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Mid-Holocene climate conditions and moisture source variations based on stable H, C and O isotope compositions of speleothems in Hungary

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ABSTRACT

This paper presents stable C and O isotope data, as well as water contents and stable hydrogen isotope compositions of inclusion-hosted water of ^{230}Th-dated stalagmites collected from the Leány and Pál-völgyi Caves of Central Hungary, within and about 50 km NW from Budapest, respectively. A good replication of contemporaneous stalagmite oxygen isotope records and their agreement with the COMNISPA record from the Eastern Alps suggest that the stalagmite oxygen isotope variation reflects past climate change. H_2O contents in the Leány stalagmite indicate a relationship with the oxygen isotope compositions and hence with climate conditions, raising the possibility of its use as a climate proxy in future studies. The stalagmites show strong negative $\delta^{18}O$ excursions for two cold periods at about 5.4–6 and 8–9 ka, whereas the oxygen isotope data are relatively high in the period of 6–7 ka, indicating warmer conditions. The stalagmite hydrogen and *d*-excess series positively correlate with the COMNISPA record, interpreted as a sign of moisture source variations. High *d*-excess values in the periods of low $\delta^{18}O$ data for the Leány Cave at about 5.4–6 and 8–9 ka suggest a shift to Mediterranean moisture source domination when the COMNISPA record indicate weaker North Atlantic Oscillation (NAO) activity. Lower *d*-excess values in the period of 6–7 ka are associated with low $\delta^{18}O$ values in the COMNISPA record, corresponding to NAO+ phase. The inferred moisture source changes are in accordance with published instrumental data covering the last hundred years and atmospheric circulation model results, and demonstrate that NAO activity influenced the climate conditions of the Carpathian Basin during most of the Holocene.

1. Introduction

Although the Holocene had been considered as a stable period compared to the entire Quaternary, climate fluctuations sometimes induced major societal/environmental changes (e.g., the Little Ice Age or Medieval Warm Period, see Mann et al., 2009). Research on these climate change events had traditionally focussed on temperature variations, whereas precipitation amount and seasonality is gaining importance in recent years (e.g., Mangini et al., 2007), requiring complex interpretation of combined data from independent proxies. Cave speleothems can record annual or even seasonal changes in climate conditions prevailing at the surface above the cave, and can grow up to hundreds of thousand years (e.g., Fairchild et al., 2006). As analytical techniques developed in the last decade, high resolution stable isotope studies in speleothem research have produced isotope records that are comparable in secular resolution and importance to the widely used ice core records (e.g., Henderson, 2006). Several stalagmites have been analysed at high resolution, rendering the establishment of composite records possible that can be used as reference paleoclimate records (e.g., Wang et al., 2001, 2008; Vollweiler et al., 2006; Mangini et al., 2007).

The Carpathian–Pannonian Region is one of the key areas for investigations on the paleoclimate conditions of the European continent as this area receives influences from the Atlantic realm, from the Mediterranean and also from the continental territories. This study provides not only carbon and oxygen isotope compositions of stalagmite calcite from caves in the central part of the Carpathian–Pannonian Basin, but also hydrogen isotope compositions of inclusion-hosted fluids that can directly reflect dripwater,

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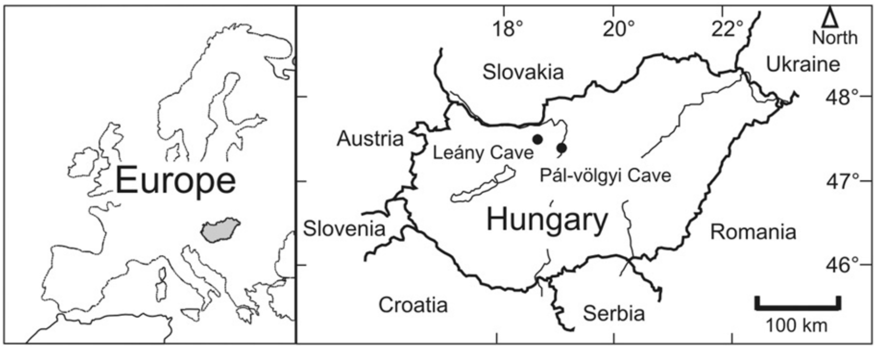

Fig. 1. Locations of the Leány and Pál-völgyi Caves in Hungary.

and hence local precipitation water, compositions. Two stalagmites, one from the Leány Cave and another from the Pál-völgyi Cave (Fig. 1), were selected to provide Holocene thermal and hydrological records in this region.

2. Locations and samples

Geological descriptions and maps for the Leány and Pál-völgyi cave systems as well as photos of the stalagmites studied are given in the Supplementary material background information. The Leány Cave (part of the Ariadne Cave System) is located in the Pilis Mountains, 30 km north-west of Budapest, in the rock group of Csévi-szirtek (Csévi Cliffs). The Ariadne system consists of a series of caves, including the Leány (Girl) Cave, Legény (Boy) Cave, Vacska Cave, and Ariadne Cave, of which total length is more than 13 km. The altitude of the cave entrance is 452 m asl (Székely, 2003), 3 km from the closest village. The vertical extension of the cave is 203 m. The cave is surrounded by natural forests along the Csévi-szirtek. The connection between the Leány Cave and Legény Cave was discovered in 1997, between this new system and the Ariadne Cave in 2006, and with the Vacska Cave in 2010.

The Pál-völgyi Cave, 14 km in length, is situated in the interior of Budapest, on the right side of Danube River, in the Buda Hills that has an altitude of 300–400 m asl. The entrance of the cave is at 206 m asl, surrounded by a belt of residences. The vertical extent of the cave is 123 m (Székely, 2003). For the present study a young, 12 cm high, 10 cm broad stalagmite of the "Vasárnapi Barlangászok" ("Sunday Cavers") passage seemed to be most suitable.

Meteorological data and cave temperatures are partly from the public database of the Hungarian Meteorological Survey and partly based on unpublished data from G. Surányi and H. Nagy for the period of 2007–2010. The area of the caves studied is characterized by continental climate with 10–50 snowy days per year. The annual precipitation is between 500 and 1000 mm, usually 600–700 mm. The wettest months are May–June and September–October. The average temperature in the surroundings of the Pál-völgyi cave is 10.5 °C, and the Leány Cave's area has an annual mean temperature of 8 °C. The temperatures of the inner passages reflect the mean annual temperatures around the caves, fluctuating between 9.9 °C and 10.6 °C in the Pál-völgyi Cave and between 7.2 °C and 8.1 °C in the Leány Cave. The Pál-völgyi Cave is a show cave, and thus frequent opening for visitors can cause higher temperature fluctuation compared to earlier times when the cave was closed.

3. Analytical techniques

The U/Th age determinations were conducted at the Department of Geosciences, National Taiwan University using standard procedures (Shen et al., 2002, 2003). 0.1–0.2 g of sample chips were dissolved in HNO_3, spiked with a mixed ^{229}Th–^{233}U–^{236}U tracer, refluxed, and taken to dryness. U and Th were co-precipitated with an Fe-carrier, then separated to form a clean U fraction and a clean Th fraction using an anion exchange resin (Shen et al., 2003). The isotopic compositions of U and Th in the solutions were measured by a multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS), Thermo Electron NEPTUNE (Shen et al., 2010) at the National Taiwan University, Taipei.

Calcite samples were drilled from cut and polished stalagmite surfaces at about 1 mm spatial resolution. Stable carbon and oxygen isotope compositions of approximately 150–200 μg carbonate samples were determined applying the carbonate–orthophosphoric acid reaction at 72 °C (Spötl and Vennemann, 2003) and using an automated GASBENCH II sample preparation device attached to a Thermo Finnigan Delta Plus XP mass spectrometer at the Institute for Geological and Geochemical Research, Budapest. H_2O contents in stalagmite samples and H isotope compositions of inclusion-hosted H_2O were determined by vacuum-crushing. Chips of 3–5 mm were placed in stainless steel tubes welded at one end, pumped to vacuum, then crushed using a hydraulic press. The released H_2O was purified by vacuum distillation and reacted with Zn at 480 °C to produce H_2 gas (see Demény, 1995; Demény and Siklósy, 2008). The H_2O contents and D/H ratios were determined using a Finnigan MAT delta S mass spectrometer at the Institute for Geological and Geochemical Research. The isotope compositions are expressed as δD, $\delta^{13}C$ and $\delta^{18}O$ values relative to V-PDB ($\delta^{13}C$ and $\delta^{18}O$ values of calcites) and V-SMOW (δD and $\delta^{18}O$ values of waters), according to the equation: $\delta = (R_{sample}/R_{standard} - 1)$. 1000, where R is the D/H, $^{13}C/^{12}C$ or $^{18}O/^{16}O$ ratio in the sample or in the international standard. The measurement precision is better than 0.15‰ for C and O isotope data based on replicate measurements of international standards (NBS-19; NBS-18) and in-house reference materials, and about 3‰ for δD values based on duplicate analyses.

4. Results

Age models of the studied stalagmites are based on ten age dates for the Leány stalagmite and three age dates for the Pál-völgyi one (Supplementary material Table S1), although the oldest of the age dating samples (Leany-Cave 1, uncorrected age of 11,300 years) was too contaminated by detrital Th to conduct correction. The age-depth model was constructed for the Leány Cave stalagmite using the StalAge algorithm of Scholz and Hoffman (2009). Two model calculations were conducted, one using all dates obtained and another one excluding two dates with large uncertainties (8960 ± 1115 years and the uncorrected age of 11,300 years). The two age model curves are equal within 5 years except for the part older than 10,270 years, which shows the reliability of the age

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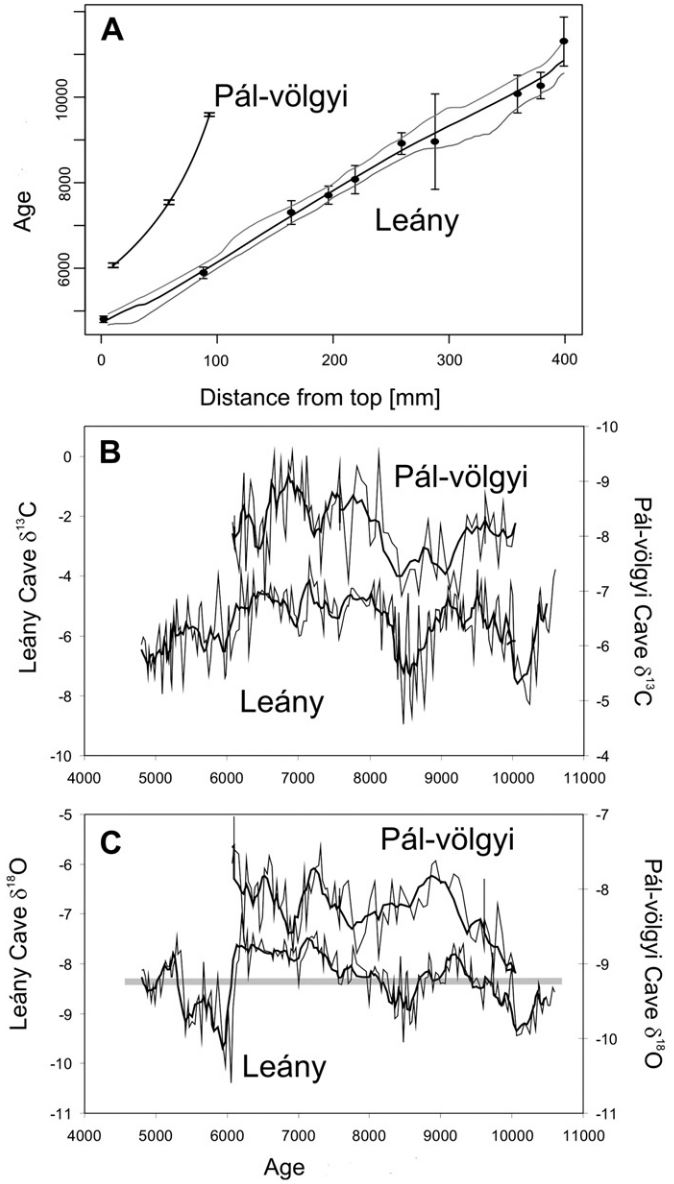

Fig. 2. Age-depth models (A), stable carbon (B) and oxygen (C) isotope compositions (in ‰ relative to V-PDB) of the stalagmites of the Leány and the Pál-völgyi Caves, Hungary, as a function of age. Horizontal grey line indicates the average oxygen isotope composition of the Leány stalagmite. Note that the $\delta^{13}C$ axis of the Pál-völgyi Cave stalagmite is reversed.

model. However, data for the part older than 10 ka will not be discussed in this paper. Fig. 2A shows the age–depth curve for the entire age data set, while the ages for the sampling points of isotope analyses were computed using the age model for selected age data (see age dates in Supplementary Table S2). Due to the small number of dates and the condensed nature, the Pál-völgyi stalagmite will be used just for comparison. Due to the limited age control, the age-depth model was constructed on the base of 2nd order curve fitting.

The carbon isotope compositions of the Leány and the Pál-völgyi Caves' stalagmites range from −9 to −3.5‰, and from −9.5 to −6.8‰, respectively (Supplementary material Table S2). Both stalagmite $\delta^{13}C$ records show systematic centennial fluctuations (Fig. 2B), but different long-term trends. Fig. 2B shows 10-point running averages that suggest an anticorrelation between the two records, although the limited age control on the Pál-völgyi stalagmite makes precise comparison of the two curves uncertain. Opposite to the $\delta^{13}C$ values, the $\delta^{18}O$ values are rather similar in the two records (Fig. 2C), both in terms of covered ranges and long-term trends. The centennial fluctuations observed for the $\delta^{13}C$ data appear also in the $\delta^{18}O$ values (Fig. 2C). Apart from the centennial fluctuations, positive and negative shifts appear in both $\delta^{18}O$ records. Positive peaks are found at 9.3 and 5.2 ka, a persistent positive plateau appears between 6 and 7 ka, whereas marked negative shifts occur at about 10 ka, 8.5, 6 and 5.4 ka (the average oxygen isotope composition of the Leány stalagmite is shown in Fig. 2C as a reference for the isotope shifts).

Weak C–O correlations appear in both records ($R^2 = 0.32$, $p < 0.000$ and $R^2 = 0.22$, $p < 0.000$ for the Leány and the Pál-völgyi stalagmite data, respectively, Supplementary material background information). Due to the condensed nature of the Pál-völgyi stalagmite, the conventional Hendy test (Hendy, 1971) was not possible due to difficulties in following and sampling the same lamina. However, the lamina thickness of the Leány stalagmite allowed us to conduct the conventional Hendy test at 6 deposition layers (Supplementary material background information). The correlation coefficients (R^2 values) for the C and O isotope compositions range from 0.01 to 0.94 (Pearson correlation coefficients and p values are listed in Supplementary material Table S2).

The H_2O contents (Supplementary material Table S2) show a systematic decrease from about 6000 ppm (at about 10 ka) to 400 ppm (at about 5 ka). On top of the long-term trend, the H_2O content rises at negative $\delta^{18}O$ shifts (Fig. 3A). Although the exact cause of this behavior is not known, the pattern is too systematic to be accidental, and raises the possibility of future application of the water content as a proxy of stalagmite growth conditions that could be explored in subsequent studies. The hydrogen isotope data of the Leány stalagmite (Supplementary material Table S2)

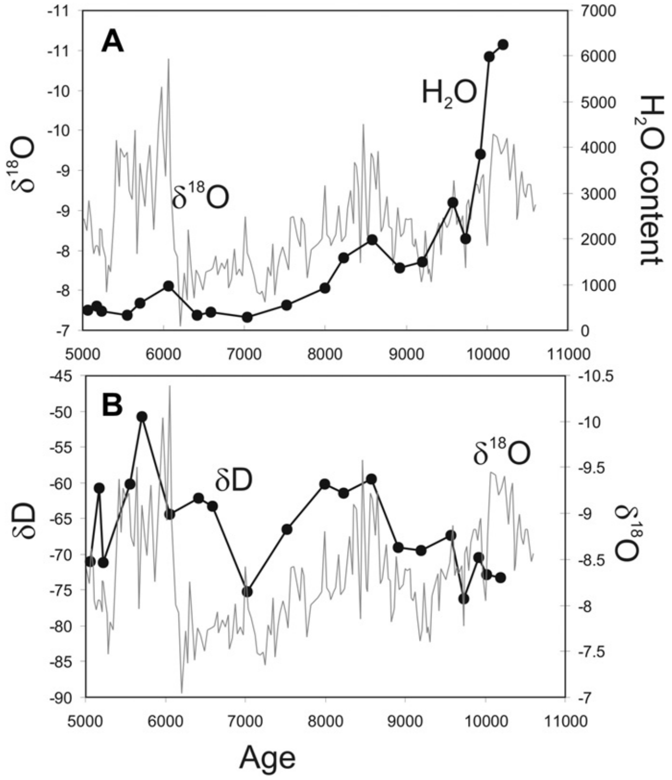

Fig. 3. (A) H_2O contents (in ppm), (B) hydrogen isotope compositions of inclusion-hosted water (in ‰ relative to V-SMOW) and $\delta^{18}O$ values (in ‰ relative to V-PDB) of the stalagmite of the Leány Cave, Hungary, as a function of age.

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range from −75 to −51‰, showing firm shifts. Although the δD data show no correlation with the δ^{18}O values at the sampling points ($R^2 = 0.03$), plotting the entire δ^{18}O and δD data set there is a tendency of low δ^{18}O associated with high δD values (Fig. 3B.).

5. Discussion

5.1. Evaluation of the observed stable isotope compositions as paleoclimate proxies

Stalagmite isotope records depend on a number of factors, including processes related to the local characteristics of the cave (Fairchild et al., 2006; Lachniet, 2009). One of the most severe influences that can modify the isotopic composition of dripwater and affect the precipitating speleothem calcite is kinetic fractionation induced by CO_2 degassing. Rapid CO_2 degassing would remove preferentially the light C and O isotopes from the dripwater and calcite–water equilibrium would not be reached. The kinetic fractionation would cause coupled positive C and O isotope shifts in the precipitating carbonate (Hendy, 1971; Mickler et al., 2004, 2006; Scholz et al., 2009). As CO_2 degassing takes place in the flowing dripwater film on the stalagmite surface, the precipitating carbonate's δ^{13}C and δ^{18}O values would increase with increasing distance from the growth axis. The kinetic effects may obscure the climate-related isotope signals, thus, stalagmites with positive δ^{13}C–δ^{18}O correlations are generally considered to be unusable for paleoclimate studies. A traditional selection criterion is the so-called Hendy test (Hendy, 1971), when C and O isotope analyses are conducted along growth laminae and in case of positive δ^{13}C–δ^{18}O correlation the isotope data are treated with caution.

The communication between the different parts of the Leány Cave, the rather large temperature fluctuation at the site of the Leány stalagmite suggesting a ventilated open system, and the slight positive δ^{13}C–δ^{18}O correlation (Supplementary material background information) may indicate kinetic fractionation during calcite precipitation. Thus, a traditional Hendy test was performed. From the six laminae analyzed from the Leány stalagmite (see Supplementary material background information), four gave R^2 values below 0.2, whereas the other two gave 0.75 and 0.94, suggesting again kinetic fractionation. However, even in those laminae with strong C–O isotope correlation, the δ^{13}C and δ^{18}O values do not increase with increasing distance from the growth axis, but rather 1–2 sampling points have positive isotope shifts. Taking the size of the dental drill used (0.6 mm), the stalagmite's average growth rate ($\sim$0.05 mm y^{-1} given by the stalagmite's length and age interval covered) and the difficulties in following the laminae exactly, it is likely that sampling points representing several years may not have perfectly overlapping years along the growth layer. The positive correlation due to potential outliers and the absence of monotonous δ^{13}C and δ^{18}O increase away from the growth axis collectively suggest that the results of the Hendy test should be treated with caution. The application of the Hendy test as a selection tool has also been challenged by recent studies (Dorale and Liu, 2009; Mühlinghaus et al., 2009; Day and Henderson, 2011). As Dorale and Liu (2009) pointed out, changes in climate conditions may also cause coupled positive δ^{13}C and δ^{18}O shifts in the precipitating calcite, and hence they suggested a Replication Test instead of or beside the conventional Hendy test. Correlations with other, independent proxy records generally strengthen the conclusions drawn from speleothem data that may reflect local rather than regional conditions (e.g., Schimpf et al., 2011). The most direct method of this assessment is comparison with other stalagmites collected in the vicinity, but in caves that show no significant ventilation. The Pál-völgyi Cave served as a possible location, where a stalagmite with a similar age range was found. The low R^2 value (0.22) for the Pál-völgyi stalagmite δ^{13}C and δ^{18}O correlation regression suggests that kinetic fractionation is not significant in this case. Comparing these C and O isotopic data with those observed in the Leány stalagmite found that the carbon isotopic compositions differ significantly from each other (Fig. 2B), whereas the oxygen isotope compositions are similar both in terms of absolute values and secular changes (Fig. 2C). As ventilation (kinetic fractionation) causes rapid CO_2 removal from the dripwater, the C isotope composition should be more affected than O isotope (cf. Mickler et al., 2004, 2006; Scholz et al., 2009). The discrepancy found in C isotope values and patterns of the two stalagmites and the relative enrichment in ^{13}C in Leány stalagmite would be in agreement with the assumed kinetic fractionation for the Leány cave, but other local effects (vegetation and soil activity, dripwater evolution, etc.) may also result in different cave behaviour in terms of carbon isotope fractionations. The observed difference, however, indicates that the δ^{13}C records can not be used as paleoclimate proxies until the exact mechanisms of carbon isotope fractionations are known for the cave sites. In contrast, the similarities in oxygen isotopic compositions indicate that the oxygen isotope compositons of Leány stalagmite was not significantly affected by kinetic fractionation and likely carry paleoclimatic information.

Another strong argument for the application of stalagmite isotope compositions as paleoclimate proxies is if a good agreement with well known reference records can be observed. For this purpose, one of the most important regional stalagmite records, the COMNISPA record was used, which is a composite δ^{18}O curve of several stalagmites from the Spannagel Cave, Austria (Vollweiler et al., 2006). The oxygen isotope compositions of the Leány and Pál-völgyi stalagmites are negatively correlated with the COMNISPA curve (Fig. 2C and Fig 4A). As the oxygen isotope composition of calcite depends not only on dripwater composition, but also on deposition temperature, the interpretation of these data alone would be difficult. Hydrogen isotope compositions may help as these directly reflect the composition of the dripwater. Interestingly, the δD pattern is opposite to the δ^{18}O trends of the Leány stalagmite with higher δD values at lower δ^{18}O periods (Fig. 3B). It follows from this behavior that the δD values should be positively correlated with the COMNISPA curve, and Fig. 4B shows that this is indeed the case. These observations collectively suggest that the oxygen isotope compositions of both the Leány and the Pál-völgyi Caves reflect climate conditions and can be used as paleoclimate proxies.

The COMNISPA curve displays a positive correlation with the HSG (Hematite Stained Grains) curve (Bond et al., 2001) that was interpreted as a sign of variations of the strength of the North Atlantic Oscillation (NAO) by Mangini et al. (2007). The similarities in isotope patterns of the stalagmites of the Leány, the Pál-völgyi and the Spannagel Caves suggest that the North Atlantic Oscillation (NAO) may have influenced the climate conditions in Central Hungary. Before further discussion of the isotope data obtained in this study, an overview of the NAO is given below.

5.2. The North Atlantic Oscillation and its effect in the Carpathian Basin

The North Atlantic Oscillation (NAO) is a major synoptic meteorological feature that affects climate conditions in northern and western Europe significantly. It is numerically defined by the air pressure difference between the dynamic centers located at the Azores and Iceland, called the NAO index (NAOI, Hurrell, 1995). At a high pressure difference (NAO+ mode) winter storm activity is stronger in Northern and Western Europe, inducing higher amount of winter precipitation in these areas, whereas at lower

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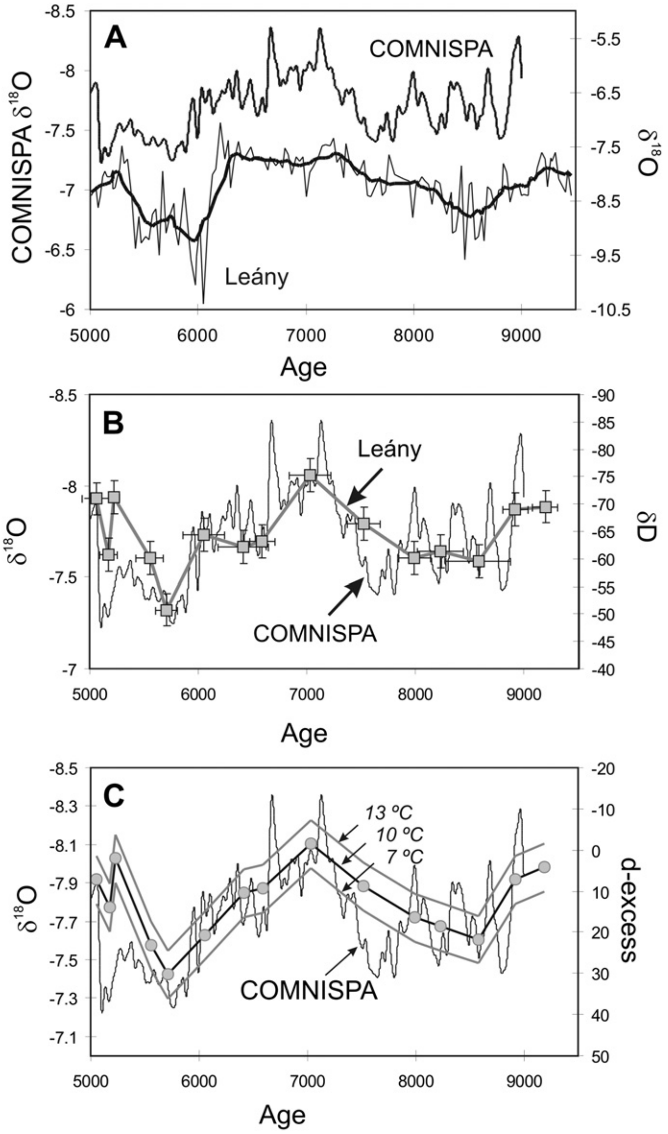

Fig. 4. Stable oxygen (A) and hydrogen (B) isotope compositions (in ‰ relative to V-SMOW) of the Leány Cave stalagmite, as well as calculated d-excess (C) values (in ‰). Uncertainties for δD data and age intervals covered by the individual analyses (see Analytical Methods and Supplementary Material Table S2) are shown for the H isotope compositions. The COMNISPA δ^{18}O record (Vollweiler et al., 2006; Mangini et al., 2007) is shown for comparison.

pressure difference storm activity and precipitation amount are increased in the Mediterranean (Hurrell and Deser, 2010). Storminess and humidity are associated with temperature changes, as during NAO+ mode more heat is transported to Western and Northern Europe. In a continent-scale study, Beranová and Huth (2007) determined correlation coefficients between NAOI and winter temperature and precipitation. Temperature-NAOI correlations are positive all over Europe, whereas precipitation has positive correlations with NAOI in Western and Northern Europe and negative ones in the Mediterranean region. The same scheme is presented by Langebroek et al. (2011), who used a general atmospheric circulation model (ECHAM5-wiso) to construct precipitation-temperature-NAOI correlation maps for Europe.

The Carpathian Basin has an intermediate position between the Western European and the Mediterranean regions. Domonkos (2003), Domonkos and Tar (2003), and Bartholy et al. (2009) have evaluated the temperature and precipitation data for Hungary for the period of AD 1901–2000 and correlated these meteorological data with the NAO index values. They found that cold and wet winters dominated during NAO− phases, while NAO+ phases are associated with warm and dry winters in the Carpathian Basin. It should be noted that temperature has a much weaker correlation with NAOI than precipitation (Matyasovszky, 2003). In terms of precipitation amount, the same relationship has been reported for Turkey for the period of 1929–1993 (Türkes and Erlat, 2003) and for Romania for the period of 1961–1996 (Tomozeiu et al., 2005). Evaluating discharge data for 510 European rivers, Wrzesinski and Paluszkiewicz (2011) showed that in the region of the Mediterranean (including the Balkan Peninsula and the Carpathian Basin) winter NAO indices are negatively correlated with river discharge data, in accordance with the precipitation amount – NAOI relationship described above.

These observations are based on correlations of instrumental records, and are consequently confined mainly to the 20th century. However, an increasing number of studies have shown that the North Atlantic Oscillation affected European climate in the whole of the last millennium. The periods of the Medieval Climate Anomaly and the Little Ice Age are thought to be characterized by persistent positive and negative modes of the NAO, respectively (Mann et al., 2009; Trouet et al., 2009; Luoto and Helama, 2010). Lake sediments from the region of the European Alps recorded changes in temperature, precipitation amount and composition in the last millennium (Teranes and McKenzie, 2001; Magny et al., 2011), and these studies suggested that in positive NAO mode (NAO+ stage) more winter precipitation from northern moisture trajectories was transported from the Atlantic Ocean to the region of the Alps. This means more winter precipitation with low δ^{18}O values, as exemplified by a negative δ^{18}O shift in lake sediments by Teranes and McKenzie (2001). The low oxygen isotope composition of the winter precipitation from northern moisture trajectories appears also in the dripwater and in the depositing stalagmite of the Spannagel Cave (Mangini et al., 2007), making the COMNISPA record a proxy for NAO activity.

5.3. Variations in moisture sources

The advantage of hydrogen isotope analysis of fluid inclusion-hosted water is that it supplies direct information on the water composition, or even on formation temperature following the concept of Zhang et al. (2008) that assumes that the meteoric water line equation is known. The meteoric water line is given by the equation $\delta D = a \cdot \delta^{18}O + b$, where "*a*" and "*b*" are location-specific constants. Domination of Atlantic moisture in the local precipitation without secondary evaporation during fallout would produce water compositions plotting along the Global Meteoric Water Line (GMWL; $\delta D = 8 \cdot \delta^{18}O + 10$; Craig, 1961). Periods with hot and dry conditions would enhance fallout evaporation such as observed in the Great Hungarian Plain, yielding Local Meteoric Water Lines with slopes between 7 and 8 and different *d*-excess values from −7.4 to +0.1‰ (Fórizs, 2005). Moisture originating from the Mediterranean region would have higher deuterium content compared to the Atlantic-sourced water that yields a separate meteoric water line (MMWL; $\delta D = 7.5 \cdot \delta^{18}O + 22$, Gat, 1980; Gat and Carmi, 1987). Using a selected meteoric water line, the measured hydrogen isotope compositions would determine the oxygen isotope composition of the water, which can be combined with the calcite composition to provide formation temperature using the fractionation equation (Friedman and O'Neil, 1977; see also Demény et al., 2010 for the selection of the fractionation equation). The formation temperatures obtained this way for the Leány stalagmite using the GMWL range from 4 to 22 °C, which is irrealistic in the cave environment. The large temperature range

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suggests that the meteoric water line varied with time and different MWL equations should be used for the temperature calculation.

The differences in these meteoric water lines is generally expressed using the deuterium-excess (also called *d*-excess) value that is defined as $d = \delta D - 8 \cdot \delta^{18}O$ (Dansgaard, 1964). Using the calculated $\delta^{18}O$ and measured δD values, the *d*-excess values could be determined by using temperature estimations. The present day cave temperature of the collection site of the Leány Cave was found to be 7.2–8.1 °C, whereas the long term annual mean temperature of the area of Hungary is about 10 °C. Cooling and warming events represented most likely less than 2–3 C° variation in the entire Holocene, thus ±3 C° variation is the extreme case. The range of 7–13 °C would certainly include the temperature variation in the Leány Cave, so this range was used in the calculations. The obtained *d*-excess data are plotted in Fig. 4C. It is apparent that the effect of temperature uncertainty is much less than the *d*-excess variation, thus, the observed pattern can carry valuable information and can be compared with other records. The *d*-excess values show a strikingly good positive correlation with the COMNISPA record (Fig. 4C), indicating that during periods with strong North Atlantic influence (NAO+ mode, low $\delta^{18}O$ values in the COMNISPA record) the rainwater approached the Local Meteoric Water Lines (at 7.5–6.5 ka), while weak North Atlantic periods (NAO−mode) enhanced the effect of Mediterranean moisture, bringing precipitation water with elevated *d*-excess values (9–8 and 6–5.5 ka periods, Fig. 4C). The inferred change in moisture origin is in accordance with the behaviour of the North Atlantic Oscillation, as during NAO+ phases moisture transport trajectories from the North Atlantic are shifted to the north, while during NAO-periods the trajectories are displaced toward the Mediterranean.

6. Conclusions

Stable hydrogen isotope compositions of inclusion-hosted H_2O, and carbon and oxygen isotope compositions of stalagmites of two caves from Central Hungary were studied in order to provide a speleothem-based paleoclimate record for the middle-early Holocene. Good replications of stalagmite isotope records from Leány and Pál-völgyi Caves, and their correspondence with the well known COMNISPA $\delta^{18}O$ record (Vollweiler et al., 2006; Mangini et al., 2007), suggest that the stable isotope time-series reflect regional climatic conditions at 10–5 ka. Variations in calculated *d*-excess values (from −7 to +30‰ obtained for a selected temperature of 10 ± 3 °C) imply the possibility of changing moisture sources. As the high *d*-excess values appear in periods when the COMNISPA $\delta^{18}O$ record indicate weak North Atlantic circulation activity (NAO−mode, Mangini et al., 2007), it can be concluded that during negative NAO phases the effect of Mediterranean moisture transport to the Carpathian Basin became more significant during the early- to mid-Holocene.

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Appendix A. Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.quaint.2012.05.035.

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