FINAL REPORT

Reconstructions of climate and environmental changes in Hungary and Croatia during the Holocene by complex investigations of speleothems

PD-121387

This PD-OTKA project (No: 121387), funded by the NKFIH, aimed to conduct a comprehensive investigation of Hungarian and Croatian caves, including monitoring of precipitation, cave monitoring, and a detailed investigation of speleothems. The project provided a salary for the Principal Investigator (PI) for three years (no research budget). Thus, the measurements and field trips were conducted within the framework of scientific cooperation or sponsored by the host institute (Institute for Geological and Geochemical Research, CSFK, ELKH).

The research project had three main directions: 1) *monitoring of precipitation*; 2) *monitoring of caves in Hungary and Croatia*; 3) *comprehensive investigation of speleothems collected in the studied caves*. Determining the processes and conditions that control and affect the stable isotope composition of the precipitation in this region is essential to reveal the climatological information that can be inherited into the infiltrating water. In addition, the comprehensive monitoring activities within the cave allow us to disentangle the complex interactions between the environmental/climatological parameters and the response inside the cave, as well as identify the "transformation" of this information to the petrographic, chemical and isotopic signal of the precipitating carbonates. Two cave systems (Cerovacke, Baraceve) were selected in Croatia, and four in Hungary (Csodabogyós, Legény, Ajándék and Béke caves) for the performance of the detailed studies.

Altogether 14 papers related to this the project were published (12) or accepted for publication (2). In five of these studies the PI is first author or the corresponding author. In addition, seven conference (national and international) abstracts were also published within the framework of the project. In addition to the published works, one has recently been submitted and two others are in the final stage of preparation.

Major results

1) Monitoring of precipitation

   Precipitation samples were collected on a monthly and daily basis at several locations in Croatia and in Hungary. Apart from the sites close to the studied caves, several others

were also monitored providing the first "comprehensive map" of the isotopic composition of the precipitation for Hungary. We determined the relationship of the stable isotope composition of precipitation with climatological parameters and the source of the moisture, which information is essential in order to interpret the stable isotope composition of speleothems and to reconstruct past temperatures. Thus, we established the δ^{18}O-T and δD-T relationships for precipitation. In addition, we constrained the seasonality of the hydrogen and oxygen isotope composition of the precipitation and determined the amount-weighted stable isotope composition of annual precipitation, and the compositions characteristic of the summer and winter half-years. Apart from the temperature and seasonal distribution of precipitation, the moisture source and its changes have the most significant impact on the stable isotope composition of precipitation (and the infiltrating water). We reconstructed the path of the air moisture from the source region by running the NOAA HYSPLIT trajectory model and determined the location where the water vapour entered the atmosphere by calculating specific humidity along the trajectories. Five possible moisture source regions for precipitation were defined: Atlantic, North European, East European, Mediterranean and continental (local/ convective). We found that the most dominant among the identified marine source regions in all stations is the Mediterranean area; while the second is the Atlantic region. The ratio of the precipitations originating from Eastern and Northern Europe seem to correlate with the geographic position of the meteorological station. Our investigation revealed systematic and significant differences between the regions, especially in the case of the most dominant marine sources: the precipitation originating in the Mediterranean regions shows more positive stable hydrogen and oxygen isotope compositions, and has systematically higher d-excess values than that originating in the Atlantic sector. Thus, if the relative contribution of the precipitation of different moisture source changes, the isotopic composition of the infiltrating water, and hence those in the drip water will change. For this reason, it is also necessary to consider moisture source changes for the interpretation of the stable isotope data of speleothems.

2) Cave monitoring

For a robust interpretation of the speleothem-based proxies it is crucial to conduct cave monitoring. Therefore, all the caves were monitored from where speleothems were collected or are planned for collection. The monitoring activities in caves included

microclimate measurements, analyses of the elemental and stable isotope compositions of drip water and precipitation, as well as stable isotope measurements of modern calcite precipitates formed on light bulbs or glass plates. The stable isotope compositions of the drip waters in all cases (except one) show systematically lower values than those found in amount-weighted annual precipitation, and show no significant seasonality (contrast to those in precipitation). Thus, the investigation of the relationship of stable isotope composition of the drip waters and the amount-weighted precipitation indicated that the epikarst above the studied sites is generally well mixed and the dominant infiltration takes place during the winter half year. Moreover, the relative contribution of winter precipitation can vary even within same cave system also reflecting the local morphology of the karst above the cave. In summary, it can be concluded that the speleothems from these locations likely record multiannual winter-biased climate trends. The d-excess values of the drip waters show an increasing trend from the Aggtelek Karst towards the Adriatic Sea, showing higher values than 10‰. These observations indicate a significant contribution of moisture originating from the Mediterranean Basin to the infiltrating water. The monitoring of the precipitation supports these findings, as among the marine moisture sources the Mediterranean is the most dominant, even relatively remotely from the sea. By investigating the fresh carbonates precipitated on the glass plates we determined the calcite-water isotope fractionations and we found significant scatter, even within individual caves. Besides these observations, it can be noted that the majority of the data fall close to the Coplen (2007) and the Tremaine et al. (2011) fractionation values in both the Croatian and Hungarian caves.

Apart from these general features, all caves and sites have their own characteristics. These site-specific features play a key role in constraining the factors that influence the carbonate precipitation and the observed chemical and isotopic variations in speleothems.

3) Climate reconstructions

In order to find speleothems that developed during the Holocene in the studied caves, we performed in-situ sampling in some cases, while in other cases we took the speleothems and sampled their bottom and upper part for trial U-Th age determinations. Unfortunately, the selected speleothems in Csodabogyós, Legény and Ajándék caves

gave significantly older ages, mainly covering the last interglacial period. Therefore, these speleothems were not suitable for the current research. For this reason, we collected speleothems from Béke Cave and involved speleothems from other caves. We were able to collect speleothems that developed during the Holocene in the selected Croatian caves (Baraceve and Cerovacke caves).

Utilizing the available speleothems, we performed comparisons among different records. The main focus was on the early (8.2 kyr event) and late (Dark Ages Cold Period (DACP); Medieval Warm Period (MWP)) Holocene climate events because the collected and studied speleothems cover these time periods.

Early Holocene

The most significant climate anomaly during the Holocene took place around 8.2 ky (cal BP), therefore it is referred to as the "8.2 kyr event". The studied speleothems (BB-speleothem and B1350) from Béke Cave includes this period. Applying the improved age model, the combined stable isotope and trace element signals revealed a complex picture of this event. It seems that this event is characterized by negative oxygen and carbon isotopic peaks around 8.2 ky (cal BP) and a positive stable isotope anomaly around 8.1 ky (following the actual event). The negative anomaly has been widely observed in the western European speleothems and other speleothems worldwide showing the global significance of this climate event. Similar to other records the observed stable isotope (d13 and d18O of calcite and dD of inclusion-hosted water) of Béke speleothems can be interpreted as a signal of cold conditions. However, followed by this negative anomaly in the stable isotope signal we recognized a significant positive peak in Béke speleothems. The positive peak is not just observed in the oxygen and carbon isotopic composition but also in the dD values obtained from inclusion-hosted water. The positive peak observed by us is not really mentioned in other European speleothem-based climate reconstructions. Unfortunately, the speleothem collected in Cerovacke Cave in Croatia did not provide age data of sufficient reliability to make a comparison with the records from Béke Cave possible.

This positive peak may reflect 1) an increase of air temperature, 2) a relative increase of precipitation during the warmer period, or 3) changes in the moisture sources. Independent evidence from Central Europe for increased temperature following the negative 8.2 anomaly has been reported based on the oxygen isotopic composition of benthic ostracods from lake sediments (Austria; Andersen et al., 2017). Moreover, in

addition to increased air temperature, it seems that the hydrological conditions also changed during the period as the Sr content and the calculated dead carbon contribution (dcp, referred from C-14 data) shows a decrease from 8100 y (cal BP). In addition, the recent monitoring of the stable isotope composition and the moisture source of the precipitation that we conducted in Hungary indicates that the different moisture sources are characterized by different stable isotopic compositions. Therefore, the observed anomaly in the isotopic composition of the speleothem can be explained also by changes in the source of the precipitation in Hungary.

Late Holocene

The speleothem from Baraceve Cave (Croatia) grew in the late Holocene covering the time period from 1600 y (cal BP) to 600 y (cal BP) with a hiatus between 1000 y (cal BP) and 850 y (cal BP). A similar hiatus was also observed in a flowstone from Béke Cave, Hungary (Demény et al., 2019) from 1050 to 650 y (cal BP) indicating that during the Medieval Warm Period (MWP) both regions are characterized by a relative dry climate. In addition, the carbon isotopic composition of the Baraceve speleothem shows elevated values at the beginning and at the end of the MWP (i.e. before and after the hiatus) reinforcing the dry condition. These observations agree well with other speleothem-based records from Europe suggesting that this Medieval Climate Anomaly is not only a regional scale climate signal.

Following the MWP the Baraceve speleothem (Croatia) recorded a wet climate condition characterized by a strong negative carbon isotope anomaly at 655 y (cal BP), which coincides with the Medieval Glacier Advances (MGA). A similar feature can be observed in the flowstone record from Kapferloch Cave (Austria, Boch and Spötl, 2011). After this event the carbon isotopic values of the Baraceve speleothem show a continuous increase until the "Little Ice Age" indicating continuous changes in the hydrological conditions towards the drier climate. Before the "Little Ice Age" the growth of the Baraceve speleothem stopped similarly to the flowstone from Béke Cave. The period before the MWP, from ca. 1550 to 1200 is often referred to as the Dark Ages Cold Period (DACP). Many papers indicate that during this period cold and wet climate condition were common in the Northern Hemisphere. Moreover, the Baraceve (Croatia) record suggests two drier and probably warmer periods within the DACP. One is centred at 1450 y (cal BP) while the other at 1280 y (cal BP). Although the former dry event is very obvious also in the flowstone record from Kapferloch Cave (Austria, Boch and

Spötl, 2011), the latest in Croatia seems to be a stronger signal. These peaks in the flowstone from Béke Cave and Kiskőhát stalagmite (Hungary) are not so obvious, which might indicate that these dry events were not significant in Hungary.

It can be concluded that the studied regions do not always show the same paleoclimate pattern in comparison with each other and with other European archives because Croatia and especially Hungary are located in the transition zone influenced by the meteorological (climatological) conditions of the Atlantic, Mediterranean and North/East regions.

Papers related to the project

1) Published papers

Czuppon Gy., Bocic N, Buzjak N, Ovari M, Molnar M. 2018. Monitoring in the Barac and Lower Cerovacka caves (Croatia) as a basis for the characterization of the climatological and hydrological processes that control speleothem formation. QUATERNARY INTERNATIONAL, pp. 1-14. (IF: 2.488)

Czuppon Gy., Demény A., Leél-Őssy Sz., Óvari M., Molnár M., Stieber J., Kiss K., Kármán K., Surányi G., Haszpra L. 2018: Cave monitoring in the Béke and Baradla Caves (Northeastern Hungary): implications for the conditions for the formation cave carbonates. INTERNATIONAL JOURNAL OF SPELEOLOGY, 47, 13-28. (IF: 1.439).

Bottyán E., **Czuppon Gy.*,** Weidinger T., Haszpra L., Kármán K. 2017: Moisture source diagnostics and isotope characteristics for precipitation in east Hungary: implications for their relationship. HYDROLOGICAL SCIENCES JOURNAL, 62:12, 2049-2060 DOI: 10.1080/02626667.2017.1358450.

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Czuppon, Gy.; Kern Z., Kármán K., Németh Sz., John Sz., Haszpra L., Kohán B., Kiss K., Siklósy Z., Polasek Zs. 2019. Hydrogen and oxygen isotope composition of cave drip waters: Implications for paleoclimate signal in stalagmite. MŰSZAKI FÖLDTUDOMÁNYI KÖZLEMÉNYEK, 88.1. 44-47.

Czuppon, Gy.; Bottyán, E ; Kristóf, E ; Weidinger, T ; Haszpra, L ; Kármán, K. 2021. Stable isotope data of daily precipitation during the period of 2013-2017 from K-puszta

(regional background monitoring station), Hungary. DATA IN BRIEF 36, 106962.

Demény, A ; Kern, Z.; Németh, A ; Frisia, S ; Hatvani, IG ; **Czuppon, Gy** ; Leél-Őssy, Sz.; Molnár, M ; Óvári, M ; Surányi, G. 2019: North Atlantic influences on climate conditions in East-Central Europe in the late Holocene reflected by flowstone compositions. QUATERNARY INTERNATIONAL, 512, 99-112.

Surić M., **Czuppon Gy.**, Lončarić R., Bočić N., Lončar N., Bajo P., Drysdale R.N. 2020: Stable Isotope Hydrology of Cave Groundwater and Its Relevance for Speleothem-Based Paleoenvironmental Reconstruction in Croatia. WATER, 12:9, 2386; https://doi.org/10.3390/w12092386.

2) Accepted papers

Czuppon Gy, Demény A., Leél-Őssy Sz., Stieber J., Óvári M., Dobossy P., Berentés Á., Kovács R. Monitoring and geochemical investigations of caves in Hungary: implications for climatological, hydrological and speleothem formation processes. in Cave and Karst Systems of Hungary (eds: Veress M., Leél-Őssy Sz.), SPRINGER BOOK.

Emese B, Erzsébet K., Weidinger T., Haszpra L., **Czuppon Gy.*** 2020. Source of the precipitation in Hungary. (A magyarországi csapadékvíz forrása) LÉGKÖR.

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3) Submitted manuscript

Czuppon Gy., Demény A., Leél-Őssy Sz., Stieber J., Óvári M., Dobosy P., Berentés Á., Richard Kovács: Cave monitoring in Hungary: an overview. CENTRAL EUROPEAN GEOLOGY.

4) Manuscript under preparation, before submission

- Significance of the air moisture source on the stable isotope composition of the precipitation in Hungary.
- Complexity of the 8.2 k.y. event as recorded in speleothem from Central Europe: implication for the climate recovery.

Papers which are partly related to the project

Demény, A.; Rinyu, L ; Kern, Z ; Hatvani, IG. ; Czuppon, Gy ; Surányi, G ; Leél-Őssy, Sz ; Shen, CC ; Koltai, G. 2021. Paleotemperature reconstructions using speleothem fluid inclusion analyses from Hungary. CHEMICAL GEOLOGY, 563. 120051.

Demény, A.; Kern, Z. ; Hatvani, I.G. ; Torma, C. ; Topál, D. ; Frisia, S. ; Leél-Őssy, S. ; Czuppon, Gy. ; Surányi, G. 2021. Holocene hydrological changes in Europe and the role of the North Atlantic ocean circulation from a speleothem perspective. QUATERNARY INTERNATIONAL, 571, 1-10.

Hatvani I.G., Szatmári G., Kern Z., Erdélyi D., Vreca P., Kanduc T., **Czuppon Gy.,** Lojen Sz., Kohán B. 2021. Geostatistical evaluation of the precipitation stable isotope monitoring network design for Slovenia and Hungary. ENVIRONMENT INTERNATIONAL. 146: 106263.

Kern Z., Hatvani G.I., **Czuppon Gy**., Fórizs I., Erdélyi D., Kanduč T., Palcsu L., Vreča P. 2020: Isotopic 'altitude' effect and 'continental' effect in modern precipitation across the Adriatic-Pannonian region. WATER, 12:6, 1797; doi:10.3390/w12061797.

Kern Z., Hatvani I.G., Erdélyi D., Vreča P., Bronić I.K., Kanduč T., Štrok M., Fórizs I., Palcsu L., Süveges M., **Czuppon Gy.,** Kohán B. 2020: Isoscape of precipitation amount-weighted annual mean tritium (3H) activity from 1976 to 2017 for the Adriatic-Pannonian region. EARTH SYSTEM SCIENCE DATA, 12 : 3 pp. 2061-2073.

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