

## **Role of Karstic and Basinal Fluids in Porosity Evolution in the Buda Hills, Hungary\***

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Search and Discovery Article #50534 (2012)

Posted January 17, 2012

\*Adapted from poster presentation at AAPG International Convention and Exhibition, Milan, Italy, October 23-26, 2011. Please refer to companion article, Characterization of Fluids and Their Products in a Recent, Fault-Related Hydrothermal System, Case Study from the Buda Thermal Karst, Hungary, [Search and Discovery article #50535 \(2012\)](#)

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### **Abstract**

The Buda Thermal Karst (BTK) is located at the boundary of an uplifted Mesozoic-Tertiary carbonate unit and the adjacent Neogene sedimentary basin. Being a “marginal” area, the BTK serves as the discharge zone of regional fluid flow. This implies that it may receive fluid components from several sources, resulting in a wide range of discharge features, including springs, caves, and mineral precipitates.

Recent- and palaeo-hydrogeology were studied in parallel to understand the effects of palaeo-fluid migration on the recent hydrogeological setting and the distribution and development of the extensive, still active hypogene cave-systems in the BTK. On the basis of geochemical analyses of recent waters, petrography and fluid inclusion analyses of palaeo-precipitates, three types of fluids from different sources were distinguished: (i) regional karst water, (ii) local karst water, and (iii) basinal fluid. Based on the HC indications detected in both the palaeo- and the recent waters and on the changes of mineral assemblages it is proposed that the basinal contribution had been continuous from the late Middle Miocene on. In addition, an increase in dissolution at the expense of precipitation from the Miocene to present suggests an evolving groundwater system in which the proportion of karst waters increased at the expense of the basinal component. Along with mixing corrosion, also aggressive gases (H<sub>2</sub>S, CO<sub>2</sub>, CH<sub>4</sub>) related to hydrocarbon accumulations in the basin have been efficient dissolving agents. The timing of the initiation of basinal fluid migration to the Buda

Hills was estimated by structural geological considerations and fluid inclusion data. Compressional tectonics is believed to have been maintaining flow from the basin side up to now.

In addition to the effects of different fluids on the formation of caves and cave minerals microbial activity has had also significant influence (as proved in the case of the recent spring caves).

Microbial activity is also reflected by extremely negative  $\delta^{34}\text{S}$  values (down to -32‰) measured in gypsum samples. Nevertheless, wide range of sulphur isotopic values revealed also the multiple sources of sulphur, i.e. basinal  $\text{H}_2\text{S}$  (~10 ‰), marine evaporite sulphate from the nearby basin (20-30‰) and sulphur compounds of the host rocks (~6 ‰).

Porosity evolution of the BTK has been most likely influenced by complex hydrogeological systems of various fluid components and affected also by microbial activity.

### **Selected References**

Kovacs-Palffy, P., and M. Foldvari, 2004, Mineralogy of the travertines in NE Transdanubia (Hungary): *Foldtani Kozlony*, v. 134/4, p. 563-587.

Muller, P., and I. Magyar, 2008, The Pannonian deposits of the Buda Mountains: *Foldtani Kozlony*, v. 138/4, p. 345-356.

Poros, Z., A. Eroess, J. Madl-Szoenyi, A. Mindszenty, F. Molnar, P. Ronchi, and A.E. Csoma, 2010, Mixing of karstic and basinal fluids affecting hypogene cave formation and mineralization in the Buda Thermal Karst, Hungary *in* L. Zaharia, A. Kis, B. Topa, G. Papp, and T.G. Weiszberg, (eds.), IMA 2010; 20<sup>th</sup> general meeting of the International Mineralogical Association: *Acta MineralogicaPetrographica Abstract Series*, v. 6, p. 465.

Scheuer, G., and F. Schweitzer, 1974, New considerations on the formation of the freshwater limestone series in the neighborhood of the Buda Mountains: *Geographical Review*, v. 22/2, p. 113-134.

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# Introduction

Buda Thermal Karst is famous for its extensive hypogenic cave-system and abundant thermal springs and spas.



**Gellért Spa**



**Molnár János Cave**

Photo by Egri

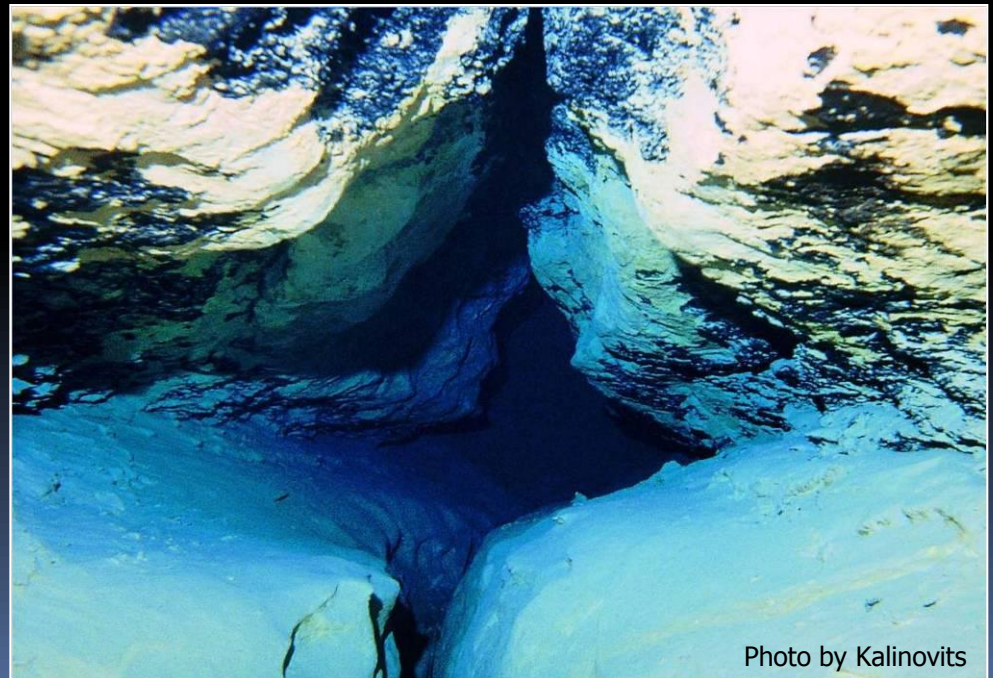
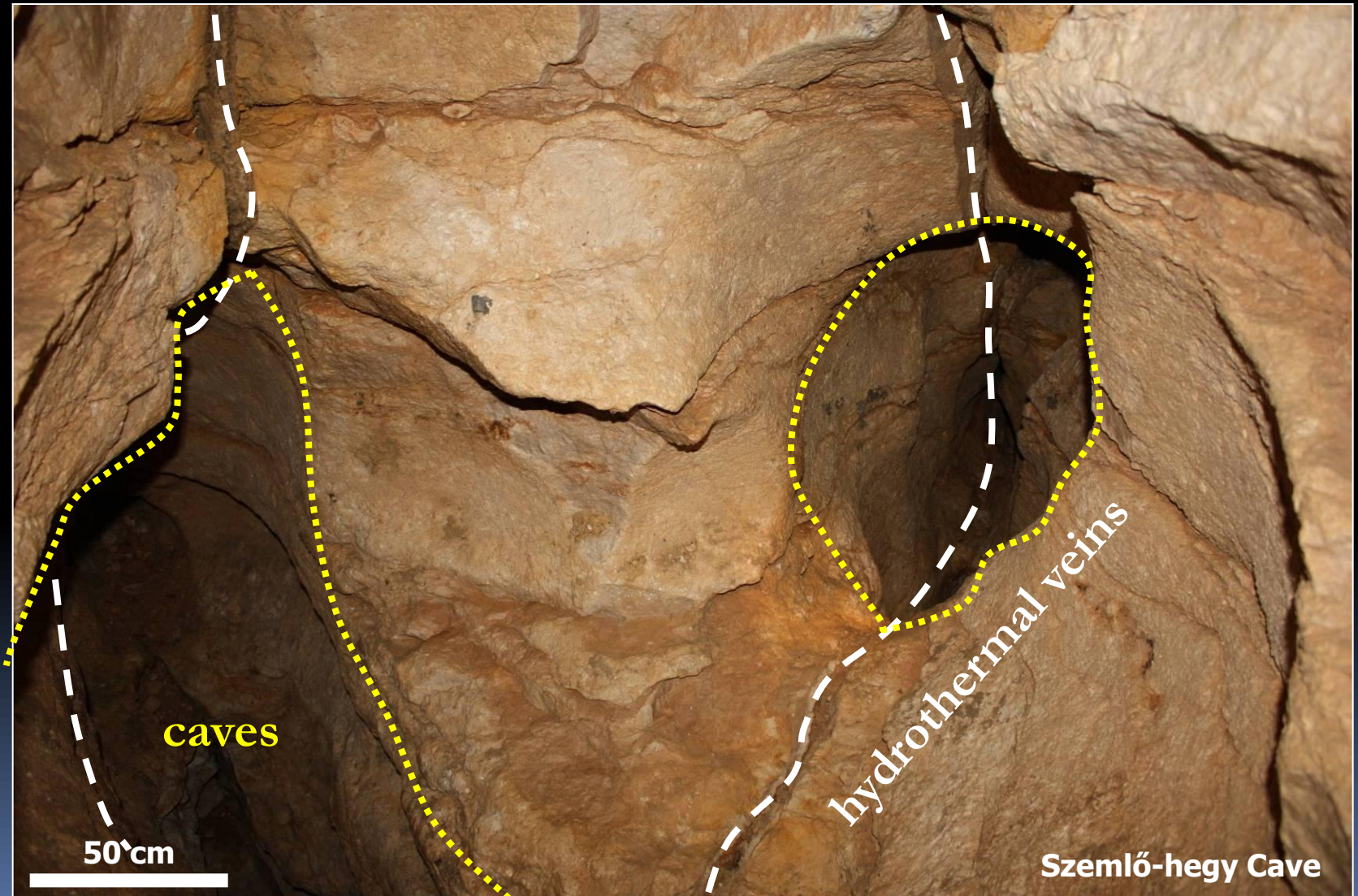


Photo by Kalinovits



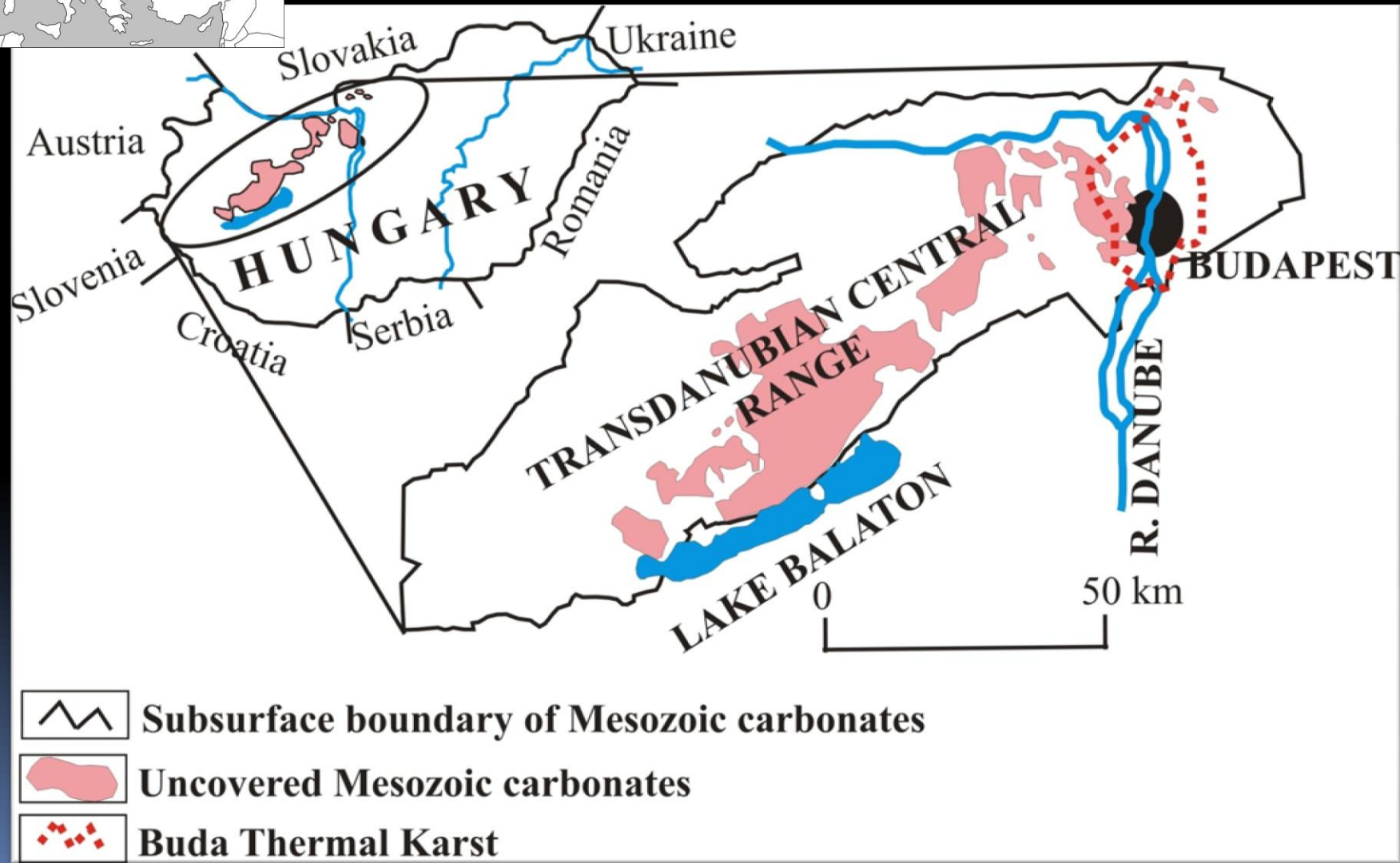
Caves are often related to former hydrothermal veins



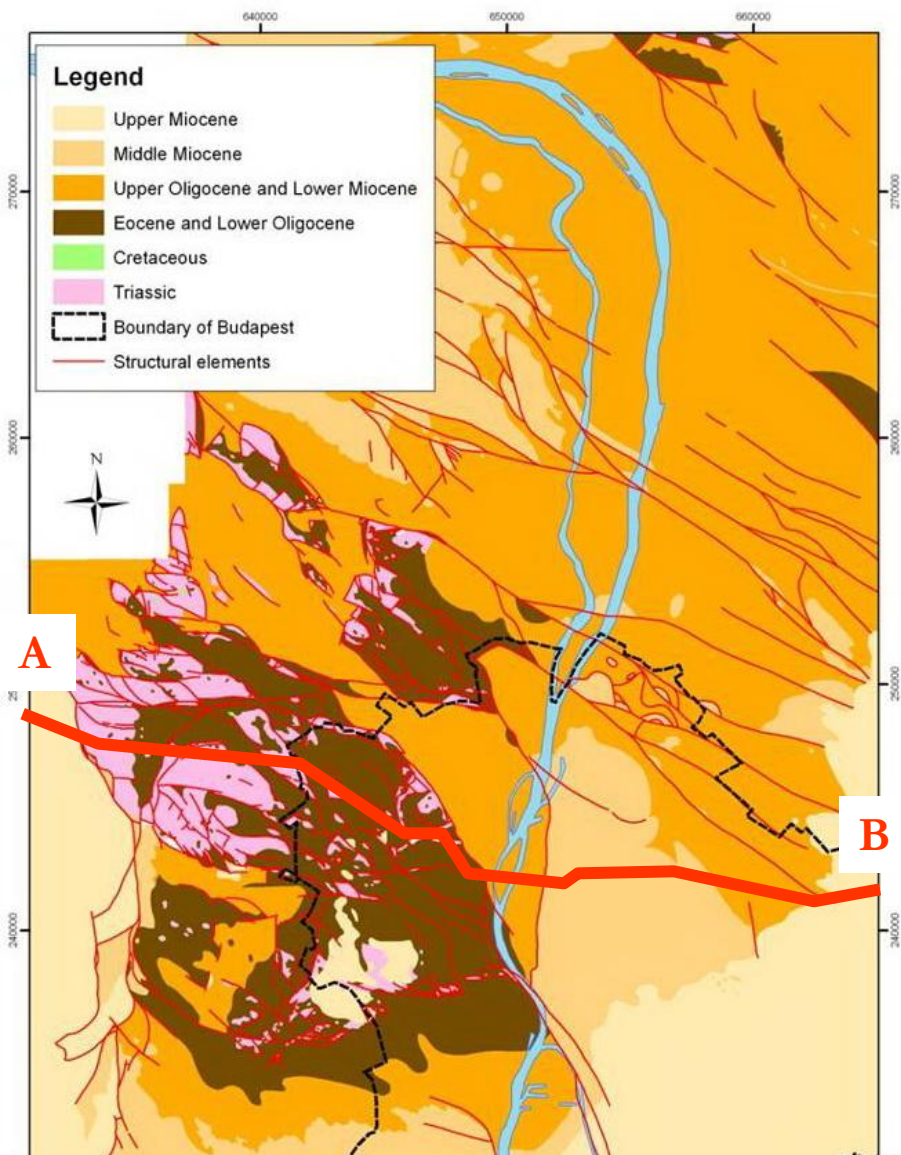
# Aims and methods

- Because of the obvious spatial relationship between vein-forming and cave-forming fluid migration, this study was aimed to understand the possible causal relationship of the two.
- Paleo- and recent hydrothermal phenomena were investigated in the Buda Hills by several previous authors (e.g. Braun, 1889; Schafarzik, 1921, Alföldi 1979, Dublyansky, 1991; Molnár and Gatter, 1994; Benkovics et al., 1999; Gál et al., 2008), however, separately only. Mixing corrosion has been considered as the most important actual cave-forming process (e.g. Kovács and Müller, 1980; Takács-Bolner and Kraus, 1989; Leél-Óssy, 1995; Kalinovits, 2006).
- We have studied **recent- and paleo-hydrogeology in parallel** to understand the effects of paleo-fluid migration on the recent hydrogeological setting and the distribution and development of the extensive hypogenic cave-systems in the Buda Thermal Karst.
- **Recent water** samples were taken from wells and springs and analysed while the composition of „**paleo-fluids**” was established by fluid inclusion studies.

# Geology of Buda Thermal Karst





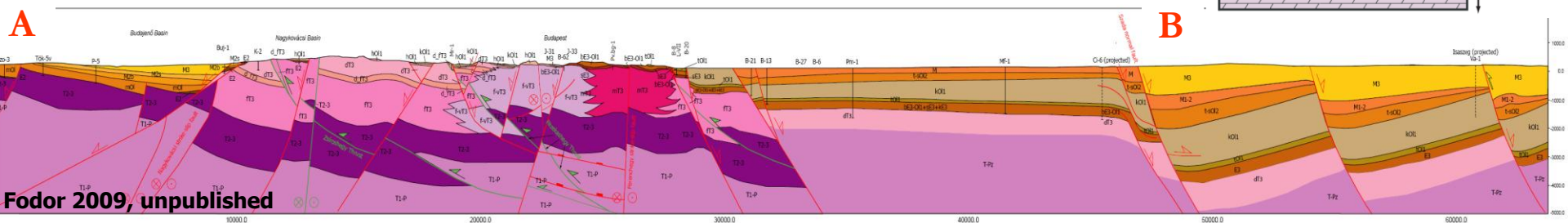
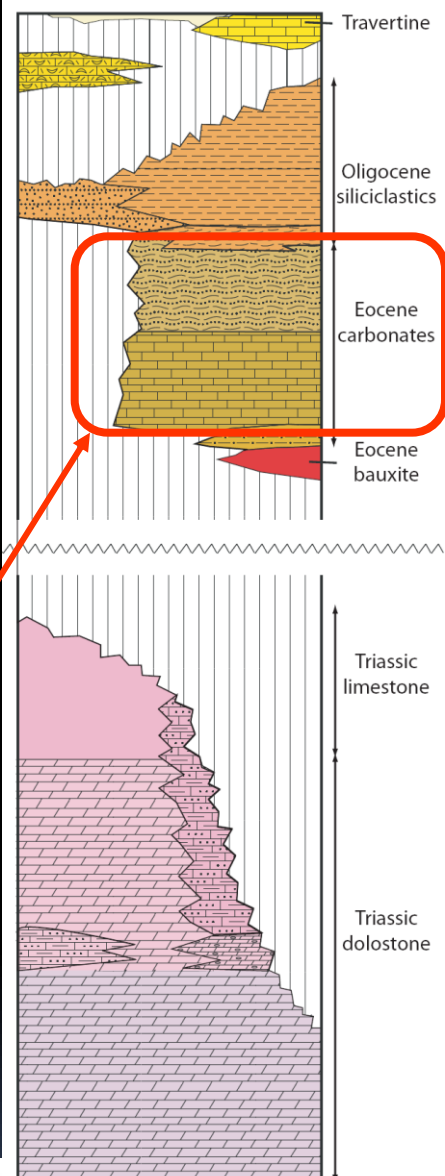


Buda Hills is an uplifted block of the Mesozoic-Tertiary basement of the Pannonian Basin

Two carbonate successions: Triassic and Eocene

Majority of the caves is hosted by the Eocene carbonates

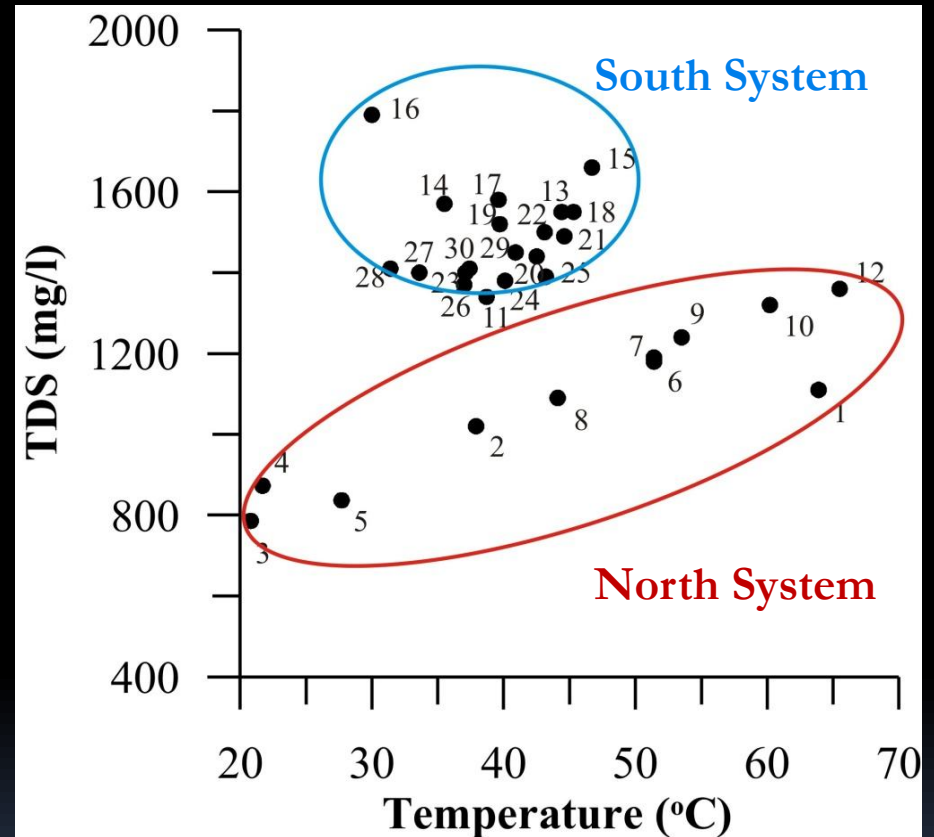
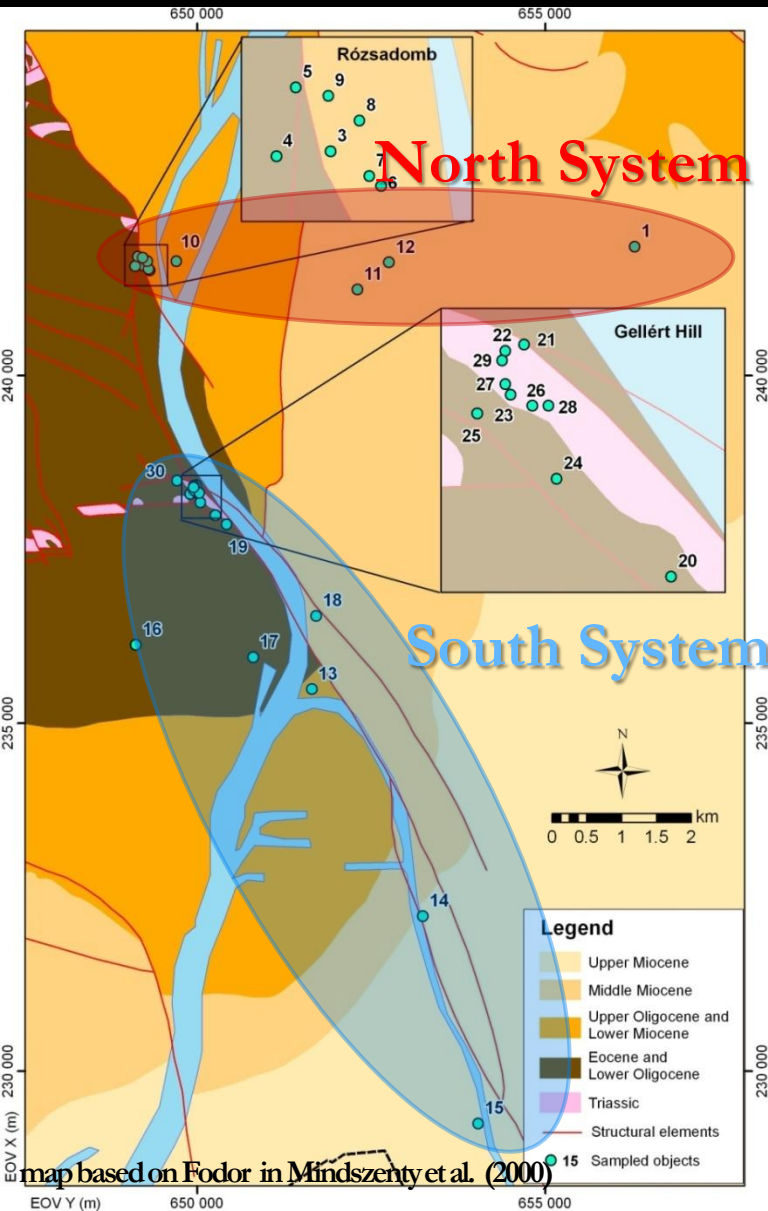
Formation of travertines from the latest Miocene on (Scheuer & Schweitzer 1974, Müller & Magyar 2008)





# Recent hydrogeological setting

## Two sub-systems: North and South



# Recent hydrogeological setting

Two sub-systems: North and South

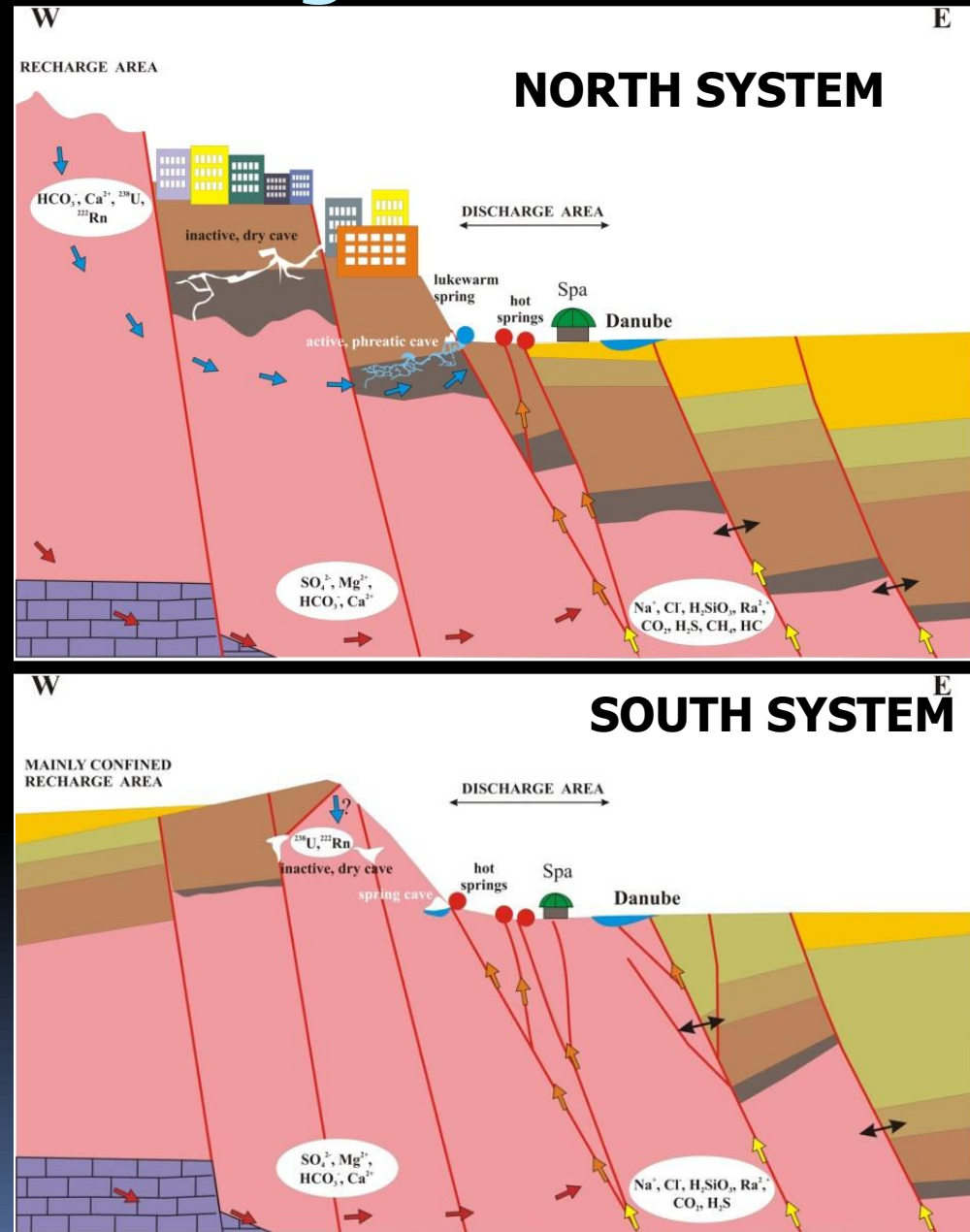
## North System:

Meteoric end member  
12°C, 775 mg/L TDS

Hydrothermal end  
member 76 °C, 1440  
mg/L TDS

## South System:

Direct discharge of  
hydrothermal waters:  
47 °C, 1790 mg/L TDS



# Recent hydrogeological setting

## Two fluid components:

### 1. Karst water

#### a, local-intermediate flow systems:

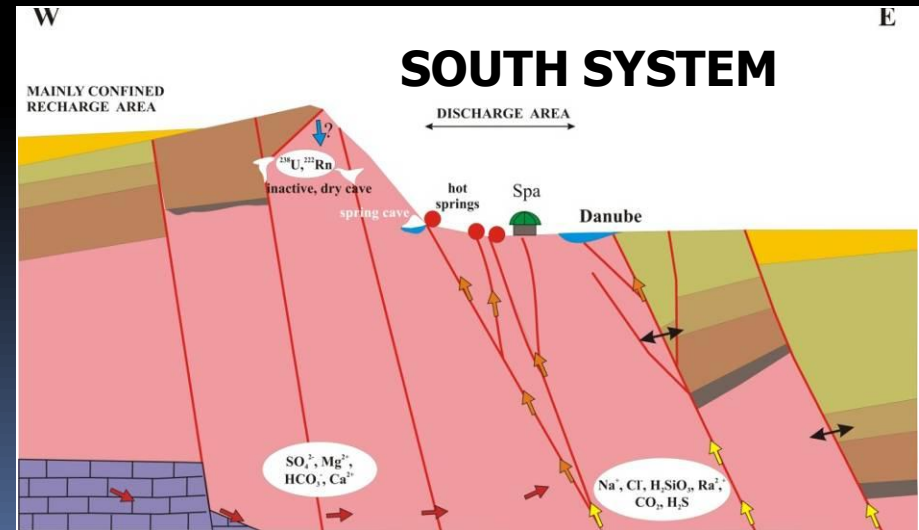
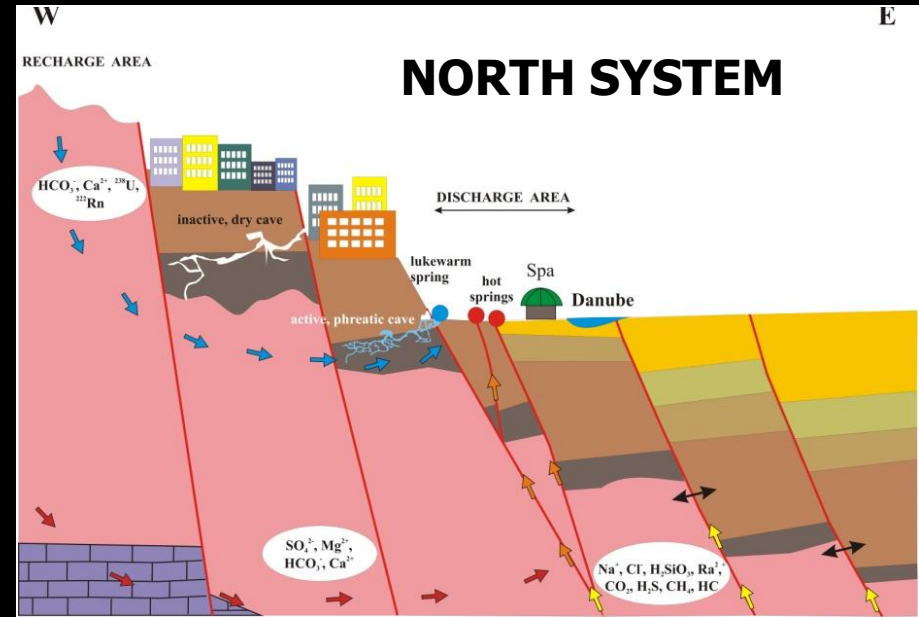
- components:  $\text{Ca}$ ,  $\text{HCO}_3^-$
- salinity: 775 mg/l
- temperature:  $12^\circ\text{C}$

#### b, regional flow systems

- components:  $\text{Ca}+\text{Mg}(\text{Na}+\text{K})$ ,  $\text{Cl}+\text{SO}_4^{2-}+\text{HCO}_3^-$
- salinity: 1440 mg/l
- temperature:  $\sim 76^\circ\text{C}$

### 2. Basinal fluid

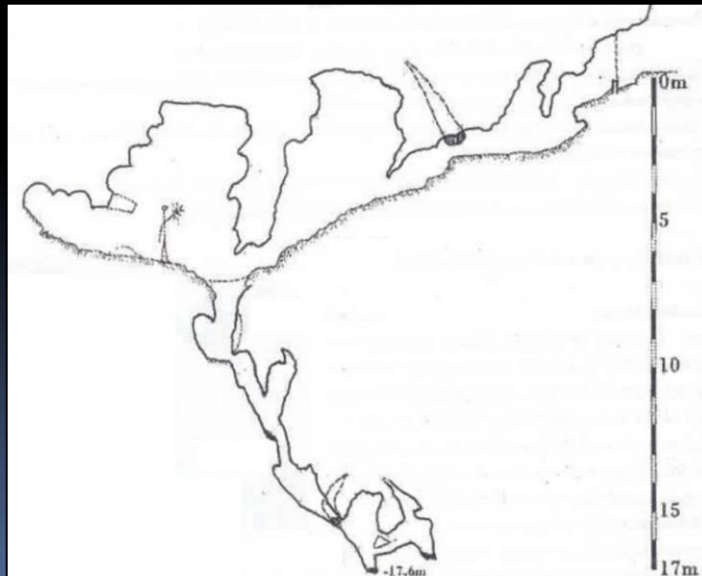
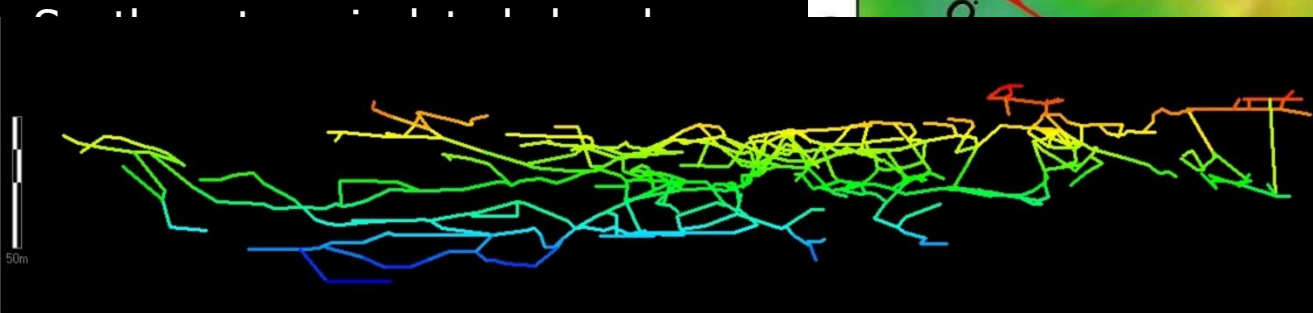
- components:  $\text{Na}$ ,  $\text{Cl}$ ,  $\text{Ba}$ ,  $\text{Sr}$ ,  $\text{F}$ ,  $\text{B}$ ,  $\text{Li}$ ,  $\text{SO}_4^{2-}$  and gases:  $\text{H}_2\text{S}$ ,  $\text{CO}_2$  +  $\text{CH}_4$  and other HC, only in the North system
- salinity:  $\sim 40\,000\text{ mg/l}$  (?)



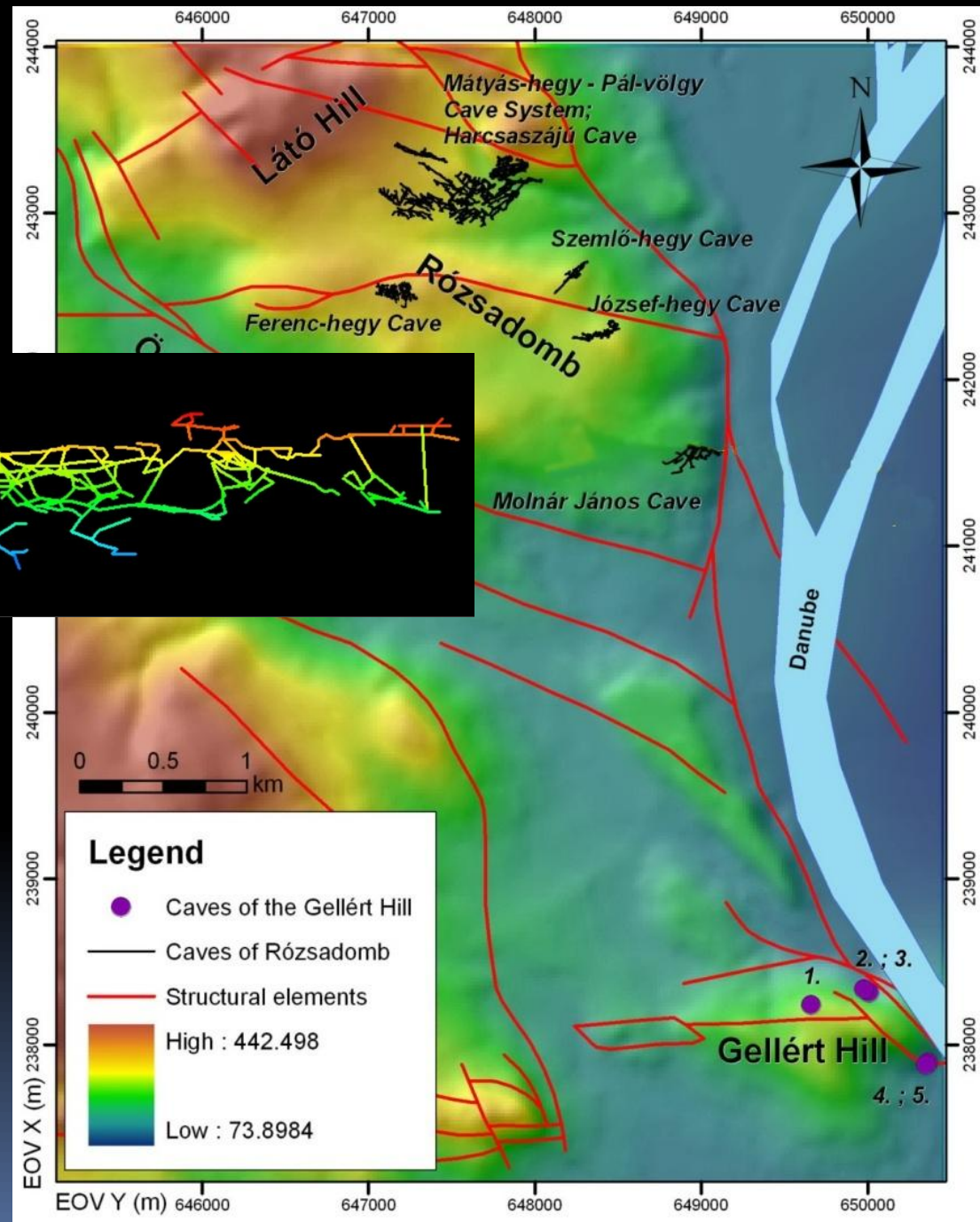


# Caves

North system: fracture-related,  
maze-like pattern



Leél-Őssy et al. 2007



# Cave forming processes

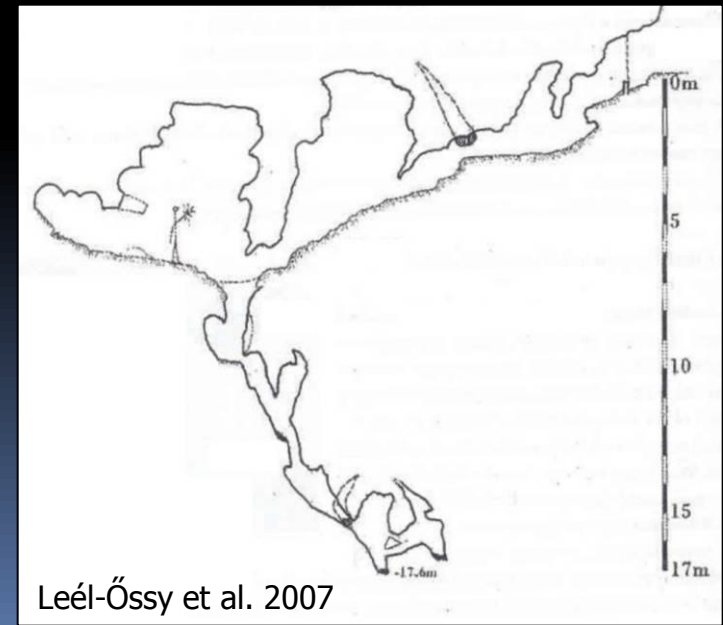
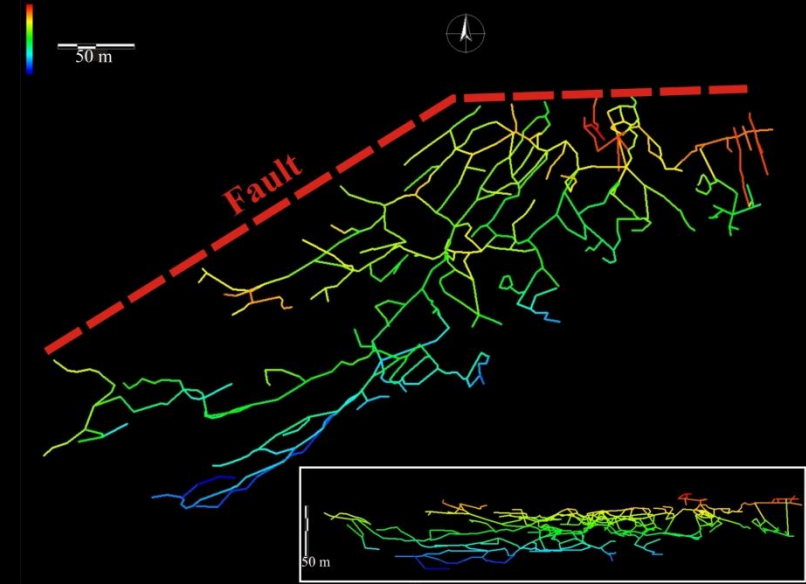
Two sub-systems: North and South

## North System:

- **Mixing corrosion**
- **Microbially mediated sulphuric acid speleogenesis**

## South System:

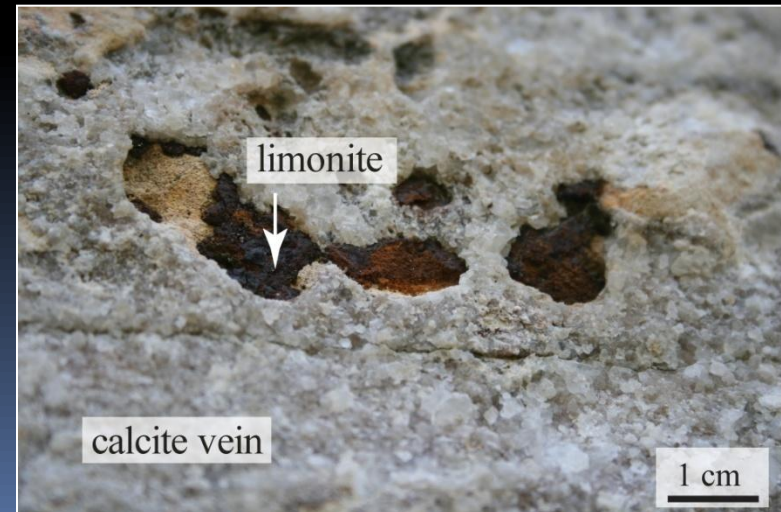
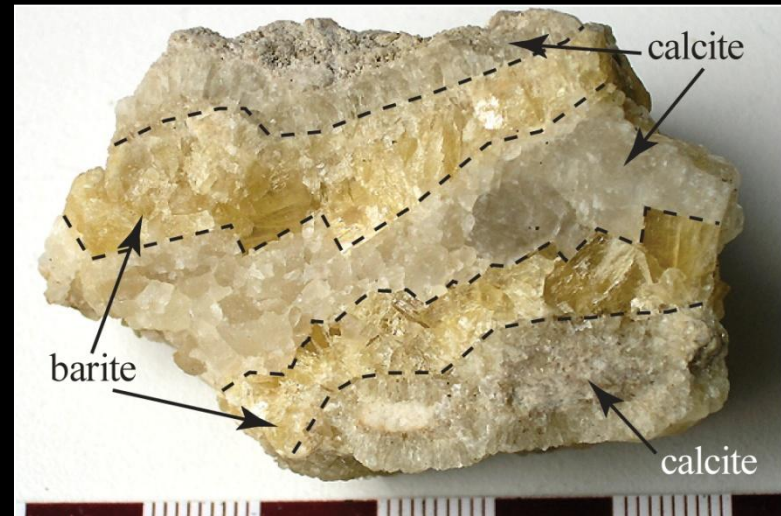
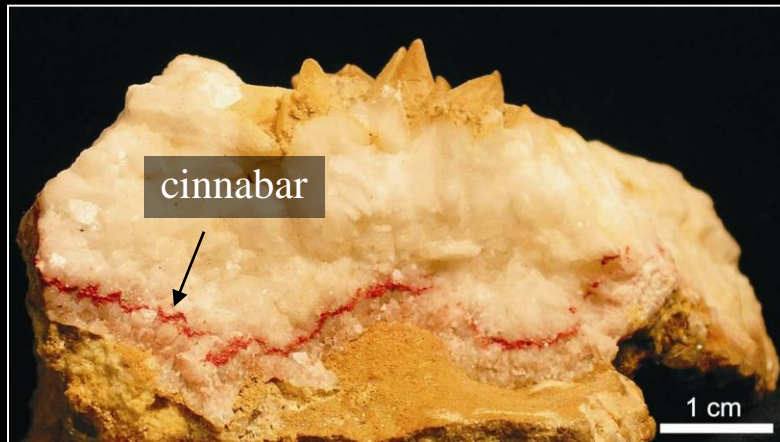
- **Microbially mediated sulphuric acid speleogenesis**





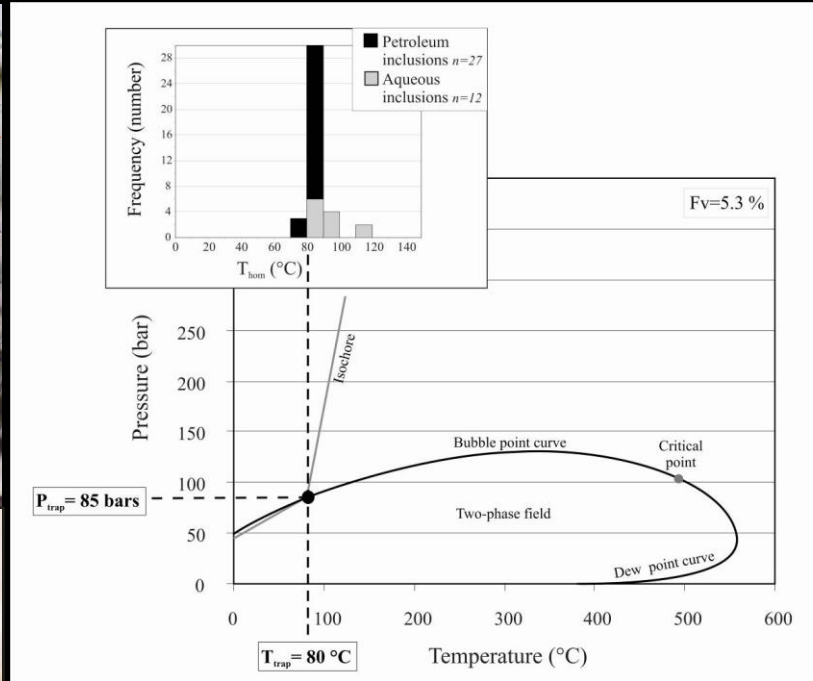
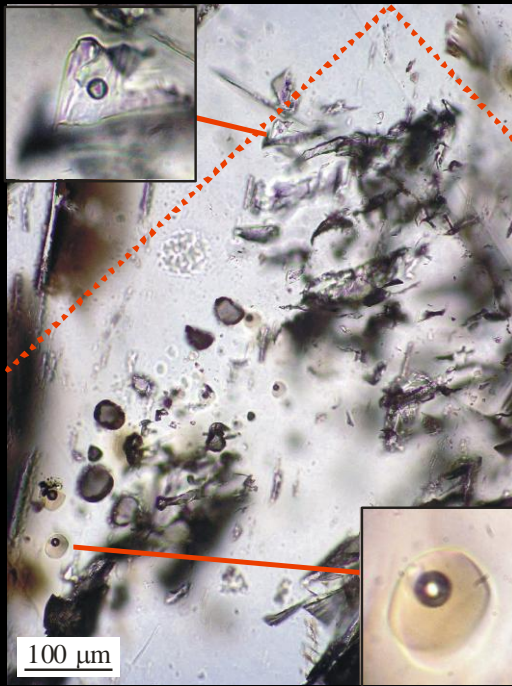
# Paleo hydrogeology

## Fracture-filling paragenesis: calcite, barite, fluorite, sulphides

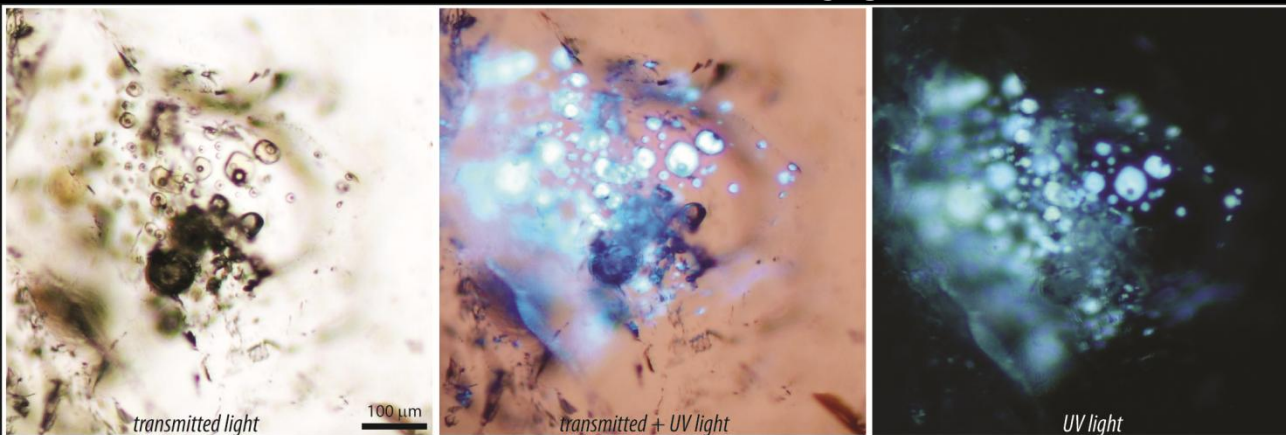




## Fluid inclusion evidence for basinal fluid contribution



Details in: Poros et al. in press,  
*IJES*



- Coeval, primary petroleum and aqueous inclusions in vein-calcite.
- Both petroleum and aqueous phases contain  $\text{CO}_2$  and minor amounts of  $\text{CH}_4$ .
- Confined system:  $P=85$  bars  $\sim 800$  m depth.
- Initiation of basinal contribution: Middle Miocene based on structural geological evidences.

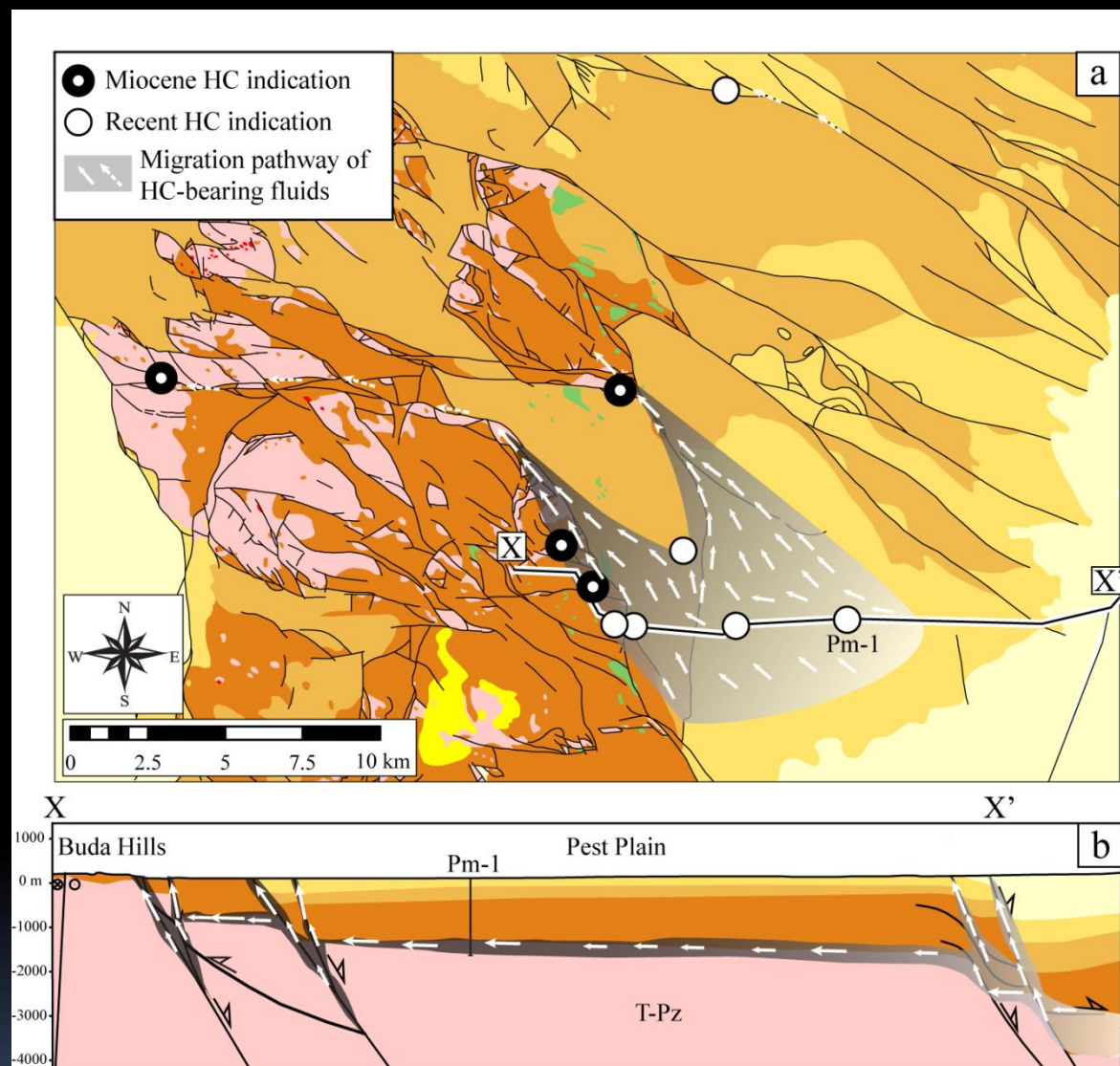
CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>S gases have migrated up towards the Buda Hills from the Miocene on.

- low salinity (<1.7 NaCl eq. wt%)
- HC only in the North

- Same subsystems were established for Miocene and recent
- Same fluid migration pathways were reconstructed for Miocene and recent
- Similar compositions of Miocene and recent basinal fluids (e.g. Na, Cl, Ba, Sr, F, Fe,  $\text{SO}_4$ ,  $\text{H}_2\text{S}$ , HC,  $\text{CO}_2$ ,  $\text{CH}_4$ )
- Pliocene-Pleistocene travertines contain fluorite and barite (Kovács-Pálffy & Földvári, 2004)
- Recent – sub-recent barite precipitation (Surányi in Leél-Össy, 2004)
- Compaction-driven upward flow converted into compressional flow from the Late Miocene due to the inversion of the Pannonian Basin.



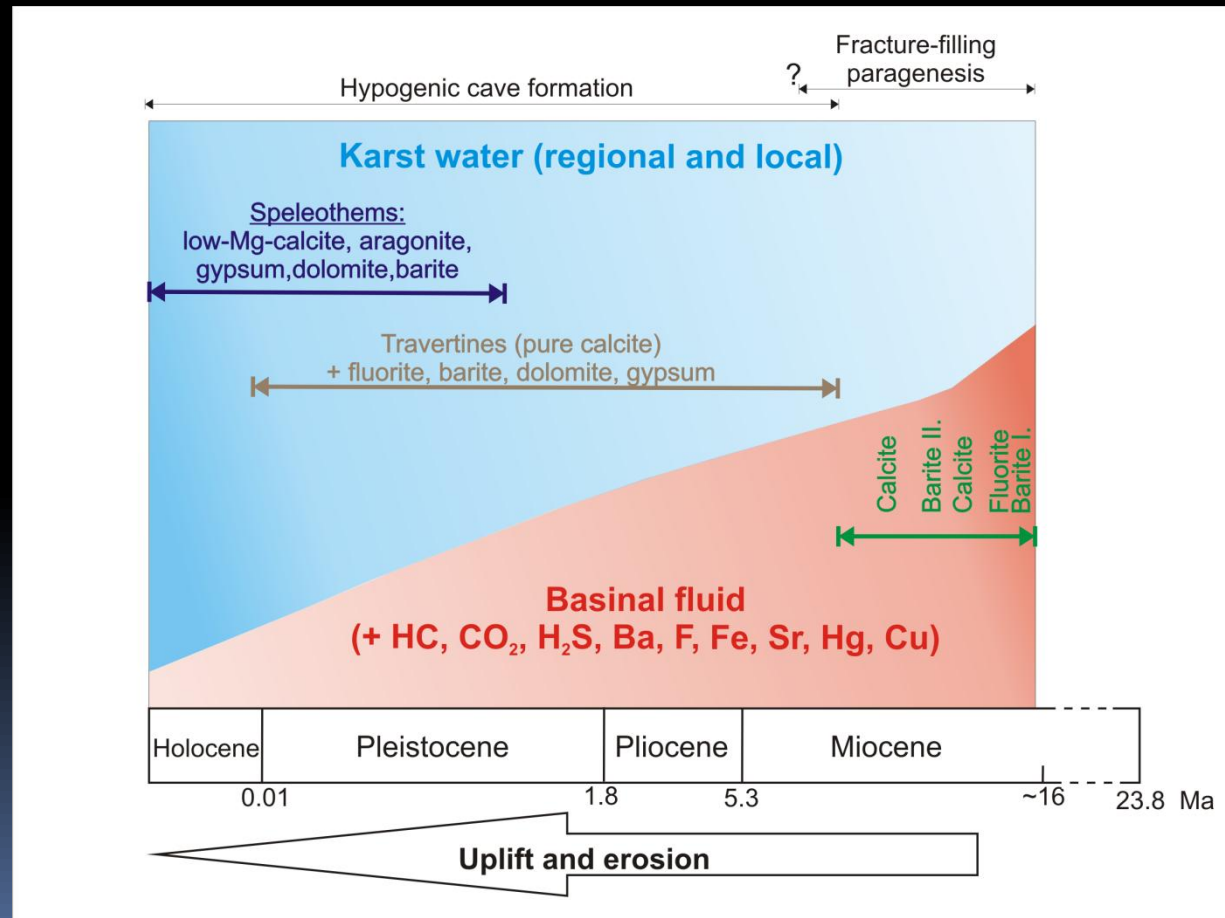
Continuous basinal fluid contribution from the Middle Miocene on



Poros et al. in press, *IJES*

## **Uplift and erosion** resulted in:

- Gradual change from confined to **unconfined aquifer**
- Increasing **direct recharge** from increasingly exposed surface
- **Increasing proportion of karst water** at the expense of basinal fluids
- Change in predominant cave forming process:
  - Agressive gases prevalent in confined conditions
  - Mixing corrosion prevalent in unconfined conditions





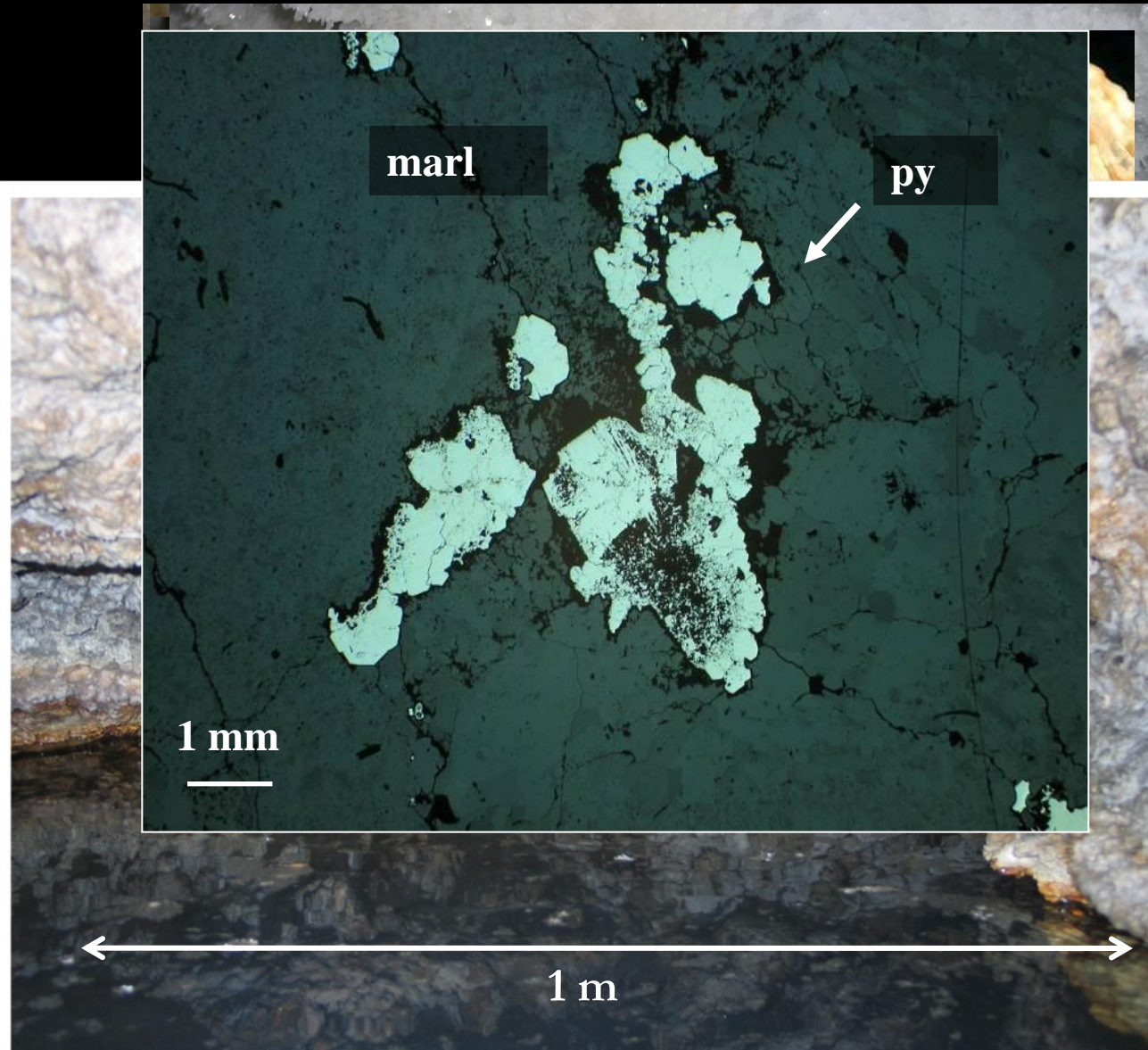
# Sulphate and sulphide minerals

- Miocene times: barite, pyrite
- Recently: gypsum

- Morphological types of gypsum:

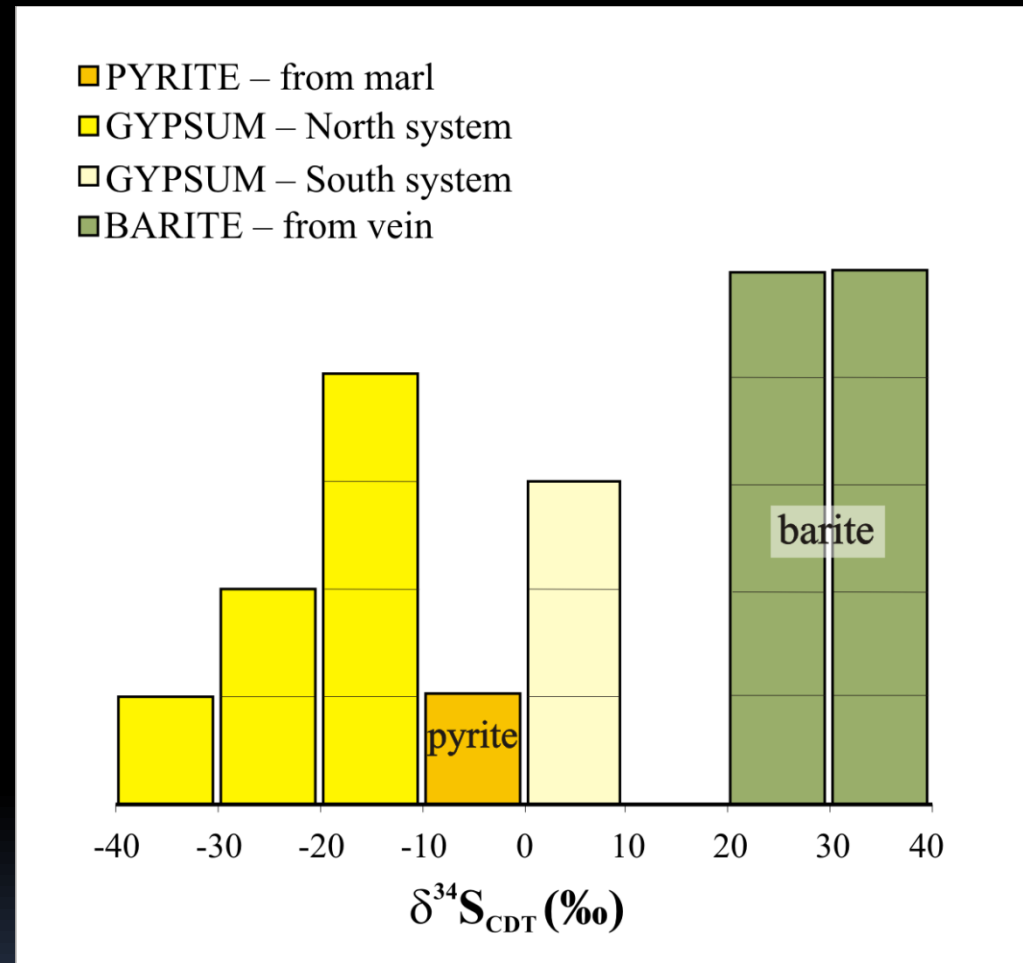
- crust of microcrystalline gypsum may or may not be bulbous,
- chandeliers and towers
- skeletal crystals,
- flower (sources of S is concentrated → pyrite oxidation),
- needle-fiber,
- coarse crystalline gypsum overgrowth (reprecipitated crust)

*Different morphologies reflect different sources of sulphur*



# Sulphur isotopes of minerals

- Sulphur isotopes of gypsum reflect the differences of the two hydrogeological systems:
  - 1# gypsum: North system (-19.2 – -32.3‰)
  - 2# gypsum: South system (2.3 – 8.7‰)
- Sources of sulphur
  - 1# barite (22.5 – 38.1‰)  
→ sulphate from marine evaporites
  - 2# pyrite in marl formed by early BSR
- Strong microbial effect (e.g. -32‰) due to the repeated redox processes (?)
- Morphological differences are most probably caused by differences in growth rate



# Conclusions

- On the basis of geochemical analyses of recent waters, and the petrography and fluid inclusion analyses of paleo-precipitates, three types of fluids from different sources were distinguished: 1) regional karst water, 2) local karst water and 3) basinal fluid.
- Continuous **basinal contribution to the Buda Thermal Karst** from the Miocene on was confirmed.
- Hydrothermal events having resulted in the formation of vein-filling minerals, travertines and hypogenic caves should be treated as different phases of the evolution of one single hydrothermal system.
- Increase in dissolution at the expense of precipitation from the Miocene to present suggests an evolving groundwater system in which the proportion of karst waters increased at the expense of the basinal component due to the uplift and erosion.
- In addition to classical mixing corrosion, other processes must have been efficient in dissolution, e.g. **corrosion effect of gas seeps** ( $\text{H}_2\text{S}$ ,  $\text{CO}_2$ ,  $\text{CH}_4$ ), the presence of microbial activity.



# Acknowledgements

- ENI S.p.A. and Shell International E&P for financial support
- Szabolcs Leél-Őssy, Sándor Kalinovits, Benedek Gál, Szilárd Regős, Csaba Leél-Őssy, Sándor Kraus for the assistance in cave mineral sampling
- Jacques Pironon (Nancy Université, France) for his help in investigations of petroleum inclusions
- Andrea Borsodi for the microbiological studies
- Orsolya Győri for useful comments

Thank you for your attention!

