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STUDY OF A PASSIVE RADON MITIGATION PROCESS AND INDOOR RADON CONCENTRATION'S TIME DEPENDENCE AFTER MITIGATION

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Abstract: The uranium mining was stopped in the Mecsek Mountains (S-Hungary) in 1997 and mine reclamation of the contaminated area began. For this purpose radiometric survey of houses in the towns of the former mining area, including settlement of Kővágószőlős, was undertaken. Long-term indoor radon monitoring by using etched track detector in dwellings was a part of the radiometric survey. Most of the dwelling houses, located close to a mine-tunnel, show a yearly average of indoor radon activity concentration higher than the EU recommendation for existing buildings (400 Bq/m^3). A typical two story brick house was selected to test a mitigation method. The house cellar showed 1950 Bq/m^3 on average of indoor radon activity concentration during the summer of 2004 (03.06.2004-30.08.2004), and in the bedroom above the cellar 450 Bq/m^3 (by etch track detectors). To reduce this high radon concentration, a new radon mitigation was completed in the house. Before mitigation, the soil radon characteristics were determined around the house, which shed light on high radon exhalation values (range 20-200 mBq/m^{2}s, average: 100 mBq/m^{2}s) and significant radium content (60-65 Bq/kg). Indoor radon concentration (averaged for 1 hour) was also monitored continuously for one month once before and three times after the mitigation technique was applied. Due to the mitigation, the initial radon concentration was decreased by 71% (average radon concentration in the same period of the year before the mitigation: 1480±74 Bq/m^3, after the mitigation: 420±33 Bq/m^3). Time dependence of radon concentration after the mitigation shows characteristic features due to the change in meteorological conditions, such as variations in temperatures. Daily dependence of the radon concentration is clearly showed by high radon activity values which occurred a few times in the afternoon governed by temperature change. High values these peaks result in a relatively high average radon level (1480±74 Bq/m^3). However, the radon concentration at nights is always significantly lower than the average.

Keywords: indoor radon concentration; mitigation; radon monitoring; passive system; daily periodicity

1. INTRODUCTION

Nationwide measurements of radon activities in the indoor air of dwellings are continuously presented all over the world. The radon concentration does vary not only with seasonal changes but also with the age, the construction mode of houses, the ventilation conditions and with specific sites and geological materials (Oufni et al., 2005). Elevated radon levels can be found more often in houses with older buildings made up of blocks, stones, muddy walls and having concrete roofs (Rafique et al., 2011). To reduce the health risk caused by indoor radon, several radon surveys have been carried out in the last decade in many countries in Europe (Denman et al., 2005). As a result of these projects houses showing radon levels higher than recommended by the EU (400 Bq/m^3) (90/143/Euratom), were selected for radon remediation or test of mitigation processes. There are several ways to reduce the radon level in buildings. Radon reduction system for prevention and mitigation require the following design criteria. It should be able to reduce the radon concentrations considerably below the reference level; safe which not create back-drafting; durable and functional

techniques during the expected life of the building; easy to monitor the performance; quiet and unobtrusive; and low cost for installation, operation and maintenance (Zeeb et al., 2009). The main aim of the mitigations and the remediation is to reduce the yearly radon level below the limit by the lowest cost. Radon mitigation strategies need to be adapted to the specific mixtures of housing and building characteristics, climate zones, radon sources, and transport mechanisms in order to be cost-effective (Zeeb et al., 2009). The commonly used radon mitigation methods include active methods like soil depressurization, under floor active ventilation, building or soil pressurization, and in very porous soils radon wells. On the other hand, there are some passive methods that do not need electric input, for instance, under floor passive ventilation, sealing of surfaces and radon membranes and barriers (Zeeb et al., 2009). The best results are achieved by combining these methods (e.g., sealing, active soil depressurization and building pressurization), according to the local features (Korhonen et al., 2000). The qualitative efficiency of the remediation process can be defined by the average radon reduction factor: ARRF=A/B, where A is average radon activity concentration after mitigation and B is average radon activity concentration before mitigation (Maringer et al., 2001). This is the percentage to which the initial radon concentration is reduced after the mitigation process. The ARRF or $R_{A/B}$ (after/before mitigation ratio) can be defined using average values of radon concentration before and after the mitigation for different time intervals. Allison et al., (2008) for instance, used the mean daily radon concentrations throughout the period of their 5-weeks study (21 days before and 14 days after the remediation).

The site of our study is located above a closed uranium mine where a remediation process has been carried out, and more than a hundred houses were examined for annual average indoor radon concentrations. A previous solid state nuclear track detector survey (Somlai et al., 2006) showed that there were approximately 30 houses where the annual radon activity concentrations exceeded the recommended action level (ICRP 65) of 600 Bq/m^3. A representative house and its vicinity, from those 30 homes studied previously, had been chosen for determining its natural radioactivity and its source of radon. These results clearly indicated the necessity of mitigation (Nagy et al., 2009).

In this paper we are dealing with results mitigation of rooms having the highest radon level in the selected house. We also tested the efficiency of the mitigation method which was designed by the vender (Dörken Ltd.) to use during the construction of a new house, however we applied it primarily as a post-construction method.

2. DESCRIPTION OF THE STUDY SITE

The area of the studied house is located in the western part of Mecsek Mountains (South Hungary) (Fig. 1). The geology of the whole site is well known as a result of the 40 years of uranium mining activity done. The village was built upon Permian sandstone called Kővágószőlős Sandstone Formation. The uranium mineralization formed at the boundary of grey reductive Kővágótöttös Sandstone and red oxidative Cserkút Sandstone. Both sandstone types are members of the Kővágószőlős Sandstone Formation (Császár, 2005).

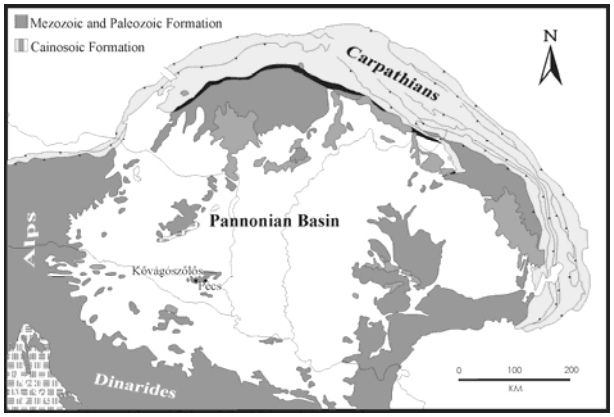

Figure 1. Map of the Carpathian Pannonian region, showing location of Kővágószőlős

The building studied is about one hundred years old, a two-story family house with a cellar, and was built of brick and local stone, basically different members of the Kővágószőlős Sandstone Formation. A garden of 700 m^2 adjacent to the house was also the object of the study area.

3. APPLIED METHOD FOR RADON MITIGATION

For the mitigation a special sealer material produced by Dörken Ltd. was applied, coupling it with a releasing pipe system coming out from the studied house. The membrane reliably drains off methane and radon gas. On the floor we used a DELTA-TERRAXX surface gas drainage system, a full-surface drainage system which is highly pressure-resistant and the membrane is perfectly rot-proof and acts as reliable drainage for radon and methane gas. On walls we used a DELTA-PT drainage system, which creates a ventilated gap between the damp basement wall and the plaster.

The collected radon gas in the membrane between the wall and the plaster is released into the air through the coupled pipe. The exhaust pipe (110 mm in diameter) is leaving the house sunk into the floor through the dining room (Fig. 2). This is a passive system, which needs no artificial ventilation.

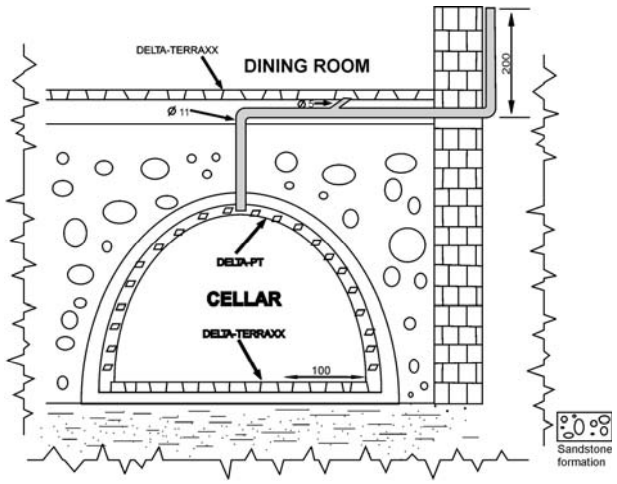

Figure 2. The cross section of the mitigated cellar. On the floor the DELTA-TERRAXX drainage system and on the vault the DELTA-PT have been used. The collected radon gas in the membrane between the wall and plaster is released through a pipe.

4. RESULTS AND DISCUSSION

4.1. Pre-mitigation measurements

Before radon reduction, a serious pre-mitigation in situ and laboratory measurement had been carried out to determine natural radioactivity levels in the selected house and its vicinity. Firstly, we completed several in situ measurements on radon concentrations of the indoor air in each room of the house, on soil gas radon concentrations in garden of the house and on radon exhalation of the soil in the garden. Secondly, in the laboratory we determined the ^{226}Ra content and the specific radon exhalation rates of the collected soil samples from two points in the garden at 20 cm below the surface (Nagy et al., 2009) (Tab. 1).

4.2. Measurements after the mitigation process

Based on our previous results (Nagy et al., 2009; Nagy, 2009) especially those of the indoor radon concentration measurements (Table 1, Fig. 3), a mitigation was necessary. This is consistent with the fact that local sandstones (Kővágószőlős Sandstone Formation) affected by U-mineralization were used as building stones for the house. To determine the mitigation efficiency, indoor radon concentration in the cellar was monitored by AlphaGUARD radon detector for one month (March, 2009) and passive CR-39 etch track detector was also used after the mitigation.

Note that this second measurement was performed exactly one year after the pre-mitigation measurement (Fig. 3) expecting similar meteorological conditions during the two experimental periods. The time dependence of the hourly average radon concentrations (measured by AlphaGUARD) are shown on figure 4.

Table 1. Results of pre-mitigation measurements based on the data of Nagy et al., 2009; and Nagy, 2009.

Measured Parameter	unit	mean	min	max
Indoor ^{222}Rn concentration in cellar (3 monthly measurement in Summer, 2004 using etch track detector) (Várhegyi personal communication, 2009)	Bq/m^3	1950	-	-
Indoor ^{222}Rn concentration in cellar (one monthly measurement in Spring, 2008, using AlphaGUARD radon monitor) (Nagy et al., 2009)	Bq/m^3	1480	85	5790
Indoor ^{222}Rn concentration in bedroom (3 monthly measurement in Summer, 2004, using etch track detector) (Várhegyi personal communication)	Bq/m^3	450	-	-
Indoor ^{222}Rn concentration in bedroom (3 monthly measurement in Winter, 2003-2004, using etch track detector) (Várhegyi personal communication)	Bq/m^3	647	-	-
Soil gas ^{222}Rn concentration (average value of 13 different measurements in 13 points within the house garden) (Nagy, 2009)	kBq/m^3	42	15	118
In situ soil ^{222}Rn exhalation (average value of 13 different measurements in 13 points within the house garden) (Nagy, 2009)	mBq/m^{2}s	104	23	194
Outdoor gamma dose rate (average value of 13 different measurements in 13 points within the house garden) (Nagy, 2009)	nGy/h	99	83	113
Soil ^{226}Ra content (average from two soil samples) (Nagy, 2009)	Bq/kg	65	-	-
Soil specific ^{222}Rn exhalation (average from two soil samples) (Nagy, 2009)	Bq/kg	12	-	-

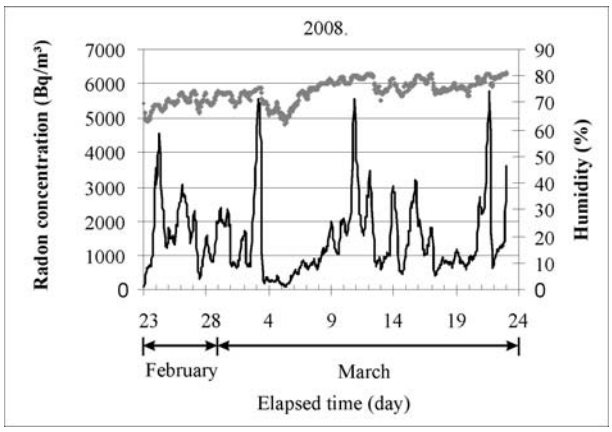

Figure 3. One month long monitoring (February 23–March 23, 2008) of the radon concentration in the mitigated cellar before the mitigation process: (—) Radon concentration (Bq/m^3); (♦) Relative humidity (%).

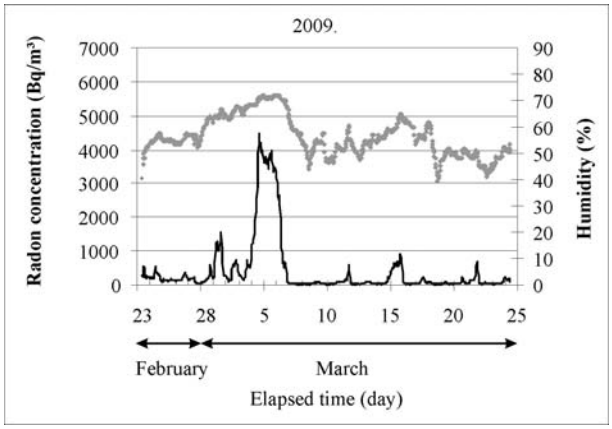

Figure 4. One month long monitoring (February 23–March 23, 2009) of the radon concentration in the mitigated cellar after the mitigation process: (—) Radon concentration (Bq/m^3); (♦) Relative humidity (%).

The monthly average indoor radon concentrations were 1480 Bq/m^3 before, and 420 Bq/m^3 after mitigation measured by AlphaGUARD, and 363 Bq/m^3 by etch track detector one year after mitigation. It means a value of 0.29 ARRF (for the calculation, we used the data measured by AlphaGUARD). The data were collected in March, however, if it is assumed that the efficiency is constant over the year the value of 0.29 ARRF can be interpreted as an overall efficiency value. This is a highly promising result from a passive method compared with results of SARAH project in Austria (Maringer et al., 2001), where two kinds of passive radon mitigation system were used and the ARRF was 0.5 (subfloor pressure decreasing system without operation of the suction fan) and 0.6 (passive subfloor ventilation system), respectively. According to a radon remediation of a two-storey dwelling by active sub-slab depressurization in the U.K. where the ARRF ($R_{A/B}$) was 0.18 in the downstairs and 0.33 in the upstairs (Allison et al., 2008), our results are fully acceptable.

Radon–barrier membranes were used to a during-construction radon remediation in U.K., at a designated 'radon affected area'. The experienced radon reduction can be characterized by a bimodal behavior, with approximately equal maxima at values of 0.3 and 0.5 (ARRF). A possible explanation for this result is that the data reflects the existence of two distributions of mitigation efficiency. One derives from well-installed membranes, exhibiting an intrinsic mitigation factor around 0.3, and the other value represents a defective membrane population, affording only 0.5 mitigation (Groves-Kirkby et al., 2006). The 0.3 ARRF with radon-barrier membrane is closely comparable to our results.

After efficient mitigation there were some time periods when high radon concentration occurred (Fig. 4). However, these peaks are limited only to some days. On other days, during the measurements shown in the diagram (Fig. 4) had quite low radon content levels. The amplitudes of the peaks are above 600 Bq/m^3 which increases the value of average radon concentration up to 420 Bq/m^3. However the average radon concentration without these days goes down to less than 150 Bq/m^3.

To understand the reason for the effects which resulted in the elevated radon concentration values, we reconsidered the location of the pipe system that releases the radon from the mitigated room (i.e., the cellar). The outside part of the pipe opens on the eastern wall of the house and has a southern direction with an inflection. We examined the wind direction during the measurement period (February 23, 2009 – March 23, 2009) using Ogimet (Weather Information Service) web based meteorological database. Data from meteorological station of Pécs-Pogány (WMO index 12942), about 20 km from our study site, had been chosen. During the period of February 23, 2009 – March 23, 2009 the one-day averaged wind directions were evaluated. We looked for the directions of SSE, SE and ESE since these directions can increase pressure on the releasing pipe. There were three days, 4, 5, and 6 of March, when wide and large peaks of the radon concentration curve occurred (Fig 4). We conclude that accidental coincidence of the wind direction and the direction of the outlet of the pipe caused increased radon concentration in the mitigated air. In this region the most probable wind direction is N or NW, which is true almost always during the year at the study site. That is why the pipe was installed on the east side of the building and the mitigation was so efficient. It is obvious that efficiency of mitigation can be improved by use a cap on the top of the pipe, which can preclude the blow of

wind into the pipe and, thus, retain radon gas in the pipe system when the wind direction is S-SW. In a place with more severe weather conditions the passive system would work with lower ARRF.

The third month long monitoring covered the second part of September and the first part of October in 2008, which was also a post-mitigation measurement. The radon concentration and the humidity values averaged for 1 hour data collection time as figure 5 shows.

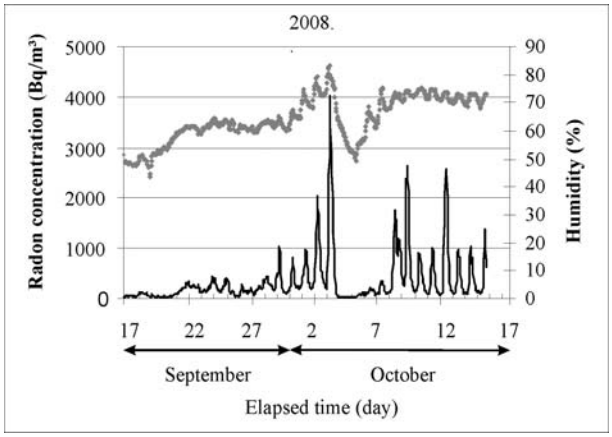

Figure 5. The time dependence of radon concentration (—) and relative humidity (♦) during 30 days in September-October (September 17– October 15, 2008) after the mitigation process.

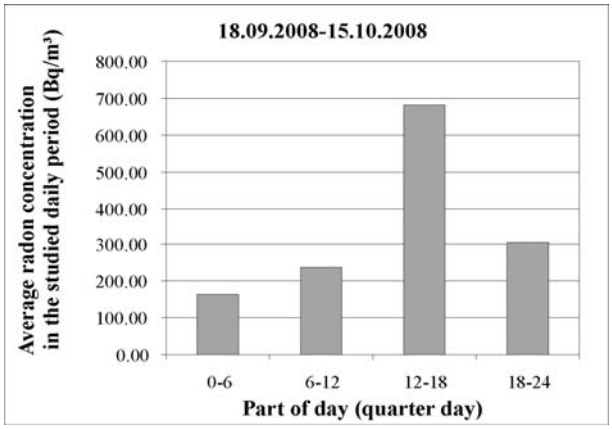

Figure 6. The daily dependence of the radon concentration during the Autumn measurement in 2008, after the mitigation process. Daily periodicity of the radon concentration for all days during the measurement is shown. The standard deviation of the values in the studied daily period: first quarter (0-6) 138, second quarter (6-12) 163, third quarter (12-18) 772, fourth quarter (18-24) 284.

The average indoor radon concentration was 344 Bq/m^3 which is below the recommended level of the EU (90/143/Euratom). However, the values are changing in a wide range between 16-4032 Bq/m^3 (Fig. 5). Variation of the radon concentration shows daily periodicity for more than half of the studied days. These peaks reach higher than 1000 Bq/m^3 radon level on 6 different days, and, on the other hand there are no peaks from 17th to 22nd of September and from 4th to 7th of October. Evaluating the properties of the peaks, for every one-day period can be divided into four quarters in a 6 hour unit: 0-6 h, 6-12 h, 12-18h, 18-24 h. It is clear that the highest radon concentration occurs always in the 3rd quarter, basically afternoon, between 12-18 hours (Fig. 6). Similar results have been found by Gäggeler et al. (1995) in Switzerland.

For the Summer period, typical diurnal variations with a peak in the afternoon are observed for ^{214}Pb, which is a radon decay product. Another diurnal variation of indoor radon concentration has been reported by Murty et al. (2010) in dwellings from Botswana. In this study the ^{222}Rn concentration was found to be high in the early morning hours and then the values decreased reaching a minimum concentration during the early afternoon hours. It is clear that variation of ^{222}Rn concentration shows a strong positive correlation with that of atmospheric pressure, humidity and temperature. Similar conclusion was summarized by Neves et al. (2009) during a Portuguese and Kolarz et al. (2009) in a Serbian study related to the daily periodicity in radon concentration. In the Portuguese study generally a daily maximum occurred in the morning, between 9 and 10 a.m., however the distribution was slightly bimodal, with some maxima occurring also at night. In this study only a significant correlation was observed between radon daily variations and outdoor temperature, although radon concentration shows also a modest relation to outdoor relative humidity, which is likely not a causal correlation because it arises via change of temperature.

In our study the daily periodicity shows different feature compared to results of Allison et al. (2008); Gäggeler et al. (1995); Murty et al. (2010) Neves et al. (2009); Paridaens et al. (2005) due to the opening direction of the exhaust pipe installed and the mitigation process. In a dwelling where soil gas radon advection has been suppressed, either by careful design and construction or by post construction remediation, it would not be expected that typical diurnal variability be exhibited (Allison et al. 2008). The afternoon peaks can be explained by differences in temperature between the outside and the indoor air. After 12 a.m. the temperature rises and becomes higher than the inside air. The air in the pipe becomes denser than the outside air and, therefore, the air circulation turns back. The colder and denser air cannot go out. In this case the releasing effect does not work, thus, the radon remains inside. In Summer this effect increases the ARRF, however the indoor levels of radon are generally low (Papaefthymiou et

al., 2003). Pattern of time dependence of radon concentration is similar to that of humidity (Fig. 4) as value of 0.56 correlation coefficient between these variables shows collected in the cellar during the one-month measurement. Similar correlations between the relative humidity of outdoor air and the radon progeny concentrations were reported by Baciu (Baciu et al., 2005) in Romania, where the correlation coefficient was 0.21 – 0.54. We also measured the relative humidity in the indoor air, however, due to the opening position of the exhaust pipe there is a link between the outdoor and indoor air. An explanation of such dependence is that a relatively high relative humidity increases the attachment rates of radon progeny to the aerosol particles as several papers stated (Raabe, 1968; El-Hussein et al., 2001).

5. CONCLUDING REMARKS

A test of a passive radon mitigation process was carried out in a cellar of an old brick house where the radon level was found higher (>1000 Bq/m^3) than the recommended EU values in previous surveys (nuclear track detector, AlphaGUARD and RAD7 radon monitor).

The applied mitigation system reduced the monthly radon level from 1480 Bq/m^3 to 420 Bq/m^3. The ARRF (average radon reduction factor) is 0.3, which is increasable by use a cap on the top of the pipe, which can preclude the blow of wind into the pipe and, thus, retain radon gas in the pipe system when the wind direction is S-SW.

The radon concentration showed diurnal variation, the values are higher in the afternoon and lower in the early morning. The afternoon peaks can be explained by difference in temperature between the outside and the indoor air. After 12 a.m. the temperature becomes higher than the inside air. The air in the pipe becomes denser than the outside air and, therefore, the air circulation turns back. The colder and denser air cannot go out. In this case the releasing effect does not work, thus, the radon remains inside.

The radon concentration and the relative humidity in the cellar during the one month post-mitigation measurement showed a weak positive correlation (a 0.56 correlation coefficient).

Based on our results, the used DELTA-TERRAXX and DELTA-PT sealer materials are applicable for radon mitigation.

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