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Radon as a natural tracer for underwater cave exploration

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ABSTRACT

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.

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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-

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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

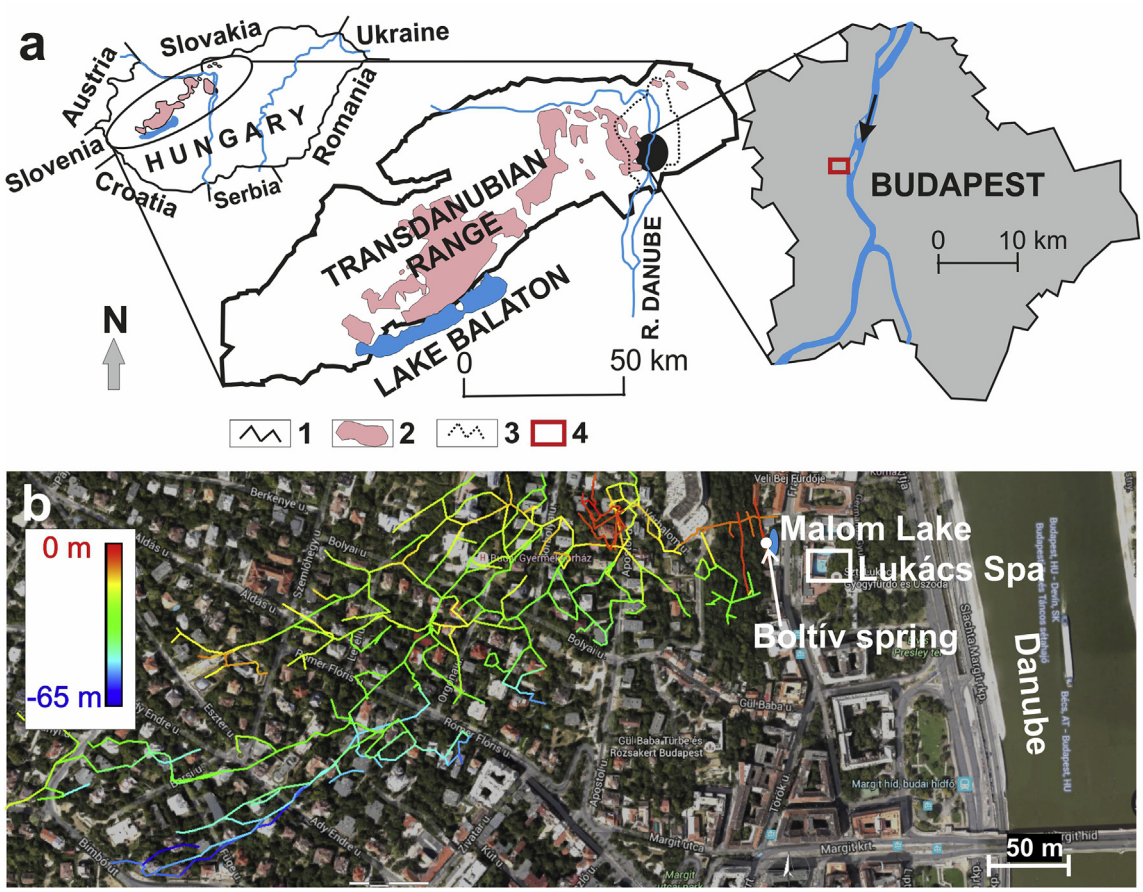

Fig. 1. a) Location of the Buda Thermal Karst in the Transdanubian Range and the study area in Budapest. Legend: 1: Subsurface boundary of Mesozoic carbonates, 2: Uncovered Mesozoic carbonates, 3: Buda Thermal Karst, 4: Study area. b) Location of the Molnár János cave, Boltív spring, Malom Lake and Lukács 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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water component and cause local radon anomalies. These iron-hydroxide containing biofilms and hence zones of high radon concentration mark the active cave forming, mixing zones. Based on previous radon measurements in the cave and in the spring (Baradács et al., 2002; Bodor et al., 2014; Erőss et al., 2012b; Restás-Göndör, 2015) it is assumed that the active mixing and cave forming zone has to be close to the spring, since the highest (71 BqL^{-1}, where the average is 44 BqL^{-1}) radon concentrations were measured there.

1.3. Objectives

The aim of the present study was to use radon as a natural tracer to locate active cave forming zones. To achieve this, underwater radon activity concentration mapping was carried out involving that part of the cave which is closest to the spring (Fig. 2). In the course of the study, active connections i.e. the existence of flow paths was suggested to match the measured radon concentrations, and these were verified using an artificial tracer.

1.4. Methods and experimental

The water samples were collected during November and December 2015 from the MJ cave, Boltív spring, Malom Lake and Lukács Spa (Fig. 1b). During 4 sampling campaigns, 42 water samples from 20 sampling sites were collected (Fig. 2). Thirteen sites were located in the MJ cave, 4 points in the spring (BS) at different positions (at the two edges and in the middle of the fracture of the spring, at shallow (1–2 m) depths and one deeper (5–6 m)), 2 sites in the lake (ML) and 1 point in the Lukács spa (LS). The latter was included to characterize the radon content of the waters which are taken by the spa through a pipe directly from the cave. The aim of the 4 sampling campaigns was to evaluate the stability of the parameters, since certain sampling sites could be reached only once during the study because of technical difficulties.

For the radon measurements the water samples were collected in the underwater cave with the help of divers, using syringes (10 mL). In the spring, bailer was used for sampling. The samples were injected into special 23 mL glass vials after the divers emerged from the cave at the surface station. These vials were prefilled with 10 mL Opti-Fluor O liquid scintillation cocktail. After the sample injection they were closed by parafilm in order to be air-tight. The radon activity was measured using a liquid scintillation TRICARB 1000 TR instrument in the laboratory under stable conditions (25 °C air temperature). The measurement protocol is included calibration by using known concentration $RaCl_2$ solution.

During sampling additional water samples were collected into 0.25 L polypropylene bottles for electrical conductivity measurements. The electrical conductivity was measured in μScm^{-1} by using a WTW multi 3430 SET G instrument (reference temperature 25 °C, error 1%) after the divers emerged from the cave at the surface station. This parameter reflects the dissolved solid content of the waters (Freeze and Cherry, 1979).

For the verification of the connection between the István-room of the cave and the spring, a NaCl solution was used. As a first step, the chloride concentrations of water samples collected from the István-room (MJ3) and from the spring (BS) were determined by titration. The necessary concentration of the NaCl solution for the artificial tracing was calculated on the basis of straight-line distance of the István-room and the spring using the cave map (Kalinovits and Kollár, 1984). The goal was to raise the dissolved solid concentration of the water in the spring to be detectable by electrical conductivity measurements. Based on the polygon map of the cave and on-site observations maximum 50 m long and 0.2 m wide passage was assumed, filled by 10 m^3 water. Based on this, 2 kg NaCl were dissolved in 7.5 L water and injected at the MJ3 site. Dataqua DA-DTK device was used for the continuous recording of the electrical conductivity (reference temperature 20 °C, error 1.5%) in the spring.

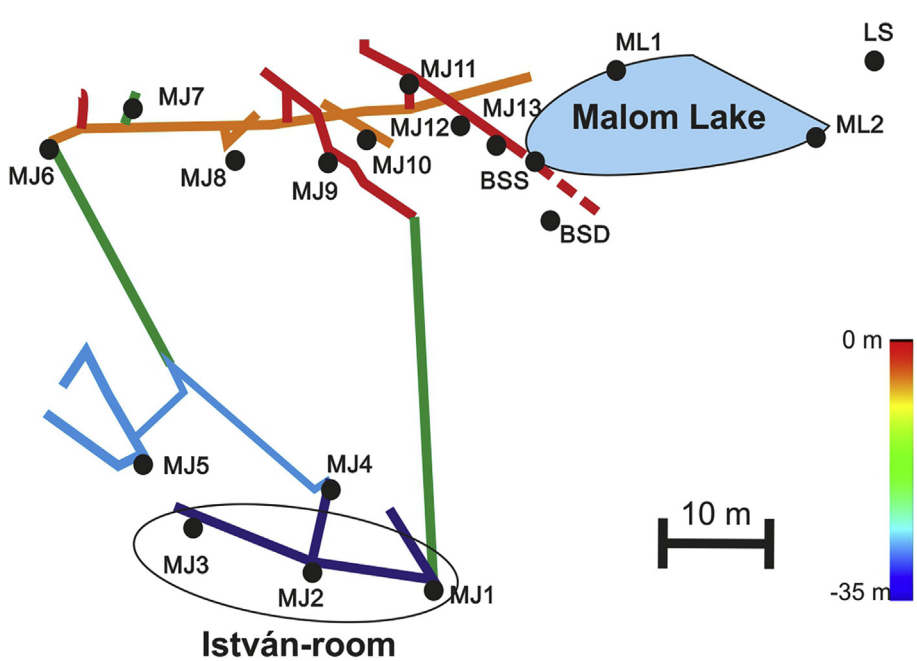

Fig. 2. Location of the sampling points. Legend: MJ: Molnár János cave, BSS: Boltív spring shallow part (1–2 m depth), BSD: Boltív spring deep part (5–6 m depth), ML: Malom Lake, LS: Lukács Spa. The shallower and the deeper cave passages are marked by different color on the map (see the color scale of the depth on the right side of the figure), the red one is the shallowest region (−5 m below surface) and the blue is the deepest (−35 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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2. Results

Table 1 summarizes the results of the 4 sampling campaigns. Where it was technically possible, repeated samples were taken in order to evaluate the stability of the parameters. However, there were some sampling sites which were sampled only once due to technical difficulties. To evaluate the representativeness of the results from those sites, the parameter stabilities of the other sites were used (Table 2). The low standard deviation values (0.0018–0.1417) allowed the comparison of sites measured at different times.

The average of electrical conductivity values (Table 2) varied between 970 and 1054 μScm^{-1}. The higher values (above 1000 μScm^{-1}) were measured in the shallower part of the cave (MJ6, MJ8, MJ9, MJ10, MJ11, MJ12, MJ13), in the Malom Lake (ML1, ML2) and in the Lukács Spa (LS), where the water arrives through a pipeline from the cave. The end of the pipeline is situated close to spring, between BSS and MJ13.

The lower values (below 1000 μScm^{-1}) were measured in the deeper cave passages (MJ1, MJ2, MJ3, MJ4, MJ5, MJ7) and at deeper zone in the Boltív spring's fracture (BSD) (Fig. 3a).

On the contrary, higher (>30 BqL^{-1}) radon activity concentrations were measured in the deeper region of the cave (MJ1, MJ2, MJ3, MJ4, MJ5, MJ7), in the Boltív spring (both in the shallow and deep part), in the Malom Lake (ML1, ML2) and in the Lukács Spa (LS). Lower values (around 20 BqL^{-1}) occurred in the shallower part of the cave (MJ6, MJ8, MJ9, MJ10, MJ11, MJ12, MJ13) (Fig. 3b). The highest concentration was measured in the Boltív spring deep region (BSD) at 84 BqL^{-1}. However, the lowest (18 BqL^{-1}) radon activity concentration was measured in close vicinity of the Boltív spring at MJ13.

3. Discussion

Previous studies established that active mixing takes place in the MJ cave area (Erőss et al., 2011; Erőss et al., 2012a,b). The inferred mixing end members have different temperatures and dissolved solid concentrations, thus they differ in density as well. The large passages of the cave (several meters in diameter) allow free convection of waters, i.e. the warmer waters (about 27 °C) can be found at shallower depth and colder waters (21–17 °C) dominate at depth. This is reflected in the electrical conductivity distribution, since higher values (reflecting higher dissolved solid content, which means greater proportion of the thermal water component) are characteristic in the shallower cave passages. However, this difference was more pronounced in the distribution of radon content.

Higher (>30 BqL^{-1}) radon concentration was measured in the deeper part of the cave (MJ1-3), where around the MJ3 site divers reported the existence of iron-hydroxide-biofilms as a potential source of radon. However, the highest activity concentrations were measured deep in the fracture of the spring (BSD), and also in

Table 1
Results of the radon activity concentration and electrical conductivity measurements of the four sampling campaigns.

Sample ID	Date	Electrical conductivity [μScm^{-1}]	Error [μScm^{-1}]	Rn^{222} [BqL^{-1}]	Uncertainty [BqL^{-1}]
BSS1	11.18.2015	995	10	58	4
BSS2	11.18.2015	994	10	49	4
BSS3	11.18.2015	988	10	57	4
MJ1	11.18.2015	970	10	32	3
MJ2	11.18.2015	969	10	37	3
MJ3	11.18.2015	973	10	45	4
MJ4	11.18.2015	965	10	34	3
MJ5	11.18.2015	967	10	36	3
MJ6	11.18.2015	1016	10	22	3
MJ7	11.18.2015	968	10	31	3
MJ8	11.18.2015	1015	10	22	3
MJ10	11.18.2015	1014	10	22	2
MJ11	11.18.2015	1011	10	25	3
MJ12	11.18.2015	1012	10	22	3
BSS1-2	12.4.2015	988	10	57	4
BSS2-2	12.4.2015	1002	10	40	3
MJ1-2	12.4.2015	980	10	32	3
MJ2-2	12.4.2015	974	10	–	–
MJ3-2	12.4.2015	973	10	44	4
MJ4-2	12.4.2015	971	10	32	3
MJ5-2	12.4.2015	973	10	36	3
MJ9-2	12.4.2015	1020	10	20	2
MJ11-2	12.4.2015	1118	11	19	2
MJ12-2	12.4.2015	1028	10	19	2
MJ13	12.4.2015	1022	10	18	2
ML1	12.9.2015	1010	10	52	4
MJ1-3	12.9.2015	982	10	37	3
MJ2-3	12.9.2015	977	10	34	3
MJ3-3	12.9.2015	979	10	50	4
MJ4-3	12.9.2015	973	10	35	3
MJ9-3	12.9.2015	1026	10	24	3
MJ11-3	12.9.2015	1032	10	22	2
MJ12-3	12.9.2015	973	10	20	2
BSD-1	12.9.2015	981	10	84	5
BSD-2	12.9.2015	981	10	71	5
BSD-3	12.9.2015	981	10	78	5
LS	12.9.2015	1020	10	38	3
ML2	12.16.2015	1015	10	42	4
ML1-4	12.16.2015	1020	10	59	4
BSD-4	12.16.2015	985	10	80	5
BSD-5	12.16.2015	983	10	81	5

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Table 2
Descriptive statistics of electrical conductivity and radon activity concentrations from the sampling sites.

	EC. Average	EC. Relative standard deviation	EC. Standard deviation	EC. Min.	EC. Max.	EC. Number of samples	Rn^{222} Average	Rn^{222} Relative standard deviation	Rn^{222} Standard deviation	Rn^{222} Min.	Rn^{222} Max.	Rn^{222} Number of samples
BSS1	992	0.0050	4.9	988	995	2	58	0.019	1.09	57	58	2
BSS2	998	0.0057	5.7	994	1002	2	45	0.136	6.10	40	49	2
BSS3	988	–	–	988	988	1	57	–	–	57	57	1
MJ1	977	0.0066	6.4	970	982	3	33	0.088	2.94	32	37	3
MJ2	973	0.0042	4.0	969	977	3	36	0.061	2.17	34	37	3
MJ3	975	0.0036	3.5	973	979	3	46	0.078	3.62	44	50	3
MJ4	970	0.0043	4.2	965	973	3	34	0.031	1.05	32	35	3
MJ5	970	0.0044	4.2	967	973	2	36	0.003	0.10	36	36	2
MJ6	1016	–	–	1016	1016	1	25	–	–	25	25	1
MJ7	968	–	–	968	968	1	24	–	–	24	24	1
MJ8	1015	–	–	1015	1015	1	25	–	–	25	25	1
MJ9	1023	0.0041	4.2	1020	1026	2	22	0.142	3.10	20	24	2
MJ10	1014	–	–	1014	1014	1	25	–	–	25	25	1
MJ11	1054	0.0538	56.7	1011	1118	3	22	0.128	2.78	19	25	3
MJ12	1020	0.0111	11.3	1012	1028	2	20	0.119	2.42	19	22	2
MJ13	1019	–	–	1019	1019	1	18	–	–	18	18	1
ML1	1010	–	–	1010	1010	1	55	0.085	4.73	52	59	2
ML2	1007	–	–	1007	1007	1	59	–	–	59	59	1
BSD	982	0.0018	1.8	981	985	5	79	0.059	4.68	71	84	5
LS	1020	–	–	1020	1020	1	38	–	–	38	38	1

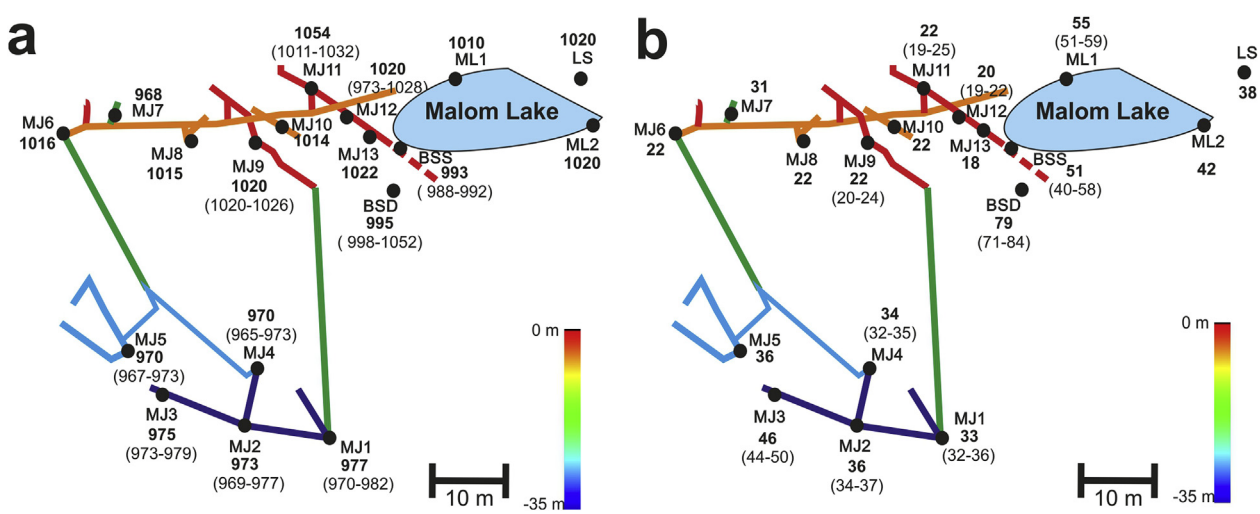

Fig. 3. a) Distribution of the electrical conductivity values (μScm^{-1}) in the MJ cave: average in bold, minimum and maximum in brackets. b) Distribution of the radon activity concentrations in the MJ cave ($Bq\ L^{-1}$): average in bold, minimum and maximum in brackets. The shallower and the deeper cave passages are marked by different color on the map (see the color scale of the depth on the right side of the figure), the red one is the shallowest region (−5 m below surface) and the blue is the deepest (−35 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.)

shallower zone of the springwater (BSS) and in the lake (ML). The lower values of the BSS and ML samples might be explained by degassing.

Based on the cave map connection (i.e. active flow) between the shallower part of the cave (MJ6 to MJ13) and the Boltív spring seems to be obvious (Fig. 2). However, both the electrical conductivity values and the radon activity concentrations point out that the spring rather has connection to the deeper part of the cave (MJ 1 to MJ3, the so-called István-room), i.e. the main portion of the spring water originates from the deeper part of the cave (Fig. 4). On the other hand, there is no mapped cave passage between the István-room and the Boltív spring.

To investigate this supposed connection between the Boltív spring and the István-room artificial tracer (NaCl solution) was injected in the István-room (MJ3) with help of divers. The original chloride concentration of the cave waters at MJ3 and in the spring (BSS) were similar (46 mgL^{-1}). The appearance of the salty water in the spring was detected by an electrical conductivity measuring instrument, which recorded that the values started to increase half an hour after the injection and increased from 855 to 1052 μScm^{-1} and decreased again. It suggests that a passage with active water flow exists between the István-room and the Boltív spring.

However, the distribution of radon in the cave passages shows lower concentrations (18–46 BqL^{-1}) compared to the spring, therefore an addition deep inflow from a hitherto unknown cave passages is assumed (Fig. 4), from which waters with high radon content arrive to the spring. These passages are assumed to be in the active cave formation zone. The existence of the deep inflow is also supported by the flow pattern observed by divers: the water arriving in the cave at MJ6 is in part diverted towards MJ4-MJ1 instead of following the obvious path MJ7-MJ13. This flow direction could be explained by a vertical upwelling near MJ1.

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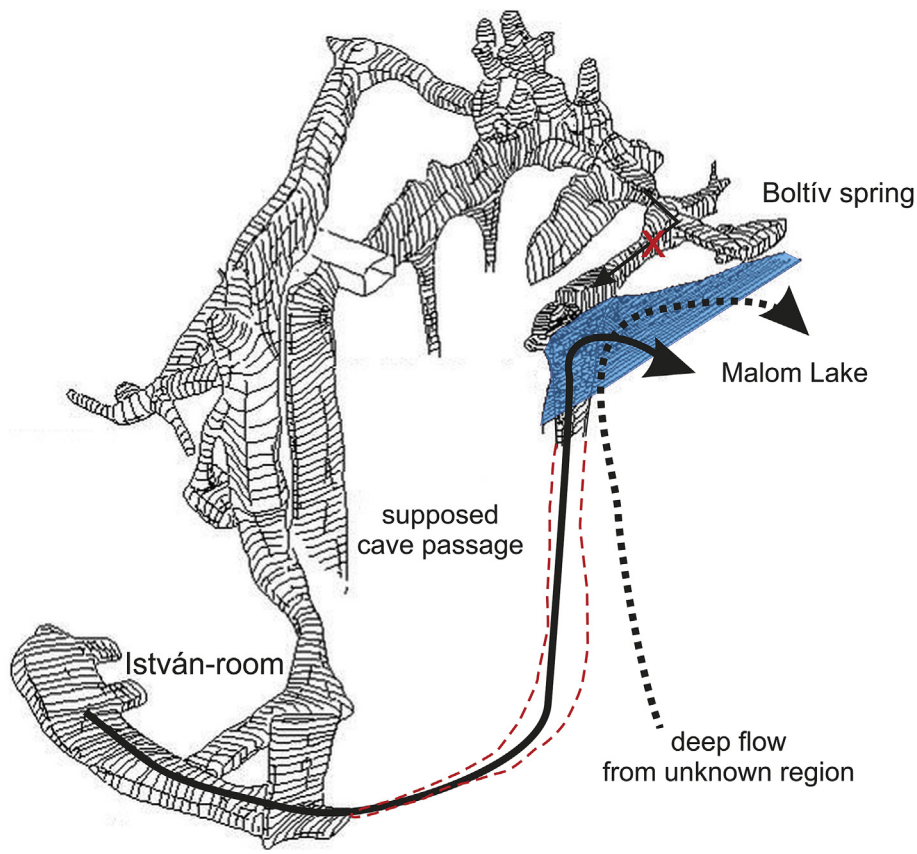

Fig. 4. The 3D model of the MJ cave closest to the Boltív spring (Kalinovits and Kollár, 1984). It shows the supposed passageway based on the artificial tracer test, furthermore illustrates the water components of the springwater suggested by the results of this study.

4. Conclusions

In this study radon mapping was carried out in the Molnár János underwater cave and its outflow point, the Boltív spring of the Buda Thermal Karst system. As the passages of the cave are part of the flow system, it provides a unique research laboratory to investigate the aquifer system and its dynamics. This study firstly used the radon activity concentration distribution for underwater cave exploration.

With the help of radon mapping, it can be concluded that one part of the Boltív spring's discharge is coming from a hitherto unknown region of the cave, which might be in the course of active formation. Moreover, based on the radon activity concentration distribution it can be established that the spring receives water rather from the deeper part of the cave (region of István-room) and seemingly has no connection with the shallower part. An active flow path was inferred by an artificial tracer (NaCl) between the István-room and the spring.

We showed that radon is a useful natural tracer in underwater caves, because differences are more pronounced compared to other chemical parameters such as electrical conductivity. In case of recently forming hypogenic caves radon concentration anomalies can assign the active mixing zones of cold and thermal waters, i.e. the ongoing cave formation. Moreover, active flow paths, underwater connections can be established using its concentration distribution which can be verified by artificial tracers.

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