Fluid Flow Systems and Hypogene Karst of the Transdanubian Range, Hungary—With Special Emphasis on Buda Thermal Karst

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Judit Mádl-Szőnyi, Anita Erőss, and Ádám Tóth

Abstract

Carbonate regions have great economic importance for water supply, oil and gas reservoirs, geothermal fluids and also Mississippi Valley-type ore deposits. Therefore, the understanding and consequences of flow pattern in carbonates require special interest. The hypogene and epigene karst areas of carbonate sequences were distinguished and associated with different orders of groundwater flow. However, the effect of confinement on flow pattern of carbonate aquifers was not fully considered in previous studies. We demonstrated the most important prerequisites and consequences of the application of the gravity-driven regional groundwater flow concept for carbonate sequences at different degrees of confinement. The results put into a frame the distribution of different springs and caves (epigene and hypogene) of the carbonate system of the Transdanubian Range, Hungary, and provide insights for better understanding of the hydrogeology of areas with similar unconfined and confined settings. Relationship among different flow regimes, distribution and character of springs and hypogene karstification processes, in addition to natural discharge-related phenomena, such as mineral and microbial precipitates, were recognized in the area of Buda Thermal Karst. This area is a natural laboratory where the connection between groundwater flow and karstification processes can be studied.

Keywords

Spring • Groundwater flow • Mineral and microbial precipitate • Confined and unconfined carbonate aquifers • Basinal fluid

1 Introduction

Deep and thick carbonate systems with different degrees of confinement constitute the most important thermal water resources in terrestrial areas outside of volcanic ranges (Goldscheider et al. 2010; Mádl-Szőnyi 2015; Mádl-Szőnyi and Tóth 2015). These deep and confined carbonate sequences are exposed to porosity enlargement processes, summarized under the term hypogene speleogenesis, involving various dissolution mechanisms, which was reviewed by Palmer (1991, 1995), Klimchouk (2000, 2012) and Goldscheider et al. (2010). Thermal and mineral waters of carbonate sequences are used by spas for recreation purposes all over the world. In addition, geothermal installations use these resources for electricity production and district heating (Mádl-Szőnyi 2015). Geothermal installations apply thermal water often with CH_4 combustion or with CO_2 sequestration (Han and McPherson 2009; Zhang et al. 2011).

Based on these considerations, it is a scientific and practical question to better understand why thermal water resources and hypogene karstification processes are so closely connected to terrestrial carbonate systems. The review paper of Goldscheider et al. (2010) dealt with this issue in a

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J. Mádl-Szőnyi (✉) · A. Erőss · Á. Tóth
József and Erzsébet Tóth Endowed Hydrogeology Chair, Department of Physical and Applied Geology, Eötvös Loránd University, Pázmány P. stny. 1/C, Budapest, 1117, Hungary
e-mail: szjudit@ludens.elte.hu

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comprehensive way. The authors pointed out the significance of the understanding of thermal water resources and hypogene karstification processes in the framework of hierarchical flow systems. In their Fig. 2 (Goldscheider et al. 2010), the hypogene and epigene karst areas are distinguished and complemented with supposed flow paths (cold, lukewarm and thermal) and different geochemical processes influencing hypogene karstification. The source of basinal fluids was supposed to lie in the basement, and the significance of terrestrial heat flow was also indicated. This figure is purely conceptual and is not based on any physically proven flow pattern.

Klimchouk (2007) was the first who put the potential dissolution processes in carbonates into the framework of the so-called Tóthian-type flow systems (Tóth 1999). The title of his Fig. 1 (Klimchouk 2007) was "Epigenic and hypogenic karst in the context of basinal groundwater flow." He also mentioned the constraints that "the figure shows mainly gravity-driven flow in an idealized homogeneous basin. In reality, most sedimentary sequences are highly heterogeneous, and gravity-driven flow interacts with other flow mechanisms." He recognized that epigene karstification corresponds to local flow systems while hypogene to regional or intermediate ones. However, based only on the two unconfined sub-basins of Tóth, the location and characteristics (chemical and temperature distribution) of thermal and lukewarm springs and the near-surface and underground locations of hypogene karstification cannot be properly explained.

This discrepancy was resolved by the conceptualization of Tóthian-type flow pattern for idealized connected confined and unconfined sub-basins based on the joint interpretation of numerical flow and heat transport simulation and spring analysis (Mádl-Szőnyi and Tóth 2015). Based on this adapted flow model in carbonate sequences, we can distinguish between those parts of the system where carbonates are partially covered with siliciclastic sediments (unconfined or semiconfined sub-basin) and those where carbonates are fully confined with dominantly siliciclastic sediments (with low hydraulic conductivity). In this system, the sub-basin where the carbonates can be found under thick siliciclastic sediments is, in fact, confined. The significance of the boundary between these two sub-basins was revealed explicitly based on flow and heat transport simulations in Mádl-Szőnyi and Tóth (2015). These two sub-basins and consequent heat and flow pattern can be handled as a new adaptation of Tóthian flow concept for carbonate sequences. With these settings, the numerically derived asymmetric flow pattern can explain the distribution of springs, the heat pattern and the mixing of basinal and fresh waters (of local, intermediate and regional flow systems) and the potential location of epigene and hypogene speleogenesis.

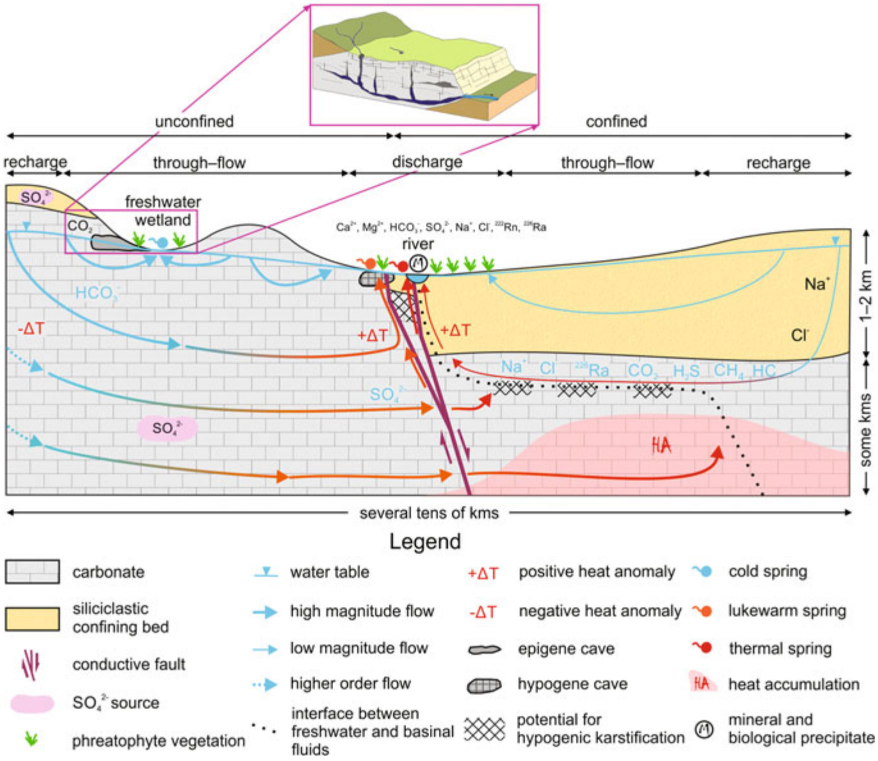

Fig. 1 Conceptual Tóthian-type GDRGF pattern for the boundary of unconfined and confined sub-basins. The consequences on flow-related manifestations imply that the shallow karst aquifer (*inset*; modified after Goldscheider and Drew 2007) is embedded into the regional flow pattern as a local system (after Mádl-Szőnyi and Tóth 2015)

The detailed explanation of Fig. 1 can be found in Mádl-Szőnyi and Tóth (2015).

The first part of this chapter demonstrates the most important prerequisites and consequences of the application of gravity-driven regional groundwater flow concept for carbonate sequences. The results are used to explain the distribution of different springs and caves (epigene and hypogene) of the carbonate system of the Transdanubian Range, Hungary, and provide insights for better understanding the hydrogeology of areas worldwide with similar unconfined and confined settings. The second part summarizes the relationship between flow regimes, springs and hypogene karstification processes for the Buda Thermal Karst. This area is a natural laboratory where the connection of groundwater flow and karstification processes can be studied.

2 Fluid Flow Systems and Hypogene Karstification in Confined and Unconfined Carbonate Ranges—Theoretical Considerations

2.1 Cross-Formational Flow and Confinement of Carbonates

Regarding the potential for the evolution of epigene and hypogene caves, the most decisive geological factor in carbonate sequences is the confinement of carbonate aquifers, in agreement with the findings of Klimchouk (2007). However, confinement usually refers only to the existence of thick siliciclastic cover above carbonate sequence, but does not reveal its influence on flow pattern and consequent fluid distribution in the system. Therefore, we have to understand the evolution of flow pattern in such regions to learn more about its effect on both epigene and hypogene karstification.

Especially, the cross-formational flow (ascending or descending vertical flow component) has great significance on hypogene karstification processes as it was highlighted by Klimchouk (2007). The direction and intensity of flow are determined by not only the hydraulic conductivity (aquifer or aquitard units) but also the driving forces determined by fluid potential (Hubbert 1940) differences.

We can assume a given basin with two (confined and unconfined) sub-basins and cross-formational flow driven by topographic differences operating in both parts. Until now, however, the physically based flow patterns were not derived for such situations because first, there were not enough measured data to reveal it and second, basin-scale numerical simulations were not available due to conceptual limitations since the applicability of equivalent porous medium (EPM) approach was formerly debated for carbonate sequences.

2.2 Heterogeneity of Permeability on Basin Scale

Owing to the heterogeneity of a karst aquifer in recharge and permeability pattern (matrix, fissured and enhanced permeability due to dissolution), difficulties arise in the adaptation of the gravity-driven regional groundwater flow (GDRGF) concept (LaMoreaux et al. 1975; Scanlon et al. 2003; Wellman and Poeter 2006). However, this issue can be overcome by focusing not on an individual aquifer and its detailed permeability distribution but on a carbonate system or basin with its enhanced permeability (see in details in Mádl-Szőnyi and Tóth 2017). In addition, heterogeneities of a basin need to be interpreted based on the consequences of artificial or natural changes of flow pattern (Tóth 1995). This requires a different basin-scale approach and methods compared to usual catchment or aquifer-scale consideration (Mádl-Szőnyi and Tóth 2015).

2.3 Hydraulic Continuity in Carbonates

Thick carbonate systems should be considered as hydraulically connected components of a carbonate basin with unconfined and confined subregions where water-table differences induce gravity-driven flow. The hydraulic continuity can be interpreted on a given timescale for a basin, if a change in hydraulic head at any point of the flow domain causes a head change at any other point, within the time interval of the observation (Tóth 1995). It was derived by Mádl-Szőnyi and Tóth (2015) that hydraulic continuity is more effective in carbonates as compared to siliciclastics due to the higher hydraulic diffusivity of carbonates. Consequently, artificial and natural changes in hydraulic head (and pressure) propagate at higher speed and over greater distances and depths in carbonate rocks than in clastic sediments.

2.4 Significance of Springs

Since springs are natural discharge features (Tóth 1971; Engelen and Kloosterman 1996; Ford and Williams 2007; Kresič and Stevanovič 2009; Tóth 2009b), they represent the termination of the underground flow paths. Consequently, their areal distribution and basic characteristics can help to understand the basinal hydraulic pattern of gravity-driven groundwater flow (which could be influenced by additional driving forces) in carbonate regions. If springs are considered in the context of flow systems, their regularities become clear. The most specific basic parameters are: elevation of spring discharge point, water temperature, total dissolved solids (TDS) and chloride content and volume discharge (Mádl-Szőnyi and Tóth 2015).

The elevation of discharge points of springs represents the hierarchy of the gravity-driven flow. Their exact location and topographic position can be used to delineate terminal zones of flow systems and to indirectly differentiate between flow systems of different orders. Cold karst springs theoretically have to discharge at the elevated parts of a basin, while lukewarm and thermal springs occur near the boundary of confined and unconfined carbonates (Mádl-Szőnyi and Tóth 2015). These springs are often related to marginal conductive structural elements.

In carbonate sequences, advective heat transport facilitated by flowing groundwater also affects the flow pattern (Domenico and Palciauskas 1973; Sass 2007). It can be simply supposed that the water temperature of springs is indicative of the order of flow, connected to local, intermediate and regional systems. Different driving forces, such as gravity, buoyancy (i.e., thermal convection or thermal density effect) can contribute to this, but their effects cannot be simply separated (Havril et al. 2016).

The water in unconfined karst systems has basically meteoric origin; therefore, the hydrogeochemical character of water in short (local) flow systems and in recharge areas of longer (intermediate or regional) flow systems has basically calcium–magnesium bicarbonate facies. The TDS for all gravity-driven flow systems generally increases along the flow paths. The ratio of SO_4^{2-}/HCO_3^{-} increases due to the decrease in the CO_2 of soil origin (Back 1966; Tóth 1999, 2009a). The sulfate can be of biotic or abiotic origin, from oxidation of sulfide minerals and hydrogen sulfide related to organic matter maturation, dissolution of gypsum and anhydrite, etc., and can appear in lukewarm and thermal waters (Langmuir 1971; Bretz 1949; Egemeier 1981; Hill 1987; Worthington and Ford 1995; Gunn et al. 2006). Toward the discharge areas (i.e., the springs), the chemical composition can change from calcium–magnesium and sodium bicarbonate–sulfate to sodium chloride in the confined karst. Thermal springs are characterized by elevated chloride content, and the deep origin of CO_2 and H_2S may also be characteristic for them (Goldscheider et al. 2010). Regarding the findings for the Transdanubian Range, the source of NaCl-type water in the systems was the thick, dominantly low-permeability siliciclastic confining layer. Its contribution to the underlying karst water is responsible for the appearance of additional basinal flow component. This "basinal fluid" can be represented by its chloride content as a conservative component of flow systems (Mádl-Szőnyi and Tóth 2015).

The magnitude, intensity and variability of spring discharge are topography dependent. Spring discharge is affected by geological conditions and climate as well. Therefore, the variability of spring discharge is also characteristic because springs fed by local flow systems supposedly display higher variability than those with a higher order of flow systems (Mádl-Szőnyi and Tóth 2015; Bodor et al. 2014). The variability of discharge of thermal springs, in general, is low, i.e., steady due to the buffering effect of the large storage capacity of the rock framework of the regional flow system (Klimchouk 2007; Tóth 2009a; Bodor et al. 2014).

2.5 Conceptual Flow Model and Connected Manifestations for an Unconfined and Confined Carbonate Basin

The flow is basically lateral in an unconfined carbonate system, with limited vertical flow components compared to siliciclastic basins, and the hierarchy of flow systems is not well developed (Fig. 1). In unconfined carbonates, the water has basically meteoric origin and the temperature of springs systematically increases toward the lowest elevation discharge point. The flow pattern at the margin of unconfined and confined carbonates is more complex due to the asymmetric recharge rates at the two sides. The "fresh" karst water, which originated from meteoric recharge through unconfined carbonates, is shifted toward the confined carbonates due to limited recharge across the confining strata. The limited meteoric recharge may flush some Cl^{-}-rich basinal fluids from the low-permeability siliciclastic cover into the underlying carbonates, therefore creating an interface between "freshwater" and "basinal fluids" under the confined sub-basin. Along this interface the upwelling basinal fluids contribute to the chemical composition of thermal and partially lukewarm springs with dominantly meteoric origin. The interface of fresh and basinal fluids governed by gravity-driven flow and modified by other driving forces can be economically important due to potential for porosity enlargement and hydrocarbon and heat accumulation. The ^{226}Ra, ^{222}Rn and dissolved gas content (CO_2, H_2S and CH_4) of thermal springs originates from the "basinal fluid component." The hypogene karstification (connected porosity enlargement) can be related to those biogeochemical processes which appear along the interface between fresh and meteoric fluids. A basin-scale conceptual model was derived to summarize and generalize these findings. This model can explain the position of cold, lukewarm and thermal springs in a basin as well as their chemical facies. In addition to the hypogene and epigene karstification, mineral precipitates can be directly connected to this generalized flow pattern (Mádl-Szőnyi and Tóth 2015).

3 Fluid Flow Systems and Hypogene and Epigene Karst of the Transdanubian Range

The hydrogeologically complex thick carbonate system of the Transdanubian Range (TR) is located in the Central Pannonian Basin, Hungary, and bounded by tectonic lines and built up by thick Paleozoic metamorphic and Permian–Cretaceous sedimentary sequences; however, the Triassic–Early Jurassic carbonates form the main karst system of the area, which is dissected by major faults and influenced by folding. The carbonate range has 300 km × 100 km in extent (Csepregi 2007) and is partially confined by thick Neogene sediments mainly at the edges (Fig. 2).

The hydraulic continuity of the TR was inferred by long-term (1950–1990) mine dewatering (Alföldi and Kapolyi 2007) which has resulted in drying up of springs at the highest elevations and caused changes in chemical composition, temperature and spring discharge at lower elevations. In addition, the hydraulic head decrease could be followed on a regional scale for the whole reservoir (Csepregi 2007).

Cluster and discriminant analyses were carried out based on a database containing geodetical, volume discharge and chemical data of 800 springs before artificial intervention took place based on Hungarian Spring Register. Spring elevation, water temperature, chloride content and discharge data were used during the evaluation (Bodor et al. 2014). The derived groups were compared with the results of numerical simulations. Finally, springs were grouped into different flow systems (Mádl-Szőnyi and Tóth 2015) (Fig. 3).

The character and pattern of springs show a systematic distribution in the Transdanubian Range. The local flow-related spring groups can be found at the highest elevation (above 175 m asl); their temperature is lower than 12.2 °C; and there is no difference between their chloride content (<6 mg/l) based on medians of the different parameters of the groups. However, their discharge volume shows marked variations (1×10^{-4} to 6×10^{-4} m^3 s^{-1}). The springs of the second group are separated from local flow systems. Their discharge elevation is higher than 120 m asl, and their temperature is elevated, around 19 °C based on medians. These springs can be connected to intermediate flow systems. Finally, the springs with highest temperature (42.3 °C) and chloride content (154 mg/l) and lowest discharge elevation (104 m asl) can be connected to regional flow systems. The discharge volume cannot be interpreted directly in terms of flow systems (Mádl-Szőnyi and Tóth 2015).

Thermal springs related to deep regional flow systems can be found only in the NE margin of the system, along the River Danube. Lukewarm springs of intermediate flow systems discharge at or close to the boundary of the confined and unconfined parts of the system, whereas local flow relates to cold springs and discharges in the unconfined part of the system (Fig. 3) (Tóth and Mádl-Szőnyi 2016).

Taking into consideration the basin-scale distribution of epigene caves, it can be concluded that they are associated with the subsurface drainage of the unconfined system. Hypogene caves were developed or are developing at the boundary of confined carbonates. Inactive or dry hypogene caves occur at higher elevation in unconfined carbonates. This indicates a considerable erosion of siliciclastic cover and downward shift of the discharge zones related to the Late Miocene uplift (Haas 2001; Dombrádi et al. 2010).

4 Buda Thermal Karst: The Regional and Intermediate Discharge Area and Hypogene Karst Region of the Transdanubian Range

The northeastern regional and intermediate discharge zone of the TR is located close to the River Danube (Figs. 2 and 3). This particular part of the system, the Buda Thermal

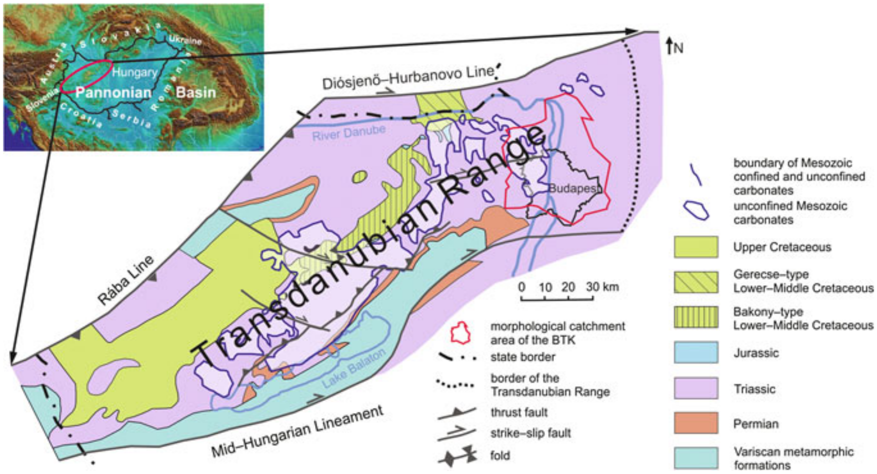

Fig. 2 Transdanubian Range Unit: boundaries of the range and the Paleozoic and Mesozoic formations with the indication of main structural features and the area of unconfined carbonates (modified after Fülöp in Haas 2001 and Fodor 2010) and also the morphologically delineated catchment area of Buda Thermal Karst (BTK) (after Mádl-Szőnyi et al. 1999)

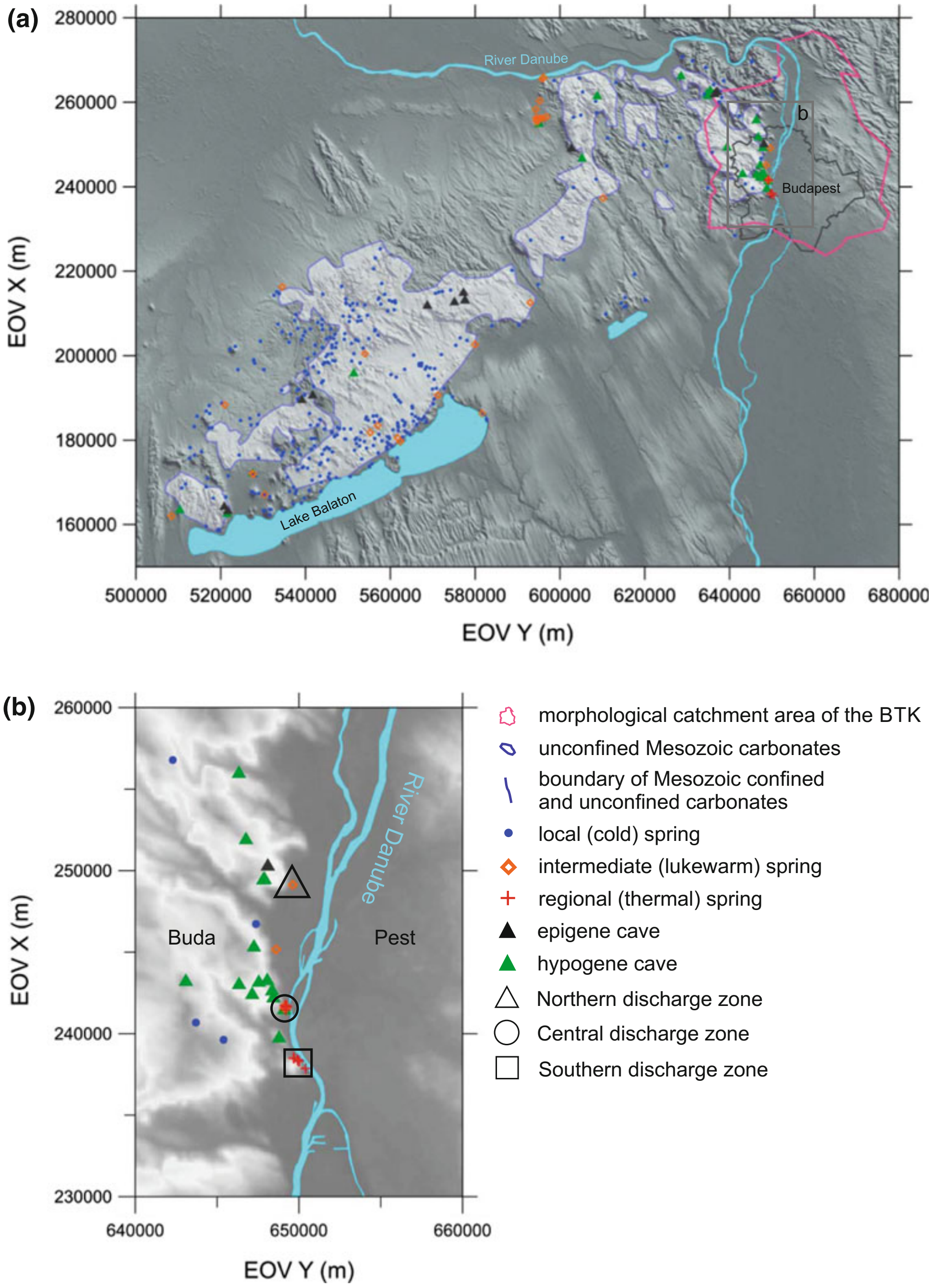

Fig. 3 **a** Surface manifestation of GDRGF for the TR. Springs are related to local (cold), intermediate (lukewarm) and regional (thermal) flow systems based on the classification of Mádl-Szőnyi and Tóth (2015). Location of epigene and hypogene caves is based on the data from the Hungarian Ministry of Agriculture and Rural Developments, Hungary (A. Gazda 2014, personal communication). The figure is complemented by the morphologically delineated catchment area of Buda Thermal Karst (BTK) (after Mádl-Szőnyi et al. 1999). **b** Spring groups and epigene and hypogene caves of Budapest and surroundings (EOV is the Hungarian National Grid which is a transverse Mercator projection—positive X is pointed to north and positive Y is pointed to east. Coordinates refer to meters)

Karst (BTK), is situated within and in the surroundings of Budapest. The area is strongly controlled by Cretaceous, Paleogene–Miocene, Miocene–Pliocene and Pleistocene structural elements, mainly normal and reversed faults trending in NW–SE, NE–SW and N–S directions (Fodor et al. 1994). These tectonic elements act as conduits or barriers and have strongly affected the paleo and recent fluid migration (Poros et al. 2012; Erőss et al. 2008; Havril et al. 2016). The tectonic boundary of unconfined and confined carbonates can be seen inside the morphologically delineated catchment area of BTK (Figs. 2 and 3a). This hydrogeological situation established the famous bath culture of Budapest and is responsible for the development of hypogene caves as well.

4.1 Gravity-Driven Flow Pattern and Fluid Components of BTK

The BTK area receives fluid from several sources (meteoric and basinal), in agreement with the conceptual model (Fig. 1). In addition to meteoric fluids, the contribution of basinal fluids at the discharge area of the BTK was first suspected based on water chemical analyses of Alföldi (1979) and was proved based on recent fluid analyses by Erőss (2010), Erőss et al. (2012a) and on mineralogical and fluid inclusion studies by Poros et al. (2012). The origin of the basinal fluid was determined to be related to the confining layers of the carbonates. The mechanisms of its contribution to spring discharge were also revealed in the form of vertical downward leakage from the confining layer to carbonates and upward flow to the springs (Mádl-Szőnyi et al. 2015; Mádl-Szőnyi and Tóth 2015).

The discharging fluids from different origins (meteoric and basinal) with different flow systems (intermediate and regional) result in a wide range of discharge features including springs, caves and also mineral precipitates. Extensive hypogene cave systems have been developed (e.g., Takács-Bolner and Kraus 1989; Leél-Őssy 1995; Leél-Őssy and Surányi 2003; Leél-Őssy 2017, in this volume) and are actively forming at present due to the interaction between fluids and the carbonate rocks (Erőss 2010; Erőss et al. 2011b). Therefore, the BTK system can be considered as a prime example of an active hypogene karst (Erőss 2010).

The natural discharge of the system is manifested mainly in the form of springs along and in the riverbed of the River Danube (Fig. 3a) forming three distinct discharge areas and are strongly influenced by tectonic pattern (Alföldi et al. 1968; Ötvös et al. 2013; Erhardt et al. 2017). The Northern discharge zone of the BTK is characterized by lukewarm springs (18–24 °C, TDS < 1000 mg/l); in the Central zone both lukewarm (21–27 °C, TDS < 1000 mg/l) and thermal springs (53–63 °C, TDS > 1000 mg/l) occur, while in the Southern discharge area of the system temporally and spatially uniform thermal water discharge (33–45 °C, TDS 1500–1700 mg/l) is characteristic (Papp 1942; Alföldi et al. 1968; Erőss et al. 2008). The lukewarm springs were evaluated as belonging to intermediate flow systems and the thermal springs as the discharge of regional flow systems based on cluster analysis (Bodor et al. 2014) and numerical simulations (Mádl-Szőnyi and Tóth 2015).

Since the second part of the nineteenth century, deep wells were increasingly used in addition to natural springs. Déri-Takács et al. (2015) evaluated the waters in Budapest by multivariate exploratory techniques, and the influence of the temperature and chloride content was the strongest in grouping springs according to their characteristics. Besides the previously distinguished three temperature-based groups, which are reflecting the natural discharge conditions, an extra group of the deep wells was determined in the confined part of the system.

A comprehensive hydrogeological study was carried out for the characterization of processes acting presently and their resulting parameters at the discharge zone of the BTK (Erőss 2010; Erőss et al. 2011b, 2012a, b). Studying the attributes of springs and wells, caves, mineral precipitates, that is, the entire range of phenomena related to discharge (Tóth 1971), information can be obtained related to the parent flow system, its hydrogeological environment and the processes taking place throughout the entire length of the flow system and in the close vicinity of the discharge zone. Moreover, the identification and understanding of recently active processes and their manifestations in this hypogene karst area will help to identify and understand paleo-phenomena both in the BTK and in other hypogene karst areas with similar settings.

The relationships between flow systems and hypogene karstification were examined for the Central and Southern discharge zone, with further input from the deep wells for the characterization of the confined part of the system in Pest (Fig. 3b). Two distinct conceptual flow (Fig. 4a, b) and cave development models were developed for the Central and Southern discharge areas.

4.2 Flow Models and Fluid Components for Central and Southern Discharge Areas

According to hydraulic and hydrogeochemical studies and analyses of radionuclides, considerable differences were identified between the flow systems and discharging waters for Southern and Central systems (Erhardt et al. 2017; Erőss et al. 2012a, b).

Radionuclides (^{226}Ra and $^{234+238}$U) as natural tracers were used to characterize the different flow systems based on their different geochemical behavior: radium is mobile in reducing, and uranium in oxidizing conditions. Discharging waters of regional flow systems are characterized by reducing conditions and therefore with negligible uranium content, whereas local flow systems represent oxidizing environments and consequent low radium content. The radium content of waters in the Southern discharge area is in the range of 221–870 mBq/l, while in the Central discharge zone it is between 53 and 591 mBq/l. The uranium values in the Southern zone vary between 11 and 33 mBq/l, while in the Central discharge zone are in the range of 10–83 mBq/l (Erőss et al. 2012b). With the aid of radionuclides (^{226}Ra and $^{234+238}$U), the mixing end-members for the discharging waters of the Central system were identified: a meteoric end-member with an average temperature of 12 °C and 775 mg/l TDS, and a thermal end-member with a temperature of 76.5 °C and 1440 mg/l TDS, respectively (Fig. 4a). The thermal end-member has also dominantly meteoric origin (regional flow component) with additional basinal fluid component based on the numerically proved conceptual model (Fig. 1). The cold meteoric end-member could be identified due to a local flow component (Mádl-Szőnyi and Tóth 2015). Taking into consideration the derived flow systems, the cold meteoric end-member and the

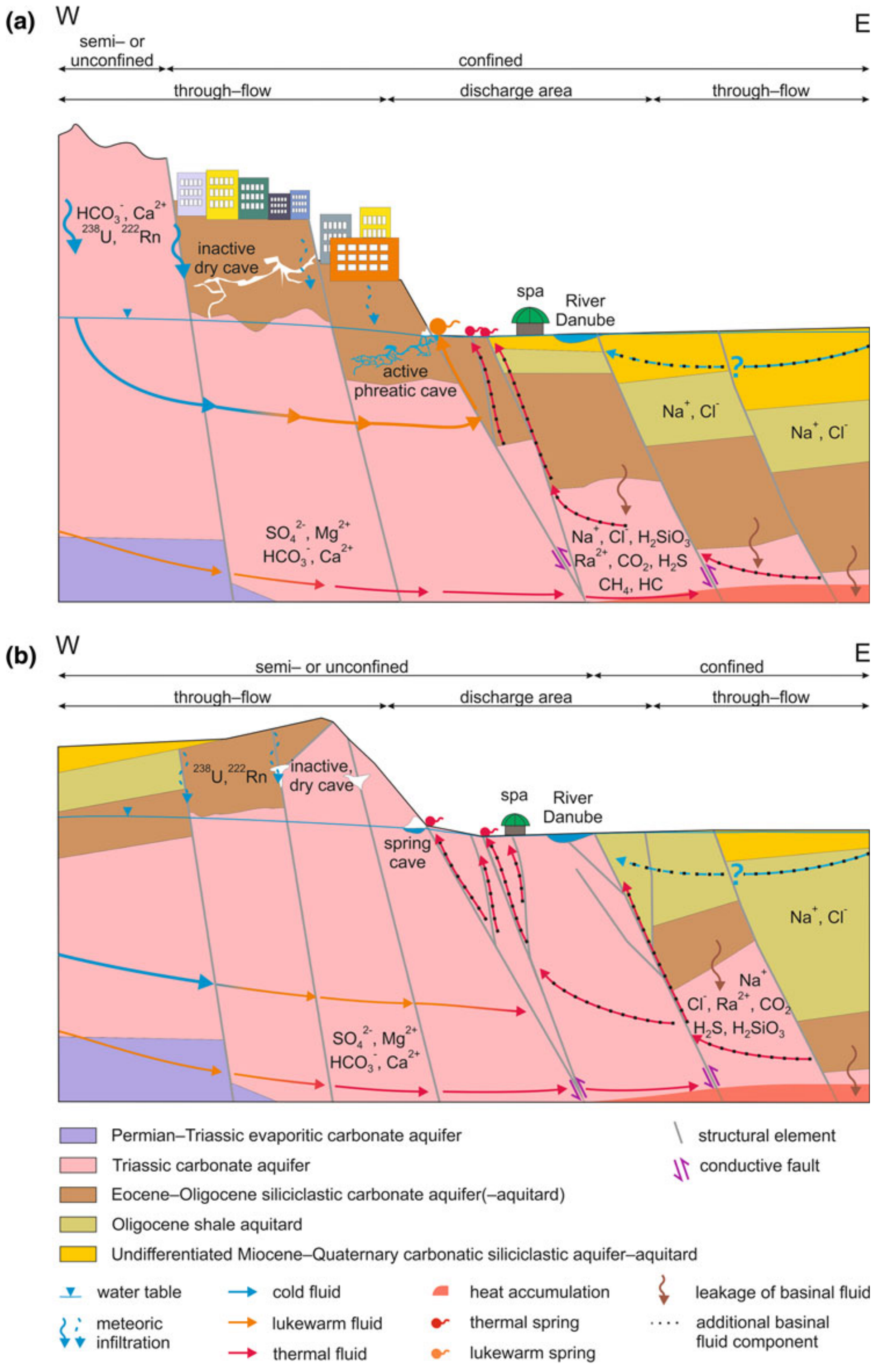

Fig. 4 Conceptual flow models: **a** Central system; **b** Southern system (modified after Erőss 2010 based on the generalized model of Mádl-Szőnyi and Tóth 2015)

thermal end-member represent different flow systems (local and regional) resulting different end-member composition.

For the Southern system, the thermal waters range from 35 to 47 °C temperature and 1400 to 1800 mg/l TDS, and only one component could be inferred with the help of the radionuclides (Erőss et al. 2012b) (Fig. 4b). Comparing the two systems, the Southern zone is characterized by elevated Ca^{2+}, Mg^{2+}, HCO_3^-, SO_4^{2-} and TDS content. These higher values correspond to lower temperatures within a narrower range (35–47 °C). The aforementioned differences highlighted between the two areas are also apparent on the distribution of parameters in relation to depth. However, with regard to Na^+ and Cl^- (i.e., the basinal fluid component) there are no differences between the two systems.

The contribution of basinal fluids to the discharging waters of BTK was identified in the form of Na^+, K^+, Cl^-, H_2SiO_3, CO_2, H_2S, CH_4 and liquid hydrocarbons (Erőss 2010; Erőss et al. 2012a; Mádl-Szőnyi and Tóth 2015). The similar Na^+, Cl^- and CO_2 content of the waters in both systems also highlights the common basinal origin. However, slight differences were also identified between the two systems regarding the basinal components, e.g., no hydrocarbon was found in the Southern system (Erőss 2010).

The geochemical and temperature difference between the sub-systems is due to the slightly different hydrogeological environment. The recharge area of Central system is composed of large exposed carbonate surfaces facilitating the recharge and resulting in a larger volume (>10,000 m^3/day) of discharge of meteoric origin (Papp 1942; Alföldi et al. 1968). As opposed to the unconfined recharge area, the discharge area can be characterized by confined conditions; therefore, the discharge of the cold meteoric karst water and the upwelling thermal fluids is structurally controlled having an important effect on the mixing of these waters. Mixing can only occur through structures. Since the recharge area of the Southern system can be characterized by a limited surface of exposed carbonates, the meteoric contribution is also limited in the discharge area. Furthermore, a significant strike-slip fault and some hydrostratigraphical changes also impede water flow from the western recharge areas toward the discharge area (Erhardt et al. 2017). The natural discharge rate of the Southern system (3200 m^3/day; Papp 1942; Alföldi et al. 1968) is thus lower when compared to the Central system, reflecting differences in the recharge conditions.

4.3 Hypogene Karstification Processes for the Central and Southern Discharge Areas

In the Southern discharge area, due to the lack of mixing members, different processes are proposed to be responsible for the formation of the caves compared to the Central area. Based on hydrogeological considerations and recent microbiological investigations (Borsodi et al. 2012; Anda et al. 2014), microbially mediated sulfuric acid speleogenesis is proposed as the dominant cave-forming process for the Southern area (Erőss et al. 2011a, 2012a). This process is further supported by actively forming gypsum crusts on the cave walls above the water table.

Based on the radionuclide study, it can be confirmed that the dominant cave-forming process in the Central area is, indeed, mixing corrosion. According to the observed discharge characteristics at the Central discharge zone, where the discharge of lukewarm and hot springs were clearly separated and tectonically controlled, mixing could only exist either by dispersion or along faults. Therefore, the dissolutional porosity also has a strong structural control, as can be seen on the cave maps, the distribution of the passages following the main structural directions. This study highlights the importance of hydrogeological investigations in the understanding of cave formation.

4.4 Mineral and Microbiological Precipitates

Besides hypogene karstification processes, mineral precipitates can also be characteristic in discharge zones. Beside travertines, calcite-rich mud and rafts, gypsum crust and iron–manganese–hydroxides seem to be the most characteristic active geochemical byproduct at the Buda Thermal Karst. Iron–hydroxides are composed by poorly crystallized ferrihydrite and goethite, based on Mössbauer spectroscopy measurements (Erőss 2010; Kuzmann et al. 2014). The occurrence of iron–manganese–hydroxide precipitates may denote the mixing of anoxic deep waters and oxygen-rich meteoric waters in case of the Central zone as these precipitates were found in deep phreatic conditions. In the southern caves, they may be indicative of the presence of an oxidation zone, as they are found at the spring outlets.

The ^{226}Ra activity of the recent iron–hydroxide precipitate is 1460–3680 (±10%) Bq/kg (dolomite background: 45 Bq/kg). It was also proved that this iron–hydroxide is the source of the ^{222}Rn content of the discharging waters (Erőss 2010; Erőss et al. 2012b). Besides radium, accumulation of iron, manganese, arsenic and other trace elements (Pb, Cr, Cu, Ni, Zn, Mo, U) as biominerals on the surface of precipitate are observed in the discharge zones (Erőss 2010; Dobosy et al. 2016). According to Tazaki (2009), microbial mats can accumulate heavy metals and radionuclides through precipitation and complexation on and within the bacterial cell surface containing carboxyl and hydroxyl groups.

Iron–hydroxide precipitates were also found associated with calcite rafts in the dry paleo-caves of the BTK. It can therefore be deduced that the same principle may apply to ancient precipitates. The mineralogical composition of these old iron–hydroxide precipitates is dominantly goethite and can also be characterized by similar trace element content as the recent ones (Erőss 2010). Accordingly, their common occurrence with calcite rafts in paleo-systems can be used as evidence of cave formation. This association thus can serve as a cave level marker and can be used to identify former hypogene cave discharge areas.

5 Summary and Conclusion

The evaluation of hypogene karstification in a flow system context can help to improve the understanding of associated processes in a comprehensive way. The numerical and conceptual adaptation of unconfined Tóthian-type flow for thick carbonate regions with unconfined and confined basinal settings (Mádl-Szőnyi and Tóth 2015) led to the identification of a very special asymmetric flow pattern with different (local, intermediate and regional) flow systems. These fluids have dominantly meteoric origin. Additionally, as a new factor, the contribution (via leakage) of basinal fluids from the confining cover in hypogene karstification was also proved and recognized. It could be also concluded that the underground interface between basinal and meteoric fluids is of special interest, because along this interface very different fluids with different origin (meteoric and basinal) can contribute to hypogene karstification (Mádl-Szőnyi and Tóth 2015). The physically based conceptual model can help to interpret the hypogene karstification as the manifestation of groundwater flow (Fig. 1).

The model is illustrated by the example of the Transdanubian Range, Hungary. The distribution of natural springs in the region can indicate the terminal zones of natural flow systems. Therefore, these discharge features have special significance in the understanding of flow patterns. Numerical simulations based on EPM approach can reveal the pattern of springs for the region (Mádl-Szőnyi and Tóth 2015). The comparison of clustering of springs and the derived flow systems for the TR led to good correlation between groups and flow systems. It was found that springs related to local flow systems appear at the elevated part of the range and are characterized by low chloride content and low, but increasing temperature toward the lower surface elevations. It was also shown that lukewarm and thermal springs appear close to and at the boundary of the confined and unconfined settings. The thermal springs appear at the lowest elevations, and they have the highest chloride content originated from basinal fluid contribution. The correlation between springs and epigene and hypogene caves as discharge features of groundwater flow was also demonstrated. The epigene caves can be found at the elevated part of the system; however, inactive hypogene caves can be found not only in the surroundings of thermal and lukewarm springs but also in a higher position than the recent boundary of unconfined and confined settings. This can be explained by the uplifting and erosion of the confining layers since the Late Miocene.

The correlation between flow systems and hypogene karstification was demonstrated on the example of Buda Thermal Karst. Hydraulic, hydrogeochemical and radionuclide studies revealed the flow pattern and fluid components for the BTK (Erhardt et al. 2017; Erőss et al. 2012a, b). Based on these data, distinct flow models were derived for the Central and Southern discharge areas (Erőss 2010; Erőss et al. 2012a). We can interpret the first model as characterized by intermediate and regional flow, while the latter as due to only regional flow. The origin of fluids is mostly meteoric, but additional basinal fluids from the confined basin also contribute. The differences in the chemical composition of discharging water can be explained by the differences in the hydrogeological environment of the BTK.

The distinct flow models could be connected to different dominant hypogene karstification processes. In the case of the Central discharge area, this is represented by mixing corrosion, while for the Southern discharge area microbially mediated sulfuric acid speleogenesis was found to be the dominant process. It was also recognized that not only karstification but also precipitation (in the form of calcite rafts and iron–hydroxide precipitate) can be correlated with the cave-forming process.

The example of Buda Thermal Karst in the frame of the Transdanubian Range demonstrates the importance of the flow system concept in hypogene speleogenesis. The flow model for confined and unconfined settings can be used to understand the connected subsurface and discharge-related processes in similar hypogene karst regions of the world. Furthermore, this knowledge can be used for planning thermal water utilization (Mádl-Szőnyi 2015).

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