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Holocene climate variability in Sicily from a discontinuous stalagmite record and the Mesolithic to Neolithic transition

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Abstract

Fabric and stable isotopic composition of a Holocene stalagmite (CR1) from a cave in northern Sicily record changes in paleorainfall in the early Holocene. High $\delta^{13}C$ stable isotope values in the calcite deposited from ca. 8500 to ca. 7500 yr ago are interpreted as reflecting periods of high rainfall. The wet phase was interrupted by two periods of multi-century duration characterized by relatively cool and dry winters centered at ca. 8200 and ca. 7500 yr ago, highlighted by low $\delta^{13}C$ and $\delta^{18}O$ values. A high variability of $\delta^{13}C$ values is recorded from ca. 7500 to ca. 6500 yr ago and indicates that the transition from a pluvial early Holocene to the present-day climate conditions was punctuated by decadal-scale periods of relatively dry winters. In northern Sicily, the traditional elements of the Neolithic appear at ca. 7700 yr ago. It is possible that changes in rainfall influenced the passage from hunter-gathering to farming and sheep-herding economies.

Keywords: Stalagmite; Sicily; Early Holocene; Paleorainfall

Introduction

The Holocene climate evolution of Sicily is relatively poorly known, despite its central Mediterranean position and its role in the history of Mediterranean civilization. To date, there is one continuous postglacial record of environmental change provided by lacustrine sediments from Lago di Pergusa, located in the central part of the island (Sadori and Narcisi, 2001; Fig. 1). This record reveals dramatic hydrological changes during the Holocene: wet climatic conditions before ca. 8000 yr BP were succeeded by a trend towards aridification, which commenced ca. 6900 yr ago (Sadori and Narcisi, 2001). A pluvial early Holocene followed by a decrease of rainfall has been recognized from lake sediments and speleothem records in a wide area around the Mediterranean Sea (Burns et al., 1998; Bar-Matthews et al., 1999; Gasse, 2000). In Sicily, the late Mesolithic (ca. 8500 yr ago) is characterized by a change in lithic assemblages and a significant increase in the diversity of food resources being exploited from a wide range of habitats (Tusa, 1992). The traditional elements of the Neolithic, as documented by the stratigraphy recovered from Grotta dell'Uzzo (Fig. 1), appeared at ca. 7700 yr ago (Mannino et al., in press), at the end of the wet early Holocene. According to Tusa (1992), indigenous hunter-gatherers with a strong Mesolithic tradition adopted certain elements of the Neolithic culture, which improved their lifestyle. What mechanisms could account for Tusa's (1992) 1000-yr-long "transition phase"? Tusa (1992) inferred that the "transition" phase was a complex process, which was possibly influenced by climate and environmental conditions.

Studies of the isotopic composition and fabrics in speleothems from Israel and southern Europe have shown that calcite deposits in caves are valuable tools for reconstructing the chronology and

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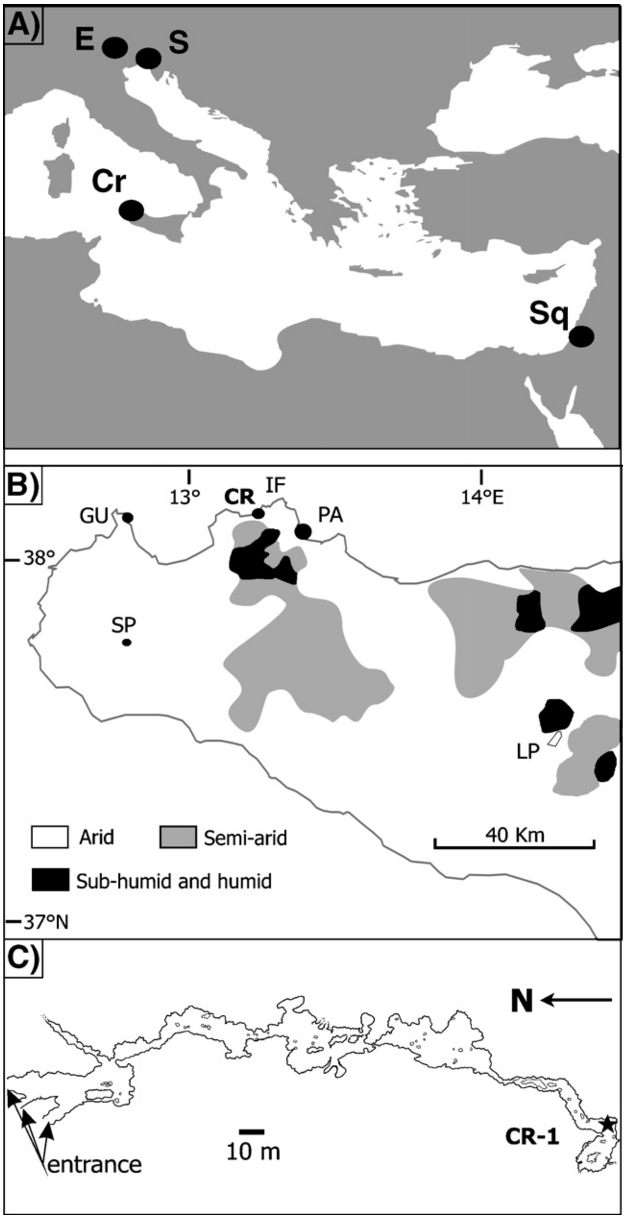

Figure 1. (A) Location of the Mediterranean sites discussed in the text. Cr = Grotta di Carburangeli; E = Grotta di Ernesto; S = Grotta Savi; Sq = Soreq Cave. (B) Western part of Sicily showing the different aridity zones (after Sadori and Narcisi, 2001) and the location of the Sicilian sites mentioned in the text. GU = Grotta dell'Uzzo; IF = Isola delle Femmine; LP = Lago di Pergusa; PA = Palermo; SP = Stretto Partanna. (C) Map of Grotta di Carburangeli and location of stalagmite CR1.

the characteristics of past climate and environmental changes in the Mediterranean (Bar-Matthews et al., 1997, 2000, 2003; McDermott et al., 1999). Bar-Matthews et al. (1999) also recognized the potential for speleothem-based studies to identify hydrological changes that had a strong impact on past societies. Here, we present petrographic and geochemical data from a Sicilian Holocene stalagmite from Grotta di Carburangeli, located in the Piana di Carini at 22 m above sea level (asl) (Fig. 1), near the archaeological site of Grotta dell'Uzzo. The aim of the study is to reconstruct the climate evolution of southern Italy, with emphasis on the early Holocene, when the transition from Mesolithic to Neolithic lifestyles occurred.

Setting and sample

Grotta di Carburangeli was chosen because it is a typical karstic cave, which occurs in carbonate rocks (many caves in Sicily developed in gypsum) and contains speleothems. Because of conservation requirements only one stalagmite (CR1) could be removed from the inner part of the cave (Fig. 1), which is overlain by approximately 8 m of fissured, partially dolomitized limestone and a 50-cm-thick brownish agricultural soil supporting Mediterranean evergreens. This inner part of the cave is characterized by a relatively stable microclimate, with air and water temperatures of 19.4 ± 0.5°C, and a relative humidity of 95 ± 5%.

The present-day climate in the area above the cave is arid (Fig. 1). The mean annual precipitation recorded at the weather station of Palermo is 619 ± 117 mm/yr (from AD 1797 to 1999). Similar rainfall has been recorded during the past 30 yr at Isola delle Femmine, a local weather station near the cave. About 76% of the annual rainfall falls between October and March, with maximum precipitation from October to January (average = 85.0 mm/month). Minimum precipitation is recorded from June to August (average = 10 mm/month). Mean annual temperature recorded at Palermo since AD 1960 is 18.2±0.5°C (20.3°C at Isola delle Femmine). Potential evapotranspiration is 820 mm/yr. The monthly average is 26 mm/month in winter, 49 mm/month in spring, 125 mm/month in summer and 73 mm/month in autumn. The hydrological balance is positive in winter (+56 mm/month) and negative in all the other seasons (−4 mm/month in spring, −115 mm/month in summer, −5 mm/month in autumn; Camuffo and Pagan, 2005). Net infiltration into the karst aquifer is restricted to major recharge events in winter (from December to March). The amount-weighted annual $\delta^{18}O$ value of rainfall recorded at Palermo is −5.5‰ (Longinelli and Selmo, 2003).

The thin rock and soil cover, and the fractured nature of the carbonate bedrock, suggest the drips that feed the stalagmites respond rapidly to rainfall changes. The delay between rainfall events and drip-rate increase is up to 2 days depending on the duration and intensity of the event. Monthly monitoring (AD 2001–2003) showed that the drip rate at five sites adjacent to the point from where CR1 was sampled varied from 20 drops/h in winter to 1 drop/h in summer. An exceptionally dry period extending from autumn 2002 to autumn 2003 resulted in the absence of active drips for about one entire year.

In regular hydrological years, aquifer recharge occurs during winter, when drip-water electrical conductivity (EC at 20°C) was ca. 830 μS/cm. Typically, at the end of winter the drip-water EC decreases to ca. 500 μS/cm. Minor recharge occurs during early spring, and the EC increases up to 600 μS/cm within 48–96 h following major rainfalls. The variability in seasonal and annual infiltration is reflected by the pattern of calcite formation on glass plates positioned beneath the drips. The glass plates positioned in December 2001 and retrieved in February 2002 were completely coated by calcite, whereas those positioned in May and retrieved in June showed less than 10% of the winter deposition. No calcite was deposited from June 2002 to October

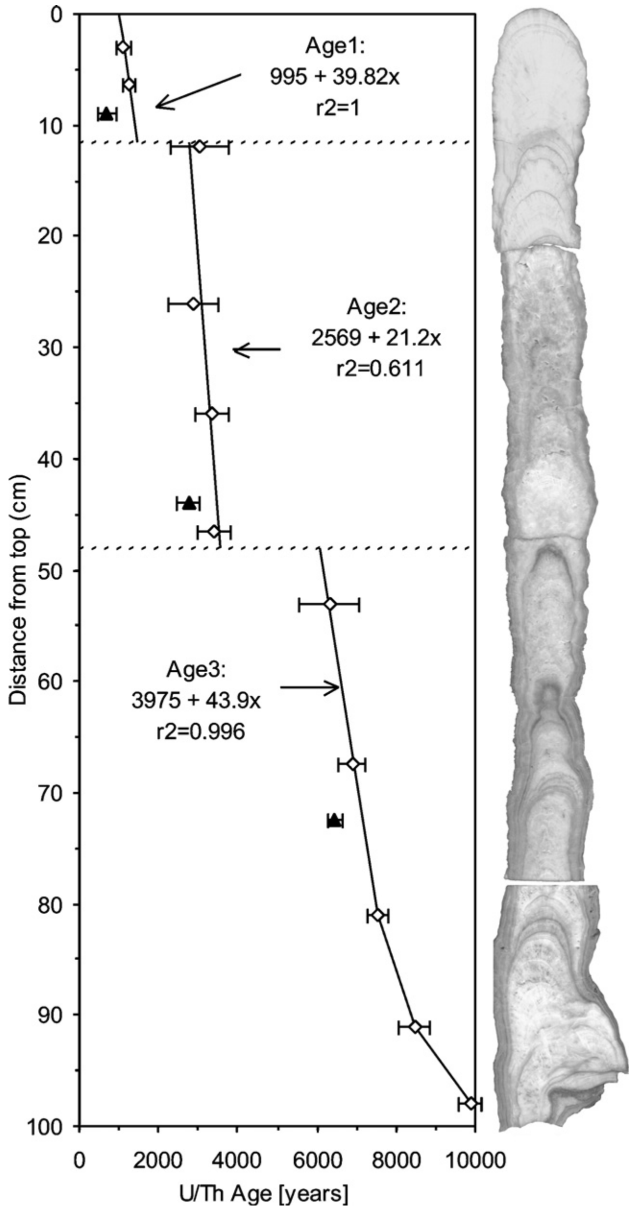

Figure 2. Axial slab and U/Th age model of stalagmite CR1. Open diamonds: data used in the age model. Squares: data not used in the age model. The two major hiatuses are highlighted by dashed lines (x = cm).

2003. Deposition recommenced in autumn 2003, reaching a maximum in winter, and a minimum in spring and summer 2004, when the in situ precipitation experiments ended.

At the time of removal, the ca. 1-m-long, candle-shaped stalagmite CR1 was lying on the floor near its clearly visible base. The morphology and length of CR1 are similar to those of the few remaining active stalagmites in the same chamber, and we infer that CR1 and the in situ speleothems are approximately of the same age.

Methods

Stalagmite CR1 was cut into two halves, one of which was further cut along the vertical growth axis. From the axial part of one of these pieces, we prepared a series of 25 thin sections that encompassed the entire vertical extent of the stalagmite. Calcite fabrics were studied in thin section by optical transmission microscopy. Calcites precipitated onto the glass plates were examined using a stereomicroscope. Subsamples for C- and O-isotope ratio analyses were micromilled along the original prisms from which the thin sections were cut. The milling increment ranged from 0.5 to 0.15 mm according to the thickness and porosity of the growth layers. In addition, we sampled individual growth layers from the axis laterally at four different levels (at 8 mm, 58 mm, 453 mm and 476 mm from the top) to determine whether significant non-equilibrium water–calcite fractionation had occurred (Hendy, 1971) (Figs. 2 and 3). The stable C- and O-isotope values were measured using a Delta^{plus}XL mass spectrometer equipped with an on-line carbonate preparation system (Gasbench II). Standardization was accomplished by using the NBS19 standard, and results are reported relative to the Vienna Peedee Belemnite (VPDB) standard. The 1σ precision of the $\delta^{13}C$ and $\delta^{18}O$ values (replicate analyses) is less than 0.10‰. Fourteen ages were

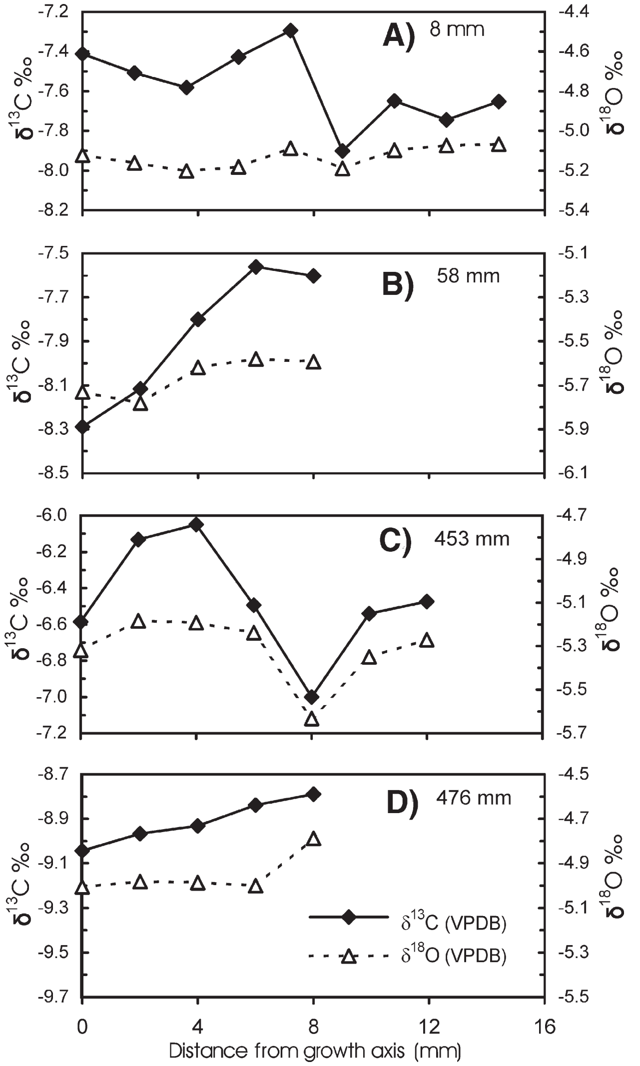

Figure 3. Results of the four Hendy tests for isotope equilibrium deposition of synchronous calcite layers. Values (in mm) refer to the distance from the top of the stalagmite.

obtained by applying the U/Th method (TIMS; procedure described in Frank et al., 2000; Neff et al., 2001).

Results

Fabrics

Most of CR1 consists of white, porous open dendritic and compact dendritic calcite fabrics (Frisia et al., 2000) arranged in 0.09- up to 1.1-mm-thick layers. These are capped by a thin (10–30 μm) compact layer composed of sub-euhedral, equant crystals (Figs. 4A–C). In the top 4 cm of CR1 a porous, open columnar fabric is predominant (Figs. 4A and B). Present-day crystals precipitated on the glass plates show large size variability, from 10 to 100 μm, and the prevalence of steep rhombohedra with macro-steps. In the middle and lower portions of the stalagmite, the fabric is more compact in the axial part (Fig. 4C), with equant crystals that fill the porosity between dendrites (Fig. 4D). At ca. 48, 64, 78 and 81 cm from the top, up to 3 cm thick grayish, translucent laminae are composed of sub-euhedral calcite mosaics (Fig. 4E). The layer from ca. 48 to ca. 46 cm from the top consists of compact dendritic and sub-euhedral calcite fabrics showing distinct red layers about 100 μm thick. These layers contain goethite, hematite and dolomite. Microscopic observations do not support an authigenic origin for the dolomite. Iron oxides and dolomite are also interspersed within the dendritic fabric from 46 to 20 cm from the top. The bottom 0.5 cm of the stalagmite consists of compact, columnar fabric.

Age data and age–depth model

The whole stalagmite is characterized by a very low U content (40–129 ppb), which yielded problems in the accuracy of the dating. The interval between 9 and 45 cm from the stalagmite top has detrital contamination, with ^{230}Th/^{232}Th activity ratios between 2.3 and 6.7. For this reason, it was necessary to correct the ages (see Table 1). Petrographic observations suggested discontinuous growth. Consequently, we dated four subsamples that were taken just above and below two visible hiatuses at 11.5 and 48 cm from the top. Eleven ages do not show inversion and are regarded as representative of the correct stratigraphy. Four U/Th analyses yielded anomalously young ages, possibly related to the small-scale selective leaching of porous calcite (Borsato et al., 2005), and were discarded. In the portions from 0 to 11.5, 11.5 to 48 and 48 to 81 cm from the top, the age model was constructed by linear regression between the U/Th-dated intervals. In the portion from 82 mm to the bottom, we used a simple linear interpolation between the three available U/Th analyses. The age model, the regression lines, the discarded U/Th analyses and the two major hiatuses are shown in Figure 2. Mean growth rates obtained from

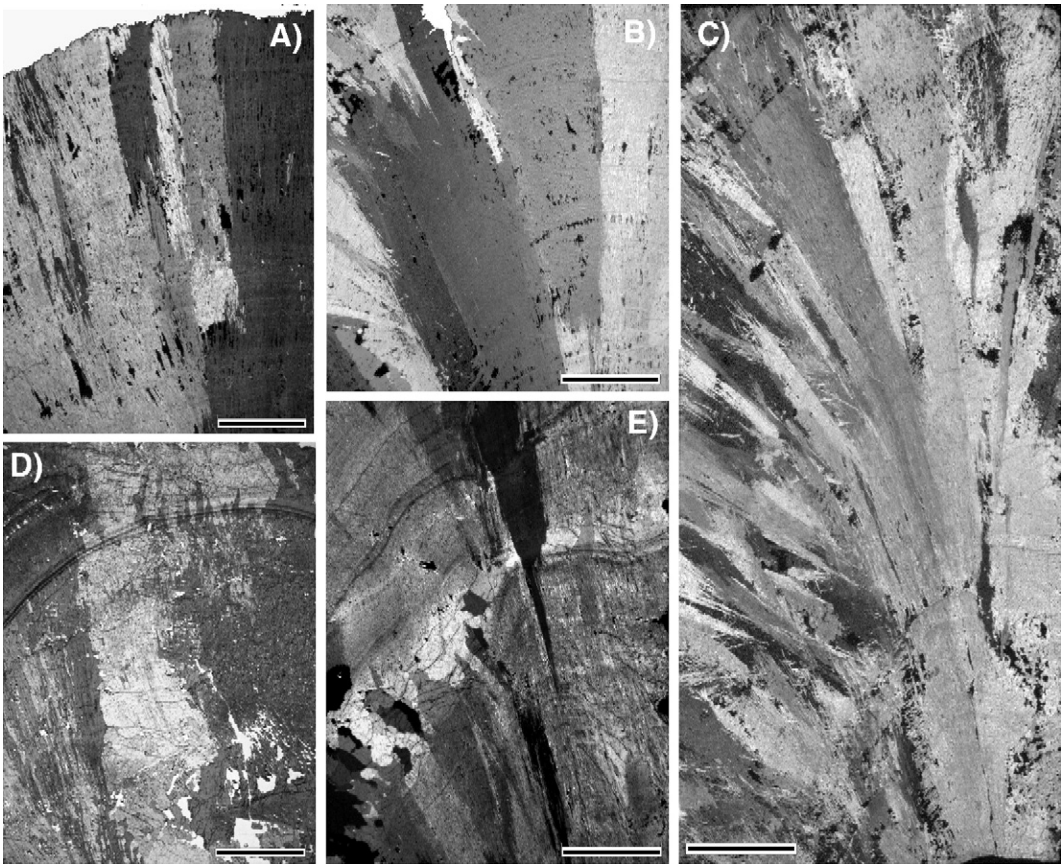

Figure 4. CR1 calcite fabrics. (A and B) Open columnar (right part of the picture, corresponding to the vertical growth axis) and dendritic; (C) predominant dendritic. Note the scaffold-like crystals, which are most visible at the bottom of the photo. (D and E) dendritic and sub-euhedral (equant) crystals. The central part of D and the bottom-right corner of E show void-filling calcite, open columnar individuals and dendritic branches. Scale bar in all photomicrographs: 5 mm.

Table 1
Results of U/Th analyses

Lab No.	Distance from top (cm)	δU (‰)	^{238}U (μg/g)	^{230}Th (pg/g)	^{230}Th/^{232}Th activity ratio	Age (uncorrected) (ka)	Age (corrected) (ka)
3416	3.0 ± 0.2	90.3 ± 7.4	0.07080 ± 0.00014	0.0141 ± 0.0018	9.0 ± 1.178	1.223 ± 0.210	1.115 ± 0.178
3418	6.4 ± 0.2	103.7 ± 9.3	0.07110 ± 0.00014	0.0150 ± 0.0018	28.9 ± 3.785	1.286 ± 0.190	1.250 ± 0.160
3268	**9.0 ± 0.2**	**90.0 ± 5.4**	**0.07461 ± 0.00015**	**0.0097 ± 0.0029**	**6.7 ± 2.018**	**0.800 ± 0.260**	**0.704 ± 0.239**
3318	12.0 ± 0.2	95.5 ± 6.9	0.06973 ± 0.00014	0.0493 ± 0.0046	2.6 ± 0.243	4.394 ± 0.650	3.051 ± 0.728
3222	26.0 ± 0.2	95.8 ± 5.1	0.04979 ± 0.00010	0.0355 ± 0.0012	2.3 ± 0.080	4.432 ± 0.700	2.901 ± 0.629
3223	36.0 ± 0.2	86.2 ± 5.3	0.04983 ± 0.00010	0.0331 ± 0.0012	4.1 ± 0.145	4.160 ± 0.395	3.349 ± 0.398
3224	**44.0 ± 0.2**	**93.7 ± 3.4**	**0.12915 ± 0.00026**	**0.0715 ± 0.0016**	**4.1 ± 0.097**	**3.430 ± 0.310**	**2.769 ± 0.294**
3270	46.5 ± 0.2	91.7 ± 11.4	0.07450 ± 0.00023	0.0426 ± 0.0047	23.0 ± 2.591	3.547 ± 0.295	3.425 ± 0.416
2746	53.0 ± 0.2	57.3 ± 12.9	0.07481 ± 0.00015	0.0758 ± 0.0033	19.6 ± 0.852	6.588 ± 0.750	6.325 ± 0.755
3225	67.5 ± 0.2	93.1 ± 6.1	0.03915 ± 0.00008	0.0440 ± 0.0018	28.4 ± 1.183	7.086 ± 0.290	6.892 ± 0.352
3226	**72.5 ± 0.2**	**96.1 ± 3.9**	**0.04710 ± 0.00009**	**0.0497 ± 0.0011**	**33.8 ± 0.766**	**6.611 ± 0.165**	**6.459 ± 0.200**
3227	81.0 ± 0.2	95.8 ± 4.1	0.04472 ± 0.00009	0.0553 ± 0.0012	24.8 ± 0.568	7.801 ± 0.220	7.556 ± 0.257
3228	91.0 ± 0.2	87.8 ± 5.9	0.04137 ± 0.00008	0.0555 ± 0.0023	70.7 ± 3.193	8.550 ± 0.345	8.456 ± 0.391
2747	98.0 ± 0.2	104.1 ± 4.6	0.05600 ± 0.00011	0.0894 ± 0.0019	35.3 ± 0.748	10.089 ± 0.275	9.870 ± 0.293

Errors are quoted as 2σ standard deviations. The correction for the detrital contamination was performed applying the following assumptions: (i) the detrital ^{238}U/^{232}Th activity ratio is close to the upper continental (mean crustal) value taking the Pb isotope evolution into account (i.e., 0.764, Th/U mass ratio of 4.1; Wedepohl, 1995); (ii) initial non-detrital ^{230}Th concentration is zero; and (iii) radioactive equilibrium exists between detrital ^{230}Th–^{234}U and ^{238}U. Bold: data that resulted in age inversions and that we discarded.

the age model were compared with the average thickness of the visible laminae. The results, reported in Table 2, indicate that the axial extension rates during the three stalagmite growth phases, as obtained with U/Th ages, are similar to the mean thickness of the visible laminae. This suggests that the visible laminae are likely to be annual.

Historical ages here are expressed in AD and BC. We chose to quote pre-historical ages in thousands of years before the year 2000. The ages obtained through the age model are thus expressed as "years ago" (e.g., Vacco et al., 2005). When comparing with data series from the literature for which the age is commonly reported in calibrated (cal) years BP, and which refer to 1950 as present, we added 50 yr to the calibrated ages. This makes a difference only for the most recent part of the record. Many speleothems, however, show annual laminae, which allow calibration with long time series of instrumental data and historical records and therefore it is becoming more and more common to start the chronology from the year 2000 rather than from 1950 (e.g., Proctor et al., 2002; Mangini et al., 2005).

Corrected radiometric ages reported in Figure 2 and Table 1 show that stalagmite CR1 grew discontinuously from ca. 9900 yr ago and ceased to grow ca.1000 yr ago. The faster axial growth rate (471 μm/yr) is recorded in the central-upper part of the stalagmite, between 11.5 and 48 cm. The two portions, from 48 to 81 cm and from 0 to 11.5 cm from the top, are characterized by growth rates of ca. 240 μm/yr (Table 2). The bottom of the stalagmite (81 to 97 cm from the top) is characterized by an axial growth rate of 50 μm/yr.

Table 2
Comparison between axial growth rates determined by U/Th dating and laminae thickness measurements for different growth intervals

Interval (cm from top)	Time span (yr)	Axial growth rate by U/Th (μm/yr)	Mean lamina thickness (μm)
0–11.5	995 – 1453	251 ± 120	234 ± 55
11.5–48	2783 – 3558	471 ± 250	393 ± 90
48–81	6082 – 7531	228 ± 55	270 ± 95
81–91	7531 – 8456	108 ± 35	
91–98	8456 – 9870	50 ± 14	

Stable isotopes

The CR1 stable isotope values are plotted against distance from the top in Figure 5. High-resolution stable isotope analyses were carried out starting at ca. 91 cm from top because the base of the stalagmite shows irregular, detrital-rich layers. The stable isotope profiles reflect the three growth phases that were identified by petrography and radiometric dating. The lower portion (91–48 cm from the top) is characterized by a high C isotopic variability, with $\delta^{13}C$ values ranging from −10.4 ‰ to ca. 0.0‰. The mean $\delta^{13}C$ value for this portion of the stalagmite is −4.4‰. Minimum of $\delta^{13}C$ values (−9.3 and −10.4‰) correspond to compact, sub-euhedral calcite layers, but another $\delta^{13}C$ minimum

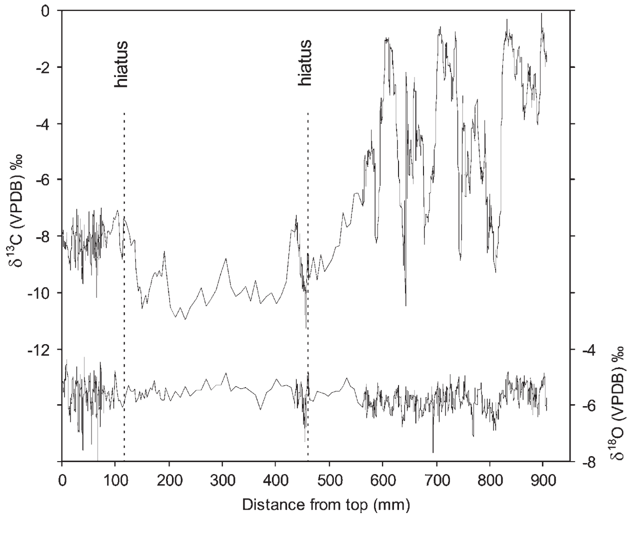

Figure 5. Stable isotope profiles of CR1 plotted against distance from the top of the stalagmite. Dashed, vertical lines show position of the two major hiatuses.

(−8.9‰) corresponds to an open dendritic fabric. Maximum δ^{13}C values commonly correspond to a compact dendritic fabric (Fig. 3C). The tests for equilibrium deposition show co-variance of δ^{13}C and δ^{18}O values immediately above the first major hiatus (Figs. 2 and 3). The second portion is characterized by more depleted average δ^{13}C values (−9.6‰) with respect to the lower portion of CR1, and δ^{13}C values ranging from −11.3 ‰ to −7.2‰. In the third, upper portion, above the second major hiatus, the average δ^{13}C value of speleothem calcite (−8.2‰) is higher than that in the middle portion, with minimum and maximum values of −10.2‰ and −7.0‰, respectively.

The δ^{18}O values range from −8.1 to −4.3‰ (average −5.7‰) and show a weak trend toward higher values from the bottom to the top of the stalagmite. The δ^{18}O values do not show marked differences in the three growth phases. The δ^{13}C and δ^{18}O show significant positive co-variance only for the upper 11.5 cm of the stalagmite, corresponding to the time intervals between 1215 and 995 yr ago (Fig. 6A; $r^2=0.537$, $P<0.0001$) and between 1450 and 1215 yr ago (Fig. 6B; $r^2=0.265$, $P<0.0001$), which indicates predominant non-equilibrium deposition. The δ^{13}C and δ^{18}O values of calcite deposited on the glass plates, which yielded sufficient material for stable isotope analyses (from December

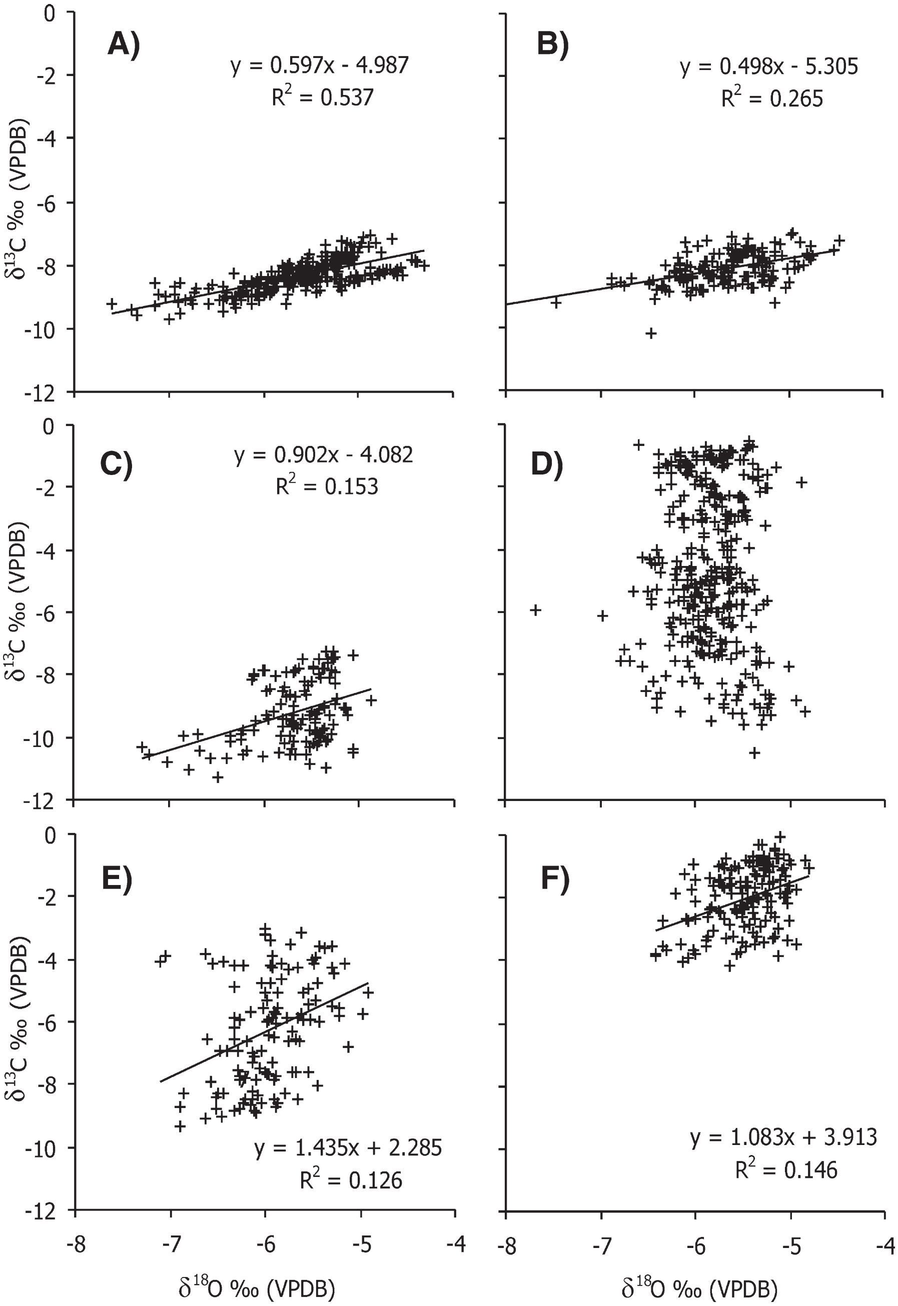

Figure 6. Stable C and O values (referred to VPDB standard) cross plots for selected time spans (in years before 2000). A = 995–1215 yr; B = 1215–1450 yr; C = 2790–3550 yr; D = 6080–7240 yr; E = 7245–7630 yr; F = 7640–8450 yr. Growth periods A and B, which correspond to the third growth phase, show marked evidence of non-equilibrium deposition along the growth axis in successive growth layers. For periods A, B, C, E and F: $P \ll 0.0001$. Part D shows equilibrium deposition.

2001 to February 2002), are −9.1‰ and −6.7‰, respectively. These values cannot be considered representative of the modern cave calcite isotopic composition but provide information on calcite formed during a regular hydrological year.

Discussion

In principle, a reliable paleoclimate record for northwest Sicily should be based on several stalagmites that were deposited continuously and under conditions of isotopic equilibrium. In the literature, however, there are many examples of paleoclimate reconstructions based on single stalagmites, some of which have been recognized as affected by non-equilibrium phenomena and discontinuous growth (e.g., Frumkin et al., 1999; Plagnes et al., 2002; Spötl and Mangini, 2002; Drysdale et al., 2004; Wurth et al., 2004). A recent compilation of published data indicates that most of the speleothem isotope records may be affected by kinetic phenomena (Mickler et al., 2006). Previous research also showed that useful paleoclimatic information can be extracted from speleothems showing low U content and detrital Th contamination (Bertaux et al., 2002; Vacco et al., 2005).

Comparison with other records from the same broad region may considerably aid in discriminating local factors from regional climate changes when only a single speleothem sample is available for analysis. By comparison with data from Soreq cave (Bar-Matthews et al., 2000), Plagnes et al. (2002) recognized the influence of hydrological activity of the eastern Mediterranean basin as far west as the south of France. Petrography also aids interpreting single stalagmite stable isotope series in terms of localized hydrology (McDermott, 2004). Published records of Holocene stalagmite stable isotope time series are available for comparison from Italy and the Mediterranean region (Bar-Matthews et al., 1997,

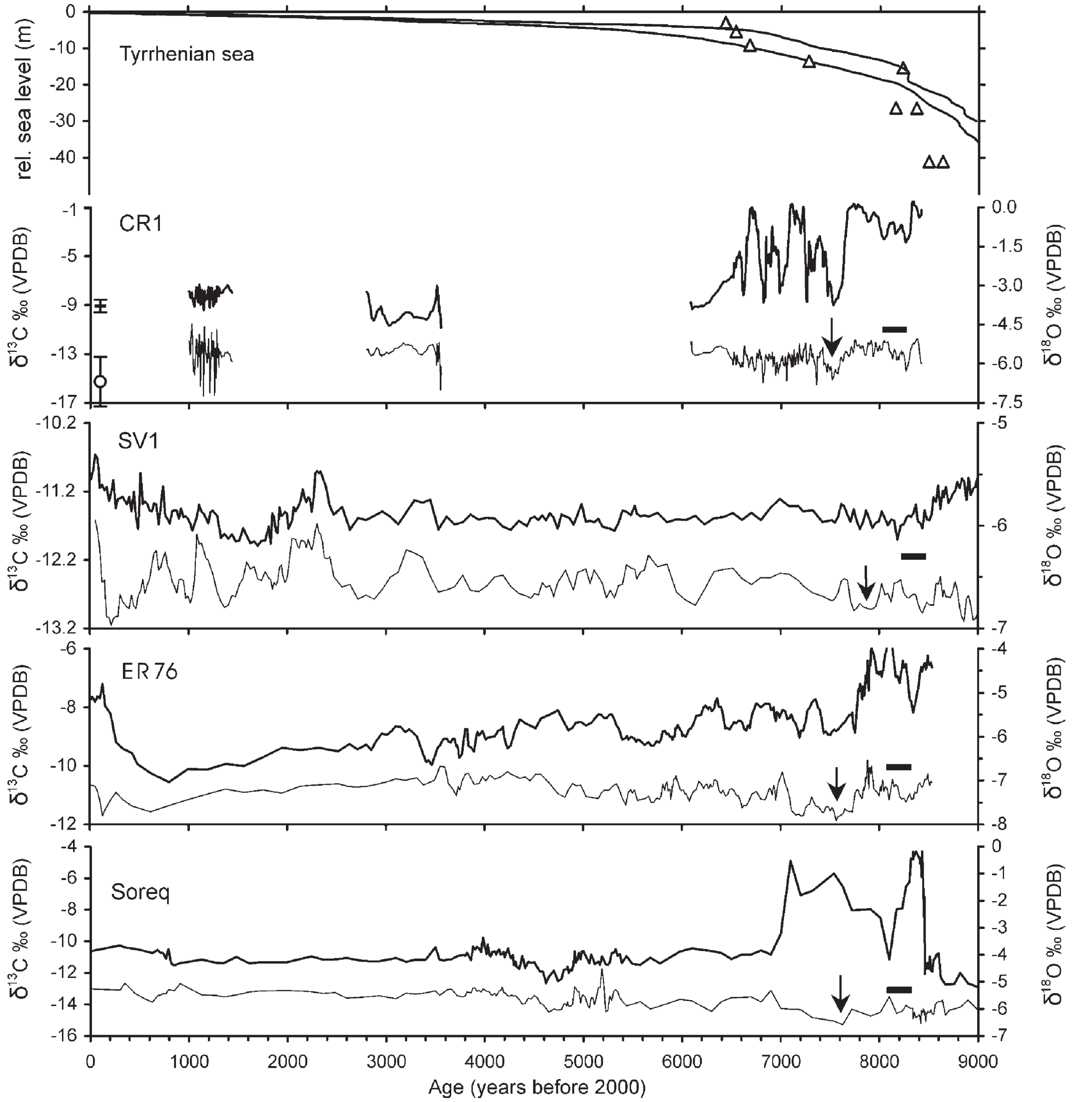

Figure 7. Comparison of Tyrrhenian sea-level changes (top panel; from Antonioli et al., 2001), stalagmites CR1 (3 points running mean), SV1 (Grotta Savi), ER 76 (Grotta di Ernesto) and Soreq cave $\delta^{13}C$ and $\delta^{18}O$ time series. For ER76, the early Holocene age model follows Frisia et al. (2001); annual lamina counting was locked to the oldest U/Th dating with the lowest 1 σ error (7.77±0.09). Consequently, a bottom age of ca. 8600 yr ago was obtained. The thick bars mark the low in $\delta^{18}O$ centered at ca. 8200 yr ago. The arrows mark a second $\delta^{18}O$ low centered at ca. 7500 yr ago. Note the weak trend toward higher $\delta^{18}O$ values from the early Holocene to the present in CR1, SV1 and Soreq Cave records.

1999; McDermott et al., 1999; Frisia et al., 2001, 2005) and render the reconstruction of paleoclimate from CR1 speleothem more reliable. Thus, we believe that robust paleoclimatic and paleoenvironmental information can be obtained from CR1 despite non-equilibrium deposition and discontinuous growth.

We tested the paleoclimate potential of CR1 by first comparing its Holocene isotope time series with those from the eastern Mediterranean Soreq cave speleothems (Bar-Matthews et al., 1999, 2000, 2003), the northern Italian Savi cave SV1 stalagmite (Frisia et al., 2005) and the alpine Grotta di Ernesto cave ER76 stalagmite (McDermott et al., 1999; Frisia et al., 2001) (see Fig. 1 for locations). The C isotope compositions of CR1, ER 76 and Soreq are commonly higher in their early Holocene portions (Figs. 7 and 8). The early Holocene δ^{13}C values from CR1, ER76 and Soreq also show a large variability, which amounts to ca. 10 ‰ in CR1, ca. 8‰ in the Soreq cave record and ca. 3‰ in ER76. It is noteworthy that the early Holocene high δ^{13}C-variability portion of ER76 consists of dendritic fabric (McDermott et al., 1999) and that the relatively large δ^{13}C variability (ca. 7‰) in Holocene portions of the Irish stalagmite CC3 (from Crag Cave, located near the Atlantic shore in SW Ireland) likewise consist of dendritic calcite (McDermott et al., 1999). Little can be said about the Soreq record, as there are no published images of the early Holocene fabrics sampled for stable isotope analyses. By contrast, the fabric of stalagmite SV1 is columnar through the period where the δ^{13}C values are relatively stable (ca. −11.5‰). All speleothems illustrated in Figures 7 and 8 showing a relatively large δ^{13}C variability and commonly higher δ^{13}C values in the early Holocene formed in cave passages beneath less than 30 m of bedrock, whereas SV1 developed ca. 70 m below the surface (Frisia et al., 2005). The timing of early Holocene δ^{13}C maxima and minima do not coincide exactly in CR1, ER76 and Soreq (Fig. 8), but it is remarkable that the relatively large amplitudes in these three records appear to be typical of the early Holocene period from ca. 9000 to ca. 7000 yr ago. In ER76, high δ^{13}C values comparable to those of the early Holocene are recorded only in the past 200 yr (Fig. 7). The early Holocene δ^{18}O records show minima centered at ca. 7500 yr ago in CR1, SV1, ER76 and Soreq (arrows in Fig. 7). A second minimum (of about −1‰) occurs at ca. 8100 to 8400 yr ago (bars in Fig. 7). The mid-Holocene portion of CR1, from ca. 3600 to ca. 2800 yr ago, shows lower oxygen and carbon isotope values than the early Holocene and a small peak centered at ca. 3100 yr ago, which roughly corresponds to similar peaks in the SV1 and ER76 records.

Comparison of the CR1 isotope record with pollen data from Lago di Pergusa (Fig. 9) shows that the trend towards lower δ^{13}C values roughly coincides with the decline of pollen concentration in the lake sediments. In addition, the two major deposition gaps in CR1 occurred during periods characterized by a decrease in the arboreal pollen percentage.

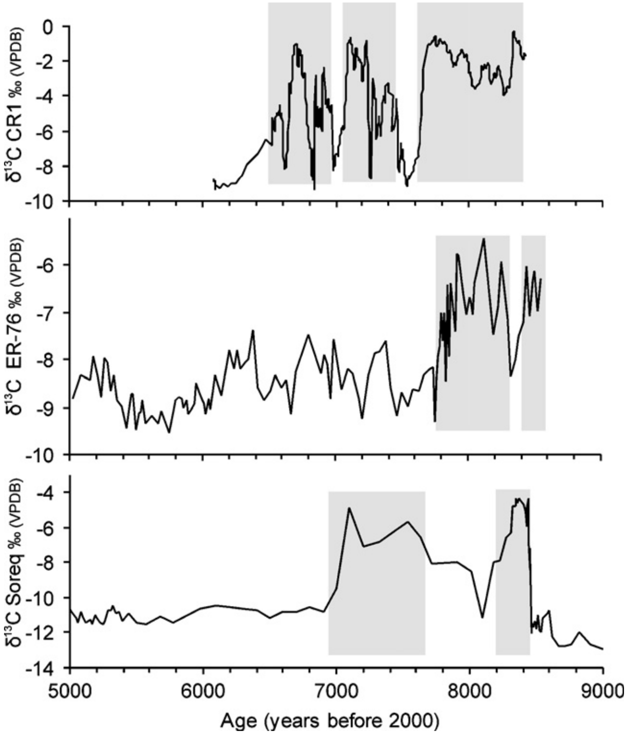

Figure 8. Close-up of the early Holocene δ^{13}C record of CR1 compared to the δ^{13}C records of ER76 and Soreq Cave. Shading marks intervals of high δ^{13}C values. Although they vary in amplitude, timing and number, they seem to be typical of the pluvial early Holocene.

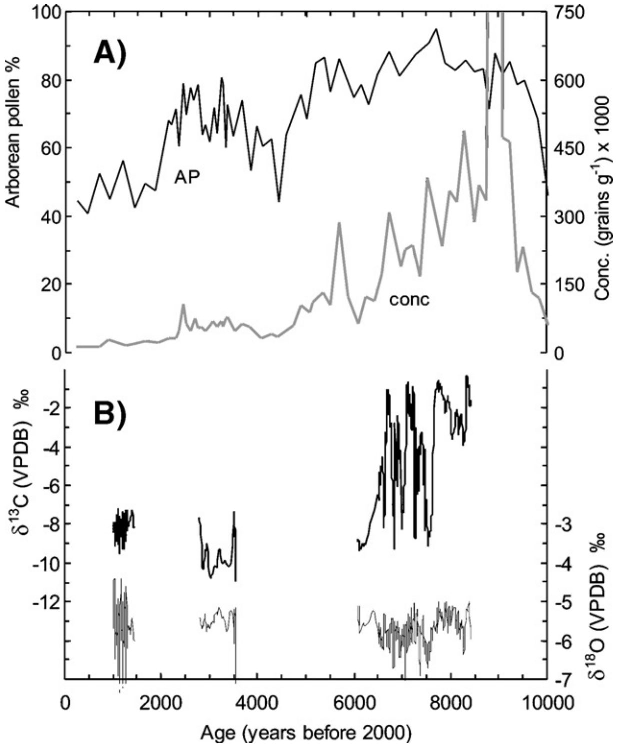

Figure 9. Comparison between the Lago di Pergusa pollen record (A) and the CR1 isotope record (B). The ages for Lago di Pergusa were calibrated using INTCAL 98 (Stuiver et al., 1998). Present was referred to as the year 2000 for a comparison with the stalagmite ages (see text for details). The decrease in total pollen concentration coincides with the trends toward lower δ^{13}C values in CR1. Maximum arboreal pollen (AP) percentages roughly coincide with the pluvial early Holocene phase.

Factors controlling δ^{13}C and δ^{18}O variations in CR1

The elevated δ^{13}C values recorded by CR1 in the early Holocene appear to reflect some regional climate or environmental changes, rather than solely local hydrological conditions. High $\delta^{13}C$ values of speleothem calcite can be the result of several factors, including Rayleigh distillation processes related to CO_2 degassing and calcite precipitation (Bar-Matthews et al., 1996; Mickler et al., 2004, 2006), forced degassing related to oscillating cave ventilation (Spötl et al., 2005), calcite deposition in the vadose zone (Baker et al., 1997), non-equilibrium isotopic incorporation due to the growth mechanisms of the dendrites (Frisia et al., 2000), short soil–water residence time resulting in hindered CO_2 hydration reaction at pH ≤ 9 (Lasaga, 1981), enhanced dissolution of the host rock during wet periods (Bar-Matthews et al., 1999), minimal soil and vegetation cover above the cave (Bar-Matthews et al., 2003; Drysdale et al., 2004), a higher proportion of C4 as opposed to C3 plants (Bar-Matthews et al., 1997) and changes in soil activity that control the production of soil $P(CO_2)$ (Genty et al., 2003).

In the early Holocene part of the CR1 record, the $\delta^{13}C$ values in the −8.0 to −11.0‰ range can be considered as representative of calcite precipitated from drip waters that were almost at equilibrium with CO_2 respired from C3 plants (McDermott, 2004). By contrast, the high $\delta^{13}C$ values might reflect the influence of CO_2 respired from C4 plants during dry periods. The pollen data from Lago di Pergusa, however, do not support a shift from C3-dominated to C4-dominated vegetation in the period between ca. 9000 and ca. 7000 yr ago. The hypothesis of high $\delta^{13}C$ values related to enhanced dissolution of the host rock seems unlikely because of the slow rate of the kinetic reactions involved (Lasaga, 1981) and the very thin rock cover above the CR1 site. Rayleigh distillation due to calcite precipitation in the soil zone commonly results in a progressive increase of calcite $\delta^{13}C$ and $\delta^{18}O$ values (Mickler et al., 2006), both of which show little co-variation in the early Holocene portion of CR1 (Fig. 6). Minimal microbial activity in the soil above the cave has been suggested to be one of the most likely causes of large shifts towards high $\delta^{13}C$ values (ca. −6‰) in a French stalagmite from Villars cave during periods of deteriorating climate (Genty et al., 2003). At a regional scale, high $\delta^{13}C$ values in cave calcites are associated with shifts towards relatively high $\delta^{18}O$ values and reduced growth rates during cold and dry extreme events, as observed in the Villars and Savi records (Genty et al., 2003; Frisia et al., 2005). In CR1, relatively high growth rates and $\delta^{18}O$ values within the range of the present-day calcite do not support cold and dry extreme conditions as responsible for high $\delta^{13}C$ values in the early Holocene portion of the stalagmite.

High $\delta^{13}C$ values in speleothem calcite coupled with low $\delta^{18}O$ values have been variously interpreted as related to hot and dry climate (Frumkin et al., 2000) or to enhanced rainfall which stripped the soil zone whereby the infiltrating waters had little interaction with soil CO_2 (Bar-Matthews et al., 2003). Bar-Matthews et al. (2003) observed that during intense storms, the dissolved inorganic carbon of cave drips has $\delta^{13}C$ values that are enriched by up to 9‰ with respect to the values observed in normal rainfall conditions. A study on modern calcite precipitated in shallow passages from Harrison Cave, Barbados, also demonstrated that the timing of maximum $\delta^{13}C$ values closely matches that of maximum rainfall and highest drip rates (Mickler et al., 2004). The same study recognized the importance of residence time of the water in the aquifer in the kinetics of CO_2 hydration and isotopic equilibration between soil CO_2 and H_2O.

Stalagmite CR1 is characterized by dendritic fabric for most of its growth, including the early Holocene portion. This fabric can form when rapid drip rates alternate with long periods of extremely slow discharge, producing $\delta^{13}C$ values up to ca. 5‰ heavier than the columnar fabric in the same speleothems (Frisia et al., 2000). In CR1, dendritic calcite shows both high and low $\delta^{13}C$ values, which excludes fabric-only effect on the carbon isotope composition. Its presence, however, is indicative of a large hydrological contrast. If the growth laminae are annual, as inferred from the number of laminae counted between dated intervals, then it can be inferred that the early Holocene was characterized by a very wet winter and a relatively dry summer.

Degassing of the $HCO^-_{3(aq)}$ reservoir alone is unlikely to be a major cause of the high $\delta^{13}C$ values because ^{13}C enrichment due to CO_2 degassing is relatively small (Mickler et al., 2004) compared to the high variability observed in CR1 calcite. However, if degassing was forced by ventilation, it may have further contributed to the 13C enrichment in CR1 dendritic fabric. In the absence of detectable air currents, cave ventilation is related to changes in atmospheric pressure and temperature differences between the cave interior and the exterior atmosphere, and controls the CO_2 fluxes in shallow caves (Hoyos et al., 1998). It has been hypothesized that gaseous CO_2 diffusing from the soil zone contributes to the CO_2 content and composition of the cave atmosphere (Spötl et al., 2005). The transfer of CO_2 from the cave to the outer atmosphere has been observed to occur in the wetter and cooler season, when atmospheric pressure is low (Hoyos et al., 1998; Spötl et al., 2005). This process controls degassing and consequently the $\delta^{13}C$ values of cave calcite. In the early Holocene, sea surface temperatures (SST) in the Tyrrhenian Sea were higher than today (Antonioli et al., 2001), and for the Mediterranean Sea the temperature change has been estimated to be up to 2.9°C (Marchal et al., 2002). The relative sea level was ca. −35 to ca. −10 m below the modern value (Fig. 7; Antonioli et al., 2001), which resulted in a greater cave to coast distance. A larger sea–land contrast can therefore be hypothesized. This could result in a change of atmospheric pressure and wind fields on a local scale, which enhanced ventilation during the rainy season and degassing in the cave, resulting in higher $\delta^{13}C$ values in the dissolved inorganic carbon $\delta^{13}C$ and ultimately in the calcite. The effects of sea level and SST appear to be possible common features that can explain similar trends in the $\delta^{13}C$ records of stalagmites developed in shallow caves of the Mediterranean region and the Irish coast. A strong seasonal contrast, possibly coupled with ventilation would account for the development of ^{13}C-enriched dendritic fabrics in the early Holocene growth phases of CR1, ER76 and the Irish CC3 stalagmites (McDermott et al., 1999).

Present-day monitoring suggests that the amount of winter rainfall is the most important parameter, which controls stalagmite growth within Grotta di Carburangeli. Maximum rainfall and high drip rates therefore seem to be the most likely causes for the high $\delta^{13}C$ values measured in the early Holocene portion of CR1 (cf. Mickler et al., 2004). We suggest that maxima in the early Holocene $\delta^{13}C$ record of CR1 reflect periods of very wet winters. During wet winters, the infiltrating waters had only short interaction with soil CO_2, degassing was possibly enhanced by ventilation (strong sea–land contrast), and kinetic phenomena likely contributed to the formation of calcite with high $\delta^{13}C$ values. By contrast, low $\delta^{13}C$ values reflect periods of relatively dry winters, when the infiltrating waters had time to equilibrate with soil CO_2 evolved from C3 vegetation, and degassing was less intense.

By assuming that the pattern of calcite precipitation remained the same until the present day, we infer that the $\delta^{18}O$ composition of fossil CR1 calcite mostly reflects changes in the isotopic signal of the winter rainfall and processes in the soil zone (McDermott, 2004). Temperature effects on the calcite–water fractionation related to cave temperature changes, however, cannot be discarded for millennial $\delta^{18}O$ variability. The Holocene $\delta^{18}O$ time series of CR1 show a slight increasing trend from the early Holocene to ca. 600 AD, when the stalagmite ceased to grow. At present, North Atlantic air masses are the dominant source of winter moisture in Sicily. Air masses originating in the North Atlantic acquire moisture when crossing the Mediterranean Sea, giving rise to abundant snowfall or rainfall in southern Italy in late autumn and winter (Pinna, 1977). As the modern calcite $\delta^{18}O$ value is in the range of fossil calcite, it is possible that the North Atlantic source of winter moisture has not changed in the past 10,000 yr. The $\delta^{18}O$ record encoded in CR1 therefore likely reflects one or more of the following: (i) changes in the $\delta^{18}O$ of the North Atlantic ocean surface; (ii) changes in air temperature in the source area and in the target area (northern Sicily); and (iii) long-term shifts of the trajectories of low and high pressure systems.

The most distinguishing feature of the ca. 600 to ca. 1000 AD part of CR1 is the co-variance of the $\delta^{13}C$ and $\delta^{18}O$ values (Figs. 6A–C). The $\delta^{13}C$ values of the mid- and late-Holocene portions of CR1 are depleted with respect to the average values for the early Holocene (Fig. 7) and are typical of calcite formed in equilibrium with CO_2 respired from C3 plants (McDermott, 2004). Co-variation can be ascribed to near-surface evaporative processes in semi-arid settings (Bar-Matthews et al., 1996) and the effects of Rayleigh-type distillation (Mickler et al., 2004). If our interpretation of the $\delta^{18}O$ variability in terms of the combined effect of isotopic composition of winter rainfall and evaporation in the soil zone is correct, shifts to higher $\delta^{18}O$ in the growth phase from ca. 600 to ca. 1000 AD should be indicative of drier winters or more ^{18}O-enriched winter rains. In this last growth phase of CR1, decadal-scale peaks of high $\delta^{18}O$ and $\delta^{13}C$ values appear to coincide (but not always) with higher ^{14}C production rate (Fig. 10). Mangini et al. (2005) attributed a positive correlation between $\Delta^{14}C$ and $\delta^{18}O$ in a stalagmite from the Alps to variability in hydrology or moisture sources. In the case of CR1, it appears that periods of lower solar activity resulted in reduced winter rainfall and enhanced evaporation in the soil zone.

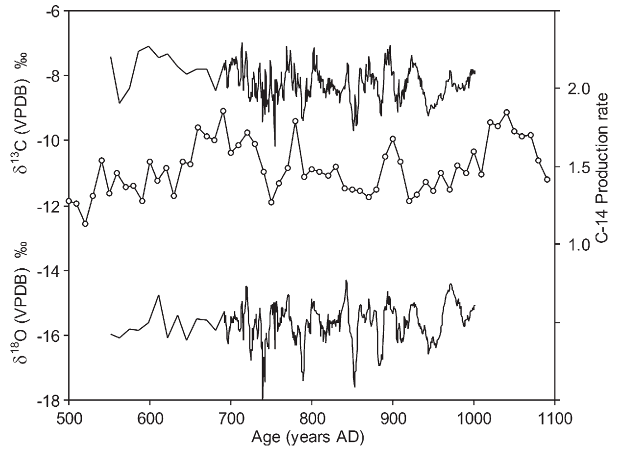

Figure 10. Stable isotope records of the third growth phase (ca. 600 to ca. 1000 AD) recorded by of stalagmite CR1 compared to the radiocarbon production rate. Higher radiocarbon production broadly coincides with calcite of higher C and O isotope values.

Climate changes and possible influences on past societies

The $\delta^{18}O$ and the $\delta^{13}C$ variability in CR1 most likely record climate-related hydrological changes. The isotope record of CR1 suggests that in the early Holocene, up to ca. 7500 yr ago, the climate in Sicily was wetter than today, and the dendritic fabric suggests strong seasonal contrast. The lacustrine record from Lago di Pergusa indicates very wet climatic conditions and strong seasonal contrast before 8000 yr ago, when the maximum expansion of arboreal biomass is recorded (Sadori and Narcisi, 2001; Fig. 9). The wettest early Holocene interval broadly coincides with "Sapropel event 1" recognized in the Soreq record (Bar-Matthews et al., 2003), higher SST in the Tyrrenian Sea and the maximum rate of Holocene sea-level rise (Antonioli et al., 2001).

Little can be said about temperature. Low $\delta^{18}O$ values at ca. 8100 to ca. 8500 yr ago, which are also present in the records of ER76, SV1 and Soreq cave, may correspond to a multi-century cooling (from ca. 8000 to ca. 8500 yr ago) related to solar output fluctuations (Rohling and Pälike, 2005). The climate deterioration from ca. 8000 to ca. 8500 yr ago is also recorded by Tyrrhenian spelean serpulids (Antonioli et al., 2001) and by a decrease in the percentage of warm species in marine sediments from the Sicilian Channel and Aegean Sea (Rohling et al., 2002; Sprovieri et al., 2003). Model results, however, show that both temperature and precipitation decreased during the "8200 event" (Morrill and Jacobsen, 2005). Thus, competing effects of temperature and rainfall rate possibly influenced the $\delta^{18}O$ value of CR1 and other Mediterranean stalagmites. Due to the influence of multiple climatic factors, therefore, the $\delta^{18}O$ records of CR1 shows little evidence for a significant "8200-yr" cooling and associated aridity with respect to other speleothem records (McDermott et al., 2001; Baldini et al., 2002). A second negative $\delta^{18}O$ peak centered at ca. 7500 yr ago could also be due to combined temperature and precipitation effects on precipitation–evaporation during a period of reduced solar activity (see Rohling and Pälike, 2005). The fabric that corresponds to more depleted $\delta^{18}O$ values shows sub-euhedral crystals filling open dendrites. This suggests a change from variable to relatively constant drip rates (Frisia et al., 2000).

If our interpretation of the stable isotope data and fabric development in CR1 is correct, the climate anomalies recorded by the concurrent negative $\delta^{18}O$ and $\delta^{13}C$ values in the early Holocene suggest weaker evapotranspiration during cooler periods. From ca. 7500 to ca. 6500 yr ago changes in hydrology show a greater amplitude with respect to the previous early Holocene phase. Wet winters were still predominant, but the overall wetter-than-today conditions were interrupted abruptly (every ca. 200 yr) by multi-decadal-long dry spells (Fig. 7). Since ca. 7500 yr ago, therefore, the duration of pluvial winter-phases shortened, and by ca. 6500 yr ago present-day climate conditions were established. The CR1 record suggests that there was a ca. 1000-yr-long transition from the early Holocene to the late Holocene climate modes.

A decrease in total pollen concentration marks the onset (ca. 6900 yr ago) of a trend towards aridification in the sediments from Lago di Pergusa (Sadori and Narcisi, 2001) (Fig. 9). Bar-Matthews et al. (2003) interpreted the Holocene δ^{18}O trend observed in the Soreq record as reflecting decreasing rainfall from ca. 7500 yr ago to the present and suggested that this agrees with lake-level and monsoon-strength data from North Africa and the Middle East. The rainfall at Soreq is mostly derived from the Eastern Mediterranean, whereas North Atlantic air masses are the dominant source of rainfall and snowfall in Sicily. The CR1 trend to very slight δ^{18}O enrichment and δ^{13}C depletion since the early Holocene suggests changes in the climate phenomena, which brought winter moisture generated in the North Atlantic to the Sicilian coast. The overall trend is consistent with the decrease in Northern Hemisphere insolation (Berger, 1978), but investigation of the causes of Holocene winter rainfall in Sicily is outside the scope of this paper.

At Grotta dell'Uzzo, the late Mesolithic commenced ca. 9000–8500 yr ago by an expansion of food resource exploitation. Marshland birds, marine fish, marine mollusks and mammals and wild legumes became part of the diet. A similar, efficient exploitation of food resources from a wide range of habitats characterizes the Mesolithic in Northern Italy (Pedrotti, 2001). By ca. 7700 yr ago, fully domesticated cattle and pigs, alongside sheep and goat, appear in Grotta dell'Uzzo sequence and are clearly associated with Neolithic material, specifically ceramics. Full Neolithization seems to have occurred at the transition from the early Holocene pluvial phase to the present-day climate. It is not clear if Neolithic colonists entered Sicily and spread their culture, or if late Mesolithic people adopted the Neolithic lifestyle becoming farmers themselves. It is, however, possible that environmental factors, such as a decrease in rainfall, could have prompted some late Mesolithic groups to adopt agriculture (K. D. Thomas, personal communication, 2006). Besides cultural evolution and immigration, it seems possible that climate changes influenced the Mesolithic to Neolithic transition. A series of excavated parallel channels at Stretto Partanna (Fig. 1), which were used ca. 7500 yr ago to divert water into artificial basins (Tusa, 1992), might also have been constructed in response to decreased rainfall.

CR1 ceased to grow ca. 6100 yr ago. The sudden interruption of calcite deposition in CR1, however, can hardly be related to severe arid conditions that persisted for about 3000 yr. In the time interval corresponding to the first depositional hiatus of CR1, speleothems continued to grow at Grotta di Ernesto, Grotta Savi and Soreq Cave (Bar-Matthews et al., 1999; McDermott et al., 1999; Frisia et al., 2005). At about 6000 yr ago, pollen records from Spain suggest a change towards "Mediterranean conditions" whereas decline in arboreal pollen at Lago di Pergusa suggests progressive aridification (Sadori and Narcisi, 2001). Thus, there is the possibility that the first hiatus in CR1 is a local response to a regional climate change. This local response could have been due to human intervention, or to natural clogging of the feeding drip. At present, we can only rely on the observed trend towards higher isotope values recorded prior to stalagmite growth cessation, which supports the "natural" hypothesis.

Calcite deposition on CR1 recommenced ca. 3550 yr ago. The calcite above the first hiatus contains iron oxides, hydroxides and dolomite, which were likely part of the insoluble epikarst and flushed into the cave once the flow path was re-established. Thus, it is possible that this growth phase documents erosion and infiltration of the pre-existing, red soil. Sedimentation at Lago di Pergusa and decrease in arboreal pollen percentage indicate dry climatic conditions from ca. 3000 to ca. 2800 yr ago, which broadly correspond to a decrease in diameter toward the top of the mid-Holocene portion of CR1 (Fig. 2). This portion stopped to grow ca. 2800 yr ago, most probably because of another local change in the hydrological pathway. A major phase of deforestation in the Bronze Age occurred at around this same period in the Mediterranean region (Oldfield and Dearing, 2003). The fabrics and mineralogy of the mid-Holocene growth phase of CR1 document stripping of the original karst land surface. During the hiatus from ca. 2800 to ca. 1400 yr ago, there must have been a significant change at the surface, most probably related to agriculturalism. Thus, it is possible that the second hiatus in CR1 deposition was a local hydrological response to a change in land use. The fabric of CR1 in the upper growth phase is open columnar, which suggests that the replacement of the original red soil by agricultural soil altered some cave parameters, such as the drip rate, or the flux of gas CO_2 from the cave to the outer atmosphere.

At present, availability of water is a central issue in Sicily, in part because of the long-term aridification trend is enhanced by inefficient water resource exploitation and pollution. Past societies probably learned to cope and respond to hydrological changes by adopting new ways of life. Future development in Sicily should build upon the lessons learned from the past, and account for the effects of solar variability on water resources. If our interpretation of CR1 record is correct, solar output has an impact on the winter rainfall and consequently recharge to karst aquifers. The fragmentary Holocene record suggests a possible relationship between solar output and the strength (or trajectory) of the winter cyclones, which reach the NW coast of Sicily. The mechanisms responsible for the amplification of the solar signal are still debated. Some models indicate a solar impact on the meridional structure of atmospheric circulation (Shindell et al., 1999). Furthermore, quantitative studies suggest changes in winter storm-track latitude in the western North Atlantic (Tinsley and Yu, 2004).

Conclusions

This study has examined the paleoclimate evolution of Sicily during the Holocene using the fabric and high-resolution stable isotope composition of a stalagmite, providing new data for this part of the Mediterranean region. Although the stalagmite grew discontinuously, and the mid and late Holocene portions show non-equilibrium stable isotope incorporation, comparison with other regional records suggest that CR1 still provides the following valuable information:

(1) In the early Holocene, from ca. 8500 to ca. 7500 yr ago, high $\delta^{13}C$ values are interpreted as indicative of wet winters. Low $\delta^{13}C$ and $\delta^{18}O$ excursions centered at ca. 8200 and ca.7500 yr ago correspond to long-term climate anomalies when both temperature and precipitation decreased. In this pluvial phase, late Mesolithic people significantly improved food resource exploitation.
(2) A hydrologically unstable period from ca. 7500 to ca. 6500 yr ago marked the transition from a pluvial early Holocene to the present arid climate. In this period, decadal-scale dry spells probably favored the adoption of Neolithic agriculturalism.
(3) The onset of CR1 stalagmite discontinuities at ca. 6000 yr and at ca. 2700 yr ago reflect local hydrological responses both to decreased precipitation and human activities.
(4) Winter rainfall in Sicily at the centennial scale was modulated by variable solar output.

Our study suggests that hydrological variability was, and still is, the most important factor that influenced societal responses to climate changes in Sicily. The relationship between decreased solar output and winter rainfall, and the recognition of the timing, duration and regional effects of large-scale climate anomalies, will be better understood when other well-dated and continuous climate proxy series become available from southern Italy. The possibility that early Holocene hydrological changes had an impact on Neolithization, however, cannot be discarded on the basis of age uncertainties, as the radiometric dating errors of speleothems, lake sediments and archaeological layers still allow overlapping of climate and archaeological events.

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