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# Climate variability in the northern Levant from highly-resolved Qadisha record (Lebanon) during the Holocene optimum.

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## Abstract

New stalagmites from Qadisha cave (Lebanon) located at 1720 m asl provides a high-resolution and well-dated record for the northern Mount-Lebanon. The stalagmites grew discontinuously from 9.2 to 5.7 and at 3.5 ka, and they show a tendency from a more negative oxygen-isotope signal at ~9.1 ka to a more positive signal at ~5.8 ka. Such a trend reflects a change from a wetter to a drier climate at high-altitudes. The  $\delta^{13}\text{C}$  signal shows rapid shifts throughout the record and a decreasing trend towards more negative values in the mid-Holocene, suggesting enhanced soil activity. On the short-term trend, Qadisha stalagmites record rapid dry/wet changes on centennial scales, with a tendency to more rapid dry events towards the mid-Holocene. Such changes are characterized by an overall good agreement between both geochemical proxies and stalagmite growth, and might be affected by the seasonal variations in snow cover. The Qadisha record is in good agreement with other Levantine records, showing more humid conditions from 9 to 7 ka. After 7 ka, a drier climate seems to affect sites at both low- and high-altitude areas. The Qadisha record reflects uniquely mountainous climate particularities compared to other records, specifically the effect of snow cover and its duration, regulating the effective infiltration.

## Keywords

Qadisha, Holocene, speleothems, geochemistry, snow cover

## 1. Introduction

The impact of future climate change in the Eastern Mediterranean (EM), a region already exposed to severe water stress on agriculture and environment, must be better constrained and may be assessed by investigating past climatic variability (Masson-Delmotte et al., 2013). The climate of the Eastern Mediterranean (EM) is influenced by weather systems originating from the North Atlantic Ocean, passing from Europe to the Mediterranean Sea. In the EM region, many records from lake and marine cores (Rohling et al., 2002; Emeis et al., 2003; Jones and Roberts, 2008; Almogi-Labin et al., 2009; Develle et al., 2010) and speleothems (Bar-Matthews et al., 2003; Verheyden et al., 2008; Cheng et al., 2015) have been used to define past climate conditions. Regional paleoclimate records suggest that the region is sensitive to large-amplitude glacial-interglacial changes and climate fluctuations on millennial to decadal timescales (Bar-Matthews et al., 2003; Almogi-Labin et al., 2009; Bar-Matthews and Ayalon, 2011). However, this sensitivity is unequal among regions because of the heterogeneity of the EM climate (Fig. 1) over short distances (Ulbrich et al., 2012). The precipitation distribution shows a high spatio-temporal variability with most of the effective moisture occurring during winter-spring seasons and being concentrated on mountainous regions. Confidently reconstructing this variability requires a dense network of precisely dated and highly resolved palaeoclimate records. Past spatio-temporal climate variability in the EM is still poorly documented due to unevenly distributed records (Burstyn et al., 2019).

In the Levantine region stretching from southern Turkey, Syria, Lebanon, Palestine, Israel to Cyprus, many studies cover the Holocene period, but few are well-dated (Bar-Matthews et al., 2003; Cheng et al., 2015) or record Rapid Climate Changes (RCC). Some are located along the coast at low altitudes like Jeita or Zalmon cave, other records such as Soreq and West Jerusalem caves are located at mid-altitudes (Western flanks of Judean Plateaus). Some lake or marsh records are located in the rain shadow of the Levantine Mountains (e.g. the Dead Sea, the Ammiq marsh in the Beqaa inner plain, the Ghab depression). Among the records covering the Holocene, a few are located in high mountainous areas, such as Mizpe Shelagim cave in Mount (Mt)-Hermon and Incesu cave in the Taurus mountains (Turkey). In the central Levant, Mt-Lebanon (3088 m) is an imposing range facing the moisture coming from the Mediterranean and creates therefore a peculiar local climate system in Lebanon. None of the records spanning the Holocene were retrieved in areas located at high altitude, except for El-Jurd marsh (Chedadi et Khater, 2016) and Yammouneh lake records, which both provide low-resolution (millennial-scale) data. To analyse the effect of mountainous climate and its variability during the Holocene, a reconstruction of past climate variations from highly resolved archives, such as speleothems is needed. These secondary cave deposits (e.g.

stalagmites) are currently considered as the most suitable terrestrial archives for establishing high resolution proxy time-series in paleoclimate research (Genty et al., 2003; Cheng et al., 2012; Fairchild and Baker, 2012).

We report new stalagmite stable-isotope data ( $\delta^{13}\text{C}$ ;  $\delta^{18}\text{O}$ ) from Qadisha cave (Lebanon) located at 1720 m asl, which provides a high-resolution and well-dated record for the northern Mt-Lebanon covering the time period from 9 to 5 ka. The combined measurements of calcite  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  and trace elements enable us to characterise the regional climate trend vs local mountainous effect on high-altitude records. Isotope measurements of fluid inclusions water ( $\delta\text{D}$ ;  $\delta^{18}\text{O}_w$ ) provide insights into the hydrological cycle and allow an estimation of mineral formation temperatures for the Holocene optimum at the high altitude Qadisha Cave.

## 2. State of the art

During the past decades, several synthesis reports were prepared on paleoclimate studies in the EM (Robinson et al., 2006; Finné et al., 2011; Burstyn et al., 2019), compiling many marine (Rossignol-Strick et al. 1999; Kallel et al. 2000; Emeis et al. 2003; Almogi-Labin et al., 2009) and terrestrial records spanning the Holocene period (Frumkin et al. 2000; Bar-Matthews et al. 2003; Verheyden et al., 2008; Develle et al. 2010, Rowe et al., 2012; Ayalon et al., 2013; Cheng et al., 2015; Gasse et al., 2015; Ünal- İmer et al., 2015; Cheddadi and Khater, 2016; Flohr et al., 2017; Carolin et al., 2019; Sinha et al., 2019; Erkan et al., 2021; Jacobson et al., 2021; Burstyn et al., 2022). Focusing more on the Levantine coast, from the Sinai and Negev desert to the southern flanks of the Taurus Mountains in Turkey, and including Cyprus, studies have revealed a general climatic state of a wet and warm Early Holocene from ~10 to ~6 ka during maximum summer insolation, coeval with the deposition of Sapropel S1. Sapropels are organic-rich layers deposited during periods of increased discharge of the river Nile (Rossignol-Strick et al. 1999; Kallel et al. 2000). This precessional time-scale effect results in isotopically lighter sea surface water due to the contribution of the  $\delta^{18}\text{O}$ -depleted Nile influx into the EM sea basin. This isotopic source effect is expressed in Soreq, Peqiin (Bar-Matthews et al., 2003; Bar-Matthews and Ayalon, 2011, Burstyn et al., 2022) and Jeita cave records (Verheyden et al., 2008; Cheng et al., 2015) with more negative oxygen isotope values following the depletion of sea surface  $\delta^{18}\text{O}$  as resolved from planktonic foraminifera (Grant et al., 2016). In the Southern Taurus mountains, both Dim (Unal-Imer et al., 2015) and Incesu cave records (Erkan et al., 2021) show a trend towards more depleted  $\delta^{18}\text{O}$  from ~10 to ~8 ka. Although the isotopic composition of the EM source is considered to have a primary effect on the  $\delta^{18}\text{O}$  signal of the terrestrial records, the isotopic  $\delta^{18}\text{O}$  depletion is amplified by increased rainfall (Emeis et al., 2003; Almogi-Labin et al., 2009), also leading to more negative  $\delta^{18}\text{O}$  values. This effect has been invoked for the Early Holocene in some speleothem records (Burstyn et al., 2019) and the Yammounh polje record (Develle et al., 2010).

The distribution of rainfall and its changes during the Holocene were not-coeval along the Levantine coast. Regional variations in the rainfall amount cause isotopic variations as attested by cave climate monitoring studies (Bar-Matthews et al., 1997; Ayalon et al., 1998; 2004; Nehme et al., 2019). Cave water isotopic values are biased towards the main infiltration period which is the winter-spring season (Nehme et al., 2019; 2020). The isotopic signal of the infiltration water is transferred into the cave calcite. Rapid seasonal, decadal or centennial isotopic variations in speleothems are therefore more likely related to rainfall amount changes (Bar-Matthews et al., 1997; Orland et al., 2014; Nehme et al., 2019) as cave temperature would not vary strongly on seasonal, annual and in deep, less ventilated caves even on decadal scales. The impact of temperature on the calcite  $\delta^{18}\text{O}$  values is about 0.2‰ per 1°C change (Demény et al., 2010; Tremaine et al., 2011; Daeron et al., 2019), whereas 200 mm change in the annual rainfall amount could cause a change in the  $\delta^{18}\text{O}$  of rainfall, and therefore of the infiltrating water by 1‰, as measured at the Soreq cave site by Bar Matthews et al. (2003).

Develle et al., (2010) and Cheng et al., (2015) found contrasting patterns of climate variability between records from northern and southern Levant. Cheng et al. (2015) emphasize an out-of-phase pattern between records from northern Levant relative to the Dead-Sea Basin (DSB). The contrasting pattern of precipitation is likely due to enhanced warm southerly-south-westerly flow which intensifies spring-winter precipitation over northern Levant (Brayshaw et al., 2011). Furthermore, the contrasting north-south precipitation pattern persists on millennial to centennial timescales, linked potentially to an enhanced (weakened) meridional circulation, which results in a wet (dry) northern (southern) Levant (Xoplaki et al., 2004), expressed by a contrasting RCC between Jeita isotopic variations and DSB levels (Cheng et al., 2015).

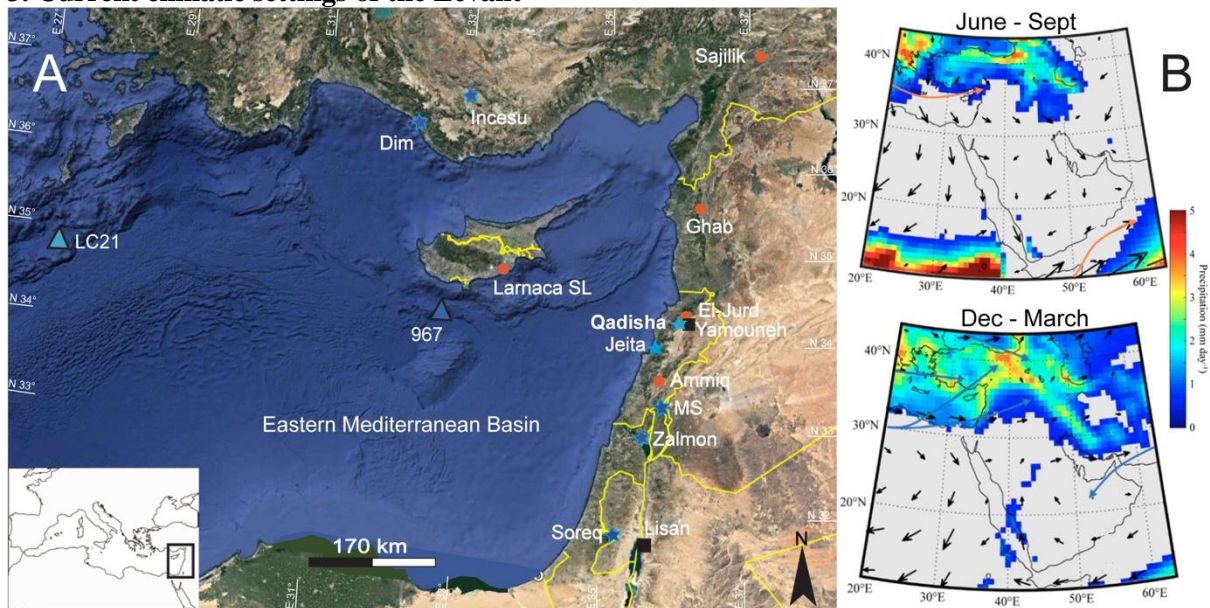
In parallel to changes in meridional circulation patterns, Cheng et al. (2015) emphasized the relevance of distinct effective infiltration (P-E) effects, enhanced by local factors (topography, vegetation) between the northern and the southern Levant. This interpretation, previously proposed by Develle et al. (2010), focusses on effective rainfall, related to precipitation – evaporation (P-E) in both soil and epikarst that affects water supply to caves (infiltration) and lakes (drainage) and thus explains differences in water balances between sites. Indeed, Develle et al., (2010) stressed on the different timing of rapid climate changes in the northern and the southern Levant.

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According to the Jeita cave and Aammiq marsh records (Hajjar et al., 2008), the major shift from humid to general dry conditions occurred around 6 ka, which is different than in the Dead Sea which level was extremely low during the entire Holocene compared to the Late Glacial Maximum (LGM). Migowski et al. (2006) do not exclude those humid conditions relative to modern times prevailed in the DSB during the early-Holocene, but the water balance contrast seen between Yammouneh and DSB records rely on different P-E conditions in both regions. For example, the Yammouneh record (Develle et al., 2010) do not reveal proxy evidence for a drought around 8 ka as observed in Jeita record (Cheng et al., 2015) at lower altitude. More to the South, the 8.2 ka event was pointed out in Soreq record (Bar-Matthews et al., 2003), but was not interpreted as a drought.

Effective infiltration (P-E) also influences proxies related to local soil and epikarst conditions such as  $\delta^{13}\text{C}$  in carbonates. Such proxies, conditioned by vegetation cover and soil microbial productivity, are additionally influenced by  $\text{CO}_2$  degassing and Prior Calcite Precipitation (PCP) related to changing cave drip rates (Fohlmeister et al., 2020). In the EM region, a contrasting pattern of hydroclimate variability between the northern and the southern Levant is exemplified by comparison of the  $\delta^{13}\text{C}$  profiles of Jeita with Peqiin and Soreq records. Although Jeita and Peqiin cave records show similar  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  isotopic trends, a notable heavy excursion in the Soreq  $\delta^{13}\text{C}$  record from ~10 to ~7 ka presents an opposite trend relative to the Jeita  $\delta^{13}\text{C}$  curve. This positive Soreq  $\delta^{13}\text{C}$  event is interpreted to reflect an extremely wet period in the southern Levant partially because of its association with lighter  $\delta^{18}\text{O}$  values (Bar-Matthews and Ayalon, 2011). Cheng et al., (2015) challenge this interpretation and suggest that the Soreq cave record could reflect drier conditions during the Early Holocene, consistent with low Dead Sea levels, The recent publication of Burstyn et al., (2022) follows the interpretation of Bar-Matthews and Ayalon, (2011) and involves a complex opposite response system of magnetic particle influx to rainfall to demonstrate the coupling between the inflow of magnetic particles ( $\text{IRM}_{\text{flux}}$ ) and  $\delta^{13}\text{C}$  in Soreq speleothems. The contrasting climate between northern and southern Levant are therefore still under debate and the factors such as topography in the local P-E response to the synoptic climate systems still not well constrained mainly due to different interpretations of the  $\delta^{13}\text{C}$  response to water availability.

### 3. Current climatic settings of the Levant



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**Figure 1.** A-Location of Qadisha Cave (this study) and other paleoclimatic records spanning the Holocene: Lisan lake (Torfstein et al., 2013a, 2013b), Soreq Cave (Bar-Matthews et al., 2003; Burstyn et al., 2022), Zalmon Cave (Keinan et al., 2019), Peqiin Cave (Bar-Matthews et al., 2003), Mizpe Shelagim (MS) Cave (Ayalon et al., 2013), Ammiq peat record (Hajjar et al., 2008), Jeita Cave (Verheyden et al., 2008; Cheng et al., 2016), Yammouneh Paleolake (Develle et al., 2010), El-Jurd peat record (Cheddadi & Khater, 2016, Ghab core (Zeist & Woldring, 1980; Yasuda et al., 2000), Sagilik peat (Sekeryapan et al., 2020), Incesu Cave (Erkan et al., 2021) Dim Cave (Unal-Imer et al., 2015), Larnaca Salt Lake (Kaniewski et al., 2020), LC21 (Grant et al., 2012), ODP 967 (Emeis et al., 2003; Scrivner et al., 2004). B- Maps showing seasonal precipitation amounts in the wider EM region. Panels at the right illustrate seasonal precipitation amounts from June to September (JJAS) and from December to March (DJFM), respectively. Grey areas indicate regions where the daily precipitation amount is smaller than 0.5 mm. Data retrieved from the ERA-Interim reanalysis dataset (1979 to 2015 CE) (Berrisford et al., 2011)

Today, the Levant region is mainly influenced by the mid-latitude westerlies, which originate from the Atlantic Ocean, forming a series of sub-synoptic low-pressure systems across the Mediterranean Sea (Gat et al., 2003; Ziv

et al., 2010). In winter, cold air plunging south over the relatively warm Mediterranean enhances cyclogenesis, creating the Cyprus Low (Alpert et al., 2005). Moist air is then driven onshore, generating intense orographic rainfall across Mt-Lebanon, Mt-Hermon and the Syrian mountains in the northern Levant. In summer, the westerly belt is shifted to the north, following the northern shift of the North-African subtropical high pressures, and the region experiences hot and dry conditions with more southward winds. In Lebanon, the annual rainfall varies between 700 and 1000 mm along the coastline and more than 1400 mm in higher mountains. The snow coverage reaches in average 5 months from December to April (Shabaan et al., 2015) in basins located at mid-latitudes (1200-2000 m) and up to 7 months from December to June (Fayad and Gascoïn, 2020) in high-altitude basins (>2500 m). As a consequence of the above circulation system, the climate is seasonal with wet winters (November to February) and dry, hot summers (May to October) with a significant influence of snow coverage (up to 75 %) on the water supply budget in karst networks and springs at high-altitudes (Koeninger et al., 2017).

A general West-East gradient in rainfall (amount, isotopic composition) from the Lebanese coastline to the inner Beqaa plain is evident in the published Local Meteoric Waterline (LMWL; Aouad-Rizk et al., 2005) as a consequence of the altitudinal effects related to the Rayleigh distillation processes (Dansgaard, 1964; Rozanski et al., 1993). This gradient is mirrored in the cave stream and dripwater isotopic composition (Nehme et al., 2019) with a clear altitudinal gradient from the coastline to the highest Mt-Lebanon peaks. The North-South Gradient at Mt-Lebanon and to the South is expressed also in the isotopic composition and amount of rainfall. The studies of Aouad-Rizk et al. (2005), Gat et al. (2005), and Saad et al. (2005) showed more positive isotopic rainfall values and a lower rainfall amount towards the south, impacting the water balance (P-E) and effective infiltration in karstic systems along the Levantine coast and mountain chains.

#### 4. Cave site and sample description

Qadisha Cave (34°14'38"N; 36°02'11"E) is located at 1720 m asl in the northern part of Mt-Lebanon (Fig. 1), in the vicinity of the highest peaks (Mt-Makmel) of the mountain chain reaching 3088 m asl. The latter with a North-East - South-West direction faces the East-Mediterranean Basin. The cave develops in Quaternary deposits derived from dolomitised Cretaceous limestone, located in the vicinity of the Qadisha catchment basin (Dubertret, 1975; Nader et al., 2006). The basin is fed by ample snowmelt and has a mean elevation of 2244 m. The duration of snow cover is variable throughout the years and reach 6 to 7 months in the Qadisha water catchment basin (Telesca et al., 2014; Fayad and Gascoïn, 2020). The cave is horizontal with more than 1076 m of explored galleries, comprising an upper relict part and lower active part with a permanent spring (discharge rate up to 1 m<sup>3</sup>/s) (Edgell, 1997). Qadisha cave, partially transformed in a tourist cave since 1934 (first show cave in Lebanon). It is located nearby the cedar forest of Bsharreh, which is believed to have had a wider forest cover in the past. Today, the Quaternary deposits above the cave, consisting mainly of scree and cemented rock debris, are covered with shrub vegetation (Dubertret, 1975). A previous monitoring study in the cave (Nehme et al., 2019) showed a mean cave-air temperature of 9.0 ± 0.5°C and a pCO<sub>2</sub> concentration of 600 ppmv. Water percolation through the cave persists generally throughout the year. Two speleothems Qad-1 and Qad-2, were retrieved from the upper gallery of the cave, which hosts many active stalagmites.

#### 5. Methods

##### 5.1. <sup>230</sup>Th dating

The chronology of Qad-1 and Qad-2 were established by 10 and 9 <sup>230</sup>Th dates, respectively (Table 1). Exploratory <sup>230</sup>Th dating was performed first at Xi'an Jiaotong University (China) in 2011 and 2014 and the rest was completed at the University of Minnesota (USA) in 2018, by using Thermo-Finnigan Neptune/Neptune plus multi-collector inductively coupled plasma mass spectrometers. The methods are identical in both laboratories (Cheng et al., 2013). Standard chemistry procedures (Edwards et al., 1987) were used to separate uranium and thorium. A triple-spike (<sup>229</sup>Th-<sup>233</sup>U-<sup>236</sup>U) isotope dilution method was used to correct instrumental fractionation and to determine U/Th isotopic ratios and concentrations (Cheng et al., 2013). U and Th isotopes were measured on a MassCom multiplier behind the retarding potential quadrupole in the peak-jumping mode using standard procedures (Cheng et al., 2013). Uncertainties in U/Th isotopic measurements were calculated offline at 2σ, including corrections for blanks, multiplier dark noise, abundance sensitivity, and contents of the same nuclides in spike solution. Corrected <sup>230</sup>Th ages assume an initial <sup>230</sup>Th/<sup>232</sup>Th atomic ratio of 4.4 ± 2.2 × 10<sup>-6</sup>, the values for a material at secular equilibrium with a bulk earth <sup>232</sup>Th/<sup>238</sup>U value of 3.8 (Cheng et al., 2013).

**Table 1.** <sup>230</sup>Th dating results of stalagmites Qad-stm1 and Qad-stm2. The error is given as 2se. U decay constants:  $\lambda^{238} = 1.55125 \times 10^{-10}$  (Jaffey et al., 1971) and  $\lambda^{234} = 2.82206 \times 10^{-6}$  (Cheng et al., 2013). Th decay constant:  $\lambda^{230} = 9.1705 \times 10^{-6}$  (Cheng et al., 2013). \* $d^{234}U = ([^{234}U/^{238}U] \text{ activity} - 1) \times 1000$ . \*\*  $d^{234}U$  initial was calculated based on <sup>230</sup>Th age (T), i.e.,  $d^{234}U_{\text{initial}} = d^{234}U_{\text{measured}} \times e^{234\lambda T}$ . Corrected <sup>230</sup>Th ages assume the initial <sup>230</sup>Th/<sup>232</sup>Th atomic ratio of 4.4 ± 2.2 × 10<sup>-6</sup>. Those are the values for a material at secular equilibrium, with the bulk earth <sup>232</sup>Th/<sup>238</sup>U value of 3.8. The errors are arbitrarily assumed to be 50%. \*\*\*CE stands for "Contemporaneous Era". Ages are defined as the year before 1950 A.D. \*age considered as outlier

Sample Number	depth (mm)	<sup>238</sup> U (ppb)	<sup>232</sup> Th (ppt)	<sup>230</sup> Th / <sup>232</sup> Th (atomic x10 <sup>-6</sup> )	$\delta^{234}\text{U}^*$ (measured)	<sup>230</sup> Th / <sup>238</sup> U (activity)	<sup>230</sup> Th Age (a) (uncorrected)	<sup>230</sup> Th Age (a) (corrected)	$\delta^{234}\text{U}_{\text{initial}}^{**}$ (corrected)	230Th Age (yr CE)*** (corrected)
Qad-stm1-0	2	435,0 ±1,0	3591 ±72	85 ±2	353,7 ±2,2	0,0426 ±0,0002	3848 ±19	3307 ±127	357 ±2	3247 ±127
Qad-stm1-8	7	606,5 ±0,6	132 ±3	5739 ±140	442,4 ±1,5	0,0756 ±0,0003	5853 ±28	5848 ±28	450 ±1	5780 ±28
Qad-stm1-7	26	575,7 ±0,9	97 ±3	7502 ±240	444,3 ±2,2	0,0764 ±0,0004	5914 ±30	5911 ±30	452 ±2	5843 ±30
Qad-stm1-6	46	615,3 ±1,0	122 ±3	6432 ±183	450,6 ±2,0	0,0774 ±0,0004	5966 ±29	5962 ±29	458 ±2	5894 ±29
Qad-stm1-5	65	665,4 ±0,7	361 ±8	2329 ±50	445,7 ±1,7	0,0767 ±0,0003	5929 ±24	5918 ±25	453 ±2	5850 ±25
Qad-stm1-4	77	535,7 ±1,0	466 ±10	1472 ±31	437,3 ±2,7	0,0777 ±0,0004	6045 ±33	6028 ±35	445 ±3	5960 ±35
Qad-stm1-3	96	572,6 ±0,6	352 ±7	2126 ±46	432,1 ±1,5	0,0792 ±0,0003	6185 ±28	6173 ±29	440 ±2	6105 ±29
Qad-stm1-2	101	546,2 ±0,6	66 ±2	11419 ±389	441,9 ±1,8	0,0840 ±0,0003	6528 ±25	6525 ±25	450 ±2	6457 ±25
Qad-stm1-1	115	643,7 ±1,1	1156 ±23	792 ±16	433,5 ±2,2	0,0863 ±0,0003	6748 ±28	6711 ±38	442 ±2	6643 ±38
Qad-stm1-00*	128	639,0 ±2,0	978 ±20	902 ±18	428,1 ±2,6	0,0838 ±0,0003	6573 ±24	6542 ±32	436 ±3	6482 ±32
Qad-Stm2-5	6	558,6 ±1,5	194 ±4	45 ±3	303,3 ±2,1	0,0009 ±0,0001	79 ±5	71 ±7	303 ±2	5 ±7
Qad-stm2-4	13	297,6 ±0,3	193 ±4	2309 ±51	397,4 ±1,7	0,0907 ±0,0004	7294 ±36	7281 ±37	406 ±2	7213 ±37
Qad-stm2-3	25	621,7 ±0,6	316 ±6	3161 ±65	406,2 ±1,4	0,0973 ±0,0002	7794 ±22	7783 ±23	415 ±1	7715 ±23
Quad 2-8.2	28.2	351,8 ±0,3	255 ±6	2284 ±50	423,4 ±1,6	0,1003 ±0,0004	7937 ±38	7922 ±39	433 ±2	7851 ±39
Qad-stm2-2	30	336,4 ±0,5	87 ±2	6980 ±178	457,5 ±2,1	0,1093 ±0,0004	8466 ±32	8461 ±33	469 ±2	8393 ±33
Qad-stm2-8.6	36	367,1 ±0,3	284 ±6	2422 ±52	483,2 ±1,6	0,1138 ±0,0004	8667 ±35	8652 ±36	495 ±2	8581 ±36
Qad-stm2-8	49	499,2 ±0,7	62 ±2	15661 ±580	481,2 ±2,2	0,1186 ±0,0004	9062 ±35	9060 ±35	494 ±2	8992 ±35
Qad-stm2-7	67	582,4 ±0,8	188 ±4	6121 ±144	484,8 ±2,1	0,1200 ±0,0004	9153 ±34	9147 ±34	497 ±2	9079 ±34
Qad-stm2-6	80	566,7 ±0,8	379 ±8	2970 ±63	489,1 ±2,1	0,1206 ±0,0004	9167 ±34	9154 ±35	502 ±2	9086 ±35
Qad-Stm2-1	106	545,4 ±0,9	207 ±4	5220 ±106	472,7 ±1,4	0,1199 ±0,0002	9218 ±20	9211 ±21	485 ±1	9145 ±21

## 5.2. Calcite and water stable-isotopes measurements

Samples for stable isotopic analyses were taken along the growth axis of Qad-1 and Qad-2 stalagmites (Fig 2) for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  measurements. Overall, 230 samples were measured in both stalagmites. Samples were drilled along the stalagmite growth axis at 2.5 mm resolution using a Merchantek Micromill mounted on a Leica microscope, with a 0.3 mm resolution for specific parts. Between every sample, the drill bit and sampling surface were cleaned with compressed air. The samples were analysed using a Nu Carb carbonate device coupled to a Nu Perspective mass spectrometer (MS) at the Vrije Universiteit Brussel, Belgium. Parts of the samples were measured at the Laboratory for Environmental and Raw Material Analysis (LERA) at Karlsruhe institute of Technology (KIT), Germany, using a Thermo Gasbench II connected to a DELTA V IRMS in continuous flow mode. All  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  values are calibrated against Vienna Pee-Dee Belemnite (V-PDB) and are reported in permil (‰). Analytical uncertainties were better than 0.1 ‰ (1 $\sigma$ ) for oxygen and 0.05 ‰ (1 $\sigma$ ) for carbon on both instruments. Percolation and stream waters as well as recent calcite samples underneath active drip water were collected previously from Qadisha cave in 2011 and 2014 (Nehme et al., 2019).

## 5.3. Trace element data

Elemental abundances were determined by laser ablation (LA)-ICP-MS, at the Institute for Geosciences, Johannes Gutenberg University Mainz, Germany, using an ESI NWR193 ArF excimer LA system equipped with the TwoVol<sup>2</sup> ablation cell, operating at 193 nm wavelength, coupled to an Agilent 7500ce quadrupole ICP-MS. Ablation was performed in line scan mode and surfaces were pre-ablated prior to each line scan to prevent potential surface contamination. Line scans were performed at a scan speed of 10  $\mu\text{m/s}$ , using a spot size of 110  $\mu\text{m}$  and a laser repetition rate of 10 Hz. Laser energy on the samples was about 3 J/cm<sup>2</sup>. Measured ion intensities were monitored in time-resolved mode and background intensities were measured for 15s. Synthetic glass NIST SRM 612 was used to calibrate element concentrations applying the preferred values given in the GeoReM database (; Jochum et al., 2005; 2012). Quality control materials (QCMs) (USGS MACS-3 and USGS BCR-2G) were used to monitor the accuracy and precision of the LA-ICP-MS analysis and calibration strategy. Raw data were processed using TERMITE (Mischel et al., 2017), an R script for data reduction. <sup>43</sup>Ca was used as an internal standard and applied at a sample Ca concentration of 390,000  $\mu\text{g/g}$  and the values reported in the GeoReM data base for the QCMs. Element concentrations determined for the QCMs had a precision of <0.02% (1 $\sigma$ ).

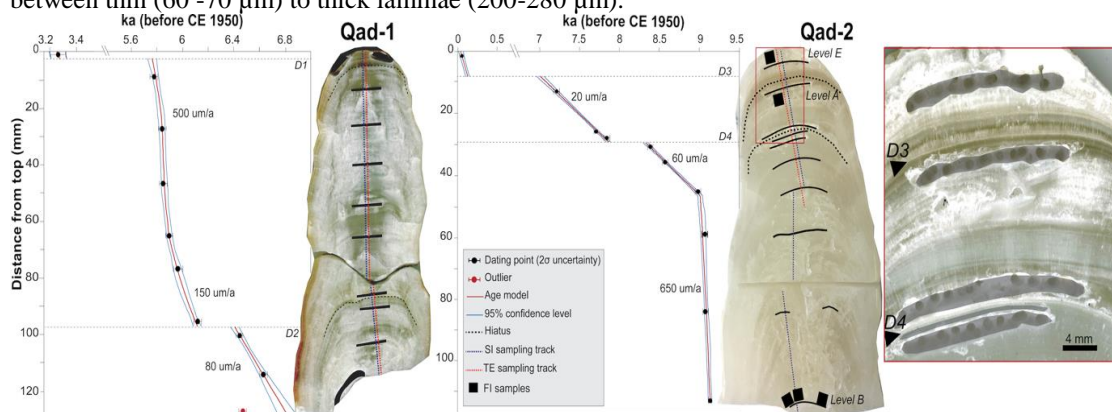
## 5.4. Fluid inclusions stable-isotope (H-O) analyses

Five calcite samples, from three levels in Qad-2 stalagmite were measured using a custom-built extraction line connected to a Picarro L2130i analyzer using cavity ring down spectroscopy (WSCRDS). This technique allows to simultaneously measure hydrogen and oxygen isotopes for minute water amounts released from calcite. The extraction line follows the design of Affolter et al. (2014) and is described in detail in Weissbach et al. (2022). In brief, the calcite samples were hydraulically crushed, and the released fluid inclusion water instantly vaporized in the heated extraction system and transferred to the analyser. Reference water injections with known  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values were used for calibration and quality control. Glass capillaries ( $\mu\text{l}$  size) were used for high-precision water amount calibration and isotopic control. The precision of fluid inclusion water analyses is dependent on the released water amount and is 0.5 ‰ for  $\delta^{18}\text{O}$  and 1.2 ‰ for  $\delta^2\text{H}$ , if fluid water amounts are >0.2  $\mu\text{l}$  and reaches 0.1-0.3 ‰ for  $\delta^{18}\text{O}$  and 0.2-0.7 ‰ for  $\delta^2\text{H}$  for fluid water amounts >1  $\mu\text{l}$  (Weissbach et al., 2022).

## 6. Results

### 6.1. Petrography and chronology

270 Both samples of 11 and 13 cm each display along their growth axis a translucent to whitish calcite with a columnar  
 271 fabric observed over most sections. Both stalagmites are highly laminated. With the aid of a camera mounted on  
 272 a Leica Microscope, images of laminae were taken at several depths and the thickness of each was measured.  
 273 Layer thickness of laminae varied between 60  $\mu\text{m}$  to 280  $\mu\text{m}$ . The laminae growth is irregular and alternates  
 274 between thin (60 -70  $\mu\text{m}$ ) to thick laminae (200-280  $\mu\text{m}$ ).



275 **Figure 2.** Age model of both Qadisha-1 (Qad-1) and Qadisha-2 (Qad-2) stalagmites using StalAge (Scholz and Hofmann,  
 276 2011). Sampling track for stable-isotopes, trace elements and location of fluid Inclusions (squares) along the growth axis  
 277 are shown on each stalagmite. Red rectangle in the upper part of Qad-2 stalagmite shows a high-resolution image of the  
 278 discontinuities D3 and D4 along the growth axis.  
 279

280  
 281 A total of 19 Uranium/Thorium ages were obtained from stalagmites Qad-1 and Qad-2 (Table 1). The age  
 282 distribution based on the StalAge model (Scholz and Hofmann, 2011) indicates that the stalagmite Qad-1 grew  
 283 from ca.  $6.643 \pm 0.038$  to  $3.247 \pm 0.127$  ka including two discontinuities (Fig. 2). The first discontinuity D1 ranges  
 284 from 3.247 to 5.787 ka (extrapolated ages) and D2 covers a shorter period from 6.108 to 6.408 ka (extrapolated  
 285 ages). Qad-2 grew from  $9.145 \pm 0.021$  to  $0.005 \pm 0.007$  ka including two discontinuities: D3 (0.005 to 7.016 ka)  
 286 and D4 (7.844 to 8.326 ka). The age model of Qad-1 was constructed with 8 ages, and one age (Qad-stm1-00)  
 287 considered as an outlier. The latter is sampled on the edge of the growth axis and is not in stratigraphical order  
 288 with the other dating points. At the base of Qad-1, the growth rate is around 60  $\mu\text{m}/\text{a}$  evolving to a higher rate  
 289 around 150  $\mu\text{m}/\text{a}$  and up to 500  $\mu\text{m}/\text{a}$  in the upper part of the stalagmite. For the Qad-2 stalagmite, all the ages  
 290 are in stratigraphical order and were used in the calculation of the age model. The growth rate at the base of Qad-2  
 291 is very high, reaching 650  $\mu\text{m}/\text{a}$  and is reduced to 60 and 20  $\mu\text{m}/\text{a}$  in the middle and upper part of the stalagmite.  
 292

## 293 6.2. Stable isotopic composition of calcite, modern water and fluid inclusions

294 Calcite  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  values were analysed at a multi-annual to decadal resolution. The  $\delta^{18}\text{O}$  values for Qadisha  
 295 record (Fig. 3) range from  $-5.7$  ‰ to  $-7.5$  ‰, with a mean of  $-6.8$  ‰. The  $\delta^{13}\text{C}$  values range from  $-6.2$  ‰ to  
 296  $-8.6$  ‰ around a mean of  $-7.5$  ‰. Both  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  values generally co-vary during the entire period from  $\sim 9.1$   
 297 to  $\sim 5.7$  ka but are distinct in the medium and long-term trends. For example, the  $\delta^{18}\text{O}$  values show an overall trend  
 298 towards more positive values from early to the Mid-Holocene, unlike the  $\delta^{13}\text{C}$  values that show a trend towards  
 299 more negative values when reaching the Mid-Holocene. Significant variations are noticeable in the stable-isotope  
 300 curves with clear  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  excursions around  $\sim 9.1$ ,  $\sim 8.9$ ,  $\sim 7.7$ ,  $\sim 7.4$ - $7.2$ ,  $\sim 6.5$  and  $\sim 5.9$  ka.

301 Stable-isotopes of Qad-2 calcite dated at  $0.005 \pm 0.007$  ka (calcite age between 1938 and 1952 CE) show an  
 302 average  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  value of  $-7.2$  ‰ and  $-6.6$  ‰ respectively. On a comparative basis, present cave and spring  
 303 water sampled in 2011 and 2014 in Qadisha Cave (Nehme et al., 2019) show average values of  $-8.6$  ‰ for  $\delta^{18}\text{O}_w$   
 304 and  $-46.9$  ‰ for  $\delta^2\text{H}_w$ .  
 305

306 **Table 2.** Qadisha fluid inclusions samples in chronological order (old to young). Stable-isotope measurements are shown in  
 307  $1\sigma$  error. Measurements with an (\*) are considered outliers due to a low water amount of less than 0.5  $\mu\text{l}/\text{g}$ . Apparent  
 308 formation temperature was determined using the calcite-water fractionation factors after Kim and O'Neil. (1997).

Qad_2 samples	Age of level [ka CE 1950]	Sample code	$\delta\text{H}^2$ [permil]	$\delta^{18}\text{O}$ [permil]	Volume [ $\mu\text{l}$ ]	Mass [g]	Water amount [ $\mu\text{l}/\text{g}$ ]	$T_f$ [ $^{\circ}\text{C}$ ]
Level B	$\sim 9.0$ - $9.1$	BQ_2_d	-41.70	-5.81	0.29	0.58	0.50	$18.7 \pm 2.5$
Level B	$\sim 9.0$ - $9.1$	BQ_2_b	-43.20	-7.65	0.73	0.78	0.94	$10.3 \pm 2.5$
Level B*	$\sim 9.0$ - $9.1$	BQ_2_a	-39.58	-5.53	0.23	0.72	0.32	$20.1 \pm 2.5$
Level A	$\sim 7.2$ - $7.3$	AQ_1	-42.82	-7.04	0.39	0.46	0.84	$12.2 \pm 2.3$
Level E*	$\sim 0.05$	EQ_1	-19.46	-6.88	0.01	0.45	0.02	$11.2 \pm 2.4$

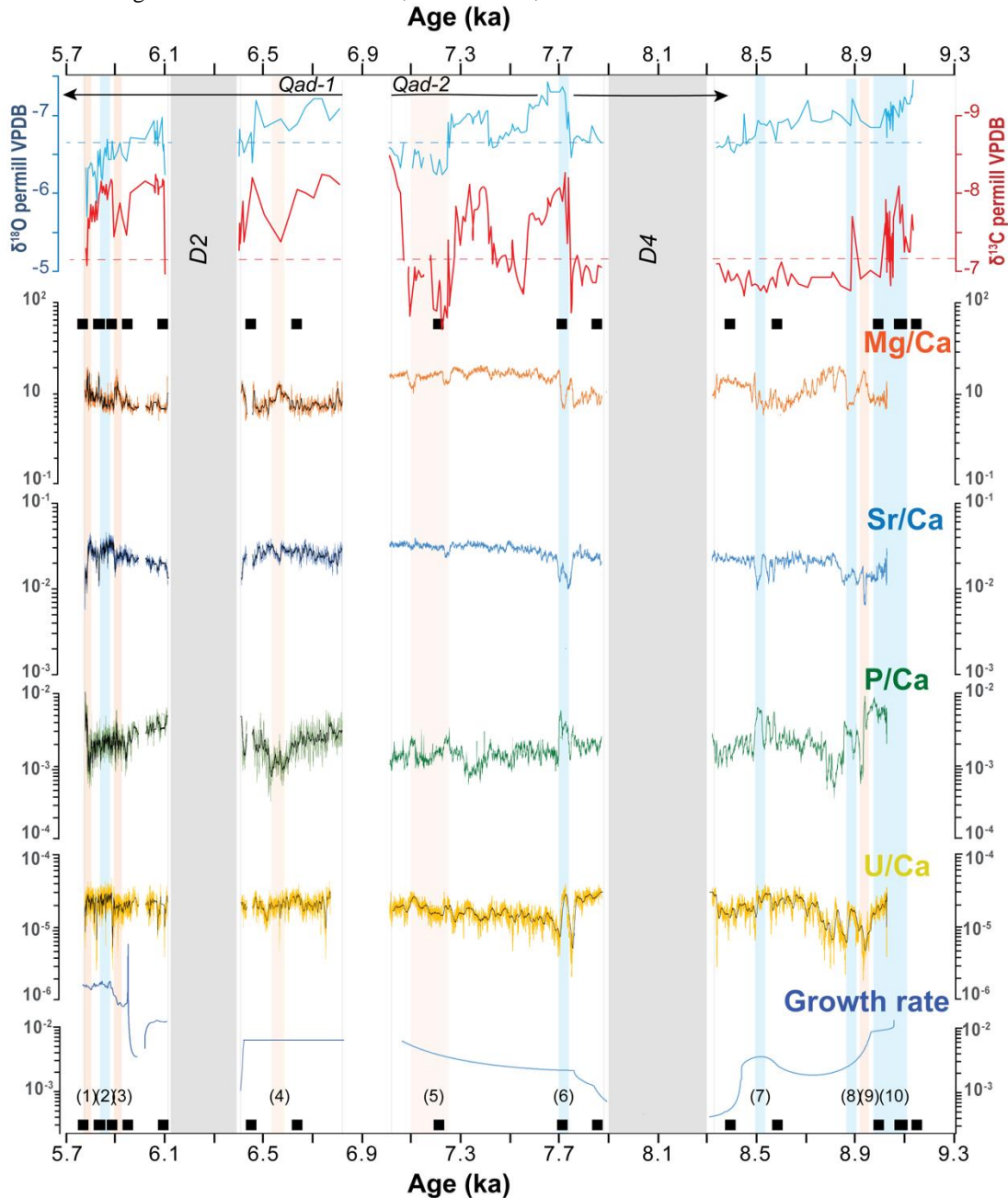
309



310 Average fluid inclusions  $\delta^{18}\text{O}_w$  and  $\delta^2\text{H}_w$  values of Qad-2 are -6.5 ‰ and -41.8 ‰, respectively. Samples taken  
 311 at approximately the same level B in the stalagmite (~9.0-9.1 ka) show a certain variability of 3.6 ‰ for  $\delta^2\text{H}$  and  
 312 2.6 ‰ for  $\delta^{18}\text{O}$ , between samples. The sample with the highest water yield shows the most negative  $\delta^{18}\text{O}$  value.  
 313 At ~7.2-7.3 ka,  $\delta^{18}\text{O}_w$  is -7.0 ‰ and  $\delta^2\text{H}_w$  is -42.8 ‰ (Table 2). Level E spanning a time period of the last century  
 314 yielded a very low amount of water and was therefore rejected.  
 315

### 316 6.3. High resolution trace element data

317 Main trace element ratios presented in Fig. 3 are all above background levels. The overall sampling resolution is  
 318 annual to multi-annual. The average elementary ratios show a slight shift between Qad-1 and Qad-2 with in  
 319 general a higher variability for Qad-1. Mg/Ca ratios show a few significant shifts that are mirrored in most other  
 320 investigated ratios (Fig. 3). Particularly strong trace elemental signals are visible around ~9.0-8.9 and ~7.7 ka,  
 321 whereas weaker signals are noticeable at ~8.5, ~7.3 & ~7.1, ~6.6 and ~5.8 ka.



322 **Figure 3:**  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  profiles of both Qad-1 and Qad-2 stalagmites with the trace elements curves (Mg/Ca, Sr/Sa, P/Ca,  
 323 U/Ca) and a moving average curve (in black) for some of the trace elements. Growth rate is displayed in logarithmic scale.  
 324 Both stable isotopes and trace elements data are plotted against time (yr CE 1950), modelled using StalAge. Black dots refer  
 325 to the dating points and the grey shading to the identified discontinuities (D2, D4). Blue rectangles highlight significant  
 326 variations towards wetter conditions in both stable isotopes and trace element data and orange rectangles highlight drier  
 327

328 conditions. Dashed lines crossing  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  profiles indicate present-day values. Rapid dry and wet events are numbered  
329 from 1 to 10.

## 330 7. Discussion

### 331 7.1. Age growth behavior of the studied speleothems

332 Both stalagmites grew during the Holocene, roughly between 9.2 and 5.7 ka, one succeeding the other and  
333 suggesting favorable (humid and warm) conditions for stalagmite deposition at the high altitude of Qadisha cave.  
334 Although growth is relatively constant over most parts of the stalagmites, several hiatuses are observed. Former  
335 studies on stalagmites from Mt-Lebanon (Verheyden et al., 2008; Nehme et al., 2015; 2018) suggest that cave  
336 calcite precipitation and thus growth in the central Levant is conditioned by effective infiltration (Cheng et al.,  
337 2015). The growth stops may be site-specific but could also reflect unfavorable conditions for calcite deposition,  
338 such as dry conditions with low effective infiltration within the epikarst or scarcer vegetation with less bio-  
339 vegetation activity. Based on the number of currently analyzed speleothems, one cannot make the distinction  
340 between one of the options.  
341

342  
343 Growth rates of both stalagmites are of the same order, reflecting similar drip rates and cave  $p\text{CO}_2$ . Similarities in  
344 the petrography of both speleothems corroborate consistent growth conditions. High growth rates of 0.15-0.6  
345 mm/a between 5.8 and 6.1 ka and of 0.3-0.9 mm/a between 9.0 and 9.15 ka allow for annual and higher resolution  
346 climate reconstruction. Growth rates of 15-50  $\mu\text{m}/\text{a}$  for most other parts of the record allows at least for multi-  
347 annual resolution using traditional micro-milling technique for stable isotope analysis. Two growth stops at 8.3-  
348 7.8 ka and 6.4-6.1 ka are supported by two clear petrographical discontinuities. The growth stops between 8.3 and  
349 7.8 ka in Qad-2 stalagmite, although supported petrographically by a thin dust layer, is not supported by changes  
350 in other proxies prior or after the hiatus. If regional aridification characterized the 8.2 ka event, as inferred from  
351 the lowland Jeita Cave record (Cheng et al., 2015), then a combination of low infiltration and scarcer vegetation  
352 within the Qadisha karst basin would make speleothem growth highly unfavorable, plausibly leading to a  
353 centennial-scale hiatus. Such a hiatus is observable in Qad-2, but cannot be attributed confidently to climatic  
354 change.  
355

356 Unlike the growth stop dated between 8.3 and 7.8 ka in Qad-2, the one dated between 6.5 and 6.1 ka in Qad-1  
357 seems to be clearly framed by changes in stable-isotope values and trace elements. More positive  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$   
358 values suggest drier conditions prior and directly after the growth stop and may record unfavorable growth  
359 conditions, although prior calcite precipitation related to local factors at the drip rate may occur and other  
360 speleothems need to be studied to confirm our hypothesis. This seems also in agreement with a change towards  
361 increased Mg/Ca values, and decreased Sr/Ca values just prior and after the hiatus.  
362

### 363 7.2. Interpreting the changes in geochemical proxies from Qadisha Cave

#### 364 a- Speleothem $\delta^{18}\text{O}$ & $\delta^{13}\text{C}$

365 In the Levant, it is now widely established that the  $\delta^{18}\text{O}$  signal is interpreted as related to effective recharge in the  
366 epikarst (precipitation amount vs evapotranspiration) and therefore infer to water balance (P-E) in the epikarst  
367 (Bar-Matthews et al., 2003; Verheyden et al., 2008; Cheng et al., 2015; Nehme et al., 2020). Higher infiltration  
368 (more positive P-E) is related to pronounced low-pressure systems with significant rainfall. Higher rainfall  
369 amounts are known to cause more negative rainfall  $\delta^{18}\text{O}$  values (Bar-Matthews et al., 2003; Aouad et al., 2004)  
370 and reduced annual rainfall (with concomitant reduced P-E) to cause higher rainfall  $\delta^{18}\text{O}$  values.  
371

372 At mid-latitudes,  $\delta^{13}\text{C}$  and growth rate in speleothems are related to biological  $\text{CO}_2$  production, which is dependent  
373 on soil and vegetation conditions in the catchment (Genty et al., 2001a; 2001b). Warmer and wetter periods usually  
374 enhance the production of biogenic,  $\delta^{13}\text{C}$ -depleted  $\text{CO}_2$  and increases the growth rate (Genty et al., 2001a; 2001b).  
375 Cold/dry conditions reduce the vegetation cover and the biogenic  $\text{CO}_2$  supply. Lower/higher calcite  $\delta^{13}\text{C}$  values  
376 and faster/slower growth rates are therefore usually indicative of soil development/disruptions and can be related  
377 to warmer-wetter/colder-drier conditions. An additional control on the  $\delta^{13}\text{C}$  is exerted by the hydrological state of  
378 the aquifer. Partial dewatering of the drip-feeding system induces longer residence times and prior calcite  
379 precipitation, leading to higher  $\delta^{13}\text{C}$  during drier periods (Fairchild and Baker, 2012).  
380

381 The stable isotopic  $\delta^{18}\text{O}$  values of both Qadisha stalagmites display a general trend from 8.5 to 5 ka towards more  
382 positive  $\delta^{18}\text{O}$  values. This general  $\delta^{18}\text{O}$  trend comprises three independent segments that include several rapid  
383 shifts towards negative values at  $\sim 9.1$  ka (10),  $\sim 8.9$  (8),  $\sim 8.5$  (7)  $\sim 7.7$  (6) ka and 5.8 (2) ka (Fig. 3). Such negative  
384 'peaks' are interpreted as wet intervals with high effective recharge (high growth intervals of the stalagmites) and  
385 a positive water-balance (P-E) (Cheng et al., 2015). Rapid changes towards more positive  $\delta^{18}\text{O}$  at  $\sim 9.0$ (9),  $\sim 7.1$ -  
386  $7.2$ (5),  $\sim 6.6$ (4),  $\sim 5.9$ (3) and  $\sim 5.7$ (1) ka, are particularly well expressed and chronologically constrained. Some of  
387 these periods interpreted here as 'drying' events, particularly at  $\sim 9.0$ (9), are characterized unusually by high

388 growth rates. Such peculiar periods seem to indicate at least locally, a wet period as confirmed by negative (lower)  
389  $\delta^{13}\text{C}$  values, prior to the short dry period of ~80 years, but with still high growth rates. Albeit with low resolved  
390 growth rates compared to the resolution of geochemical proxies (annual for trace elements and decadal for the  
391  $\delta^{13}\text{C}/\delta^{18}\text{O}$ ), such peculiar drying events preceded by short wet events reflect a rapid decadal variability, inferring  
392 to a shift in the distribution of effective infiltration.

393

### 394 **b- Trace elements**

395 In general, the (trace) element concentrations of Mg, Sr, P, and U in calcite are controlled by hydrological  
396 processes and less by cave temperature (Roberts et al., 1998). Speleothem Sr/Ca ratios constitute a sensitive proxy  
397 for infiltration changes in the epikarst, with higher values being recorded during times of reduced effective  
398 moisture availability (Fairchild and Treble, 2009). Other mechanisms can explain Sr mobility within the epikarst  
399 and include changes in weathering rates, aerosol input, soil activity that can induce high-frequency variability on  
400 short timescales (decadal to annual) (Verheyden et al., 2000; Fairchild et al., 2006; Sinclair et al., 2012; Baker et  
401 al., 2021). For Mg/Ca ratios in speleothems, it is common to attribute an increase as a response to prior calcite  
402 precipitation (PCP) during times periods of low effective infiltration (Verheyden et al., 2000; Tooth and Fairchild,  
403 2003; McDermott et al., 2004; Fairchild et al., 2006). Since Sr/Ca ratios are similarly influenced by PCP, Mg and  
404 Sr ratios are often positively correlated.

405

406 An exception to this behavior is given in the presence of dolomitized limestone in the karst aquifer, which is the  
407 case in the Qadisha basin, where processes of incongruent calcite dissolution and/or differentiated soil input may  
408 play a role (Roberts et al., 1998; Hellstrom and McCulloch, 2000; Huang et al., 2001; Sinclair et al., 2012,  
409 Jamieson et al., 2016).

410

411 Phosphorous and U are less commonly studied but increasingly used as proxies in the speleothems archive. This  
412 element is generally mobilized from soils during early autumnal/winter storms. High P/Ca in speleothems reflect  
413 maximum soil infiltration during heavy rainfalls at the start of wet seasons (Borsato et al., 2007). The P/Ca ratio  
414 can be complemented by other palaeo-hydrological proxies such as Ba/Ca or U/Ca. U is generally leached from  
415 the bedrock during high rainfall events and results in high U/Ca into the calcite. (Ayalon et al., 1999; Treble,  
416 2009)

417

418 In the Qadisha speleothems, the trace element concentrations vary at maximum by two orders of magnitude. Wet  
419 and dry episodes identified by stable-isotopes and growth rate changes (section 7.2.a, Fig. 3) can overall be linked  
420 to changes in the trace element concentration, with generally high Mg/Ca, and low Sr/Ca, P/Ca and U/Ca  
421 corresponding to higher stable-isotope values, suggesting an overall common driver, although with different  
422 sensitivities between proxies. Therefore, we assign in the Qadisha speleothems the periods with high  $\delta^{13}\text{C}/\delta^{18}\text{O}$   
423 and Mg (and generally low Sr, P and U) to dry periods, with a lower water recharge in the epikarst, and the  
424 opposite changes to wet periods. At 9.0 ka and 7.7 ka, significant changes in  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values, as well as in  
425 U and P concentrations may suggest the occurrence of important flushing episodes.

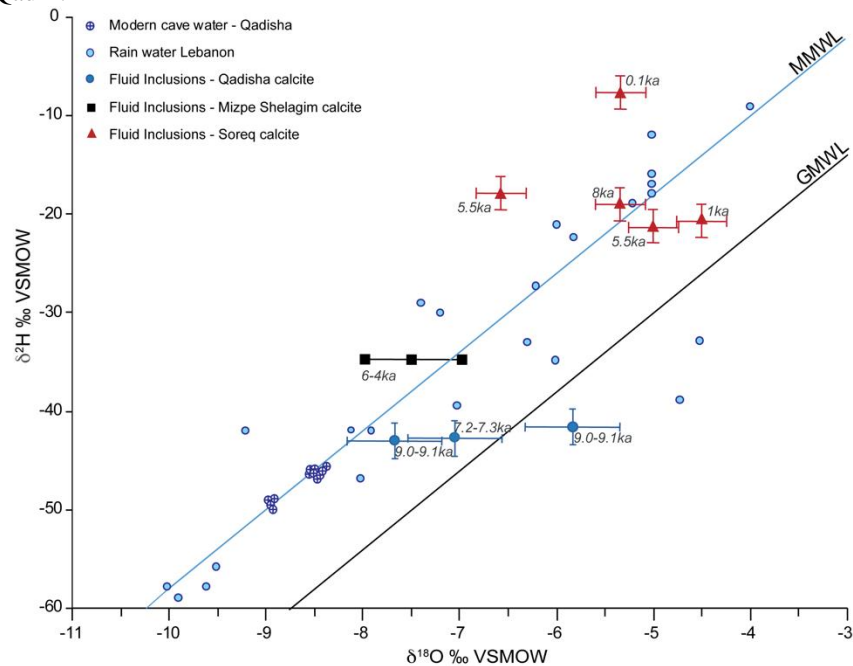
426

### 427 **7.3. Infiltration water isotopes based on fluid inclusion analysis**

428 Isotope measurements of fluid inclusions water ( $\delta\text{D}$ ;  $\delta^{18}\text{O}_w$ ) can provide insights into the hydrological cycle and  
429 allow estimation of mineral formation temperatures. Three successful fluid inclusion analyses from Qad-2  
430 stalagmite with water amount  $>0.5\ \mu\text{l/g}$  were retained here (Table 2).  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values of water sampled in  
431 pools, cave streams and drip water inside Qadisha cave vary between -8.4 and -9.0, and between -45.7 ‰ and -49.5  
432 ‰, respectively. The d-excess of the cave water is between 21.4 and 22.6 ‰ (Nehme et al., 2019). As the cave  
433 water represents an average of the annual precipitation, rainfall values in contrast show a higher variability from  
434 -12.6 to +1.2 ‰ and -86 to +11.6 ‰ for  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values, respectively (Aouad et al., 2004). d-excess values  
435 are between 2 and 27 ‰ with an average close to that of the cave water (Aouad et al., 2004), indicating a generally  
436 higher d-excess for Lebanon compared to the GMWL. Although FI values fall within the modern-day range of  
437 rain water, compared to modern-day drip water from Qadisha, they show important variability. Especially, the  
438 samples from the B-level (9.0-9.1 ka) raise questions about the lateral coherency of the isotopic composition of  
439 FI water. The variability between different levels (B versus E) may be related to differences in isotopic  
440 composition of the rain and therefore the fluid inclusions. Overall, the results are more positive than today's  
441 isotopic composition of drip waters. Following our above interpretation, these values dated within the interval of  
442 7.2-7.3 ka and 9.0-9.1 ka, represent isotopic values of water precipitated close to dry events (5) and (9) identified  
443 above based on their more positive calcite  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values (Fig. 4) and may suggest short-term drier  
444 conditions in the generally wetter Early- to Mid-Holocene. A higher fraction of summer rainfall contribution to  
445 the water budget with a higher isotope value compared to winter snow with its lower isotopic composition (Aouad  
446 et al., 2004) may be another possibility to explain this isotopic observation. In Soreq Cave, Early-to Mid-Holocene  
447 fluid inclusion isotope values are comparable or slightly more negative compared to modern-day values

448 (Matthews et al., 2021). This contrasting effect could be related to Sapropel S1 that has a significant effect on the  
 449 isotope values in the southern Levant (Bar-Matthews et al., 2003) but is not particularly expressed in the northern  
 450 Levant (e.g. the Jeita record; Cheng et al., 2015) where rainfall amounts are generally higher. In addition, snowfall  
 451 is of minor importance for the infiltration budget at Soreq cave whereas it is today the dominant water source in  
 452 the region around Qadisha cave.

453 The fluid inclusion isotope values of various Early- to Mid-Holocene speleothems along the Levant become more  
 454 negative from South (Soreq Cave) to the North (Qadisha Cave), in agreement with a dominating altitudinal effect  
 455 and in addition by an increase in average rainfall towards the Lebanon mountains (Nehme et al., 2019). The  
 456 difference in drip water  $\delta^{18}\text{O}$  values between Soreq and Qadisha cave is  $\sim 4\text{‰}$ , which is close to the expected  
 457 difference from altitude alone. The elevation difference is about 1400 m which explains an isotopic shift of 2.8-  
 458 4.2 ‰ at a lapse rate of 0.2-0.3 ‰/100 m (Clark and Fritz, 1997). The difference between measured fluid inclusion  
 459 isotope values of Holocene samples from the Soreq and Qadisha cave is similar. Although hardly resolvable with  
 460 the current set of fluid inclusion data, rainfall amount could also play a role. For example, Mizpe Shelagim is  
 461 situated almost at the same elevation as Qadisha cave, but about 120 km to the South and shows a more positive  
 462  $\delta^2\text{H}$  value than Qad-2.



463 **Figure 4.** Graph showing the fluid inclusion stable-isotope of Qadisha stalagmite (this study) in comparison with FI from  
 464 Mizpe Shelagim (Ayalon et al., 2013) and Soreq record (Matthews et al., 2021), all plotted against the Global (Rozanski et  
 465 al., 1993) and Mediterranean meteoric waterlines (Gat et al., 2003). Modern rain water of Lebanon (Aouad et al., 2005; Saad  
 466 et al., 2005) are plotted in blue circles respectively. Modern cave water of Qadisha cave is plotted in crossed rectangles and  
 467 circles respectively.  
 468  
 469

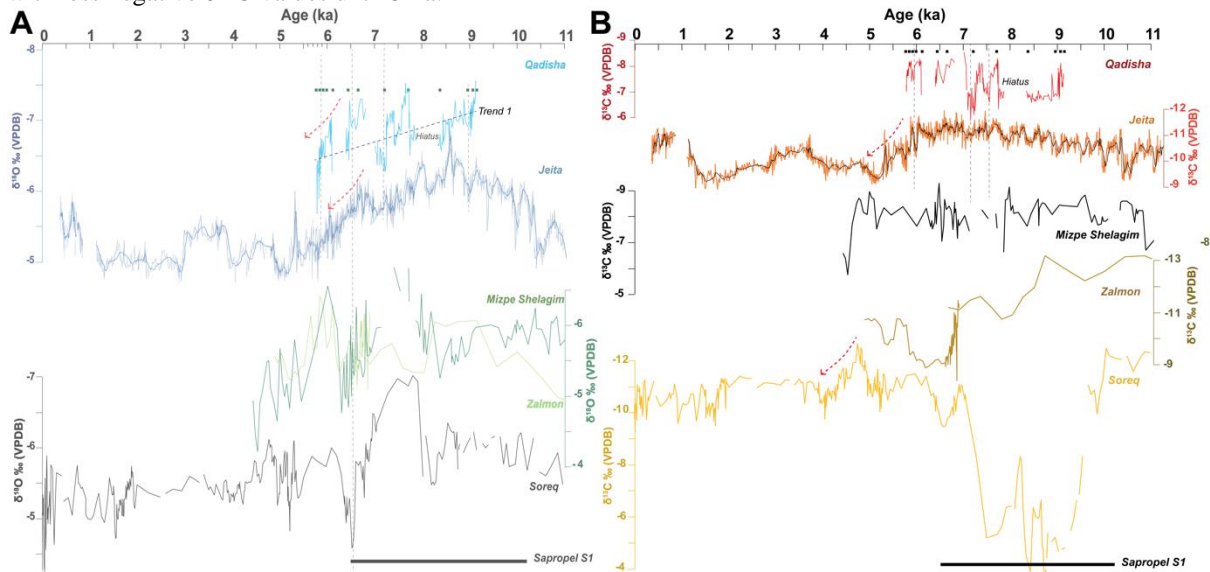
#### 470 7.4. The climatic conditions during the Holocene optimum

##### 471 a- Global and regional drivers of climate variability during the Holocene Optimum

472 The Qadisha stalagmites grew from 9.2 to 5.7 ka, albeit with a discontinuous trend, and show a general tendency  
 473 from more negative  $\delta^{18}\text{O}$  values during the Early Holocene towards more positive  $\delta^{18}\text{O}$  values at the end of the  
 474 Holocene optimum. This trend reflects a change from a wetter (more positive P-E) towards a drier climate  
 475 (reduced P-E) in this high-altitude area. This trend is also noticeable at low altitude, for example in the Jeita  $\delta^{18}\text{O}$   
 476 record, and in other speleothem records from the Southern Levant, e.g., Mizpe Shelagim and Soreq Cave (Fig. 5A).  
 477 The carbon signal in Qadisha indicates values reaching  $-8\text{‰}$  during wet peaks and shifts to  $-6\text{‰}$  during dry events  
 478 (Fig. 5B), reflecting a higher soil and vegetation activity during humid periods. The  $\delta^{13}\text{C}$  curve overprints the  
 479 short-term changes with a slow long-term increasing trend (Fig. 5B) towards more negative values when reaching  
 480 the Mid-Holocene, suggesting overall improved bio-pedological soil activity in the Makmel mountains from 9 to  
 481 6 ka.  
 482  
 483

484 The two investigated Qadisha stalagmites started to grow during the period of maximum summer insolation  
 485 (Berger and Loutre, 1991) and a high SST in the Eastern Mediterranean, reaching  $\sim 18^\circ\text{C}$  during the Holocene  
 486 optimum (Fig. 6) (Emeis et al., 2003; Scrivner et al., 2004). From 10 to 6.5 ka, the deposition of Sapropel 1 in the  
 487 Eastern Mediterranean occurred as a result of increased discharge of the river Nile, leading to isotopically lighter

488 sea surface water. The contribution of the Mediterranean source effect as the only factor in the  $\delta^{18}\text{O}$ -depleted  
 489 signals in terrestrial records has been debated in the last decades. Recent studies on Yammouneh lake (Develle et  
 490 al., 2010), Soreq (Grant et al., 2016) and Zalmon caves (Keinan et al., 2019) show a contribution of rainfall amount  
 491 in changes of the  $\delta^{18}\text{O}$  signal throughout Glacial-Interglacial cycles and even during the sapropel events, by  
 492 extracting the source signal (the  $\delta^{18}\text{O}_{\text{g.rubber}}$ ) from the  $\delta^{18}\text{O}_{\text{calcite}}$  of speleothems or  $\delta^{18}\text{O}_{\text{ostrac}}$ . Signal of lake ostracods.  
 493 An increased precipitation over Mt-Lebanon during the Sapropel event is noticeable with depleted  $\delta^{18}\text{O}_{\text{lake}}$   
 494 (corrected from the source) signal reaching  $-11\%$  in Yammouneh lake, from 9.2 to 8.3 ka. Likewise, the  
 495 uncorrected  $\delta^{18}\text{O}_{\text{ostrac}}$  signal of the lowland Sagilik II peat record (Sekeryapan et al., 2020) in the northern part of  
 496 the Levant, albeit a low-resolved curve, reach the most depleted ostracod  $\delta^{18}\text{O}$  values between 9 and 7.7 ka. The  
 497 well-dated lowland Jeita Cave record in central Lebanon show the most depleted values for the whole record  
 498 around 8.5 ka, followed by a sharp  $\delta^{18}\text{O}$  drop (less negative values) from 8.5 to 8 ka and then a general decrease  
 499 with less negative  $\delta^{18}\text{O}$  values until 5 ka.



500  
 501 **Figure 5.** Plots of the Qadisha stable-isotope curve in comparison with other speleothem records in the Levant. A. Carbon  
 502 isotope curves. B. Oxygen isotope curves. Black dots represent U-Th dating points in the Qadisha record.  
 503

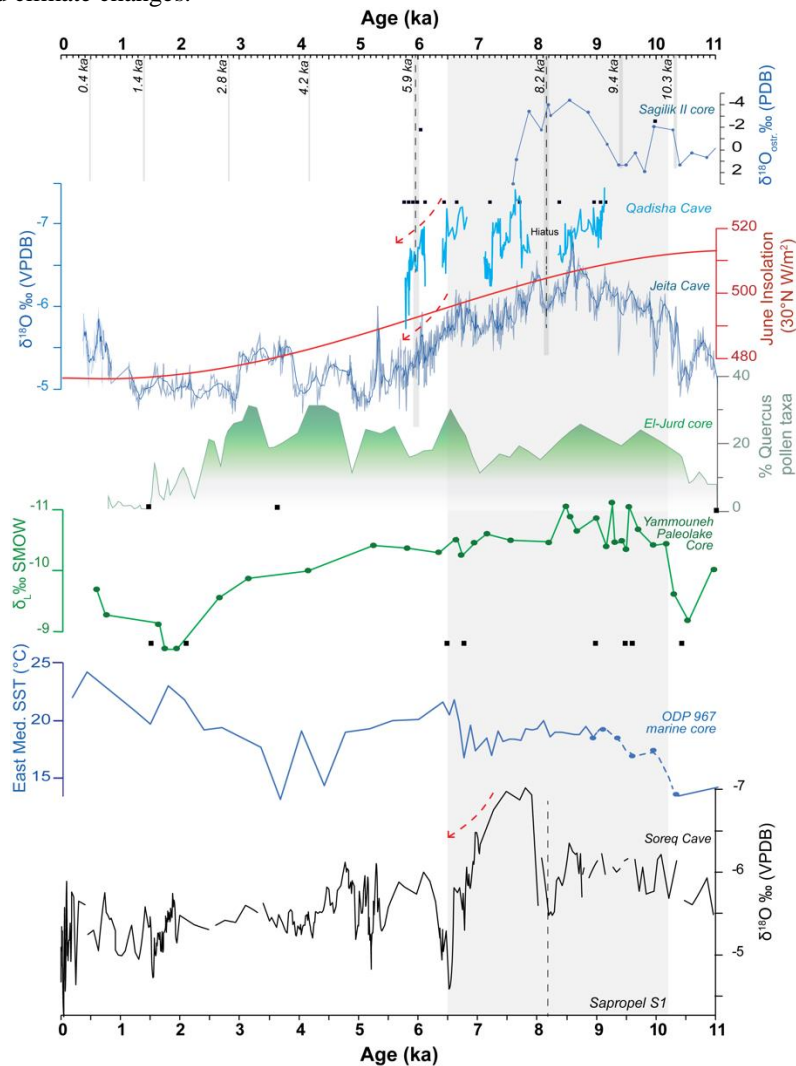
504 More locally in the Makmel mountains, the Quercus pollen taxa in Al-Jurd peat core (Cheddadi and Khater, 2016),  
 505 located 23 km North of Qadisha cave, show a relatively high pollen taxa from 10 to 8 ka, followed by a reduced  
 506 Quercus pollen percentage from 8 to 6.5 ka, and marked later by an increase of Quercus pollen % at 6.5, 5.5, 4.5  
 507 and 3 ka (Fig. 6). Albeit a low-resolved curve, the pollen record of Al-Jurd peat core seems to show a higher  
 508 percentage of Quercus pollen during the Mid-Holocene (6.5 to 3 ka) rather than in the Early-Holocene period  
 509 from 9 to 6 as indicated by several speleothems. The Quercus Forest extension after 6.5 ka is rather puzzling in  
 510 the local paleoclimate scheme of (speleothems) the Makmel mountains, but could suggest wetter/warmer  
 511 conditions with improved bio-pedogenic activity in soils coeval with a reduced snow season and/or decreased  
 512 snow cover enabling the extension of the forest. During the Early Holocene, slightly colder/dryer conditions (than  
 513 the mid-Holocene) might have prevailed with the persistence of longer snow cover period, impacting the length  
 514 of the plant growing season but enhancing speleothem growth with an important recharge from the snow melt  
 515 during summer. Soil erosion is enhanced during short melt seasons on the long-term trend in the Makmel  
 516 mountains, and would explain the reduced Quercus Forest in the area. In similar alpine context such as in Rio  
 517 Martino cave, Piedmont Alps, Italy (Regiaterri et al., 2019), it was suggested that an increase in the persistence  
 518 of snow cover may also impact the  $\delta^{13}\text{C}$  by reducing the length of the plant growing season, leading to reduced  
 519 biogenic  $\text{CO}_2$  supply and therefore to less negative  $\delta^{13}\text{C}$  signal in speleothems. Above the Qadisha site, the Cedar  
 520 Forest stretching over same altitude than in El-Jurd site, may have had similar reduction/extension dynamics  
 521 during the Early/Mid-Holocene and thus leading in general from  $\delta^{13}\text{C}$  signal (avg:  $-7\%$ ) in Qadisha speleothems  
 522 in the Early-Holocene to a slightly more negative  $\delta^{13}\text{C}$  signal (avg:  $-8\%$ ) in the Mid-Holocene.  
 523

### 524 **b- Centennial-scale climate variability**

525 Rapid climatic changes of centennial scale are recorded in some of the highly-resolved stalagmites along the  
 526 Levantine coast. Cheng et al., (2015) detected nine cold/dry events in the Jeita record during the Holocene coeval  
 527 with those in the North Atlantic sediments (Bond et al., 2001). The Bond events in the Jeita record are interpreted  
 528 as dry events inferred from both  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  variations (Cheng et al., 2015) (Fig. 5 and 6). Some of the events,  
 529 such as the  $\sim 5.1$  ka, noticeable in the Jeita record, is also recorded as a dry event in Anatolian Lake records  
 530 (Roberts et al., 2011). The 8.2 ka, recorded in the Jeita stalagmite, corresponds to a  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  excursion (Fig.

531 4) in Soreq cave (Bar-Matthews et al., 2003). Such event is however not noticeable in other speleothem records  
 532 along the Levantine coast, nor in Yammouneh lake (Develle et al., 2010). In Qadisha cave, the 8.2 ka cold event  
 533 is not expressed or may be related to the growth stop.  
 534

535 Other events, at ~8.9-9.0 ka, ~7.1 and ~5.9 ka, (Fig. 4) in the Jeita record are also visible in the Qadisha record  
 536 with more positive stable-isotope values (dry events), but with less clear changes in the related trace element  
 537 pattern. In contrast, rapid climate changes interpreted as wet events in Qadisha Cave, such as the most prominent  
 538 7.7 ka wet period is not noticeable in the Jeita record but corresponds to the major peak in the Soreq record (Fig.  
 539 5A). The inconsistency in recording rapid climate change events between Levantine records may be due to  
 540 different proxy resolution (e.g., smoothing in case of too low resolution) or differences in the local archive  
 541 sensitivity on rapid climate changes.



542  
 543 **Figure 6.** Comparison of the Qadisha record with regional records in the EM region. From top to bottom: Sagilik II  
 544  $\delta^{18}O_{\text{Ostracods}}$ , Turkey (Sekeryapan et al., 2020), Qadisha  $\delta^{18}O_c$  record, Lebanon (this study) Jeita  $\delta^{18}O_c$  record, Lebanon (Cheng  
 545 et al., 2015), the Summer Insolation curve (Berger and Loutre, 1991), Quercus Pollen taxa % from El-Jurd Core (Cheddadi  
 546 & Khater, 2016),  $\delta^{18}O_l$  Yammouneh paleolake record (Develle et al. 2010), East Mediterranean SST, ODP 967 (Emeis et al.,  
 547 2003; Scrivner et al., 2004), Soreq  $\delta^{18}O_c$  record (Bar-Matthews et al., 2003)  
 548

549 **c- Altitudinal and snow cover effect on the significance of climate signals**

550 In the Levant, most of the published speleothem records are located at low altitudes facing the Mediterranean Sea,  
 551 except for Incesu and Mizpe Shelagim records located at 1615 m and 2180 m respectively. Only at Mizpe  
 552 Shelagim a plausible link between the effect of snow cover at high altitudes and speleothem growth was  
 553 established on Glacial-Interglacial timescales. During the Holocene interglacial, the snow cover in mountain areas  
 554 participates actively in the epikarst water-budget, feed drip sites, and favor speleothem growth in caves located at  
 555 high altitudes, as well as affect the vegetation dynamics. Whereas temperature remains close to 0°C during winter  
 556 over large areas of Mt-Lebanon (Fayad and Gascoïn, 2020), the soil beneath the snow does not freeze under

557 present winter conditions, except during exceptional cold events. Comparing the Qadisha records, with those  
558 located at low altitudes, helps in deciphering altitudinal trend and local particularities.  
559

560 The Qadisha stalagmites show more negative  $\delta^{18}\text{O}$  values (Fig. 5A) than those recorded by the Jeita stalagmite  
561 related to the altitudinal effect expressed in the rain and cave water isotopic signals (Nehme et al., 2019). Also,  
562 the contribution of the snow to the infiltration and the percolation water in caves (Aouad-Rizk et al., 2005) may  
563 lead to more negative drip water and calcite  $\delta^{18}\text{O}$  values. Noteworthy is that the Qadisha stalagmites display a  
564 larger amplitude in the  $\delta^{18}\text{O}$  changes and therefore have a more dynamic response to changes than the Jeita record.  
565 This behavior may be related to the higher altitude and the enhanced convective rainout at mountain ranges as  
566 well as to the generally lower  $\delta^{18}\text{O}$  values of snow. An increase in low pressure systems can disproportionately  
567 increase the rainfall/snow amount at high-altitude mountain regions. Qadisha records can therefore indicate better,  
568 or at least more clearly rapid climate changes than the low-elevation Jeita speleothems. In contrast, short-term dry  
569 conditions may be better recorded by the lower-elevation archive sites as they are more sensitive to droughts and  
570 PCP which enhances the isotope and element signal. An example is the drought event recorded in Soreq, Mizpe  
571 Shelagim and Zalmon Cave around 6.5 ka which is well expressed in these caves, but less well expressed in  
572 Qadisha Cave.  
573

574 More to the South, Zalmon and Soreq cave records indicate changes in the  $\delta^{13}\text{C}$  values of more than 4‰  
575 throughout the Holocene (Fig. 5B). This drastic shift in the  $\delta^{13}\text{C}$  signal of Zalmon cave is mirrored by the  $\delta^{13}\text{C}$   
576 signals in other records, with a trend towards less negative  $\delta^{13}\text{C}$  values by the end of the Holocene optimum.  
577 Although marked by a high  $\delta^{13}\text{C}$  shift, the vegetation in northern Galilee indicates a reduced bio-pedological  
578 activity after 8.5 ka, whereas the soil activity seems to be less variable throughout the Holocene optimum in Jeita  
579 coastal site until 6 ka. In the case of Qadisha Cave, the carbon isotope signal reflects a higher sensitivity of the  
580 vegetation on humidity changes with step-changes (< 2‰), similar to Mizpe Shelagim (Mt Hermon). In Soreq  
581 cave located to the south, the shift to less negative carbon values that are closer to the bedrock signal throughout  
582 the Holocene optimum, is interpreted as reflecting a soil denudation by intensive storms. The vegetation cover  
583 appears to be more stable at Jeita and changes drastically only during larger climatic shifts (e.g. 6 ka), whereas  
584 the soil system at high-altitude sites (Qadisha and Mizpe Shelagim) is more sensitive to rapid changes in the water  
585 budget and the snow cover dynamics and indicate short-term erosional phases between 7 and 6 ka.  
586

## 587 **8-Conclusions**

588 New stalagmites from Qadisha cave (Lebanon) located in the high Mt-Makmel, North of Mt-Lebanon, provide a  
589 well-dated record for the northern Levant with high-resolution geochemical proxies ( $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  values, trace  
590 element concentrations). The Qadisha stalagmites grew from 9.2 to 5.7 ka, albeit with discontinuities, and show  
591 a general tendency from more negative  $\delta^{18}\text{O}$  values during the Early Holocene towards more positive  $\delta^{18}\text{O}$  values  
592 at the end of the Holocene optimum. This trend is in agreement with a change from a wetter to a drier climate in  
593 this high-altitude area. The  $\delta^{13}\text{C}$  values show rapid shifts along the record and a decreasing trend towards more  
594 negative values when reaching the Mid-Holocene. The slight trend towards more negative values suggests overall  
595 improved bio-pedological soil activity in the Makmel mountains during the Mid-Holocene. Discrepancies  
596 between pollen data showing a wetter period at 7-6 ka than at 7-9 ka, unlike speleothems showing an overall wet  
597 Early Holocene are interpreted as related to a reduced snow cover season and thickness since 6.5 ka, explaining  
598 the extension of *Quercus* at 6.5 ka. On the short-term climate trend, Qadisha stalagmites record rapid dry and wet  
599 changes on centennial scale from 9 to 5 ka, with a tendency for drier conditions towards the mid-Holocene.  
600

601 Qadisha record is in good agreement with other Levantine records, such as the one from Jeita Cave, showing  
602 overall more humid climate conditions from 9 to 7 ka in the region. After 7 ka, a drier climate seems to affect sites  
603 at both low and high-altitude areas. The Qadisha record reflects particularities of a mountainous climate compared  
604 to other records: *i*) more negative stable-isotopic values than the Jeita record due to the altitudinal rainout effect,  
605 *ii*) a larger amplitude in the oxygen isotopes reflecting a more dynamic response to rapid climate changes than  
606 records located at lower altitudes, *iii*) the effect of snow cover and duration, regulating the effective infiltration in  
607 the Makmel area.  
608

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616

617 **10- CRediT author statement**

618 **C. Nehme:** conceptualization, writing-original draft preparation, field investigation and sampling, analysis;  
619 **S. Verheyden:** conceptualization, field investigation, sampling, reviewing and editing; **T. Kluge:** analysis,  
620 software validation, funding, writing, review and editing; **F.H. Nader:** field investigation and sampling,  
621 review and editing ; **R.L. Edwards :** analysis, validation, funding; **H. Cheng :** analysis, funding ; **E. Eiche:**  
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623

624

625 **11-References**

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## Editor's comments

1. Please include (i.e., repeat) the abstract in the text of the manuscript itself.

this was done in the revised final draft

2. I believe "northern Lebanon" (as in the title), not "Northern Lebanon" is correct--unless the latter is a formally defined locality. Please maintain consistency throughout the manuscript.

the term Northern was corrected in the text and the capitalized word was replaced by "northern"

3. The journal doesn't use "kyr", just "ka" (thus, for example, the "8.2 ka event")

Kyr was replaced by ka in the text.

4. Do you really need two dozen citations on lines 59-63? Unless this is a review article, your point could surely be made with a more selective list.

the list was shortened and the unnecessary citations were removed from the reference list

5. If you use "CE" then "yr" is extraneous. See, for example, line 269. Although the journal doesn't have formal guidance, "CE 1950" is a more intuitive rendering than "1950 CE". In any case, maintain consistency throughout.

In the text, the ages were converted all to ka and the term "kyr" was replaced by "ka". See correction model in lines 283, 285 and 302.

6. Figure 1a needs lat/long tics.

This was added in figure 1

7. Tables 1 and 2 need corrected headings for the ages.

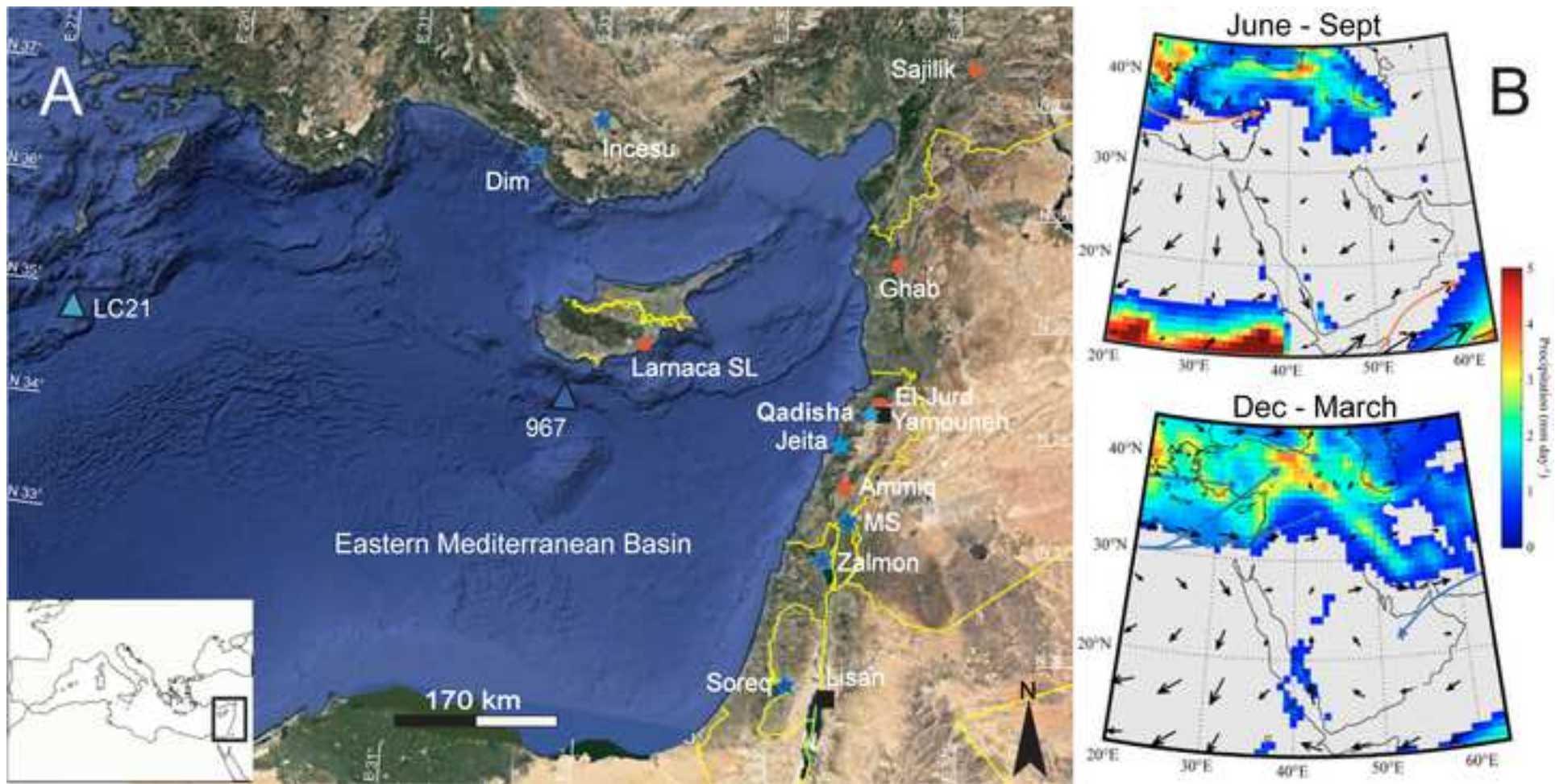
The corrected reference to AGE was changed in the headings of table 1 and 2.

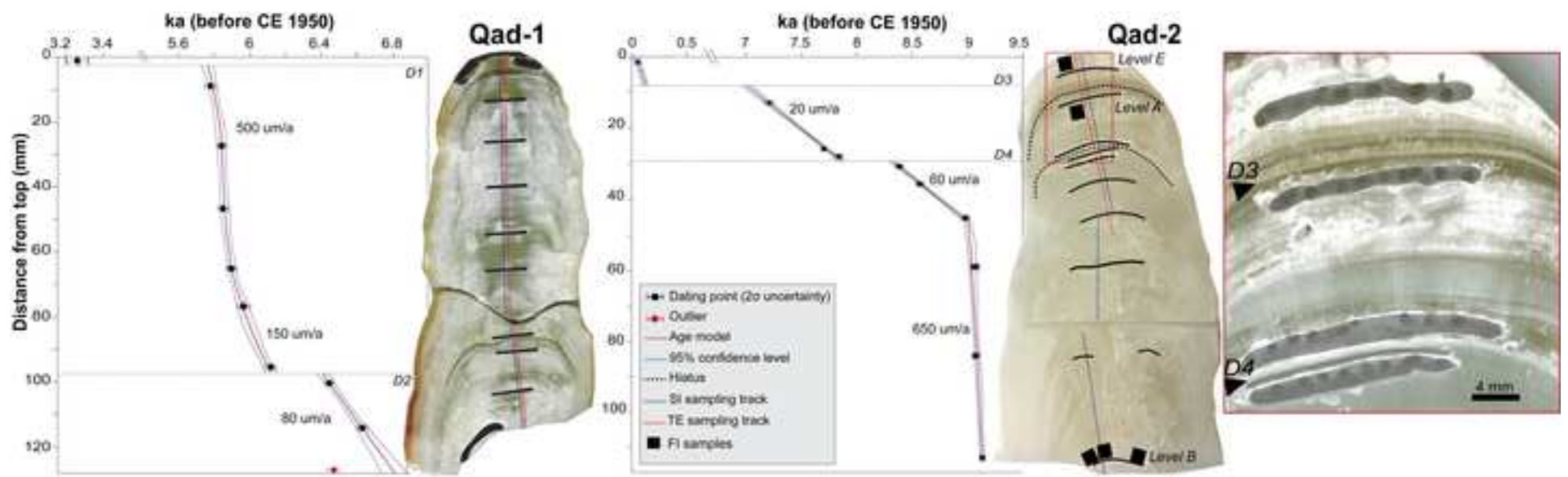
8. Figures 2, 3, 5, and 6 have four different ways of expressing what may (or may not) be the same thing. If you're giving an AGE, then simply "yr" (or "ka"), or if identifying the datum is critical, "yr (before CE 1950)". If you're giving a DATE, then "Date (CE)" would be correct. You should probably choose one framework and stick with it for all figures. In any case, no "yrs" and no "kyr".

In the figures, the reference to ages in the axis was changed into "Age (ka)". only in figure 2, we put Age (ka before CE 1950) and the figure 2 refers to the age model.

Figure 1

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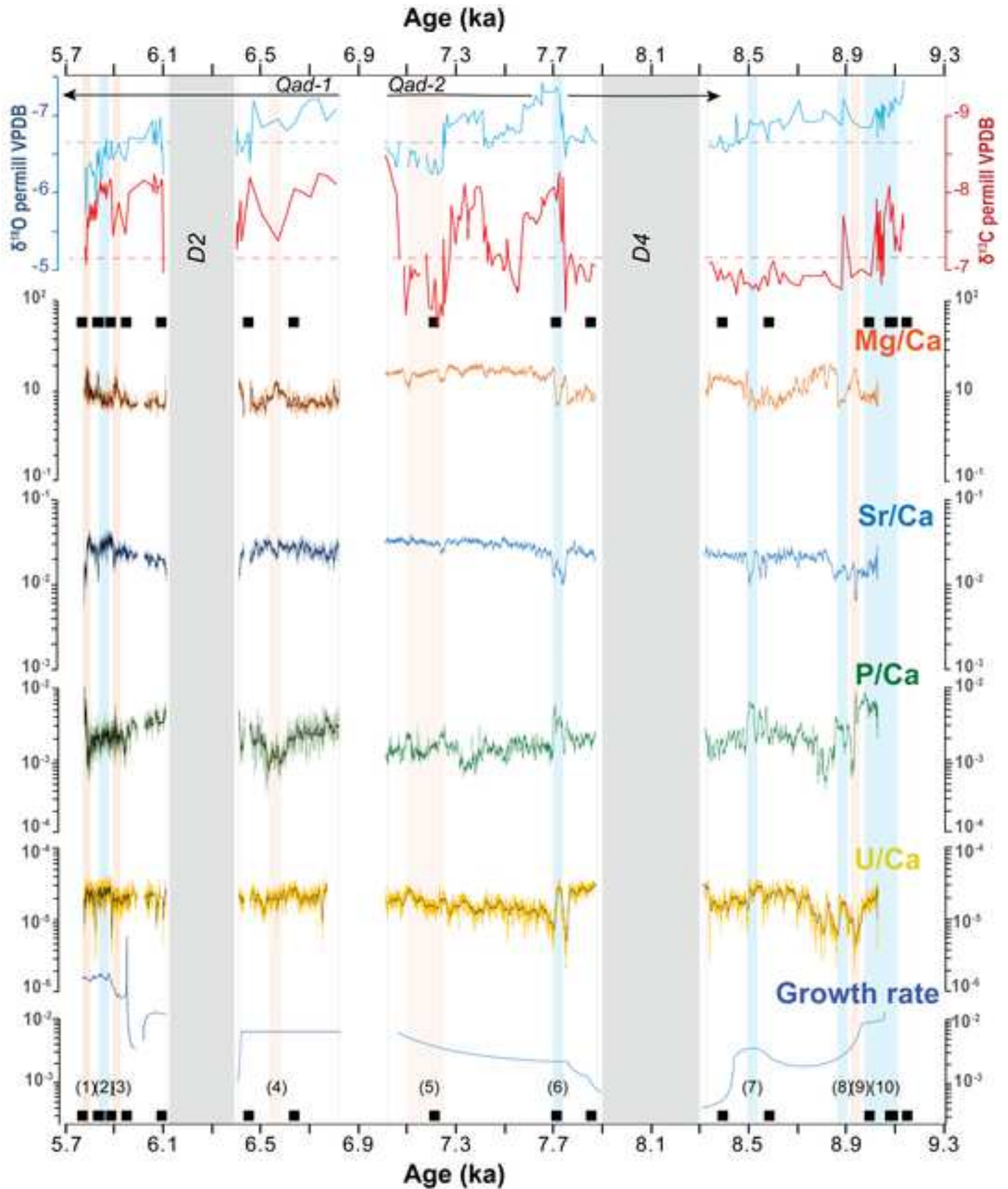


Figure 4

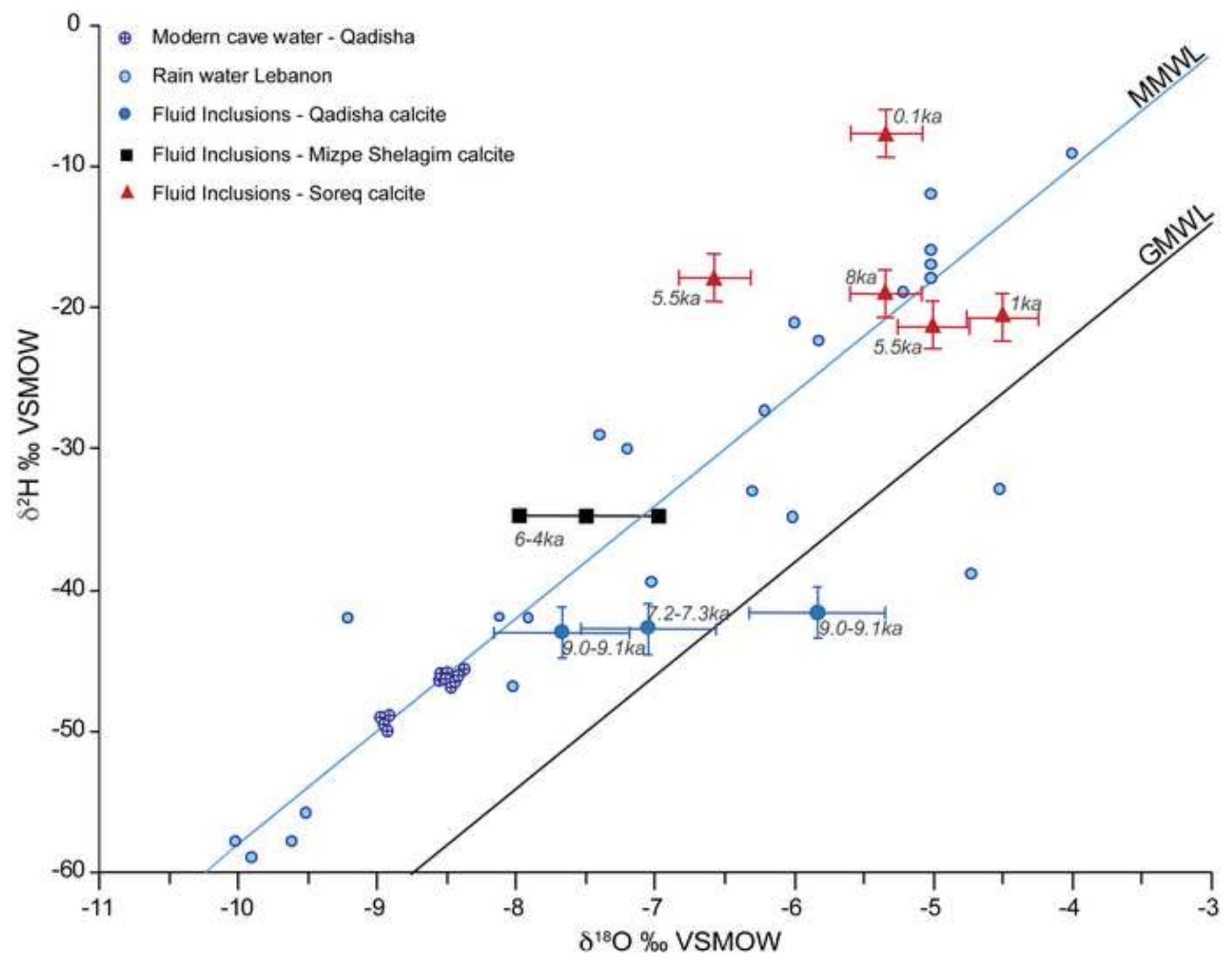




Figure 5

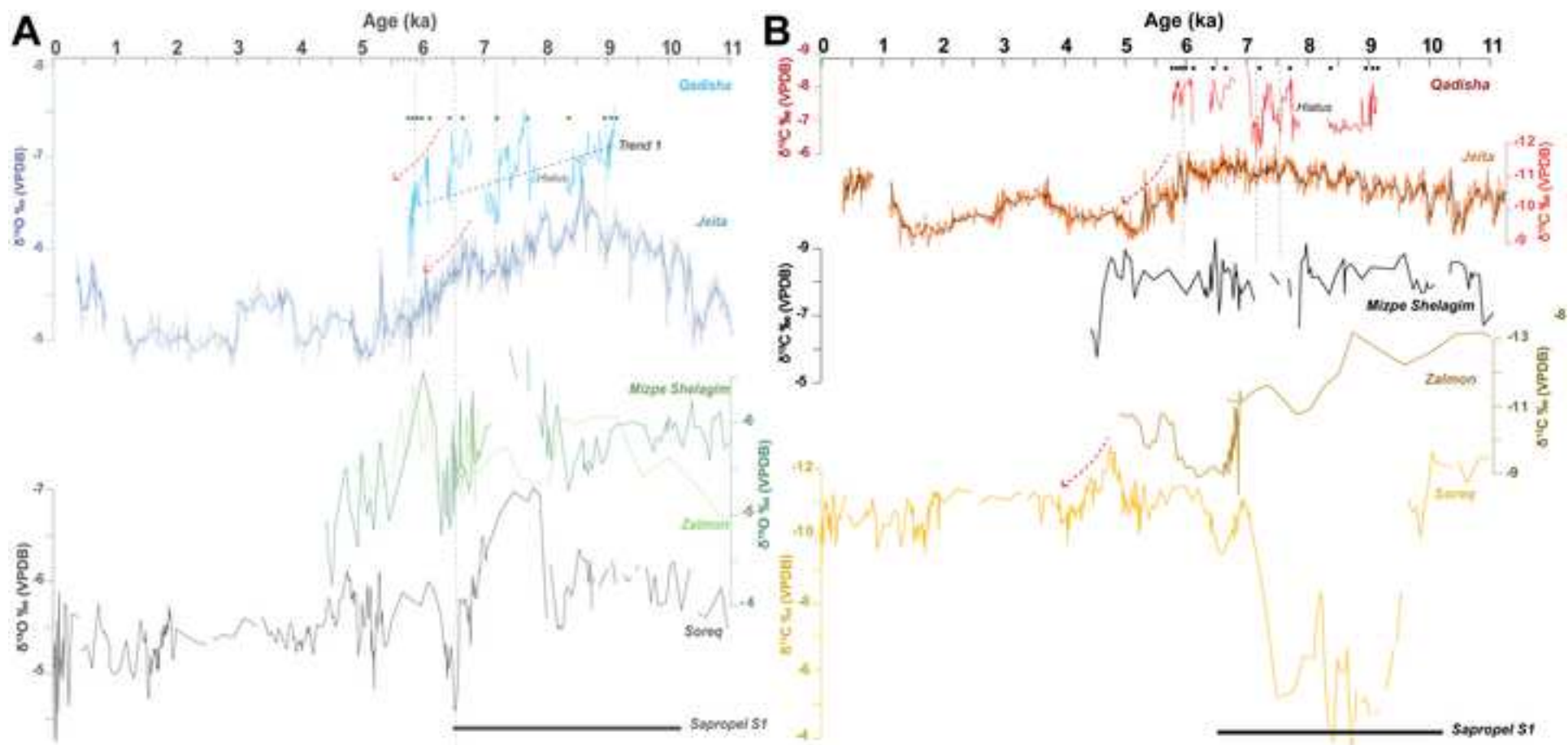
[Click here to access/download;Figure;Fig.5\\_Regional SI\\_cave records.png](#)

Figure 6

Click here to  
access/download;Figure;Fig.6\_Regional\_Levant\_records.png

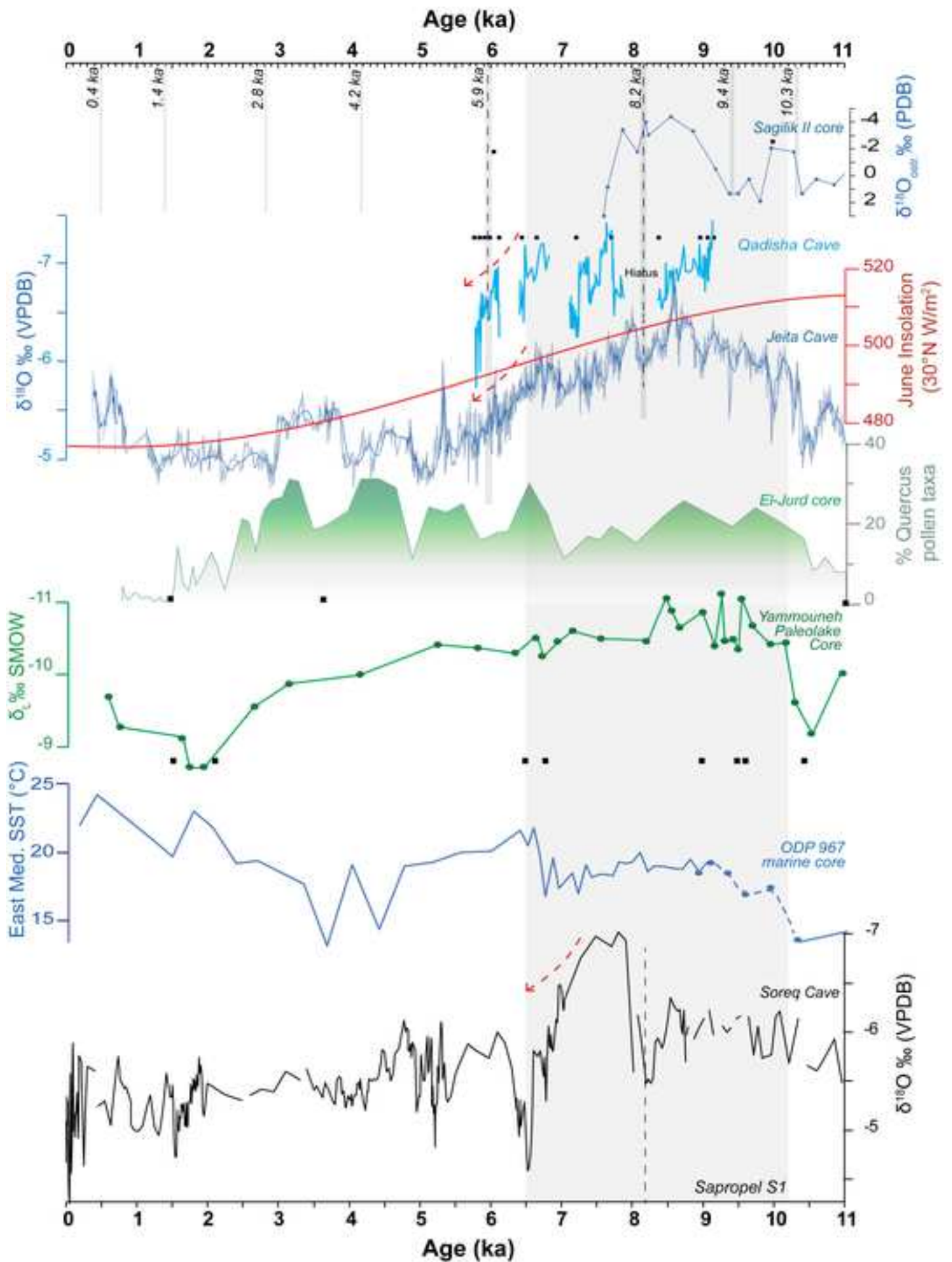


Table 1

error is  $2\sigma$  error.

Sample Number	depth (mm)	$^{238}\text{U}$ (ppb)	$^{232}\text{Th}$ (ppt)	$^{230}\text{Th} / ^{232}\text{Th}$ (atomic $\times 10^{-6}$ )	$\delta^{234}\text{U}^*$ (measured)	$^{230}\text{Th} / ^{238}\text{U}$ (activity)	$^{230}\text{Th}$ Age (a) (uncorrected)
Qad-stm1-0	2	435.0 $\pm$ 1.0	3591 $\pm$ 72	85 $\pm$ 2	353.7 $\pm$ 2.2	0.0426 $\pm$ 0.0002	3848 $\pm$ 19
Qad-stm1-8	7	606.5 $\pm$ 0.6	132 $\pm$ 3	5739 $\pm$ 140	442.4 $\pm$ 1.5	0.0756 $\pm$ 0.0003	5853 $\pm$ 28
Qad-stm1-7	26	575.7 $\pm$ 0.9	97 $\pm$ 3	7502 $\pm$ 240	444.3 $\pm$ 2.2	0.0764 $\pm$ 0.0004	5914 $\pm$ 30
Qad-stm1-6	46	615.3 $\pm$ 1.0	122 $\pm$ 3	6432 $\pm$ 183	450.6 $\pm$ 2.0	0.0774 $\pm$ 0.0004	5966 $\pm$ 29
Qad-stm1-5	65	665.4 $\pm$ 0.7	361 $\pm$ 8	2329 $\pm$ 50	445.7 $\pm$ 1.7	0.0767 $\pm$ 0.0003	5929 $\pm$ 24
Qad-stm1-4	77	535.7 $\pm$ 1.0	466 $\pm$ 10	1472 $\pm$ 31	437.3 $\pm$ 2.7	0.0777 $\pm$ 0.0004	6045 $\pm$ 33
Qad-stm1-3	96	572.6 $\pm$ 0.6	352 $\pm$ 7	2126 $\pm$ 46	432.1 $\pm$ 1.5	0.0792 $\pm$ 0.0003	6185 $\pm$ 28
Qad-stm1-2	101	546.2 $\pm$ 0.6	66 $\pm$ 2	11419 $\pm$ 389	441.9 $\pm$ 1.8	0.0840 $\pm$ 0.0003	6528 $\pm$ 25
Qad-stm1-1	115	643.7 $\pm$ 1.1	1156 $\pm$ 23	792 $\pm$ 16	433.5 $\pm$ 2.2	0.0863 $\pm$ 0.0003	6748 $\pm$ 28
Qad-stm1-00*	128	639.0 $\pm$ 2.0	978 $\pm$ 20	902 $\pm$ 18	428.1 $\pm$ 2.6	0.0838 $\pm$ 0.0003	6573 $\pm$ 24
Qad-STM2-5	6	558.6 $\pm$ 1.5	194 $\pm$ 4	45 $\pm$ 3	303.3 $\pm$ 2.1	0.0009 $\pm$ 0.0001	79 $\pm$ 5
Qad-stm2-4	13	297.6 $\pm$ 0.3	193 $\pm$ 4	2309 $\pm$ 51	397.4 $\pm$ 1.7	0.0907 $\pm$ 0.0004	7294 $\pm$ 36
Qad-stm2-3	25	621.7 $\pm$ 0.6	316 $\pm$ 6	3161 $\pm$ 65	406.2 $\pm$ 1.4	0.0973 $\pm$ 0.0002	7794 $\pm$ 22
Quad 2-8.2	28.2	351.8 $\pm$ 0.3	255 $\pm$ 6	2284 $\pm$ 50	423.4 $\pm$ 1.6	0.1003 $\pm$ 0.0004	7937 $\pm$ 38
Qad-stm2-2	30	336.4 $\pm$ 0.5	87 $\pm$ 2	6980 $\pm$ 178	457.5 $\pm$ 2.1	0.1093 $\pm$ 0.0004	8466 $\pm$ 32
Qad-stm2-8.6	36	367.1 $\pm$ 0.3	284 $\pm$ 6	2422 $\pm$ 52	483.2 $\pm$ 1.6	0.1138 $\pm$ 0.0004	8667 $\pm$ 35
Qad-stm2-8	49	499.2 $\pm$ 0.7	62 $\pm$ 2	15661 $\pm$ 580	481.2 $\pm$ 2.2	0.1186 $\pm$ 0.0004	9062 $\pm$ 35
Qad-stm2-7	67	582.4 $\pm$ 0.8	188 $\pm$ 4	6121 $\pm$ 144	484.8 $\pm$ 2.1	0.1200 $\pm$ 0.0004	9153 $\pm$ 34
Qad-stm2-6	80	566.7 $\pm$ 0.8	379 $\pm$ 8	2970 $\pm$ 63	489.1 $\pm$ 2.1	0.1206 $\pm$ 0.0004	9167 $\pm$ 34
Qad-STM2-1	106	545.4 $\pm$ 0.9	207 $\pm$ 4	5220 $\pm$ 106	472.7 $\pm$ 1.4	0.1199 $\pm$ 0.0002	9218 $\pm$ 20

U decay constants:  $\lambda_{238} = 1.55125 \times 10^{-10}$  (Jaffey et al., 1971) and  $\lambda_{234} = 2.82206 \times 10^{-6}$  (Cheng e  
 $\delta^{234}\text{U} = ([^{234}\text{U}/^{238}\text{U}]_{\text{activity}} - 1) \times 1000$ . \*\*  $\delta^{234}\text{U}_{\text{initial}}$  was calculated based on  $^{230}\text{Th}$  age (T), i.e.,  $\delta^{234}\text{U}_{\text{initial}}$   
Corrected  $^{230}\text{Th}$  ages assume the initial  $^{230}\text{Th}/^{232}\text{Th}$  atomic ratio of  $4.4 \pm 2.2 \times 10^{-6}$ . Those are the values  
equilibrium, with the bulk earth  $^{232}\text{Th}/^{238}\text{U}$  value of 3.8. The errors are arbitrarily assumed to be 50%.  
\*\*\*CE stands for ‘‘Contemporaneous Era’’. Ages are defined as the year before 1950 A.D.  
\* age considered as outlier

<sup>230</sup> Th Age (a) (corrected)	$\delta^{234}\text{U}_{\text{Initial}}$ ** (corrected)	230Th Age (yr CE)*** (corrected )
3307 ±127	357 ±2	3247 ±127
5848 ±28	450 ±1	5780 ±28
5911 ±30	452 ±2	5843 ±30
5962 ±29	458 ±2	5894 ±29
5918 ±25	453 ±2	5850 ±25
6028 ±35	445 ±3	5960 ±35
6173 ±29	440 ±2	6105 ±29
6525 ±25	450 ±2	6457 ±25
6711 ±38	442 ±2	6643 ±38
6542 ±32	436 ±3	6482 ±32
<hr/>		
71 ±7	303 ±2	5 ±7
7281 ±37	406 ±2	7213 ±37
7783 ±23	415 ±1	7715 ±23
7922 ±39	433 ±2	7851 ±39
8461 ±33	469 ±2	8393 ±33
8652 ±36	495 ±2	8581 ±36
9060 ±35	494 ±2	8992 ±35
9147 ±34	497 ±2	9079 ±34
9154 ±35	502 ±2	9086 ±35
9211 ±21	485 ±1	9145 ±21

t al., 2013). Th decay constant:  $\lambda_{230} = 9.1705 \times 10^{-6}$  (Cheng et al., 2013).

$$I = \delta^{234}\text{U}_{\text{measured}} \times e^{\lambda_{234}T}$$

for a material at secular

Table 2

Qad_2 FI	Age of level [ka CE]	Sample code	$\delta H_2$ [permil]	$\delta^{18}O$ [permil]	Volume [ $\mu$ l]	Mass [g]	Water amount [ $\mu$ l/g]	$T_{fi}$ [ $^{\circ}C$ ]
Level B	9.0-9.1	BQ_2_d	-41.70	-5.81	0.29	0.58	0.50	18.7 $\pm$ 2.5
Level B	9.0-9.1	BQ_2_b	-43.20	-7.65	0.73	0.78	0.94	10.3 $\pm$ 2.5
Level B*	9.0-9.1	BQ_2_a	-39.58	-5.53	0.23	0.72	0.32	20.1 $\pm$ 2.5
Level A	7.2-7.3	AQ_1	-42.82	-7.04	0.39	0.46	0.84	12.2 $\pm$ 2.3
Level E*	0.05	EQ_1	-19.46	-6.88	0.01	0.45	0.02	11.2 $\pm$ 2.4

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This piece of the submission is being sent via mail.