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Long-term time series of environmental tracers reveal recharge and discharge conditions in shallow karst aquifers in Hungary and Slovakia

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ABSTRACT

Study region: Three karst regions in Hungary and Slovakia.
Study focus: Time series of environmental tracers both in the groundwater recharge and discharge provide important insights into how a karst water system works. The aim of the present work was to study the response of discharging karst waters to recharge using time series of environmental tracers, such as tritium, stable water isotopes, noble gases and SF_6.
New hydrological insights for the region: Our results show that sampling frequency plays a significant role in detecting short residence times (months): the seasonality of ^{18}O isotope composition of a selected karst spring indicates a 10 % contribution of recent water with a residence time of half a year. The contribution of an older component can be proven by the decrease of the tritium content of the waters, which compares to the decreasing trend of the tritium time series of the precipitation. However, the tritium concentrations are just slightly lower than those of the precipitation and the recharge water, hence the residence times of these shallow springs are supposed to be short. $^3H/^3He$ and SF_6 apparent ages confirm this to be between 0 and 10 years, with a median of 1.4 years. Our study demonstrates that long-term time series are preferable to provide better estimation to the age distribution than individual, short-term investigations.

1. Introduction

Karst aquifers are a valuable groundwater resource, as they provide drinking water to about 25 % of the world's population (van Beynen, 2011). Karst systems are a vulnerable environment, having high permeability due to their conduit flow component, which allows for very fast transport of contaminants (Foster et al., 2013) (Fig. 1). Karst waters are a very important source for drinking water, especially in countries possessing large karst areas, and lacking access to drinking water resources.

When studying the properties of shallow groundwater aquifers, the recharging as well as the discharging water has to be

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investigated (Katz et al., 1997; Clark and Fritz, 2013; Hartmann et al., 2013; Brenčič and Vreča, 2016). The relationship of groundwater in the initial and the final state gives us valuable insights into how the flow system works (Lee and Krothe, 2001; Barbieri et al., 2005; Long et al., 2008). In a shallow groundwater system, like an epigene karst aquifer, the change in the recharge water (from precipitation) is evidently reflected in the groundwater, and in the discharge water (i.e. springs) (Bonacci et al., 2016; Sun et al., 2016; Sappa et al., 2018; Stroj et al., 2020).

To improve the understanding of the groundwater flow systems in karstified carbonates, the basic concepts of hydraulic flow have to be considered which are in accordance with 'hydraulic continuity' (Tóth, 1995). The groundwater flow patterns are controlled largely by topography, and the groundwater movement has been conceptualized to occur within local, intermediate and regional flow systems (Tóth, 1963). In the case of small drainage basins (local to intermediate groundwater flow systems), water table configuration is considered as the main driving force of groundwater flow, and the recharge and discharge properties of karst aquifers are defined mainly by the geological structure (faulted, fractured zones). The spring represents an output of the groundwater flow system combining numerous theoretical flow pathways. Although the subsurface processes are less evident, numerical simulations are used to improve better understanding of the flow systems, and to calculate residence times, for instance, using particle-tracking (Wang et al., 2016) and or numerical simulation (Molson and Frind., 2012; Janos et al., 2018). Investigation of the transport and geochemical processes through the rock matrix is also an important step between the infiltration and the leakage (Szijártó et al., 2021).

In the last few decades, stable and radioactive isotopes have become powerful tools usually used as natural tracers in order to solve many problems in karst hydrogeology, like management of karst water resources (Criss et al., 2007), vulnerability of karst aquifers (Trček and Zojer, 2010), karst engineering, etc. (;Goldscheider, 2015; Vasić et al., 2020). Evaluation of different isotope composition can be applied to obtain information about the behaviour of the karst aquifer, such as the origin of the groundwater, the mixing properties and dynamics, the age distribution as well as aquifer processes occurring within the geological structure, response of recharge to discharge (Mook, 2006). Knowledge of the groundwater residence time is of great importance, since accurate age determinations can give us an insight to what extent the resource that we want to use is renewable, or whether it is a limited deep aquifer and a "geological" reserve, which does not have active interaction with different ambient water bodies including surface waters (Criss and Davisson, 1996; Criss et al., 2007)

Since karst aquifers are vulnerable to pollution, the response of the water quality to a potential surface contamination has to be taken into account. As a first approach, the mean residence time of the spring water can be estimated using environmental tracers. The mean residence time in a karstic aquifer represents an average age composed of the waters of different residence times contributing to the water discharge at the spring. To constrain the residence time of these waters, a higher sampling frequency within a longer time period is recommended to reveal both short and long-term patterns in the stable or radioactive isotope compositions and/or other chemical tracers of the springs. When using transit times based on environmental tracers in hydrological systems, long-term time series are recommended (Einsiedl, 2005; Stewart and Morgenstern, 2016).

The aim of this study was to examine the response of discharging karst waters using time series of environmental tracers in order to determine the age distribution of the karst springs. Additionally, we intended to study shallow karst aquifers representing local groundwater flow systems, focusing on the investigation of how the recent change of the tritium input function of the precipitation could be used in shallow karst aquifers. Springs in three, mainly autogenic karst areas were studied in central Europe (Hungary and Slovakia). Groundwater samples for environmental tracers were taken in different frequencies between 2008 and 2018, and the time

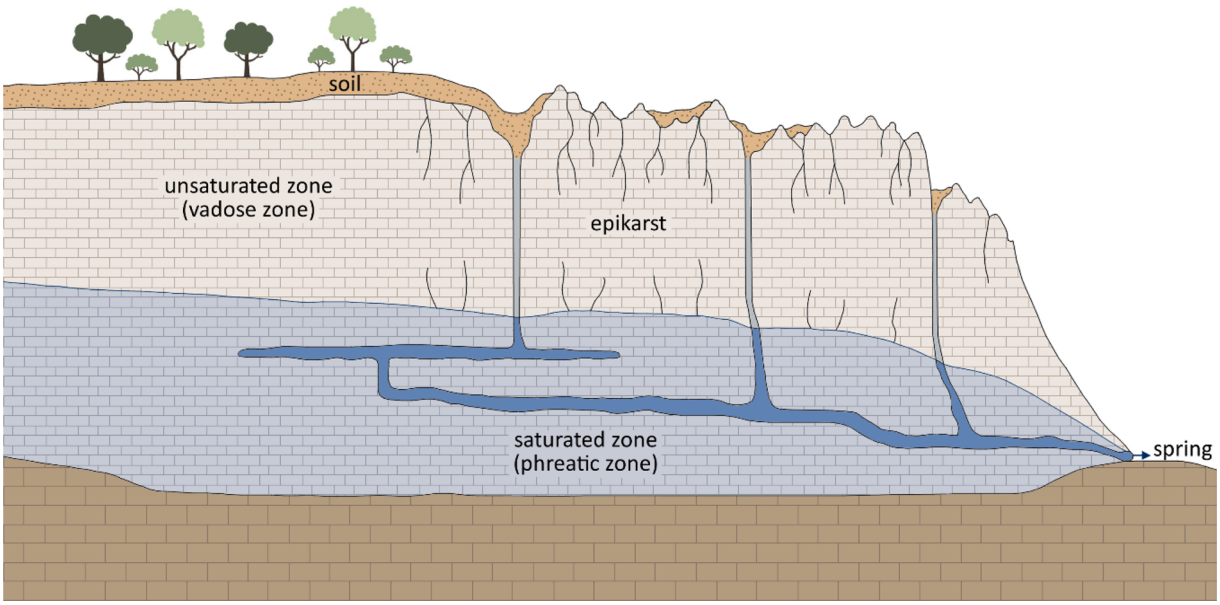

Fig. 1. A schematic representation of a karst water aquifer and conduit system.

series were compared to long-term isotope records of the precipitation.

2. Site description

2.1. Mecsek mts

Mecsek Mountains are located in southern Hungary, central Europe (Fig. 2d). The geological structure of Western Mecsek is characterized by an anticline with an east-west line of strike. The well-karstified Triassic limestone and dolomite of the anticline are particularly stressed, fragmented and faulted (Barta and Tarnai, 1999). The geological structure of Eastern Mecsek is characterised by a syncline, composed of Middle and Upper Jurassic and Lower Cretaceous limestone that is less prone to karstification. Although geologically part of a single block, topographically the karstic rocks of Western Mecsek can be found in three different zones and are mainly covered by brown forest soil with clay illuviation (Hoyk, 1999) and C3-type plants.

In Western Mecsek, hydrologeological investigations in the recharge area of the largest springs have shown a heterogeneous karst water table elevation between 180 and 260 m. Vadose passages up to 100 m in depth (Spiral cave) are known in the area and the thickness of the karstified vadose zone has been estimated to be on average ~50 m. Based on dye tracing studies Rónaki (1972) suggested that 80 % of the recharge area of Anyák and Melegmányi springs was karstic, while Kánya and Mariska springs are fed by an autogenic recharge area. To our knowledge, no dye tracing was carried out Eastern Mecsek (Rónaki, 2007).

The Mecsek Mts. are the warmest mountain range in Hungary, characterised by mean annual air temperature of 8.8 °C on the top of the mountain range (535 m asl). The area receives 500–700 mm precipitation annually and the local meteoric water line (LMWL) has been identified as $\delta^2H = 7.84 \cdot \delta^{18}O + 6.77$ (Mersich et al., 2003; Fórizs et al., 2020). Summer and winter precipitation is characterised by higher and lower stable isotope values, respectively. Although some of the air masses arrive via the Mediterranean Sea, long-term (2005–2012) precipitation monitoring in the area suggests that the primary origin of the moisture sources is the Atlantic (Fórizs et al., 2020).

2.2. Bükk mts

The geology of the uplands zone of the Bükk Mts., Hungary (Fig. 2c), includes Middle and Upper Triassic limestone. The majority of the area is hosted by the Bükkfennsík Formation (anchimetamorphosed platform facies limestone). The southern strip of the catchment

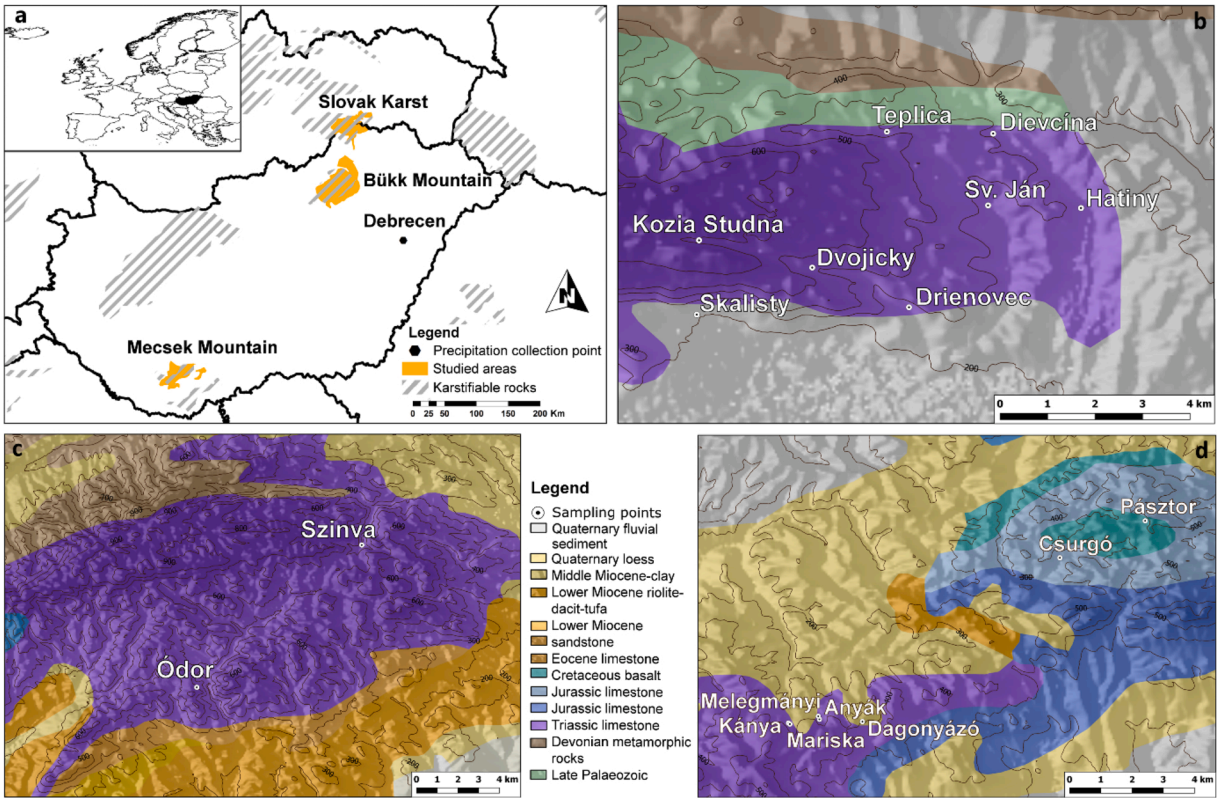

Fig. 2. (a) The three study sites in Hungary and Slovakia,(b) the Slovak Karst as a part of the Gömör-Torna Karst System, (c) Bükk Mountains and (d) Mecsek Mountains. The precipitation samples for tritium were collected in Debrecen, eastern Hungary.

consists of the Felsőtárkányi Formation (intrashelf basin facies thick bedded limestone with marl and chert intercalations), while the northern strip is made up of the Fehérkői Formation (thick-bedded platform facies limestone). There are a large number of karst springs registered in the Bükk area. The annual precipitation is 700–850 mm (Spinoni et al., 2015). The long-term average discharge of the Bükk springs is 2.25 m^3/s. One of the main springs, the Szinva is used as a drink water resource for over 100,000 inhabitants. The total catchment area (including allogenic recharge) is 280 km^2. The long-term average recharge calculated for the Bükk is 32.4 %, while on the karstic upland area the annual recharge is assumed to exceed 36 % (Kovács et al., 2015). The uplands area has a large number of sinkholes following linear patterns. Kovács et al. (2015) demonstrated that these features delineate water table conduits at depth, and that the size of low-permeability matrix blocks in between karst conduits range between 200 and 500 m. During drought periods, karst water level drops, concentrated flow ceases within karst conduits, and large-scale diffuse flow starts becoming dominant in the system (Kovács et al., 2015). This sort of scale-change of flow systems is characteristic of the Bükk uplands.

2.3. Slovak karst

Slovak Karst is the largest continuous karst area of Slovakia situated on the south-eastern boundary, which extends into Hungary as the Aggtelek Karst, forming the Gömör-Torna Karst System (Fig. 2). Together they present the largest plateau karst area of central Europe. The site is composed of dolines, karren fields, vertical and horizontal caves. The area is divided by deep canyons and gorges into seven individual units (plateaus). The easternmost plateau, Jasovská Plateau, is a separate geomorphological subunit (ca. 60 km^2). The gently sloping plateau surface is situated in the height of 650–500 m asl (Gessert, 2016). The main part of the area, similar to the whole Slovak Karst, consists of the Wetterstein limestone (Middle Triassic). The lower part of the plateau is built up by Steinalm and Waxenec limestone (Triassic). Tectonic lines are predominantly in the W-E direction. On the north edge of the plateau the ascending Gutenstein limestone and dolomite is visible as the result of the Silický príkrov Nappe overthrust (Mello Elečko et al., 1996). The plateau is divided lengthwise by the deep tectonic depression of the Miglinc valley, which relates to the lamellar structure of the area, it is a part of the Rožňavská línia Fault. Its slopes ascend different non-karstic rock of Jurassic age. This valley is a very clear delineation of apparently independent hydrological units (Mello et al., 1997). The south edge of the plateau is bounded by steep subvertical tectonic faults.

Most of the plateau area is covered by rendzic leptosols that are typical soils for this karstic mountain range. Average annual rainfall is 660–990 mm (Climate Atlas of the Slovak Republic, http://klimat.shmu.sk/kas/). Jasovká Plateau has a very clear hydrological

Table 1
The elevation of the investigated springs and the individual recharge areas.

Spring name	Mountain	Coordinates (°; WGS84)	Elevation of the spring (m asl)	Elevation of the recharge area (m asl)	Average discharge (l/min)
Anyák	Mecsek	N46.136331, E18.225253	343	380–460	20
Melegmányi	Mecsek	N46.137597, E18.224769	323	380–460	2
Dagonyázó	Mecsek	N46.135372, E18.243036	406	420–440	0.5
Kánya	Mecsek	N46.134792, E18.212419	336	360–470	50
Mariska	Mecsek	N46.134344, E18.212997	336	360–470	3
Csurgó	Mecsek	N46.203033, E18.324794	344	360–400	1
Pásztor	Mecsek	N46.218306, E18.360114	442	460–540	30
Ódor	Bükk	N47.987756, E20.508944	490	520–590	2
Szinva	Bükk	N48.079495, E20.615506	343	360–640	21,000
Dievcína	Slovak Karst	N48.674292; E20.976133	268	270–450	3
Drienovec	Slovak Karst	N48.624719; E20.951983	246	260–650	720
Dvojicky	Slovak Karst	N48.636070; E20.924453	410	420–610	3
Hatiny	Slovak Karst	N48.653040; E21.001224	234	240–300	720
Kozia Studna	Slovak Karst	N48.643858; E20.892019	574	590–700	1.2
Skalisty	Slovak Karst	N48.622627; E20.891384	210	240–630	1400
Sv. Ján	Slovak Karst	N48.653800; E20.974680	267	270–600	1800
Teplica	Slovak Karst	N48.674852; E20.945785	305	325–600	2100

boundary. Karst springs have variable water yields, many of which are only seasonal. However, the strongest springs (with average discharge of 20–50 l/s) are outlets from underground passages (Hochmuth and Gessert, 2017) (Table 1).

3. Methods

3.1. Sampling strategy

Between 2006 and 2020, water samples for environmental isotope tracers (^{3}H, ^{18}O, ^{2}H, noble gases) were taken regularly from karst springs in the three study areas. Five of the monitored springs (Kánya, Mariska, Anyák, Melegmányi and Dagonyászó springs) are located in the Western Mecsek and two springs (Csurgó and Pásztor springs) are located in Eastern Mecsek. Kánya and Mariska are located a few hundred meters from each other and the same is true for Anyák and Melegmányi springs (Table 1). In Bükk Mts., we focused on two springs: Szinva and Ódor. The Szinva catchment extends over a 12 × 3 km elongated area of 37 km^2. The Szinva spring is located at 343 m asl. The Ódor spring is located to the western-south from the Szinva by 30 km, at an elevation of 490 m asl. Eight karst springs were regularly investigated in the Slovak Karst. The elevation of recharge area in the three study sites compares to each other ranging from 240 to 700 m asl. The mean discharge rates are also shown in Table 1. These latter values were determined using the bucket method in the Mecsek Mts. and at Ódor-spring, while the discharge of Szinva and other springs in the Slovak Karst were calculated from water velocities using flow profiling. To reveal long changes in isotope properties, seasonal sampling frequency was chosen for the Mecsek Mts. and Slovak Karst. One specific spring (Anyák) was chosen to perform monthly sampling to observe whether there was a contribution of infiltration of very short residence time. Ódor and Szinva-springs were sampled in a 1−2 year time steps, since the searching for the long-term trend was of interest in Bükk Mts.

3.2. Analytical methods

Isotope analyses were performed at the Isotope Climatology and Environmental Research Centre (ICER), Institute for Nuclear Research, Debrecen, Hungary. Karst water samples have been analysed for tritium using the ^{3}He-ingrowth method with a special ^{4}He isotope dilution technique (Palcsu et al., 2010; Papp et al., 2012). The uncertainty of the method is 0.1–0.3 TU in the range of 5–10 TU.

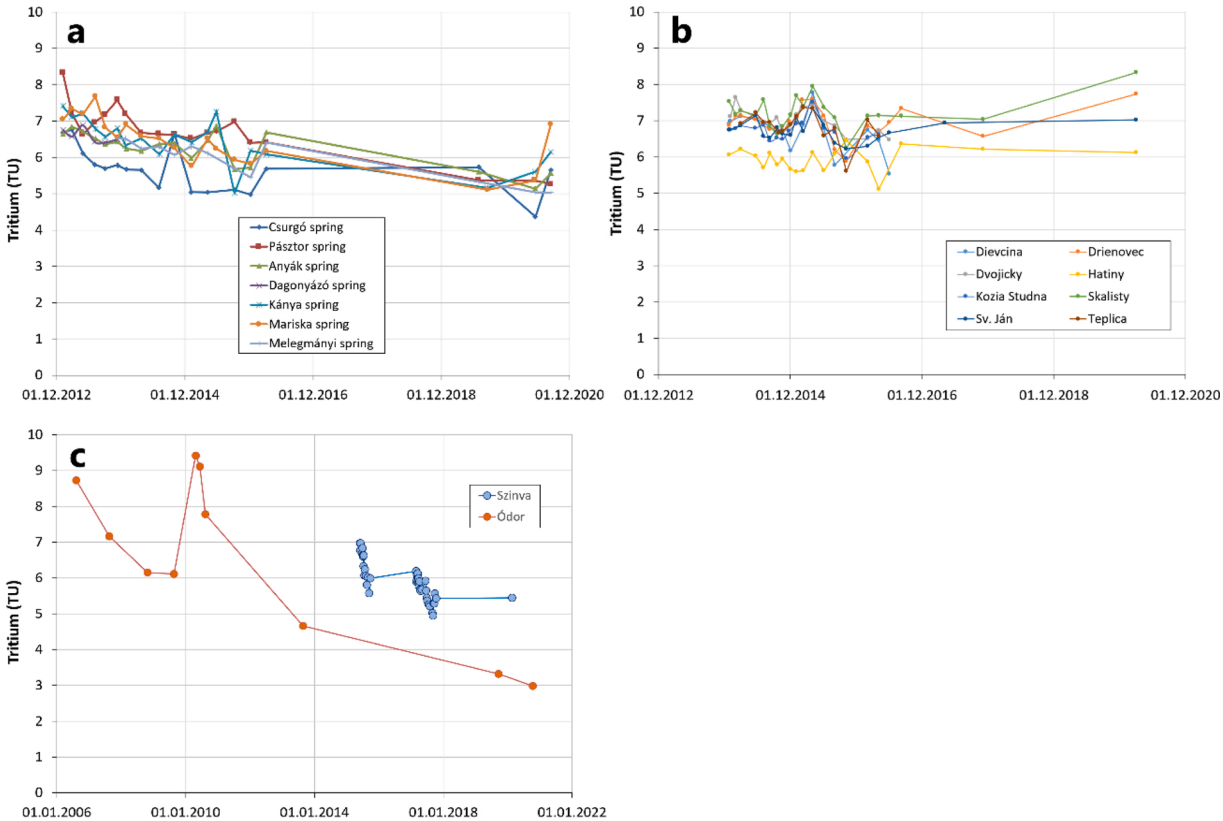

Fig. 3. Tritium time series of the karst springs in the three study sites: a) Mecsek Mts., b) Slovak Karst, c) Ódor-spring and Szinva-spring, Bükk Mts. Note that X-axes have different scales.The decreasing trend can also be seen in case of the other two study sites. Karst spring waters in the Slovak Karst have tritium values from about 7.0–7.5 TU in December 2013 to 6.5–7.0 TU in early 2016 (Fig. 3b). Afterwards, the decreasing trend cannot be seen anymore, in fact, the values of 2019 are slightly higher than before.

Stable isotope ratios (δ^2H and δ^{18}O) have been determined either by isotope ratio mass spectrometry (Vodila et al., 2011) or cavity enhanced absorption spectrometry (Czuppon et al., 2018). For hydrogen and oxygen isotope ratios, both methods have a measurement error of 1.5‰ and 0.1‰, respectively. In order to calculate ^{3}H/^{3}He apparent ages of the water, samples have also been taken for noble gas analysis. Helium (including ^{3}He/^{4}He), neon, argon, krypton and xenon concentrations have been analysed mass spectrometrically with a standard deviation of 1–2 %, as described by Papp et al. (2012).

3.3. Calculation of apparent ages

Tritium and noble gas measurement allow us to calculate ^{3}H/^{3}He apparent ages. The tritiogenic ^{3}He component can be decomposed from the measured noble gas concentrations and ^{3}He/^{4}He ratio, and then the ^{3}H/^{3}He age is calculated by "Equation 1″, where t is the age, λ is the decay constant of tritium, $^3H_{meas}$ is the measured tritium concentration, and the $^3He_{trit}$ is the tritiogenic component of dissolved ^{3}He (Schlosser et al., 1988; Palcsu et al., 2017).

$$t\left({}^3H/{}^3He\right) = \frac{1}{\lambda}\ln\left(\frac{{}^3H_{meas} + {}^3He_{trit}}{{}^3H_{meas}}\right) \tag{1}$$

The accuracy of the age calculations is 1−2 years. Several samples have been analysed for SF_6 in the Institute for Environmental Physics (IUP), Heidelberg, Germany, by gas chromatography (Friedrich et al., 2013), and then SF_6 apparent ages have been calculated based on the comparison of atmospheric mixing ratio of SF_6 in the previous decades (Darling et al., 2012)

4. Results and discussion

4.1. Tritium

The tritium values of the karst spring samples are slightly decreasing in each study sites in the investigated period (Fig. 3). In Mecsek Mts. the tritium of all springs are significantly decreasing from 7.0 to 8.0 TU to 4.5–5.5 TU during seven years (2013–2020) (Fig. 3a). Most of the springs have similar tritium concentrations; the only exception is Csurgó spring with a slightly lower tritium content. The tritium concentrations of the springs are compared to that of the precipitation and the recharge water. Fig. 3 shows the tritium concentration of the monthly precipitation taken in Debrecen, East Hungary. The arithmetic mean as an average of the monthly precipitation is 10.4 TU for the whole data set of the 19 years of collection. The time series of tritium in monthly precipitation shows a clear seasonality (Palcsu et al., 2018), i.e. the winter precipitation has a significantly lower tritium content than the summer precipitation. Although, the sampling location for the precipitation tritium time series is about 300 km apart the Mecsek Mts. (100 km from the Bükk Mts., and 150 km from the Slovak Karst) (Fig. 2), we consider this to be representative of the precipitation falling at each study site. The water samples in Mecsek Mts. were taken at 3–4 months intervals in the first three years, and then less frequently. Yet, in the first three years, the seasonality of the tritium concentration of the precipitation, and hence recharge water, cannot be observed in the discharge of the karst aquifer.

In the Bükk Mts. two karst springs have been investigated. A 14-year long, but less frequent sampling was performed at Ódor-spring (Fig. 3c). The tritium concentration of Ódor-spring was first observed to be decreasing from 2006 until 2009 (from 9 to 6 TU), and then it was rising by the next year, and then decreasing again.

The tritium concentration in 2019–2020 reached 3.0 TU. Another karst spring in Bükk Mts., the Szinva spring was sampled at weekly resolution for a discharge study in 2015 and then in 2017 again. In each individual year, the tritium time series show a decreasing trend, in fact, the tritium values are significantly lower in 2017 than in 2015 (Fig. 3c). Note that the accuracy of the tritium analyses is between 0.11 and 0.22 TU for these water samples, hence these changes in tritium are significant. An additional sample was analysed in 2020, but the tritium concentration cannot be significantly distinguished from the last results of 2017.

To summarize, in each study site the tritium concentrations of the karst waters show a long-term decreasing trend. The exception are two springs in the Slovak Karst, that have higher tritium concentration in 2020. Springs with the lowest tritium concentration might have the longer mean residence time. Similar decreasing trends can be seen in spring waters of other shallow karst aquifers (Kluge et al., 2010a; Al-Charideh, 2011; Ozyurt et al., 2014). Although the tritium concentration of precipitation is decreasing throughout the 1990′s, after 2005 it remains almost stable. The decreasing trend in the tritium time series can be attributed to these following processes: i. the bomb contribution having higher tritium content is getting negligible in the aquifer due to wash-out and radioactive decay, ii. the contribution of recharge is weak due to, perhaps, less precipitation or higher evaporation/transpiration rate, iii. the trend is following the trend in the precipitation which is modulated by the solar cycle. Palcsu et al. (2018) recently demonstrated that the tritium concentration of the precipitation does have a link to the solar cycle.

The higher annual tritium concentrations between 2006 and 2009 (see Fig. 6) can be attributed to weak solar magnetic activity, when more cosmic galactic particles could reach the Earth's atmosphere producing more cosmogenic nuclei, like tritium (Palcsu et al., 2018). To distinguish and confirm which process is responsible to the decrease of tritium, other environmental tracers have to be evaluated.

4.2. ^{18}O isotope composition

In karst springs, seasonal patterns in stable hydrogen and oxygen isotope composition of water are frequently observed and used to

reveal short response from recharge to discharge (Mance et al., 2014; Hu et al., 2015; Jeelani et al., 2015; Malík et al., 2015; Cao et al., 2018). However, as previously discussed, we did not observe any seasonality in the tritium time series. Stable isotope values of Mecsek and Slovak Karst springs do not exhibit a clear seasonal cycle, corroborating the absence of intra-annual variability in tritium levels. However, as samples have been taken at 3–4-month intervals, the lack of seasonality in both environmental tracers may be a result of under-sampling. Fig. 4a and b show the oxygen isotope composition of the Mecsek and the Slovak Karst, respectively. These two sites have slightly different oxygen isotope signature, the springs in the Slovak Karst have more positive values by 0.5–1.0 ‰. This can be attributed to several factors: the lower elevation or the higher mean annual temperature of the Slovak Karst, and moisture source regions or the recharge seasonality are different at the two areas. To confirm the absence or presence of seasonality in the springs, a frequent, 3–4-week sampling period was performed at Anyák-spring (Mecsek Mts.) for ^{18}O analysis from September 2015 until December 2018.

The main assumption is that if the ^{18}O of the local precipitation is varying according to the seasonality (Fig. 4c), the spring water has a very young component, it must be observed in the time series of the karst water as a seasonal change. Contrary to the previous observations when the sampling has not been done less frequently (in 3–4-month steps), the δ^{18}O time series of Anyák spring shows a significant periodic variation. As a result of a visual comparison of the two time series, this variation is shifted by 0.5 years from the seasonal variation of the precipitation (Fig. 4d), indicating a significant contribution of a fresh water component with a residence time of potentially half a year. In principle, the age of the fresh component can be n+0.5 years (n is an integer), but as can be seen later, the overall mean residence time of the water is very short. When estimating the yield of this very young component, one has to take a look at the magnitude of the variation in the δ^{18}O of the spring water and precipitation. The spring water is varying in a range of 0.6‰ (between -10.0‰ and -9.4‰), while the ^{18}O composition of precipitation covers a range of 6‰ (between -12‰ and -6‰) (Fig. 4c and d). Considering the variations of δ^{18}O values, and assuming the older component has a constant stable isotope composition (i.e. well mixed), the first order assumption for the contribution of the youngest water is at least 10 %. On the other hand, the average δ^{18}O of Anyák spring is -9.75‰ and the δ^{18}O values are varying around this value, while the unweighted and the weighted (by the precipitation amount) mean averages of δ^{18}O of the precipitation at six stations in the Mecsek Mts. are -8.82 and -8.66‰, respectively. Eq. (2) shows how different averages are calculated. Subscipts "s" and "w" denote summer and winter. Monthly precipitation amounts and an additional weighing factor are referred as "p" and "x", respectively.

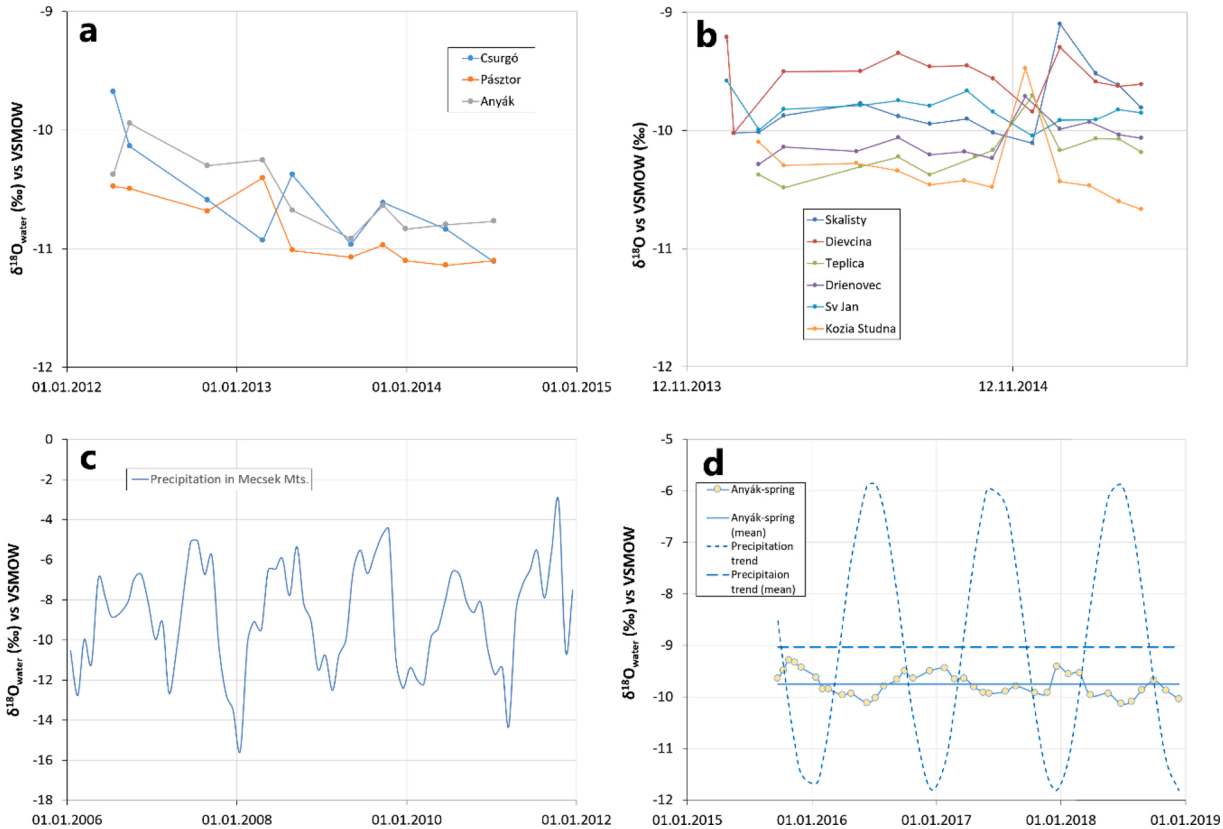

Fig. 4. Oxygen isotope composition (δ^{18}O) of karst springs and precipitation samples: a) karst springs in the Mecsek Mts., b) karst springs in the Slovak Karst, c) monthly precipitation in the Mecsek Mts. (Fórizs et al., 2020), d) time series of the Anyák-spring and a sinusoidal curve fitted to the δ^{18}O values of the local precipitation.

$$\delta^{18}O_{pr} = \frac{\sum_s \delta^{18}O_s \cdot p_s + \sum_w \delta^{18}O_w \cdot p_w \cdot x}{\sum_s p_s + \sum_w p_w \cdot x} \quad (2)$$

The spring has a lower δ^{18}O value than the precipitation average, this suggests that the winter precipitation prevails in the recharging water. In Hungary, the summer months have more precipitation, hence, when calculating mean annual average δ^{18}O values weighted by the precipitation amount, the δ^{18}O will be higher than the arithmetic average of the monthly values. An additional weighing factor (refer to "x" in Eq. (2)) for the hydrologic winter half-year (November to April) was modelled to obtain close match to the one of the long-term average of the spring. If we give an additional weighing factor of 5 (after a trial and error searching), as a best fit parameter for the cold months, an average value (-9.74‰) is obtained, which is reliable for the long-term average for the Anyák-spring (-9.75‰).

This approach gives an assumption that the precipitation fallen in the winter half-year prevails to the recharge by a factor of five than that of the summer months (from May to October). The same approach will be used when estimating the tritium concentration of the recharge water based on the monthly tritium values and precipitation amounts.

4.3. Recharge component analysis

To detect the origin of the water in the springs, and reveal the contribution of the re-evaporation of local waters, the relationship of ^{18}O and ^{2}H has been examined. The local meteoric water line in the Mecsek Mts. is $\delta^2H = 7.84 \cdot \delta^{18}O + 6.77$ (Fórizs et al., 2020). The δ^2H-δ^{18}O values of the spring waters are slightly above the LMWL, indicating a small contribution of re-evaporation to the local precipitation, although this effect has a minor importance (Fig. 5).

As one could see in Figs. 2 and 3, the tritium values of the springs are decreasing and lower than that of the precipitation. This allows us to restrict an estimation to the age distribution of the waters. The ^{18}O time series of one specific karst spring (Anyák, Mecsek Mts.) indicates a half-year residence time for the recent precipitation with a contribution of around 10 %. The tritium time series and other dating methods, such as ^{3}H/^{3}He (Kluge et al., 2010b; Szűcs et al., 2015; Miller et al., 2017; Palcsu et al., 2017)and SF_6 age determinations (Friedrich et al., 2013; Delbart et al., 2014) help us estimate the contribution of the older components in the range of years. Fig. 6 shows all tritium concentrations for each study sites and the precipitation for Debrecen (eastern Hungary). Additionally, an estimation for the tritium time series of the recharge water is also given in Fig. 6. The annual mean tritium values of the recharge estimation have been composed by weighing the monthly tritium values with the precipitation amount and an additional factor of 5 for the months within the winter half-year (see the chapter of ^{18}O isotope composition above). In Fig. 6, the lower tritium values of the spring waters indicate that the mean residence time of the water is not zero. If we want to estimate a residence time of the water, the tritium concentrations of the water and the precipitation have to be compared. The average tritium concentrations (arithmetic means) of the precipitation in Debrecen, eastern Hungary, and in Krakow, Southern Poland are 10.8 TU (Palcsu et al., 2018) and 10.0 TU (GNIP database, IAEA), respectively. We are aware that the isotope composition of precipitation does not reflect that of infiltrating water, because the summer contribution is smaller than that of the other seasons due to evaporation and transpiration. In case of the Debrecen precipitation, the arithmetic mean of the monthly tritium is 10.8 TU, while the mean tritium concentration weighted by the precipitation amount is 11.5 TU (note that there is more precipitation in summer than in winter). While using an additional weighing factor of 5 for the winter hydrological half-year, as was derived from the stable isotope composition (see above), the mean is 9.9 TU. As for a first estimation of the mean residence time of the springs in Mecsek Mts., this latter weighted mean tritium concentration is used to the calculation. According to the radioactive decay law, one can obtain mean residence times between 1 and 12 years. The youngest is the spring in the Slovak Karst (0–2 years), Mecsek spings and Szinva have similar ages (0–4 years), while the oldest is Ódor (4–12 years). However, this calculation assumes that the tritium concentration of the recharge water, i.e. the weighted average of the precipitation does not change. Obviously, this is not absolutely true. Figs. 3 and 6 show that the tritium concentration of the precipitation in the investigated duration do change. After the 1990′s, this change is apparently solely due to the solar cycle (Palcsu et al., 2018). The attenuation of the bomb peak cannot be seen any more in the precipitation. In Fig. 6, an apparent conformity between the long-term

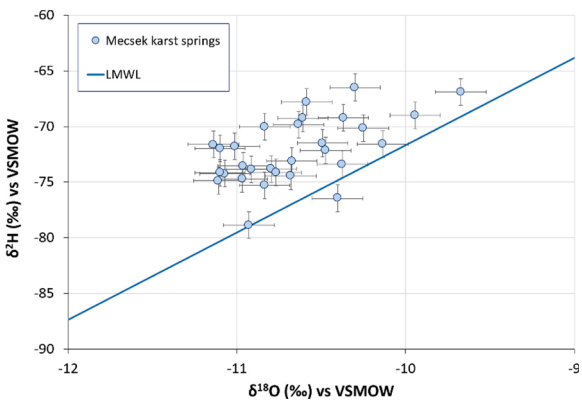

Fig. 5. Oxygen and hydrogen isotope plot for the studied karst springs in Mecsek Mts and the local meteoric water line (LMWL).

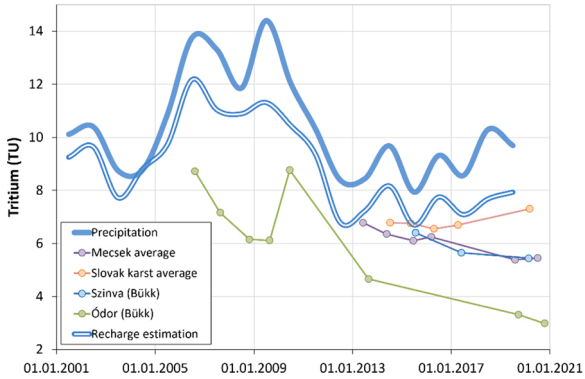

Fig. 6. Tritium time series of the precipitation and the karst springs (averaged for the Mecsek and the Slovak Karst).

tritium trends of the spring waters and the precipitation/recharge water can be seen.

An important question resulting from our study is whether these variations in the spring water tritium time series can be attributed to the solar cycle (Morgenstern et al., 2010). If this is correct, the long-term change of the tritium time series in the precipitation due to the solar cycle could be used to constrain the age distribution of shallow karst springs, when applying long-term time series for karst waters. To answer the question of the presence of the solar cycle in the time series, the mean residence times of the waters have to be taken into account.

4.4. Apparent ages

Additional age tracers have been used to further analyse the residence time. In the Mecsek Mts., samples for dissolved noble gases were taken between 2013 and 2015, and then $^3H/^3He$ apparent ages were calculated (Fig. 7a). These ages are varying between 0 and 10 years. The distribution of the age values is exponential (see the histogram in Fig. 7b). The median of these 47 $^3H/^3He$ ages is 1.4 years. Note that some tritiogenic ^3He might have been lost during sampling when the water yield was too small. As these springs are open discharges, it is possible that some helium degassing could have occurred during sampling, however we consider this process to be negligible. Although these young ages can be considered as minimum ages, these $^3H/^3He$ ages indicate that the mean residence time of the water are up to 10 years. To verify this, in July 2013 water samples for SF_6 determination were taken at four springs in Mecsek Mts. The calculated SF_6 apparent ages (corrected for excess air amount determined from the noble gas temperature calculation) agree very well to the $^3H/^3He$ ages in all cases, and both were from 0 to 1.4 years, indicating that those discharging waters were indeed very young (Table 2).

4.5. Source of variability in spring water tritium

The long-term tritium monitoring data of three different shallow karst aquifers shows in most cases decreasing tritium concentrations, and thus exhibits a similar pattern to the natural variability of tritium concentration in precipitation. In these three karst areas, significant higher changes in either air temperature or precipitation amount, evaporation-transpiration rate and recharge rate cannot be seen which would be responsible to the change of the distribution of younger and older water ages in the aquifer, thus

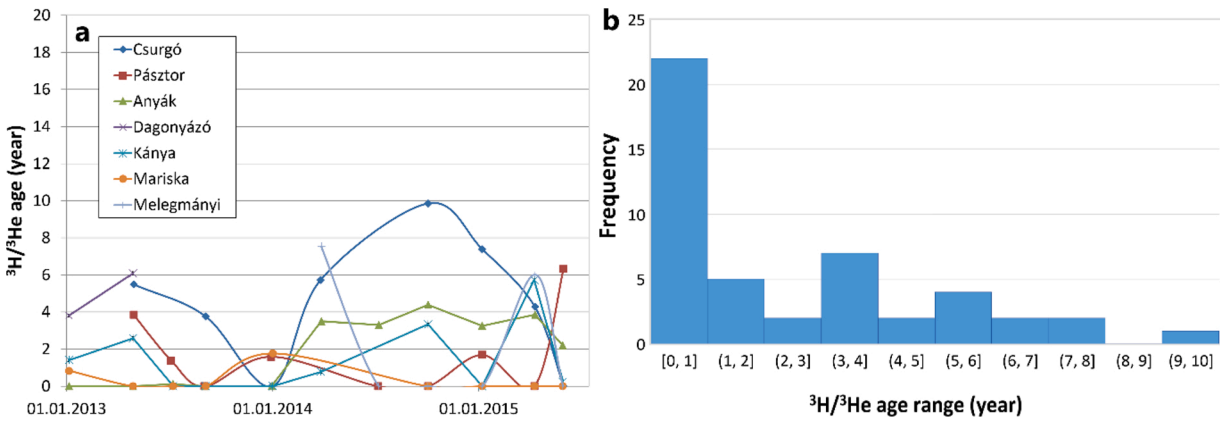

Fig. 7. (a) $^3H/^3He$ apparent ages of the springs in Mecsek Mts in time. The uncertainties of these ages are between 1.3 and 1.8 years. (b) The histogram of the 47 individual $^3H/^3He$ ages.

Table 2
Comparison of $^3H/^3He$ and SF_6 apparent ages of four springs in July 2013.

	Sampling date	$^3H/^3He$ age (year)	SF_6 age (year)
Mariska	9 July 2013	0.0 ± 1.3	0.0 ± 1.0
Anyák	8 July 2013	0.1 ± 1.5	0.0 ± 1.0
Pásztor	5 July 2013	1.4 ± 1.3	1.0 ± 1.0
Kánya	9 July 2013	0.0 ± 1.4	contaminated

resulting in lower tritium concentrations. One explanation for this effect can be the attenuation of the bomb peak contribution. The aquifers contain waters of different residence times, thus the older components used to include more tritium from the nuclear weapon tests, while the tritium concentration of the younger components is free of bomb tritium, and has already reached the natural level (Palcsu et al., 2018). As time goes, the contribution of the bomb tritium gets less significant. Moreover, the radioactive decay provides an additional decrease of tritium. The decreasing signature is the strongest in case of Ódor-spring (Bükk Mts.), as it also has the lowest tritium concentration. This strongest decrease and the lowest tritium amount are in accordance with the highest mean residence time. The other springs in Bükk and Mecsek Mts. show similar pattern. Even if a slight contribution of very recent waters (detected by the frequent analysis of $\delta^{18}O$) might contribute to the age distribution of the springs (as can be seen in Anyák-spring, Mecsek Mts.), the decreasing trends are similar to each other, and can be attributed to the depletion of the bomb contribution. However, those springs whose tritium concentrations are higher, i.e closer to that of precipitation and the estimated recharge values, may follow another trend as well (i.e. having some increase from 2016 onwards). As the $^3H/^3He$ and SF_6 apparent ages indicate that the spring water in Mecsek Mts. is very young (1.4 years as a medial of the age distribution), it is expected that the long-term changes of tritium in precipitation, and hence the recharge water, is reflected more directly in the spring waters. Since the precipitation time series are modulated by solar activity (Palcsu et al., 2018), the solar cycle should be observed in the tritium time series of the karst waters. The highest tritium concentrations can be seen in the Slovakian karst springs (Fig. 6). The individual samples as well as the annual averages are increasing in tritium from the year 2016. Similar pattern can be observed in the precipitation and the recharge estimate: the tritium is increasing from 2015 probably due to the weakening solar activity, as expected. However, even though the signatures compare well, we do not conclude the elevated tritium of the Slovakian spring is directly linked to the increase of the precipitation. The 6-year time series is still not long enough to decide whether the tritium in the precipitation modulated by the solar activity can be observed in shallow karst aquifers.

5. Conclusion

Time series of environmental tracers have revealed important information on the recharge and discharge conditions in three karst aquifers in Hungary and Slovakia. Frequent sampling (2–4 weeks) of spring waters can help us determining the contribution of recent infiltration. In the case of spring waters from Mecsek Mts. in Hungary, the 10 % of the discharge water has a residence time of half a year. Additionally, the recharge conditions can be also estimated using the stable isotope composition of the water. Weighing isotope data of the winter and summer months separately provides us with an adequate estimation to investigate the behavior of the recharge during the winter and summer half-year. This approach, namely using a weighing factor of 5 for the winter months, has been used when estimating the tritium concentration of the recharge water based on the monthly tritium values and precipitation amounts. The apparent ages vary between 0 and 10 years, showing and exponential distribution in the $^3H/^3He$ ages of the Mecsek springs. The median of the $^3H/^3He$ ages is 1.4 years. Using the tritium values and the estimation of the tritium time series of the recharge water, mean residence times have been estimated. The youngest is the spring in the Slovak Karst (0–2 years), Mecsek spings and Szinva have similar ages (0–4 years), while the oldest is Ódor (4–12 years).

An important question resulting from our study is whether the variation of tritium in the spring water can be attributed to the solar cycle. Even though the signatures compare well, we do not conclude the elevated tritium of the Slovakian springs or some Mecsek springs is directly linked to the increase of the precipitation. The 6-year time series is still not long enough to decide whether the tritium in the precipitation modulated by the solar activity can be observed in shallow karst aquifers. The solar-activity signal would make possible the determination of a few year residence times using long-term times series of environmental tracers, including tritium, however further studies and longer time series are needed for this. Thus, additional samples for tritium will be taken from each spring in these study areas twice a year, and hopefully, after obtaining longer time series, we can confirm the absence or presence of the solar component in the shallow karst waters.

CRediT authorship contribution statement

László Palcsu: Conceptualization, Methodology, Investigation, Data curation, Writing - original draft, Funding acquisition. **Alena Gessert:** Investigation, Writing - review & editing. **Marianna Túri:** Investigation. **Attila Kovács:** Data curation. **István Futó:** Investigation. **Judit Orsovszki:** Investigation. **Anita Puskás-Preszner:** Investigation. **Marjan Temovski:** Investigation, Writing - original draft. **Gabriella Koltai:** Conceptualization, Methodology, Investigation, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.ejrh.2021.100858.

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