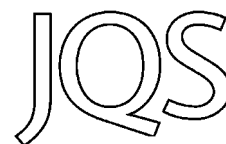


Multiproxy record for the last 4500 years from Lake Shkodra (Albania/Montenegro)



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Received 25 January 2012; Revised 5 June 2012; Accepted 7 June 2012

ABSTRACT: A multi-proxy record is presented for approximately the last 4500 cal a BP from Lake Shkodra, Albania/Montenegro. Lithological analyses, C/N ratio and $\delta^{13}\text{C}$ of the organic and inorganic carbon component suggest that organic matter and bulk carbonate are predominantly authigenic. The $\delta^{18}\text{O}$ record of bulk carbonate indicates the presence of two prominent wet periods: one at ca. 4300 cal a BP and one at ca. 2500–2000 cal a BP. The latter phase is also found in southern Spain and Central Italy, and represents a prominent event in the western and central Mediterranean. In the last 2000 years, four relatively wet intervals occurred between ca. 1800 and 1500 cal a BP (150–450 AD), 1350–1250 (600–700 AD), 1100–800 (850–1150 AD), and at ca. 90 cal a BP (1860 AD). Between ca. 4100 and 2500 cal a BP $\delta^{18}\text{O}$ values are relatively high, with three prominent peaks indicating drier conditions at ca. 4100–4000 cal a BP, ca. 3500 and at ca. 3300 cal a BP. Four additional drier events are identified at 1850 (ca. 100 AD), 1400 (ca. 550 AD), 1150 (800 AD) and ca. 750 cal a BP (1200 AD). The pollen record does not show changes in accordance with these episodes owing to the poor sensitivity of vegetation in this area, which is dominated by an orographic rainfall effect and where changes in altitudinal vegetation belts do not affect the pollen rain in the lake catchment. However, since ca. 900 cal a BP a significant decrease in the percentage arboreal pollen and in pollen concentrations suggest major deforestation produced by human activities. Copyright © 2012 John Wiley & Sons, Ltd.

KEYWORDS: Lake Shkodra; late Holocene; Mediterranean; palaeoclimate; stable isotopes.

Introduction

Future climate predictions suggest that changes in rainfall and water resources will have important socio-economic and political impacts over the Mediterranean region (Lionello *et al.*, 2006). Therefore, understanding the past hydrological variability in this region is an essential prerequisite for establishing future climate scenarios. The last ca. 5000–6000 years is regarded as being particularly relevant because the boundary conditions of the climate system have not changed substantially (in comparison with larger glacial–interglacial changes or at the beginning of the Holocene), and represents the period when an environment and climate comparable with that of today was established (e.g. Wanner *et al.*, 2008).

However, in the Mediterranean basin the long history of human occupation and activities makes it problematic to discriminate unequivocally between climate and non-climatic influences on the environment, especially during the mid to late Holocene (e.g. Roberts *et al.*, 2004, 2011a; Magny *et al.*, 2012). One means by which to sidestep this problem is through the application of stable isotope analyses to environmental archives (e.g. speleothems, lake sediments, pedogenic carbonates), whose isotopic composition is mostly independent of direct anthropogenic activity (Roberts *et al.*, 2010). In particular, stable isotopes on lake deposits can be used to assess the spatial coherency of the climate changes across the

Mediterranean region, especially given the wide geographical distribution of lakes. Recently, the number of available records has increased, allowing the compilation of a regional synthesis (Roberts *et al.*, 2008). However, many records possess poor chronologies (Zanchetta *et al.*, 2007) and low resolution (for the late Holocene in particular there is poor geographic coverage), implying that the palaeohydrological and palaeoclimatic information for most of the Mediterranean region is incomplete and thus poorly understood. Here we seek to redress this situation by presenting a high-resolution (multi-decadal) multiproxy record from the freshwater coastal Lake Shkodra (Fig. 1) for the last ca 4500 cal yr BP.

Site description

Lake Shkodra is a 45-km-long, 15-km-wide, shallow lake (5 m mean depth) situated at ca. 5 m a.s.l. (Fig. 1). It covers part of a flat depression surrounded by NW–SE-elongated relief (up to 2750 m a.s.l.). The catchment consists of carbonate rocks (limestones and dolomites), with minor exposures of siliciclastics, which supply sediments to the Moraca River, the main surface inflow to the lake. The only outlet of the lake is the Bojana River, which flows to the Adriatic Sea. The area is dominated by a Mediterranean-type climate. Rainfall received in the lake catchment (Podgorica and Shkodra meteorological stations) generally ranges between 2000 and 2800 mm a⁻¹ but some areas receive over 3000 mm annually on average. Summer aridity is pronounced with a mean monthly rainfall of <50 mm for July. The average annual air temperature is ca.

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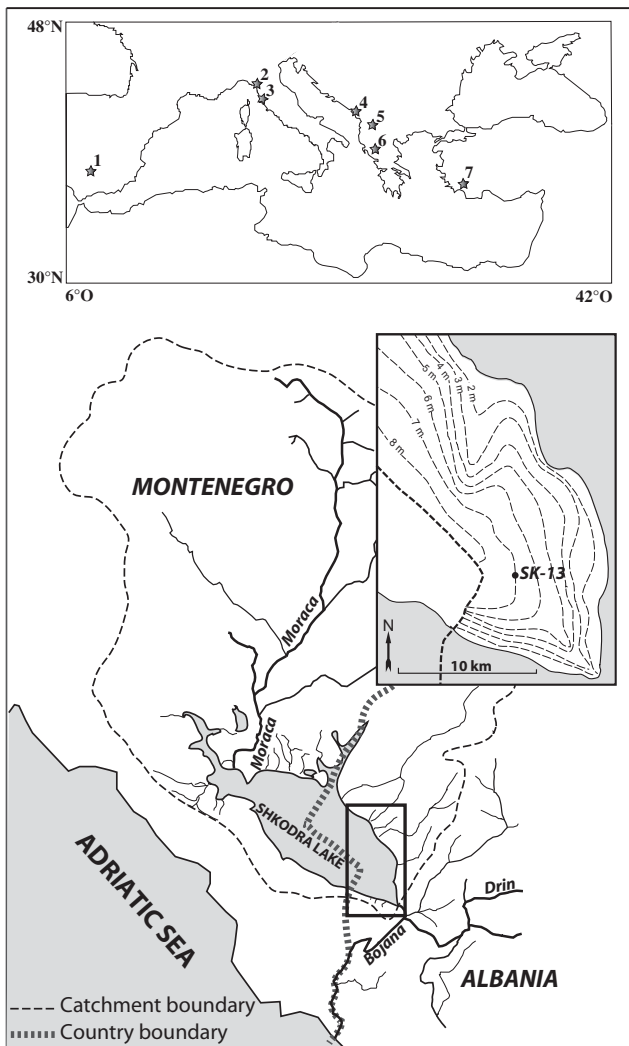


Figure 1. Location map of Lake Shkodra and core location (lower panel) and location of the sites quoted in the text (upper panel): 1, Zonjara Lake; 2, Renella Cave; 3, Accessa Lake; 4, Shkodra Lake; 5, Prespa and Ohrid lakes; 6, Ioannina (Pomvotis); 7, Gölhisar Lake.

15°C. Occasional flash floods combined with a relatively high catchment/lake ratio (11:1) and the bathymetry can produce large (up to 5 m) lake-level fluctuations, such as those observed between 1956 and 1970 (Lasca *et al.*, 1981; Keukelaar *et al.*, 2006).

The water residence time in the lake is estimated to be ca. 120 days (Keukelaar *et al.*, 2006). A significant karst system with submerged springs contributes to inflow to the lake (Karaman and Beeton, 1981), which compensates for summer evaporation. This plus the rapid residence time suggests that water level is not dominated by evaporative effects. van Welden *et al.* (2008) studied the last 500 years of sedimentation of the lake from two short cores (SK-17 and SK-06) and showed that this period was characterized by undisturbed sedimentation of fine-grained carbonate-dominated material, despite the incidence of destructive earthquakes that affected the area in 1905 and 1979.

Materials and methods

Several cores were retrieved in September 2003 using a UWITECTM platform and coring device. Core SK13 (7.8 m long) was selected due to its more central position within the lake and for the evident continuity in sedimentation (Fig. 1). The chronological framework was established by four ¹⁴C accelerator mass spectrometry measurements (Table 1)

Table 1. Radiocarbon data, tephra layers and depths from core SK13. Note that the age of the Agnano Mt Spina tephra (AMS) is reported as the best estimate performed by Blockley *et al.* (2008).

Lab. label/Tephra	Core depth (cm)	Age, ¹⁴ C a BP	Age, cal a BP (2σ)
SacA	17	200 ± 30	305–0
Poz-15211	115	745 ± 30	728–661
Poz-15212*	288	1695 ± 30	1694–1535
Pollena tephra	300–295	—	1478 ¹
FL tephra	514	3150 ± 60 ²	3215–3480
Avellino tephra	571	3530 ± 40 ³	3920–3690
Poz-15214	614	3760 ± 35	4238–3990
AMS tephra	726–716	—	4690–4300 ⁴

¹Rosi and Santacroce (1983); ²Coltelli *et al.* (2000), ³Zanchetta *et al.* (2011), ⁴Blockley *et al.* (2008). * This age has been excluded from the age model for the obvious incongruence with the historical age of the tephra.

obtained from terrestrial plant macrofossils and on the basis of the presence of four tephra layers of known age (Sulpizio *et al.*, 2010; Fig. 2A). The ¹⁴C ages were calibrated using the IntCal 04 curve (Reimer *et al.*, 2004; Table 1). The youngest tephra is historically documented (the AD 472 'Pollena' eruption, Rosi and Santacroce, 1983) whilst for the others the best estimated ages available in the literature have been used (Table 1). The age model was discussed in detail by Sulpizio *et al.* (2010) and for completeness of information is reported in Fig. 2(B). The age model indicates that the sedimentation rate is, on average, ca. 0.2 cm a⁻¹. One sample (thickness ca. 1 cm) corresponds to ca. 5 years.

Samples for stable isotopes on bulk carbonate were collected every 5 cm (indicating a sample resolution of ca. 30–50 years) and dried in an oven at 50°C for 48 h. Samples were gently disaggregated and sieved at 100 μm to separate biogenic remains (e.g. ostracods and shells) from the sediments and then the fraction below 100 μm was powdered and homogenized. A preliminary test on ten random samples was performed to verify the effect on the presence on organic matter. Aliquots of the homogenized sample were digested for 24 h with H₂O₂ to oxidize organic matter. One aliquot was analysed for ¹³C/¹²C and ¹⁸O/¹⁶O ratios without pre-treatment. Because we do not find significant differences between treated and untreated samples, the entire succession was analysed without further pre-treatment following the recommendation of Wierzbowski (2007). Herein we refer to the isotopic composition of these samples as 'bulk', and represented by the notation δ¹⁸O_c and δ¹³C_c. Measurements were made using a GV Instruments GV2003 continuous-flow isotope-ratio mass spectrometer at the Advanced Mass Spectrometry Unit, University of Newcastle, Australia. Samples were digested in 105% phosphoric acid at 70°C. Mass spectrometric measurements were made on the evolved CO₂ gas. Results were normalized to the Vienna Pee Dee Belemnite scale using an internal working standard of Carrara Marble (NEW1–Newcastle), previously calibrated against the international standards NBS18 and NBS19. Mean analytical precision for both δ¹⁸O and δ¹³C is better than 0.1‰.

One aliquot of the sample was treated with 10% HCl to remove carbonate, then washed several times with deionized water to neutral pH, and dried again at 40°C. The isotopic composition of bulk organic matter was obtained at the IGG-CNR of Pisa by producing CO₂ by combustion using a Carlo Erba 1108 elemental analyser, interfaced to a Finnigan DeltaPlusXL via the Finnigan MAT Conflo II interface, calibrated using a within-run laboratory standard (graphite and ANU-sucrose). Average analytical reproducibility for these

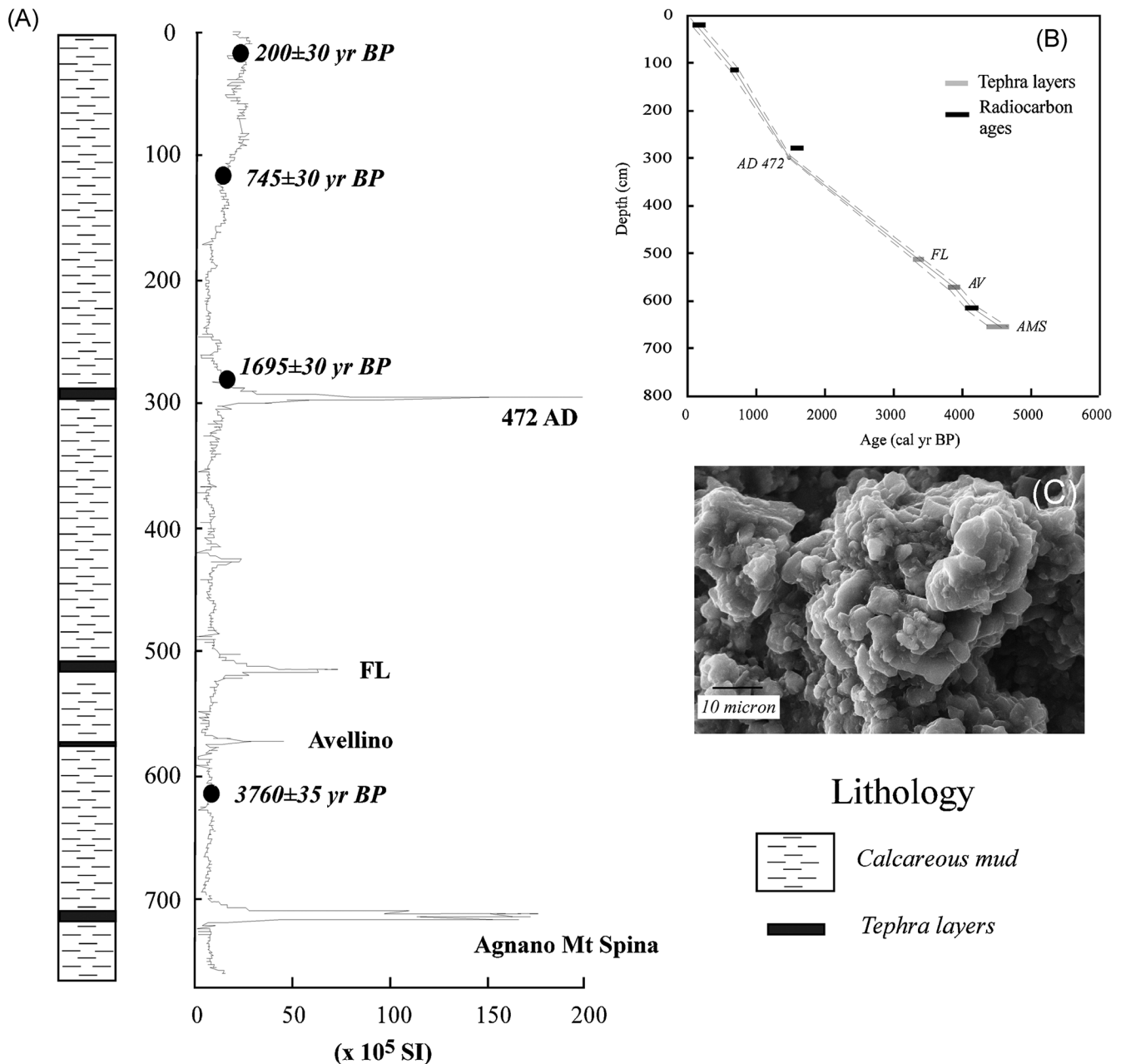


Figure 2. (A) Lithology, magnetic susceptibility, tephra layers and position of radiocarbon dates for core SK13; (B) age model for core SK13; (C) SEM image of sediments – scale bar = 10 μm . Note the calcite crystal.

samples was $<0.1\%$. Concentrations of total carbon (TC), total organic carbon (TOC) and total nitrogen (TN) were measured with a Carlo Erba 1108 elemental analyser, with measurements calibrated against an Acetanilide standard (precision generally $<0.1\%$). The high-resolution carbonate content of Lake Shkodra was determined by combining hydrochloric acid to the laser grain-sizer according to the method of Arnaud (2005). Randomly selected samples ($n=20$) were ground to a fine powder and analysed by X-ray diffraction for defining the main mineral component. A few selected samples at different depths were mounted on glass and observed under a scanning electron microscope (SEM) coupled to an energy-dispersive X-ray analyser (Fig. 2C).

The isotopic composition of regional meteoric precipitation and lake isotope data are necessary for proposing a model for the interpretation of the oxygen isotope composition of lake carbonate (e.g. Baroni *et al.*, 2006; Leng *et al.*, 2010b). As there are no local monitoring programmes for the stable isotopes of local rainfall and lake waters, we have used the IAEA data (<http://ishohis.iaea.org> for Dubrovnik, Komiza-Vis island,

Zadar Malinka-Krk island) and those reported by Longinelli and Selmo (2003) and Anovski (2001) to obtain a regional estimate. In addition, lake-water samples were collected sporadically between 2008 and 2011 (Table 2). The oxygen isotopic composition was determined by the water- CO_2

Table 2. Water isotopic composition of Lake Shkodra and the inlet, the Moraka River.

Date	$\delta^{18}\text{O}$ (‰)	$\delta^2\text{H}$ (‰)
August 2008 (Moraca River)	-7.43	-44.1
August 2008 (lake water)	-5.84	-33.6
November 2009 (lake water)	-6.01	-36.2
June 2010 (lake water)	-7.69	-49.9
June 2010 (Moraka River)	-9.33	61.7
June 2010 (Moraka River: mouth)	-8.70	-56.5
May 2011 (lake water)	-7.12	-42.4
May 2011 (lake water)	-7.21	-44.3
May 2011 (lake water)	-7.25	-44.2

equilibration mass spectrometry method at 25 °C using a Finnigan MAT252 at IGG-CNR. For hydrogen isotope analysis, waters were reduced to H₂ at 450 °C using Mg and analysed using a Europa Scientific GEO 20-20 mass spectrometry. Water isotope data are reported with respect to V-SMOW.

Pollen samples were prepared at ca. 10- to 20-cm intervals. About 0.5 g of dry sediment was chemically treated with HCl (30%), HF (40%) and hot NaOH (10%). *Lycopodium* tablets were added to each sample to calculate the pollen concentration. More than 50 samples were analysed under the microscope, with counts of ca. 300 terrestrial pollen grains on average per sample. The pollen content was very rich and generally well preserved. All pollen count data were reduced to the arboreal pollen/non-arboreal pollen (AP/NAP) curve.

Results

Lithology

Core SK13 core is dominated by relatively massive light-brown calcareous mud (Fig. 2A). X-ray diffraction (XRD) and SEM analyses on the non-clay fraction indicate that the prominent mineral phase throughout the core is calcite followed by minor quartz. The carbonate fraction is mainly composed of calcite crystals of ca. 10 µm (Fig. 2C), typical of bioinduced precipitation (e.g. Kelts and Hsü, 1978). XRD for some samples revealed a peak just above background value at 30.80–31.00 2θ, which may correspond to the principal peak of dolomite. An estimate performed with XRD using mixtures of known amounts of calcite and dolomite suggest that the dolomite content, if present, should be significantly less than 1%.

Visual inspection and magnetic susceptibility investigations from the core have previously revealed the presence of four tephra layers (Table 1), which have been extensively discussed by Sulpizio *et al.* (2010). The CaCO₃ content shows large variability (Fig. 3) with the highest values at ca. 1000 cal a BP (ca. 60%), but mirroring generally the trend of δ¹³C_c (i.e. when calcite increases δ¹³C_c decreases and vice versa, Fig. 3). This is particularly evident between 3500 and 3000 cal a BP or at ca. 1000 cal a BP where lowest δ¹³C_c values correspond to highest CaCO₃ content or since 1000 cal a BP when δ¹³C_c values increase and percentage carbonate progressively decreases. Statistically, this negative correlation is very poor considering the whole record ($r^2 < 0.2$), but it is higher in the first 1000–1500 years ($r^2 = 0.7$ and 0.5, respectively) and between 3200 and 3700 years ($r^2 = 0.5$). Three prominent spikes with very low CaCO₃ values are centred at ca. 1430, 2550 and 4350 cal a BP. The first and the last correspond relatively well to episodes of tephra deposition (Pollena and Agnano Mt Spina). Although the macroscopic tephra thicknesses were excluded from the age model, volcanoclastic sedimentation, settling of the finest tail of the tephra and bioturbation appear responsible for these spikes having diluted the carbonate fraction. Excluding these intervals, the carbonate content record shows five main intervals. After a first phase with a decreasing trend from ca. 4500 to ca. 4200 cal a BP, values remain relatively high (ca. 50%) and stable up to ca. 3700 cal a BP, after which an interval with lower percentage carbonate occurs, lasting until ca. 1250 cal a BP. In this interval there are four main phases of significant decrease, two, as already noted, at ca. 2250 and 1430 cal a BP and the other centred at ca. 1700 cal a BP and lasting ca. 200 years. The longer phase of lower values corresponds to the first part of this interval, lasting ca. 1000 years (3700–2700 cal a BP). At ca. 1250 there is a sharp increase in carbonate content reaching the highest values at ca. 1050 followed by progressively lower values.

TOC (Fig. 3) shows a first phase, ending at ca. 2500 cal a BP, that is characterized by values between ca. 3 and 6% followed

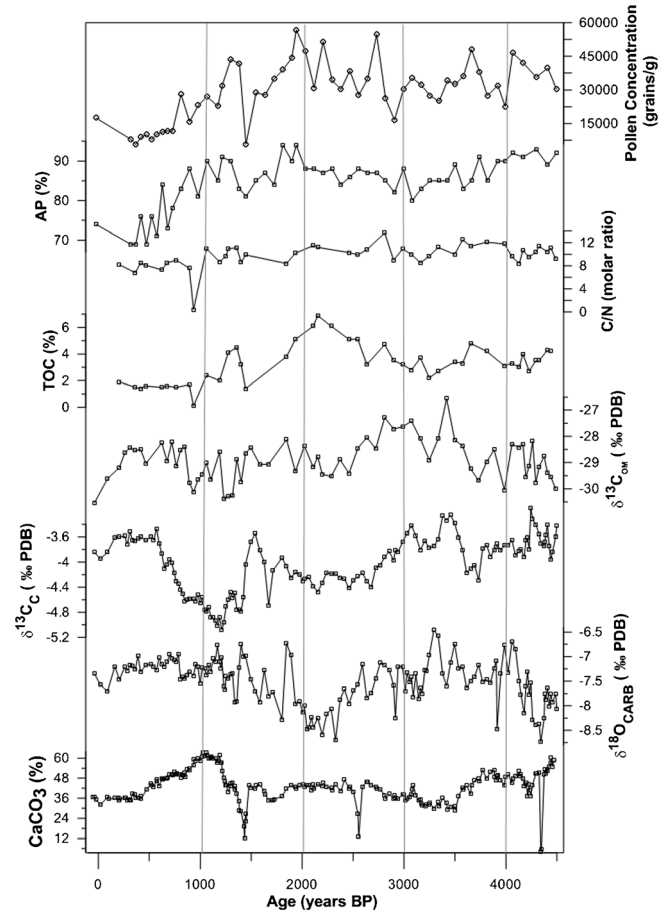


Figure 3. Proxy data from core SK13 plotted versus age (cal a BP). δ¹⁸O_c and δ¹³C_c correspond to bulk carbonate; δ¹³C_{om} corresponds to organic matter; AP, arboreal pollen percentage.

by an interval displaying a sustained increase and reaching the highest values of the record at ca. 2100 cal a BP (8.7%). A decrease follows this peak, reaching a low at ca. 1450 cal a BP, before rising again. The lowest value occurs at ca. 950 cal a BP. The last part of the record is fairly constant at around 2%. The C/N ratio is, on average 8.3 ± 1.7. From ca. 4500 to 900 cal a BP they are slightly higher (8.9 ± 1.0) than the interval after ca. 900 cal a BP (6.8 ± 0.6).

Stable Isotope Record

The δ¹⁸O_c record shows a variation of more than 2‰ (from ca. -8.7‰ to -6.4‰, Fig. 3). From ca. 4500 to 1100 cal a BP the record shows frequent and large fluctuations. After ca. 1100–1200 cal a BP the variability progressively decreases and after ca. 700 cal a BP the values remain close to ca. -7.2‰. Two intervals with, on average, ¹⁸O-depleted values occur between ca. 4500–4100 and ca. 2600–2000 cal a BP. More ¹⁸O-enriched intervals occur between ca. 4100 and 2600 cal a BP and from ca. 2000 cal a BP onward.

The δ¹³C_c time series shows less short-term variability but a more complex structure over the long-term, compared with the δ¹⁸O_c record. The δ¹³C_c values have a range of ca. 2‰ (from -5.1‰ to -3.1‰). Between ca. 4500 and 2900 cal a BP the values are generally higher when compared with the interval between ca. 2900 and 1700 cal a BP. A decreasing trend starting at ca. 1400 cal a BP reaches the lowest δ¹³C_c values at ca. 1200 cal a BP. Following this, there is a long trend of increasing values up to ca. 600 cal a BP followed by relatively invariant values up to the top of the core.

The carbon isotope composition of the organic matter ($\delta^{13}\text{C}_{\text{om}}$) oscillates between ca. -30.5 and -26.5% (Fig. 3). Between ca. 3600 and 2600 cal a BP there is a clear phase characterized by higher (on average) values, followed by an interval of lower values at ca. 2500–2100 cal a BP. A phase of relatively higher values again occurs at 850–300 cal a BP. More prominent lower values instead occur at the base of the record, at ca. 4000, 1300 and 900 cal a BP and at the top of the record.

Pollen Data

Here we discuss only the AP and total pollen concentrations. AP and total concentrations show a relatively high correlation ($r^2 = 0.52$). AP% is mostly greater than 85% up to ca. 900 cal a BP, after which it decreases down to ca. 70% (Fig. 3). The record starts with high AP percentages then progressively decreasing with the first notable reduction at ca. 3100 cal a BP, after which they increase again to the highest values between ca. 1950 and 1800 cal a BP. Following this, a sharp decline occurs, with another prominent reduction at ca. 1450 cal a BP, followed by a recovery before the final decrease. Similar to AP%, total pollen concentrations show a strong decline from ca. 900–1000 cal years BP up to the top of the record. Before this decline, the concentration values show a first phase of increase (ca. 4500–4000 cal a BP), followed by a rapid decline centred at ca. 3900 cal a BP. After a short period of recovery (centred at ca. 3600 cal a BP), there is an interval with lower values between ca. 3500 and 2800 cal a BP, with the lowest values of this interval at ca. 2900 cal a BP. Between ca. 2700 and 1900 cal a BP there is an interval characterized by the highest concentration values, although large fluctuations are observable (e.g. the sharp declines at ca. 2550 and 2100 cal a BP). From ca. 1900 cal a BP there is a long decline of pollen concentration, reaching one of the lowest values at ca. 1450 cal a BP, and after a rapid recovery lasting about 100 years there is a final dramatic decline in total pollen concentration.

Discussion

Oxygen Stable Isotope Interpretation

The possible presence of trace amounts of dolomite suggests clastic contamination from the lake catchment. Significant input of clastic carbonates limit the use of stable isotope data from the bulk fraction as a palaeolimnological proxy (e.g. Leng *et al.*, 2010a). However, we note that: (i) if any, the contamination is relatively minor, as suggested by XRD ($\ll 1\%$ or below the detection limit); (ii) there is no covariance between $\delta^{18}\text{O}_c$ and $\delta^{13}\text{C}_c$, which could be expected if contamination is significant and its input varies through time (Hammarlund and Buchardt, 1996), even if a positive correlation between clastic contaminant and isotope composition it is not always obvious (see, for instance, the data for Lake Pamvotis, discussed by Leng *et al.*, 2010a); (iii) there is a crude negative association (statistically low for the whole record) between $\delta^{13}\text{C}_c$ and carbonate content (Fig. 3) – this would be difficult to attribute to high bedrock carbonate contamination in the absence of a positive correlation of $\delta^{18}\text{O}_c$ and $\delta^{13}\text{C}_c$; (iv) $\delta^{13}\text{C}_c$ and $\delta^{13}\text{C}_{\text{om}}$ show, for most of the record, a positive association (again not statistically significant), suggesting that both organic matter and carbonate originate from within the lake; and (v) the C/N ratio (on average <10) suggests that organic matter has originated mainly from within the lake (e.g. Meyers and Lallier-Vergès, 1999): if a large amount of clastic carbonate was present it should have been washed into the lake accompanied by terrestrial organic matter, the ratio of which would be higher.

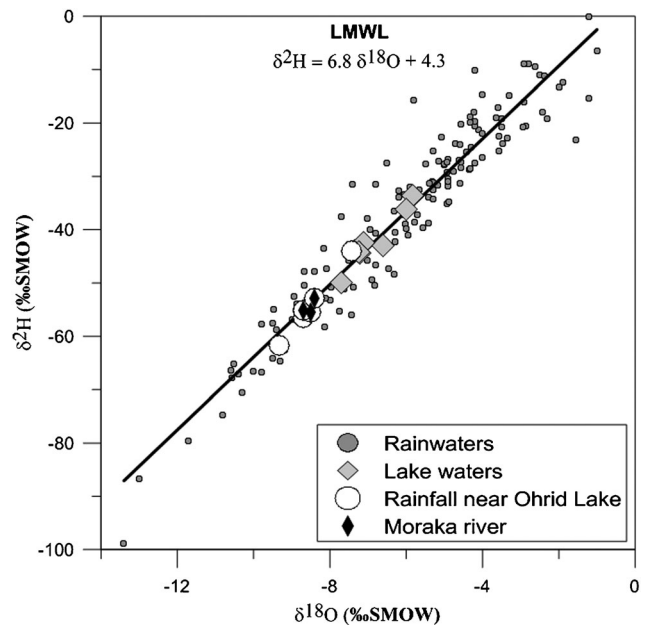


Figure 4. $\delta^2\text{H}$ versus $\delta^{18}\text{O}$ for the area under study. Monthly rainfall data are from <http://ishohis.iaea.org>, Longinelli and Selmo (2003) and Anovsky (2001).

The few water samples collected in the lake plot along the regional meteoric water line (Fig. 4), indicating that lake waters are, if anything, minimally affected by evaporation. This is not completely surprising owing to the very short residence time of the lake water and the presence of submerged springs that undoubtedly dampen the effects of evaporation during the summer months. Preliminary data on diatoms (N. Roberts, unpublished data) indicate that freshwater conditions occurred throughout the investigated period.

The large lake-level variability monitored in the last few decades (Keukelaar *et al.*, 2006) and the relatively large oscillations observed in the $\delta^{18}\text{O}_c$ record suggests that the system is sensitive to hydrological changes, but the isotopic balance is not affected by local evaporation. Thus, the isotopic composition of the lake water is likely to respond mainly to changes in the isotopic composition of the inflow waters, which in turn are a proxy for the isotopic composition of meteoric precipitation in the catchment.

The analyses of rainfall isotopic data from the region show a complex behaviour (Fig. 5), and the data collected by Anovski, (2001) for Lake Ohrid (Fig. 1) can be considered, as a first approximation, representative of the recharge area of the Moraca River (Fig. 1), as shown in Fig. 4. Although the temperature effect and the relationship with the amount of precipitation are not particularly strong, there is a tendency for rainfall with higher isotopic composition during the summer at the time of higher temperatures and lower precipitation; the opposite prevails during winter (Fig. 5). Spring and autumn have intermediate values. Therefore, these figures indicate that lake water could be forced towards lower isotopic values during cool periods, especially during autumn/winter/early spring, when accompanied by higher precipitation: in other words, a low summer-to-non-summer precipitation ratio. By contrast, a shift towards a higher ratio of summer-to-non-summer precipitation, and/or higher summer temperatures, should favour heavy-isotope enrichment in the lake waters. As most of the precipitation in the area occurs during the winter months, increased winter recharge may produce an average decrease of lake water $\delta^{18}\text{O}$, and vice versa. This should be partially affected by the change in the fractionation factor of

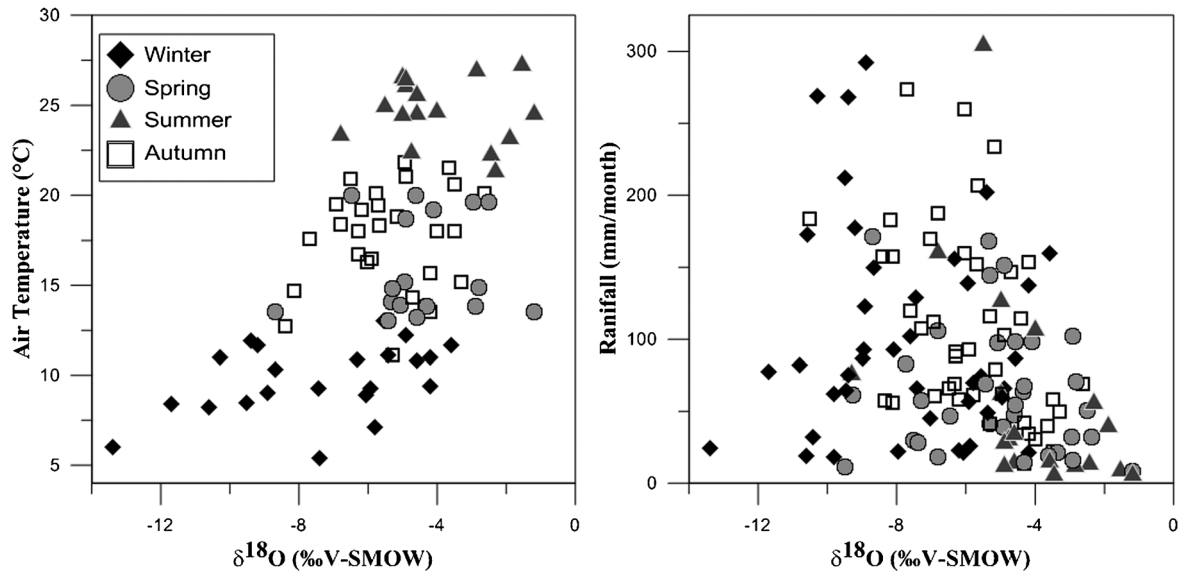


Figure 5. Isotopic composition from different stations for the region showing the dependence of isotopic composition on temperature and precipitation. Data are from <http://ishohis.iaea.org>, Longinelli and Selmo (2003) and Anovsky (2001).

calcite–water with temperature at the time of calcite precipitation (usually during the warmer part of the year, e.g. Leng and Marshall, 2004). Interestingly, many Mediterranean stations show a $\delta^{18}\text{O}_p/T$ of ca. $0.2\text{‰}\text{ }^{\circ}\text{C}^{-1}$ (e.g. Bard *et al.*, 2002; Zoto, 2006), including the data set shown in Figs. 4 and 5 ($0.22\text{‰}\text{ }^{\circ}\text{C}^{-1}$), even if the correlation for these stations is not particularly high ($r^2=0.28$), partly due to the relatively short period of measurements. Because the fractionation factor for carbonate precipitating close to isotopic equilibrium dictates a $\delta^{18}\text{O}_c/T$ gradient of ca. $-0.2\text{‰}\text{ }^{\circ}\text{C}^{-1}$ in the range ca. $10\text{--}30\text{ }^{\circ}\text{C}$ (e.g. Kim and O'Neil, 1997), this indicates that the temperature effect on rainfall and on calcite precipitation could cancel out each other (e.g. Bard *et al.*, 2002). Therefore, the $\delta^{18}\text{O}_c$ record in Lake Shkodra is potentially a near-pure signal of the change in the isotopic composition of the recharge water, given the effect of the short residence time and the presence of recharge by submerged springs. The changes in the isotopic composition of $\delta^{18}\text{O}_c$ should record changes in the lake's seasonal recharge patterns, although this can be complicated by altered storm-track trajectories, which themselves could alter rainfall isotope composition (e.g. Celle-Jeanton *et al.*, 2001).

The most striking feature of the $\delta^{18}\text{O}_c$ record is the long-term trend showing two prominent periods of wetter conditions centred at ca. 4300 and 2100 cal a BP, which are followed rapidly by $\delta^{18}\text{O}_c$ increases indicative of drier conditions. The drier phase between ca. 4100 and 2500 cal a BP is punctuated by several oscillations suggesting unstable hydrological conditions. Particularly prominent are drier conditions occurring at ca. 4100–4000, 3500 and 3300 cal a BP. The following wetter period lasted ca. 500 years and ended abruptly at ca. 2000 cal a BP, and was rapidly followed by a short period at ca. 1850 cal a BP (ca. 100 AD) of drier conditions.

Three evident short phases of wetter conditions occurred between ca. 1800 and 1500 cal a BP (150–450 AD, spanning the Roman Empire), 1350–1250 cal a BP (600–700 AD, at the beginning of the Dark Ages), 1100–800 cal a BP [850–1150, during the Middle Ages – possibly representing the so-called Medieval Climate Anomaly (MCA)] and at ca. 90 cal a BP. Four prominent drier events occur at ca. 1850 cal a BP (ca. 100 AD), 1400 cal a BP (ca. 550 AD), 1150 cal a BP (ca. 800 AD), and a longer period of relatively high $\delta^{18}\text{O}$ values lasting from ca. 780 to 200 years a BP, at ca. 750 cal a BP (ca. 1200 AD). This last period covers most of the Little Ice Age.

Interpretation of carbon isotope composition of organic matter and carbonate, C/N ratio and carbonate content

The $\delta^{13}\text{C}_c$ and carbon isotope composition of organic matter ($\delta^{13}\text{C}_{om}$) are broadly covariant (Fig. 3), supporting the notion that organic and inorganic carbon originated from the same dissolved inorganic carbon pool (DIC; Hollander and McKenzie, 1991). The average difference between $\delta^{13}\text{C}_c$ and $\delta^{13}\text{C}_{om}$ is 24.9 ± 0.8 , in agreement with fractionation from the same DIC source (e.g. Hollander and McKenzie, 1991). The fact that this difference does not vary significantly implies that neither the eutrophic state of the lake nor the level of CO_2 consumption varied significantly (Hollander and McKenzie, 1991; Mayer and Schwark, 1999). The C/N ratio largely matches this interpretation, indicating that lacustrine organic matter prevails. After ca. 900 cal a BP the C/N ratio decreases, suggesting that the lake sediment shows increased proportion of algal matter with a lower C/N ratio compared with that of vascular lacustrine plants, or a concurrent decrease in the terrestrial input, if present (e.g. Meyers and Lallier-Vergès, 1999). The $\delta^{13}\text{C}_c$ and carbonate content, with an apparent negative correlation for some intervals (Fig. 3), may suggest, however, that the amount of primary productivity changed in the lake. This is supported by the TOC data.

Factors influencing the isotopic composition of DIC in karstic lakes are many and varied (Hollander and McKenzie, 1991; Mayer and Schwark, 1999; Leng and Marshall, 2004). Usually, ^{13}C -depleted intervals reflect an increased input of CO_2 derived by oxidation of organic matter whereas higher values suggest increasing equilibration with atmospheric CO_2 and/or hard waters from the catchment (i.e. those strongly affected by dissolution of old carbonates). Rivers normally carry lower $\delta^{13}\text{C}$ DIC values coming primarily from the leaching of soil CO_2 (e.g. Mook and Tan, 1991), which can change in lakes due to a progressive equilibration with atmospheric CO_2 as well as plant photosynthesis, respiration and microbially mediated carbon cycling (e.g. Hollander and Smith, 2001). However, karst springs can carry CO_2 derived from dissolution of old marine carbonate, which is usually more ^{13}C -enriched than 'respired' CO_2 . Intervals characterized by lower $\delta^{13}\text{C}_c$ and $\delta^{13}\text{C}_{om}$ at Shkodra could indicate a persistent input of CO_2 originating from soil CO_2 via river flow, whereas intervals

characterized by higher values may indicate increased equilibration with the atmosphere and more ^{12}C consumption by biological activity and/or relatively more recharge by submerged springs.

Pollen record

Pollen results indicate that the physiognomy of the vegetation was not overly affected by the hydrological changes inferred by the oxygen isotopes, with percentage changes in AP showing rather moderate variability, ranging mainly from 84% to 94% from ca. 4500 to 900 cal a BP. This is not surprising taking into account the orography of the area and the disposition of the altitudinal vegetation belts. Pollen concentration values, by contrast, suggest that important decreases in forest biomass have occurred. However, a general tendency for pollen concentration and AP% to decrease follows the general trend of increasing $\delta^{18}\text{O}$, possibly suggesting progressive drying (Fig. 3).

The first significant pollen concentration minimum occurs at ca. 4000 cal a BP, close to but not exactly matching the oxygen isotope peak, followed by a minimum at ca. 3300 cal a BP in good agreement with the $\delta^{18}\text{O}_\text{c}$ record. A further important minimum occurs at ca. 2900 cal a BP, which does not correlate with the $\delta^{18}\text{O}_\text{c}$ record. At ca. 1400 cal a BP a significant decrease is well matched by a drier phase identified in the $\delta^{18}\text{O}_\text{c}$ record. The start of the prominent decline of both AP% and pollen concentration at ca. 900 cal a BP seems likely to have been generated by phases of deforestation due to human activity starting in the Middle Ages. A first relatively large increase in pollen concentrations matches the arid phase in the $\delta^{18}\text{O}_\text{c}$ record, abruptly followed by the minimum cited above. This suggests that the pollen response lags the isotope response, notwithstanding the different resolution of these two proxies. The maximum at ca. 3600 cal a BP is in agreement with a relatively wetter phase indicated by isotope data.

Perhaps one of the most interesting features is the interval containing the highest concentration values that corresponds to the wetter period identified in the $\delta^{18}\text{O}_\text{c}$ record. The highest values are reached just prior to the abrupt end of this interval. The relative maximum centred at ca. 1300 cal a BP is again close to a wet phase as suggested by the $\delta^{18}\text{O}$.

Regional Significance of the Record

The oxygen isotope record in SK13 is one of the most robust and informative proxies from a palaeohydrological point a view, as the other proxies seem to indicate long-term trends that are difficult to interpret owing to the relatively sparse information on modern lake behaviour. Pollen percentages seem strongly influenced by the orography of the area: any climatic change is probably accommodated by changes in the altitudinal position of the vegetation belts but without wholesale changes in the pollen rain in the lake, even if a progressive decrease in the pollen concentration and AP% since 2000 years a BP (mirroring the progressive increase of the $\delta^{18}\text{O}_\text{c}$ values) may indicate a progressive drying. However, total pollen concentration appears to be more informative than AP%, correlating better with $\delta^{18}\text{O}$. Yet this correlation is not entirely consistent, suggesting that the vegetation may have a threshold of hydrological sensitivity somewhat different to the lake $\delta^{18}\text{O}_\text{c}$.

Overall, the interval between ca. 4200 and 2500 cal a BP is one of higher $\delta^{18}\text{O}_\text{c}$ bracketed by two intervals characterized by lower $\delta^{18}\text{O}_\text{c}$ values. At Lake Malik (Albania), Fouache *et al.* (2010) described a phase between ca. 4000 and 2600 cal a BP of lower lake levels with deposition of peat. A similar pattern

was discussed by Roberts *et al.* (2011b) for stable isotope records in lakes of the eastern Mediterranean, notwithstanding differences in the quality of the various chronologies. In particular, the isotopic record of Lake Gölhisar (Eastwood *et al.*, 2007) shows two humid phases separated by a prominent dry event centred at ca. 3500 cal a BP.

By contrast, the interval between ca. 2500 and 2000 cal a BP with lower $\delta^{18}\text{O}_\text{c}$ values indicates wetter conditions. This interpretation is also supported by the pollen data. We note that during this interval there is an increase in TOC accumulation, which could indicate increased preservation of organic matter. This interval is also coincident with a decrease in $\delta^{13}\text{C}_\text{c}$. Lake level at Lake Malik was higher between 2600 and 2000 cal a BP (Fouache *et al.*, 2010), whilst at Lake Accesa (Tuscany; Magny *et al.*, 2007) level was higher between ca. 2800 and 2000 cal a BP (Fig. 6). Most notably, this period coincides with the highest phase of lake level and amount of precipitation in southern Spain reconstructed in Zoñar Lake (Martín-Puertas *et al.*, 2010). In Fig. 6 the Zoñar Lake Rb/Al ratio is interpreted as a proxy for runoff input to the lake (Martín-Puertas *et al.*, 2010). We also note that this interval corresponds to a phase of lower haematite-stained grain (HSG) deposition in subpolar cores of the North Atlantic (Fig. 6).

At the base of the SK13 record we see evidence for a humid phase centred at ca. 4300 cal a BP and lasting about 200 years. Although not completely captured in its entirety, analogies can be found in other records in the central Mediterranean. A multi-proxy speleothem record from Buca della Renella (Italy) (Drysedale *et al.*, 2006) indicates wetter conditions around this time. Magny *et al.* (2009, and references therein) discuss a wet phase at ca. 4500–4300 cal a BP, including a period of increased floods in Spain, and soil development in Tunisia. At Lake Malik (Albania), sedimentological evidence indicates high lake levels between ca. 4200 and 4100 cal a BP (Fouache *et al.*, 2010).

At ca. 4000–4100 cal a BP there is a prominent arid event. Our chronology is substantially in agreement with that of the Buca della Renella multi-proxy record (Drysedale *et al.*, 2006) and this arid phase has been identified in the aforementioned record of Spanish flood history (Thorndycraft and Benito, 2006) and soil formation in Tunisia (Zielhofer and Faust, 2008). Many pollen records of central Italy probably also capture this event (e.g. Sadori *et al.*, 2004, 2011). According to Di Rita and Magri (2009), this event is consistent with an increase in summer drought with progressive expansion of the North Africa anticyclone zone over the central Mediterranean Basin. Additionally, the Shkodra record may further suggest a decrease of rainfall during the colder months. It is important to note that this event is well recorded outside the Mediterranean at lower and higher latitudes (Drysedale *et al.*, 2006). It is also identified in the HSG record from subpolar cores of the North Atlantic (Bond *et al.*, 2001), even if it does not appear among the most prominent of such events (Fig. 6).

Two further significant phases of drier conditions occur at ca. 3500 and 3300 cal a BP. These events correspond to a peak in the HSG record and the phase of cooling recorded in core LC21 in the Aegean Sea following the deposition of the Thera tephra (Rohling *et al.*, 2002). In the alluvial deposits around the ancient city of Gibala (coastal Syria) Bretschneider and Van Lerbege (2008) found pollen evidence for a severe drought between ca. 3.3 and 2.7 cal ka BP, and Kaniewski *et al.* (2010) have suggested that this arid phase may have produced region-wide crop failure corresponding to the Late Bronze Age collapse.

Four shorter phases of wetter conditions occurred at ca. 1800–1500 cal a BP (150–450 AD, during the Roman Empire), 1350–1250 (600–700 AD, at the beginning of the Dark

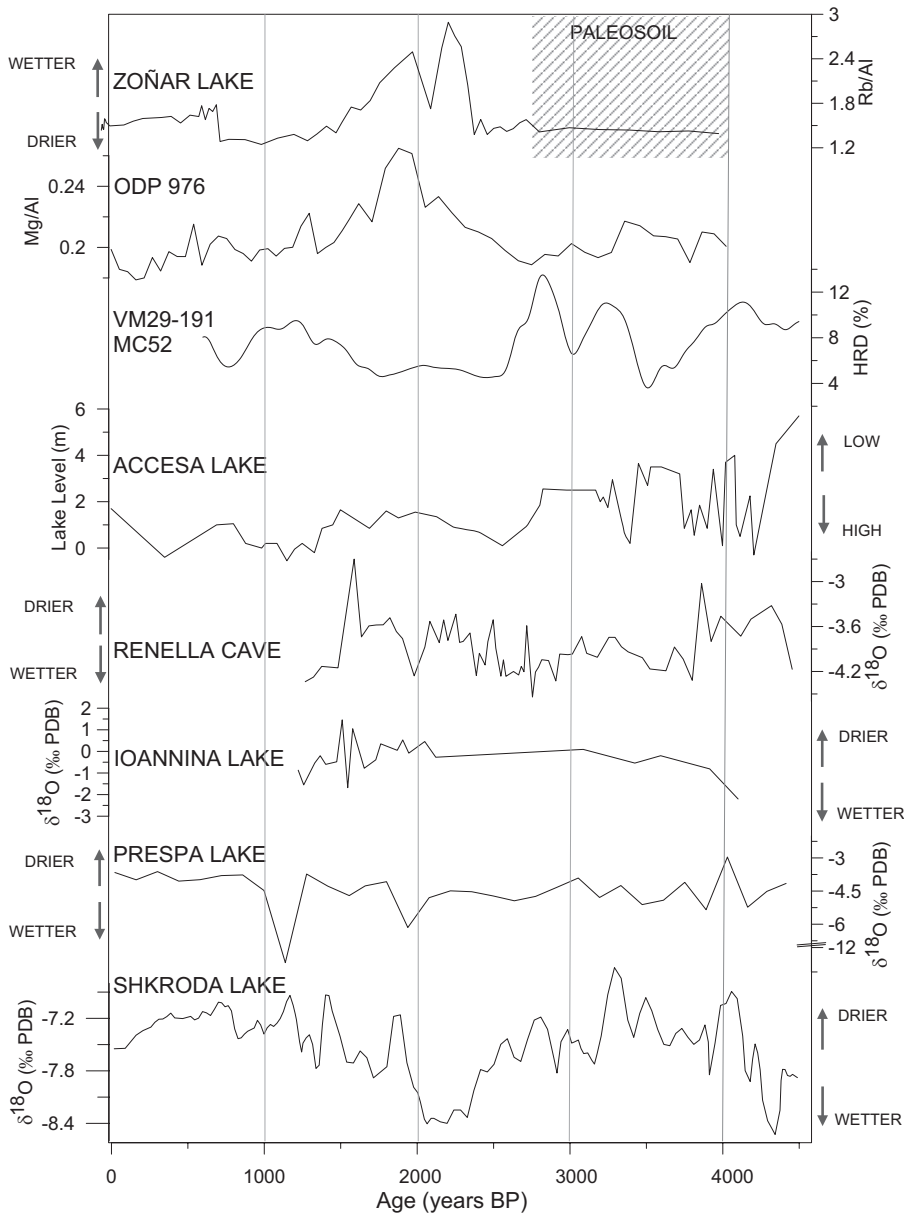


Figure 6. Comparison among different proxies discussed in the text and the $\delta^{18}\text{O}$ record of Lake Shkodra. Zoñar Lake Rb/Al record: Martín-Puertas *et al.* (2010); marine core ODP 976 Mg/Al record: Jimenez-Espejo *et al.* (2008); marine cores VM29-191 and MC52 HSG (%) record: Bond *et al.* (2001); Accessa Lake level record: Magny *et al.* (2007); Renella cave $\delta^{18}\text{O}$ record: Drysdale *et al.* (2006); Ioannina Lake $\delta^{18}\text{O}$ record: Frogley *et al.* (2001); Prespa Lake $\delta^{18}\text{O}$ record on bulk carbonate: Leng *et al.* (2010b); Shkodra $\delta^{18}\text{O}$ record on bulk carbonate fraction (this study). The Shkodra record is three-point averaged compared with Fig. 3.

Ages), 1100–800 (850–1150 AD, during the Middle Ages – possibly representing the MCA – Bradley *et al.*, 2003) and centred at ca. 90 cal a BP, i.e. at ca. 1860 AD. According to the interpretation proposed under our age model, the MCA is represented by a slightly wetter interval. It is interesting to note that this period does not show any prominent feature compared with the last 2000 years of our record. However, the accuracy of our age model is questionable for the last ca. 1000 years and this interval needs to be better resolved in future studies using, for example, other cores which may contain this interval at a higher resolution (van Welden *et al.*, 2008).

Four prominent drier events occur at ca. 1850 (100 AD), 1400 (550 AD), 1150 (800 AD) and ca. 750 cal a BP (1200 AD). The event at ca. 1150 cal a BP is in good agreement (within age errors) with an arid period between ca. 1200 and 1100 cal years BP reported by McMillan *et al.* (2005) from speleothems in two caves of southern France. Still within age error is the prominent arid event that can be inferred from the $\delta^{18}\text{O}$ of bulk carbonate from Lake Ohrid (Leng *et al.*, 2010b). The drier conditions which appear to extend from ca. 780 to 200 cal a BP with the higher oxygen isotope values centred at ca. 750 cal a BP cover a significant part of the Little Ice Age.

An interesting feature of the interval between 2000 and 300 cal a BP is the progressive reduction in magnitude of the $\delta^{18}\text{O}_c$ excursions. This may correspond to a progressive, long-term aridification, supported somewhat by pollen data, even if we cannot rule out other contributing factors: for instance, the residence time of the lake water due to damming of the outlet, or the lake ‘catchment’ effect triggered by a change in land use due to human activity, as suggested by the pollen data for the last 900 years. Additionally, human impacts could have promoted catchment erosion, increasing clastic carbonate input to the lake. van Welden *et al.* (2008), on the basis of grain-size and magnetic data from cores SK-17 and SK-06, reported a substantial increase in clastic input during the Little Ice Age. This may have increased input of clastic carbonate to the lake, dampening the isotopic signature of *in situ*-precipitated CaCO_3 . However, the possible increase in clastic carbonate input during the last 2000 years is not supported by the trend in $\delta^{13}\text{C}_{\text{om}}$, which shows a decrease in the last 2000 years, and the C/N ratios, which suggest an increase of algal organic matter in the last 1500 years, and finally $\%\text{CaCO}_3$, which shows a decrease in the last 1000 years BP. In addition, the $\delta^{13}\text{C}_{\text{om}}$ record reveals that the interval with the lowest values was

between ca. 1500 and 500 cal a BP. Taken together, these patterns are difficult to interpret in terms of increases in the terrestrial input of carbonate into the lake.

Figure 6 also shows $\delta^{18}\text{O}_c$ records from Lakes Prespa (Leng *et al.*, 2010b) and Lake Ioannina (Pamvotis) (Frogley *et al.*, 2001), compared with the Lake Shkodra record. The three records show evident arid/wet/arid oscillations between ca. 2000 and 1000 cal a BP, although differences in the resolution and chronology do not provide sufficient evidence for correlating these oscillations. In addition, the Prespa record shows a clear peak with higher $\delta^{18}\text{O}_c$, which is nearly coincident with the similar event at ca. 4100 cal a BP in Shkodra.

Although a recent synthesis of studies suggests the existence of a regional pattern of hydrological variability (e.g. Roberts *et al.*, 2008, 2011b) it is clear that a fine-scale of resolution and excellent chronological control are necessary for improving our knowledge of climatic changes over the Mediterranean Basin, including the Balkans. However, these correlations may indicate that the Shkodra $\delta^{18}\text{O}$ record is not representative for the Balkans in general because more agreement is found with records outside of the study area (e.g. Gölhisar and Zoñar lakes).

Conclusion

This study has shown that Lake Shkodra can potentially help with reconstruction of the palaeohydrological evolution of the region thanks to the high sedimentation rate, with high authigenic carbonate component, its rapid residence time and low evaporative effect on the water isotopic composition. In particular, the Shkodra $\delta^{18}\text{O}_c$ record indicates the presence of two main wet phases: one centred at ca. 4300 cal a BP, and one between ca. 2500 and 2000 cal a BP. The latter phase is well expressed in southern Spain, Central Italy and, tentatively, in Turkey, and possibly represents a prominent event in the Mediterranean. Four minor wetter intervals have occurred over the last 2000 years: between ca. 1800 and 1500 cal a BP (150–450 AD), 1350–1250 (600–700 AD), 1100–800 (850–1150 AD), and, less so, at ca. 90 cal a BP. Between ca. 4100 and 2500 cal a BP the $\delta^{18}\text{O}_c$ values are relatively high, with three prominent peaks indicating drier conditions at ca. 4100–4000, ca. 3500 and centred at ca. 3300 cal a BP. The event found at Shkodra at 4100–4000 cal a BP confirms this event as a prominent one in Mediterranean records. Four additional drier events were identified at ca. 1850 (100 AD), 1400 (550 AD), 1150 (800 AD) and ca. 750 cal a BP (1200 AD). The MCA may have been wetter, but is not particularly prominent when compared with the last 2000 years. On the other hand, part of the Little Ice Age seems to have been drier, with wetter conditions towards the end, but better resolution and dating for this interval is necessary to substantiate this. Since ca. 900 cal a BP, human impact becomes more apparent in the pollen data, and may also have affected evolution of the lake.

Acknowledgements. Shkodra lake coring was funded by a NATO Science for Peace project (SFP 977 993), which was initiated and managed by F. Jouanne. A.W. and C.B. thank their colleagues who made the coring possible, especially J. L. Mugnier, R. Koci and S. Bushati. This research was also partially supported by INGV-DPC 2005–2007 projects (V-3 subproject on explosive activity of Somma-Vesuvius – Tephra dispersion, Leaders R. Santacroce and G. Zanchetta). We thank M. Guidi and L. Dallai (IGG-CNR) for assistance during the analytical work. C. Martin-Puertas is acknowledged for Zoñar lake data and for useful discussion. We thank M. Magny and an anonymous reviewer for useful suggestions, which greatly improved the quality of the manuscript.

Abbreviations. AP, arboreal pollen; DIC, dissolved inorganic carbon; HSG, haematite-stained grain; MCA, Medieval Climate Anomaly; NAP, non-arboreal pollen; SEM, scanning electron microscopy; TC, total carbon; TN, total nitrogen; TOC, total organic carbon; XRD, X-ray diffraction.

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