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Radon as a natural tracer for underwater cave exploration

Katalin Csondor [a], Anita Erőss [a, *], Ákos Horváth [b], Dénes Szieberth [c]

[a] Department of Physical and Applied Geology, Institute of Geography and Earth Sciences, Eötvös Loránd University, Pázmány Péter Sétány 1/c, 1117 Budapest, Hungary
[b] Department of Atomic Physics, Institute of Physics, Eötvös Loránd University, Pázmány Péter Sétány 1/a, 1117 Budapest, Hungary
[c] Department of Inorganic and Analytical Chemistry, Budapest University of Technology and Economics, Műegyetem Rakpart 3, 1111 Budapest, Hungary

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A B S T R A C T

The Molnár János cave is one of the largest hypogenic caves of the Buda Thermal Karst (Budapest, Hungary) and mainly characterized by water-filled passages. The major outflow point of the waters of the cave system is the Boltív spring, which feeds the artificial Malom Lake. Previous radon measurements in the cave system and in the spring established the highest radon concentration (71 BqL^{-1}) in the springwater. According to previous studies, the origin of radon was identified as iron-hydroxide containing biofilms, which form where there is mixing of cold and thermal waters, and these biofilms efficiently adsorb radium from the thermal water component. Since mixing of waters is responsible for the formation of the cave as well, these iron-hydroxide containing biofilms and the consequent high radon concentrations mark the active cave forming zones. Based on previous radon measurements, it is supposed that the active mixing and cave forming zone has to be close to the spring, since the highest radon concentration was measured there. Therefore radon mapping was carried out with the help of divers in order to get a spatial distribution of radon in the cave passages closest to the spring. Based on our measurements, the highest radon activity concentration (84 BqL^{-1}) was found in the springwater. Based on the distribution of radon activity concentrations, direct connection was established between the spring and the István-room of the cave, which was verified by an artificial tracer. However, the distribution of radon in the cave passages shows lower concentrations (18–46 BqL^{-1}) compared to the spring, therefore an additional deep inflow from hitherto unknown cave passages is assumed, from which waters with high radon content arrive to the spring. These passages are assumed to be in the active cave formation zone. This study proved that radon activity concentration distribution is a useful tool in underwater cave exploration.

1. Introduction

1.1. Radon as a natural tracer

Radon is a radioactive, noble gas with atomic number 86. It has many isotopes, but only three of them are naturally occurring: ^{222}Rn, ^{220}Rn (thoron) and ^{219}Rn (actinon), with half lives of 3.82 days, 54.5 s and 3.9 s, respectively. Due to the very short half-life of thoron and actinon, ^{222}Rn is the most frequently-used environmental radon isotope, which is the daughter element of the ^{226}Ra in the ^{238}U decay series. In the text below radon always means ^{222}Rn.

Radon is widely used in air and in aquatic environments to study dynamic processes (Wilkening, 1990; Quindos Poncela et al., 2013). It is often used in caves (Hakl et al., 1997; Cigna, 2005) to study natural ventilation (e.g. Wilkening and Watkins, 1976; Fernández et al., 1986; Perrier et al., 2004) based on the concentration differences in the cave air and in the atmosphere. It is useful to investigate the recharge dynamics of karst aquifers where the high radon concentration periods indicates that the soil water or water having transited through the soil zone is rapidly transferred to the saturated zone (Savoy et al., 2011). Radon (in this case both ^{222}Rn and ^{220}Rn) is used as a tool to estimate probabilities for geophysical risk events such as earthquakes or volcanic activity. Radon anomalies prior to earthquakes have usually been observed in soil gas as well as in groundwater or in springs (Nevinsky et al., 2015; Oh and Kim, 2015).

As radon is naturally found in groundwater and has a short half-

* Corresponding author. Tel.: +36 1 381 2125; fax: +36 1 381 2130.
E-mail addresses: kacc91@gmail.com (K. Csondor), anita.eross@geology.elte.hu, anita.eross@geoleogy.elte.hu (A. Erőss), akos@ludens.elte.hu (Á. Horváth), denes.szieberth@mail.bme.hu (D. Szieberth).

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life, it is a useful time-tracer for hydrogeological systems with relatively short flow distances and/or high flow velocities, e.g. for karst aquifers. The transit time of such flow systems is comparable with the half-life of radon (Eisenlohr and Surbeck, 1995). Radon is also an excellent tracer of interaction of groundwaters and surface waters because of the concentration differences in these reservoirs. It has been used both to assess infiltration of surface waters into aquifers (Hoehn and von Gunten, 1989; Hamada and Komae, 1998), and as a tracer of groundwater discharge into surface waters such as streams, lakes or even the ocean (Cook et al., 2006; Burnett et al., 2001, 2003, 2010; Swarzenski, 2007).

Together with other members of the ^{238}U decay chain (^{226}Ra, $^{234+238}$U), radon can be used to characterize different order flow systems and mixing processes based on the different geochemical behavior (Hoehn, 1998; Gainon et al., 2007; Erőss et al., 2012b, 2015; Cozma et al., 2016). Here we present a new application of radon in underwater cave exploration and in characterization of flow directions.

1.2. Study area and previous measurements

The capital city of Hungary, Budapest, has a unique karst system, the so-called Buda Thermal Karst (BTK), which was shaped mainly by the discharging thermal waters. These thermal waters established the famous bath culture of the city, as well as formed the hypogenic cave systems of the area. The Molnár János cave (MJ cave) is one of the largest as well as the youngest member of hypogenic caves in the BTK and is mainly characterized by water-filled passages (Kalinovits, 2006; Leél-Őssy and Surányi, 2003; Surányi et al., 2010).

The BTK is developed at the northeastern margin of the Transdanubian Range, in the regional discharge zone of its carbonate aquifer system (Fig. 1a) (Mádl-Szőnyi and Tóth, 2015). The MJ cave is located at one of the main discharge areas of the BTK and its passages are part of the active flow systems. The major outflow point of the waters passing through the cave system is the Boltív spring (BS), which feeds the artificial Malom Lake (ML) (Fig. 1b). This spring is one of the few natural springs of the BTK area where the dynamics of the aquifer system can be studied. Behind it the MJ cave is offering a unique possibility to investigate the flow system inside the aquifer.

Previous hydrogeological studies (Erőss et al., 2011; Erőss et al., 2012a,b; Ötvös et al., 2013) established that in the MJ cave area, mixing of waters with different temperatures and geochemical compositions takes place and this process is responsible for the formation of the cave. With the aid of radionuclides (^{222}Rn, ^{226}Ra, $^{234+238}$U) the mixing end members (meteoric: 12 °C, 775 mgL^{-1} total dissolved solids (TDS); thermal: 76.5 °C, 1440 mgL^{-1} TDS) were determined (Erőss et al., 2012b). As a result of mixing of these waters, iron-hydroxide containing biofilms form (Borsodi et al., 2012; Erőss, 2010; Erőss et al., 2012b; Mádl-Szőnyi and Erőss, 2013), and these efficiently adsorb radium from the thermal

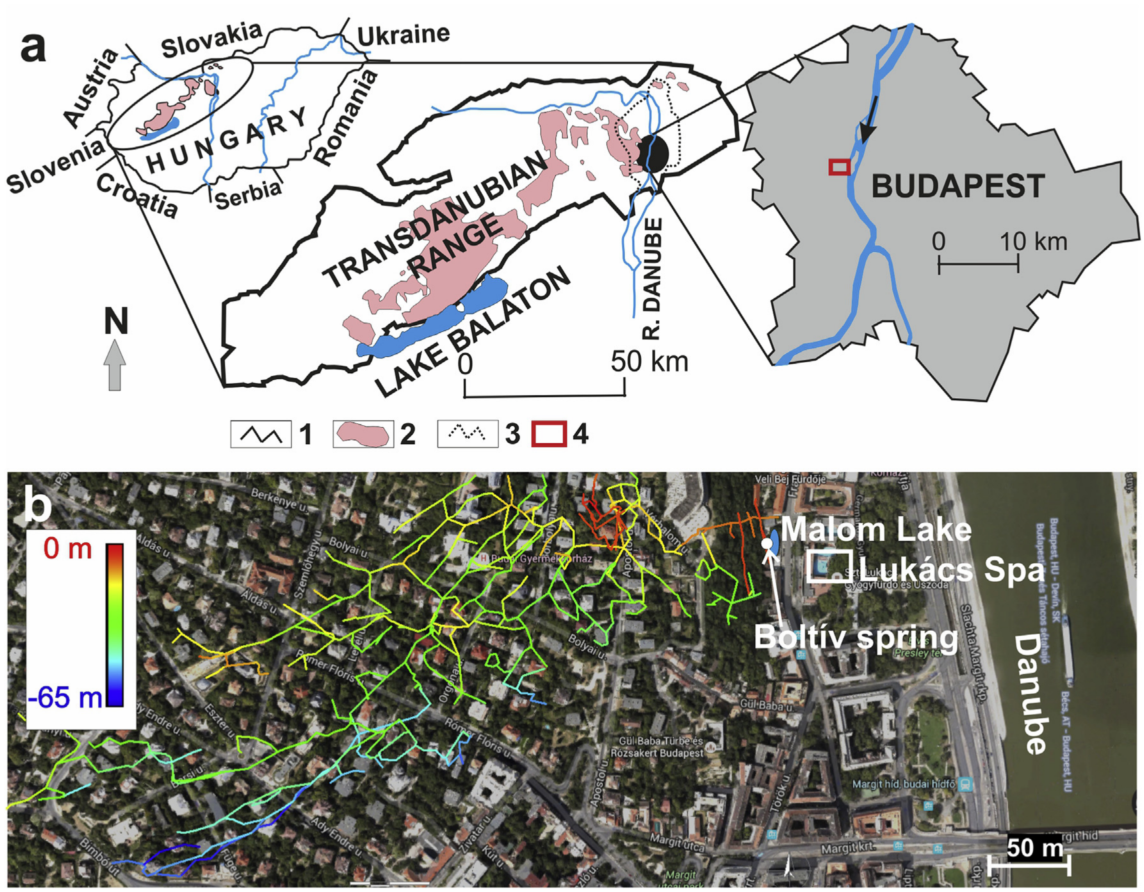

Fig. 1. a) Location of the Buda Thermal Karst in the Transdanubian Range and the study area in Budapest. Legend: 1: Subsurface boundary of Mesozoic carbonates, 2: Uncovered Mesozoic carbonates, 3: Buda Thermal Karst, 4: Study area. b) Location of the Molnár János cave, Boltív spring, Malom Lake and Lukács Spa in Budapest. The shallower and the deeper cave passages are marked by different color on the map (see the color scale of the depth on the left side of the figure), the red one is the shallowest region (−5 m below surface) and the blue is the deepest (−65 m below surface). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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