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# Radionuclides in the waters of the Bükk region, Hungary

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**Abstract** The aim of the present study is to characterize the distribution of the $^{238}U$ decay series radionuclides, focusing on uranium, radium, in the cold, lukewarm and thermal karst waters in the Bükk region. This is a novel approach to characterize fluids and understand their mixing in regional discharge zones of carbonate aquifers. Based on the measurements, simple flow system, without mixing, is supposed, and the elevated radium content might be connected to hydrocarbon reservoir fluids. The results may support the improvement of protection strategies of springs and wells involved in water supply.

**Keywords** Radium · Uranium · Groundwater · Karst · Thermal water

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## Introduction

Natural radionuclides of the $^{238}U$ decay series, uranium, radium and radon are ubiquitous in groundwater [1]. Besides the dosimetric importance of the content of these isotopes in subsurface water [2–6] they are efficient natural tracers of groundwaters and their mixing processes [7–11], based on their different geochemical behaviour: radium is mobile in reducing, uranium is in oxidizing conditions. This causes fractionation along flow paths.

This is a novel approach to characterize fluids and understand their mixing in regional discharge zones of carbonate aquifers, where different order flow systems convey waters with different temperature, composition and redox-state to the discharge zone [9]. Discharging waters of regional flow systems are characterized by elevated total dissolved solid content (TDS), temperature and by reducing conditions, and therefore with negligible uranium content, whereas local flow systems have lower TDS and temperature, represent oxidizing environments, and therefore their radium content is probably low. This method was demonstrated and successfully applied in case of the regional discharge area of the Buda Thermal Karst system in Budapest [9].

### The aims of the work

The Bükk region, in the northeastern part of Hungary (Fig. 1a), is one of the largest karst areas of the country, where the water supply relies mainly on karst water resources. Moreover, this area is famous for its thermal springs and wells, all around the foothills of the Bükk Mountains, which are used for both balneological and heating purposes. Therefore the protection and sustainable use of these resources is an important issue, which requires

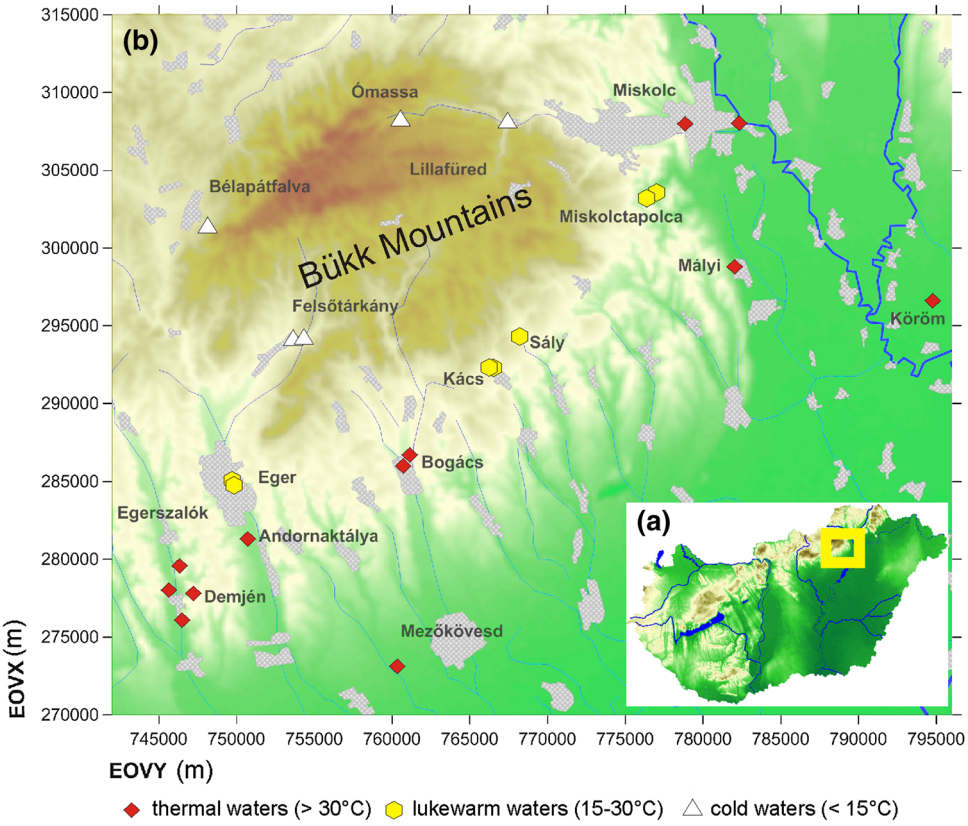



**Fig. 1** Location of sampling sites and temperature distribution of the sampled waters in the Bükk region, Hungary

good understanding on the hydrogeological functioning of the karst system.

The aim of the present study is to apply and test natural radionuclides as natural tracers on another large karst area with thermal water discharge after Budapest. During this study, water samples were collected and analysed for radionuclides and for the main anions and cations.

Here we show the distribution of uranium and radium in the cold, lukewarm and thermal karst waters in the Bükk region based on 31 samples (Fig. 1b). With the application of radionuclides as natural tracers the better understanding of the relationship of cold and thermal karst waters, i.e. the functioning of the karst system is expected.

## Methods

The water samples were collected during February and March 2014. In-situ physicochemical parameters (specific electrical conductivity, temperature, pH, Eh, dissolved $O_2$) were recorded during sampling on the field by WTW Multi 3410 multi-parameter measurement device. For radium and uranium measurements, 0.25 L PET bottles were used during sampling. The samples for radium and uranium have not been filtered nor acidified as it was intended to measure what is naturally in solution. The samples were kept cool until delivery to the laboratories.

Radium and uranium were measured with alpha spectrometry using Nucfilm discs, which are coated by selectively adsorbing thin films [12]. The Ra-discs made of selectively adsorbing $MnO_2$ thin films on polyamide substrates. Exposing a disc for 6 h to an untreated, stirred 100 mL water sample extracts typically more than 90 % of the radium. The U-discs are made of polycarbonate, which is covered with epoxy resin fixing a finely ground ion exchange resin (Diphonix®). For uranium measurements the water samples were acidified with concentrated (85 %) formic acid that guarantees that uranyl-$CO_2$ complexes are broken up. The U-discs were exposed to 100 mL acidified, stirred water for 24 h. After 24 h >90 % of the uranium activity present in the sample is adsorbed. The dried discs were measured with a solid-state alpha detector. Detection limit is 5 mBq $L^{-1}$ for $^{226}Ra$ and for $^{238}U + {}^{234}U$ as well, when measuring for 1 day. The measurement results are displayed in Table 1.

## Results and discussion

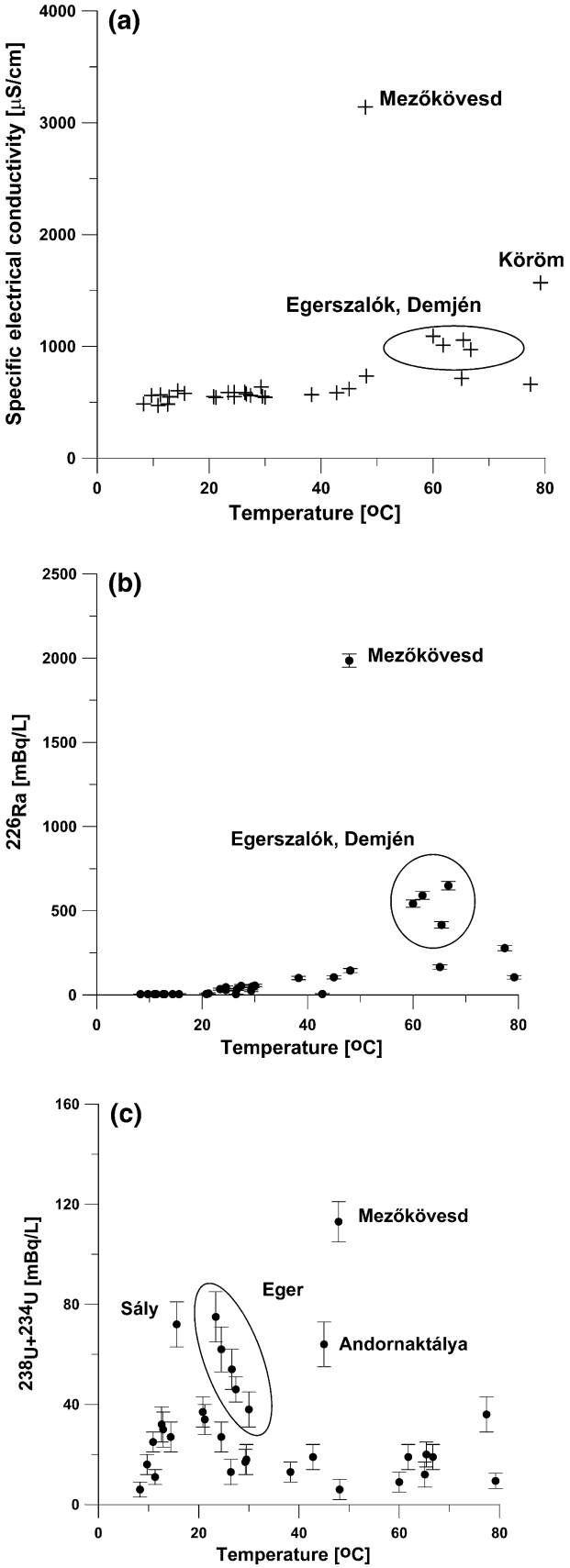
The specific electrical conductivity (EC) values provide information about the dissolved solid content of the waters,

**Table 1** The specific electrical conductivity (EC), temperature (T) (both recorded on the field), uranium and radium activity concentrations of the sampled waters (0 error means detection limit)

| Sample ID | Date of sampling | Location | EC ($\mu S\ cm^{-1}$) | T (°C) | $^{226}Ra$ ($mBq\ L^{-1}$) | Error ($mBq\ L^{-1}$) | $^{238}U + ^{234}U$ ($mBq\ L^{-1}$) | Error ($mBq\ L^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| KOR | 26.02.2014 | Köröm | 1,568 | 79.2 | 105 | 10 | 9 | 3 |
| FON | 26.02.2014 | Miskolc | 585 | 42.8 | 6 | 3 | 19 | 5 |
| TBF | 26.02.2014 | Miskolc-tapolca | 638 | 29.3 | 26 | 5 | 17 | 5 |
| TUJ | 26.02.2014 | Miskolc-tapolca | 566 | 11.3 | 5 | 0 | 11 | 3 |
| ANF | 26.02.2014 | Lillafüred | 561 | 9.7 | 5 | 0 | 16 | 4 |
| GAF | 26.02.2014 | Lillafüred | 485 | 8.3 | 5 | 0 | 6 | 3 |
| MZS1 | 14.03.2014 | Mezőkövesd | 3,140 | 47.9 | 1,985 | 39 | 113 | 8 |
| MALY | 14.03.2014 | Mályi | 660 | 77.4 | 278 | 15 | 36 | 7 |
| TSZ | 14.03.2014 | Miskolc-tapolca | 589 | 26.4 | 5 | 0 | 13 | 5 |
| ERZS | 14.03.2014 | Miskolc | 568 | 38.3 | 101 | 10 | 13 | 4 |
| DE1 | 19.03.2014 | Egerszalók | 1,057 | 65.4 | 416 | 19 | 20 | 5 |
| EGV3 | 19.03.2014 | Eger | 588 | 23.4 | 35 | 6 | 75 | 10 |
| EGV1 | 19.03.2014 | Eger | 574 | 26.6 | 34 | 6 | 54 | 8 |
| AT8 | 19.03.2014 | Andor-naktálya | 620 | 45 | 105 | 10 | 64 | 9 |
| B2 | 19.03.2014 | Felsőtárkány | 483 | 12.6 | 5 | 0 | 32 | 7 |
| EGV4 | 19.03.2014 | Eger | 560 | 27.4 | 54 | 8 | 46 | 5 |
| SZIK | 19.03.2014 | Felsőtárkány | 473 | 10.9 | 5 | 0 | 25 | 4 |
| BELT | 20.03.2014 | Bélapátfalva | 549 | 12.9 | 5 | 0 | 30 | 7 |
| BAR | 20.03.2014 | Eger | 589 | 24.5 | 30 | 6 | 62 | 9 |
| TÜK | 20.03.2014 | Eger | 553 | 29.5 | 50 | 7 | 18 | 6 |
| TÖR | 20.03.2014 | Eger | 544 | 30 | 56 | 7 | 38 | 7 |
| ES I. | 20.03.2014 | Eger | 550 | 24.5 | 46 | 7 | 27 | 6 |
| DHE | 20.03.2014 | Demjén | 1,010 | 61.8 | 591 | 23 | 19 | 5 |
| EKB | 20.03.2014 | Demjén | 970 | 66.7 | 649 | 25 | 19 | 5 |
| BOG1 | 20.03.2014 | Bogács | 714 | 65.1 | 166 | 13 | 12 | 5 |
| CSEM | 20.03.2014 | Bogács | 735 | 48.1 | 145 | 12 | 6 | 4 |
| KEN | 20.03.2014 | Demjén | 1,091 | 60 | 542 | 22 | 9 | 4 |
| SÁLY | 25.03.2014 | Sály | 579 | 15.6 | 5 | 3 | 72 | 9 |
| KÁCS-NAGY | 26.03.2014 | Kács | 603 | 14.4 | 5 | 0 | 27 | 6 |
| KDISZ | 28.03.2014 | Kács | 550 | 20.8 | 6 | 4 | 37 | 6 |
| KTÜK | 30.03.2014 | Kács | 543 | 21.2 | 9 | 4 | 34 | 6 |

which reflects the transit time of groundwater and the rock-water interaction. The thermal well in Mezőkövesd has the highest EC (3,140 $\mu S\ cm^{-1}$) (Table 1). Otherwise, only the thermal wells in Egerszalók, Demjén and Köröm show EC above 1,000 $\mu S\ cm^{-1}$. Majority of the samples is characterized by EC below 800 $\mu S\ cm^{-1}$ in a wide temperature range (8–77 °C) (Fig. 2a). These lower EC values accompanied by higher temperature may indicate deep but relatively short flow path and homogeneous aquifer lithology, where considerable heat amount is collected but the transit time may not allow acquiring high dissolved solid content. This might be the consequence of the close vicinity of recharge and discharge areas and their considerable elevation difference (500–600 m).

Similarly to the EC, the highest values both for radium (1,985 $mBq\ L^{-1}$) and uranium (113 $mBq\ L^{-1}$) were measured in the Mezőkövesd thermal well (Fig. 2b, c). Regarding the radium content, majority of the samples is characterized by less than 200 $mBq\ L^{-1}$ activity concentrations and increasing radium content can be observed




◀**Fig. 2** EC (**a**), radium (**b**) and uranium (**c**) data plotted versus temperature

with increasing temperature. Thermal wells around Egerszalók and Demjén have higher values, between 400 and 700 mBq $L^{-1}$.

Since the thermal wells around Egerszalók, Demjén and in Mezőkövesd are related to hydrocarbon exploration and known hydrocarbon fields are located in these regions, the elevated radium values might be connected to hydrocarbons, as radium is commonly enriched in saline (high EC) formation fluids contained within or in the vicinity of hydrocarbon reservoirs [13, 14]. The higher EC values in these samples support this idea.

The uranium contents are low, usually below 80 mBq $L^{-1}$, majority of the data is even below 40 mBq $L^{-1}$. However, these values correspond to a wide temperature range (8–79 °C). The water supply wells and springs in Eger, Andornaktálya and Sály show uranium values between 40 and 80 mBq $L^{-1}$. The presence of uranium may indicate the presence of freshly infiltrated, oxygenated waters.

## Conclusions

These first results of a comprehensive sampling in the Bükk region characterize the distribution of radium and uranium in cold, lukewarm and thermal waters (springs and wells) of the area. According to the measured low EC values, which are accompanied by high temperature, simple flow system with deep but short flow paths and no interaction with other aquifer lithologies can be supposed in the area. This is reflected by the radium content of the waters as well: increasing temperature is accompanied by increasing radium content. Exceptions are those thermal wells, where hydrocarbon fields characterized by reservoir fluids with elevated dissolved solid content are in the vicinity. The strongly reducing environment explains the elevated radium content as well. There is no apparent mixing trend based on the radionuclides, however, further samplings and local evaluations are in progress. As the karst waters are important resources in the region, with the application of this novel method in case of carbonate aquifers, our results may contribute to the better understanding of the functioning of the karst system, which is fundamental for the sustainable use.

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## References


1. Hoehn E (1998) Radionuclides in groundwaters: contaminants and tracers. In: Herbert M, Kovar K (eds) Groundwater quality: remediation and protection. IAHS Publ. No. 250, pp 3–9
2. Somlai J, Horvath G, Kanyar B, Kovacs T, Loczi T, Nenyei A (2002) Concentration of $^{226}$Ra in hungarian bottled mineral waters and the committed effective dose due to intakes of them. J Environ Radioactiv 62:235–240
3. Kovács T, Bodrogi E, Dombovari P, Somlai J, Nemeth C, Capote A, Tarján S (2004) $^{238}$U, $^{226}$Ra, $^{210}$Po concentrations of bottled mineral waters in Hungary and their committed effective dose. Radiat Prot Dosim 108:175–181
4. Erden PE, Dirican A, Seferinoglu M, Yeltepe E, Sahin NK (2014) U-238, U-234 and Ra-226 concentrations in mineral waters and their contribution to the annual committed effective dose in Turkey. J Radioanal Nucl Chem 301:159–166
5. Kovács T, Bodrogi E, Somlai J, Jobbágy V, Patak G, Németh C (2003) Ra-226 and Rn-222 concentrations of spring waters in Balaton Upland of Hungary and the assessment of resulting doses. J Radioanal Nucl Chem 258:191–194
6. Alshamsi DM, Murad AA, Aldahan A, Hou X (2013) Uranium isotopes in carbonate aquifers of arid region setting. J Radioanal Nucl Chem 298:1899–1905
7. Gainon F, Surbeck H, Zwahlen F (2007) Natural radionuclides in groundwater as pollutants and as useful tracer. In: Bullen TD, Wang Y (eds) Water-rock interaction: proceedings of the 12th international symposium on water-rock interaction, Kunming, China, vol 1, 31 July–5 August 2007, Taylor and Francis, London, pp 735–738
8. Swarzenski PW (2007) U/Th series radionuclides as coastal groundwater tracers. Chem Rev 107:663–674
9. Erőss A, Mádl-Szőnyi J, Surbeck H, Horváth Á, Goldscheider N, Csoma AÉ (2012) Radionuclides as natural tracers for the characterization of fluids in regional discharge areas, Buda Thermal Karst, Hungary. J Hydrol 426–427:124–137
10. Grabowski P, Bem H (2012) Uranium isotopes as a tracer of groundwater transport studies. J Radioanal Nucl Chem 292:1043–1048
11. Chawla D, Jacob N, Mohokar HV (2014) Identification of groundwater discharge in Cuddalore coast, Tamil Nadu using radium isotopes. J Radioanal Nucl Chem 300:29–32
12. Surbeck H (2000) Alpha spectrometry sample preparation using selectively adsorbing thin films. Appl Rad Isot 53:97–100
13. Collins AG (1975) Geochemistry of oilfield waters. Elsevier, Amsterdam
14. Peterson RN, Viso RF, MacDonald IR, Joye SB (2013) On the utility of radium isotopes as tracers of hydrocarbon discharge. Mar Chem 156:98–107