Complex hydraulic and geochemical evaluation of the Villány thermal karst area

PD 116227 OTKA Final Report

Rationale and aims of the project

The porosity evolution of carbonate rocks and the development of karst systems are strongly coupled with groundwater flow, as these rocks are highly soluble. Since groundwater is considered as a geologic agent (Tóth 1999), karst phenomena including caves, springs, and the karst system itself can be interpreted as manifestations of flowing groundwater. Therefore, hydrogeology has a crucial role in karst research which can highlight the connections of different karst phenomena.

Deep, regional carbonate aquifers probably constitute the most important thermal water resources outside of volcanic areas (Goldscheider et al. 2010), however they are important as hydrocarbon reservoirs as well. Modern karst systems where the fluids, as well as their dynamics and effects can directly be investigated, deliver analogues and important parameters for reservoir diagenetic studies.

Owing to the favourable geographical, geological and geothermal conditions many of the regional carbonate reservoirs in Hungary are characterized by surface outcrops as well as by a subsurface continuation – a covered part – serving as basement of adjacent sedimentary basins. The basin fill sediments above the carbonate rocks as heat insulators play an important role in the fact, that many of the karst areas are characterized by natural thermal water discharge, usually at the boundary of outcropping carbonates and adjacent basins. Furthermore, important geothermal resources can develop in their covered part, in their foreland. These marginal, tectonically controlled areas between the two reservoir parts are favourable sites for the development of hypogene caves, where the morphology clearly reflect the effects of upward flowing waters and the minerals show the effect of thermal waters. These areas are favourable sites of mixing corrosion as well, because different order flow systems convey waters with different characteristics to here and their interaction with each other and the carbonate rocks often resulted in cave development.

The Villány thermal karst system in South Hungary is characterized by outcropping Mesozoic carbonates (unconfined reservoir part in the Villány Hills) which continue shallow covered (i.e., confined position) within the southern foreland area, then gradually deepens towards the Drava Basin, forming a thick (up to 1700 m) karst reservoir. Groundwater of the karst reservoir is mainly used for *drinking water supply* (e.g. Siklós, Mánfa, Villány etc.), *balneological purposes* with long tradition (Harkány, Siklós, Beremend), but there is also an example for *geothermal heating system* in the wider region (in Bóly). Natural groundwater discharge is mainly connected to the foothills of the Villány Hills (cold (10°C) karst springs in Máriagyüd) and to the uplifted basement carbonate blocks in the foreland of the Villány Hills represented by lukewarm springs (Büdöstapolca (20 °C), Siklós (24 °C) and Kistapolca (24 °C)). Thermal water discharge (around 50°C) was observed on the surface in form of a swamp in the area of Harkány. This area was drained, and Hungary's first thermal well was constructed to capture the thermal water in Harkány (Zsigmondy 1873). Nowadays all the natural springs are captured and most of them are intensely used for water abstraction, except the springs in Kistapolca.

There are several caves connected to uplifted basement carbonate blocks in the foreland of the Villány Hills or to the margin of the hills, where the morphology and the minerals decorating the cave walls indicate the effect of thermal waters (Dezső et al. 2004; Rónaki 2000; Takács-Bolner 1985; Takács-Bolner in Székely 2003a; Takács-Bolner in Székely 2003b; Vigassy et al. 2010). Some of them, the largest ones, were discovered by human activity (quarrying): Nagyharsány Cave, Beremend Cave. The Beremend Cave is connected to karst aquifer even today.

The groundwater, the thermal water resources and the caves of the Villány karst system were hitherto investigated separately. However, all these phenomena belong to one single system, and they can be evaluated only if their context is understood, i.e. if their common cause is revealed: the pattern of groundwater flow and its thermal and geochemical characteristics. Based on the experiences on the Buda Thermal Karst system in Budapest, the *gravity-driven regional groundwater flow approach and the concept of hydraulic continuity* is applicable in karst systems (Erhardt et al. 2017; Erőss et al. 2012a; Mádl-Szőnyi et al. 2017, 2018; Mádl-Szőnyi and Tóth 2015), therefore, this approach and the measured data based hydraulic evaluation method were used in the Villány karst area in contrast with the traditionally applied aquifer focused hydrogeological approach. This basin-scale hydrodynamic approach treats the flow field (including aquifers and aquitards) as a whole. During the interpretation of fluid potential anomalies, the effect of hydrostratigraphy or geology (permeabilities) can directly be concluded. *This approach and the measured data based hydraulic methods were first applied in the area during this project*. Groundwater hydraulics supported by hydrogeochemistry can help (i) delineate the flow regimes (discharge and recharge zones), (ii) determine the origin and driving forces of different fluid components, (iii) evaluate the role of structural elements and lithological heterogeneities in fluid flow, and (iv) identify and characterize recent rock-water interactions and processes, such as cave formation.

Based on the previous considerations, the aims of the project in the Villány thermal karst area were:

1. Evaluate the groundwater flow system based on measured hydraulic data.

2. Characterize the geochemical composition of the waters, use natural tracers (chemical components) to identify different fluid components.

3. Evaluate the cave forming processes.

Results of previous studies – initial information

In the first year of the research project the main focus was on literature survey (including papers, monographs, reports, maps etc.) and archival data collection. BSc students were involved to this retrospective hydrogeological research phase, who submitted their BSc thesis about their literature study (Kremmer 2016; Csobaji 2016). Many unpublished hydrogeological reports and maps were found from the area related to water management of drinking water supply systems and the thermal spas. A comprehensive report about the Harkány, Matty and Büdöstapolca water resources from 2008 included a new, yet unpublished, conceptual model of the area (Csicsák et al. 2008), which indicated an interplay between the Drava sedimentary basin and the carbonate suite of Villány. As a driving force of the basinal fluids tectonic compression and compaction was mentioned and as indicators of basinal fluids organic compounds (hydrocarbon traces) were designated in the thermal waters of Harkány. Lorberer and Rónaki (1978) and Liebe and Lorberer (1981) also suggested other reservoir connections form north-northwest (Szigetvár-Bogádmindszent and West Mecsek).

Results of regional groundwater flow system evaluation

Following the results of these above mentioned reports, and according to the original goals of the project, the measured data based hydraulic and geochemical evaluation was extended to Drava-basin and to the direction of the Mecsek Mountains, composing a 4571 km^2 large area (Fig. 1).

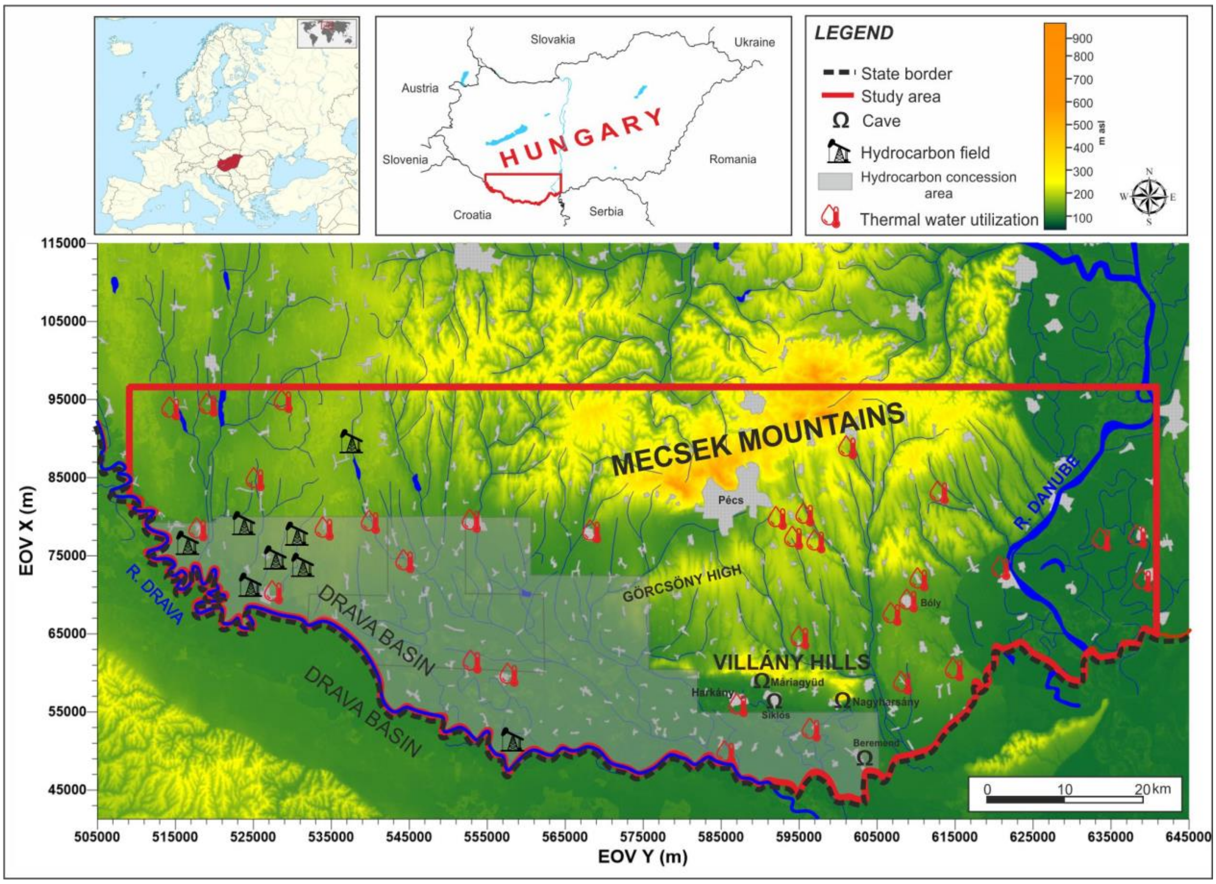

Fig. 1 Location of the regional study area in South Transdanubia with the indication of groundwater-related resources and phenomena

This method required enormous archival data collection of the paper-based documentation of water and hydrocarbon wells from data repositories (Mining and Geological Survey of Hungary, General Directorate of Water Management of Hungary). Nevertheless, a digital database was compiled from these data for the whole area, which can be used also for further studies facilitating a huge scientific and practical potential. MSc students were involved in data collection and in the hydraulic and geochemical evaluation and processed different parts of the whole study area (Csondor 2016; Zádeczki 2018; Csobaji 2018).

During the measured data based hydraulic assessment 44 pressure-elevation (p(z)) profiles, 6 tomographic fluid potential maps and 3 hydraulic cross sections had been constructed to determine the vertical and horizontal fluid flow conditions. Hydrochemical data analyses were carried out based on both archival and measured (i.e. measurements completed during this project) chemical data to support the hydraulic conclusions.

These results of the regional-scale study were summarized in a paper "*Katalin Csondor, Brigitta Czauner, Lehel Csobaji, Orsolya Győri, Anita Erőss: Characterization of the regional groundwater flow system in South Transdanubia (Hungary) to understand karst evolution and development of hydrocarbon and geothermal resources*" and submitted to the *Hydrogeology Journal* (submitted on10. February 2020, review arrived on 30. March, revision with minor issues is completed; IF (in 2018): 2.401). The main conclusions regarding 1) the regional hydraulic and geochemical evaluation of the area and 2) the cave forming processes derived from the regional-scale studies are summarized here as follows.

In the shallower regions of the study area down to about z=(-500) m asl vertical flow directions usually reflect the topography. Namely, downward flow conditions can be observed in the topographically higher areas, and horizontal and upward flow conditions in the topographically lower areas. Consequently, gravitational flow systems could be identified, thus recharge (downward flow), midline (horizontal flow) and discharge (upward flow) areas (Fig. 2).

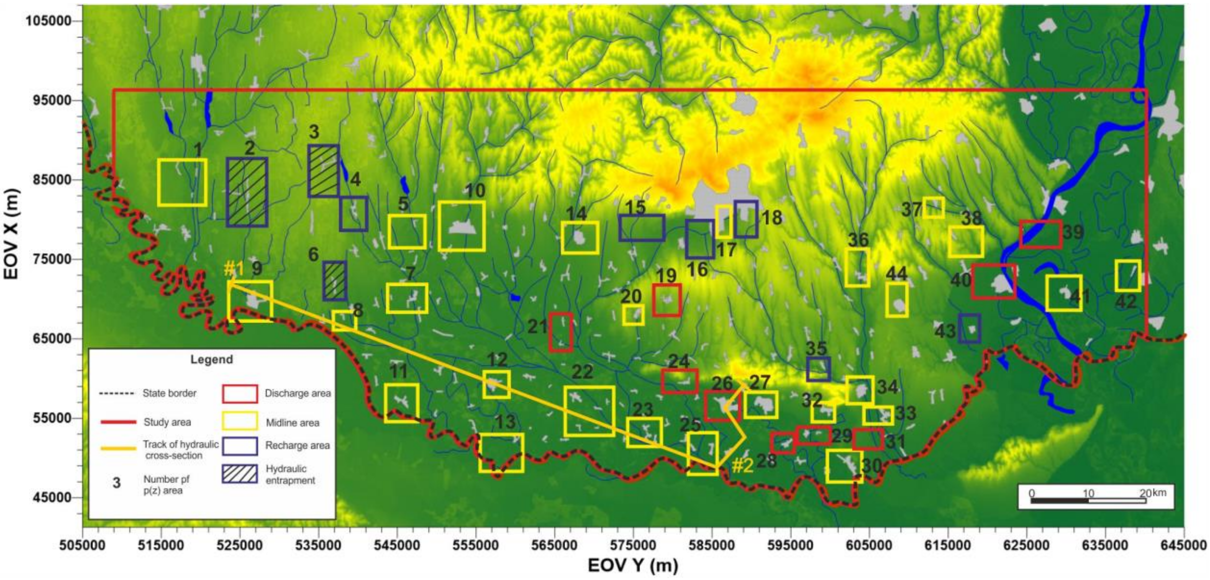

Fig. 2 Vertical flow conditions in the areas of p(z) profiles from the land surface down to z=(-500) m asl

Proven by the available data, penetration depth of downward flow (in recharge areas) is around z=(-400)-(-500) m asl. Below this elevation a dominantly lateral flow with some vertically upward flow component could be determined beneath the shallow recharge and discharge areas as well around z=(-500)-(-1000) m asl. These upward flows are still gravitational flows considering the magnitude of h values and the lack of overpressure. On the whole, horizontal flow conditions are dominant within the gravitational flow system of the study area, while the regional flow direction reflects the topography and tends toward S-SE, i.e. from the mountainous and hilly regions towards the regional base levels of the Drava and Danube Rivers. Although it is worth mentioning that discharge areas cannot be found along the Drava River, opposite to the Danube River (Fig. 2). On the other hand, in the deeper regions of the study area below about z=(-1500)-(-2000) m asl, which are represented by data only in the Drava Basin, an overpressured flow system could be identified with significant overpressures (max. 66%) and vertical pressure gradients (max. 41.58 MPa km^{-1}). Its vertical flow component is dominantly upward, whilst fluids are driven laterally from the deeper sub-basins towards their margins, i.e. northeast, east and southeast, which do not reflect the topography (Fig. 3). Transition zone of the gravitational and overpressured flow systems could be determined between about z=(-1500) and (-2000) m asl where the overpressure seems to be totally dissipated.

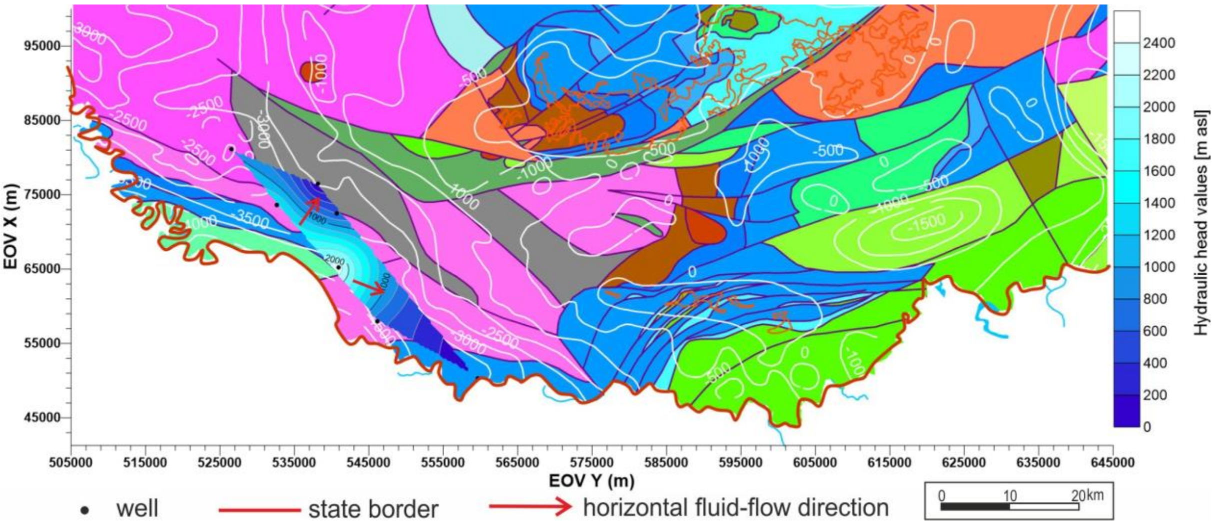

Fig. 3 Tomographic fluid-potential map #6: fluids are driven from the deeper sub-basins towards their margins, i.e. northeast, east and southeast. Background: Pre-Neogene basement map (Haas et al. 2010)

In the eastern half of the study area data were available only from shallow depth (above z=(-200) m asl) where gravity-driven upward flows could be identified, particularly in the southern foreland of Villány Hills. However, existence of the overpressured flow system is strongly probable below about z=(-2000) m asl in the eastern half of the study area as well, particularly in the sedimentary basin parts, i.e. in the southeastern continuation of the Drava Basin (in the south of Villány Hills) and in the southwestern corner of the Great Hungarian Plain (in the east of Mecsek Mts. and Villány Hills). This theory can be supported by the proven presence of overpressured flow systems within the northwestern part of the Drava Basin (described by the present project) and in the Great Hungarian Plain's sub-basins (Tóth and Almási 2001; Mádl-Szőnyi and Tóth 2009; Czauner and Mádl-Szőnyi 2013; etc.) based on the geological similarity of these areas.

Comparison of the study area with the Buda Thermal Karst system (Budapest, Hungary)

Boundary zone of the dominantly unconfined Mesozoic carbonates of the Buda Thermal Karst (BTK) and confined deep carbonates within the basement of the Hungarian Paleogene Basin (HPB) could serve as analogy for the southern foreland of Villány Hills (sfVH). The first complex and measured data based hydraulic evaluation of the BTK area was carried out by Erhardt et al. (2017). As a result, gravitational flow systems, hydraulic continuity, and the modifying effects of aquitard units and faults were identified in the karst area similarly to the sfVH. The significant conclusion according to which the thicker siliciclastic cover causes greater vertical pressure gradient thus upward flow within the underlying carbonate aquifer was drawn in the BTK and sfVH as well. Also the evolution history of the karst area and the caves (uplift initiated evolution of the topography-driven groundwater flow related to the inversion of the Pannonian Basin) and demonstrability of the basinal fluid component with hydrocarbon traces show similarities in the two areas (Poros et al. 2012; Mádl-Szőnyi et al. 2018). However, a significant difference can also be determined between the confined deep carbonates of the sfVH-Drava Basin system and the BTK-HPB system. Namely, in the former normal and overpressured conditions were detected with lateral and upward flows, while in the latter normal and underpressured conditions were identified by Mádl-Szőnyi et al. (2019). They explained the underpressure by erosional disequilibrium reflecting geologically transient flow field, and by the flow impeding effect of the aquitard units within the siliciclastic confining layers. On the other hand, in the Drava Basin overpressure could be generated by the interaction of (i) tectonic compression and vertical compaction as fluid flow driving forces, and (ii) the flow impeding effect of the aquitard units within the siliciclastic confining layers, similarly to the S-SE sub-basins of the Great Hungarian Plain (Tóth and Almási 2001; Czauner and Mádl-Szőnyi 2013). Actually, effect of tectonic compression on the flow pattern could be the most significant in the Drava Basin compared to the other sub-basins of the Pannonian Basin, since inversion of the basin first started in the southwestern part of it at the end of the Miocene (Bada et al. 2007). Afterwards, the onset of inversion gradually migrated towards more internal parts of the system resulting in transpression (local shortening) in the western parts of the Great Hungarian Plain, while in the eastern parts transtension (local extension) still dominates. As a consequence of the difference between the pressure conditions significant distinction can be made regarding the origin of the basinal fluid components of the two areas as well. In case of the BTK-HPB system, underpressure of the confined carbonates drives the dominantly *basinal fluids of the confining strata* into the underlying carbonate aquifers via vertical leakage. On the other hand, in the sfVH-Drava Basin system overpressure of the confined carbonates drives the *basinal fluids upward and towards the marginal areas* of the basin.

As a further result, comparison of the study area with the Buda Thermal Karst allows for the definition of key factors, which determine the connection of the marginal carbonate reservoirs with the Pannonian Basin, i.e. (i) tectonic regime and related geographical evolution, (ii) flow impeding effect of the aquitard units within the siliciclastic confining layers. It can be also concluded that the basin evolution, particularly the inversion had a crucial role in the initiation of the flow system of these marginal karst reservoirs and their karstification (cave forming) processes.

Local scale phenomena derived from the regional groundwater flow system evaluation

Based on the evaluation of regional groundwater flow systems, also notable local scale phenomena could be explained particularly in the southern foreland of Villány Hills (sfVH), where hydraulic, karstification, and hydrochemical phenomena could be jointly interpreted. Geologically, the basement built-up by carbonates and is situated in uncovered (i.e., unconfined) position within the Villány Hills, and in a shallow covered (i.e., confined) position within the southern foreland area, then gradually deepens towards the Drava Basin. Hydraulically, this confined or unconfined situation of carbonates proved to be decisive in the evolution of vertical flow directions. During the regional groundwater flow system analysis gravity-driven lateral and upward flow conditions were identified in the area (Fig. 2). Lateral flows could be determined where the carbonate basement outcrops or has a very thin sedimentary cover (p(z) #27, 30, 32 and 34 on Fig. 2), whilst upward flows could be identified where the siliciclastic sediments cover the carbonates in greater thickness (p(z) #26, 28, 29 and 31 on Fig. 2). Since the siliciclastic cover usually has lower hydraulic conductivity than that of the basement carbonates, it can be concluded that the thicker cover causes greater vertical pressure gradient thus upward flow within the underlying aquifer.

Regional groundwater flow system analysis also helps to better understand the cave forming processes. Those caves in the Villány Hills which show the effect of upwelling thermal waters (i.e., hypogene caves) can be found in uncovered carbonates at the margin of the Villány Hills (Nagyharsány, Máriagyüd in Fig. 1) and in uplifted basement blocks (Siklós, Beremend in Fig. 1). According to the hydraulic evaluation, their *formation cannot be related to the present day flow systems* since lateral flow conditions dominate within these areas (p(z) #27 Siklós, #30 Beremend, and #32 Nagyharsány in Fig. 2), moreover they are mostly situated above the water table. Formation of the caves is rather connected to the inversion of the Pannonian Basin where the compressional stress field resulted in the uplift of the Mecsek Mountains, Villány Hills and some basement blocks, which were still covered with sediments. As the recharge through the uncovered hilly ranges and carbonates initiated the gravity-driven flow systems, these waters could meet with compression-driven upwelling basinal thermal waters at the marginal areas of the Villány Hills and uplifted basement blocks. The mixing of different fluids could result in cave formation due to the mixing corrosion phenomena. As the direct recharge through the uncovered hilly ranges became more and more pronounced, the ratio of basinal fluids decreased, the caves were flooded by “fresh” waters and the thermal water related, hypogene cave formation period has stopped, also because some of the caves were disconnected from the water table due to ongoing uplift. This scenario is supported by stable isotope measurements performed during this project from the cave filling waters of the Beremend Cave, which showed similar values to the present day precipitation (Fig. 18., see details later in the text).

Furthermore, within the sfVH there is a special place around Harkány where effects of the presumably existing overpressured flow system, i.e. groundwater flows derived from the Drava Basin could be identified. The geochemical uniqueness of the thermal waters of Harkány was explained by Lorberer and Rónaki (1978) and Liebe and Lorberer (1981) with fluid contribution from the north-northwest. Later, the conceptual model of Csicsák et al. (2008) already indicated an interplay between the Drava Basin and the carbonate suite of Villány, whilst as indicators of basinal fluids $\delta^{18}O$ and δD values, ^{14}C ages, and organic compounds (hydrocarbon traces) were designated in the thermal waters of Harkány. The present study proved both presumed flow directions based on the analyses of measured data. Namely, on p(z) profile #26 (Fig. 2) upward flow direction, whilst on hydraulic cross section #2 (Fig. 4) a supposedly fault-related positive potential anomaly (i.e., elevated h values) can be observed. This flow pattern could be determined within the gravitational flow system since data were available only above z=(-200) m asl, however support and contribution of the overpressured system can be presumed at least along the fault zone, which approaches the land surface within the elevated carbonate basement block. Further evidence could be that in the sfVH the Harkány area is located the nearest to the Drava Basin’s margin which forms a bay around Harkány.

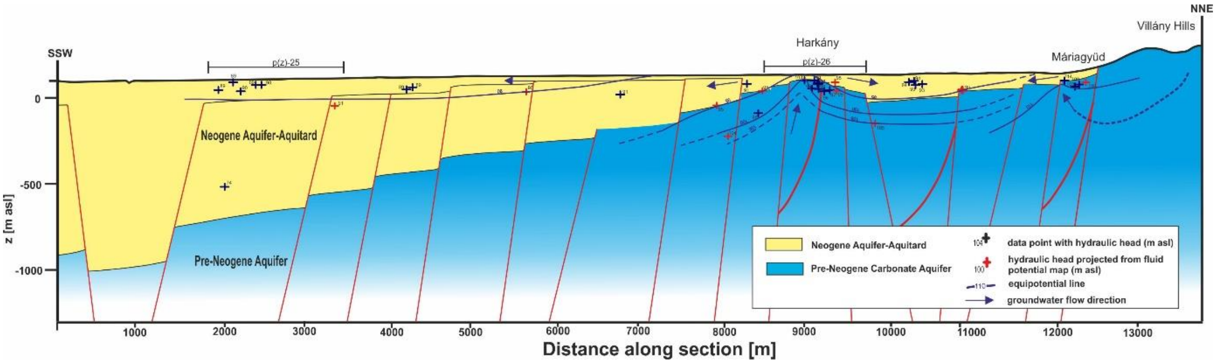

Fig. 4. Hydraulic cross section showing the flow field in the southern foreland of the Villány Hills through Harkány

The effect of the basin can be also seen on the temperatures of the thermal waters in Harkány (62°C), which is the highest groundwater temperature within the sfVH (maximum temperature value of the other wells in p(z) profiles # 27-30 and #32 is 33°C). In addition, based on the hydrochemical analysis groundwaters of Harkány of Ca-Na, HCO_3-Cl-SO_4 facies and their higher chloride and sodium content represent a transition between the karst waters of Ca-Mg, HCO_3 facies and the deep basinal fluids of Na-K, Cl-SO_4-HCO_3 facies (Fig. 5). In other words, it is a mixed water with a deep basin component as well. However, based on the gas chromatographic measurements performed during this project organic components were not detected in the waters of the Harkány Spa wells, therefore the findings of Csicsák et al. (2008) could not be confirmed. Among the radionuclides, radium shows the highest concentration in the area of Harkány (Table 1). This can be also an indication (a natural tracer) of basinal fluid contribution in form of strongly reduced waters similarly to other marginal karst areas (BTK – Erőss et al. 2012b; SW foreland of Bükk– Erőss et al. 2015).

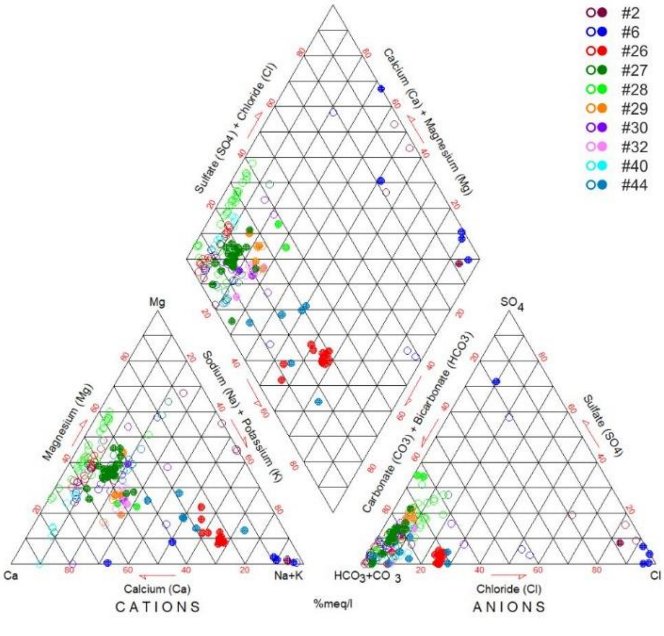

Fig. 5 Piper diagram of water samples from the areas of p(z) profiles presented in Fig. 2 (#2, 6, 26-30, 32, 40, 44). Full circles: water samples from the Pre-Neogene Aquifer, empty circles: water samples from the Neogene Aquifer-Aquitard group. Red symbols indicated the area of Harkány (p(z) area #26)

Table 1. Results of the radionuclide measurements

Sampling point	***ORP [mV]***	***^{222}Rn [Bq/l]***	***^{222}Rn error [Bq/l]***	***^{226}Ra [mBq/l]***	***^{226}Ra error [mBq/l]***	***^{234}U+ ^{238}U [mBq/l]***	***^{234}U+ ^{238}U error [mBq/l]***
Beremend cave1	-	<5 Bq/l	1	21	5	-	-
Beremend cave2	118	<5 Bq/l	0	12	3	16	4
Beremend1	7	21	3	158	12	30	6
Bóly1	-	<5 Bq/l	1	<5 mBq/l	2	12	3
Büdöstapolca1	-60	25	3	60	8	27	5
Büdöstapolca2	-224	43	4	29	5	173	13
Egyházasharaszti1	-34	11	2	100	10	25	5
Egyházasharaszti2	-50	18	2	<5 mBq/l	2	16	4
Harkány1	-354	43	5	140	12	46	7
Harkány2	-345	25	3	69	8	66	8
Harkány3	-347	27	3	196	14	45	7
Harkány4	-365	29	3	230	15	54	7
Kistapolca lake	-140	14	2	25	5	15	4
Kistapolca1	-28	-	-	30	6	18	4
Kistapolca2	-111	14	2	25	5	50	7
Márfa1	612	<5 Bq/l	2	13	4	93	10
Matty1	-46	29	3	76	9	33	6
Siklós1	569	21	3	9	3	73	9
Villány1	150	5	2	5	2	45	7
Villány2	138	<5 Bq/l	1	<5 mBq/l	2	42	6
Villánykövesd1	174	<5 Bq/l	1	6	3	22	5

Regarding the north-northwestern origin of groundwaters proposed by Lorberer and Rónaki (1978) and Liebe and Lorberer (1981), it can be supported by the flow directions on fluid-potential maps #1-4 (Fig. 6) down to about z=(-1000) m asl. Regarding the karst water component, Villány Hills represent the primary recharge area, but regional scale flows derived from the Mecsek Mountains could also reach the Harkány area based on the flow directions on fluid-potential maps #1-4 (Fig. 6).

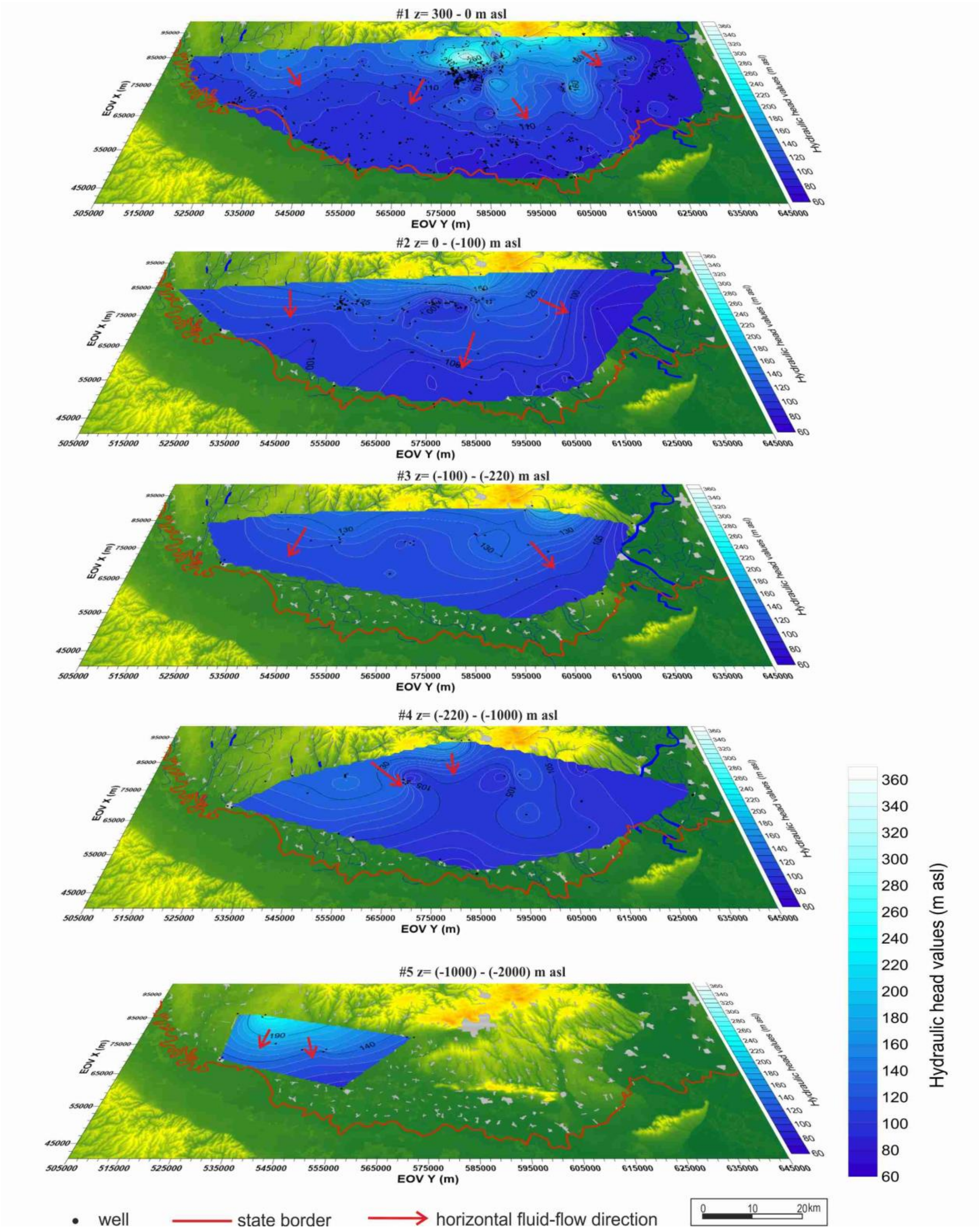

Fig. 6 Tomographic fluid-potential maps of the regional study area. Background: topographical map.

Finally, as a result of the complex measured data based analysis of the Harkány area, a generalized flow field model could be established in Fig. 7 based on hydraulic cross section on Fig. 4 and the related phenomena.

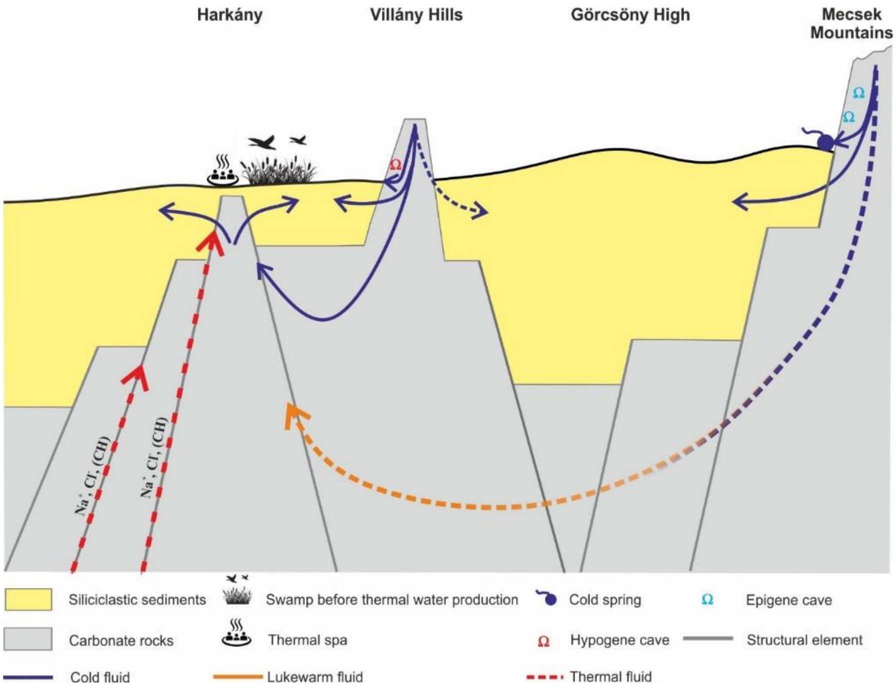

Fig. 7. Generalized flow field model of the Harkány area based on the measured data based hydraulic study

Results of studies related to cave formation in the local study areas: Kistapolca and Beremend

Beside the conclusions of the regional hydraulic study regarding the cave formation in the area, local scale studies were performed in order to evaluate the recent cave forming processes in detail. MSc students were involved in these measurements as well, resulted in an MSc thesis (Godányi 2018).

The karst springs in the little village of Kistapolca (Fig. 8) are captured, but the only naturally outflowing springs in the Villány karst area nowadays. Springs are the surface manifestations of groundwater flow systems (Tóth 1971). Their appearance and physico-chemical parameters represent the flow system as a whole and report on the rock-water interactions along the flow path. Nowadays most of the springs lost their natural character, because of anthropogenic influence, i.e. they are captured and intensely used. Therefore the rather rare natural springs are of great importance to study the flow system of an area (Groves 2007). Moreover, natural springs usually maintain groundwater-dependent ecosystems in forms of creeks, lakes, wetlands, etc., which also emphasizes their value.

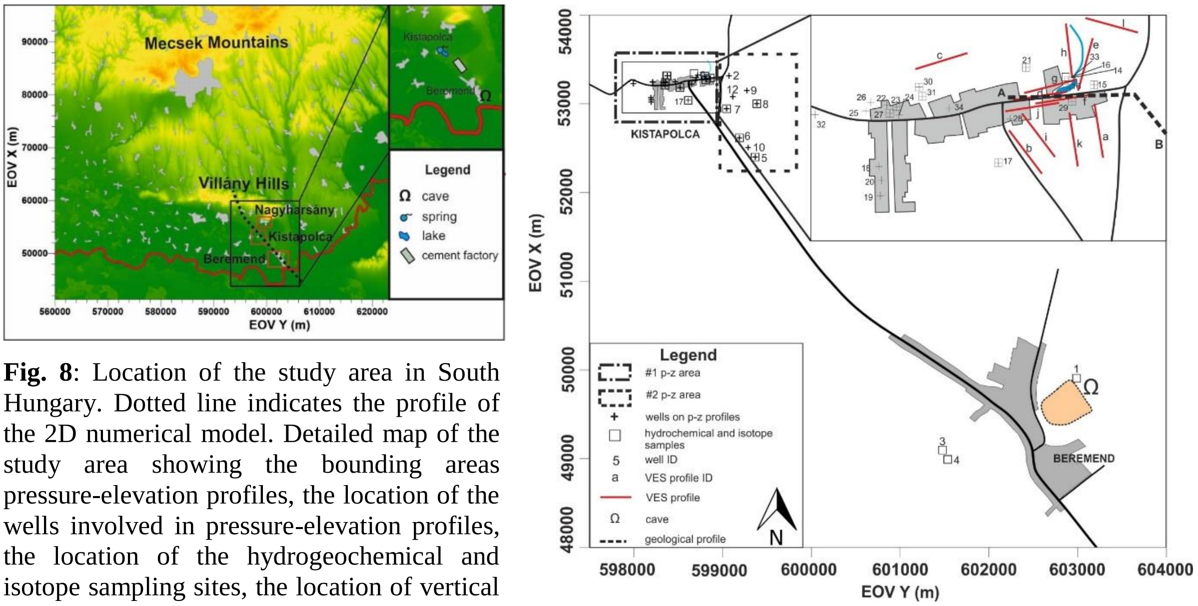

Fig. 8: Location of the study area in South Hungary. Dotted line indicates the profile of the 2D numerical model. Detailed map of the study area showing the bounding areas pressure-elevation profiles, the location of the wells involved in pressure-elevation profiles, the location of the hydrogeochemical and isotope sampling sites, the location of vertical electrical sounding (VES) profiles, the Beremend Cave and the location of A-B profile

The temperature of the springs in Kistapolca are about 22-24 °C, well above the annual mean temperature (11°C). Previous studies by Rónaki et al. (1967), Lorberer and Rónaki (1978) suggested that the lukewarm temperature is the result of mixing between thermal and cold karst waters. To investigate this question is of great importance since only 6 km away from Kistapolca can be found one of the most protected hypogene caves of Hungary, the Beremend Cave (Fig. 8), which still have underwater passages. The morphology and the minerals decorating the cave walls indicate the effect of thermal waters (Takács-Bolner 1985, Takács-Bolner in Székely 2003a, Vigassy et al. 2010). The question can be raised whether the lukewarm spring water temperature is the result of mixing, and whether this mixing together with the resulting cave formation is active even today, in the recent hydrogeological settings. Since understanding of karstification requires clarifying the groundwater flow conditions in an area (Klimchouk 2007, 2012), these questions can be answered by studying the only natural lukewarm springs and its hydrogeological background in the vicinity, i.e. in Kistapolca.

The springs in Kistapolca also maintain a lake, which has been dried up several times in the last few years. Flow system understanding can also help to explore the reason of these dry periods and constrain the mechanism, as groundwater-dependent ecosystems maintained by the different order flow system's discharge react differently to climatic changes or anthropogenic effects (Havril et al. 2018).

The aim of this local study was to explore the hydrogeological characteristics of the Kistapolca area in order to (1) understand the origin of the elevated temperature; and (2) to analyse its relevance to hypogene cave formation; and (3) to evaluate the vulnerability of the lake in Kistapolca as a groundwater-dependent ecosystem maintained by the springs. The paper entitled "*Groundwater flow system understanding of the lukewarm springs in Kistapolca (South Hungary) and its relevance to hypogene cave formation*" by Anita Erőss, Katalin Csondor, György Czuppon, József Dezső and Imre Müller was published in the *Environmental Earth Sciences* journal, which summarizes these results (submitted in June 2019, accepted for publication on 17. February 2020; available online 5. March 2020; IF (in 2018): 1.871; the paper is published with full open access: https://link.springer.com/article/10.1007/s12665-020-8870-3). Here only the essence of the study is highlighted.

To explore the hydrogeological characteristics of the Kistapolca area different techniques, including geophysical, geochemical and stable isotope measurements, evaluation of continuous-time series of water level, temperature, and electrical conductivity data, and hydraulic evaluation of vertical flow conditions by pressure-elevation profiles were combined. Since the appearance of the springs in Kistapolca is thought to be connected to an uplifted carbonate basement block, geophysical methods (Vertical Electrical Soundings (VES)) were applied first to investigate this question in detail (Fig. 9).

Pressure-elevation profiles were constructed based on archival and recently measured well data to investigate the groundwater flow direction, i.e. prove the upward flow conditions in the area, which may explain the existence of the springs. The main ions and stable isotopes report on the geochemistry and rock-water interactions, the continuous-time series of water level, temperature and electrical conductivity data of the springwater reflect the dynamics of the flow system. Finally, the possibility of that scenario, in which the Villány Hills infiltrated meteoric water can be heated up and discharge in Kistapolca with elevated temperature, was tested by numerical simulation.

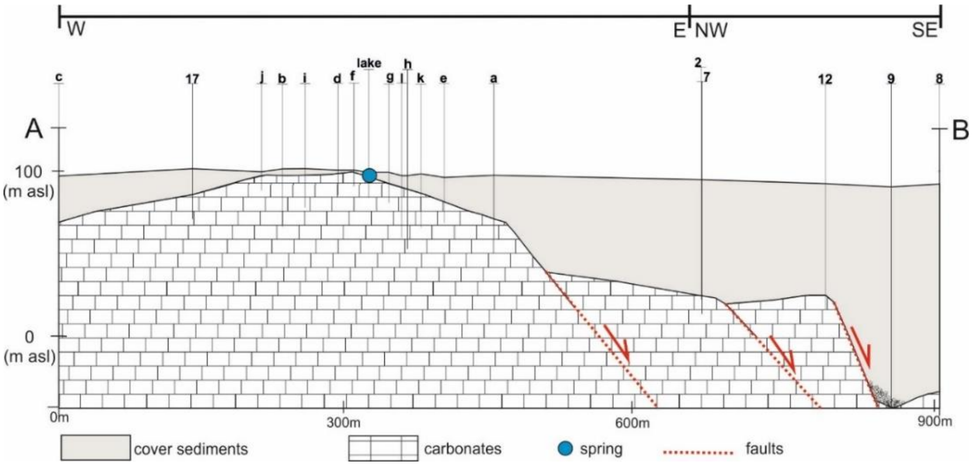

Fig. 9: A-B simplified geological profile through Kistapolca (see location on Fig. 8) based on the results of vertical electrical soundings and borehole stratigraphic data showing the situation of the carbonate basement under the sediment cover.

Based on the VES measurements and stratigraphic data of boreholes, the position of the carbonate basement block was refined in Kistapolca compared to the results of the geophysical survey in the 1960s (Rónaki et al. 1967). The results show, that the basement situated in the highest position, i.e. closest to the surface in the close vicinity of the lake, which explains the existence of the springs (Fig. 9). The pressure-elevation profiles showed greater than the hydrostatic (γ_{dyn}=10.45 MPa km^{-1}), i.e., super-hydrostatic vertical pressure gradient in Kistapolca, which refers to the upward vertical flow direction. This also supports and explains the existence of springs. This method as part of the gravity-driven regional groundwater flow (GDRGF) concept's methodology (Tóth 2009) was previously commonly applied only in sedimentary basins. However, recent investigations (Mádl-Szőnyi and Tóth 2015; Erhardt et al. 2017) highlighted its applicability in karst areas. Pressure-elevation profiles were applied by Erhardt et al. (2017) in a similar, marginal karst area (Buda Thermal Karst), where the springs and discharge areas were verified by this method. The results in Kistapolca further strengthen the applicability of flow system analysis methodology in karst areas.

The upward flow and the discharge effect of karst waters into the cover sediments are also evident from the isotopic composition of dug wells and springwater, that are characterized by low hydrogen and oxygen isotope values ($\delta D \approx -77.9‰$, and $\delta^{18}O \approx -10.9‰$). This is reflected also by the bicarbonate and calcium dominance in these wells. The Ca+Mg/HCO_3 ratio is close to 1, implying also carbonate dissolution-related origin of these waters (Zaidi et al. 2015). The visually recognizable connection between precipitation events and the water level increase on the continuous time series indicates the dynamic response of the thick carbonate reservoir on precipitation in the form of regional water level increase (Fig. 10). The stability of electrical conductivity and temperature over time in the mainspring, however, indicates that local infiltration, local flow system have no or little direct, immediate effect on the spring's parameters. This is also in accordance with the lowest measured isotopic values of the spring and the wells in Kistapolca, as it is discussed above. This suggests the influence of higher-order flow systems rather than the local recharge conditions in the wells and springs of Kistapolca, which is also supported by the numerical simulation (Fig. 11).

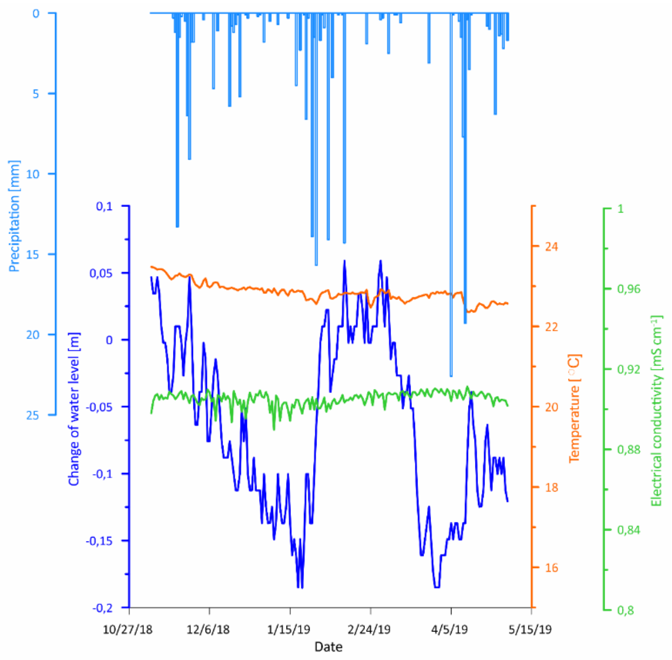

Fig. 10. Continuously recorded water level change, electric conductivity and temperature data of the mainspring (lines) and the daily precipitations in Nagyharsány (bars).

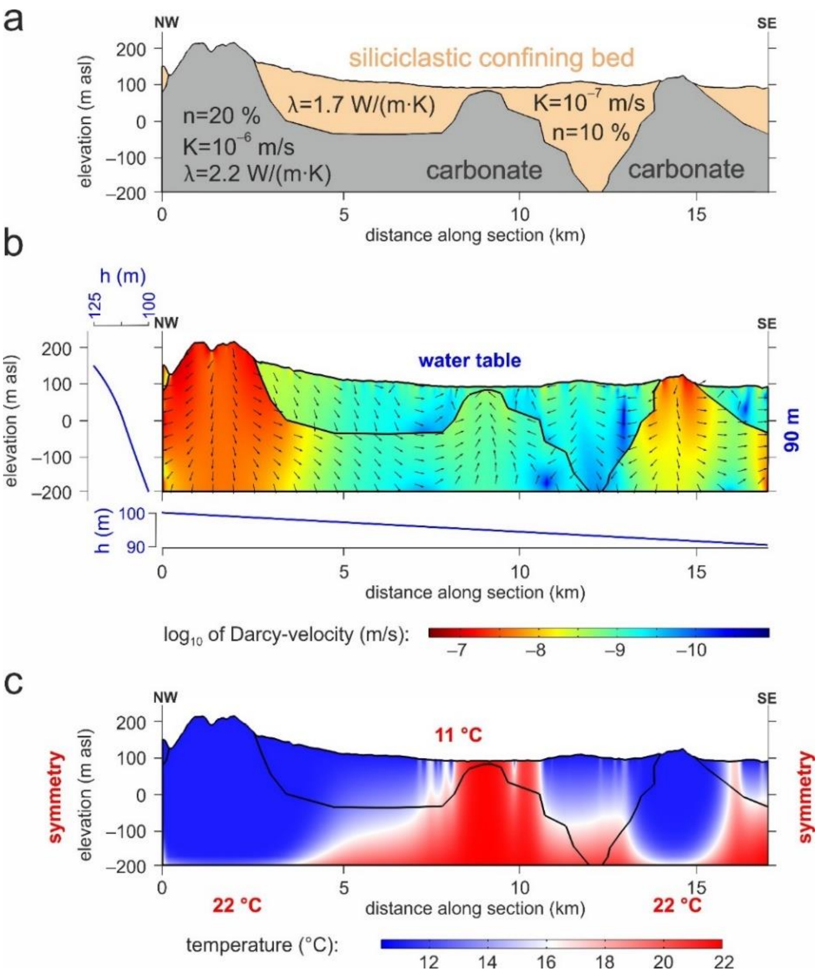

Fig.11: Numerical simulation results of groundwater flow and heat transport. a) Model geometry indicating parameters of siliciclastic confining bed and carbonate: K - hydraulic conductivity, n - porosity, λ - thermal conductivity. b) The intensity of groundwater flow with the normalized flow vectors and boundary conditions. c) Temperature distribution and the applied boundary conditions of heat transport

The small scale, 5-10 cm water level changes recorded at the mainspring in Kistapolca might be the effect of the water abstraction of the nearby cement factory and the waterworks in the village, which have together yearly about 120-145-190x10^6 m^3 abstracted water amount in the last three years, showing an increasing trend (data provided by the General Directorate of Water). Therefore, the groundwater-dependent ecosystem of the lake fed by the spring can rather suffer from this anthropogenic effect. Climate change i.e. reduced recharge can further strengthen this effect, as large groundwater abstraction superimposed on regional water level decrease may result in longer and more common dry periods of the lake, as it was theoretically investigated by Havril et al. (2018).

In summary, the combination of geophysical, geochemical, hydraulic methods enabled to deliver explanations to the existence of the springs, and multiple pieces of evidence that the spring is fed by a higher-order - possibly intermediate - flow system without or limited influence of local flow system. These findings are also supported by numerical simulation. The results regarding the present day cave formation in the area indicate that mixing of different waters is not an active process today. In addition, the vulnerability of the groundwater-dependent lake ecosystem was evaluated as well.

On site measurements in the Beremend Cave

Since its discovery in 1984 the Beremend Cave is one of the most protected caves in Hungary. Its morphology and special mineral assemblage show the effects of thermal waters (Takács-Bolner 1985; Takács-Bolner in Székely 2003a; Vigassy et al. 2010), thus belong to the unique group of hypogene caves in Hungary. Its hydrogeological relevance is that the cave reaches the karst watertable offering a window to the aquifer, studying its actual processes, including cave forming, mineral precipitation. The cave is situated in middle of an active limestone quarry (Fig. 12). Because of its uniqueness and the operating quarry, its research is strongly limited. It took 9 months until we got permission for research allowing maximum four visits per year.

Fig.12: Aerial photo of the protected limestone block of the Beremend cave within the quarry made with DJI Spark drone

To acquire better understanding about the actual processes of the hypogene cave of Beremend, a two-year-long experiment (*originally not planned in the proposal*) has been started in March 2018. 24 limestone tablets - made of the host rock of the cave - were installed in the cave in three positions: under the watertable, at the water/air interface and in the air (Fig. 13). One tablet from each location is planned to be removed in every three months and the changes of the rock surfaces are planned to be investigated by scanning electron microscopy. Rock surfaces were recorded also prior the experiment for comparison.

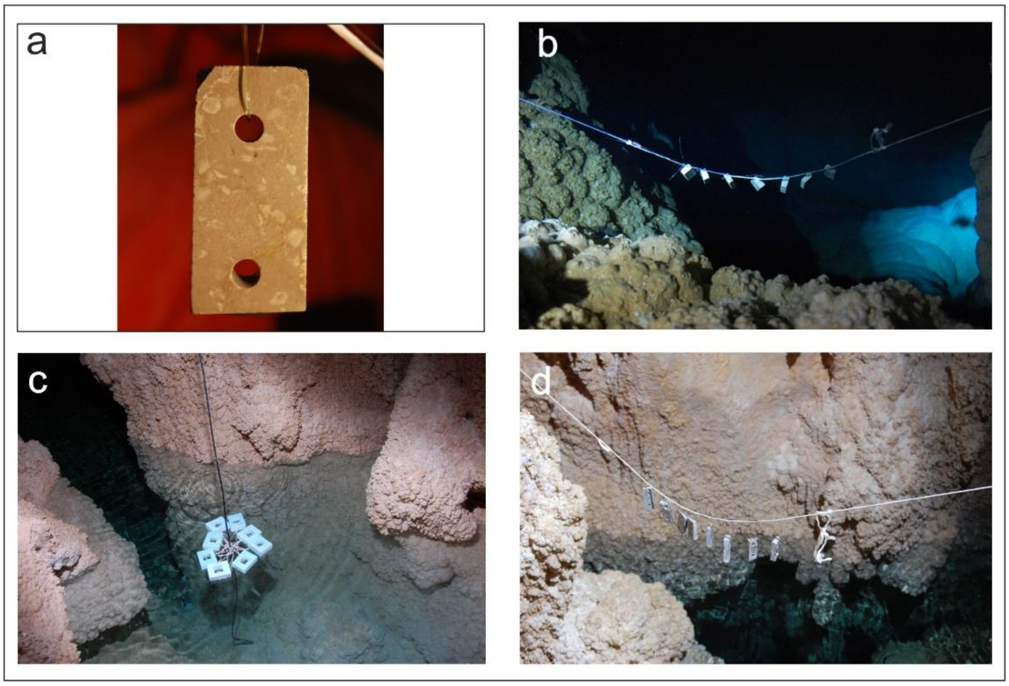

Fig.13: a: the host rock samples (2x5 cm in dimension); b: limestone tablets under the watertable c: limestone tablets on the water/air interface d: limestone tablets in the air

There are modifications related to obstacles in the course of the experiment. Due to both the a priori limited access and the active quarrying in the close vicinity of the cave, only 5 sets of samples were removed as yet (01.06.2018, 19.10.2018, 03.05.2019, 06.09.2019, 28.02.2020). Additionally, due to the extreme low water levels (see details later), the samples at the water/air interface and under the water table had to be replaced. As we did not have access to the same scanning electron microscopy after the start of the experiment and the pictures taken by an another instrument cannot be compared with the original ones, all the samples will be investigated at once at the end of the experiment with the originally used scanning electron microscope. The experiment will continue until the end of 2020.

In addition, to better understand the processes in the cave and enhance the experiment, continuous measuring devices were installed in the cave air and into the water. The device in the water recorded the water level, water temperature, electrical conductivity. These time series were enhanced by daily precipitation data from Nagyharsány-Konkoly, which were acquired through cooperation with József Dezső (University of Pécs) (Fig. 14).

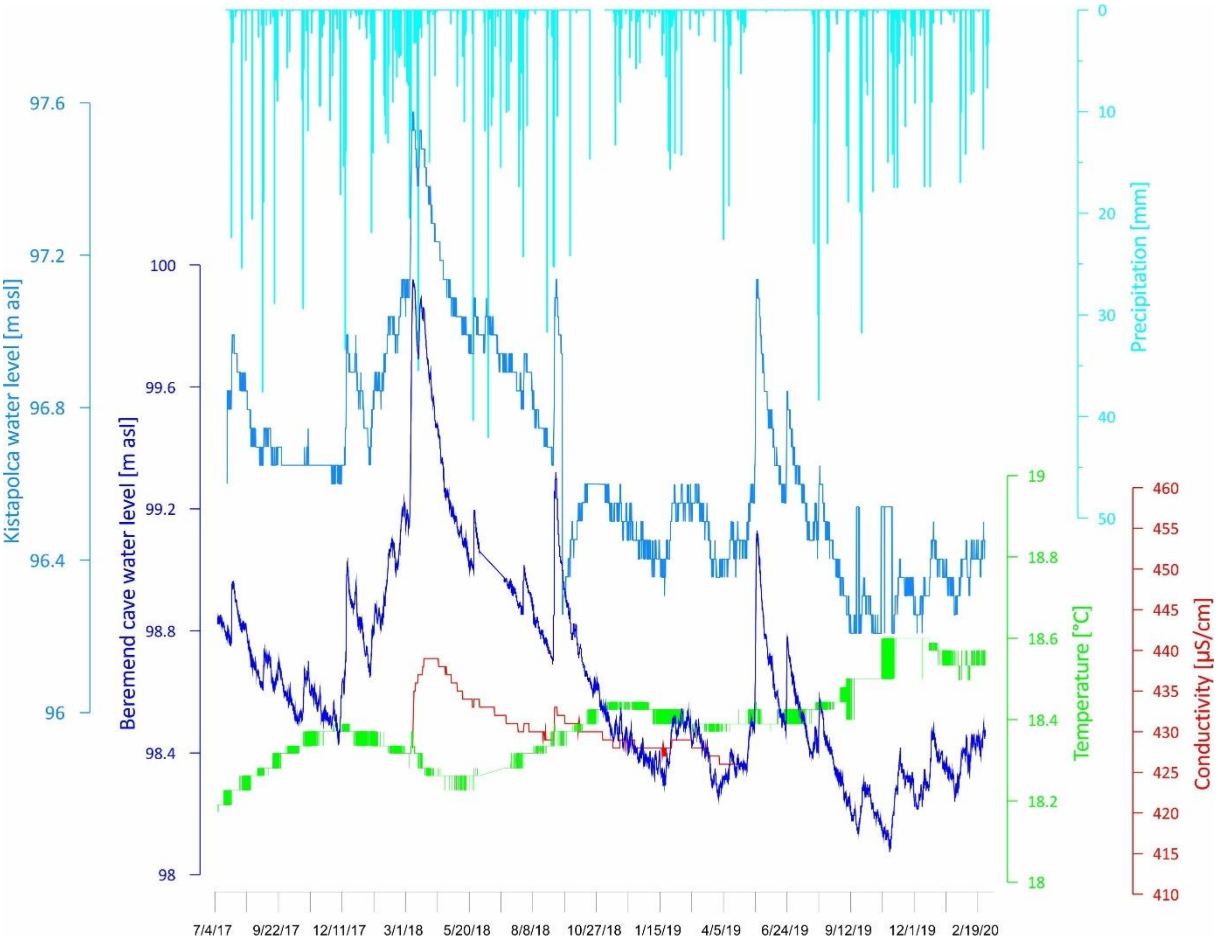

Fig.14: Continuously recorded temperature, electrical conductivity and water level data in the Beremend Cave. Additionally, continuously recorded water level data of the main spring in Kistapolca is displayed.

Based on the continuous data, no significant changes in temperature and conductivity values are observed. Temperature of the cave filling water varies between 18.2 and 18.5 °C, the electrical conductivity range is 425-439 µS/cm. Stability of these parameters indicates that local infiltration has no significant effect on the water's temperature and electrical conductivity. However, there is a clear connection between precipitation and water level changes which refers to the dynamic response of the karst reservoir on precipitation. Similar observations were made in the Kistapolca spring (see Fig. 10). The water level changes in the cave are moving together with the Kistapolca spring water level data, which means that they are well connected, belong to the same system, i.e. there is no relevant hydraulic barrier between the two measuring points. Thus, studying the easier accessible spring in Kistapolca is also relevant related to the processes in the aquifer in the vicinity of the cave.

In the cave extreme water level decreases were observed during the visits in the last years (Fig. 14, Fig. 15). In the same periods in Kistapolca the overflow of the spring stopped, and the lake dried up. In the last 3 years the abstracted water volume of the cement factory and the waterworks in Kistapolca has been continuously increased (120 →145 →190 × 10^6 m^3; data provided by the General Directorate of Water), which may cause this low-water periods. However, to investigate this question in detail, including the possible long term decrease also in precipitation amount, data of all available monitoring wells in the region and the precipitation will be investigated for longer time interval (5-10-20 years depending on data availability) in cooperation with József Dezső (University of Pécs), as a new research direction based on the findings of the project.

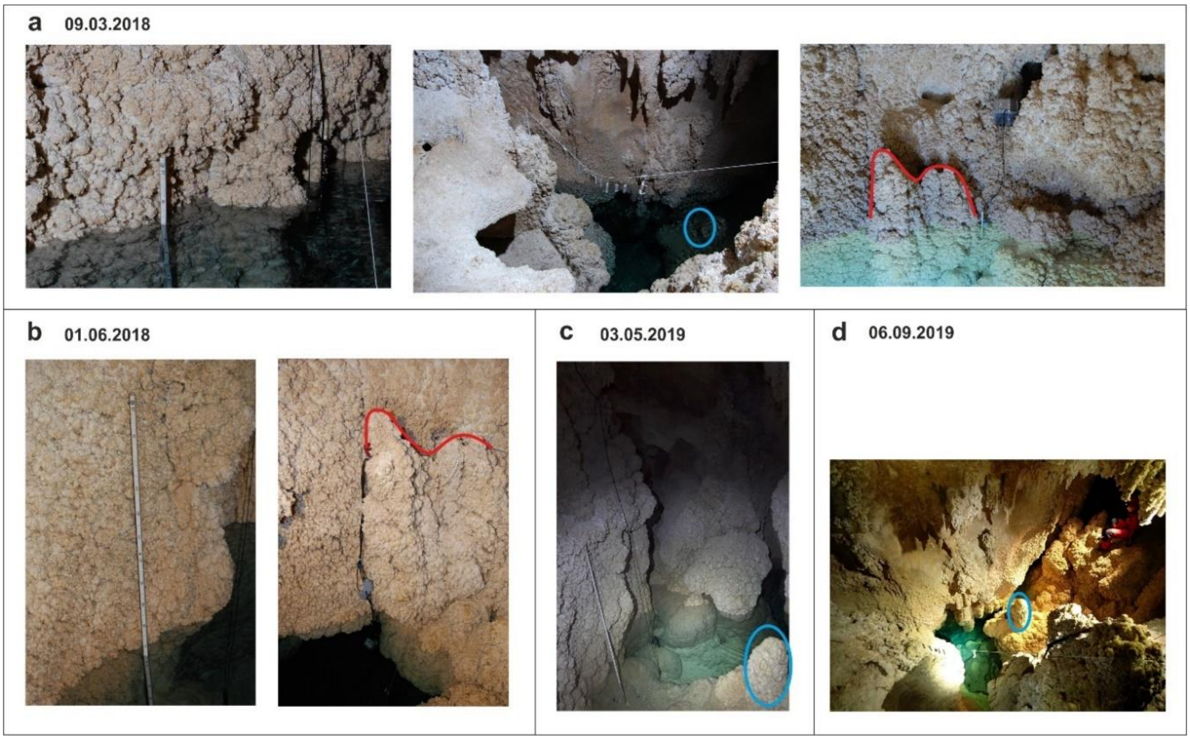

Fig. 15: a: normal (09.03.2018) 99.869 m asl, and extremely low (b: 01.06.2018 water level 99.061 m asl; c: 03.05.2019, water level 98.359 m asl; d: 06.09.2019, water level: 98.242) water levels observed in the cave. The maximum water level difference is 1.627 m.

For experimental purposes a continuously measuring device was developed by Prof. Heinz Surbeck, which is able to continuously record the gas saturation, total dissolved oxygen concentration, oxygen partial pressure, electrical conductivity, temperature and CO_2 concentration in the water and relative humidity, atmospheric pressure, temperature and oxygen content in the air (Fig. 16). These parameters would enable to better understand the precipitation-dissolution processes and may serve as input parameters for geochemical modelling. In cave conditions (high relative humidity, lack of electricity) this was the first time in Hungary that these parameters were continuously recorded. The instrument was operated by a solar panel from the surface, so there was no need to place a battery in the cave, through which the extreme protection of the cave was fulfilled. With some technical interrupts the instrument operated between 09.03.2018 and 18.10.2018. Unfortunately, the sensor placed into the water and included the most important parameter, the CO_2 concentration did not work despite all efforts. We must admit, that the limited allowed visits were not ideal for testing this instrument in this cave. Further work and testing periods are needed and are in progress not related to the Villány karst area.

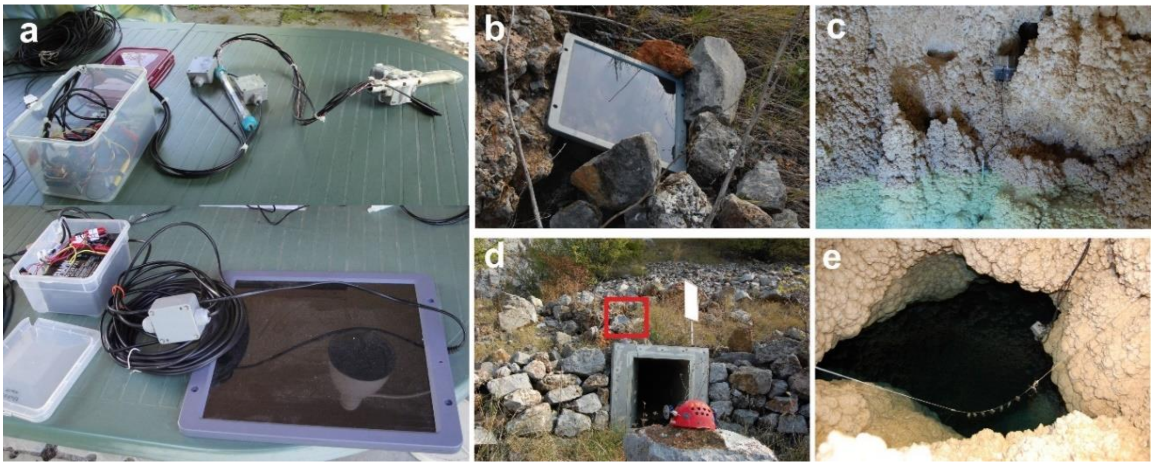

Fig. 16: a: the instrument with the in air and in water sensors; b-d: location of the solar panel on the surface near the cave entrance; c-e: the location of the instrument in the cave

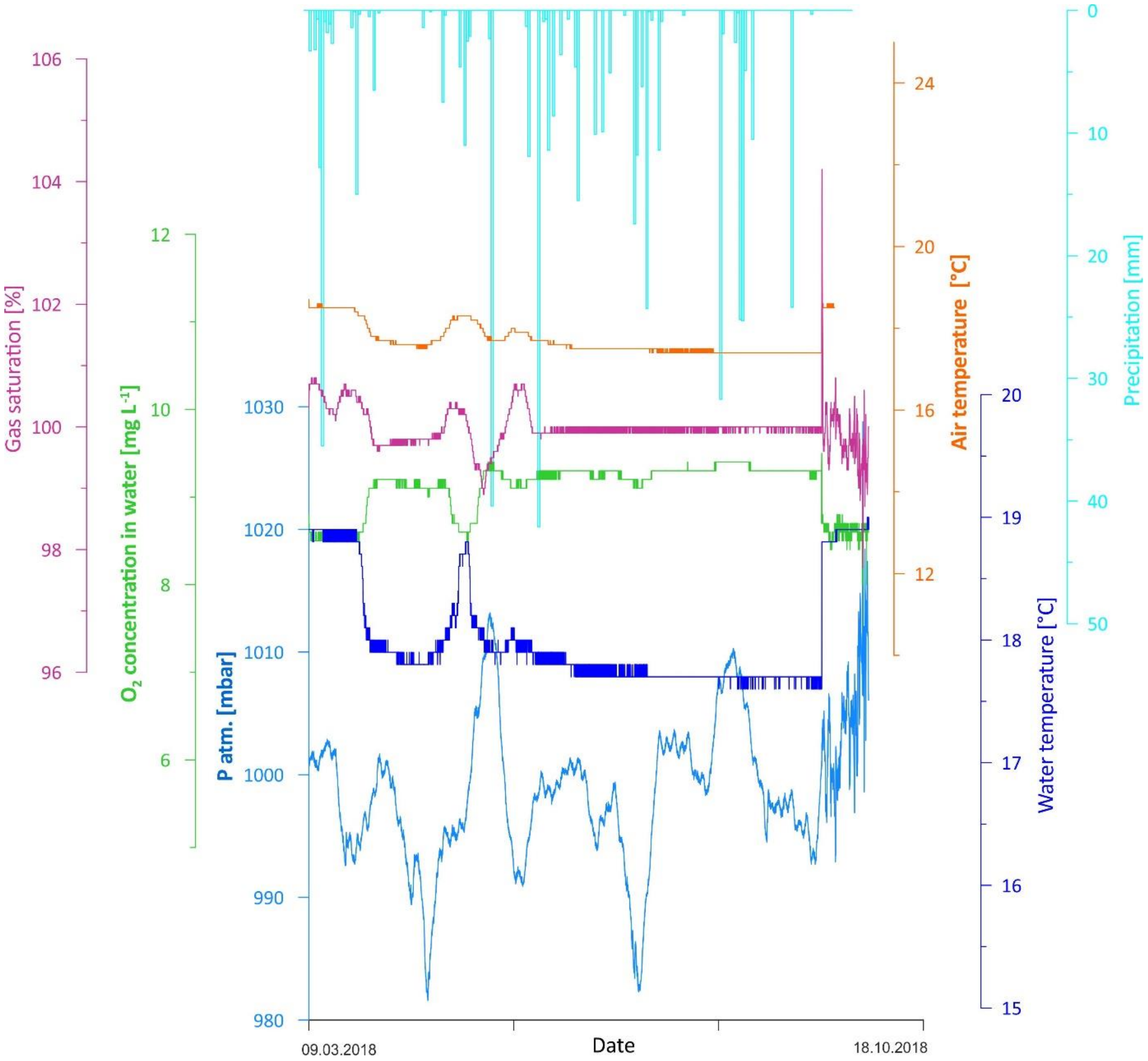

Fig. 17. Continuously measured parameters in the Beremend Cave by a newly developed measurement device – results of the first test period

In every cave visit the physico-chemical parameters were recorded on site to compare the data with the continuous measurements. Additionally, water samples were collected for chemical and stable isotope analyses (Table 2). Since water chemistry data are rather rare or include very few parameters from the cave, these analyses provide significant contribution to the scientific database related to this cave.

Table 2. Results of on site and chemical analyses of water samples from the Beremend Cave

#	Date	Temp. [°C]	EC [µS/cm]	pH	DO [mg L^{-1}]	DO [%]	ORP [mV]	*δD*	*δ18O*	*HCO_3 [mg L^{-1}]*	*Ca [mg L^{-1}]*	*Mg [mg L^{-1}]*	*Cl [mg L^{-1}]*	*SO_4 [mg L^{-1}]*	*K [mg L^{-1}]*	*Na [mg L^{-1}]*	*TDS [mg L^{-1}]*
1	09.03.2018	18.1	425	8.28	89	8.4	118	- 63.84	- 8.90	210	37	21.9	7.98	35.5	2.59	19.8	366
2	01.06.2018	18.1	428	8.34	103	10.1	37.9	-	-	202	37.6	20.5	9.6	25	3	26	323.7
3	19.10.2018	18.3	423	8.3	90	9.1	50.6	- 63.75	- 8.88	215	58.2	14.8	11.8	30	3	19	351.8
4	03.05.2019	18.3	421	7.31	84	7.9	-	- 64.42	- 8.92	205	39	23	7.4	35	3	21	333.4
5	06.09.2019	18.3	477.6	8.01	77	7.3	192.5	-	-	196	34.4	22	20.9	33	3	19	328.3
6	28.02.2020	18.4	478.5	8.24	49	4.6	140.2	-	-	177	36.3	27.8	14.6	65	3	21	344.7

#	Date	NO_2 (mg/l)	NO_3 (mg/l)	NH_4 (mg/l)	Li (µg/l)	Be (µg/l)	B (µg/l)	Al (µg/l)	Ti (µg/l)	V (µg/l)	Cr (µg/l)	Mn (µg/l)	Fe (µg/l)	Co (µg/l)
1	09.03.2018	<0.1	15.8	<0.1	19.3	n.d.	93.9	20.80	n.d.	0.74	1.07	0.86	<0.005	<0.2
4	03.05.2019	<0.030	16	<0.020	32.76	<1.0	114.61	<10	2.04	<1.0	<1.0	<5.0	<10	<1.0

#	Date	As (µg/l)	Se (µg/l)	Sr (µg/l)	Mo (µg/l)	Cd (µg/l)	Sn (µg/l)	Sb (µg/l)	Ba (µg/l)	Pb (µg/l)	U (µg/l)	Ni (µg/l)	Cu (µg/l)	Zn (µg/l)
1	09.03.2018	<1	<1	563	0.44	n.d.	n.d.	0.11	31.40	<0.2	0.36	<1	1.56	1.73
4	03.05.2019	<1.0	<1.0	609.15	<1.0	<0.4	<0.4	2.86	36.69	1.85	<1.0	<1.0	33.53	10.97

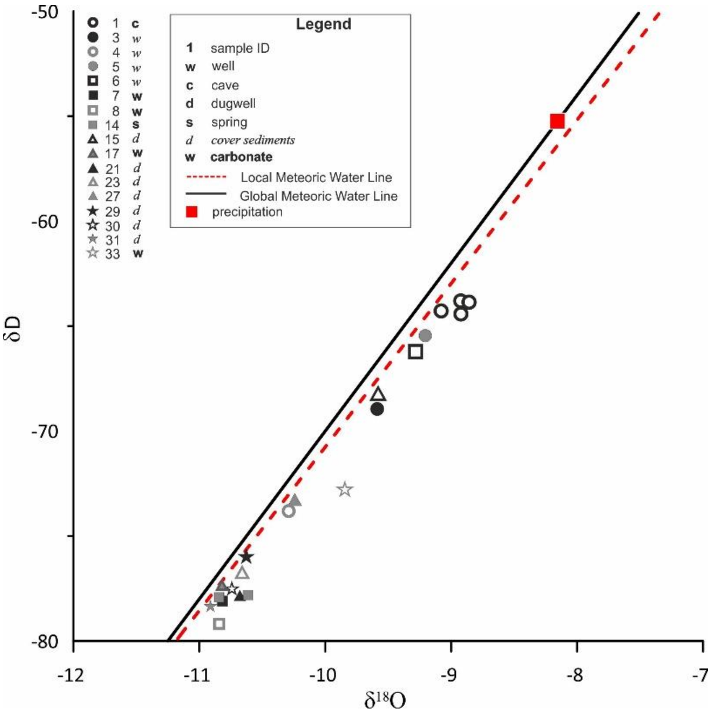

Fig. 18: Isotopic composition of the sampled waters (data in Table 2). GMWL and LMWL indicates the global meteoric water line and the local meteoric water line (Czuppon et al. 2017), respectively. As a reference, the annual amount weighted isotopic composition of precipitation is also indicated (Czuppon et al. 2017).

The isotopic composition of water samples from the Beremend Cave show the most positive values among the samples collected in Beremend and Kistapolca, plotting closest to those that is observed in the recent precipitation implying lack or very limited the effect of higher-order flow systems. The measured $\delta^{18}O$ value of the water in the cave is in accordance with the measurements ($\delta^{18}O$: -8.8 ‰) of Vigassy et al. (2010). The lowest electrical conductivity value (425 µS/cm) together with the isotopic composition closest to the precipitation in the water of Beremend Cave support the scenario showing the effects of recharge and local flow conditions. The numerical simulation is also in good agreement with these observations. It can be concluded, that the karst water present in the Beremend Cave nowadays does not show the effect of deep flow systems and thermal waters implying that significant mixing of deep and local waters might already be inactive as today.

The cave is rather influenced by local processes, which is reflected also by the geochemical composition of the cave filling water. However, these should be evaluated in detail in order to keep the protection of the peculiar thermal water-related mineral assemblages of the cave.

A publication is planned to summarize the results in Beremend after all rock samples are removed from the cave and it is planned to be submitted to Acta Carsologica (IF: 0.756 in 2018).

Summary

During the project hydraulic and geochemical evaluation was completed for a regionally extended area around the Villány Hills, in South Transdanubia. The gravity-driven regional groundwater flow system approach, the concept of hydraulic continuity and the measured data based hydraulic evaluation method were first applied in the area during this project. As a result, the pattern of groundwater flow and its thermal and geochemical characteristics explored the connection between the hitherto separately investigated phenomena and resources, such as caves, springs, karst system, geothermal and hydrocarbon resources. Beside the characterization of regional-scale fluid flow directions, the interpretation of regional flow field allowed for local-scale conclusions regarding 1) the uniqueness of the Harkány thermal waters 2) evolution of the karst system, timing of cave formation and recent cave formation. The study confirmed that in marginal karst areas the evolution of the adjacent sedimentary basin has a crucial role in the evolution of the karst system itself. Local-scale studies in Kistapolca and Beremend provided further details related to the recent processes and cave formation. In case of the Beremend Cave, the first hydrogeology-focused research has been performed during this project. The project results can be directly applied in the exploration and sustainable utilization of groundwater-related resources, such as thermal waters and hydrocarbons in the regionally extended research area.

Scientific potential and the economic outcomes of the project’s results

The study area in South Transdanubia (Fig. 1) is the cradle of thermal water abstraction, since Hungary’s first thermal well was constructed in this region, in Harkány, by Vilmos Zsigmondy in 1866 (Zsigmondy 1873). Currently there are about 50 wells yielding thermal water above 30°C temperature and utilized by spas, agriculture and industry (Fig. 1). Thermal water is exploited for balneological purposes without reinjection and only very few heating systems use this technique (e.g., in Bóly in the studied region). This limits the amount of heat and water that can be utilized from a reservoir causing non-sustainable production (Rybach and Mongillo 2006). Unfavourable effects of excessive thermal water production (e.g., significant drop of hydraulic head, temperature, yield, changes in water chemistry) have already been reported at various parts of the Pannonian Basin (Szanyi and Kovács 2010; Tóth et al. 2016; Rotár-Szalkai and Ó Kovács 2016). The sustainable utilization of the thermal waters and the long term productivity of the reservoirs require the clarification of the path(s) of the groundwater flow as well as the driving forces (Mádl-Szőnyi and Simon 2016). Reinjection and further production possibilities of thermal water can be evaluated and planned only if the regional hydrodynamics are known.

Beside thermal water, considerable hydrocarbon resources are also connected to the sedimentary basin-fill of the Pannonian Basin. In South Transdanubia there are also several hydrocarbon fields connected to the Drava Basin (Kőrössy 1989; Saftic et al. 2003) (Fig. 1). Basin scale groundwater flow systems affect the petroleum systems as a result of geologic agency of groundwater flow (Tóth 1980, 1999). Despite this fact traditionally stratigraphical, tectonical, lithological traps and their combination are only considered. However, hydraulic traps could also have an important role (Tóth 1988). In the deeper sub-basins of the Pannonian Basin a so called overpressured flow system can be observed below the gravitational flow systems (Tóth and Almási 2001). The overpressure is the result of vertical compaction and lateral tectonic compression, and usually drives fluids in upward direction. Where overpressured upward flow meets with gravitational downward flow of recharge areas, regional fluid-potential minimum zones evolve, which can serve as hydraulic traps for hydrocarbons (Tóth 1988). Therefore, regional groundwater flow system analysis could help hydrocarbon exploration as well, as it was demonstrated by Verweij and Simmelink (2002) and Verweij et al. (2012), and also successfully applied in the eastern part of the Pannonian Basin (Czauner and Mádl-Szőnyi 2013).

Based on the regional scale evaluation of fluid flow systems, considerable conclusions could be drawn regarding the geothermal and hydrocarbon potential of the regional study area (Fig. 1). From a hydrodynamic point of view, thermal water or geothermal energy production need less energy investment in the zones of lateral and particularly upward flows. Resources of the overpressured flow system are non-renewable and can be produced only with reinjection with high pressure. However, production from gravity-driven flow system can be sustainable, though long-term productivity usually requires reinjection even into this system with less energy input in the zones of lateral and particularly downward flows (Mádl-Szőnyi and Simon 2016). Based on the present study, favourable areas and depth zones can be determined for production and reinjection as well. P(z) profile #44 demonstrates the pressure conditions of Bóly town where a geothermal cascade system based on a 1500 m deep production well and an injection well in the same reservoir supplies the district heating system. Pressure data represent through-flow conditions (i.e., midline area) down to z=(-1500) m asl, which can be explained by the topographically transient position of the area between the Mecsek Mts. (as recharge area) and the lowlands of Danube and Drava Rivers (as discharge areas). On the other hand, the lateral flow conditions allow of the thermal water production with relatively high yield and reinjection with low pressure. Based on the example of Bóly, areas of through-flow conditions are hydraulically favorable for the installation of neighboring production and injection wells, for instance the areas of p(z) profiles #9, 10, 12, and 14 where through-flow conditions are proven down to z=(-1100), (-800), (-1600), and (-1600) m asl, respectively.

Regarding the hydrocarbon potential, where overpressured and superimposed gravitational upward flows meet with gravitational downward flow of recharge areas, regional fluid-potential minimum zones evolve, which can serve as hydraulic trap for hydrocarbons (Tóth 1988). In the study area a hydraulic entrapment zone was identified in the surroundings of p(z) profiles #2, 3, and 6 around z=(-500) m asl. In other words, this zone represents the upper boundary of vertical hydrocarbon migration, thus entrapment of hydrocarbon is more probable in these areas also in greater depth. As a consequence or evidence, producing hydrocarbon fields are known from this region. Cooperation with Orsolya Győri (TDE ITS Ltd.) during the regional scale hydraulic evaluation of the Drava Basin part of the regional study area in form of an MSc thesis and related scientific paper designate already the economic outcomes of the project's results.

During the project large amount (i.e. thousands) of paper-based archive well documentations found in different repositories were processed and digitized. The acquired digital database and the associated interpretations significantly contribute to the scientific success and competitiveness of the PI's research group. The database and interpretations may provide fundamental information for further academic researches, as well as for the industry sector, such as hydrocarbon and geothermal exploration.

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Erőss Anita

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Anita Erőss

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