Rowe et al., Multi-proxy speleothem record of climate instability during the early last interglacial in southern Turkey

<u>Highlights</u>

- A speleothem from Turkey provides a climate record through Termination II;
- Isotopic evidence shows rainfall increased at the start of the last interglacial;
- A dry period lasting ~200 years occurred early in the last interglacial;
- The isotopic structure of the dry event is similar to the Holocene 8.2 ka event;
- A synchronous climate anomaly is identified in other European speleothems;



1	Multi-proxy speleothem record of climate instability during the early last
2	interglacial in southern Turkey
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14	
15	Abstract: A stalagmite from Dim Cave in southern Turkey contains a climate record
16	documenting rapid and significant changes in amounts of precipitation between ~ 132 ka and
17	\sim 128 ka, during the penultimate glacial – interglacial transition. Some U-Th dates have been
18	compromised by carbonate dissolution but rigorous selection and tuning to $\delta^{18}O$ records from
19	other speleothems has generated a robust age model. Growth rate was initially very slow but
20	a rapid increase at ~129 ka was accompanied by strong negative trends in $\delta^{18}O$ and $\delta^{13}C$, a
21	combination implying the onset of much wetter conditions. Isotopic values at ~ 129 ka
22	suggest that groundwater recharge rates and biogenic activity in the soil zone exceeded those
23	of the early Holocene. A significant isotopic enrichment event at ~128 ka, during which there
24	was alternating aragonite and calcite deposition, documents a strong drying event with a

duration that lasted ~200 years. A concurrent decrease in ⁸⁷Sr/⁸⁶Sr ratios indicates increased 25 26 groundwater residence times and the cumulative evidence suggests amounts of rainfall fell 27 from well above to slightly below present-day levels. Similar δ^{18} O enrichment events are present in coeval speleothem records from southwest France and the Northern Alps, and 28 29 these, together with pollen evidence from Italy, Greece and the Iberian margin of drier 30 conditions at this time, imply that a climate anomaly extended across the northern 31 Mediterranean borderlands. The timing, duration and structure of this episode are consistent 32 with marine evidence of strong North Atlantic cooling early in the last interglacial and there 33 is a resemblance to the Holocene 8.2 ka event recorded globally in many proxy-climate archives. 34 35 36 Keywords: Stalagmite; Termination II; stable isotopes; climate anomaly; aragonite; Mediterranean. 37 38 39 **1. Introduction** 40 41 Glacial to interglacial transitions involve complex changes in climate on decadal to 42 millenial timescales as large land-based northern hemisphere ice sheets disintegrate, raising 43 sea level and stimulating changes in ocean and atmosphere circulation patterns. The nature 44 and timing of climate changes during the Late Glacial - Early Holocene transition in the 45 North Atlantic region is known in considerable detail from multiple sources (Lowe et al., 2008) such as high resolution stable isotope data from Greenland ice cores (Steffensen et al., 46 47 2008) and lacustrine cores (Von Grafenstein et al., 1999), palynological profiles (Allen et al., 48 1999; Bottema, 1995; Lotter et al., 2000), coleopteran records (Coope and Lemdahl, 1995; Coope et al., 1998) and marine sediment cores (McManus et al., 2004; Peck et al., 2008). 49

50 These have identified rapid switching between stadial and interstadial conditions prior to the 51 start of the Holocene at 11.7 ka (Walker et al., 2009) as erratic ice sheet disintegration 52 periodically disrupted the North Atlantic meridional overturning circulation (MOC), with 53 consequent strong and widespread impacts on global climates. In contrast, the penultimate 54 transition into the last interglacial (LIG), between ~132 ka and ~128 ka, is known in less 55 detail, mainly because the Greenland record does not extend beyond the mid-LIG (Andersen 56 et al., 2004), fewer high resolution terrestrial sites of relevant age survive for study and 57 chronologies are generally less well constrained. These limitations compromise both the 58 detection of short-lived climatic events in many terrestrial palaeoclimate archives, and their 59 correlation with possible equivalents in the more complete marine records.

60

61 In the Mediterranean, the penultimate glacial-interglacial transition is recorded in diverse 62 proxy-climate records including stable isotopes and growth rates in speleothems (Bar-63 Matthews et al., 2003; Drysdale et al., 2009), lacustrine palynological profiles (Allen and 64 Huntley, 2009; Brauer et al., 2007; Milner et al., 2012; Sinopoli et al., 2018; Tzedakis, 2003), variations in shelf sediment flux (Toucanne et al., 2015) and multi-proxy marine core data 65 66 (Grant et al., 2012; Jimenez-Amat and Zahn, 2015; Kandiano et al., 2014; Martrat et al., 2014; Rohling et al., 2015). A general trend towards more negative δ^{18} O values is generally 67 68 seen in Mediterranean speleothems through the transition, a pattern which is linked via 69 atmospheric water vapour of predominantly marine origin to a similar trend in ocean water 70 due to rising sea levels (Rohling et al., 2015). Concurrent increases in speleothem growth rates (Drysdale et al., 2009) imply higher groundwater infiltration rates and soil pCO₂ as a 71 72 consequence of increasing rainfall between ~132 ka - 128 ka stimulated by rising sea surface 73 temperatures (SST) (Kandiano et al., 2014; Martrat et al., 2004). A strengthening African 74 Summer Monsoon increased freshwater outflow from the River Nile and North African wadi

75 systems, contributing substantially to the formation of Sapropel S5 at ~129 ka (Grant et al., 76 2012: Ziegler et al., 2010). It has been suggested that freshwater input to the Mediterranean 77 was dominated by African sources and that additional contributions were negligible (Osborne 78 et al., 2010), and some modelling simulations suggest that Eemian European storm tracks 79 were shifted further north than at present (Kaspar et al., 2007). However, there is also 80 evidence of a significant runoff contribution from the Mediterranean northern borderlands 81 (Rohling et al., 2015; Toucanne et al., 2015) which has been linked to increased winter rains 82 in the Mediterranean during periods of precession minima (Bosmans et al., 2015; Kutzbach et 83 al., 2014). Pollen evidence shows that Mediterranean sclerophyllous vegetation increased and 84 peaked very early in the LIG, demonstrating that summers were hot and dry and therefore 85 that increased rainfall was confined to the winter months (Milner et al., 2012), implying an 86 Atlantic moisture source for the precipitation rather than an incursion of summer monsoonal 87 air from the south. Marine records from the North Atlantic indicate significant instability in 88 the MOC during the glacial-interglacial transition (Irvali et al., 2012; Mokeddem et al., 2014; 89 Nicholl et al., 2012), and this is likely to have stimulated abrupt changes in atmospheric 90 circulation patterns downstream over Eurasia. Identification and high resolution capture of 91 such rapid climate events in contemporary terrestrial archives is currently limited, although 92 there has been recent progress towards integrating marine, pollen and speleothem data into a 93 coherent climatic framework (Tzedakis et al., 2018).

94

95 Here we use oxygen, carbon and strontium isotope data, together with petrography, from a 96 speleothem from Dim Cave in southern Turkey to infer changes in rates of groundwater 97 recharge through the penultimate glacial-interglacial transition. These changes imply 98 significant variability in climate, especially in rainfall, similar to early-mid-Holocene

99 fluctuations which are linked to instability in the North Atlantic MOC (Cheng et al., 2009b;100 Daley et al., 2011).

101

102 **2.** Cave setting and regional climate

103

104 Dim Cave (36° 32' 27" N, 32° 06' 32" E) is located 235 metres above sea level in a spur 105 on the southern wall of the Dim River valley ~6 km from the Mediterranean coast of southern 106 Turkey, 11 km east of Alanya and 145 km south-east of Antalya (Fig. 1). It is a linear fault-107 controlled fossil phreatic cave formed in dolomitised Permian limestone which constitutes the 108 upper unit of the Alanya Nappe (Okay and Ozgul, 1984). The single main passage ~360 m 109 long and 10-15 m wide, shows considerable vertical development along fissures, and 110 terminates in a small subterranean lake which is perched on a bed of impermeable schist. 111 Shoreline evidence suggests that the lake level has previously been ~ 0.5 m higher. The 112 epikarst varies in thickness from a few metres to several tens of metres and is overlain by thin 113 soils which support a mainly coniferous woodland with an understorey of bushes and a sparse 114 ground cover. The dominant vegetation units of the region are generally red pine and maquis (Kurt et al., 2015). Relative humidity in the cave is >90 % (Baykara, 2014) and the 115 116 temperature is 18-19°C, indistinguishable from the average annual mean 1963-2004 117 temperature of 18.4°C at the coastal station of Antalya, 130 km WNW of Dim Cave and 50 m 118 above sea level (IAEA/WMO), and from temperatures in Soreq Cave (385 m a.s.l.), Israel, 119 and winter-spring SST in the Eastern Mediterranean (EM) (Bar-Matthews et al., 2003). 120 Although the cave is richly decorated, much of it is presently dry with seepage waters 121 restricted to a few locations mainly associated with major fissures. Modern active straw 122 stalactites selected for analysis were 100% aragonite (identified by X-ray diffraction, XRD), 123 implying that under present conditions this is the dominant speleothem-forming mineral. This

contrasts with the predominantly calcite mineralogy of the speleothems from the cave used to reconstruct a continuous climate record for the period 90 - 10 ka during which aragonite was only recorded between $\sim 80 - 75$ ka (Unal-Imer et al., 2015; Ünal-Imer et al., 2016).

127

128 Turkey lies in a zone of transition between the mid-latitude westerlies and the sub-tropical 129 high pressure belt, which dominate in winter and summer respectively, and its climatology is 130 complex (Spanos et al., 2003). In southern Turkey, as in most of the Mediterranean, summers 131 are very dry and there is a pronounced winter rainfall maximum. Atlantic depressions reach 132 the Eastern Mediterranean through Western and Central Europe and via the Western 133 Mediterranean Basin and regenerate in centres of cyclogenesis located in the Gulf of Genoa, 134 the Aegean and off southern Cyprus (Alpert et al., 1990; Karaca et al., 2000; Krichak and 135 Alpert, 2005; Kutiel et al., 2002; Türkeş, 1998; Türkeş et al., 2008). The intensity and tracks 136 of winter depressions, and therefore rainfall amounts, are influenced by regional pressure 137 patterns, primarily the North Atlantic Oscillation (NAO) (Türkeş and Erlat, 2003), an Eastern 138 Atlantic-Western Russian (EAWR) pressure pattern (Krichak and Alpert, 2005) and a North 139 Sea-Caspian Pattern (NSCP) (Kutiel and Benaroch, 2002). Consequently, winter rainfall 140 amounts in the EM are ultimately determined by regional circulation patterns beyond the 141 Mediterranean Basin.

142

143 Antalya receives over 70% of its annual rainfall (~1100 mm) between November and 144 February and 98% between October and May (Fig. S1). The mean November – February 145 δ^{18} O value from intermittent Antalya precipitation data from 1963 – 2004 is -5.8±0.6‰_{VSMOW} 146 (IAEA/WMO), identical to the mean δ^{18} O value of -5.7±0.3‰_{VSMOW} from five sets of six 147 drip and pool water samples collected from Dim Cave and a nearby large spring in March,

148 May and December 2009 and January and February 2010. These plot along a meteoric water 149 line (MWL) (Fig S2) defined by $\delta^{2}H = 7.0 \cdot \delta^{18}O + 14.2$. 150 (1) 151 This compares with $\delta^{2}H = 7.3 \cdot \delta^{18}O + 12.9$ 152 (2) 153 for Antalya monthly precipitation 1963-2001 (Dirican et al., 2005), and $\delta^{2}H = 7.2 \cdot \delta^{18}O + 10.8$ 154 (3) 155 for groundwaters on the windward (southern) side of the Taurus Mountains (Schemmel et al., 156 2013). The lower slope of Dim Cave water may indicate some mixing with a residual element 157 of evaporated water within the epikarst. The proximity of the data to the Mediterranean 158 Meteoric Water Line (MMWL) $\delta^2 H = 8.0 \bullet \delta^{18} O + 22$ 159 (4) 160 rather than the Global Meteoric Water Line (Fig S2) points to the Mediterranean as the 161 principal moisture source for the seepage waters. The Antalya and Taurus Mountain meteoric 162 water lines are intermediate between the global and Mediterranean lines, probably indicating 163 a significant contribution from moisture sources beyond the Mediterranean Basin. Incomplete monthly data for Antalya from 1963 to 2004 suggest that δ^{18} O in precipitation declines by 164 about 0.8‰ per 200 mm of rainfall, compared with 1.0±0.1‰ per 200 mm at Soreq Cave, 165 166 Israel from twelve years of annual data (Bar-Matthews et al., 1997) and 1.6±0.2‰ per 100 167 mm reported for monthly data from the Central Mediterranean (Bard et al., 2002). This 168 negative relationship between δ^{18} O in precipitation and rainfall amount (the "amount effect") (Dansgaard, 1964)) is generally considered to strongly influence δ^{18} O in Mediterranean 169 170 speleothems on sub-orbital timescales. 171

172 **3. Materials and Methods**

173

174 *3.1 Speleothem Description*

175

176 Dim 1 is a fossil stalagmite, 568 mm long and between 110 and 180 mm in diameter (Fig. 177 2a). Dim 1 toppled before 8.0 ka, as it has a small aragonite stalagmite (Dim 3; mineralogy 178 from XRD) on its upper side which grew between 8.0 and 7.0 ka (Table 1), and it was 179 missing its top and several mm of its base when collected. Longitudinal sectioning revealed 180 discrete growth layers. A growth axis offset of ~5 mm occurs at 460 mm distance from top 181 (dft), and at 259 mm and 131 mm dft the axial laminae flatten to define obvious 'stalagmite 182 top' morphologies. This is particularly clear at 131 mm dft where the stalagmite top was 183 rimmed by 'edge ramparts' that defined a <5 mm deep pool (Fig. 2b). There is no evidence 184 that growth stopped at any of these morphological surfaces. A prominent band of paler, 185 creamy carbonate is present between 103 and 87 mm dft (Fig. 2b) and this carbonate can be 186 followed into flanking layers that encase the 'core' of the speleothem (Fig. 2b). In the axial 187 part of the stalagmite, at 87 mm dft, the upper surface of this pale, creamy carbonate band 188 displays a clear corrosion surface interpreted as a hiatus (Fig. 2b). Above this surface there is 189 a 10 mm zone where pale creamy carbonate near the flanks appears to be replaced by darker 190 carbonate in the axial zone (Fig. 2b). Moreover, above the corrosion surface the growth axis 191 is offset by ~20 mm. The speleothem diameter narrows slightly towards the top and 192 culminates in a broken surface.

193

194 *3.2. U-Series Dating*

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196 Twenty five 100-150 mg samples were taken for U-series dating using a hand-held drill.
197 All were proximal to the central growth axis except 52-1, -2, -3, -4 and -5 which were taken

from the flanking pale creamy-coloured aragonite layers that could be traced into the growth
axis, except 52-3 which stratigraphically was a few mm higher (Fig. 2a). Laboratory and

200 instrumental methods are detailed in Supplementary Material.

201

202 3.3. Carbonate $\delta^{18}O$ and $\delta^{13}C$ analyses

203

204 421 carbonate samples each weighing \sim 70 µg were drilled at 1 or 2 mm intervals along the 205 growth axis using a 0.5 mm hand-held dental drill. The measurements were carried out in the 206 Stable Isotope Laboratory at the University of East Anglia, UK, on a Europa SIRA II dual inlet isotope ratio mass spectrometer following reaction with 100% phosphoric acid at 90° C, 207 208 using an on-line 'common acid bath' system. An internal laboratory standard ($\delta^{13}C =$ $1.99\%_{VPDB}, \delta^{18}O = -2.05\%_{VPDB}$ calibrated against NBS19 ($\delta^{13}C = 1.95\%_{VPDB}, \delta^{18}O = -$ 209 210 2.20‰ _{VPDB}) was measured with the samples in each batch. Measurement precision based on 211 the standard deviation of repeat analyses of the standard (n=7) was better than 0.08% for 212 both oxygen and carbon. Isotopic data are recalculated to an acid reaction temperature of 213 25°C and aragonite δ^{18} O values corrected by -0.29‰ to account for the differing calcite and 214 aragonite acid fractionation factors (Lachniet, 2015). Final values are reported as parts per 215 thousand (‰) deviations relative to VPDB. 216

217 *3.4. Strontium isotope analysis*

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219 Strontium isotope analyses were carried out at the NERC Isotope Geosciences Facility,

220 Keyworth, UK. Samples were taken adjacent to the growth axis and leached in 1% acetic acid

in order to remove labile strontium. Samples were then leached in warm 10% acetic acid in

222 order to dissolve carbonate material. The first batch of samples were not spiked, but

subsequent samples were spiked with an ⁸⁴Sr tracer to allow calculation of the Sr

224 concentration in addition to the isotope composition. Following conversion to nitrate, Sr was 225 separated using EICHROM Sr-Spec ion exchange resin, loaded on single Re filaments using 226 a TaO activator, and analysed using a Thermo Scientific Triton mass spectrometer operating 227 in multi-dynamic mode. Nine analyses of the NBS987 standard gave a value of $0.710251 \pm$ 228 0.000003 (4.9 ppm, 1-sigma) at the time of analysis.

229

4. Results

231 *4.1. Petrography*

232

Dim 1 has a mixed mineralogy of calcite and aragonite as identified petrographically and confirmed by X-ray diffraction. Axial parts of Dim 1 are mainly inclusion-poor columnar calcite, but are encased by flanks of pale creamy-pink aragonite-rich layers (Fig. 2a). These flanking layers thin and pinch out downward, and above ~88 mm dft they thicken and project above the broken stalagmite top (Fig. 2b). Detailed relationships between these aragonite-rich flanking layers and the axial part of the stalagmite are described after the more obvious changes in the axial part have been outlined.

240

The axial stalagmite 'core' consists largely of grey-brown columnar compact (C) calcite, which in some places have length to width ratios approaching the elongated columnar (Ce) fabric (Fig. 3a) of (Frisia, 2015). Toward the edges of the calcite 'core', C fabrics can grade into, or inter-finger with, pale cream columnar open (Co) fabrics (Frisia, 2015), a relationship similar to that described in a speleothem from Jeita Cave, Lebanon (Verheyden et al., 2008). These paler Co calcites also extend across most of the width of the axial 'core' at depths of

119-133 mm and 201-204 mm dft, and contain columnar crystals with shorter c-axes than thegrey-brown C calcites.

249

250 At ~103 mm dft there is a prominent, sharp, boundary with millimetre-scale, step-like 251 relief forming a central 'horst', followed by a 7 to 13 mm zone of mixed aragonite and calcite 252 alternating in millimetre-scale layers grading into mainly aragonite on the flanks. This is the 253 pale creamy zone seen in slabbed specimen (Fig. 2b). In the axial part of Dim 1 where the 254 isotope transect was drilled, the first 2.0 mm of this zone is dominantly (90%) bundles of 255 acicular aragonite that nucleated directly on underlying columnar crystal terminations with c-256 axes normal to stalagmite extension direction (Fig. 3b); spaces between bundles are filled 257 with non-orientated aragonite needles (Fig. 3c). At topographic highs on this boundary the C 258 calcites of the central 'horst' show sharp lateral change to aragonite (Figs. 3c and d). The next 259 4 mm are dominantly (>90%) calcitic C fabrics which then grade back to a layer of pure 260 aragonite needles 2 mm thick (Fig. 4a). A 1 mm thick C calcite (100%) layer follows (Fig. 261 4a), and above this pure aragonite needle fabrics return for 1.5 mm with sharp but irregular 262 upper boundary that in places cuts out the entire aragonite layer into the underlying 1 mm 263 thick C calcite layer. The vertical transition between aragonite and calcite in this zone is not 264 always sharp and the layer thicknesses given above vary laterally over centimetre-scale 265 distances with the calcite layers wholly replaced by aragonite in some places. Within this 266 zone there are multiple horizons of linear inclusions arranged parallel to growth that occur in 267 both the aragonite and calcite fabrics, in the latter often associated with isolated aragonite 268 needles (Fig. 4b and c).

269

270 On the flanks of this zone the mineralogy is almost purely aragonite, present up to 0.5 mm 271 lower than the basal boundary in the axial part. The aragonite fabrics are dominantly acicular

bundles or spherulitic aggregates of needle crystals (Fig. 4d) with a highly irregular upper
boundary (Fig. 5a). At the boundary, calcite C crystals or more mosaic fabrics about the
aragonite needle fabrics (Fig. 5a).

275

The upper boundary of the mixed mineral zone in the axial 'core' (~87 mm dft; Fig. 4b
and c) is sharp but undulose with underlying crystals showing ragged terminations (Fig. 4c).
Above this, the growth axis is offset and stalagmite diameter reduces. Above the upper
boundary, pale cream Co calcite is the dominant fabric, although in places directly above the
boundary the basal 200-500 µm contains patchy equant microspar fabrics (Fig. 4c).

281

282 At depths of 9 mm, 36 mm, 103 mm, 114 mm, 120 mm, 143 mm and 262 mm dft, thin 283 horizons of aragonite project into the axial calcite zone from the flanking layers but typically 284 die out before reaching the axis. These horizons have sharp lower and upper boundaries and 285 pass laterally into sub-horizontal, undulose, dark 'lines' (<1 µm thick), usually inclusion-rich 286 and parallel to growth in the central axis (Fig. 5b-e). The fluid inclusions are typically 287 spherical (Fig. 5c) and range in size between $<1 \mu m$ and 15 μm . Some, but not all, of these 288 dark lines and inclusion trails are associated with relict aragonite needles (Fig. 5d); some 289 grade laterally into darker and thicker layers (mean \sim 70 µm) that define crystal terminations 290 (Fig. 5e) and in places appear to corrode the underlying calcite. These dark lines and layers 291 occur at irregular intervals along the stalagmite but are not necessarily defined by inclusion 292 trails or confined to locations where remnants of aragonite persist.

293

294 4.2. Uranium-series

295

The U-series data (Table 1) reveal negligible levels of ²³²Th, U concentrations in aragonite 296 297 that are 10-60 times higher than in calcite, and calculated ages between 117 ka and 137 ka 298 (Figs. S3, S4). However, the dates do not consistently lie in stratigraphic order, implying a 299 degree of isotopic mobility in some samples. This is most likely related to the dark "lines" 300 and fluid inclusion trails identified at intervals within the stalagmite (Section 6, below) and 301 which are often associated with corrosion of underlying crystal terminations. Petrographic examination shows the presence of such features at sample locations 29-10, 41-8, 41-6, 29-7, 302 41-5, 41-4, 33-6, 41-2, 41-1 and 33-5, (Table 1). 303

304

305 *4.3. Oxygen and carbon isotopes*

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307 The δ^{18} O and δ^{13} C profiles (Fig. 6a, b) are broadly similar (linear regression of the two 308 data sets yields $r^2 = 0.47$), although the latter shows a stronger overall negative trend, and 309 both capture a strong positive excursion beginning at ~143 mm dft and culminating in the 310 hiatus at 87 mm dft, following the switch from calcite to predominantly aragonite deposition. 311 Minimum δ^{18} O and δ^{13} C values occur above the hiatus, after which they increase towards the broken stalagmite top. δ^{18} O values below 240 mm dft lie mainly between -5.5‰ and -6.5‰, 312 313 falling subsequently to between -6.5‰ and -7.0‰ before increasing rapidly above 140 mm to a maximum of -5.0% at 95 mm dft within the aragonite-rich layer. Above the hiatus δ^{18} O 314 315 values around -7.8‰ gradually increase to -6.5‰ at the broken top. Typically, the values in 316 Dim 1 are $\sim 0.5\%$ more negative than those recorded in Dim Cave speleothem E2 between 13 317 ka and 10 ka during the last glacial-interglacial transition (Unal-Imer et al., 2015). In 318 comparison, aragonite from modern straw stalactites and mid-Holocene stalagmite Dim 3 319 have δ^{18} O values between -3.6‰ and -5‰. The structure of the carbon isotope record shows a more distinct negative drift from the base to 140 mm dft than the oxygen. δ^{13} C values in the 320

321	lower 100 mm lie mainly around -9‰, comparable to those recorded in Dim Cave
322	speleothems between 13.0 ka and 11.5 ka (Unal-Imer et al., 2015), and fall to -11.5‰, similar
323	to early Holocene, in two step-wise depletions at \sim 460 mm and \sim 320 mm dft, although
324	several brief ~3‰ positive excursions also occur within this phase. Above ~140 mm $\delta^{13}C$
325	increases erratically to a maximum of -3.3‰ at 96 mm depth within the mixed mineralogy
326	zone. Immediately above the hiatus, values of around -13.8‰ rise to -12‰ at the broken
327	stalagmite top. These are lower than in modern straw stalagmites (-7‰ to -10‰), and in Dim
328	3 where values increase from between -6.5‰ and -8.0‰ at ~8.0 ka to -3.4‰ at ~6.9 ka.
329	
330	Five of the eleven isotope samples in the zone of mixed mineralogy immediately below
331	the hiatus at 87 mm dft (Fig. 2b) are aragonite and three have mixed mineralogy. Theoretical
332	calculations and experimental and empirical evidence indicate that $\delta^{18}O$ in aragonite is

333 enriched by about 0.8% relative to calcite precipitating from the same parent water

(Fohlmeister et al., 2018; Grossman and Ku, 1986; Kim et al., 2007; Tarutani et al., 1969). 334

335 Four of the five aragonites in Dim 1 are isotopically lighter than two of the calcite samples

336 and a 0.8% correction generates implausibly high data scatter compared to the coherent non-

337 corrected structure (Fig. 6a). Consequently, no adjustment has been applied to the aragonite

338 δ^{18} O values. δ^{13} C enrichments in aragonite of 1.0% - 2.5% relative to calcite have been

339 reported from several locations (Frisia et al., 2002; Holmgren et al., 2003; McMillan et al.,

340 2005; Morse and Mackenzie, 1990; Romanek et al., 1992), and recent analysis identified an

341 offset of 1.16±0.46‰ (Fohlmeister et al., 2018). However, to remain consistent with the δ^{18} O

data, the aragonite δ^{13} C values have also not been modified. 342

343

344 4.4. Strontium Isotopes

345

346	87 Sr/ 86 Sr ratios (Fig. 6c) are primarily influenced by the local limestone (87 Sr/ 86 Sr =
347	0.70716) and overlying soil (87 Sr/ 86 Sr = 0.70845), although regional sources such as sea spray
348	and far travelled aeolian dust are also potential contributors (Zhou et al., 2009) (Fig. 7).
349	Speleothem values lie on a mixing line, one end member of which is the limestone bedrock
350	and the other a composite source with a ratio higher than that of the soil (Fig. 7)
351	(Supplementary Material). Ratios in Dim 1 are comparable to those measured in the Mid-
352	Holocene Dim 3 speleothem (0.70821-0.70827) although encompassing a greater range, and
353	rather lower than modern soda straws which have a composition similar to modern overlying
354	soil. The ⁸⁷ Sr/ ⁸⁶ Sr pattern along the speleothem shows an abrupt fall between 370-260 mm
355	depth and a sharp drop at 140-115 mm immediately followed by a rapid increase (Fig. 6c).
356	
357	5. Discussion and Interpretation
358	
359	5.1. Petrography
360	
361	Columnar compact (C) fabrics are primary calcites that tend to form in thin water films with
362	slow drip rates and enhanced degassing under well-ventilated conditions (Frisia, 2015), while
363	transition to columnar open (Co) fabrics may suggest progressively less efficient degassing,
364	probably as water film depth increases (Kendall and Broughton, 1978). In temperate climates
365	C fabrics form under low (up to 0.35) CaCO ₃ saturation states and low drip water Mg
366	concentrations (Mg/Ca ratio <0.3) (Frisia and Borsato, 2010). However, the tendency toward
367	Ce fabrics also suggests that Mg/Ca ratios were at times >0.35 (Frisia, 2015) and possibly
368	above 0.85 (Gonzalez et al., 1992).
369	

370 Aragonite in the mixed-mineral axial zone shows little evidence of preserved dissolution-371 reprecipitation fabrics indicative of neomorphic alteration (Frisia, 2015; Frisia and Borsato, 372 2010; Martín-García et al., 2019). Rather, aragonite growth directly off the underlying C 373 crystals and abundant evidence of both lateral and vertical transitions from C calcite into 374 acicular aragonite (Fig. 4a) suggest concurrent competitive growth (A. Kendall, pers. com., 375 2015), i.e. co-precipitation of both polymorphs (McMillan et al., 2005; Railsback et al., 1994; 376 Spötl et al., 2002). Lateral polymorph co-precipitation, and vertical switching between 377 aragonite and calcite is likely controlled by variations in Mg/Ca ratios close to the calcite 378 inhibition threshold (Frisia et al., 2002; Riechelmann et al., 2014; Rossi and Lozano, 2016) 379 albeit probably 'conditioned' by low parent water CaCO₃ supersaturation state (De 380 Choudens-Sánchez and Gonzalez, 2009). Variable rates of groundwater recharge through the 381 dolomitised epikarst would achieve the required variations in drip water MgCa ratios. The 382 presence of acicular aragonite indicates very low drip rates (Frisia, 2015), usually indicating 383 relatively dry conditions. The predominance of aragonite, and its spherulitic morphology, in 384 the flank areas of Dim 1 relative to the axial part suggests that increasing fluid Mg/Ca ratios 385 resulting from axial calcite precipitation (Fairchild et al., 2000) may influence aragonite 386 precipitation, although enhanced evaporation of the flanking water film could also have been 387 a factor (Railsback et al., 1994).

388

The down-cutting nature of both the lower and upper boundaries to the mixed-mineral layer and the ragged aragonite crystal terminations below the upper boundary indicate corrosion and dissolution. Patchy presence of calcite microspar immediately above this boundary is probably a remnant dissolution-reprecipitation fabric (see e.g. (Frisia et al., 2002)) although the lateral impersistence of the microspar suggests that, mostly, neomorphic alteration fabrics were wholly removed by dissolution, followed by later precipitation of primary Co calcites.

395 The only other place where calcite fabrics are clearly neomorphic is in the flanking aragonites396 (Fig. 5a).

397

398 The presence of inclusions in an otherwise inclusion-poor stalagmite implies some disruption 399 to the normal growth conditions which were largely not conducive to trapping water, perhaps 400 because of almost perfect crystal coalescence on the crystal growth surfaces (Kendall and 401 Broughton, 1978). Some linear inclusion trails in C calcites are associated with relict 402 aragonite needles, which are interpreted as partial dissolution fabrics followed by calcite 403 regrowth largely as a primary precipitate, entombing aragonite relics. Dissolution of primary 404 aragonite near the growth axis was thus common, and inclusion trails contained within calcite 405 crystals adjacent to preserved aragonite layers almost certainly mark the position of former 406 aragonite layers. Typically, the inclusion-rich horizons are undulose, a geometry probably 407 deriving from unevenly corroded crystal edges. The thicker, darker, sub-horizontal lines are 408 also interpreted as dissolution surfaces as shown by the etching and corrosion of underlying 409 calcite crystal terminations. These dissolution-precipitation phases probably mainly represent 410 seasonal to sub-decadal events rather than significant breaks in growth.

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412 *5.2. U-series Systematics*

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414 5.2.1 Age Model
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The age inversions (Table 1, Fig. S3) most likely result from U-series decay chain disruption due micro-scale corrosion events in the axial zone, caused by aggressive drip waters, which have been identified at several horizons. Five samples (the 52-series in Table 1) were taken from discrete pristine aragonite layers on the flanks of Dim 1 which could be 420 traced into the growth axis (except 52-3 which narrowly post-dates the broken stalagmite 421 top), although some are replaced in the axial zone by a dissolution horizon where aragonite 422 has been removed. This approach was possible only in the upper part of Dim 1 where the 423 aragonite layers are sufficiently thick, and four samples lie stratigraphically above the hiatus 424 at 87 mm dft and one below (52-5). Whilst not in strict stratigraphic order, the dates all 425 overlap at 2 s.d. (average 128.3±0.4 ka), suggesting rapid speleothem growth at this time. No 426 petrographic evidence has been found indicating likelihood of isotopic disturbance in these 427 aragonites. Furthermore, differential U isotope mobility in a suite of samples in which U 428 concentrations vary by up to a factor of six would be likely to generate considerable age 429 scatter, and it is highly improbable that samples subjected to any significant disturbance 430 could still return such convergent ages. Therefore, although limited U mobility cannot be 431 discounted, the 52-series (aragonite) dates are likely to closely represent the true sample ages. 432

433 Since the 52-series (aragonite) dates establish the age of the upper part of the speleothem 434 as ~ 128 ka (Table 1), it follows that the stalagmite must be older than this below 96 mm 435 depth and that the ten dates younger than 127.2 ka (the lower 2σ limit of 52-2) can 436 confidently be rejected (Fig. S3). Significantly, these ten dates are from sample locations 437 which petrographic evidence of linear surfaces and inclusion planes show to have been 438 subject to episodic corrosion (Table 1). Sample 29-9 (136.2 ka), which is clearly an outlier, is 439 also rejected. An age model has therefore been constructed from the remaining fourteen dates 440 using StalAge (Scholz and Hoffmann, 2011) and this additionally rejects 37-3 (130.5 ka) as 441 an outlier (Fig. 8). No geochemical or petrographic grounds have been identified on which to 442 reject any of the final 13 dates used in the age model, which is considered to reliably 443 represent the speleothem chronology within the errors of the method. Following a period of slow growth (~30 mm ka⁻¹) from ~132 ka, a very rapid growth rate of ~400 mm ka⁻¹ is 444

445 implied for the upper part of the speleothem, almost as fast as the rates recorded in

speleothem CC5 from Corchia Cave at around 128 ka (Drysdale et al., 2009). The duration of the prominent hiatus at 87 mm depth cannot be resolved by the aragonite dates and hence was less than the 2σ dating errors (~0.7 ka, see also Section 5.5) and it has been disregarded in the construction of the age model.

450

The general validity of this model is supported by evidence from $(^{234}U/^{238}U)_0$ ratios (Fig. 9) 451 452 which fall from ~ 2.0 near the base of the speleothem to a plateau of ~ 1.4 at about 168 mm 453 depth, similar to the mid-Holocene values of Dim 3 (1.284 to 1.302). Although some calculated ages are in error by up to $\sim 8\%$ (Table 1), resultant $(^{234}U/^{238}U)_0$ values are modified 454 455 by <1%. This declining pattern resembles that from speleothems from Israel that grew over the past 25 ka and which showed simultaneous reductions in both $(^{234}U/^{238}U)_0$ and $\delta^{18}O$ as 456 457 rainfall increased through the transition from the last glacial into the Holocene (Kaufman et 458 al., 1998). A similar correspondence is seen in Dim 1 (Fig. 9), which is consistent with 459 speleothem growth commencing at a time when groundwater was passing through weathered 460 but unleached soils and epikarst after a dry phase, followed by preferential removal of ²³⁴U 461 from recoil-damaged sites under wetter conditions, leading to a subsequent gradual reduction in $(^{234}U/^{238}U)_0$. $\delta^{18}O$ data from Corchia Cave (Drysdale et al., 2009) and Tana che Urla Cave 462 463 (TCU) (Regattieri et al., 2014) show that warm and wet conditions were established in 464 southern Europe by ~131.5 ka and stable isotope values in Dim 1 when growth commenced 465 are similar to those recorded from Dim Cave for the Pleistocene-Holocene transition (Unal-466 Imer et al., 2015). This suggests that Dim 1 started growing at a time of climatic transition 467 and the basal date of ~132 ka is compatible with that inference and also with the observed 468 pattern of (²³⁴U/²³⁸U)₀ evolution, since Marine Isotope Stage (MIS) 6, like MIS 2, was a 469 period of relative aridity.

470

471 5.2.2. U-Th Open System Processes

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473 The U-Th isotope disruption in Dim 1 is most likely related to redistribution of high U 474 concentrations from discrete aragonite horizons following their partial dissolution soon after 475 deposition, and a notable feature is that nine of the eleven rejected ages are lower than 476 predicted by the age model. An investigation into the impact of diagenetic alteration, 477 including post-depositional dissolution, on U-Th speleothem ages has identified 478 circumstances in which infiltration waters could potentially corrode speleothem carbonate 479 (Scholz et al., 2014). These are (1) mixing corrosion caused by two or more saturated 480 solutions becoming under-saturated w.r.t. calcite on mixing; (2) pCO_2 in the cave atmosphere 481 being higher than in the soil zone; (3) seepage waters failing to reach saturation w.r.t. 482 carbonate due to high infiltration rates; (4) uptake of CO_2 by seepage waters from the cave 483 atmosphere following their isolation from the soil atmosphere under closed system 484 conditions. The first two conditions require a cave to be poorly ventilated, which is unlikely 485 in this case since Dim Cave has a relatively large natural entrance. But condition (3), and 486 perhaps (4), could occur periodically on decadal to centennial timescales in the wet winter-487 dry summer climate of the Eastern Mediterranean. Speleothem surfaces could suffer leaching 488 by aggressive drip waters following either torrential autumn-winter rains, or substantial 489 lowering of groundwater storage levels in the epikarst. Some recent studies have examined 490 the effects of later diagenetic replacement of aragonite by calcite on U distribution 491 (Domínguez-Villar et al., 2017; Martín-García et al., 2019), and the likely role of micro-voids 492 as pathways for U migration during aragonite to calcite conversion was identified in a 493 speleothem from Corchia Cave, northwest Italy (Bajo et al., 2016). There, modelling showed 494 that early conversion of even small percentages of (high U) aragonite to (low U) calcite in

LIG speleothems can increase apparent ages by $10^3 - 10^4$ ka. In contrast, the rejected dates in 495 496 Dim 1 are mainly younger than their true ages as predicted by the age model. Microprobe 497 analyses of speleothems from a cave in the Pyrenees documented U redistribution during 498 aragonite to calcite transformation, and U/Th dating returned ages both older and younger 499 than expected, possibly implicating thorium migration (Ortega et al., 2005). An age 10 ka less 500 than predicted was also recorded in a mixed-mineral speleothem from Morocco at an 501 aragonite-calcite transition (Wassenburg et al., 2012). Despite these valuable studies, the 502 behaviour of U-Th isotopes under open-system conditions in speleothems remains poorly 503 understood and the specific processes controlling isotopic redistribution in Dim 1 are unclear, 504 although early aragonite dissolution is strongly implicated. No replacement of aragonite by 505 calcite is evident from petrographic analysis, and U loss is unlikely to be a significant factor 506 given the dominance of lower, rather than higher, calculated ages relative to age model 507 estimates.

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509 5.3. Controls on oxygen and carbon isotopes

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511 5.3.1. Oxygen isotopes.

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513 Climatic controls on speleothem δ^{18} O are complex (Lachniet, 2009; McDermott, 2004) 514 particularly at times of global transition from glacial to interglacial states when the ocean-515 atmosphere system undergoes significant reorganisation, as during the period of Dim 1 516 deposition. Mediterranean stalagmites typically record decreases of 1‰ to 4‰ through this 517 time period (Bar-Matthews et al., 2003; Drysdale et al., 2009; Regattieri et al., 2014), 518 dominated by the negative δ^{18} O trend in ocean source water (Marino et al., 2015; Rohling et 519 al., 2015), and increasing rainfall (the "amount effect") as a consequence of a strengthening

520 hydrological cycle (Bar-Matthews et al., 2003; Bard et al., 2002; Dansgaard, 1964; Drysdale 521 et al., 2009; Regattieri et al., 2014; Rowe et al., 2012; Unal-Imer et al., 2015; Zanchetta et al., 522 2014). Modelling studies have identified a particularly strong connection between higher SST 523 in the EM and increased precipitation over the Anatolian Peninsula (Bozkurt and Sen, 2011). 524 Once marine surface water δ^{18} O values stabilised at interglacial values, the amount effect became the principal driver of high frequency variability in speleothem δ^{18} O signals. In a 525 526 climate with strongly seasonal rainfall, fluctuating precipitation amounts in winter are likely to dominate Mediterranean speleothem δ^{18} O records, which therefore become sensitive 527 528 recorders of changes in local water balance. Periods of reduced winter rainfall generate 529 isotopically enriched groundwater and elevated δ^{18} O in speleothem carbonate, and may create 530 a negative water balance in semi-arid regions leading to evaporation in the unsaturated zone 531 and enhanced isotopic enrichment (Markowska et al., 2015). Winter rainfall amounts in 532 Turkey are strongly influenced by depression track trajectories, which are steered by regional pressure patterns (Dirican et al., 2005; Kutiel et al., 2002; Saris et al., 2010; Türkeş and Erlat, 533 534 2003), and hence Eastern Mediterranean speleothem δ^{18} O profiles are related to changes in 535 regional atmospheric circulation. Changes in air mass trajectories have been invoked as long 536 term controls on isotopes in precipitation in the Eastern Mediterranean (Unal-Imer et al., 2015). 537

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539 5.3.2. Equilibrium Deposition

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541 The average δ^{18} O composition of a large pool, the cave lake and drip waters from five sites 542 collected during 2009-2010 was -5.7±0.3‰ _{VSMOW} (Fig. S2). This compares with -6.2‰ for 543 water from a large spring just below the cave, which probably receives flow from higher 544 catchments inland from the cave. The predicted equilibrium value for aragonite precipitating

545 from the cave waters at 18-19°C is about -5.7‰_{PDB} (Kim et al., 2007), whereas the average 546 δ^{18} O compositions of two active aragonite straw stalactites from Dim Cave are -4.2±0.1‰ PDB (n=6) and -4.4 \pm 0.1‰_{PDB} (n=6). When plotted as 1000ln α vs 10³T⁻¹, the stalactite data lie 547 548 above aragonite-water equilibrium lines which have been determined experimentally 549 (Grossman and Ku, 1986; Kim et al., 2007) (Fig. S5a). Empirical data from speleothem 550 calcite-water measurements (Tremaine et al., 2011) are not consistent with the experimentally 551 derived calcite-water equilibrium relationship commonly used in speleothem studies (Kim 552 and O'Neil, 1997), and it may be the case that water-carbonate fractionation factors for 553 speleothems are higher than the calculated experimental values (Coplen, 2007; Dietzel et al., 554 2009) (Fig. S4A). It has been proposed that a "window of equilibrium" may exist for 555 aragonite deposits in the zone $\pm 0.5\%$ either side of computed aragonite-water equilibrium 556 lines (Lachniet, 2015). The Dim stalactites are enriched by ~1.4‰ and therefore lie beyond 557 this limit (Fig. S5B) and modern aragonite deposits in Dim Cave appear not to be forming in 558 equilibrium with seepage waters. Hendy Tests (Hendy, 1971) were carried out along growth 559 layers at 277 mm, 94 mm (within the aragonitic layer) and 22 mm depth. These show only weak correlations between δ^{18} O and δ^{13} C, no systematic enrichment of δ^{18} O away from the 560 growth axis and δ^{18} O variability along growth layers not exceeding 0.6% (Fig. S6) and may 561 562 suggest that kinetic fractionation during the early LIG, under different hydrological 563 conditions, was rather limited. The weaknesses of this test are, however, widely 564 acknowledged (Dorale and Liu, 2009; Fairchild et al., 2006; Lachniet, 2009, 2015) and it is 565 probable that proximity to equilibrium deposition varied temporally as hydrological 566 conditions changed.

567

568 5.3.3. Carbon isotopes.

569

570 δ^{13} C values in speleothems are strongly influenced by the intensity of biogenic activity in 571 the soil zone above the cave, but may be modified by processes within the epikarst prior to 572 the emergence of seepage water into the cave atmosphere (Fairchild et al., 2006; McDermott, 573 2004). Root respiration, organic matter decay and microbial activity in densely vegetated soils generate CO₂ containing isotopically light carbon, which in areas dominated by C3 574 plants will typically yield speleothem δ^{13} C values of -8‰ to -14‰ (Dorale et al., 1992; 575 576 Hendy, 1971). Less negative values (-6‰ to +2‰) may indicate reduced biogenic-derived 577 CO₂ due to sparser vegetation cover and/or a greater contribution from isotopically heavier 578 atmospheric CO₂ and carbonate bedrock (Genty et al., 2003) or, in certain regions, a higher 579 proportion of C4 plants (Baker et al., 2002). However, rapid CO₂ degassing of groundwater 580 and/or evaporation in air-filled voids above the cave can lead to prior calcite precipitation 581 (PCP) (Fairchild et al., 2000), also resulting in carbon isotope enrichment of calcite along the 582 deposition pathway and ultimately in higher speleothem δ^{13} C values (Frisia et al., 2011). 583 Such local, site-specific, factors can significantly increase both the frequency and amplitude 584 of carbon isotope variability in a speleothem relative to coeval oxygen isotope data, which 585 may dilute the impact and increase the ambiguity of the δ^{13} C climate signal.

586

587 5.4. The Penultimate Deglaciation in the Mediterranean

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589 Growth in Dim 1 began around ~132.0 ka, broadly coinciding with trace element evidence of 590 increasing rainfall at TCU Cave in northern Italy (Regattieri et al., 2016), and continued 591 through the latter part of the penultimate glacial-interglacial transition. The stable isotope 592 data show an erratic pattern of increasingly negative values into the early LIG (Fig. 10a,b) in 593 common with other speleothem records from the Mediterranean region, NW Europe and 594 China which document all or part of the transition (Fig. 11c,d,e). Marine SST and δ^{18} O,

595 which strongly influence speleothem δ^{18} O signals, also show marked variability over this 596 time period (Fig. 11f,g,h). There are inherent difficulties in integrating marine and 597 speleothem chronologies, and thus in directly correlating terrestrial and marine records 598 (Govin et al., 2015), but the strong hydrological links between marine source waters and 599 speleothems have been utilised to synchronise land and marine archives (Drysdale et al., 600 2009), and to refine marine chronologies (Jimenez-Amat and Zahn, 2015; Marino et al., 2015). Between ~132.1 ka and ~130.2 ka, δ^{18} O values in Dim 1 oscillate between -5.5‰ and 601 602 -6.5% on centennial timescales with declining amplitude (Fig. 10a), whilst δ^{13} C values show 603 less variability and lie mainly between -8‰ and -10‰ (Fig. 10b), neither record showing any strong trend. Data from ODP976 show Mg/Ca SSTs oscillating between 14°C and 18°C (Fig. 604 605 11f), and G. Bulloides δ^{18} O fluctuating by ~0.5‰ between 132.0-130.5 ka superimposed on a 606 strong negative trend starting at ~133.0 ka (Fig. 11h) (Jimenez-Amat and Zahn, 2015). The 607 instability evident in the marine data may relate to cold water incursions into the 608 Mediterranean from the Atlantic during H11 (Jimenez-Amat and Zahn, 2015). Variability of 609 ~4°C in SSTs would strongly influence regional rainfall amounts, the isotopic composition of 610 which would initially be controlled by that of marine surface water. The presence of thin 611 aragonite layers within the predominantly calcitic Dim 1 implies periodic increases in 612 groundwater residence times and lower drip rates (Frisia and Borsato, 2010) and supports the 613 isotopic evidence for periodic fluctuations in effective rainfall amounts at this time in the 614 Eastern Mediterranean.

615

Between ~130.2 ka and ~129.4 ka on the Dim timescale, increases in δ^{18} O and δ^{13} C values of up to 0.9‰ and 3.5‰ respectively are associated with a shift in the growth axis and indicate a period of drier conditions which may be related to the ~4°C temperature drop recorded in the marine Mg/Ca data at ~130.6 ka (Jimenez-Amat and Zahn, 2015), towards the

620 end of H11 (Fig. 11f). Contemporaneous pollen records from Lake Van in eastern Turkey 621 (Pickarski et al., 2015) demonstrate increasing abundance of *Pistacia* between 131.2-129.1 622 ka, indicating dry summer and mild winter conditions. After 129.4 ka, δ^{18} O and δ^{13} C values 623 fall by 1.5% and 2.0% to -7.5% and -12% respectively, although these trends are erratic and brief positive excursions recur, especially in the δ^{18} O record at ~128.7 ka. The decreases in 624 Dim 1 δ^{18} O and δ^{13} C coincide with strong δ^{18} O negative trends in Corchia Cave (Drysdale et 625 626 al., 2009) and Soreq Cave speleothems (Bar-Matthews et al., 2003). Depletions of ~0.5‰ in 627 G. Bulloides δ^{18} O (Jimenez-Amat and Zahn, 2015) (Fig. 11h) imply some influence from a 628 marine moisture source, but nevertheless the Dim 1 oxygen and carbon data taken together 629 indicate an increasingly wetter and more densely vegetated local environment. These changes 630 correspond with the onset of the East Asian Monsoon (EAM) at the beginning of the LIG 631 which is recorded at 129.0 ka in China by large abrupt negative δ^{18} O shifts in speleothems D4 632 from Dongge Cave (Kelly et al., 2006) (Fig. 11d) and SB25 from Sanbao Cave (Cheng et al., 633 2009a). However, the continued presence within this phase of transient positive stable isotope 634 excursions in Dim 1 demonstrates that the brief drier intervals remained a characteristic of 635 climate in the Eastern Mediterranean at this time.

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At 130 ka -129 ka a general increase in extension rates occurs in speleothems Dim 1 (Fig. 637 638 8), Corchia CC5, and TCU D4, (Drysdale et al., 2009; Regattieri et al., 2014) and deposition 639 of stalagmite K1 2010 commenced in Kaanan Cave, Lebanon (Nehme et al., 2015). Marine Mg/Ca and alkenone records imply rapid $\sim 5^{\circ}$ C increases in SST at ~ 129 ka (Fig. 10f,g) 640 641 (Jimenez-Amat and Zahn, 2015; Marino et al., 2015) which may have stimulated significant 642 increases in Mediterranean rainfall, narrowly predating the onset of sapropel S5 at ~128.3 ka 643 (Grant et al., 2016). The wetter phase at Dim Cave persisted for several hundred years until 644 ~128.6 ka when δ^{18} O and δ^{13} C values increase rapidly but irregularly to -5.3% and -3.5%

645 respectively (Fig. 10a,b), implying strong reductions in groundwater recharge and soil 646 productivity. Such elevated δ^{13} C values and the switch from calcite to aragonite during 647 maximum oxygen and carbon isotope enrichment indicate increases in PCP and Mg/Ca ratios 648 as a consequence of longer groundwater contact time within dolomitic bedrock. Aragonite 649 deposition was rare in Dim Cave through the last glacial-interglacial cycle but did occur 650 between 80-75 ka, at which time δ^{18} O and δ^{13} C values peak at -3.5‰ and -5.0‰ respectively 651 (Unal-Imer et al., 2015; Ünal-Imer et al., 2016), comparable to those recorded in the 652 aragonite of Dim 1.

653

654 ⁸⁷Sr/⁸⁶Sr values experienced a two-phase decrease, separated by a slight recovery, at 655 ~128.7 ka. The initial fall coincides with the negative trends in δ^{13} C and δ^{18} O signifying 656 increasing precipitation, and the second with the strong positive stable isotope excursions and 657 change to aragonite deposition, indicative of drier conditions (Fig. 10c). The first phase may 658 be analogous to the significant drop in Sr isotope values seen in marine core ODP 658C at 659 \sim 12.5 ka following a reduction in the dust supply as the African Monsoon strengthened at the 660 beginning of the Holocene African Humid Period, (Cole et al., 2009). The expansion of 661 vegetation across North Africa at the beginning of the LIG following intensification of the Asian and African Monsoon circulations at 129 ka would similarly have reduced long range 662 aeolian dust transport at this time. Alternatively, lower ⁸⁷Sr/⁸⁶Sr ratios may derive from 663 664 increased groundwater infiltration and intensified leaching within the epikarst, although this 665 might be partially counteracted by simultaneous release of more radiogenic Sr from the soil zone (Fig. 7). The second phase of ⁸⁷Sr/⁸⁶Sr decrease, during the period of oxygen and carbon 666 667 isotopic enrichment, is compatible with increased groundwater residence times and 668 consequent increase in water-bedrock interaction following substantial reductions in annual 669 recharge.

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671 Some regional pollen data also contain evidence of a drier phase at ~128 ka, although 672 chronologies for these records are less secure (Govin et al., 2015; Zanchetta et al., 2016). At 673 Lago Grande di Monticchio (LGdM), in southern Italy, arboreal pollen abundances decrease 674 between 128.15 ka and 127.90 ka (Allen and Huntley, 2009; Brauer et al., 2007), and a brief 675 (decadal to century-scale) reduction in temperate tree pollen occurs at Tenaghi Phillipon 676 (TP), northeast Greece, at ~128.4 ka just prior to arboreal-dominated taxa reaching fully 677 interglacial abundances (Milner et al., 2012). However, no equivalent event is seen at 678 Ioannina, northwest Greece (Tzedakis et al., 2003). Marine core MD95-2042 on the Iberian 679 Margin (Sanchez Goni et al., 1999) records peak abundance of warm humid-temperate pollen 680 occurring early in the LIG, preceded by a "Younger Dryas-like event". The precise date of 681 this event is uncertain, but it probably lies between 128 ka and 129 ka (Govin et al., 2015) 682 and may correlate with the transient declines in arboreal pollen at LGdM and TP. A decline 683 in temperate tree pollen is also recorded at this time in core MD01-2444 from the Portuguese 684 Margin where it is correlated with cold water event 28 (C28) at \sim 128.7 ka (Tzedakis et al., 685 2018).

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The oxygen, carbon and strontium isotope excursion peaks in Dim 1 are immediately 687 688 followed by the prominent hiatus, which occurs as the isotopic indicators signal increasing 689 moisture availability (Fig. 6). The break in growth may denote a second, more severe, phase 690 of rainfall reduction which caused growth to stop, following which renewed heavy rainfall 691 and rapid infiltration of under-saturated groundwaters eroded the stalagmite cap. 692 Alternatively, carbonate erosion may have occurred, without prior cessation of growth, due to 693 a sudden intensification of rainfall which substantially increased flow rate through the 694 epikarst, and forced under-saturated, aggressive, seepage water into the cave. In either case,

695 the brief time-interval represented by non-deposition (see below) suggests complete or partial 696 removal of ~1-3 mm of aragonite in the axial zone. Above the hiatus, calcite deposition 697 recommenced along an offset axis, with strong negative trends in both δ^{18} O and δ^{13} C 698 reaching minimum values of -7.8‰ and -13.8‰ respectively at ~128.5 ka. These are 1-2‰ 699 more negative than early Holocene values (Unal-Imer et al., 2015), implying wetter 700 conditions and well-developed soil and dense vegetation cover above the cave at that time. 701 ⁸⁷Sr/⁸⁶Sr ratios also reach values similar to, or slightly higher, than before the dry phase, 702 probably due to an increased soil contribution. Stable isotope data from LIG speleothems 703 generally show that minimum values, indicating maximum moisture availability, are attained 704 at ~128 ka, at the beginning of the interglacial. In the Mediterranean this pattern is seen in 705 Corchia Cave (Drysdale et al., 2009) and Soreq Cave (Bar-Matthews et al., 2003), and also in 706 China, at Dongge Cave (Kelly et al., 2006). The Dim 1 stable isotope data broadly form the 707 same pattern as seen in other contemporary speleothems, but appear to reach minimum values 708 ~ 0.6 ka before those in other speleothems (Fig. 11a-e). It is highly unlikely that the 709 geochemical signals in this speleothem could be out of phase with those in all other 710 speleothems across the region and it is probable that the age model over-estimates ages 711 towards the top of Dim 1 by a few hundred years. Detailed comparison of δ^{18} O structure 712 through the positive excursion with speleothem data from France and Austria (Section 5.5) 713 supports this interpretation. Subsequent increases of about 1‰ and 1.5‰ in δ^{18} O and δ^{13} C 714 respectively, trends widely seen in other contemporary speleothems (Fig. 11c-e), are likely to 715 document reducing precipitation.

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717 5.5. Nature and origin of the positive isotopic anomaly

The rapid increases in δ^{18} O and δ^{13} C values and abrupt fall in 87 Sr/ 86 Sr between 128 ka and 719 720 129 ka, culminating in a switch from calcite to alternating aragonite-calcite precipitation, 721 clearly record a major environmental change in Dim Cave. A change from calcite to 722 aragonite mineralogy in a speleothem is generally a strong indicator of reducing effective rainfall (Railsback et al., 1994; Wassenburg et al., 2012) and the isotopic evidence supports 723 this interpretation. The large amplitude of the δ^{18} O anomaly (~2.0‰) is similar to that 724 725 reported from Soreq Cave for the Younger Dryas (YD) (Bar-Matthews et al., 2003) and to the 726 shift from YD to early Holocene values in speleothem Dim-E2 (Fig. S7) (Unal-Imer et al., 727 2015). However, the two events differ significantly in duration and also because YD 728 speleothem δ^{18} O signals incorporate strong moisture source and temperature effects deriving 729 from complex atmosphere-ocean changes associated with deglaciation (Baldini et al., 2015). 730 Nevertheless, a marked increase in aridity across Eurasia during the YD is widely recognised 731 (Belli et al., 2017; Brauer et al., 2008; Rach et al., 2014; Wang et al., 2001), although the 732 presence of drier conditions in the Eastern Mediterranean at that time is debated (Hartman et 733 al., 2016), and the Dim 1 and YD events may, therefore, have a similar origin, namely a 734 cooling of the North Atlantic and slowdown of the MOC (Ritz et al., 2013), the impacts of 735 which were most evident in winter.

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Early LIG δ^{18} O enrichment episodes also occur in European speleothems, most prominently in BD Cave in southwest France (Couchoud et al., 2009) and Schneckenloch Cave, Austria (Moseley et al., 2015) where stalagmites BD-Inf and SCH-5 record increases of 0.85‰ and 1.60‰ respectively in δ^{18} O values between 128.4 ka and 128.0 ka (Fig. 12). Within the 2 s.d. dating errors, these are compatible with the ~128.6 ka age of the positive δ^{18} O and δ^{13} C peaks in Dim 1 and there are strong similarities in the structures of these isotopic events which suggest that they probably have a common origin. Consequently, it is

744 unlikely that the isotopic and petrographic shifts seen in Dim 1 can be attributed to cavespecific factors. Transferring this part of the δ^{18} O record to the more robust BD timescale 745 746 (Fig. 12) using prominent isotopic features as tie points (Fig. S8), the ~100 year timespan of 747 the main positive peak in BD Cave provides an estimate of about 20 years for the missing 748 aragonite deposition represented by the Dim 1 hiatus. The equivalent phase of the 749 Schneckenloch Cave event also lasted about 100 years (Fig. 12). Since the hiatus in Dim 1 750 occurs during the negative-trending limb of the excursion and neither BD-Inf nor SCH-5 show a reversal of that trend in the equivalent location, it is unlikely that growth in Dim 1 751 752 ceased due to drip-water starvation prior to carbonate corrosion. Whilst the Dim and BD 753 oxygen isotope signals are interpreted as recording fluctuations in rainfall amounts, the 754 Schneckenloch data are considered primarily to record changes in local atmospheric 755 temperature (Moseley et al., 2015). Evidently the Northern Alps experienced a warming 756 event at a time of reduced rainfall on the Atlantic seaboard and in the Eastern Mediterranean, 757 possibly due to northward advection of warm Mediterranean air carrying isotopically 758 enriched water vapour during a temporary weakening of the zonal westerly flow. 759 Alternatively, the positive δ^{18} O anomaly might represent a change in rainfall seasonality from 760 a winter to a summer maximum, perhaps as a consequence of reduced winter cyclogenesis. 761

The identification of a cool dry phase in southwest France (Couchoud et al., 2009), coeval within error with the Dim 1 isotopic enrichment event, suggests that the origin of that event lies in the North Atlantic rather than within the Mediterranean. Variations in North Atlantic ocean circulation, particularly the MOC, are strongly linked to SSTs which in turn influence moisture supply downstream over Eurasia. Speleothem stable isotope data from the Eastern Mediterranean document abrupt changes in precipitation amounts through the last glacial period related to variations in sea ice cover and SSTs in the North Atlantic (Bar-Matthews et

769 al., 2003; Drysdale et al., 2009; Rowe et al., 2012; Unal-Imer et al., 2015). A 3°C SST drop 770 occurs in the ODP976 Mg/Ca record at ~128.3 ka (Fig. 11f) which has been likened to a 771 Younger Dryas-type event (Jimenez-Amat and Zahn, 2015), although a similar drop is not 772 seen in the alkenone record and Mediterranean SSTs remained well above values recorded 773 between 132 ka - 129 ka during which time Dim 1 was mainly precipitating calcite. The 774 Mg/Ca data may, however, represent an attenuated signal from a larger North Atlantic event. 775 A recent study (Tzedakis et al., 2018) has attempted to correlate positive U/Th-dated δ^{18} O 776 excursions in Corchia Cave speleothem with reductions in arboreal pollen abundance in 777 marine core MD01-2444 from the Portuguese Margin and subsequently to correlate the 778 stratigraphy of that core with others from the North Atlantic containing evidence of cold 779 water incursions (Galaasen et al., 2014; Mokeddem et al., 2014; Nicholl et al., 2012). A 780 strong association is found between North Atlantic cooling, disruption of the MOC and arid 781 phases in the Mediterranean as manifested by reduced tree pollen abundances and positive 782 δ^{18} O shifts in the Corchia Cave speleothem records. One such cold water incursion (C28) 783 with a duration of ~ 300 years is identified at ~ 128.7 ka, close to the ages assigned to the Dim. 784 BD and Schneckenloch Cave isotopic enrichments. This event provides a potential forcing 785 mechanism for modifying the mid-latitude westerly circulation pattern and inducing winter 786 rainfall reductions across Northwest Europe and the Mediterranean.

787

The duration and structure of the Dim event are strikingly similar to the well documented Holocene 8.2 ka event recorded in Greenland ice cores (Fig. S9) and in speleothems from China, Brazil and Oman (Cheng et al., 2009b), although rather muted in Mediterranean records. That event may have resulted from an outburst flood from glacial Lake Agassiz into the Labrador Sea, or from the collapse of the Hudson Bay ice saddle (Matero et al., 2017), which temporarily disrupted the North Atlantic MOC, the impact of which persisted in the

794 isotope records for ~200-300 years (Cheng et al., 2009b). The origin of the C28 event is not 795 known but is presumably related to high latitude ice melting, and in this context the poly-796 phase isotope pattern and duration are interesting as it occurs near the beginning of the LIG rather than towards the middle, as in the Holocene. There appears to be less evidence for 797 798 widespread global cooling early in the LIG compared to that available for the Holocene 8.2 799 ka event, which may reflect an absence of persistent or extensive NA winter sea ice during 800 the particularly warm LIG (Galaasen et al., 2014). Sea ice effectively suppresses winter 801 cyclogenesis and has a strong cooling effect downstream as illustrated by weaker Asian 802 Monsoon circulation during glacial periods and Heinrich Stadial Events, but colder SSTs in 803 the absence of sea ice have much less impact on atmospheric circulation patterns. The 804 speleothem evidence is compatible with a cold but largely ice-free North Atlantic leading to 805 winter rainfall reductions in NW Europe and the Mediterranean.

806

807 **6.** Conclusions

808

809 Stable isotope records from stalagmite Dim 1, which grew through the latter part of TII 810 between ~132 ka and ~128 ka, reflect a combination of changes in marine boundary 811 conditions and increases in precipitation as the hydrological cycle strengthened in response to rising global temperatures. Strongly negative δ^{18} O values indicate that the onset of the LIG in 812 813 southern Turkey was wetter than the early Holocene, supporting previous evidence from 814 contemporary Mediterranean stalagmites. Speleothem growth in Dim Cave appears 815 particularly sensitive to fluctuations in seasonal groundwater recharge and episodes of 816 aragonite deposition and subsequent partial removal by under-saturated drip waters show that 817 there were frequent brief drier cycles followed by rapid infiltration of renewed heavy rainfall. 818

A phase of strong oxygen and carbon isotope enrichment and lower Sr isotope values, lasting for 200-300 years and centred around 128.1 ka, incorporates a switch from calcite to alternating aragonite/calcite deposition, demonstrating a more severe and prolonged reduction in (winter) precipitation. Rainfall amounts fell from significantly above to rather below present day totals. This drier phase is succeeded by the wettest conditions in the record, signified by minimum δ^{18} O and δ^{13} C values, causing rapid groundwater infiltration and corrosion of the stalagmite upper surface.

826

827 The dating and structure of the positive isotopic anomaly suggest that it correlates with 828 positive oxygen isotope peaks in stalagmites from Bourgeois-Delaunay Cave, southwest 829 France and Schneckenloch Cave, Austria. These peaks are probably associated with cold 830 water event C28 in the North Atlantic and their poly-phase isotopic structures strongly 831 resemble Northern Hemisphere speleothem δ^{18} O records of the Holocene 8.2 ka event which 832 was also caused by an outburst of glacial freshwater into the North Atlantic. The ~128 ka 833 event it is not widely recognised in terrestrial archives within the Mediterranean Basin or 834 globally, possibly because the temporal resolution of many climate records is inadequate to 835 capture such a brief episode.

836

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838

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848

- 849 Appendix A. Datasets and Supplementary Information
- 850 Supplementary data for this article can be found online at:

851

852 **References**

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Figure Captions

Figure 1. (a) Location of sites referred to in the text: 1. Bourgeois-Delaunay Cave; 2. Schneckenloch Cave; 3. Core MD01-2444; 4. Core ODP976; 5. Corchia Cave and Tana che Urla (TCU) Cave; 6. Lago Grande di Monticchio; 7. Tenaghi Phillipon; 8. Dim Cave; 9. Soreq Cave. (b) Location of Dim Cave; note proximity to coast; (c) Plan of Dim Cave (http://www.dimcave.com.tr/magara.htm).

Figure 2. (a) Section through stalagmite Dim 1. Solid line follows the corrosion surface and dashed line the onset of mixed mineralogy (see below). Dotted lines follow stable isotope sampling tracks. Note occasional shifts of growth axis. Filled ovals show locations of U/Th dated samples. Open red ovals are locations of dated samples from a different face projected onto this section. Black rectangle encloses the scanned area of an adjacent slab shown in fig. 2b. (b) Scanned slab of the upper 230 mm of Dim 1, offset from the central growth axis. The darker grey colours are calcite, while the paler, cream colours are either aragonite or mixed aragonite/calcite. Note the clear stalagmite top morphologies at 131 mm dft. A prominent band of paler, creamy carbonate (mixed aragonite and calcite) is present (white arrow) between 103 and 87 mm dft. The upper surface of this band displays a clear corrosion surface (CS) and above this surface (RHS of image) there is a 10 mm zone where aragonite near the flanks (black arrow) appears to be replaced by darker calcite in the axial zone. The growth axis is offset (GAO) by ~20 mm above the corrosion surface. Flanking layers are mainly aragonite mineralogy (AFL).

Figure 3. Dim 1 thin section photomicrographs; all are crossed polarised images with scale bars of 500 µm length. a) Columnar compact (C) calcite, typical of the lower part of the stalagmite. The primary calcite fabrics are inclusion poor with length to width ratios approaching elongated columnar (Ce) fabric. b) Sharp lower boundary (axial part ~103 mm) to zone of mixed aragonite and calcite showing bundles of acicular aragonite that nucleated directly on underlying columnar crystal terminations. c) As b) showing spaces between aragonite bundles filled with non-orientated aragonite needles. White arrow points to 1 mm high 'step' in the boundary, formed by dissolution, where C calcite shows a sharp change to aragonite needle fabrics (right of arrow tip). d) Detail of mineralogical change shown in c) where the near vertical dissolution surface cuts primary C calcites, and primary aragonite needle fabrics have grown adjacent to the sub-vertical surface.

Figure 4. Dim 1 thin section micrographs. a) Crossed polarised image of vertical transitions from calcite (c) to aragonite needle fabrics (a) in the zone of mixed aragonite and calcite. There is little evidence for neomorphism and the fabrics are interpreted as indicative of syndepositional switching of primary mineralogies; scale bar is 500µm. b) Crossed polarised image of mostly aragonite needle fabrics in the upper part of the zone of mixed aragonite and calcite (upper left) into aragonite. The boundary between the top most aragonite layer and the overlying C calcites can be seen at top right of the image; scale bar is 500 µm. c) Crossed polarised image showing detail of the upper boundary between aragonite and overlying C calcites. Note the ragged terminations to aragonite needles, and presence of a zone of equant microspar (EM) directly above the boundary, and in places between aragonite needles. Faint inclusion trails are again evident in the aragonite; scale bar is 500 µm. d) Plane light image of flank zone aragonite spherulite fabrics (whole field of view) forming a clumped texture. Scale bar is 1 mm.

Figure 5. Dim 1 thin section photomicrographs. a) Crossed polarised image showing detail of irregular upper boundary (white arrows) between spherulitic aggregates of aragonite (a) needle crystals and overlying neomorphic mosaic calcites (c) in the flank zone; scale bar is 500 μ m. Plane light images of: b) linear inclusion trails in Co calcites making prominent dark lines and layers; c) spherical fluid inclusions in a linear trail; d) inclusion trails associated with relict aragonite needle fabrics (arrow); and e) dark sub-horizontal layers defining terminations of underlying Co calcite crystals. Scale bars are 500 μ m.

Figure 6. (a) δ^{18} O, (b) δ^{13} C and (c) 87 Sr/ 86 Sr data vs. distance from top of Dim 1. Heavy lines in (a) and (b) are 5-point running means. Vertical dashed line marks the position of the corrosion surface. Vertical grey shading shows region of mixed aragonite-calcite mineralogy. Heavy vertical black bars in (a) and (b) show range of modern values measured in active aragonite straw stalactites.

Figure 7. 87 Sr/ 86 Sr values of likely sources contributing to the Dim 1 signal. Saharan dust, with a value of ~0.7200, is not plotted. The upper end member represents the combined composition of all input components except limestone bedrock (Supplementary Material).

Figure 8. Age - depth relationship derived from StalAge (Scholz and Hoffmann, 2011) using 14 dated samples shown by petrographic analyses not to have experienced post-depositional modification (see Table 1 and text for discussion). Horizontal dashed line shows location of hiatus. ♦ denotes aragonite samples.

Figure 9. Relationship between $(^{234}\text{U}/^{238}\text{U})_0$ and $\delta^{18}\text{O}$ along the Dim 1 growth axis. Uranium isotope ratio errors are within symbol perimeters. Trend line is a fifth order polynomial to highlight the general data pattern. Vertical axes are inverted.

Figure 10. (a) δ^{18} O, (b) δ^{13} C and (c) 87 Sr/ 86 Sr data plotted on the Dim 1 U-series timescale. Heavy line is 5-point running mean. **Figure 11.** Relationship of Dim 1 δ^{18} O and δ^{13} C records to other contemporaneous paleoclimatic archives encompassing the penultimate glacial-interglacial transition. Records are plotted on their own timescales. (a) Dim 1 δ^{18} O record; black bar represents typical 2 σ errors on U-Th dates ($\pm \sim 0.7$ ka); (b) Dim 1 δ^{13} C record; (c) δ^{18} O profile from speleothem CC5, Corchia Cave, N.W. Italy (Drysdale et al., 2009); (d) δ^{18} O record from stalagmite D4, Dongge Cave, China (Kelly et al., 2006); (e) δ^{18} O record from Bourgeois-Delaunay Cave, southwest France (Couchoud et al., 2009); (f) Mg/Ca SST record from marine core ODP976, Alboran Sea (Jiminez-Amat and Zahn, 2014); (g) Alkenone SST record from marine core ODP976 (Marino et al., 2015); (h) δ^{18} O G. Bulloides record from marine core ODP976 (Jiminez-Amat and Zahn, 2014). Blue vertical line: termination of Heinrich Event 11 at 130 ka (Marino et al., 2009); Green line: base of sapropel S5 at 128.3 ka (Grant et al., 2016). Black dashed lines link the Dim 1 positive δ^{18} O event to suggested correlatives in European and Chinese speleothem records.

Figure 12. Positive δ^{18} O anomalies in speleothems from Schneckenloch Cave (Moseley et al., 2015), Dim Cave (this study) and BD Cave (Couchoud et al., 2009). Schneckenloch and BD data are plotted on their own timescales; Dim data has been tuned to the BD timescale using δ^{18} O peaks in the isotopic profiles as tie points (Fig. S8). The events are synchronous within U-series dating errors.

















Figure 7



Figure 8



Figure 9



Figure 10



Figure 11



Figure 12

Rowe et al. Table Caption

Table 1. U-series dating results for stalagmites Dim 1 and Dim 3. Samples excluded from Dim 1 age modelling are shaded. Aragonite samples are in bold. Underlined laboratory ID numbers identify samples in which indications of corrosion have been observed in thin section petrography. Corrected ages BP are before 1950; corrections for detrital contamination are <0.1 ka. AR = Activity ratio.

Tabl	e 1	١.
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Lab ID	Distance from top [mm]	U [maa]	²³² Th [ppb]	²³⁰ Th/ ²³⁸ U AR	²³⁴ U/ ²³⁸ U AR	(²³⁴ U/ ²³⁸ U)₀ AR	²³⁰ Th/ ²³² Th AR	Age uncorrected [ka]	±2σ	Age corrected [ka BP]	±2σ
	[]	[[]]]	1919-001			Dim 1		[]		[]	
E2 2	0	1.06	0.025	0 0248/26)	1 2042/14)	1 4365/10)	3266.0	127 806	0 699	107 916	0 699
29-10	1	0.43	0.323	0.89519(24)	1 3100(15)	1 4310(10)	3811.0	117 531	0.588	117 454	0.588
<u>52-70</u>	3	3 28	0.681	0.00010(24)	1 3101(14)	1 4461(19)	13836.8	128 825	0.500	128 759	0.687
41-8	5	0.42	0.419	0.8985(24)	1.3161(15)	1 4400(19)	2748.2	117 284	0.597	117 201	0.597
52-4	11	4 66	0.279	0.9448(26)	1 3133(15)	1 4503(19)	48083 1	128 540	0.689	128 477	0.689
52-1	67	6.36	3.963	0.9400(28)	1.3106(15)	1.4456(20)	4586.8	127.899	0.741	127.825	0.741
37-5	74	0.38	0 749	0.9384(25)	1 3017(14)	1 4345(19)	1462 5	129 302	0.682	129 199	0.681
29-9	84	0.77	0.345	0.9264(27)	1.2552(14)	1.3747(19)	6309.1	136.164	0.809	136.092	0.809
41-6	91	2.50	0.179	0.9225(24)	1.3328(17)	1.4666(21)	39093.3	119.761	0.610	119.698	0.610
29-7	91	1.86	0.374	0.91834(23)	1.3286(15)	1.4605(20)	17299.4	119.544	0.579	119.479	0.579
52-5	97	4.14	0.988	0.9455(25)	1.3156(15)	1.4532(20)	12051.1	128.265	0.688	128.198	0.688
<u>41-5</u>	106	0.25	0.116	0.9227(25)	1.3160(18)	1.4467(22)	6012.2	122.776	0.659	122.704	0.659
<u>41-4</u>	144	0.17	0.190	0.9453(26)	1.3248(20)	1.4641(25)	2589.3	126.462	0.729	126.377	0.728
37-3	214	0.17	0.989	1.0216(34)	1.3980(15)	1.5751(21)	520.3	130.597	0.831	130.422	0.830
<u>33-6</u>	215	0.17	0.265	1.0029(30)	1.4024(18)	1.5734(23)	1955.9	125.531	0.729	125.439	0.729
<u>41-2</u>	265	0.15	0.136	1.0193(28)	1.4527(20)	1.6364(24)	3354.3	120.685	0.642	120.606	0.642
102-2	309	0.15	0.565	1.1388(30)	1.5509(19)	1.7928(25)	923.2	129.114	0.663	128.986	0.665
102-1	356	0.15	0.639	1.0805(29)	1.4841(19)	1.6950(24)	780.4	128.212	0.670	128.072	0.672
<u>41-1</u>	403	0.17	0.170	1.1998(32)	1.6451(23)	1.9213(29)	3565.6	126.312	0.672	126.234	0.672
33-4	437	0.16	0.450	1.2531(35)	1.6952(21)	1.9994(28)	1340.2	128.695	0.696	128.590	0.696
37-1	449	0.19	0.204	1.2540(42)	1.6904(18)	1.9949(28)	3461.8	129.518	0.791	129.439	0.791
37-4	474	0.13	0.176	1.3112(36)	1.7523(19)	2.0879(27)	2852.3	130.732	0.682	130.650	0.682
<u>33-5</u>	481	0.14	0.439	1.2191(38)	1.7162(23)	2.0050(30)	1216.6	120.081	0.681	119.973	0.681
29-6	552	0.10	0.380	1.2289(69)	1.6383(24)	1.9282(42)	1023.0	132.761	1.363	132.642	1.362
37-2	564	0.10	0.266	1.2413(42)	1.6615(18)	1.9586(28)	1430.6	131.515	0.829	131.412	0.828
Dim 3											
33-9	7	5.29	0.280	0.0799(02)	1.2780(05)	1.2836(05)	4634.3	7.004	0.017	6.939	0.017
33-10	40	5.66	0.230	0.0926(02)	1.2952(14)	1.3020(14)	6858.8	8.045	0.022	7.980	0.022

SUPPORTING ONLINE MATERIAL FOR

Climate instability during the early last interglacial recorded by multiple proxies in a speleothem from southern Turkey

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U-series methods

This File includes:

Strontium Isotope End Member Compositions

Figs S1-S9

References

Speleothem isotope data

Cave water isotope data

1. U-Series Dating Methods:

Dating was carried out at the NERC Isotope Geosciences Laboratory, Keyworth, UK. U isotope data were initially obtained on an Axiom multi-collector inductively coupled mass spectrometer (MC-ICP-MS), but the bulk of the U and Th data were obtained on a Thermo Neptune Plus MC-ICP-MS using an Aridus II desolvating nebulizer and standard-sample bracketing. 200-250 mg samples were dissolved in HNO₃, and spiked with a mixed ²²⁹Th-²³⁶U tracer and equilibrated overnight. Following oxidation of organics in 15 M HNO₃ and 30% H₂O₂, U and Th were pre-concentrated by Fe co-precipitation using a FeCl solution. Samples were loaded on cleaned and equilibrated columns in 7 M HNO₃ and Th eluted in 8 M HCl followed by U elution in 0.2 M HCl. After an initial separation on AG-1 x 8, the separated Th aliquots were further purified using a second pass through AG-1 x 8 columns,

while separated U was purified on UTEVA columns following Andersen et al. (2008). The U and Th separates were subjected to repeated oxidation steps and taken up in 1 ml 0.2 M HCl - 0.05 M HF and centrifuged prior to mass spectrometry.

U mass bias correction used measurements of CRM 112a spiked with a ²³³U/²³⁶U tracer (IRMM 3636), while SEM gain was monitored using measured ²³⁴U/²³⁵U of mass biascorrected unspiked CRM 112a analyses. Mass-bias and spike-corrected ²³⁴U/²³⁸U values of the spiked CRM 112a runs were used as a check on the SEM gain. Hydride and tailing corrections followed (Hiess et al., 2012) and were on the order of 2 ppm of the adjacent peaks and very consistent on a timescale of several days. Mass bias and SEM gain for Th measurements were corrected using an in-house ²²⁹Th-²³⁰Th-²³²Th reference solution calibrated against CRM 112a. Th isotopes were measured in static multi-collection mode with ²²⁹Th and ²³²Th measured on Faraday detectors and ²³⁰Th on an SEM. Total ²³⁸U and 232 Th blanks were <10 pg and <4 pg and were negligible relative to the sample U and Th. Standard accuracy (within 0.1%) and reproducibility (within 0.2%) of $^{234}U/^{238}U$ was monitored by replicate analyses of Harwell uraninite HU-1. Replicate measurements of the reference solution showed 229 Th/ 230 Th accuracy and reproducibility to be ± 0.2 -0.3% for 230 Th ion beams > 5000 cps. Data reduction was carried out using in-house Excel spreadsheets incorporating the revised average ²³⁵U/²³⁸U ratio of 137.818 (Hiess et al., 2012), and U-Th ages were calculated using the decay constants of (Cheng et al., 2013).

2. Strontium Isotope End Member Compositions

If the limestone host rock is considered to be one endmember of a mixing line, a regression line fitted to a plot of ⁸⁷Sr/⁸⁶Sr vs. 1/Sr concentration and projected to the 1/Sr zero intercept reveals the composition of the other end member (or the integrated composition of

multiple other sources) (Ayalon et al., 1999; Goede et al., 1998; Verheyden et al., 2000). Regression of the Dim 1 calcite Sr data and of Dim 1 and Dim 3 aragonite data converge on a ratio of 0.70864. This is more radiogenic than the soil and indicates a contribution from sea spray (87 Sr/ 86 Sr = 0.70930) and/or far travelled, probably Saharan, dust (87 Sr/ 86 Sr =0.72200, (Cole et al., 2009)). Ratios lie closer to the radiogenic end member than to the limestone and the fractional contribution of that source can be calculated from

$$E = 1 - (R_E - R_M) / (R_E - R_L)$$
(5)

Where E is the relative input from the exogenic end member, R_E is the Sr-ratio of that end member (0.70864), R_L is the Sr-ratio of the host rock (0.70716) and R_M is the measured Sr ratio of the speleothem sample. E varies from ~0.79 where Sr-ratios are highest between ~440 mm depth and the base to ~0.65 at the minimum value at 114 mm.

3. Supplementary Figures



Figure S1. Average monthly rainfall and temperature data for Antalya for period 1963 to 2004. (IAEA/WMO). Rainfall is strongly seasonal with a winter maximum.



Figure S2. Oxygen and hydrogen isotope relationships for meteoric waters. Dim Cave Data: $\delta^2 H = 7.0 * \delta^{18} O + 14.2$; Antalya Meteoric Water Line: $\delta D = 7.3 * \delta^{18} O + 12.9$ (Dirican et al., 2005); Mediterranean Meteoric Water Line: $\delta D = 8.0 * \delta^{18} O + 22$, (Gat and Carmi, 1970); Global Meteoric Water Line: $\delta D = 8.0 * \delta^{18} O + 10$, (Craig, 1961); Groundwaters from the southern side of the Taurus Mountains: $\delta D = 7.2 * \delta^{18} O + 10.8$, (Schemmel et al., 2005).



Figure S3. U-Th ages plotted against depth below stalagmite top. The dates cluster around MIS 5e but are not in stratigraphic order, implying disruption in the uranium decay chain in some samples. Boxes enclose 52-series aragonite sample ages, which are considered reliable (see text and Table 1). Dates above the horizontal line are younger than the aragonite dates near the stalagmite top and are therefore rejected from the age model, as is the date of 136 ka which is clearly an outlier.



Figure S4. Dim 1 U-series data plotted in ${}^{234}U/{}^{238}U$ vs. ${}^{230}Th/{}^{238}U$ space. All except two points fall within, or overlap at 2 σ , the space between 120 ka and 140 ka isochrons. Blue dashed line is a least squares regression through the data (R² = 0.98). 2 σ error bars lie within the data points.



Figure S5. (a) Aragonite-water equilibrium fractionation lines. Blue: (Kim et al., 2007) corrected for acid fractionation factor (Lachniet, 2015); black (Grossman and Ku, 1986). Red line is calcite-water equilibrium fractionation line (Kim and O'Neil, 1997). Purple line is empirical speleothem calcite-water equilibrium fractionation line (Tremaine et al., 2011), offset from the experimentally derived Kim and O'Neil (1997) line. Blue squares are stalactite straws from Dim Cave; (**b**) Aragonite equilibrium equations plotted as $10^3 \text{ In } \alpha \text{ vs.}$ temperature. Black line: $1000 \text{In} \alpha_{\text{aragonite-water}} = 17.88 \pm 0.13 (10^3/\text{T}) - 30.76 \pm 0.46$ (Kim et al.,
2007); blue line: $1000 \text{In}\alpha_{\text{aragonite-water}} = 18.34 (10^3/\text{T}) - 31.954$ (Grossman and Ku, 1986); red line: +0.5‰ upper boundary below which aragonite can be reasonably inferred to have precipitated in, or close to, isotopic equilibrium with its parent water (Lachniet, 2015). Black diamonds show active aragonite stalactites from Dim Cave, which appear to be precipitating out of equilibrium with seepage waters.



Figure S6. Oxygen and carbon isotope data from Hendy Tests (Hendy, 1971). A: 277 mm, B: 94 mm, C: 22 mm below top. Oxygen and carbon data are poorly correlated, oxygen variability is ≤0.6‰ and there is no systematic enrichment away from the central axis (0 on horizontal axis). Carbon: open circles, dashed line; oxygen: filled diamonds, solid line.



Figure S7. δ^{18} O and δ^{13} C records from speleothem Dim-E2 through the last glacialinterglacial transition (Unal-Imer et al., 2015). The δ^{13} C values and general structure of the record are similar to that part of Dim 1 below the positive isotope excursion. The δ^{18} O values, however, are consistently less negative by $\geq 0.5\%$ than in Dim 1 below the excursion, and the isotopic pattern has a simpler expression. The magnitude of the Dim 1 δ^{18} O positive anomaly is similar to the difference between YD and early Holocene values in Dim-E2, and that of the δ^{13} C is much larger. The YD and the Dim 1 anomaly occupy different chronological positions in the deglacial sequences and are not equivalent, but the comparison suggests that the anomaly records a significant climatic event.



Figure S8. Positive oxygen isotope anomalies in Dim 1 and BDinf plotted on their own timescales. The age offset is ~500 years and lies within the typical 2 s.d. error of the Dim 1 U/Th dates (\pm ~700 years). Strong similarities are evident between the isotopic patterns and dashed lines connect inferred congruent data points. The Dim 1 age model compresses the data into an improbably short time period around the discontinuity whereas the BDinf timescale is more realistic and also closely agrees with the Schneckenloch Cave age model (Fig. 12) (Moseley et al., 2015).



Figure S9. (a) Dim 1 δ^{18} O positive anomaly plotted on the Bourgeois-Delaunay Cave timescale (Couchoud et al., 2009); (b) Greenland ice core δ^{18} O negative anomaly at 8.2 ka (Thomas et al., 2007). Heavy line is 3-point running mean. The Greenland "8.2 ka event" is detected globally in stalagmite δ^{18} O data (Cheng et al., 2009b) as a consequence of abrupt regional changes in temperature or rainfall. The architecture of the two data sets is broadly similar and they occupy comparable timespans of ~200 years, although the Greenland event occurs later in the interglacial. Changes in North Atlantic circulation, especially the MOC, are directly implicated in the Greenland and speleothem 8.2 ka isotopic anomalies and it seems likely that similar events occurred early in previous interglacial periods.

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