn and weekend peaks is evident in all caves.

Acknowledgements

We address special thanks to Prof.dr. Ioan Balintoni for useful discussions and competent reviews on the geological issue of the paper.

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Received at: 11. 01. 2013
Revised at: 15. 06. 2013
Accepted for publication at: 25. 06. 2013
Published online at: 27. 06. 2013