A late Holocene climate record in stalagmites from Modrič Cave (Croatia)

D. RUDZKA,1* F. MCDERMOTT1 and M. SURIĆ2

1UCD School of Geological Sciences, University College Dublin, Belfield, Dublin 4, Ireland
2Department of Geography, University of Zadar, Tuđmanova 24 I, 23000 Zadar, Croatia

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ABSTRACT: Few terrestrial Holocene climate records exist from south-eastern Europe despite its important geographical position as a transitional climatic zone between the Mediterranean and mainland continental Europe. Here we present new petrographic and stable isotope data for two Holocene speleothems from Modrič Cave, Croatia (44°15′N, 15°32′E), a coastal Adriatic site (120 m inland). Modern meteorological and cave conditions have been monitored for 2 years to understand the links between climate variability and stable isotope time-series records in speleothems. Typical of a Mediterranean-type climate, a negative water balance exists between April and September, so that recharge of the aquifer is restricted to the winter months. The weighted mean δ18O of the rainfall is −5.96‰ (2σ = 2.83), and the weighted mean Δ/D rainfall value is −36.83‰ (2σ = 19.95), slightly above the Global Meteoric Water Line, but well below the Mediterranean Meteoric Water Line. Modern calcite from the tops of each stalagmite exhibits δ18O values that are close to isotopic equilibrium with their respective drip water values. Unfortunately, the relatively young ages and low uranium contents (ca. 50 p.p.b.) of both stalagmites hamper the use of U-series dating. Radiocarbon dates have been used instead to constrain their chronology using a dead carbon correction. Apart from some Isotope Stage 3 material (ca. 55 ka), both stalagmites were deposited during the late Holocene. Climatic conditions during the late Holocene are inferred to have been sufficiently wet to maintain stalagmite growth and any hiatuses appear to be relatively short lived. Inferred changes in the stalagmite diameters during deposition are linked to δ13C and δ18O variations, indicating alternating periods of drier and wetter conditions. Drier conditions are inferred for the late Roman Ages warm period and the mid-Medieval Warm Period. Wetter conditions are associated with the Little Ice Age. Copyright © 2012 John Wiley & Sons, Ltd.

KEYWORDS: cave monitoring; Croatia; late Holocene; paleoclimate; speleothems.

Introduction

Published Holocene palaeoclimate records for the circum-Mediterranean region indicate significant differences between the eastern and western basins (e.g. Bar-Matthews et al., 1997; Magy et al., 2009; Martin-Puertas et al., 2010; Brayshaw et al., 2011; Giraudi et al., 2011; Kuzucuoglu et al., 2011; Peyron et al., 2011; Zanchetta et al., 2011). Holocene climate reconstructions from Croatia can provide a link between the two Mediterranean sub-basins. Although numerous studies of the Holocene terrestrial vegetation history of this central Mediterranean region have been published (Sadori et al., 2011), many of the lake sediment sequences on which reconstructions are based are poorly dated, or are sampled at a resolution that is too coarse to discern sub-millennial late Holocene environmental change. An additional complication is that many terrestrial late Holocene pollen records are compromised by anthropogenic effects (e.g. deforestation and cultural changes), making it difficult to disentangle human- and climate-induced vegetation change in the region (e.g. Di Rita and Magri, 2009; Mercuri et al., 2011).

Information about past climates in the region has also been derived from Adriatic Sea sediment cores. By contrast with the lake sediment records, many of the measurable proxies in these marine cores (e.g. planktonic and benthic foraminifera assemblages) are relatively unaffected by human-induced changes on the adjacent continents, particularly in the more distal sites (Oldfield et al., 2003; Sangiorgi et al., 2003; Piva et al., 2008). In the study by Piva et al. (2008), for example, foraminiferal assemblages, along with palaeomagnetic secular variations and aquatic pollen indicators were investigated in a set of marine sediment cores from the Western Adriatic shelf and the Southern Adriatic deep basin to reconstruct a high-resolution record of environmental and climatic conditions for the past 6000 years. The abundance of foraminifera species indicative of warm/dry conditions was found to increase markedly during climatic optima such as the Medieval Warm Period (MWP) and the Roman Ages, events for which there is a significant body of independent documentary, archaeological and historical evidence. Thus, repeated peaks in the occurrence of Globigerinoides sacculifer (an oligotrophic, shallow-water dweller typical of warm tropical environments, which currently only appears in the Western Mediterranean at the end of summer) reflect warm conditions associated with the MWP and Roman Ages. Similarly, the last occurrence of G. sacculifer marks the onset of the Little Ice Age (LIA), recognized as a cold and relatively wet period in this region. According to Piva et al. (2008), two peaks in Valvuliniera complanata (an opportunistic benthic species associated with a high availability of organic matter as a result of increased river discharge) match well with the coldest phases of the LIA: the so-called Fernau (1590–1630 AD) and Napoleon (1810–1820 AD) intervals. Cold and humid intervals were thus linked to a substantially increased river discharge, suggesting enhanced rainfall during the cold phase of the LIA.

Speleothem records have the potential to give a perspective on the terrestrial environmental changes that may have accompanied the changes recorded in the marine realm discussed above. However, relatively few studies have been published on speleothems from this part of Europe. Previous work from Croatia focused mainly on submerged samples from submarine caves along the Eastern Adriatic coast (Surić and Juračić, 2010). Horvatiničić et al. (2003) reported δ13C and δ18O time-series records for Holocene speleothems and tufas from Croatia (Plitvice Lakes, Zrmanja, Krupa and Krka Rivers) and Slovenia (Postojna Cave, and Podstojnjski Creek). Most samples were dated using the 14C method, but a few from the
Krka and Plitvice areas were dated by U-series (Srdoč et al., 1994; Horvatiničić et al., 2000). Most of the speleothems and all of the tufas indicate late Holocene deposition, although some stalagmites commenced growing during the Late Glacial (ca. 15–12 ka).

A recent study of speleothems from three Croatian caves (Lončar et al., 2011) revealed depositional intervals similar to those reported by Surić and Juračić (2010); however, some samples also indicate Holocene deposition (4 ka to present) similar to that reported here.

Rogerson et al. (2011) presented a stalagmite record from Ponor Jazbina v Rovnjah, Slovenia, for the period ca. 9–0.5 ka. Relatively arid early Holocene conditions were inferred on the basis of slow growth rates and pronounced Sr/Ca maxima. Of more relevance to the present study is the record for the last 4 ka from this Slovenian site. Changes in δ18O and Sr/Ca in this Slovenian stalagmite suggest considerable late Holocene climate variability. Lower δ18O values, coupled with minima in Sr/Ca, are associated with well-documented cool climatic phases such as the LIA and the Late Roman Ages/early Dark Ages, implying reduced aridity. By contrast, higher δ18O values and maxima in Sr/Ca ratios are associated with the Medieval Warm period, interpreted by Rogerson et al. (2011) to reflect increased aridity. These authors also suggested that millennial-scale climate changes during the Holocene were caused by shifts in atmospheric circulation, resulting in greater precipitation during cool phases, and reduced precipitation during the warmer periods.

A stalagmite from Ceremosnja Cave, eastern Serbia, dated using the radiocarbon method due to its low uranium content, preserves a record back to 2300 ± 40 14C a BP (Kacanski et al., 2001). Its palaeoclimate record based on stable isotope data indicates two warm periods between ca. 2275 and 2050 a and 1480–960 a, terminating with a colder phase from 960 a to the present. Gradually decreasing δ18O values in this speleothem during the last 2000 years were interpreted as a cooling trend, but their suggested 5 °C decrease in temperature is difficult to reconcile with the available pollen data for the region (Davis et al., 2003; Wanner et al., 2008). Thus, changes in δ18O are, at least in part, likely to reflect temporal changes in the oxygen isotope ratio of precipitation rather than temperature change alone.

This study presents new data for two speleothems (MOD-21 and MOD-22) from Modrič Cave, Croatia with the aim of reconstructing aspects of late Holocene climate change in a region that can provide links between the two Mediterranean sub-basins. Stalagmite MOD-22 was the more suitable for palaeoclimate studies, and is the main focus of this paper.

Site and sample description

Modrič Cave (44°15’24.6"N, 15°32’14.16"E) was discovered in 1985 close to the eastern Adriatic coastline of Croatia, approximately 30 km north-east of Zadar (Fig. 1). This sub-horizontal cave developed along a faulted contact zone between the Adriatic and the Dinaric structural complexes of the Upper Cretaceous Adriatic carbonate platform (Herak, 1986). The limestone thickness above the cave ranges from ca. 1 to 30 m, and its entrance (1.8 x 1.3 m) faces the coast, at an elevation of 32 m above sea level (m a.s.l.) (Surič et al., 2010). The cave extends eastwards and branches into two main passages (Fig. 1; Miko et al., 2002). All investigations reported
here were conducted in the north passage. The south passage contains evidence for human activity (bones and pottery) as well as bone fragments of Upper Pleistocene fauna (skull of cave bear Ursus spelaeus; Malez, 1987).

The region is characterized by a Mediterranean temperate humid climate with hot summers (Cia type; Köppen, 1936). Vegetation consisting of C3 plants, mainly scrubby grassland with small isolated bushes, is developed on a thin (<0.5 m) terra-rossa soil containing limestone fragments.

In June 2008, two in situ stalagmites (MOD-21 and MOD-22) were retrieved from the cave (Fig. 1). In January 2009, three temperature loggers and two Stalagmate drip loggers were installed to monitor cave environmental conditions and characterize the hydrological behaviour of the drip sites that fed the two sampled stalagmites. Additionally, a Pluvimate rainfall gauge was installed outside the cave, approximately 500 m from the entrance. Water samples from two drip feeders (MODW-21, MODW-22) and rainfall were collected monthly from July 2008 to June 2010 for isotopic analysis.

Drip site MODW-22, the feeder drip of stalagmite MOD-22, is located ~150 m from the cave entrance (Fig. 1), approximately 22 m below the surface. Drip site MODW-21, the feeder drip to stalagmite MOD-21, is located ~210 m from the entrance, at a depth of 22 m below the surface. This chamber is well isolated from the rest of the cave by two constrictions: no. 3 and no. 13 (Fig. 1).

The cave air temperature and the drip-rates at the MODW-21 and MODW-22 sites were monitored from January 2009 to October 2010 using Tinytag Aquatic temperature loggers, with a resolution of 0.01 °C. The drip logger at MODW-22 failed in 2009, and the data are available from November 2009 to October 2010 only. Due to periodic clogging of the external Pluvimate gauge, data from the two nearby meteorological stations, Zadar-Zemunik (88 m a.s.l. and 23 km from Modrič; Croatian Meteorological and Hydrological Service, 2010) and Starigrad-Paklenica (12 m a.s.l. and 8 km from Modrič; Croatian Meteorological and Hydrological Service, 2010), were used to calculate the water balances at the cave site using the Thornthwaite evapotranspiration model (Thornthwaite, 1948; McCabe and Markstrom, 2007). All three sites exhibit the highest temperature in July and August (ca. 25 °C), and a strong water deficit during the summer months. Recharge of the aquifer thus appears to be restricted to the winter and early spring (November–March).

Stalagmite MOD-22, the primary focus of this paper, was an actively growing, 28-cm-long stalagmite when collected (Fig. 2A,B). Its feeder drip is a long (approx. 1.5 m) single stalactite. The opaque white calcite in MOD-22 is softer and more porous than in MOD-21. Along the whole stalagmite there are several darker layers (ca. 1–5 mm thick), some of which include visible clay-rich horizons.

Speleothem MOD-21 was an actively growing 23.5-cm-long stalagmite located approximately 210 m from the cave entrance under the Jellyfish formation (Fig. 2C,D). During its early stages of deposition MOD-21 appears to have had two feeder drips, although all analyses are focused along the central axis of the stalagmite. Analysis of visible changes in the carbonate petrography of MOD-21 and radiocarbon dating revealed an erratic growth rate. The record from this speleothem is shown in detail in the supporting information (Appendix S1).

Methods
A 0.1-mm-diameter dental drill bit was used to obtain ~3 mg of carbonate powder from stalagmite MOD-22 at 2.5-mm intervals along its growth axis, producing 111 samples. The time interval between successive drill holes represents approximately 16 years on average in MOD-22, but this varies between 4 and 26 years, depending on the growth rate.

Oxygen and hydrogen isotope measurements on the water samples were carried out at SILLA (Stable Isotope & Luminescence Laboratory) at the University of Birmingham, UK. All water data were normalized to V-SMOW standards. Analysis of the first batch (ca. 50%) of stable isotope measurements on the carbonates was carried out at the Stable Isotope Laboratories at Royal Holloway University of London (UK), and the second batch was analysed by the Isot-Analytical laboratory (UK). All data for carbonates are reported relative to the V-PDB standard.

A small number of U-series isotope measurements were carried out at University College Dublin, using a ThermoFisher Neptune multi-collector inductive coupled plasma mass spectrometer equipped with an Aridus desolvation nebulizer. Sample preparation prior to column chemistry involves suspending ca. 200 mg of calcite powder in deionized H2O in a 15-mL teflon Savillex beaker and spiking with a
mixed spike \((^{233}\text{U}-^{236}\text{U}-^{229}\text{Th})\). After spiking, samples were dissolved gradually in 7 M HNO\(_3\) and left to equilibrate for ca. 24 h. BioRad AG 1 × 8 Resin (200–400 mesh) ion exchange columns were used for U and Th separation and purification.

All radiocarbon measurements were conducted by the Poznan Radiocarbon Laboratory, Poland, using a 1.5 SDH-Pelletron Model Compact Carbon accelerator mass spectrometry using the Oxalic Acid II (OxII) standard (Goslar et al., 2004). Radiocarbon dates were calibrated using the age calibration program OxCal4 (Bronk Ramsey, 2009) with IntCal 09 calibration curves (Reimer et al., 2009).

**Results**

**Cave air temperature**

Mean annual air temperature for the years 1961–2000 from the Zadar Puntamika and Starigrad meteorological stations (1992–2010) is 14.43 and 16.00 °C respectively. Meteorological data from the Starigrad station (ca. 8 km NW from Modrič and 12 m a.s.l.) were used as a proxy for external air temperature at the Modrič site.

Temperature logger TL1, located nearest the cave entrance (Fig. 1), recorded an average air temperature of 12.96 °C (2σ = 4.1), but this site partly reflects seasonal variations in external air temperature with a range of ca. 7.42 °C and a delay of ca. 2 months relative to the seasonal changes in external air temperature. By contrast, temperature logger TL2, deployed at the MODW-22 drip site (Fig. 1), exhibits a much more stable temperature with a mean annual value of 15.46 °C (2σ = 0.08). The amplitude of seasonal variations at this site is c. 0.19 °C, markedly lower than at TL1. As expected, temperature logger TL3 (at drip site MODW-21, Fig. 1) shows the most stable temperature of all three sites. Small temperature variations do occur at this site, but their amplitude (0.09 °C) is very small. The mean cave air temperature value calculated from one full year of data from TL3 is 15.64 °C (2σ = 0.04). Overall, air temperature in the interior of the cave (loggers TL2 and TL3) records relatively constant temperatures that are similar to those of the nearby Starigrad meteorological station.

**Drip rate and rainfall data**

The objective of this part of the study was to characterize the hydrological behaviour of the drip sites (MODW-21 and MODW-22) that feed stalagmites MOD-21 and MOD-22 to aid with the interpretation of the stalagmite stable isotope data. Data from both drip loggers, along with rainfall data from the Pluvimate logger at Modrič, were converted from drips per hour to millilitres per day based on the radius of the feeding stalactites (Collister and Mattey, 2008). These data were compared with the rainfall record inferred for the Modrič site (Fig. 3) and with two other meteorological stations: Zadar-Zemunik (NOAA, 2010) and Starigrad (Croatian Meteorological and Hydrological Service, 2010). In general, the rainfall events occur at the same time, but the magnitude of precipitation events at Starigrad is greater than that recorded at Modrič. The Modrič precipitation record is in reasonable agreement with that from Zadar. Summer months tend to be relatively dry at Modrič over the short monitored period, with the exception of July 2009, when a heavy rainfall event was recorded. A similar rainfall event occurred simultaneously at the Zadar station.

Drip site MODW-22 is very slow (average ca. one drip every 4.77 min), and it remained relatively insensitive to precipitation events over the period of its monitoring from October 2009 to September 2010 (Fig. 3). The drip logger data are in good agreement with field observations in June 2008, when MODW-22 was observed to drip approximately every 5 min, before and after rainfall. Unfortunately, the MODW-22 record from January to October 2009 is unreliable due to the logger failure, and is not shown in Fig. 3. During the period from November 2009 to February 2010, the drip rate remained quite constant with the average value of 19.23 mL day\(^{-1}\) (average drip rate of 1 drip/6.21 min), but showed a sharp increase in mid-February, followed by a gradual decrease to an average value
of 30 mL day\(^{-1}\) (ca. 1 drip/3.52 min). At the beginning of July 2010, recording was interrupted for 9 days. From mid-July to September 2010, drip MODW-22 shows a lower average drip rate of 19 mL day\(^{-1}\). The average amount of water entering through this drip site during the entire period is ca. 25.19 mL day\(^{-1}\) (755.80 mL per month).

Drip MODW-21 is faster than MODW-22, with an average drip rate of 48.28 mL day\(^{-1}\), but is much more sensitive to rainfall events. The amount of water dripping at this site is very variable, but overall there is a gradual decrease from the winter towards the summer months (Fig. 3). Drip rates at MODW-21 increased with a ca. 2- to 5-day delay in response to intense rainfall events, as seen in the Starigrad and Zadar meteorological stations (Pluvimate logger was blocked). This delay is approximately constant, and is especially prominent during the period from January to May 2009. From June to October 2009 MODW-21 appears to be unresponsive to rainfall events, possibly due to a high evapotranspiration rate and a negative water balance at this time of year.

During the periodic visits to the cave for water collection it was noticed that both drip sites never dried up completely. Overall, the drip rate data from MODW-21 and MODW-22 are in good agreement with the monthly water balance calculations using the Thornthwaite evapotranspiration model (Thornthwaite, 1948; McCabe and Markstrom, 2007) based on the record from meteorological stations that indicate a strong summer water deficit and recharge of the aquifer mostly during the winter months. The smoother trends of MODW-22 and the persistence of its drip flow during summer 2010 points to a greater fracture-controlled storage component than at MODW-21. In the scheme of Smart and Friedrich (1987), both sites are classified as ‘seepage flow’ drips.

### Drip and rain water isotope data

The residence time of water in the bedrock above the cave and the nature of the moisture sources can be investigated using drip water and rainfall D/H and \(\delta^{18}O\). Knowledge of the water residence time in the aquifer is crucial to detect possible seasonal biases in the speleothem \(\delta^{18}O\) signal. A drip site fed by water with a very short residence time, for example, could be biased towards the seasonal rainfall isotope signal associated with the wet season. As discussed below, this appears unlikely for the Modrič drip sites studied here.

The \(\delta^{18}O\) and D/H data for rainfall (Fig. 4) sampled monthly for nearly 2 years at Modrič, exhibit some seasonal variability, with lower values in the winter months (weighted mean \(\delta^{18}O = -6.93\%\), mean D/H = -43.30\% SMOW) and higher values in the summer months (mean \(\delta^{18}O = -5.30\%\), mean D/H = -32.50\% SMOW). The weighted mean \(\delta^{18}O\) is -5.96\% (2\(\sigma = 2.83\)), and the range of \(\delta^{18}O\) is 5.38\% V-SMOW. The weighted mean D/H rainfall value is -36.83\% (2\(\sigma = 19.95\)).

By comparison with the rainfall data, the oxygen isotope ratios at MODW-21 are buffered, and show only limited seasonal variability of ca. 1.08\% V-SMOW (Fig. 4). Thus, the annual mean \(\delta^{18}O\) for this site for the year 2009 is -6.40\% (2\(\sigma = 0.54\)). Site MODW-22 shows somewhat more seasonal variability (ca. 2.18\%, V-SMOW) during the period analysed (June 2008 to September 2010), and its annual mean value for the year 2009 is -5.19\% (2\(\sigma = 1.6\)). The mean \(\delta^{18}O\) from drip site MODW-22 perhaps coincidentally shows a value close to the weighted summer mean \(\delta^{18}O\) of rainfall from Modrič, while mean \(\delta^{18}O\) from MODW-21 is close to the weighted winter mean \(\delta^{18}O\) of rainfall. Drips at the MODW-22 site show higher \(\delta^{18}O\) values in winter (December–March) and lower values in summer and appear to be strongly

**Figure 4.** A 2-year record of water isotopes (D/H and \(\delta^{18}O\)) from Modrič rainfall water and the two drip sites: MODW-21 and MODW-22. Arrows labelled ‘A’ and ‘C’ represent the mean \(\delta^{18}O\) and D/H for drip waters from sites MODW-22 and MODW-21, respectively. Arrows labelled ‘B’ represent weighted mean \(\delta^{18}O\) and D/H in the rainfall. This figure is available in colour online at wileyonlinelibrary.com.
out of phase with rainfall values (Fig. 4). The partial preservation of seasonal \( \delta^{18}O \) variability in the MODW-22 drip data suggests a ‘piston-flow’ behaviour in which newly recharged rainfall pushes through previously stored, partly mixed water.

The annual mean D/H value of MODW-21 drip water for the year 2009 is \(-38.33\%\) (\(2\sigma = 4.51\)), and that from MODW-22 for the same period is \(-28.15\%\) (\(2\sigma = 13.41\)). Overall, the \( \delta^{18}O \) and D/H values for drip waters from MODW-21 are more strongly buffered than those from MODW-22.

The D/H and \( \delta^{18}O \) data for drip and rainfall waters were also used to investigate the vapour sources of the rainfall that predominantly recharge these sites. The Global Meteoric Water Line (GMWL) and the Mediterranean Meteoric Water Line (MMWL) are shown in Fig. 5(A), along with the Modrič drip and rainfall data. This figure shows that the Modrič water samples plot closer to the GMWL (D/H = \(8^\circ\delta^{18}O + 10\)) than the MMWL (D/H = \(8^\circ\delta^{18}O + 22\)). The Local Meteoric Water Line (LMWL) is given by D/H = \(8^\circ\delta^{18}O + 13\). Also shown for comparison is the LMWL for the Zadar region (Vreca et al., 2006). For comparison, the averaged data from the Croatian, Italian and Slovenian GNIP sites are also shown in Fig. 5(A) (IAEA/WMO, 2006). Figure 5(B) illustrates an inverse correlation between the average monthly rainfall amount and \( \delta^{18}O \) of rainfall at the Zadar GNIP station that influenced speleothem \( \delta^{18}O \).

**Age model of MOD-22**

Unfortunately, the Modrič stalagmites are characterized by low uranium contents (typically ca. 50–70 p.p.b. in MOD-22 and ca. 30 p.p.b. in MOD-21), young ages and relatively high \( ^{232}Th \) contents, making U-series dating impossible (Table 1). Attempts to apply corrections for the detrital contamination in the Holocene U-series age determinations for MOD-22 (Table 1) were unsuccessful, because of their very low \( ^{230}Th/^ {232}Th \) ratios (typically <3) and young ages. Corrected ages were very sensitive to the choice of \( ^{230}Th/^ {232}Th \) in the detrital component and as a result meaningful ages could not be calculated. Corrected ages that were in line with the corrected and calibrated \(^{14}C \) ages for MOD-22 (discussed below) required detrital \( ^{230}Th/^ {232}Th \) values of ca. 1.2–1.5, within the range of 0.8 ± 0.8 (\(2\sigma\)) commonly used for detrital thorium corrections (Richards and Dorale, 2003).

![Figure 5](wileyonlinelibrary.com)
Table 1. U-series measurements performed on stalagmites MOD-21 and MOD-22.

| Sample    | Distance (mm) | $^{235}$U (pMC) | Raw $^{14}$C age (BP) | DE applied (%) | $^{235}$U (pMC) after DE correction | Calibrated $^{14}$C age (cal BP 1950) | Calibrated $^{14}$C age (AD/BC) | Uncorrected age (ka) | Corrected age (ka) | Error (ka) |
|-----------|---------------|-----------------|-----------------------|----------------|-------------------------------------|--------------------------------------|---------------------------------|---------------------|-------------------|------------|
| MOD-22-R1 | 145           | 25              | 181 ± 0.08            | 23.7 ± 0.2     | 246 ± 0.73                          | 298 ± 0.84                           | 120 ± 0.08                      | 28 ± 0.08            | 120 ± 0.08         | 1.0E-05   |
| MOD-22-R2 | 267.5         | 71.18 ± 0.03    | 2810 ± 35             | 12.5 ± 0.1     | 91.3 ± 0.02                         | 276 ± 0.02                           | 126 ± 0.02                      | 100 ± 0.02           | 91.3 ± 0.02        | 1.0E-05   |
| MOD-22-R3 | 39.2          | 71.18 ± 0.03    | 2810 ± 35             | 12.5 ± 0.1     | 91.3 ± 0.02                         | 276 ± 0.02                           | 126 ± 0.02                      | 100 ± 0.02           | 91.3 ± 0.02        | 1.0E-05   |
| MOD-22-R4 | 145           | 25              | 181 ± 0.08            | 23.7 ± 0.2     | 246 ± 0.73                          | 298 ± 0.84                           | 120 ± 0.08                      | 28 ± 0.08            | 120 ± 0.08         | 1.0E-05   |
| MOD-22-R5 | 35            | 84.68 ± 0.31    | 1375 ± 30             | 12.5 ± 0.1     | 94.0 ± 0.02                         | 271 ± 0.02                           | 126 ± 0.02                      | 100 ± 0.02           | 94.0 ± 0.02        | 1.0E-05   |
| MOD-22-R6 | 35            | 84.68 ± 0.31    | 1375 ± 30             | 12.5 ± 0.1     | 94.0 ± 0.02                         | 271 ± 0.02                           | 126 ± 0.02                      | 100 ± 0.02           | 94.0 ± 0.02        | 1.0E-05   |

Table 2. Results from $^{14}$C measurements on MOD-22. Measured $^{14}$C activity ($\Delta^{14}$Cm) was corrected for the dilution effect (DE). The raw $^{14}$C age was calibrated to years BP (1950) and AD/BC using OxCal (Bronk Ramsey, 2009) and calibration curves from Reimer et al. (2009).

| Sample    | Distance (mm) | $^{235}$U (pMC) | Raw $^{14}$C age (BP) | DE applied (%) | $^{235}$Th/$^{232}$Th (activity ratio) | Error (cal BP) | Calibrated $^{14}$C age (cal BP 1950) | Corrected age (AD/BC) | 95.4% probability (age BC/AD) |
|-----------|---------------|-----------------|-----------------------|----------------|--------------------------------------|--------|--------------------------------------|------------------------|-----------------------------|
| MOD-21-U1 | 225           | 30.21           | 4.3E-06               | 0.86           | 0.88                                 | 6.8E-03 | 62.297                               | 0.733                  | 1.419                        | 1.4E-03                  | 55724                       | 4593 1602 |
| MOD-21-U2 | 225           | 30.21           | 4.3E-06               | 0.86           | 0.88                                 | 6.8E-03 | 62.297                               | 0.733                  | 1.419                        | 1.4E-03                  | 55724                       | 4593 1602 |
| MOD-21-U3 | 225           | 30.21           | 4.3E-06               | 0.86           | 0.88                                 | 6.8E-03 | 62.297                               | 0.733                  | 1.419                        | 1.4E-03                  | 55724                       | 4593 1602 |
| MOD-21-U4 | 225           | 30.21           | 4.3E-06               | 0.86           | 0.88                                 | 6.8E-03 | 62.297                               | 0.733                  | 1.419                        | 1.4E-03                  | 55724                       | 4593 1602 |

For these reasons, radiocarbon and U-series dating methods have been combined in an attempt to constrain a chronological model for these speleothems (Tables 1 and 2, Table S1 in Appendix S1). Because radiocarbon activities in stalagmites are strongly affected by the incorporation of ‘dead’ carbon from limestone and aged soil-derived carbon, raw $^{14}$C activities must be first corrected for this reservoir effect (called here as a dilution effect – DE) prior to calibration. Dead carbon values can vary between and within different cave sites, depending on vegetation cover, soil productivity, hydrological factors and limestone dissolution rates (e.g. Genty et al., 1999, 2001; Genty and Massault, 1999; Rudzka et al., 2011). The age calibration program OxCal (Bronk Ramsey, 2009) was used to calibrate the $^{14}$C data using calibration curves from Reimer et al. (2009). Calibrated dates reported in Table 2 and Table S1 (Appendix S1) were chosen with the highest probability.

For stalagmite MOD-22, the dead carbon value was estimated using a quasi-linear growth rate model for the sample, with the top surface of the stalagmite anchored to the present day (active when collected). The uncorrected $^{14}$C data to the DE value (black solid curve in Fig. 6) from stalagmite MOD-22 increase monotonically with distance, indicating quasi-linear growth rates. This points towards relatively stable soil carbon turnover and relatively constant limestone dissolution rates. The ca. 1050-year offset when comparing these $^{14}$C data with the last data point (anchored by the year of collection) (Fig. 6) is taken to reflect the ‘dead carbon’ effect for this sample. This corresponds to a dead carbon value of approximately 12.5% (Table 2, Fig. 6). Probability density functions of calibrated dates from stalagmite MOD-22 calculated after a correction to the DE value of 12.5% are shown in Fig. 7.

The six $^{14}$C measurements from MOD-22 indicate that it was a fast-growing, late Holocene stalagmite. Deposition commenced around 331 ± 96 AD (1619 ± 96 cal a BP) (Table 2), and continued at an almost linear rate prior collection in 2008.

Figure 6. Calculation of dilution effect (DE) of $^{14}$C data from stalagmite MOD-22. Dashed curve shows calibrated $^{14}$C data after applying DE = 12.5% correction taken directly from the MOD-22 growth rate. Black solid curve represents $^{14}$C data for MOD-22, uncorrected and incorrected to the DE value.
Its average growth rate is 304 μm year⁻¹. In detail, however, this rate varies between the dated intervals, with a noticeable increase in growth rate between 1345 ± 59 and 1580 ± 96 AD (605 ± 59 and 370 ± 96 cal a BP), at a distance 155–35 mm from the top.

**Speleothem isotope data**

`Hendy tests` on MOD-22 (Fig. 8A) show no correlation between δ¹⁸O and δ¹³C along the examined laminae, indicating no evidence for strong disequilibrium (kinetic) isotope fractionation effects. The stable isotope record for stalagmite MOD-22 shows only moderate variations along the whole time series (Fig. 8B). δ¹⁸O varies by about 2‰ and δ¹³C by about 4‰. The mean δ¹⁸O value is −4.11‰ V-PDB (2σ = 0.93), and the mean δ¹³C value is −7.37‰ V-PDB (2σ = 1.74). Through the whole period of deposition of MOD-22 there are two distinctive trends of increasing δ¹⁸O, first in the interval between 245 and 190 mm from the top and again between 130 and 85 mm from the top. δ¹³C shows at least five increasing trends through the time series (black arrows, Fig. 8B).

Based on the modern drip-water δ¹⁸O values for both stalagmites MOD-21 and MOD-22, and the present-day cave temperature, model δ¹⁸O values for modern calcite were calculated using equations from Craig (1965), Friedman and O’Neill (1977), Kim and O’Neill (1997), Coplen (2007), Dietzel et al. (2009) and Tremaine et al. (2011). Values obtained based on the Friedman and O’Neill (1977) and Kim and O’Neill (1997) calculations are ca. 1‰ higher than the measured δ¹⁸O values for MOD-21 and MOD-22. Values obtained using calculations from Dietzel et al. (2009) are ca. 0.5‰ higher than measured values for MOD-21 and MOD-22. Calculations based on the equations of Craig (1965), Coplen (2007) and Tremaine et al. (2011) seem to reflect the measured δ¹⁸O values to within 0.65‰, implying calcite deposition close to isotopic equilibrium (e.g. McDermott et al., 2011).

**Interpretations**

Overall, there are no high-amplitude seasonal variations in the cave air temperature at sites MODW-21 and MODW 22 (ca. 0.09 and 0.19 °C, respectively), indicating that both of these sites faithfully record the mean annual air temperature of the region averaged over multi-annual time scales. Drip sites MODW-21 and MODW-22 both display a ‘seepage flow’ character in the Smart and Friedrich (1987) classification. However, the MODW-21 drip site responds more rapidly to rainfall events, particularly from autumn to early spring, suggesting some ‘seasonal drip’ character. The presence of a small storage water reservoir and a ‘seepage flow’ component in the MODW-21 drip site could account for its behaviour from January to May 2009. The fact that most of stalagmite MOD-21 grew rapidly over a short time interval in the late Holocene (Fig. S2B) strongly suggests relatively constant water infiltration during this period.
By contrast with MODW-21, drip site MODW-22 is characterized mostly by a ‘seepage flow’ character (Fig. 3). The drip rate increases between February and March 2010, and after that it decreases very gradually, remaining essentially insensitive to rainfall events. This type of drip rate behaviour suggests aquifer re-charge during the winter, and its slow exhaustion during the subsequent months.

Rainfall at the Modrić site is probably derived from a combination of several sources. Data for the cave and meteoric waters are close to the GMWL on a plot of D/H and $d^{18}O$, which indicates a predominance of moisture from the Atlantic Ocean. Data from the MODW-21 drip site plot entirely on the LMWL of Vreća et al. (2006); however, data from drip site MODW-22 plot above this LMWL, possibly suggesting slower infiltration through the soil and consequently greater evaporation effects. The latter effect causes possible isotopic enrichment prior to infiltration (Wackerbarth et al., 2010).

Drip waters from MODW-21 do not show large temporal variations in either D/H or in $d^{18}O$, suggesting relatively efficient mixing of waters above the drip site, despite its more ‘flashy’ hydrological response (Fig. 3). The marked attenuation of the seasonal meteoric water D/H and $d^{18}O$ signals suggests a relatively long (multi-annual) residence time for the water feeding MODW-21. While the MODW-21 drip waters do not follow the D/H and $d^{18}O$ of rainfall, the site is hydrologically responsive to the rainfall events, suggesting the presence of a fracture-controlled stored water reservoir (buffering of isotopic signal).

Site MODW-22 exhibits higher D/H and $d^{18}O$ values during the winter, and appears to be out of phase with the seasonal trends in meteoric $d^{18}O$ and D/H. Overall, mean $d^{18}O$ from MODW-22 displays has a value similar to that of summer rainfall mean $d^{18}O$, and is higher than the mean $d^{18}O$ value for MODW-21. Delayed and attenuated seasonal signals in the isotopic record of MODW-22 and a relatively constant drip rate suggest piston-flow-type behaviour with incomplete mixing between the summer- and winter-recharged waters. Nonetheless, its hydrological behaviour indicates the presence of an important base-flow component that is not seen in MODW-21 (Fig. 3). Assuming that the present-day hydrological (drip-rate) characteristics of these sites have remained relatively constant, the drip monitoring data indicate that the two sites have different hydrological thresholds. Thus, site MODW-21 and by inference stalagmite MOD-21 would be expected to cease

Figure 8. (A) Hendy test for stalagmite MOD-22. Each sample set was drilled from the single growth layer (distances are measured from the top: A, 25 mm; B, 85 mm; C, 135 mm). Distance was measured from the stalagmite central axis (schematic diagram inserted). (B) Stable isotope record with the position of the radiocarbon dates for MOD-22. This figure is available in colour online at wileyonlinelibrary.com.
dripping (growing) during prolonged (multi-anual) drought periods, whereas the more important storage component of MODW-22 could permit continued dripping (and growth) during prolonged dry intervals.

Age models based on $^{14}$C dates and DE corrections indicate that both stalagmites were deposited in the late Holocene, apart from some Marine Isotope Stage 3 material (ca. 55 ka) near the base of MOD-21 (supporting information, Appendix S1). The climatic conditions during the late Holocene were therefore sufficiently wet to maintain stalagmite growth, and any hiatuses in MOD-22 appear to be relatively short.

Because the radiocarbon chronology depends on the ‘dead carbon’ correction, it should be used cautiously for palaeoclimatic interpretations. Nonetheless, an attempt was made to identify intervals of wetter and drier conditions (Fig. 9) based on petrography and stalagmite diameter. Using the rationale of Railsback et al. (2011), wetter periods are inferred when a growth layer (or layers) flow down and drape over a previously deposited layer on the flanks of the stalagmite, whilst drying trends are inferred when layers are narrower and are perched upon previously deposited layers. Based on these assumptions, we infer at least five intervals of different climatic regimes in MOD-22 (drying trends – arrows in Fig. 9).

Based on the corrected and calibrated $^{14}$C ages, the approximate duration of previously recognized distinctive climatic intervals recorded during the late Holocene [e.g. Roman Warm Period (RWP), Dark Ages cold phase, MWP and LIA] are also shown (Fig. 9).

Drier conditions, recognized by a gradual narrowing of stalagmite MOD-22 during deposition, are usually associated with trends towards higher $\delta^{13}$C (e.g. at ca. 512 ± 76 AD and 900 ± 76 AD, Fig. 9). At least five such drying-out trends are highlighted on Fig. 9. In this interpretation, two very pronounced drier episodes occur in the lower part of MOD-22. This is consistent with the more compact character of the calcite. Using the available chronology, these two dry episodes appear to correspond relatively well with the RWP and the transition from the Dark cold phase Ages to the MWP, and are in agreement with Piva et al. (2008) and Rogerson et al. (2011) who suggested warmer and drier conditions during these two late Holocene periods in the Adriatic region. Drier/warmer conditions during the transition from the Dark Ages to the MWP are also noted in the stalagmite record from Ceremosinja Cave, eastern Serbia (Kacanski et al., 2001).

Overall, relatively wet conditions dominate the middle to upper part of MOD-22 (between 1348 ± 59 and 1580 ± 96 AD) and growth rates were exceptionally high. In this interval the calcite is more porous and overlapping layers are more pronounced (Fig. 9), and the subsequent layers wrap over the previously deposited layers for almost the whole length of the speleothem (marked with dashed lines highlighting the growth layers on the stalagmite scan in Fig. 9). However, two shifts to higher $\delta^{13}$C values during this interval are noted and are interpreted as drying-out trends. $\delta^{18}$O tends to be relatively low during this interval, consistent with wetter conditions and an influence of rainfall amount on $\delta^{18}$O (Fig. 5B). This wet interval inferred from MOD-22 indicates increased rainfall and colder conditions at the beginning of the LIA in agreement with Piva et al. (2008) and Rogerson et al. (2011).

Conclusions

Dead-carbon-corrected and calibrated radiocarbon ages from the two stalagmites indicate that deposition occurred mostly during the late Holocene, aside from some Marine Isotope Stage 3 material (ca. 55 ka) at the base of stalagmite MOD-21 (supporting information, Appendix S1). Drip site MODW-22, the feeder to stalagmite MOD-22, indicates a water storage component, which is reflected in the continuous growth of MOD-22. This permitted continuous stalagmite growth during prolonged (multi-anual) dry periods such as the RWP and MWP. Overall, the data indicate alternating wet and dry conditions during the late Holocene. Drier conditions inferred for the late RWP and early MWP are
also consistent with data by Piva et al. (2008) for the region. Inferred drier conditions during the MWP are consistent with the suggestion that this interval was dominated by a persistent positive North Atlantic Oscillation, leading to drier conditions in the circum-Mediterranean region (Trouet et al., 2009).

The hydrological data for drip site MODW-21, the feeder to stalagmite MOD-21, indicate a minimal water storage component consistent with its erratic growth history. In order to sustain speleothem growth at this site, sustained relatively wet conditions are essential. These conditions probably existed during the interval when MOD-21 grew very rapidly, as indicated by the two almost identical radiocarbon dates at points along the growth axis that are ca. 70 mm apart (supporting information, Appendix S1). However, the absolute timing of this interval is uncertain. Piva et al. (2008) and Magny et al. (2009) have indicated that the LIA was a wet period in this region. This wet phase is consistent with the fast growth rates observed in stalagmite MOD-22 during this interval (Fig. 7), and with its stable isotope data and growth layer geometry. It is possible that MOD-21 also grew rapidly during the LIA, but this would require an unusually high DE value (ca. 20%) to correct its 14C dates (supporting information, Appendix S1).

Overall, the study indicates relatively constant climatic conditions during the late Holocene. However, there is evidence from accelerated deposition rates and growth layer geometry for a wetter period during the early LIA compared with the late Holocene average. Relatively linear growth rates in MOD-22 during the inferred drier intervals indicate that changes in hydrological conditions were not sufficient to cause prolonged cessations of speleothem deposition.

Supporting information
Additional supporting information can be found in the online version of this article:
Appendix S1. Isotopic record from speleothem MOD-21
Fig. S1. Uncalibrated and uncorrected age model for MOD-21
Fig. S2. Hendy test for stalagmite MOD-21, and interpretation of stable isotope records from stalagmite MOD-21.
Table S1. Results from 14C measurements on stalagmite MOD-21.

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Abbreviations. DE, dilution effect; GMWL, Global Meteoric Water Line; LIA, Little Ice Age; LMWL, Local Meteoric Water Line; MWP, Medieval Warm Period; MiMWL, Mediterranean Meteoric Water Line; RWP, Roman Warm Period.

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