Study of the dynamics and sources of the radon concentration in caves of Buda Hill

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Summary of the Ph.D. dissertation

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Introduction

Radon-222 is an inert radioactive isotope with a half-life of 3.8 days. Being the decay product of radium-226, it belongs to the uranium-238 series. As ^{226}Ra can be present in the environment, in soils, bedrocks and building materials in various activities radon is widely observed permanently from soils, rocks and building materials. When radon gas is inhaled densely ionizing alpha particles can interact with biological tissues in the lungs. The health effect of radon is most notably lung cancer (DARBY et al., 2005). Therefore, to study and measure the radon concentration is very important to understand its risk.

My major goal was to determine the time dependence of radon concentration in the Pálvölgy show cave to understand the exchange pattern of the cave air with the outdoor air based upon ^{222}Rn concentrations and to determine the factors that most affect the ^{222}Rn concentration in the cave air. For these during my Ph.D. research I carried out different physical measurements in the Pálvölgy Cave and in the Molnár János Cave located in Budapest (Hungary). The results deepen our knowledge on the nature of radon gas and provide new data on radon studies in caves.

Methodology

To define the source of the radon concentration, leakage water and clayish cave sediment samples were collected (from drilling and from the upper layer). The radon and thoron exhalation rate (by RAD 7 and radon accumulation chamber) and radioactive isotope content (^{226}Ra, ^{232}Th, ^{40}K, by high purity germanium detector) of clayish cave sediments and the composition of constituent (X-ray powder diffraction and scanning electron microscopes) were defined in laboratory. The radon concentration of leakage water samples was measured by liquid scintillation spectrometry.

To define the time dependence of radon concentration, the radon concentration constantly for one and a half years was measured using an AlphaGuard radon monitor with an integration time of 1 hour. This equipment is a portable ionization chamber permitting the continuous monitoring of radon concentration and other meteorological parameters such as indoor air temperature (°C), indoor pressure (mbar) and indoor relative humidity (%). The same meteorological parameters were measured simultaneously outside the cave by a FWS 20 meteorological station approximately six meters far from the cave entrance.

Results

The natural radioactivity in caves of Buda Hill, especially in the Pálvölgy Cave has been studied in details. Based on the results, several conclusions can be drawn, as follows:

(1). On the basis of the results of the specific ^{226}Ra, ^{232}Th and ^{40}K activity (^{226}Ra:~20-40 Bq/kg, ^{232}Th:~10-30 Bq/kg and ^{40}K:~100-400 Bq/kg), radon and thoron exhalation, and soil (clay) gas radon concentration measurements, the data show typical values for average soil. However the clayish cave sediments show higher values than the wall rocks of the caves (limestone, marl) (^{226}Ra:~10 Bq/kg, ^{232}Th:~2-5 Bq/kg and ^{40}K:~10-35 Bq/kg). Due to the elevated radon exhalation and ^{226}Ra content of clayish cave sediments, it is assumed that the source of the radon in the cave is the clayish cave sediment.

(2). A remarkable difference in the radon exhalation between the clayish cave sediments, formed on the marl and on the limestone, was observed. The radon exhalation rate of the clayish cave sediment, collected from the Buda Marl, shows the highest value. In accordance with the previous theses point, the radon concentration shows two times higher values in the section of the cave located in the Buda Marl than those in the Szépvölgy Limestone. These results suggest that the Buda Marl and the clayish cave sediments, derived from Buda Marl, are considered as the most likely radon sources.

(3). I observed a seasonal variation in radon concentration of leakage water in the Pálvölgy cave. The radon concentration of water samples is the lowest in winter and maximum values were measured in summer. There is no equilibrium between the radon concentration of water and air. The ratios are always higher (0.4-8) than the equilibrium value (~0.23-0.27). This means that the leakage water has higher radon content. However because of the negligible volume of the water related to the air the radon concentration of the water does not influence the radon concentration in the cave air.

(4). My study confirmed that the radon concentration inside the cave strongly depends on the outdoor air temperature. The correlation coefficient between the radon concentration and outdoor air temperature was 0.76 at the 2nd measurement point in the Pálvölgy Cave between 27.10.2009-22.12.2011. Similar values (0.6-0.75) were obtained by other scientists world-wide.

(5). I observed that the correlation between the radon concentration inside the cave and the outdoor air temperature is not constant for the whole year, in winter period this relation is stronger than in the summer period. The lowest correlation coefficient (monthly average values of the correlation coefficient between the hourly radon concentration and outdoor air temperature) was found in July-September period (0.3-0.4), the highest value (0.7) was observed in December.

(6). It can be stated that if the outdoor air temperature is lower than the cave air temperature, which in the Pálvölgy Cave is 10-11 °C and in the Molnár János cave is 21 °C, the air is flowing from outside into the cave, and the radon concentration is low in the cave . However, in reverse case, when the outdoor air temperature is higher than the cave air temperature, the fresh air cannot flow into the cave, because the air is flowing out from the cave, so the new air is coming through the cracks and gaps of the rocks inside the cave, and the radon concentration increases.

(7). Besides the seasonal variation of radon concentration a clear daily (24 hours) and a 12 hours periodicity was found. I confirm that the cause of the daily periodicity is the periodicity of the outdoor air temperature. The 12 hours periodicity could be associated with tidal effect, because it can influences the radon emanation of rocks and soils by the tidal deformation of the crust.

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