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Isotopic climate record in a Holocene stalagmite from Ursilor Cave (Romania)

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ABSTRACT: The PU-2 stalagmite from Ursilor Cave provides the first dated Romanian isotope record for the Holocene. The overall growth rate of the speleothem was 3.5 cm kyr^{-1}, corresponding to a temporal resolution of 142 y between each isotope analysis. The 'Hendy' tests indicate that isotopic equilibrium conditions occurred during the formation of PU-2, and hence that it is suitable for palaeoclimatic studies. The relationship between δ^{18}O and temperature was found to be positive. This can be interpreted either as rain-out with distance from the west-northwest ocean source of evaporation or shifts in air mass source with changing North Atlantic Oscillation indices. Applying five U–Th thermal ionisation mass spectrometric (TIMS) dates to a 17.5 cm isotope profile (δ^{18}O and δ^{13}C) along the stalagmite growth axis enabled a tentative interpretation of the palaeoclimate signal over the past 7.1 kyr. Spikes of depleted isotopic δ^{18}O values are centred near ca. 7, ca. 5.2 and ca. 4 ka, reflecting cool conditions. The record shows two warm intervals between ca. 3.8 and ca. 3.2 ka (the maximum warmth) and from ca. 2 to ca. 1.4 ka, when the δ^{18}O values were less negative than present. The 'Holocene Climate Optimum' spanning the time interval from ca. 6.8 to ca. 4.4 ka is not well expressed in the PU-2 stalagmite. Individual spikes of lighter δ^{13}C are interpreted as indicative of periods of heavy rainfall, at ca. 7, ca. 5.5, and ca. 3.5 ka. The overall trend to lighter δ^{13}C in the PU-2 stalagmite may reflect a gradual decrease in water–rock interaction. The results demonstrate that the effect of North Atlantic oceanic changes extended to the investigated area. Nevertheless, some differences in temporal correlation and intensity of stable isotopic response to these climatic events have been found, but the exact nature of these differences and the underlying mechanism is yet to be determined. Copyright © 2002 John Wiley & Sons, Ltd.

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KEYWORDS: speleothem; U-Th TIMS; Holocene; isotope; crystal fabric; palaeoclimate; Romania.

Introduction

Continental records of mid-European Holocene climatic conditions are scarce (Willis, 1994; Willis *et al.*, 1995; Onac and Lauritzen, 1996; Carbonnel *et al.*, 1999; Grafenstein *et al.*, 1999; Denèfle *et al.*, 2000; Feurdean *et al.*, 2001; Björkman *et al.*, 2002). Most Romanian Holocene sites rely on pollen stratigraphy but they have low temporal resolution and inadequate dating control (Diaconeasa and Farcas, 1996; Farcas *et al.*, 1999). The present paper assesses the potential for using the isotopic record of cave calcites as a tool for climatic reconstruction in this region over the past 7.1 kyr based on a stalagmite from Ursilor (Bears) Cave, northwest Romania. For this purpose we applied U–Th thermal ionisation mass spectrometric (TIMS) dating to the δ^{13}C and δ^{18}O profile of this stalagmite and analysed microscopically the calcite fabrics. The results were then compared with other terrestrial and marine records.

Stable oxygen and carbon isotope analysis of speleothems has been used for more than ten years to reconstruct climatic and environmental changes (Lauritzen and Lundberg, 1999a). However, quantitative interpretation of isotope variations in terms of climate or vegetation changes is hampered by a limited understanding of physical and chemical processes controlling the global and local isotope behaviour (Rozanski *et al.*, 1992). The oxygen isotopic signature of speleothems is often related to climate, because infiltrating meteoric water is ultimately responsible for calcite deposition within the cave: where calcite is deposited in isotopic equilibrium and

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the percolation pathways are relatively short, the δ^{18}O of speleothem calcite has been interpreted as reflecting the isotopic composition of precipitation and the mean annual temperature (Dorale *et al.*, 1998; Lauritzen and Lundberg, 1999b). Carbon isotopic signature is controlled by many complex processes, among which are biogenic production of soil CO_2, rainfall, drip rate and bedrock interaction (Baker *et al.*, 1997; Dorale *et al.*, 1998; Lauritzen and Lundberg, 1999a; Repinski *et al.*, 1999; Linge *et al.*, 2001a). The crystallographic fabric sometimes reflects cave microclimate and climate conditions at the surface (Lauritzen and Onac, 1999; Frisia *et al.*, 2000).

Geographical and geological setting

The Ursilor Cave is located in the northwestern part of Romania (80 km southeast of Oradea) at 482 m a.s.l. (Fig. 1, inset), on the west-facing slope of the Apuseni Mountains. The cave has no known natural entrance, having been discovered accidentally in 1975 by blasting in a marble quarry. The first exploration revealed that the cave preserved important palaeontological remains (mainly *Ursus spelaeus*) and also a great variety of speleothems. Consequently, it was gated and fitted for tourism, and has become the most important show cave in Romania.

The cave is formed in Upper Jurassic (Tithonic) recrystallised limestone and consists of 1500 m of large passages developed along two distinct levels, located at 100 to 200 m beneath the surface (Fig. 1). The lower one, still active, hosts most of the cave bear remains; therefore it is preserved as a scientific reserve. The upper level is no longer hydrologically active; it is highly decorated and open for tourist access (Rusu, 1981). The two artificial entrances are well sealed in order to preserve the original cave microclimate so that the upper level remains free from vigorous air flow and maintains a stable temperature. The mean annual temperature in the cave is 9.81 °C (ranging between 9.52° and 10.1 °C), and the relative humidity, measured over one year at 11 stations throughout the cave, is high at 95–100% (Racovita *et al.*, 1999). During the same time-interval, the relative humidity at the stalagmite sampling point (S7) was almost constant (99–100‰), and the temperature varied between 9.8 and 10 °C.

The modern climate around Ursilor Cave is mild and humid, predominantly influenced by west-northwest oceanic air masses. Occasionally, and for short periods of time (months), this dominant air flow is replaced by northeasterly winds and the climate becomes relatively dry and cool. The mean annual precipitation is 950 mm (based on a 21-y record from Pietroasa hydrometric station located in the cave vicinity (Oraseanu I, 2001, personal communication), and the annual average temperature is 9.7 °C (National Institute for Meteorology and Hydrology (INMH)). The vegetation above and around the cave consists of a C3 deciduous association dominated by *Quercus, Carpinus* and *Juglans*. This climatic regime is the distal expression of the westerly depression system that controls the climate of much of northwestern Europe. The dominance of latitudinal or zonal air flow versus meridional air flow is controlled by the North Atlantic Oscillation (NAO), the balance between the subtropical high pressure centre near the Azores and the subpolar low pressure centre south and east of Greenland (Lamb and Peppler, 1987; Cook *et al.*, 1998). If the index is positive, northern and western Europe is dominated by zonal circulation with onshore, westerly and, locally, southwesterly frontal systems bringing stormy, wet conditions with mild temperatures. With a negative index northeasterly circulation is enhanced bringing cold air to southeastern Europe with more offshore air flow, drier conditions and lower mean temperatures.

Rozanski *et al.* (1992) observed a strong relationship between isotopic composition of rainfall and temperature for European stations, with a δ^{18}O temperature coefficient of ca. 0.6‰ $°C^{-1}$. The average temperature and isotopic composition of rainfall for Vienna (the longest record available) was 9.9 °C and −9.8‰ (−9.9‰ weighted mean) versus VSMOW (Vienna Standard Mean Ocean Water). Continued rain-out of the air mass gives an average isotopic composition close to the cave site of −10.3‰ (−9.9‰ weighted mean) (Rozanski *et al.*, 1993). Because of the association of the air flow regime with temperature, zonal circulation is likely to produce precipitation of higher isotopic composition compared with meridional circulation. The scale of variation for European stations over the 30-yr period of study, 1960–1990, was ca. 2‰ (Rozanski *et al.*, 1992). The range for the cave site would thus be ca. −9 to −11‰. Comparing the variations in isotopic composition of rainfall with NAO fluctuations (Hurrell, 1995), a negative NAO index (e.g. 1960s) corresponds to a reduction of ca. 1‰ and a positive NAO (e.g. 1987–1990) with an increase of ca. 1‰.

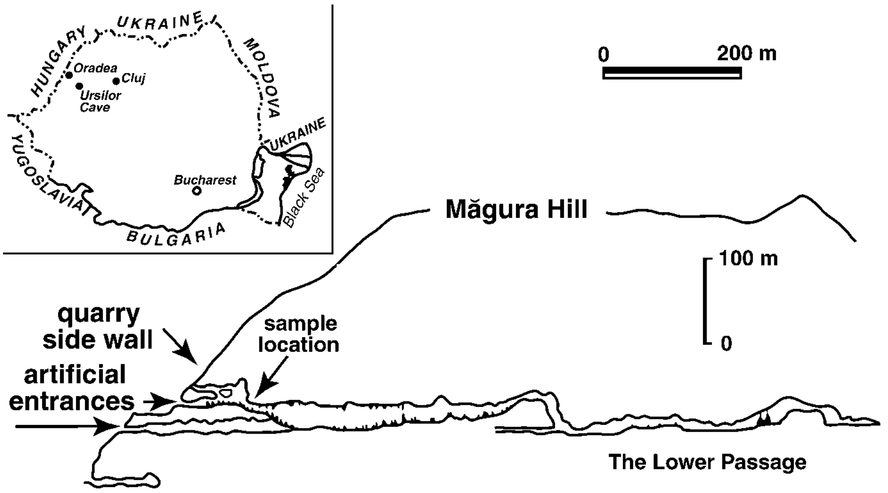

Figure 1 Cross-section through the Ursilor Cave showing the location of the PU-2 sample

Site and sample description

The PU-2 sample is a 72.5-cm-long stalagmite that was removed from Candles Passage (Fig. 1) when the cave was developed for tourism. This part of the cave has the highest density of stalagmites per square metre recorded in any Romanian cave.

The distinctive physical features of PU-2 are the alternating dense, optically clear calcite layers and white-opaque laminae. The growth axis of the upper 4 cm of this speleothem has shifted ca. 1.5 cm with respect to the previous growth axis (Fig. 2) but neither macro- nor microscopic observations revealed any obvious erosional hiatus. No other visible hiatuses or detrital bands were observed along the stalagmite. The uppermost 17.5 cm of the PU-2 stalagmite (the focus of the present study) has a rather homogeneous structure, whereas the rest of it shows several internal corrosion areas. The off-centred 'cap' is composed of hundreds of laminae. Many authors have found such layers to be annually deposited (e.g. Baker *et al.*, 1993; Genty and Quinif, 1996; Holmgren *et al.*, 1999). In order to test if the laminations are annual, a thin section was made of the uppermost 2.2 cm of the stalagmite.

Crystal morphology was classified macroscopically into six zones on the basis of visual changes in texture (Fig. 2). Nine specimens collected along the isotopic profile were prepared for thin-section investigations from what appeared macroscopically to be the main crystal fabric zones (CCF and DCF in Fig. 2). Three of the samples (at the base of the isotopic profile, 4.5 cm above the base, and 1.5 cm below the 'cap') show dendritic fabrics (Frisia *et al.*, 2000) (Figs 2 and 3), whereas all the others consist of elongated calcite crystals making up typical stalagmitic columnar fabric (Kendall and Broughton, 1978; Onac, 1997) (Fig. 4).

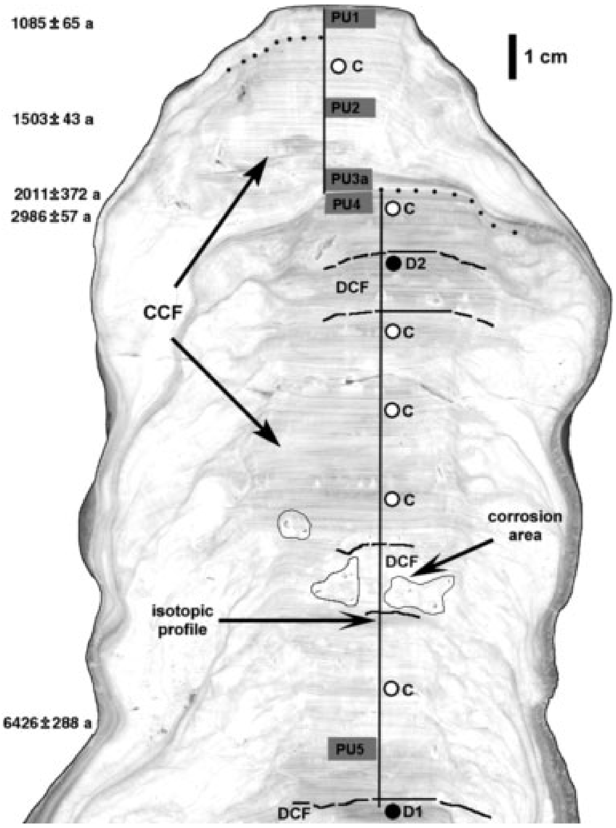

Figure 2 Structure of the upper part of the PU-2 with the location of the specimens used for thin-section analyses (open circle, columnar fabric; solid circle, dendritic fabric; CCF, columnar crystal fabric zone; DCF, dendritic crystal fabric zone), U–Th dated samples (shaded boxes), isotopic profile and 'Hendy' tests (solid dot series)

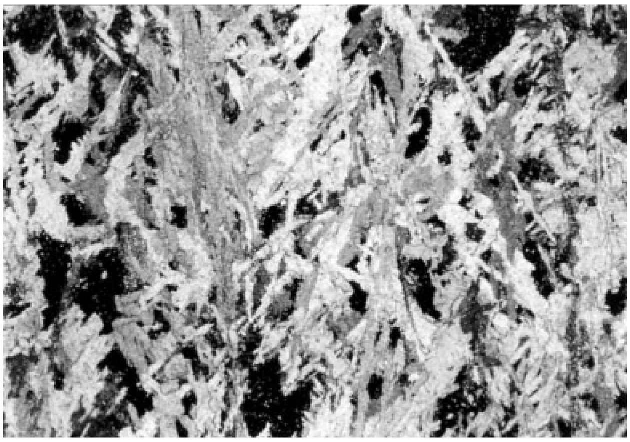

Figure 3 Photomicrograph showing dendritic fabric (D1 in Fig. 2; crossed nicols, ×40)

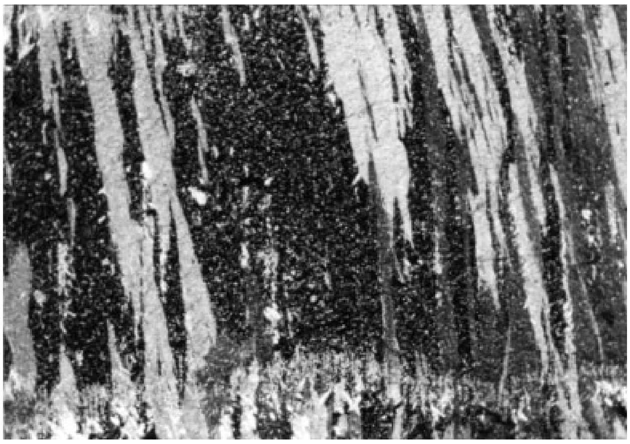

Figure 4 Photomicrograph of elongate calcite crystals building up columnar fabric (typical for any of the C marked samples in Fig. 2; crossed nicols, ×40)

U-series dating and stable isotope analyses

Preliminary dating of seven subsamples by U–Th alpha-spectrometry showed that PU-2 started to grow at 19 ± 2 (1σ) ka (Onac and Lauritzen, 1996), but the errors associated with the alpha dates were relatively high (average 1σ error of ca. 17%), owing to the very low U content (0.03–0.065 ppm). Therefore, we re-dated, by mass spectrometry, the upper, most homogeneous part of the stalagmite that covers only the past 7.1 ka.

Five TIMS subsamples (2.2–3 g) were taken (Fig. 2) as follows: at 16 cm from the top, close to the base of the isotopic profile (PU5); below (PU4) and above (PU3) the hiatus marking the 'cap'; in the middle of the 'cap'(PU2), and in its uppermost part (PU1). The samples were composed of pure white and/or translucent calcite with no visible detritus. Mass abundances of ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{229}Th, ^{230}Th and ^{232}Th were measured on a Finnigan 262 RPQ instrument in dynamic mode. The chemical preparation and TIMS runs followed the procedures described in Lauritzen and Lundberg (1999b).

Subsamples for stable isotopic analysis (80–90 μg each) were taken at 5-mm intervals along the growth axis of the uppermost 17.5 cm of the PU-2 stalagmite, the profile shifting to one side following the shift in the drip point (Fig. 2). These were analysed for oxygen and carbon isotopic ratios in CO_2 expelled in a hot H_3PO_4 line, on a Finnigan 251 mass spectrometer at the GMS Laboratory, University of Bergen. The reproducibility of the system is 0.06‰ for $\delta^{13}C$ and 0.07‰ for $\delta^{18}O$, based on replicate measurements of an internal carbonate standard. Results are reported as ‰ versus PDB (Pee Dee (Formation) Belemnite), using the NIST (National Institute of Standards and Technology) (NBS (National Bureau of Standards)) 19 standard as reference.

Results and discussion

Time-scales and growth rates

The time-scale used for the isotope stratigraphy is based on U-series dates, with suitable interpolation for individual sample points. The dating results are shown in Table 1. All errors are $\pm 2\sigma$. Despite the extremely low U content (0.016–0.024 ppm), the five subsamples yielded ages in correct stratigraphical order. All samples show low $^{230}Th/^{232}Th$ ratios that may indicate a certain detrital contamination (discussed below).

The five dates apparently indicate that calcite deposition ceased at ca. 3.2 ka and that the upper part of the stalagmite (the 'cap') grew following a precipitation hiatus of ca. 1000 yr. We consider that local circumstance (probably neotectonics) rather than a climatic event triggered this halt. This assumption is supported by the presence within the same area of tens of other stalagmites displaying the same 'shifted cap'. However, it is possible that the depositional hiatus did not actually occur (or was not as long as 1000 yr) because the shift of the drip point ca. 1.5 cm meant that, for a long time, the drips slid rapidly off the steep side of the speleothem. Therefore, we may assume that it was only after the new 'plateau' formed that the normal stratigraphy was re-established and the new axis of growth could be traced confidently. Further evidence in favour of a short hiatus comes from the U and Th systematics (Table 1): these remain constant across the break and, indeed, throughout the section studied.

Calculation of growth rates based on the ages calculated (i.e. with no consideration of detrital contamination) yields an order of magnitude higher growth rate for the topmost half of the stalagmite's 'cap' than for its lower half. According to these calculations the growth rate increased from ca. 3.5 cm kyr^{-1} in the lower section (between subsamples PU5 and PU4) to more than 12 cm kyr at the top (between subsamples PU2 and PU1). Although minor tectonism may have caused a change in the drip rate or route, such an increase in the growth rate is not supported by any change in the textural morphology of the growth laminae. If, instead, we assume some detrital contamination and 'correct' the ages using an assumed initial $^{230}Th/^{232}Th$ value (initially we used 1.5, following Schwarcz, 1980), a more uniform growth rate ensues for the 'cap', increasing only from ca. 3 cm kyr^{-1} to ca. 4.6 cm kyr^{-1}. These values are also closer to that of ca. 3.5 cm kyr^{-1} determined for the section of the stalagmite below the hiatus, where the growth rates using uncorrected versus corrected ages are not significantly different (Fig. 5a).

In order to test which age model would be more appropriate in our case (i.e. corrected versus uncorrected), we performed laminae counting for the uppermost section of the stalagmite, located between subsamples PU1 and PU2, where the difference between the uncorrected and corrected ages would reach ca. 68%. The result of our counting yielded a value of 417 laminae. If we assume that these laminae are annual as proven by Baker *et al.* (1993), Genty and Quinif (1996), Holmgren *et al.* (1999) on other Holocene stalagmites, it is evident that the *corrected values* would be more appropriate for a reliable age model as the time interval between PU2 and PU1 would span ca. 432 yr instead of only ca. 15 yr! We then further refined the correction procedure so that the difference in age between PU1 and PU2 conformed to the true measured difference. This yielded an initial $^{230}Th/^{232}Th$ value of 1.41. This value was applied to the other three dates to give the corrected ages shown in Table 1. The growth rates calculated according to this model vary between 2.9 cm kyr^{-1} and 4.7 cm kyr^{-1}.

The overall growth rate of 3.5 cm kyr^{-1} is consistent with measured Holocene stalagmite growth rates from other parts of Europe (Franke and Geyh, 1971; Genty and Quinif, 1996; Mihevc and Lauritzen, 1997; Baker *et al.*, 1998; McDermott *et al.*, 1999; Vesely, 2000; Linge *et al.*, 2001b; Shopov Y., 2000, personal communication), and accords well with the theoretical estimates given by Dreybrodt (1999).

Isotopic signatures and climatic interpretation

The record of oxygen isotopic variations in cave calcite can be interpreted as a record of changing fractionation in response to climatic changes. Fractionation effects at lower temperatures are increasingly more pronounced; the effect of this temperature control varies with the part of the ocean–vapour–precipitation–drip water–crystallisation system that is dominantly affected. They may be summarised briefly as follows. Where the source of vapour remains constant, low temperature of the evaporating ocean water causes depletion of the vapour (Dansgaard, 1964; Clark and Fritz, 1997), and low temperature of the landmass over which the vapour travels causes increased depletion of the vapour during rain-out. If these controls are not significant, then low temperature in the cave itself may have the opposite effect, causing enrichment during the crystallisation process.

Table 1 Uranium-series TIMS dates for stalagmite PU-2 (ages in italic were used for interpolations)

Name and position (centimetres from top)	U (ppm)	^{232}Th (ppm)	Initial					Age (ka$\pm 2\sigma$)	Corrected age[a]
			$^{234}U/^{238}U$	$^{234}U/^{238}U$	$^{230}Th/^{234}U$	$^{230}Th/^{232}Th$	$^{234}U/^{232}Th$		
PU1 (0–0.5)	0.021	0.021	1.26 ± 0.11	1.258 ± 0.001	0.0140 ± 0.0006	5.0 ± 0.2	341 ± 20	1.53 ± 0.06	*1.085 ± 0.065*
PU2 (2–2.5)	0.021	0.008	1.24 ± 0.004	1.249 ± 0.004	0.0154 ± 0.0004	12.5 ± 0.3	807 ± 29	1.69 ± 0.04	*1.503 ± 0.043*
PU3a (3.5–4)	0.022	0.011	1.26 ± 0.001	1.266 ± 0.001	0.02 ± 0.0033	14.5 ± 2	712 ± 166	2.22 ± 0.4	*2.01 ± 0.372*
PU4 (4–4.5)	0.024	0.016	1.27 ± 0.002	1.27 ± 0.002	0.0297 ± 0.0005	15.5 ± 0.3	519 ± 13	3.28 ± 0.06	*2.98 ± 0.057*
PU5 (16–16.5)	0.016	0.010	1.2 ± 0.003	1.230 ± 0.003	0.0597 ± 0.0024	34.4 ± 1.5	576 ± 34	6.7 ± 0.3	*6.42 ± 0.288*

[a] Ages were corrected assuming a $^{230}Th/^{232}Th$ initial ratio of 1.41.

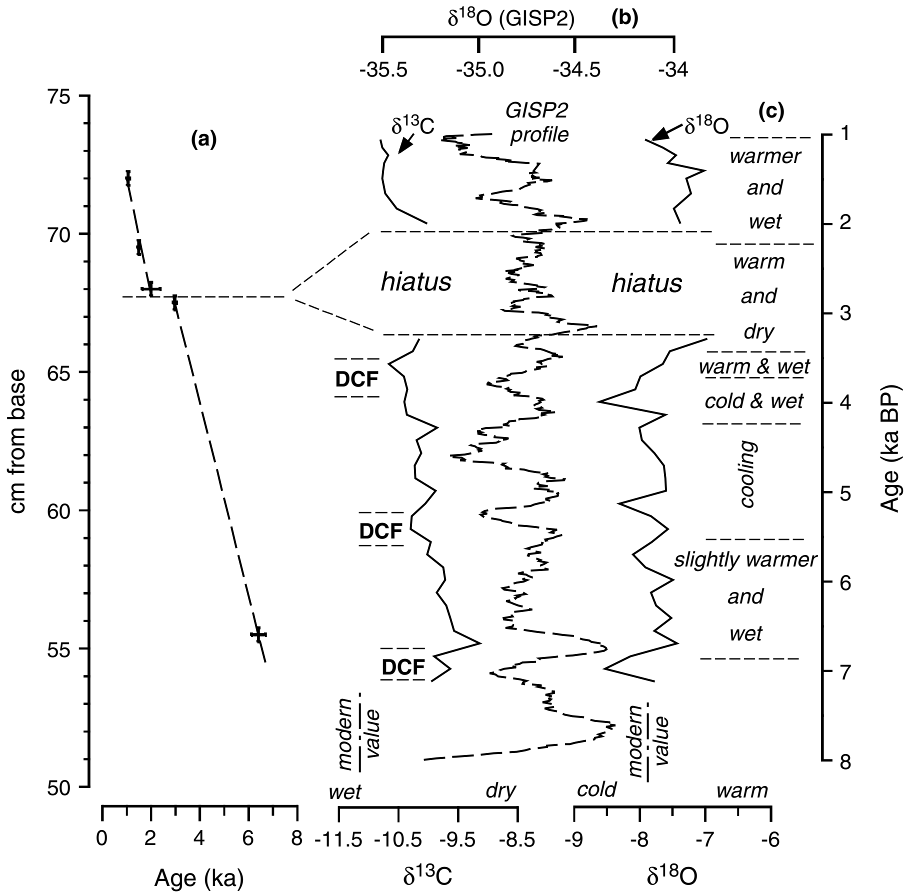

Figure 5 (a) Diagram showing the age of PU-2 versus height above its base. (b) The calibrated isotope time-series of PU-2 compared with the GISP2 curve based on a 25-point running mean. (c) Summary of Holocene climatic changes in western Romania based on pollen and archaeological records (compared with the present)

Changes in the level of interaction of drip waters and rock during percolation causing enrichment is another possible, but usually minor, effect. Where the source of the vapour changes then the properties and distance of the source may have the most significant impact on the isotopic composition of the vapour.

In an inland site such as this one, rain-out effects are most likely to be the strongest control (Rozanski *et al.*, 1993); the depletion becomes more marked as air masses move across cooler continental land masses. Climatically, Romania is a sensitive region of Europe, lying towards the eastern limit of Atlantic air mass penetration into the European continent. The air masses follow an eastward progressing rain-out trajectory over the low-relief European land mass. Precipitation along the western border of Romania is isotopically depleted (the modern long-term mean ca. 300 km to the north is −10.3‰ (versus VSMOW) compared with a mean of −5.0‰ at about the same latitude in southwest Ireland, average temperature 10.4 °C: Rozanski *et al.*, 1993), and it becomes more depleted if regional temperatures are reduced, as a result of either/both increased rain-out and/or a shift in vapour source with negative NAO index. Therefore, in this region isotopically more negative values of δ^{18}O in speleothems that are precipitated under equilibrium conditions are expected to reflect a decrease of temperature; i.e. oxygen isotopes and temperature should show a positive relationship. Two tests are required: the first is to see if the calcite is precipitated under equilibrium conditions; the second is to test the relationship of $\delta^{18}\mathrm{O}_{\mathrm{c(calcite)}}$ with temperature.

Although the value of performing the 'Hendy' test is often questioned owing to its pitfalls (Talma and Vogel, 1992; Lauritzen and Lundberg, 1999a; Linge *et al.*, 2001a), the PU-2 stalagmite was tested for isotopic equilibrium conditions along two growth bands (see Fig. 2). The variations in $\delta^{18}\mathrm{O}_{\mathrm{c}}$ versus $\delta^{13}\mathrm{C}_{\mathrm{c}}$ along each layer show no positive correlation and no enrichment in δ^{18}O towards the flanks of the two layers. Thus, isotopic equilibrium conditions during the time of PU-2 deposition are confirmed.

In order to test whether the relationship of $\delta^{18}\mathrm{O}_{\mathrm{c}}$ is negative or positive with respect to temperature, we compared trends in the δ^{18}O time-series with known, dated climatic events recorded close to the site. Two well-documented episodes with colder-than-present temperatures occurred at about 7 ka and about 4 ka (Grove, 1979, 1988; Starkel, 1991; Veit, 1993; Magyari *et al.*, 2001—see discussion below). Both episodes are represented in this sample as deviant $\delta^{18}\mathrm{O}_{\mathrm{c}}$ values, which are more depleted than present-day values. This strongly suggests that the temperature-sensitivity of the drip water is *positive* and dominates over the cave temperature fractionation. Thus the standard for inland caves of a positive relationship of temperature and calcite isotopes is confirmed for this site.

If we accept the composition of drip water as the dominant local control on δ^{18}O, its shifts are best interpreted in terms of either degree of Rayleigh fractionation of air masses during advection towards Romania or shifts in air mass source. Lighter δ^{18}O then corresponds to greater fractionation and hence larger temperature differences from source to Romania or else to a meridional air flow associated with colder temperatures. The isotopic gradient is obvious when comparing the $\delta^{18}\mathrm{O}_{\mathrm{c}}$ values from Ursilor Cave with those recorded in a stalagmite from Crag Cave (Ireland) (McDermott *et al.*, 1999; 2001). The

$\delta^{18}O_c$ values of the PU-2 stalagmite vary typically around -7.79 ± 0.82‰, whereas those from Crag Cave stalagmite vary around -3.26 ± 1.75‰ (McDermott *et al.*, 2001). These values mirror the gradient in isotopic values of precipitation shown above.

Carbon isotopes in calcite are related to soil processes that often are mediated by climatic events, but not necessarily in a simple way. If vegetation is known (usually from other proxies such as pollen) to have shifted its photosynthetic pathway, then long-term changes in the carbon isotopic composition of speleothems may reflect shifts in the type of vegetation overlying the cave (Dorale *et al.*, 1998, 2002; Holmgren *et al.*, 1999; Repinski *et al.*, 1999). If the vegetation has not changed, wet/dry episodes may still cause depletion/enrichment of the carbon isotopes as soil water balance and cave drip rates change. In many cases crystallographic information can offer an insight into relative humidity levels because textural changes often relate to changes in temperature and water availability (Frisia *et al.*, 2000).

Two features of the $\delta^{13}C$ record must be addressed: the first-order trend showing an overall shift towards more depleted values with time; and the second-order variations around this trend. In this region, pollen evidence (e.g. Farcas, 2001; Björkman *et al.*, 2002; see discussion below) suggests that the vegetation has remained C3 throughout the Holocene: thus we can dispense with problems of shifts in photosynthetic pathways. The trend towards increasing depletion with time suggests that the drip waters may be isotopically less influenced by exchange with, or dissolution of, the isotopically heavy carbon in the bedrock (ca. 0‰), possibly related to shorter residence time in the rock above the cave over time or an increasingly more open dissolution system (which correlates with the increased growth rate observed in the upper part of the sample).

The second-order variations presumably relate to variations in fractionation, drip rates and/or biogenic soil CO_2; these may be elucidated by examining the crystal fabric. Disequilibrium fabric can be associated with inhibited growth, typically associated with dry conditions and therefore likely also to be associated with enriched isotopic values for both carbon and oxygen (Bar-Matthews *et al.*, 1996; Fairchild *et al.*, 2000). Alternatively it can be associated with abnormally rapid growth, often caused by very wet episodes. The disequilibrium dendritic crystal fabric (DCF) in PU-2 is accompanied by second-order shifts to lighter $\delta^{13}C$ values (see Fig. 2); therefore we presume that evaporative enrichment did not occur and that the disequilibrium instead might be associated with wet conditions. Two of the bands of DCF are associated with spikes of lighter $\delta^{18}O$; these are the two largest spikes of the whole record correlated with the cold, wet conditions at ca. 7 ka and ca. 4 ka. The central band of DCF is associated with a spike of lighter carbon but *heavier* oxygen. Under normal conditions a positive NAO index brings warm, wet conditions to Romania with isotopically heavy rainfall. The higher-than-normal cave drip rate might then produce DCF associated with heavier $\delta^{18}O$ values. However, if the whole of the North Atlantic region had cooled then a positive NAO index would still bring wet conditions, but the isotopic composition of that rainfall would be depleted significantly because of the substantially increased fractionation at source in addition to the fractionation during rain-out over a cooler land surface. The faster cave drip rate would reduce the residence time for water in the epikarst zone and thus reduce water–rock interaction, explaining the lighter $\delta^{13}C$ values. If this explanation is correct, then we can interpret the carbon isotopes of PU-2 to relate to rainfall intensity, the wet episodes indicated by the lighter $\delta^{13}C$ values.

In the absence of any other data, we have concluded that in the PU-2 stalagmite:(i) oxygen isotopic shifts relate directly to regional temperature changes; (ii) carbon isotopic shifts relate to cave drip rate changes, here associated with changing rainfall; (iii) disequilibrium DCF indicates high drip rates; and (iv) columnar fabric indicates lower drip rates (either warm or cold conditions). Thus, correlation between $\delta^{18}O$, $\delta^{13}C$ and textural changes in speleothem fabric may reflect climate-dependent changes in water availability and temperatures around the cave (Frisia *et al.*, 2000). A comparison of this record with other records of climate change allows an assessment of the veracity of this interpretation.

Holocene climatic indications for northwest Romania

The oxygen isotope record of PU-2 shows a first-order trend with an average isotopic composition of -7.8 ± 0.8‰ with no overall slope (Fig. 5b). If the relationship between isotopes and temperature is assumed to be positive as stated above, then we can interpret our isotopic record by comparing it with the present-day $\delta^{18}O$ values of calcite taken from stalactite tips in the vicinity of PU-2 at -7.8 ± 0.5‰.

The first spike on the PU-2 appears at ca. 7 ka when the $\delta^{18}O$ value dropped to -8.31‰. The presence of DCF (D1 in Figs 2 and 3) associated with lighter $\delta^{18}O$ values were interpreted as reflecting a cooling event accompanied by increased rainfall (higher drip rate in the cave). Around this time period, several other proxies from a west–east European transect indicate similar wet and/or cooler episodes. The low $\delta^{18}O$ values recorded in a speleothem from Ernesto Cave (Italy) were interpreted as reflecting the Cerin wet phase from Jura and the French Alps (McDermott *et al.*, 1999). Similar wet conditions are quoted by Veit (1993) from the Austrian Alps, where solifluction processes were very active around ca. 7 ka. This also was a time when the Vistula River seems to have increased its discharge (Starkel, 1991). The cold drop at ca. 7 ka is well marked on the GISP2 profile and coincides with the first 'Sahara aridity' cold period (Perry and Hsu, 2000).

Between ca. 6.8 and ca. 4.8 ka, the $\delta^{18}O$ signal suggests a period with temperatures that are comparable with the present ones. This time interval is known as the 'Holocene Climate Optimum'. With one exception, over this period all four calcite samples analysed on thin-sections (open circles in Fig. 2) displayed columnar fabrics, suggesting equilibrium conditions and constant water supply (Fig. 4) (Onac, 1997; Frisia *et al.*, 2000). Pollen analyses from different sites in northwest Romania show a mixed *Corylus* and *Picea* forest, confirming the warm and normal-wet conditions during the first part of the 'Holocene Climate Optimum' period (Farcas, 2001; Feurdean *et al.*, 2001; Björkman *et al.*, 2002). The pollen-based reconstruction from other European sites also suggests a warm mixed-forest biome during this period (Kremenetski, 1995; Allen *et al.*, 2002).

The 'Holocene Climate Optimum' interval recorded on PU-2 stalagmite is interrupted by a low $\delta^{18}O$ spike at ca. 5.2 ka and ends with a drop towards the lowest values of $\delta^{18}O$ of the entire Holocene at 4 ka (Fig. 5b). The cold spike at ca. 5.2 ka corresponds with renewed glacier activity in Europe (Grove, 1979, 1988) and also to a marked trough in the GISP2 record. Furthermore, these events fall within the long-term (ca. 5.5 to ca. 4 ka) temperature decline and precipitation increase documented by Magyari *et al.* (2001) in the Carpathian Basin based on pollen- and mollusc-derived reconstruction, and also by the apparent cooling of the sea surface in both the northeast Atlantic and the Mediterranean Basin (Marchal *et al.*, 2002). At 4 ka a cold and dry event has been recognised in many parts of the Northern Hemisphere (Perry and Hsu, 2000). About the same time most lakes throughout central and northwestern Europe were at minimum levels (Harrison and Digerfeldt, 1997). By contrast, around the 4 ka event the climate conditions in northwest Romania and adjacent regions were cool but humid as inferred from various pollen spectra (Boscaiu and Lupsa, 1967; Magyari *et al.*, 2001) and fluvial environments (Starkel, 2002).

The onset of a climatic amelioration that commenced at ca. 3.8 ka and reached its maximum at ca. 3.2 ka in our speleothem (beginning of the Bronze Age) is reflected by a marked shift in δ^{18}O values. At the very beginning of this period both δ^{18}O and δ^{13}C values were still low and the last band of DCF formed in the upper part of PU-2 (see Fig. 2). Correlation between these three parameters suggests favourable wet and warm conditions, as also indicated by the speleothem growth frequency in northwest Romania (Onac and Lauritzen, 1996). Archaeological evidence from different parts of Romania also indicates that the beginning of the Bronze Age was relatively wet and warm (Motzoi-Chicideanu, 1986), gradually becoming warm and dry until ca. 2.1 ka (Iuliu, 1992).

Shortly after ca. 3.5 ka the growth of the PU-2 stalagmite ceased for about ca. 1.2 ka. This event correlates well with the pollen data from northwest Romania, which suggests, between ca. 3.3 and ca. 2.2 ka, low peat accumulation rates and a sharp increase in relative abundance of *Carpinus* and *Fagus* pollen grains, hence, drier conditions (Feurdean *et al.*, 2001). The time interval when PU-2 growth ceased appears to have been wet in the Mediterranean region (Harrison and Digerfeldt, 1997; McDermott *et al.*, 1999) and in different regions of central Europe (Jäger, 2002; Starkel, 2002). Therefore, we consider local circumstances rather than a climate event triggered its halt.

In the upper 4 cm (ca. 2 to ca. 1.1 ka) the speleothem is composed of CCF calcite building up annual layers and so we inferred a fairly constant drip rate. The isotope profile in the uppermost part of the stalagmite shows a warm peak at ca. 1.4 ka corresponding to one of the first short-lived episodes of climatic warming recorded during the 'Little Climatic Optimum' period (Bell and Walker, 1992).

Conclusions

We assume for this interpretation that climate operated during the Holocene as it does today, where a predominantly zonal west-northwest atmospheric circulation causes relatively warm and wet conditions, and a predominantly northeast circulation causes cooler, drier conditions. This is reflected in changing δ^{18}O values of rainfall and in turn cave drip waters and calcite; higher values for $\delta^{18}O_c$ thus indicate higher temperatures. Although the relationship is not so clear-cut, we interpret changing δ^{13}C to indicate changing water–rock interaction and thus rainfall intensity: lower $\delta^{13}C_c$ values indicate higher rainfall. In this sample it appears that disequilibrium dendritic crystal fabric is associated with high drip rates. Based on these interpretations we have tried to reconstruct the climatic history of northwest Romania for the past 7.1 kyr as indicated by the relatively low-resolution isotopic record from the PU-2 stalagmite.

The oxygen isotope record of PU-2 exhibits an apparent similarity between the timing of intervals of rapid changes shown by our data and records from across Europe. The shifts centred at ca. 7, ca. 5.2 and ca. 4 ka show episodes of low δ^{18}O and are interpreted as reflecting cold events. The two spikes of higher δ^{18}O occurring at ca. 3.2 and ca. 1.4 ka signify warm climatic conditions. These episodes interrupted an otherwise constant climate with conditions very similar to those of today. However, some contrasting climatic patterns have been found between Romania and western Europe, as our record does not show the marked cooling of ca. 5.3–4.5 ka; nor does it show the 'conventional' Holocene Climate Optimum at ca. 6.8–4.4 ka as a time of noticeably warmer-than-today temperatures.

Altogether, the oxygen isotopic profile of PU-2 stalagmite plotted in Fig. 5b reveals some climate variability throughout the mid- and late Holocene of Romania. Comparison of this record with other climate proxy records throughout Europe indicates that there is a temporal correlation between these major changes and north Atlantic events. Thus, our record suggests that the closely coupled Northern Hemisphere ocean–atmosphere system of the last glacial and Holocene period extended its influence at least as far as central southeastern Europe.

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