BABEȘ-BOLYAI UNIVERSITY

FACULTY OF BIOLOGY AND GEOLOGY

Late Pleistocene climate variability

recorded in stalagmites from Romania

Doctoral Thesis

Abstract

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1. Introduction

Although widespread in Europe and around the world, the use of speleothems in palaeoclimatology is still in its infancy in Romania. Over the last two decades, only few peer-reviewed publications and PhD theses studied the climate information recorded by speleothems from Romania (Lauritzen and Onac, 1995; Onac, 1996; Onac and Lauritzen, 1996; Lauritzen and Onac 1999; Onac, 2001; Tămaș and Causse, 2001; Onac et al., 2002; Constantin, 2003; Tămaș, 2003; Tămaș et al., 2005; Constantin et al., 2007).

This study aims at increasing the paleoclimate knowledge in Romania by investigating speleothems that formed during the Late Pleistocene and Holocene in three areas: Rodnei, Făgăraș and Mehedinți Mountains.

In the study of Holocene climate variability, we combine regional pollen-based temperature reconstructions (Davis et al., 2003) with new speleothem oxygen isotope data from Ascunsă Cave (Mehedinți Mountains, south-western Romania) and published records from the larger Mediterranean realm. Thereby, we attempt to quantitatively constrain the relative magnitude of hydrologic change in speleothem oxygen isotope records. We focus on the average millennial climate state prior and after a globally observed Holocene transition from approximately 6 ka to 4 ka, bracketed by climatic events (Mayewski et al., 2004), which is ubiquitous in lake-records across the Mediterranean (Roberts et al., 2008, 2011).

To study the climate changes in Romania during the Marine Isotope Stages 4-2 (MIS 4-2), we used two stalagmites from Ascunsă and Izvorul Tăușoarelor (Rodnei Mountains) caves, in order to draw a comparison between two climatically different regions. The main focus of this case study is the Marine Isotope Stage 3 (60-30 ka), a period of abrupt climate changes that were first documented from Greenland ice cores. In terms of speleothem studies, no European records to cover the entire MIS 3 period are available. Spötl and Mangini (2002) published an isotope profile from a stalagmite collected in Kleegruben Cave (Austrian Alps) that deposited between 57 and 46 ka. Genty et al. (2003, 2010) and Weiner et al. (2009) published isotope profiles from the Villars Cave (SW France), but these either present hiatuses or they do not offer a good resolution throughout MIS 3. Fleitmann et al. (2009) published an isotope profile from Sofular Cave (Turkey) that covers MIS 3 from 50 ka to present.

The importance of climate change studies over the MIS 3 is given by the necessity of understanding modifications in the Earth system that occur during large amplitude temperature changes over short periods of time.

The MIS 5e, a very important period of the Late Pleistocene, received only few studies in Romania. The published isotope data come only from speleothems and lack both precise age-depth models and good resolution. In this study we present two stalagmite isotope profiles from the Mehedinți and Făgăraș Mountains. While stalagmite CG1 (Mehedinți Mountains) formed over the period 132-115 ka, stalagmite M3-R2/1 formed during the warmest part of the last interglacial, in the alpine region. Studying past climate changes in high mountain environments is important because of their highly sensitive conditions when compared to the surrounding regions, giving them a character of climate islands (Hedberg, 1964). Therefore, climate archives in the high mountain areas are prone to better record large scale climate dynamics by being less subjected to regional influences.

2. Methods and Materials

Out of several dating techniques that are currently in use, only few can be applied to speleothems. These are radiometric methods (e.g., U-Th, ^{14}C) based either on the disintegration of certain radioactive nuclides or on the cumulated effect of their activities (luminescence, electron spin resonance). Because all our ages were generated using the U-Th method, this is the sole technique to emphasize below.

Natural radioactive decay chains are three and start with ^{238}U, ^{235}U and ^{232}Th, each of them ending with a lead stable isotope. Between the two end-members of these chains there are a number of daughter nuclides, that are also radioactive and which give the significance of U-series disequilibrium methods in geochronology (Ivanovich and Harmon, 1992).

Fractionation of different nuclides in these chains following natural phenomena brings the decay chains out of equilibrium, their re-equilibration being a function of time. Fractionation is defined as the partitioning of isotopes between two substances or phases of the same substance with different isotopic ratios (Hoefs, 2009).

Another important characteristic is that the parent isotope that stands at the beginning of the decay chain has a half-life and abundance greater than any of the daughters. Their half-lives are equal or larger than the Earth's age (^{238}U – 4.47 x 10^9 years and ^{232}Th – 1.41 x 10^{10} years). In the case of ^{235}U, the half-life is 7.04 x 10^8 years.

In this work we studied stalagmites from caves in Romania, whose most important characteristics are outlined below.

Ascunsă Cave is located on the eastern slopes of Mehedinți Mountains (45º N, 22°36'' E, 1050 m alt.) in south-western Romania (Fig. xxxxx). It is a 400 m long and over 200 m deep contact cave, mainly carved by stream erosion into a Turonian-Senonian wildflysch (melange) unit located below thick (40-100 m) Upper Jurassic-Aptian limestone bed (Codarcea et al., 1967).

The cave from Izvorul Tăușoarelor (hereafter Tăușoare Cave) is situated in Rodna Mountains, northern Romania (47º 26' 46'' N, 24º 31' 36'' E), at an altitude of 950 m. It is a long (>18 km) and deep cave (-356, +105 m; Papiu, 2007). The cave is hosted in Upper Eocene limestone that has frequent pyrite and black shale inclusions (Viehmann and Șerban, 1963; Onac, 1987).

M3-R2 Cave is located at 2435 m a.s.l. in the Făgăraș Mountains (Southern Carpathians, Romania, 45° 34' 26" N, 24° 38' 57" E) at relatively shallow depth (less than 10-15 m below surface). The cumulated length of its galleries is 86 m and the deepest point 20 m (Giurgiu, 2006). The cavity develops in a stripe of crystalline limestone interbedded with amphiboles and garnet-rich micaschists (Codarcea and Stancu, 1968).

The Cave from Godeanu Quarry (abbreviated GC) is located at 45º 01' 27'' N and 22º 39' 50'' E, at 620 m asl, just a few meters below the surface, in Upper Jurassic-Aptian limestone (Codarcea et al., 1967). It was opened during quarrying activities and it is in fact a geode of very small size (0.5 x 0.5 x 2 m), inaccessible to humans.

From the caves detailed above, we analyzed five stalagmites, which were dated using the U-Th method and from which stable isotope samples were analyzed in order to reconstruct climate characteristics.

Stalagmite POM2 (Ascunsă Cave) is 77.4 cm long (assigned distances from -0.2 cm at the base to 77.2 cm at the top) and composed of well-laminated and compact white calcite. In total, fifteen U-Th samples were measured from the entire length of the stalagmite. A total of 151 stable isotope samples were hand drilled at 5 mm resolution using a 0.5 mm drill bit.

Stalagmite POM1 (Ascunsă Cave) was retrieved from the White Chamber, at about 80m from the entrance and 40 m below the surface. It is ~30 cm long and composed of alternations of dense dark calcite and lighter, less dense calcite. The growth axis presents several slight direction changes. Stalagmite POM1 was sampled in such a way to bracket the most important changes in axis orientation and petrography. Due to the low U content, these samples weigh around 100 mg. For stable isotope analysis, POM1 was sampled at 0.3 mm resolution (900 samples).

Stalagmite 1152 (Tăușoare Cave) was retrieved from the Chamber of the Balls, is 27 cm long and is formed by dark-brown dense calcite. The growth axis of stalagmite 1152 is stable for the first 23 cm (measured from base), shifting its direction by a few degrees and then maintaining it for the last 4 cm.

As stalagmite 1152 presents only few visible discontinuities or significant layers, the sampling was done at 2 cm resolution, using a 0.5 mm drill bit. Only clear changes in orientation or layer coloring at 19 and 23 cm were bracketed by twin samples in order to test for hiatuses. Compared to the base and top samples, that weigh ~50 mg, all others weigh between 5-10 mg. For stable isotope analysis, the stalagmite was micromilled at 0.25 mm (1069 samples).

Stalagmite M3-R2/1 (M3-R2 Cave) was collected from the main chamber of the cave, 15 m from the entrance. It is 5 cm long and composed of compact, white translucent calcite and presents a thin outer crust.

Seven U-Th samples averaging 100 mg were hand drilled at equal intervals along the growth axis. For stable oxygen and carbon isotopes, 197 samples were micromilled at an interval of 0.25 mm.

The CG1 (CG Cave) stalagmite is 20 cm long and is composed of well-laminated dense calcite of varying light colors. It was sampled from a debris and red clay accumulation resulted after the destruction of the front part of CG Cave by quarrying. Two U-Th ages were obtained on its base and top. For stable isotope analysis, 200 samples were micromilled at a resolution of 1 mm.

3. Climate changes over the past 8,200 years reconstructed from a southern Romanian speleothem

The Holocene growth model of stalagmite POM2 uses eleven U-Th age estimates with typical dating uncertainties ranging between 1 and 6 % (2 σ). The stalagmite was active at the time of sampling, thus the age at the top (77.4 cm) is considered to be 0 ka and used as additional tie point in the growth model calculation.

The two isotopic profiles display three main features: an apparent lack of trend during the Early Holocene (8.2-6 ka), a trend towards higher values during the Middle Holocene (6-4 ka) and a slightly decreasing trend during the Late Holocene (4-0 ka), interrupted by a short period of low isotope values from 3.2 to 3.0 ka (Fig. 6).

To better place in a regional context our stable isotope values, we compared the δ^{18}O profiles of stalagmites POM2 (this study), PP10 (Constantin et al., 2007), PU-2 (Onac et al., 2002) and V11-22 (Tămaș et al., 2005) with those calculated by McDermott et al. (2011) for low altitude European caves at 22° E longitude (Fig. 1).

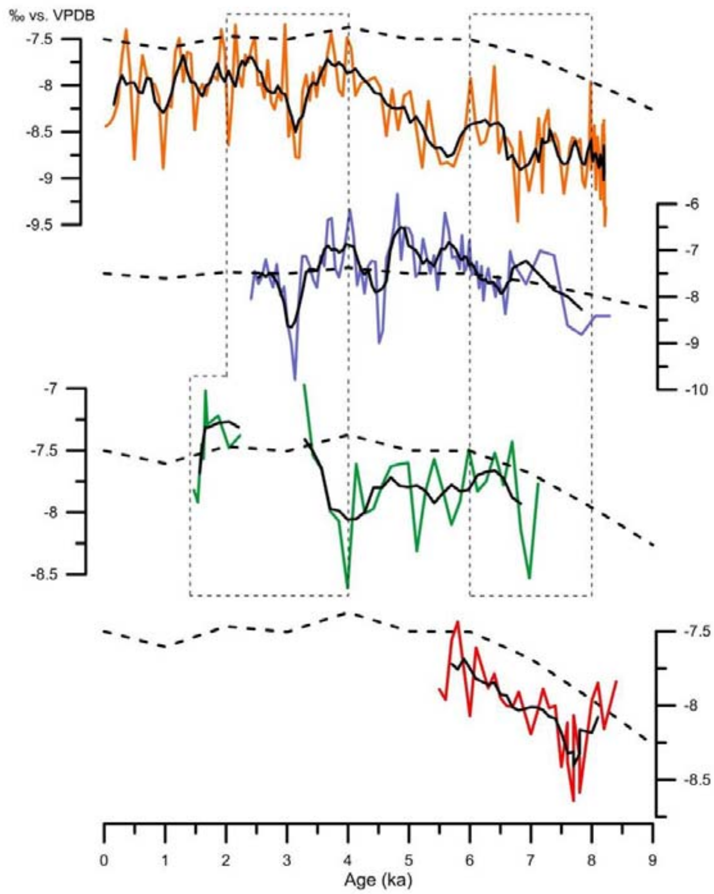

Fig. 1 – Comparison between δ^{18}O in stalagmites POM2 (orange), PP10 (blue), PU-2 (blue) and V11-22 (red) and isotopic values predicted by McDermott et al. (2011) for 22° E longitude (dashed line). Dashed line boxes represent the time windows (2-4 and 6-8 ka) on which the average isotopic values were calculated.

For the Mid Holocene, we wanted to see if the trend observed in our data was produces only by temperature variations or if there were other factors involved. The first step in constraining the temperature effect on oxygen isotopes is to assess the change of meteoric water δ^{18}O ($\Delta^{18}O_{mw}$) in response to variations of air temperature. Thus, we calculated the magnitude of annual (TANN), summer (MTWA) and winter (MTCO) temperature change over the mid-Holocene (6-4 ka) using pollen temperature reconstructions of Davis et al. (2003). For this, we used two time windows that are delimitating our study interval, namely 8-6 and 4-2 ka, and have the role of overcoming inconsistencies in chronology and time lags of environmental response between different regions. By multiplying these values with the temperature dependence factor of Rozanski et al. (1993), we obtained the theoretical change in summer and winter rainfall $\delta^{18}O$ over the course of the mid-Holocene.

The second step in constraining the temperature effect was to resolve how the change in cave air temperature impacted the isotopic signature of the precipitated calcite ($\delta^{18}O_c$). We thus employed the equation of Tremaine et al. (2011) to calculate $\delta^{18}O_c$ for the 8-6 ka and 4-2 ka windows:

$$1000 \ln \alpha = 16.1(103T-1) - 24.6 \qquad (3)$$

Further, we determined the combined isotopic effect that temperature variations had both during rain/snow events and during calcite precipitation.

The results of the calculations are plotted in Figure 2. From this graph, it is obvious that measured isotope values from sites affected by the Western Mediterranean (Clamouse, Renella and COMNISPA) or the Atlantic (Urșilor) are within the temperature constrained values.

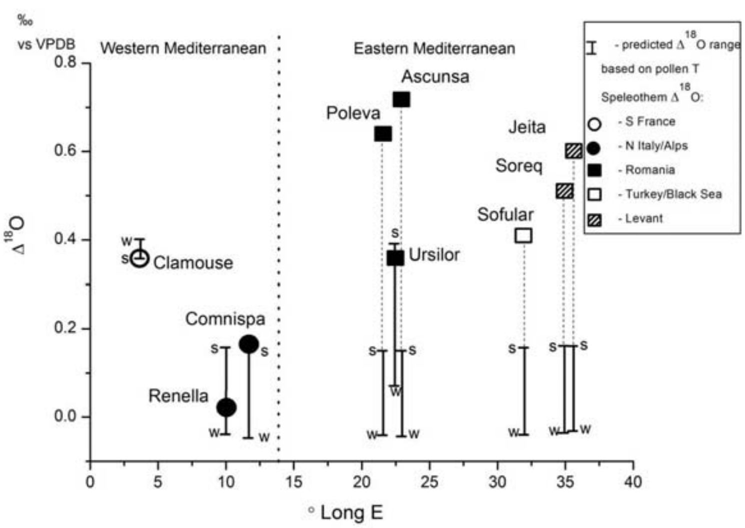

Fig. 2 – Comparison of isotopic changes in stalagmites from different longitudes in Europe and predicted isotopic change in winter and summer precipitated calcite, in the interval 4 to 6 ka.

At the same time, sites in Turkey (Sofular), Israel (Soreq), Lebanon (Jeita) and southern Romania (Poleva and Ascunsă) that are affected by the Eastern Mediterranean, plot higher than the constrained ranges. It appears that, during the mid-Holocene, the Eastern Mediterranean and its surroundings witnessed, apart from rising temperatures, an alteration of the hydrological cycle.

This change could be due to local effects, but nevertheless occurred at regional scale. One such effect could be the evaporation of rainwater prior to or shortly after infiltration in soil. In the absence of evaporation, rainwater would infiltrate through soil and epikarst preserving its original $\delta^{18}O$ signature. A likely regional factor that could have had the same consequence on measured speleothem $\delta^{18}O$ during the Middle Holocene would be the isotopic enrichment of one of the Mediterranean.

The trend towards higher $\delta^{13}C$ values observed during the mid-Holocene in the POM2 stalagmite overlaps the one observed for $\delta^{18}O$; it very likely reflects a reduction in plant activity as a consequence of decreasing water availability. This assumption implies an increase in dry conditions at the cave site, either annually or during the vegetation season. A comparison with stalagmite PP10 (Fig. xxxxxxx) reveals similar isotopic trends in both records, providing supporting evidence towards a regional decrease of water resources during the mid-Holocene.

4. Climate variability during the marine isotope stage 3 in northern and southern Romania

Following the growth model calculation, it is shown that stalagmite 1152 (Tăușoare Cave) formed between 67.7 ka and present, with a hiatus from 20 to 15 ka. From 67.7 to 30.4 ka the growth rate was 2.6 µm/year and increased to 8.6 µm/year between 30.4 and 20 ka. The sampling resolution allowed us to attain an average temporal resolution of the isotope record of ~100 years between 67.7 and 30.3 ka, and ~30 years between 30.4 and 20.0 ka.

Stalagmite POM 1 formed between ~47 and ~11 ka. In this study we are focusing our analysis on the best dated period, 47.2 to 30.8 ka. The average resolution of the POM 1 record is 22 years.

At Tăușoare Cave, MIS 3 $\delta^{18}O$ values are almost identical to the Late Holocene ones. We interpret the high $\delta^{18}O$ values recorded during MIS 4-2 to likely reflect reduced aquifer recharge and cave dripping over the cold season and consequently, mainly summer conditions.

Only a handful of published stalagmite studies report high $\delta^{13}C$ values like those at Tăușoare Cave. Among them, Kleegruben, Spannagel and Milchbach caves in the Austrian Alps. As these caves had at times no soil cover, hence no soil CO_2 available (Holzkämper et al., 2005; Spötl et al., 2006; Spötl and Mangini, 2007; Luetscher et al., 2011), the values were interpreted to reflect bedrock-derived CO_2 (via sulfuric acid dissolution of limestone). At Antro del Corchia Cave (Italy), such elevated carbon isotope values mirror poor soil development (Zanchetta et al., 2007).

The geological setting at Tăușoare Cave is prone to a similar chemical mechanism to that in the Alps. The chemical reactions imply the oxidation of pyrite (FeS_2) present in limestone and its interbedded black shales to sulfuric acid (H_2SO_4). As percolating water is acidified by hydrogen ions, it dissolves the surrounding carbonate host rock, releasing bicarbonate ions (HCO_3^-).

Therefore, under this scenario there is no need of biogenic CO_2 for either dissolving bedrock or precipitating speleothems. This implies that during MIS 4-2, soil cover might have not been present above the cave. As the $\delta^{13}C$ value of the Eocene carbonate host rock is 0.1‰ and is believed to have remained stable, it means that the positive values recorded throughout MIS 4-2 might have been kinetically enriched. Thus, in the absence of soil produced CO_2, kinetic fractionation could be responsible for $\delta^{13}C$ variability.

A second scenario, that includes the existence of soil cover, also needs the presence of the above mentioned chemical mechanism and of kinetic fractionation. In a dissolution process that relies only on soil derived CO_2, $\delta^{13}C$ would have values close to -3 - -4‰, but at Tăușoare Cave they would be further enriched to the positive values measured by us. In this scenario, the variability of $\delta^{13}C$ could be imposed by soil CO_2 production processes. Following this scenario, we can argue that during MIS 4-2, at Tăușoare Cave soil processes were weaker than in the Holocene, when we observe lower values, thus a higher contribution of plant-derived CO_2.

The scenarios outlined above reveal that the controls of $\delta^{13}C$ values at Tăușoare Cave are more complex than usual. One of these factors is the extent of water-rock interaction, which is related to the amount of infiltrating water (a reflection of the annual rainfall) and residence time of percolating water. In turn, these are controlling the magnitude of sulfide oxidation and host-rock derived HCO_3^-.

While the correlation coefficient (R^2) along the entire growth axis of stalagmite 1152 is 0.12 (n=1055) and would be indicative of low kinetic fractionation, a more detailed analysis realized as a running R^2 on 50 values, reveals that, for the studied period (67-20 ka), the two proxies are well correlated, indicating the presence of kinetic fractionation conditions at the stalagmite growth site.

Based on the fact that stronger kinetic fractionation has a hydrological component (slow drip rate and low relative humidity) and follows a chemical process controlled by water availability, we assumed that high values of the correlation coefficient reflect less rainfall at Tăușoare Cave site. Hence, this second-order proxy (derived from two first order proxies: $\delta^{18}O$ and $\delta^{13}C$) can be used as an indicator of relative rainfall amount, with high values being indicative of kinetic fractionation, hence drier conditions at the cave site.

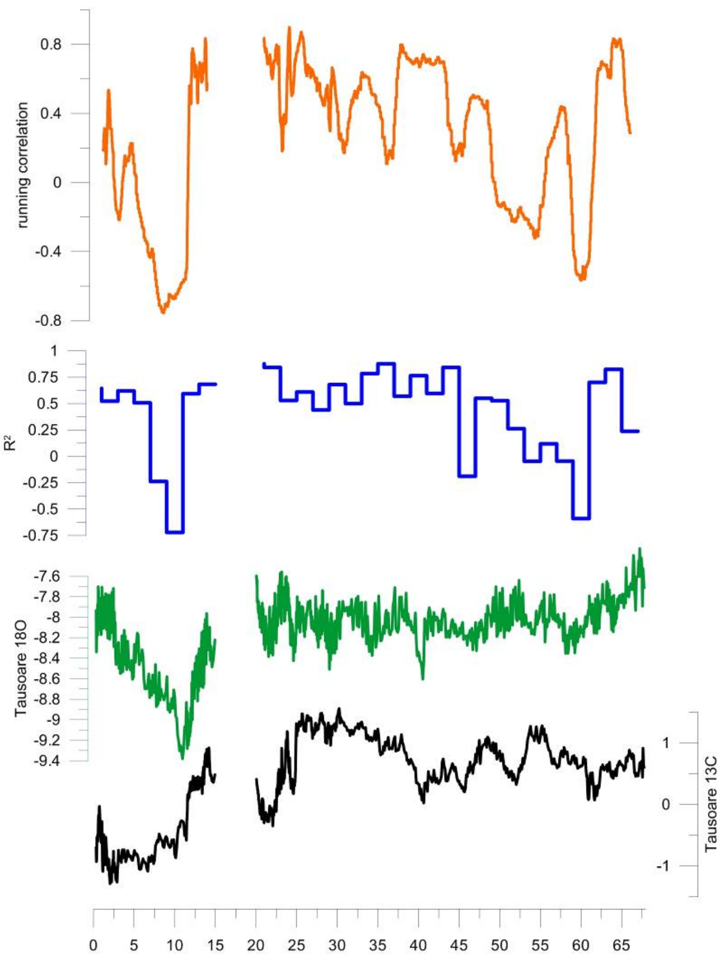

Fig. 3 – Tăușoare Cave $\delta^{13}C$ (black) and $\delta^{18}O$ (green) profiles, correlation coefficient (R^2) on 2000 year segments (blue) and 50 points running R^2 (orange).

Following our interpretation, it appears that the interstadials were generally wetter than stadials, with GIS 17 being the wettest in our record. Wet interstadials also include GIS 14, 12, 8, 4.1 and 2, but they fall on a consistently descending trend in rainfall amount.

Oxygen isotope values recorded during MIS 3 at Ascunsă Cave are close to the Holocene ones and are inversely correlated to global temperature records such as NGRIP (Andersen et al., 2006). Following the same judgment as in the case of Tăușoare Cave, we suggest that at this site calcite precipitation took place mostly during summer, as winter's frozen soil would have prevent cave drip water recharge. Thus, $\delta^{18}O$ generally reflects the isotopic signature of Atlantic derived summer meteoric water and the low variability of summer temperatures inferred by Ampel et al. (2010) in Western Europe during MIS 3. During two Greenland stadials, $\delta^{13}C$ values reached the upper limit of what is expected for carbonates deposited in equilibrium with CO_2 respired from C_3 plants (McDermott, 2004), in this case 2.9‰. Stalagmite POM 1 ceased its growth during H3, when the highest values occurred.

The fact that $\delta^{18}O$ is inversely correlated with annual temperatures and presents high isotope values during cold periods shows that seasonality of infiltration is the main driving force behind isotopic change. A possible scenario to produce this variability is represented by variations in the length of the cold season. During warmer years, the cold season could be shorter, thus allowing for more depleted waters to infiltrate in late autumn/early spring, while during colder years infiltration could be limited only to the warmest months, resulting in higher isotope values. Thus, for MIS 3, we can use the $\delta^{18}O$ data from Ascunsă Cave to infer changes in seasonality and annual temperature variability.

The low signal variability hampers the precise observation of Dansgaard/Oeschger cycles, except for the D/O 8 (GIS 8) at Ascunsă Cave, which is clearly expressed in both $\delta^{18}O$ and $\delta^{13}C$ time series. This interstadial starts with a sudden and large (~5‰) $\delta^{13}C$ rise, while oxygen values rise only by 1.5‰. During D/O 8, changes in oxygen values precede those in $\delta^{13}C$ by about 200 years. This rise in both isotope values was interrupted briefly for 50-100 years, a common feature in other isotope records such as NGRIP or Sofular (Andersen et al., 2005; Fleitmann et al., 2009). During GIS 8, the annual temperatures are the highest and most constant in the record, implying that environmental conditions were almost similar to Holocene ones at Ascunsă Cave.

Heinrich events are better expressed in our speleothem records, revealing different impacts of these rapid climate events. HE 6, probably the least studied (Hemming, 2004), appears to have lasted from ~62.5 to ~60.5 ka, but an exact estimation of the length is difficult due to the lack of clear isotopic expression.

In both our stalagmite isotope time series, the beginning of the HE 5 appears around 44 ka, similar to a clearly defined event on both $\delta^{18}O$ and $\delta^{13}C$ from Sofular Cave. In Ascunsă

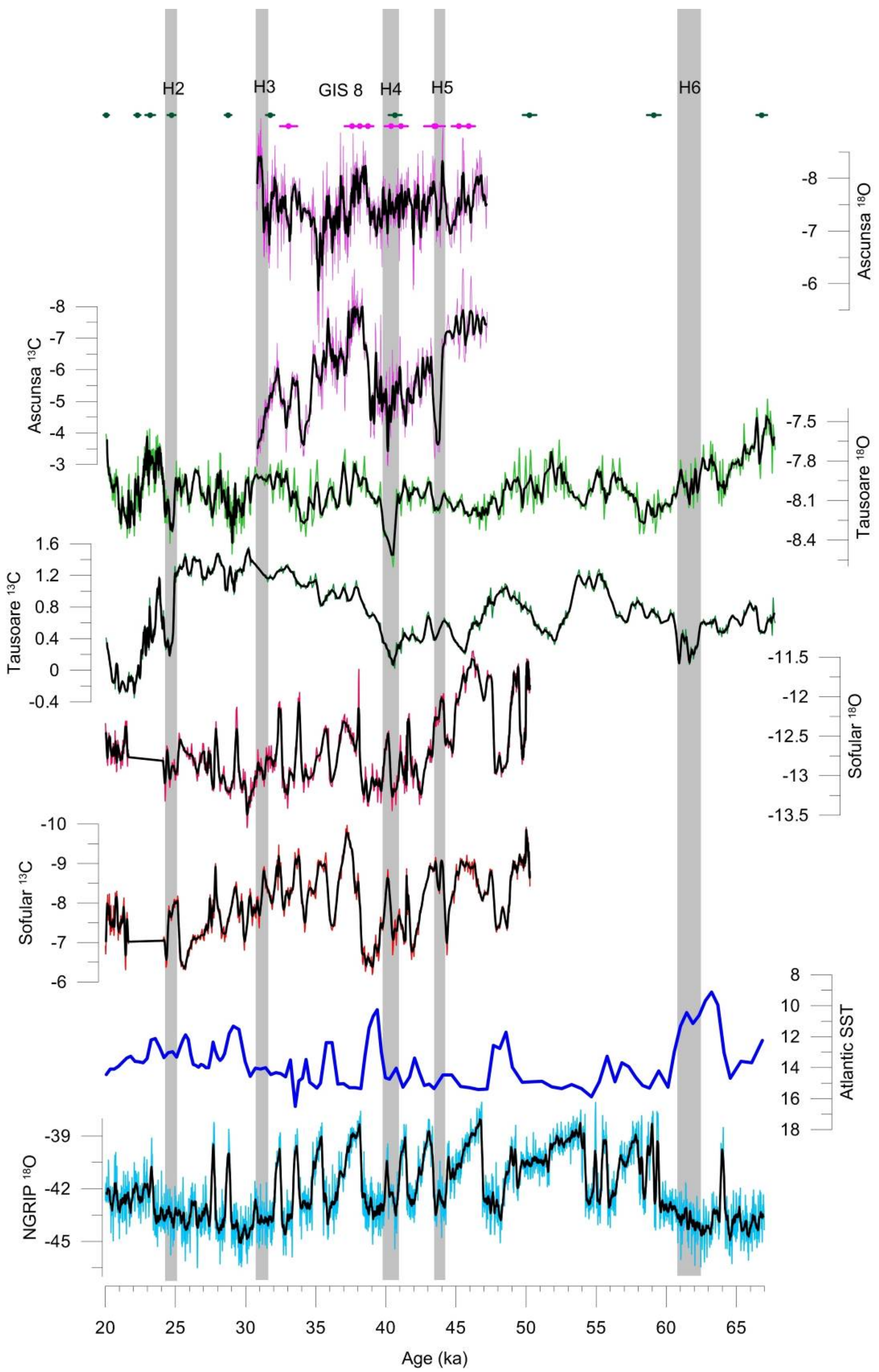

Fig. 4 – Comparison between stable isotope profiles in Tăușoare and Ascunsă caves and other records.

Cave, HE 5 produces the strongest signal in the $\delta^{13}C$ record, appearing as a rapid 3.5‰ enrichment. At Tăușoare Cave the isotopic signal related to HE 5 is muted, although still visible and simultaneous with the signal at Ascunsă Cave.

The strong depletion event at 40 ka in Tăușoare $\delta^{18}O$ data could be linked to ^{18}O depleted surface waters in the North Atlantic. This episode most probably occurred during the Heinrich Event 4 due to rapid and strong input of ^{18}O depleted melt water from the disintegrating Laurentide ice-sheet. These melt waters were responsible for cooling of the north-Atlantic surface waters, further lowering the isotopic value of the vapor originating here.

The HE 3 (~31 ka, Hemming, 2004) is difficult to recognize at Tăușoare Cave but is well-defined at Ascunsă Cave as a very cold event that halted the stalagmite growth, similar to stalagmites from Villars Cave (Genty et al., 2003, 2010). As mentioned before, extremely low values are recorded at this time in the $\delta^{13}C$ data set.

Finally, the HE 2 is easily discernable at Tăușoare in both carbon and oxygen isotope values and is centered at 24.5 ka. It appears as a ^{13}C- and ^{18}O-depletion of 1‰ and 0.4‰, respectively.

The isotope data from Tăușoare Cave were analyzed using the Morlet wavelet in order to reveal periodicities in their variability (Fig. xxxx). The most striking feature is the appearance of a 2300 year periodicity during the MIS 2, from ~31 to 20 ka, in both $\delta^{18}O$ and $\delta^{13}C$ time series. The 2300 year cycle is well documented from Greenland (Mayewski et al., 1997; Rohling et al., 2002) and the Aegean Sea; at this latter location the cycle is actually of ~2500 years (Rohling et al., 2002). The period between 31 and 20 ka is coincident with a peak in summer insolation at 45ºN (Laskar et al., 2004) and a three-fold increase in the depositional rate of the stalagmite, indicating a possible summer climate modulation by solar activity during high insolation periods. The impact of increased solar activity also appears to have an effect on the speleothem deposition processes.

5. Climate variability at middle and high altitudes in Romania during MIS 5e

As presented in a previous chapter, for this case study we used two stalagmites, CG1 (Mehedinți Mountains) and M3-R2/1 (Făgăraș Mountains).

The base and top samples of stalagmite CG1 returned U-Th ages of 131.9 ± 1.9 ka and 113.7 ± 2 ka, respectively, revealing that it formed during the whole extent of MIS 5e.

The oxygen isotopes follow a descending trend throughout the growth period, interrupted by several millennial-scale events characterized by isotopically heavy carbonates. Among these, the most important are between 125 and 122 ka and from 121 to 119 ka. The carbon isotope values follow an ascending trend throughout the growth period, interrupted by periods with low $\delta^{13}C$ values, of which the most significant one is from 123 to 121 ka. It is worth noting that both $\delta^{18}O$ and $\delta^{13}C$ profiles are virtually equal in absolute values and amplitude with the Holocene values of stalagmite POM2 from the nearby Ascunsă Cave, which lies about 5 kilometers to the south-west. This could imply comparable conditions during MIS 5e and Holocene in this area.

The M3-R2/1 $\delta^{18}O$ values show low variability in the first ~400 years of growth but afterwards they begin to fluctuate, illustrating relatively high amplitudes. Overall, the values of the whole record show no trend and have an average of -8.4‰. Around 124 ka there is an increase towards higher values, resembling a similar step recorded at almost the same time in France (Couchoud et al., 2009) and Italy (Drysdale et al., 2009). The $\delta^{13}C$ values show low variability during the first ~400 years and a weak trend towards lighter values (from -7.8‰ to -8.4‰). From 125 ka onward, the time series is punctuated by a number of short-term fluctuations of up to 2.2‰ on a trend towards heavier values (from -8.4‰ to -6.0‰).

Although the two isotopic profiles of stalagmite CG1 have different trends, they seem to correlate during major isotopic events. In order to check for the degree in which they do so, we calculated a nine point running correlation coefficient (R^2) over the whole data range (Fig. 5). All kinetic processes lead to high isotope values in speleothems, but in stalagmite CG1 the highest correlation is encountered during events with low δ values, clearly showing that kinetic fractionation did not play an important role in CG1's isotope variability. Ruling out this possibility, we argue that both $\delta^{18}O$ and $\delta^{13}C$ are reflecting regional climate variability and are less influenced by local factors. In Fig. 5, one can see that summer insolation at 45ºN is the main driving force for $\delta^{13}C$ values during MIS 5e. Although it appears that higher organic activity (denoted by lower isotopic values) parallels the increase of insolation, it is not readily discernable if summer insolation controlled this productivity directly through temperature or via increased precipitation.

Oxygen isotope values of stalagmite M3-R2/1 (~2400 m asl, ~ –8.4‰) are lower than the average MIS 5e value of the LFG-2 stalagmite from the Western Carpathians (Lauritzen and Onac, 1999, ~550 m asl, ~ –7‰) and equal to that of stalagmite CG1 (~620 m asl, –8.4‰). The difference between LFG-2 and M3-R2/1 could only partially be explained by the altitude effect (Clark and Fritz 1997; Lachniet 2009 and references therein), as this effect would imply isotopic differences as large as 5‰. Even if the altitude effect did play a role, it is probably hidden by another effect, such as seasonal isotopic variations of the percolating waters. These changes are driven by a decrease or lack of infiltration and thus carbonate deposition during winter, with the bulk of percolation and calcite precipitation occurring during the warm season. Thus, the high isotope values of M3-R2/1 reflect a decrease or lack in winter contribution to annual infiltration and hint towards a longer cold season and snow cover compared to other lower altitude sites. The fact that the oxygen isotopic signal shows reduced amplitude throughout the studied interval, points to the fact that the extent of the cold season did not change dramatically during this period.

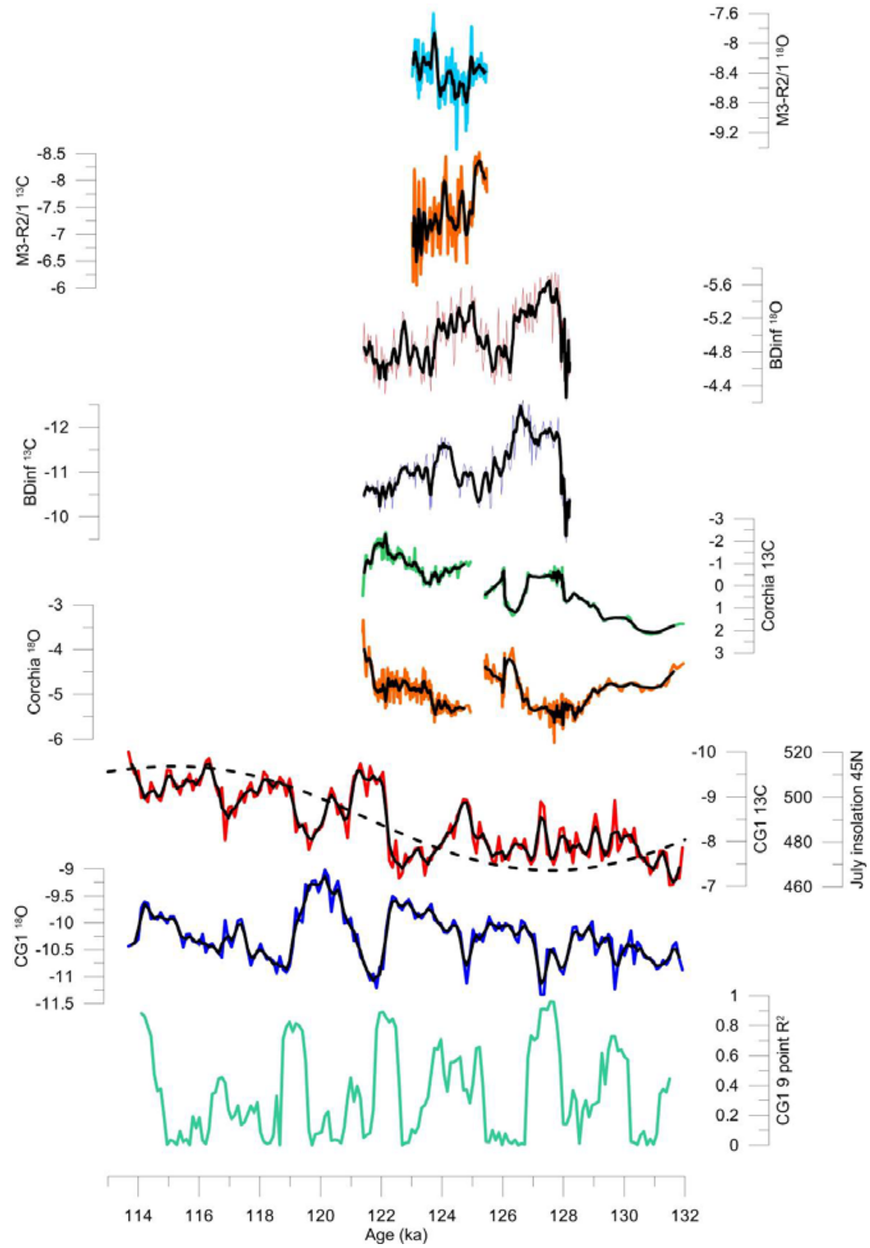

Fig. 5 – Comparison between stalagmites from CG, Corchia, Bourgeois-Delunay, and M3-R2 caves. Also plotted are the 9-point running R2 for stalagmite CG1 and July insolation at 45 N. Note that some scales are reversed!

In the $\delta^{13}C$ profile of stalagmite M3-R2/1, relatively high values compared to stalagmite CG1 imply less developed vegetation cover, whereas the ascending trend suggests decreasing plant activity throughout the entire growth period. Between 123.5 and 123.0 ka, we visually identified three well-defined centennial-scale cycles with a period of ~150 years.

The period of higher isotope variability that starts at ~125 ka at M3-R2 Cave coincides with high oxygen isotope values at CG Cave and with the sea level interval (SLI) #4 (~124-122 ka) defined by Hearty et al. (2007) as a stagnation of the interglacial level at ~ +3 - +4 m, after a drop that occurred around 125 ka (SLI #3). In the Red Sea, Rohling et al. (2008) identified a period of continuous sea level rise between 124.7 and 123.3 ka, with a rate of 1.7 m per century, likely triggered by global temperatures that were 2°C higher than today. The compilation of global sea level indicators by Kopp et al. (2009) identifies a peak in global sea levels at 124 ka. During the same time interval, high deposition rates were recorded by speleothems in France (125.3-123.8 ka, Couchoud et al., 2009), Italy (125.0-123.7 ka, Drysdale et al., 2009) and Israel (127-124 ka, Bar-Matthews et al., 2003).

The Multi Taper Method (Percival and Walden, 1993) revealed that, at 99% confidence level, 80- and 167-year cycles are visible in the carbon values, while oxygen data contain periods of 62 and 107 years. The Morlet wavelet analysis (Torrence and Compo, 1998) shows the locations where these periodicities are stronger. In case of carbon values it shows centennial cycles present at the end of the growth period, consistent with their visual identification. The 80-year carbon cycle is stronger in the middle part of the record, whereas the oxygen data show significant periodicities only in the first half of the record.

The 80-year periodicity in the $\delta^{13}C$ record is close to the Gleissberg cycle seen in solar proxy records (Sonet et al., 1990), suggesting a control of solar variability on the $\delta^{13}C$ signal. While the changes in the solar irradiance associated with solar cycles are generally too small to explain the observed changes in the climate system, various mechanisms have been proposed to link solar variability with climate. A plausible link between solar variability and climate in Romania is through the North Atlantic Oscillations which influences Romania's winter climate (Bojariu and Giorgi, 2005) and has been shown to respond to solar forcing.

In the oxygen data, the 62-year periodicity can be linked to the Atlantic Multidecadal Oscillation, a feature that is believed to reflect North Atlantic – atmosphere variability on a scale of 55-70 years (Knudsen et al., 2011; Ionița et al., 2013). The other strong periodicity in oxygen values, 107 years, could be related to solar variability, as it is similar to the 105.75 year cycle which, like the Gleissberg cycle, is a modulation of the 11 year sunspot cycle (Damon and Jirikowic, 1992).

6. Conclusions

In this thesis we studied climate variability during the Late Pleistocene and Holocene in Romania based on stalagmite stable isotope analysis from caves in Rodnei, Făgăraș and Mehedinți Mountains.

The stable isotope record of POM2 stalagmite allowed us to identify centennial to millennial climate change during the Holocene. Between 6 and 4 ka, $\delta^{18}O$ gradually shifted towards higher values and, by using a novel approach, we determined that this shift was not reflecting only rising temperatures, but that other factors might have been involved.

Our approach is based on using pollen-based temperature reconstructions to constrain the isotopic values of calcite that would vary controlled only by temperature. Isotope values measured on stalagmite POM2 were then compared to these constrained values, revealing that the mid-Holocene enrichment was 0.56‰ greater than the maximum values produced only by rising temperatures. Further, we extended the calculation to other speleothem records in the Eastern and Western Mediterranean and western Romania. We show that, during the Middle Holocene (6-4 ka), in Western Mediterranean speleothems $\delta^{18}O$ responded only to temperature change. At the same time, in the areas influenced by the Eastern Mediterranean, other processes such as enhanced evaporation rates or the gradual enrichment in the isotopic composition of Eastern Mediterranean surface waters could have contributed to the observed isotope change.

Additional evidence is conveyed by comparing the $\delta^{13}C$ profiles from Ascunsă and Poleva caves, both showing a gradual and regional decrease in water availability to plants.

Finally, we tackled two rapid climate changes, at 8.2 and 3.2 ka. In POM2 stalagmite, the 8.2 ka event is characterized by a very high growth rate, whereas the $\delta^{18}O$ and $\delta^{13}C$ values show low variability; however the 3.2-3.0 ka period is well defined by a 1.5‰ depletion in $\delta^{18}O$. By constraining the isotopic variability produced by temperature, we show that during the 8.2 ka event Ascunsă and V11 caves were influenced only by summer conditions. During the 3.2 ka event, Ascunsă Cave isotope data reflect changes in winter temperature and a lower and possibly more depleted input of Mediterranean moisture.

For the study of MIS 3 climate variability, we used two stalagmites from Ascunsă and Izvorul Tăușoarelor (Rodnei Mountains) caves and drew a comparison between two climatically different regions. The study of these two coeval stalagmites from northern and southern Romania reveals significant climate fluctuations during MIS 4-2. We argued that at both caves the aquifer recharge was restricted to the warm season and that variability in oxygen isotopes values at both caves is related to seasonality variations.

We used the correlation coefficient between carbon and oxygen isotopes at Tăușoare Cave as a second-order proxy for warm season rainfall amount. There is a good agreement between this derived proxy and the oxygen isotope signal at Villars Cave (France) and it also matches reconstructed annual precipitations from the Velay sequence, France. Following our interpretation, it appears that the interstadials were generally wetter than stadials, with GIS 17 being the wettest in our record. Wet interstadials also include GIS 14, 12, 8, 4.1 and 2, but they fall on a consistently descending trend in rainfall amount.

Concerning the rapid environmental changes recorded during MIS 4-2 by our stalagmites, the variability of D/O cycles appears muted whilst the Heinrich Events have different expressions in our proxies. Only HE 2, 3, 4, and 5 are clearly visible, whereas HE 3 and 6 had a lesser impact, consistent with studies in the marine domain.

We also revealed the existence of the 2300 years solar cycle in both $\delta^{18}O$ and $\delta^{13}C$ time series, between 31 and 20 ka. This period is also coincident with a peak in summer insolation at 45ºN and a three-fold increase in the depositional rate of the stalagmite, indicating a possible summer climate modulation by solar activity during high insolation periods.

In the study of MIS 5e climate variability, we showed that speleothem formation in the mid-altitude cave CG (Mehedinți Mountains) took place between 132 and 113 ka, without any known hiatus. While isotope values resemble those of the Holocene, we inferred that environmental characteristics were similar to those found today in the same area. Further, we showed that stalagmite CG1 was not affected by kinetic fractionation and faithfully recorded regional climate variability.

The growth period of the high altitude stalagmite M3-R2/1 (Făgăraș Mountains) is coincident with a well-documented global warm peak during MIS 5e. The stalagmite carbon and oxygen data reveal decadal and centennial climate variability in the alpine region of Romania between 125 and 123 ka. Furthermore, the high resolution of the isotope record allowed us to identify decadal and centennial periodicities in the two isotope proxies, revealing a modulation of the climate by solar activity.

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