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1	A Stalagmite Test of North Atlantic SST and Iberian Hydroclimate Linkages over the Las
2	Two Glacial Cycles
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### Abstract

Close coupling of Iberian hydroclimate and North Atlantic sea surface temperature (SST) during recent glacial periods has been identified through the analysis of marine sediment and pollen grains co-deposited on the Portuguese continental margin. These reconstructions have lacked a directly-dated, site-specific record of continental Iberian climate spanning multiple glacial cycles as a point of comparison. Here we present a high-resolution, multi-proxy ( $\delta^{13}$ C,  $\delta^{234}$ U, and growth dynamics) stalagmite record of hydroclimate from two caves in western Portugal across the majority of the last two glacial cycles (~230 ka). At orbital and millennial scales, stalagmite-based proxies of hydroclimate covaried with sea surface temperature (SST), with positive carbon isotope excursions and/or growth hiatuses indicating reduced effective moisture coincident with suppressed SST during major ice-rafted debris events and Greenland stadials, in agreement with changes in palynological reconstructions of continental climate. The Portuguese stalagmite record also reveal intervals during which the magnitudes of hydroclimate changes appear to have been somewhat decoupled from SST.

### 1. Introduction

The Portuguese continental margin is an important location for understanding oscillations in paleoceanographic conditions over orbital and millennial-scales (e.g., Voelker and de Abreu, 2011). Here, marine sediments record basin-wide oceanographic signals while co-deposited pollen grains track coeval vegetation changes occurring across Iberia. Integrated analysis of these proxies has revealed a close coupling of North Atlantic SST, regional climate, and Iberian ecosystems during the last three glacial cycles, including changes in vegetation dynamics (Sánchez Goñi et al., 2002; Tzedakis et al., 2004; Roucoux et al., 2006; Martrat et al., 2007;

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Naugthon et al. 2007; Sánchez Goñi et al., 2008), atmospheric circulation (Sánchez Goñi et al., 2013), and fire frequency (Daniau et al., 2007). One commonly applied palynological metric is the abundance of temperate tree pollen, which rises during warm and wet conditions associated both with interglacials and Greenland interstadials, concomitant with shifts in Iberian margin SST (Sánchez Goñi et al., 2002; Tzedakis et al., 2004; Combourieu-Nebout et al., 2009; Fletcher et al., 2010; Chabaud et al., 2014). However, the nature of such land-sea connections is partially obscured by the size of catchments from which the pollen are derived, with some reaching into central Iberia and spanning a range of environmental settings subject to varying climate influences (Martin-Vide and Lopez-Bustins, 2006; Naughton et al., 2007) (Fig. 1). Testing the links between terrestrial and marine systems requires continental climate proxies that provide precisely-dated and high resolution rainfall-sensitive time series spanning tens of millennia, but such records remain rare in Iberia, particularly near the west Iberian margin (Fletcher et al., 2010; Moreno et al., 2012; Stoll et al., 2013). Here we present a stalagmite record of three proxies for hydroclimate – growth dynamics,  ${}^{13}C/{}^{12}C$  ratios ( $\delta^{13}C$ ), and  $^{234}\text{U}/^{238}\text{U}$  ratios ( $\delta^{234}\text{U}$ ) – spanning the majority of the last and penultimate glacial cycles ( $\sim$ 230 ka) at two cave sites in western Portugal. This time series offers a rare, site-specific continental record capable of examining the coherence of SST controls on Iberian climate and ecosystem dynamics across glacial and interglacial periods. Few speleothem records exist in western Iberia, and this new speleothem record provides a continental proxy of hydroclimate linked to regional oceanographic conditions.

78 2. Samples and Regional Setting

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We report the analysis of six stalagmites (BG41, BG66, BG67, BG68, BG611, BG6LR) from Buraca Gloriosa (BG; 39°32'N, 08°47'W; 420 m a.s.l.) and one stalagmite (GCL6) from Gruta do Casal da Lebre (GCL; 39°18'N, 9°16'W; 130 m a.s.l.), two caves in western Portugal (Fig. 1). Environmental conditions in BG and GCL are well suited for speleothem paleoclimate reconstruction. Each cave contains only a single, small (<1 m<sup>2</sup>) entrance that restricts airflow and maintains stable temperatures (~1°C change between summer and winter seasons) and high relative humidity (~100% perennially) throughout the year, despite the fact that drip rates are highly variable, corresponding to both seasonal climatology and individual rainfall events (Fig. 2). BG and GCL are located in western Portugal, an area within the Meso-Mediterranean bioclimatic zone that dominates much of Iberia (Fig. 1). This region is characterized by strong seasonality, with warm, dry summers and cool, wet winters (Fig. 3) associated with the winter westerlies (Blanco Castro et al., 1997). In contrast, the Atlantic zone, north of the Douro River, is cooler, wetter, and less strongly seasonal. In the Pleistocene, the transition between these zones likely shifted southward with Mediterranean-type vegetation restricted to refugia (Rey Benayas and Scheiner, 2002). Pollen is transported to the west Iberian margin largely via major stream systems including (from north to south) the Douro, Tagus, and Sado rivers, with large watersheds (79,000, 81,000, and 7,650 km<sup>2</sup>, respectively). The Tagus and Sado are primarily responsible for pollen deposited southwest of Portugal, while the Douro plays an important role in delivering pollen to the more northwesterly sites (Fig. 1). Prevailing wind patterns likely prevent substantial transport of pollen from Iberia to the western Portuguese margin (Naughton et al., 2007).

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Today, the hydroclimate of western Iberia is tightly coupled with the winter North Atlantic Oscillation (NAO) (Fig. 4), an atmospheric dipole that strongly influences precipitation across much of western Europe and that more broadly reflects the strength and positioning of the Azores high-pressure system, which steers storm tracks contained within the westerlies into or north of Iberia (e.g., Trigo et al, 2002; Paredes et al., 2006; Trouet et al., 2009; Cortesi et al., 2014). The NAO is typically measured as the NAO index, which is calculated using atmospheric pressure differences between Iceland and Lisbon (or the Azores) (Barnston and Livezey, 1987). The nature of the influence of the NAO varies across Iberia, but it is strongly correlated to rainfall in western Portugal (Fig. 4). A positive NAO index is associated with a larger pressure gradient and elevated Iberian aridity. Iberian precipitation has also been linked closely to SST in regions ranging from the western North Atlantic to the Iberian margin (Lorenzo et al., 2010) where ocean circulation is dominated by the southward-flowing Portugal Current and the near-coastal and north-flowing Iberian Poleward Current, two systems that transport pollen from river mouths along the continental shelf (Fig. 1).

## 3. Materials and Methods

Stalagmite chronologies were constructed with a total of 76 <sup>230</sup>Th dates obtained at the University of New Mexico (Table 1) using the methods of Asmerom et al. (2010). For GCL samples, corrections for unsupported <sup>230</sup>Th were made with the global <sup>230</sup>Th/<sup>232</sup>Th crustal silicate value of 4.4 ppm (±50%), and for BG, corrections were made using a ratio of 13.5 ppm (±50%), a value determined from isotopic analysis of cave drip water. Powders ranging from 50-150 mg transferred into 30 ml Teflon beakers, weighed, dissolved in 15N nitric acid, and then spiked with a mixed <sup>229</sup>Th-<sup>233</sup>U-<sup>236</sup>U tracer and processed using column chemistry methods. U and Th

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measurement on the Thermo NEPTUNE MC-ICP-MS. U and Th isotopic ratios were aspirated into the MC-ICP-MS using a Cetac Aridus II low flow desolvating nebulizer and run as static routines. All isotopes of interest were measured in Faraday cups, except for <sup>234</sup>U and <sup>230</sup>Th, which were measured in the secondary electron multiplier (SEM). Gains between the SEM and the Faraday cups were determined using standard solutions of NBL-112 for U and an in-house <sup>230</sup>Th-Th standard for Th which was measured after every fifth sample; chemistry blanks reveal U and Th blanks below 20 pg. Ages are reported using two standard deviation errors and are reported relative to the year AD 1950. Age models were developed via multiple polynomial interpolations between dated intervals using the COPRA age modeling software (Breitenbach et al., 2012), except for BG68, the chronology of which was determined by linear interpolation between the two U-series dates that represent a short interval of stalagmite growth (Fig. 5). A total of 1,490 stable isotope analyses were performed on calcite samples milled from the central axis of each stalagmite. After milling, powders were weighed ( $\sim 200 \ \mu g$ ) and transferred to reaction vessels that were flushed with ultra-pure helium. Next, samples were digested using H<sub>3</sub>PO<sub>4</sub> and equilibrated overnight (~15 hours) at 34°C before being analyzed. Isotopic ratios were measured using a GasBench II with a CombiPal autosampler coupled to a ThermoFinnigan Delta Plus XL mass spectrometer at Iowa State University. A combination of internal and external standards were run after every fifth sample, as well as before and after each batch, in order to ensure reproducibility. Oxygen and carbon isotope values are presented in parts per mil (‰) relative to the Vienna Pee Dee Belemnite carbonate standard (VPDB). Precision is better than 0.1% for both  $\delta^{13}$ C and  $\delta^{18}$ O.

fractions were dissolved in 5 ml of 3% nitric acid and transferred to analysis tubes for

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The nature of the calcite comprising the BG samples alternates between a faster-growing, white, fibrous mineralogy and slower-growing, dense, clear fabric. In some areas of three samples - BG6LR, BG67, and BG68 - sharp changes between the two forms within the same growth horizons mark intervals of recrystallization. BG6LR, which grew discontinuously over much of the last glacial cycle, suffered from alteration of early Holocene (~10-7 ka) material, which was therefore excluded from this analysis. BG67 is characterized primarily by fibrous calcite that has been recrystallized to clear, dense calcite in a narrow band ascending through its core. U/Th dates from the fibrous calcite on the margins of the growth surface reveal open system behavior and thus this portion of BG67 was also excluded. BG68 is composed entirely of clear calcite but contains intervals of porosity, possibly originating from secondary alteration. We restricted our sampling to the central area of the stalagmite where calcite is clear, dense, and contains well-defined lamina. Growth position changed at numerous times in several of these stalagmites (Figure S1), and our sampling strategy accounted for these changes so as to consistently collect samples for stable isotopic analysis from the top surface (cap) of each stalagmite. Environmental conditions were measured at both cave sites over a two-year period, with data measured in three-hour intervals near the areas where the stalagmites were deposited. Temperature and relative humidity were obtained using HOBO U23 automated sensors while barometric pressure was recorded with a HOBO U20L. Drip rates were monitored with a Stalagmate acoustic drip counter (Collister and Mattey, 2008) (Fig. 2)

167 **4. Results** 

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quarters of the last 230 ka, with periods of deposition interrupted by numerous hiatuses of varying length. The longest gaps in this record span 210-198, 160-149, 106-87, 71-61, and 44-36 ka, and integrating these records yields a quasi-continuous time series from MIS 9 to present. Additional periods of deposition are present but were excluded from this analysis due to signs of secondary alteration that raise questions about the accuracy of associated U/Th dates (Fig. S1). The large number of hiatuses and also changes in growth direction complicate construction of a reliable chronology in some intervals. However, multiple stalagmites span the same growth periods, allowing for replication tests of coeval material. Replication between stalagmites of similar age is arguably the single most reliable method for evaluating the impacts of climate vs disequilibrium influences on isotopic ratios (Dorale and Liu, 2009). The similar carbon isotopic trends and values across many of the areas of overlap argue for a consistent climate signal as the primary driver of isotopic variability (Fig. S2). Hydroclimate is reconstructed here using a combination of stalagmite growth intervals and isotopic compositions. In some instances, deposition of multiple stalagmites was punctuated by hiatuses of similar time spans, a relationship that suggests links to changes in hydroclimate rather than random drip site-specific variability. Complementing growth intervals are stalagmite carbon isotopic ratios, which are impacted by a wide array of factors including vegetation type (plants utilizing the C<sub>3</sub> vs C<sub>4</sub> photosynthetic pathway) (Dorale et al., 1992) and density over the cave (Hellstrom and McCulloch, 2000), soil respiration rates (Genty et al., 2003), out-gassing of dissolved CO<sub>2</sub> from infiltrated water in voids in the epikarst under dry conditions (prior calcite precipitation; Baker et al., 1997), rate of CO<sub>2</sub>-degassing from water entering the cave (Breitenbach et al., 2015), and isotopic disequilibrium during carbonate crystallization (e.g.,

<sup>234</sup>U-<sup>230</sup>Th dating of BG and GCL stalagmites reveals growth across approximately three

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Fairchild et al., 2006). Terrestrial deposits preserving pollen spectra spanning substantial portions of the last glacial cycle are not well known from western Iberia (Gómez-Orellana et al., 2008; Fletcher et al., 2010; Moreno et al., 2012), and thus pollen in ocean sediments represents a particularly important continental climate record. Pollen obtained from the Iberian margin reveal small percentages of *Poaceae*, the family including the majority of C<sub>4</sub> plants, and therefore a persistent and overwhelming majority of C<sub>3</sub> vegetation throughout the last glacial cycle including between Greenland stadials (GS) and interstadials (GI) and across Heinrich stadials (HS) (d'Errico and Sánchez Goñi, 2003; Tzedakis et al., 2004; Desprat et al., 2006; Sánchez Goñi et al., 2008; Sánchez Goñi et al., 2013; Margari et al., 2014) (Fig. 6). Thus, increases in carbon isotopic ratios are interpreted here as primarily reflecting a combination of desaturation of voids above the cave and decreased organic CO<sub>2</sub> production within the soil zone, both of which are consistent with a vegetative response to cooler, more arid climates (Baker et al., 1997; Genty et al., 2003). Supporting this interpretation is the observed covariation of  $\delta^{234}$ U and  $\delta^{13}$ C values at both millennial and orbital scales (Fig. 6). During infiltration, <sup>234</sup>U is selectively mobilized relative to <sup>238</sup>U from the crystal lattices in carbonate bedrock owing to its displacement by alpha recoil during decay from <sup>238</sup>U (Chabaux et al., 2003; Oster et al., 2012). Decreases in effective precipitation and/or bedrock dissolution rate, both of which are associated with increased aridity, have been tied to elevated speleothem  $\delta^{234}$ U values (Hellstrom and McCulloch, 2000; Plagnes et al., 2002; Polyak et al., 2012), and are interpreted similarly here. Thus, as precipitation/effective moisture decreased, stalagmite  $\delta^{13}C$  and  $\delta^{234}U$  values increased. As differences in  $\delta^{234}U$  values between stalagmites may arise from distinct infiltration pathways, we restrict this part of the

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213 analysis solely to stalagmite BG6LR, which represents the longest individual stalagmite record 214 of this time series. 215 Oxygen isotope variability largely tracks that exhibited by carbon isotopes (Fig. S2). 216 However, interpreting changes in oxygen isotope composition in this system during intervals of 217 profound climatic change such as marked the last glacial period is complicated by the multiple 218 factors that influenced  $\delta^{18}$ O values of precipitation at these sites, including shifts in air 219 temperature and rainfall amount (Fig. 3), as well as the potential for changes in atmospheric 220 moisture sources on synoptic and seasonal scales (Moreno et al., 2014; Gimeno et al., 2012). 221 Oxygen isotopic ratios in stalagmites may also be influenced by kinetic effects (Mickler et al., 222 2004) and evaporation above the cave (Denniston et al., 1999). For these reasons, the carbon 223 isotopic record of BG/GCL represents the primary focus of this analysis. The reproducibility of 224 carbon isotope ratios between coeval BG stalagmites argues that their  $\delta^{13}$ C values may be viewed 225 as an integrated time series not substantially impacted by inter-sample isotopic offsets (Fig. 6). 226 However, incorporating the single GCL stalagmite into the BG record is complicated by site-227 specific cave hydrological effects, local controls on vegetation and soil formation, and different 228 bedrock  $\delta^{13}$ C values between the two caves that affect the values, although not the trends, of 229 stalagmite carbon isotopic ratios (Fairchild et al., 2006). As a result,  $\delta^{13}$ C values of this stalagmite are plotted separately in some figures to better track reconstructed Iberian SST. 230 231 The consistency and coherence among carbon (and oxygen) isotope values of coeval stalagmites support the interpretation that  $\delta^{13}$ C values robustly record a paleoenvironmental 232 233 signal (Dorale and Liu, 2009; Ridley et al., 2015). Some offsets exist, however, the most notable 234 of which is the shift toward higher  $\delta^{13}$ C values at the MIS 5e/6 transition (~130 ka) in stalagmite 235 BG611 that contrasts with the sharp decrease in carbon isotopic ratios in BG67 (Fig. 6).

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Assessing equilibrium crystallization in modern calcite/drip water pairs at BG is complicated by the substantial variability in integrated, months-long dripwater samples (average  $\delta^{18}$ O value of -  $3.4 \pm 1.1\%$ ) and any seasonal biases in calcite crystallization that at present remain poorly constrained. However, combining these data with the average cave temperature (14.2±0.5°C) yields modeled calcite values of -3.3±1.6‰ (Kim and O'Neil, 1997), a value that lies in broad agreement with the oxygen isotopic composition of recently deposited BG stalagmite calcite (-3.1‰).

# 5. Environmental Conditions at BG/GCL and Links to the Iberian Margin

Over the last several glacial cycles, oceanographic conditions along the western Iberian margin varied at millennial and orbital time scales in close correlation with the North Atlantic (Roucoux et al., 2005; Daniau et al., 2007; Sánchez Goñi et al., 2008; Darfeuil et al., 2015). Abrupt changes in SST reflect a balance between southward expansion of cold, subpolar waters and northward migration of subtropical water masses (de Abreu et al., 2003). During the particularly cold conditions characterizing HS and GS, Iberian margin SST decreased by as much as 9°C (to as much as 13°C below present values; de Abreu et al., 2003), with these changes helping to position the arctic or subarctic front at ~39°N, the same latitude as BG and GCL, thereby reducing atmospheric moisture to Iberia (Eynaud et al., 2009; Voelker and de Abreu, 2011). These oceanographic/atmospheric interactions would have influenced speleothem growth and carbon isotopic values at BG and GCL through impacts on effective moisture, vegetation density, and prior calcite precipitation. Indeed, the composite BG/GCL record documents a strong coherence, at both orbital and millennial scales, between Portuguese hydroclimate, vegetation, and Iberian margin SST during the last two glacial cycles (Fig. 6; Fig.

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7). Stalagmite  $\delta^{13}$ C and  $\delta^{234}$ U values covary with baseline changes in SST, and are sensitive to 259 260 gradual SST shifts. Millennial-scale changes in stalagmite carbon isotope ratios are more pronounced, with increased  $\delta^{13}$ C and  $\delta^{234}$ U values marking deterioration of Iberian climate 261 262 associated with onset of cold, dry conditions during HS (Eynaud et al., 2009; Naughton et al., 263 2009; Voelker and de Abreu, 2011) (Fig. 7). 264 Growth hiatuses characterize HS3, HS4, and HS11, suggesting complete cessation of 265 stalagmite deposition. Vegetation patterns during maximal IRD deposition on the Iberian margin 266 reveal not only dramatically reduced forest cover but also a pronounced expansion of semi-desert 267 plants, reflecting a further increase in aridity (e.g. Sánchez Goñi et al., 2000; Roucoux et al., 268 2005; Naughton et al., 2009). In particular, the long hiatus between HS7 and HS6 coincides with 269 a period characterized by very low arboreal pollen abundances and expansion of semi-desert 270 plants (Roucoux et al., 2005) and overlaps with the some of the coldest SST of the last 70 ka as reconstructed using  $U_{37}^{k'}$  at core MD95-2042, a core from off the southwestern Iberian coast 271 272 (Darfeuil et al., 2016). 273 Whether an absence of speleothem deposition is a result of pronounced reductions in 274 precipitation, an extension of below freezing temperatures that limited infiltration (Vaks et al., 275 2013; Fankhauser et al., 2016), or variations in infiltration pathway/drip position is unclear. 276 Pollen transfer functions from MD95-2042 (Fig. 1) suggest winter temperatures dropped below 277 0°C during HS and annual precipitation was reduced by up to 50% (from 800 mm to 500-400 278 mm during HS3, HS4, and HS5) (Sánchez Goñi et al., 2002). Applying this temperature 279 reconstruction to western Portugal is complicated, however, by the broad area across which these 280 pollen were sourced. Permafrost reconstructions (Vandenberghe et al., 2014) argue against the 281 hypothesis that continuous sub-zero temperatures inhibited infiltration and stalagmite growth.

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We thus suggest that the hiatuses observed at BG and GCL were driven largely by reductions in precipitation. This inference is supported by the discontinuous nature of stalagmite deposition during MIS 7 (221-193 ka). At this time, growth occurred in three intervals - MIS 7a, 7c, and 7e - punctuated by hiatuses during periods of ocean cooling (Fig. 2) and that also coincide with reductions in aquatic pollen and expansion of semi-desert plants (Desprat et al., 2006; Roucoux et al., 2006; Martrat et al., 2007; Voelker and de Abreu, 2011). During MIS 6, an absence in BG stalagmite deposition from ~160-149 ka (Fig. 6) occurs at the same time as massive seasonal discharges from the Fleuve Manche river and coldest continental climates and SST (157-154 ka) as determined from pollen and foraminifera from core MD01-2444 (Margari et al., 2014; Fig. 1). Stalagmites from Villars Cave, southwestern France, also contain hiatuses at 67-61, coincident with HS6, and 79-76 ka (Genty et al., 2003), similar in timing to hiatuses in the BG record at 71-60 and 80-78 ka (Fig. 1; Fig. 2). Additionally, a suite of stalagmites from multiple caves in northwestern Spain reveals punctuated growth, with periods of stalagmite deposition and/or elevated growth rates occurring during high Northern Hemisphere summer insolation or during GI (Stoll et al., 2013) (Fig. 1). Together, these data sets demonstrate a lack of stalagmite growth at times similar to those punctuating the BG/GCL record, suggesting SST controls on regional hydroclimate (Fig. 6). Stalagmite carbon isotopic variations also track SST changes. Prominent positive carbon isotopic excursions define the Younger Dryas (YD), HS2, HS5, HS6, and HS8, consistent with diminished concentrations of arboreal pollen in cores from the Iberian margin, and document particularly cold and dry conditions (Sánchez Goñi et al., 2000; Roucoux et al., 2005; Sánchez Goñi et al., 2008) (Fig. 7). Reduced stalagmite  $\delta^{13}$ C values document enhanced effective moisture from 170-160 and 145-135 ka, tracking peaks in temperate tree pollen and alkenone-

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305 based SST. Similarly, the sharp isotope shift marking the YD in the Portuguese stalagmite is 306 consistent with the highest resolution SST record from the Iberian margin that reveals 307 pronounced cooling during this time (Rodrigues et al., 2010). 308 Hydroclimatic shifts associated with GS and GI are also clearly expressed in some 309 portions of the BG carbon isotope record, with the magnitude of carbon isotopic variability 310 associated with GS/GI transitions ranging from 3-6% (Fig. 8). Pollen from the Iberian margin 311 has been demonstrated to track GS/GI variations, with prominent changes occurring in relative 312 abundances of Mediterranean and Atlantic forest and steppe vegetation (e.g., Sánchez Goñi et al., 313 2008). Other European stalagmite records have identified GI/GS events from the last glacial 314 period (Genty et al., 2003; Spötl et al., 2006; Boch et al., 2011; Moseley et al., 2014), but the 315 level of resolution recorded in the BG/GCL time series has not been clearly identified previously 316 in western Iberia. Interesting, many GS/GI oscillations are not readily apparent during MIS 3, 317 perhaps due to either insufficiently fine temporal resolution or a subdued hydroclimatic response. 318 A carbon isotope time series (albeit with low temporal resolution) of a flowstone from southeast 319 Spain does not present clear evidence of either GI or most HS during the last glacial cycle, 320 although it does contain a clear expression of HS11 (Hodge et al., 2008) (Fig. 1). And while 321 some Iberian lakes and peat bogs document environmental changes concurrent with HE, no 322 single record, including one of the longest - the 50 ka time series from the Fuentillejo maar, 323 south-central Spain - contains a consistent signal for all HS (Vegas et al., 2010; Moreno et al., 324 2012) (Fig. 1). 325 Despite these similarities between the BG/GCL and SST/pollen records, some points of 326 divergence also exist which, while possibly attributable to site-specific environmental shifts or 327 in-cave processes, also raise questions about non-stationarity in the connections between ocean

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spike in forest taxa occurring at the start of interglacials, decreasing thereafter for the next 5-10 kyr (Tzedakis et al, 2004; Desprat et al., 2007). This early interglacial peak is a common feature in several time series including the Antarctic δD (Petit et al., 1999) and CH<sub>4</sub> records (Loulergue et al., 2008), and in stalagmite isotopic ratios from the eastern Mediterranean (Bar-Matthews et al., 2003) and southern France (Couchoud et al., 2009) (Fig. 2). However, the BG/GCL  $\delta^{13}$ C record lacks this feature. Next, stalagmite  $\delta^{13}$ C values are lower during GI 20-22 (MIS 5a/4; 84-72 ka) than in either the Holocene or MIS 5e, suggesting that maximum warmth and precipitation were not coincident with peak summer insolation ( $\sim$ 127 ka) (Fig. 6). BG/GCL  $\delta^{234}$ U values support this interpretation and compare well with the carbon isotope ratios from the same samples, which are substantially lower during GI 20-22 than in the Holocene. This interval is of particular interest given that Atlantic forest pollen, which has been used as a proxy for air temperature, was decoupled from SST across northwestern Iberia during cold events (C18-C20) (Rousseau et al., 2006; Rasmussen et al., 2014). This decoupling is interpreted to reflect a weakened control of SST on Iberian atmospheric temperature that, in turn, drove enhanced transport of atmospheric vapor to the high latitudes, amplifying production of ice sheets in the early stages of the last glacial cycle (Sánchez Goñi et al., 2013); this process has also been demonstrated for an earlier interglacial (MIS 19; Sánchez Goñi et al., 2016). And while the BG/GCL  $\delta^{13}$ C record generally tracks SST, an anomalously large  $\delta^{13}$ C response marks ice rafting event C24 (111-108 ka), with  $\delta^{13}$ C values rising ~5% higher than expected based on the observed scaling with SST (Fig. 7). One possible explanation for these relationships is disequilibrium effects on speleothem isotopic ratios, although overlap of coeval stalagmites generally supports a consistent climatic

conditions and Iberian climate (Fig. 7). For example, some marine cores reveal a prominent

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mechanism. Alternatively, changes in the nature of the NAO may have occurred over millennial time scales, with persistent positive (negative) NAO modes having been proposed for GS (GI) (Moreno et al., 2002; Sánchez Goñi et al., 2002; Daniau et al., 2007) as well as centennial periods during the Holocene (Chabaud et al., 2014), which resulted in sustained changes in precipitation regime. The dynamics of the NAO and Azores High pressure system are not well understood prior to the historical era (Trouet et al., 2009), and the BG/GCL record cannot address this question independently. However, Justino and Peltier (2005) modeled the NAO during the LGM and found fundamental differences as compared to modern conditions, including pronounced southerly, rather than westerly, atmospheric flow during positive NAO phases. One test of past NAO dynamics involves spatial variations in precipitation across Iberia. Rainfall variability in eastern Iberia is less closely tied to the NAO than is western Iberia and instead reflects other climatic phenomena including the El Niño-Southern Oscillation (Rodó et al., 1997), helping to produce an east-west precipitation gradient. Pollen spectra from the southwest Portuguese margin and those from the Alboran Sea in the western Mediterranean (Fig. 1) reveal a pronounced precipitation gradient during HS. This interpretation is supported by varying abundances of aeolian debris (including charcoal) deposited in marine deposits in the western Mediterranean that may reflect distinct wind patterns associated with NAO modes (Combourieu-Nebout et al., 2002), talthough contradictory results were obtained by Daniou et al. (2007). And Naughton et al. (2009) identified distinct climatic phases within HS consistent with changes in the NAO phasing. Additional speleothem records offering greater spatial coverage across Iberia will provide a more robust test of the underlying drivers of millennial-scale hydroclimatic changes during recent glacial periods.

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### 6. Conclusions

Throughout most of the last two glacial cycles, hydroclimate and vegetation dynamics at BG and GCL closely tracked North Atlantic SST over both orbital and millennial scales. Enhanced aridity characterized HS, as evidenced by hiatuses in stalagmite growth or elevated carbon isotopic ratios. While not fully understood, evidence suggests that  $\delta^{13}$ C variability at these scales was tied to enhanced prior calcite precipitation or depressed soil respiration rates. Differences between the structure of the stalagmite and SST records during some time intervals suggest that land-sea connections across Iberia may have varied temporally and spatially. In order to further test this hypothesis, additional speleothem records should be developed from central and southern Iberia to better constrain spatial patterns of glacial hydroclimate variability.

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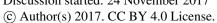
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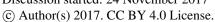
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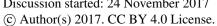
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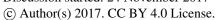






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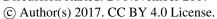






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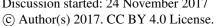






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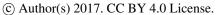






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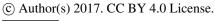




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635	Figure Captions
636	Figure 1. Average annual precipitation (mm) of the Iberian Peninsula for years AD 1901-
637	2009 (GPCC v. 6; Schneider et al., 2013) relative to cave study sites (white stars: GLC =
638	Gruta do Casal de Libre; BG = Buraca Gloriosa). Rectangle denotes location of northwest
639	Spain cave sites (NWSC) (Moreno et al., 2010; Stoll et al., 2013); FM = Fuentillejo maar (Vegas
640	et al., 2010) and GT = Gitana cave (Hodge et al., 2008); VC = Villars Cave (Genty et al., 2003)
641	located just north of map. Also shown are locations of marine cores discussed in text and GNIP

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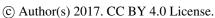




642 station at Porto. Bathymetric contours shown in grey (m). Location of currents after Voelker et 643 al. (2010). 644 645 Figure 2. Environmental monitoring at BG. A. Comparison of rainfall data from Monte Real 646 weather station (proximal to BG) and drip rates at BG. B. Temperature and humidity at BG. C. 647 Temperature and humidity profiles at GCL. 648 Figure 3. Oxygen isotopic composition of precipitation and rainfall amount (lefthand 649 panels) and air temperature (righthand panels). Data collected at IAEA/GNIP site in Porto, 650 Portugal (see Fig. 1 for location). Oxygen isotope data represent multi-year averages of monthly 651 means. Note that righthand y-axis in upper left panel is inverted in order to illustrate inverse 652 nature of rainfall and precipitation oxygen isotopic composition. 653 Figure 4. Iberian rainfall anomalies associated with the NAO. Precipitation anomalies, 654 averaged for November-March, between the positive NAO winter of 1988/89 (top panel) or the 655 negative NAO winter of 2009/2010 (bottom panel) relative to 1901-2013. Data from GPCC (v7) 656 (Schneider et al, 2013). Yellow stars denote cave sites in this study: BG = Buraca Gloriosa; GCL 657 = Gruta do Casal de Lebre. FM = Fuentillejo maar (Vegas et al., 2010) and GT = Gitana cave 658 (Hodge et al., 2008). 659 660 Figure 5. COPRA-derived age models for BG/GCL stalagmites. Black lines represent mean 661 of calculated age models while red lines denote 95% confidence intervals. See Table 1 for 662 specific ages and isotopic ratios. Not shown is the age model for BG68, which is based on only 663 two dates and thus was determined using linear interpolation. Orange squares represent "dummy

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664 ages" that were included in order to be able to extrapolate below some hiatuses, which is only 665 possible with at least two dated points. These dummy ages are added using the simple 666 assumptions of i) stratigraphically correct growth slope (i.e. higher = younger) and ii) samples 667 physically below a hiatus must be older than the oldest age above that hiatus. Conservative errors 668 were added to account for the unknown "true" age of the stalagmite at these points. 669 670 Figure 6. Comparison of Portuguese stalagmite hydroclimate proxies with regional and 671 global climate records from the last two glacial cycles (a) Ice-rafted debris abundance from 672 North Atlantic ODP Site 980 (McManus et al., 1999 using Hulu cave time scale as presented in 673 Barker et al., 2011); (b) BG/GCL stalagmite carbon isotopic time series with each stalagmite represented by a different color; <sup>230</sup>Th ages and 2 s.d. errors shown as circles with horizontal 674 675 lines. Uranium isotopic ratios for stalagmite BG6LR denoted by blue diamonds. Increased  $\delta^{13}$ C 676 and δ<sup>234</sup>U values indicate enhanced aridity; (c) Alkenone-based Iberian margin SST 677 reconstruction (core MD01-2443; Martrat et al., 2007 adapted to synthetic Greenland chronology 678 of Barker et al., 2011 by Hodell et al., 2013). Also shown are intervals between periods of 679 speleothem growth at caves in northern Spain (Stoll et al., 2013) and Villars Cave (Genty et al., 680 2003). See Fig. 1 for locations; (d) Temperate forest pollen abundance from marine cores listed 681 in figure (Tzedakis et al., 2004; Sánchez Goñi et al., 2008; Sánchez Goñi et al., 2013; Margari et 682 al., 2014); (e) Synthetic Greenland oxygen isotopic record (Barker et al., 2011); (f) EPICA Dome 683 C atmospheric methane record (Loulergue et al., 2008) and marine isotope stages. 684 685 Figure 7. Covariations of Iberian hydroclimate, Iberian margin SST, and Iberian pollen. Top Panel: Composite BG/GCL stalagmite  $\delta^{13}$ C time series and Iberian margin  $U_{37}^{k'}$  SST 686

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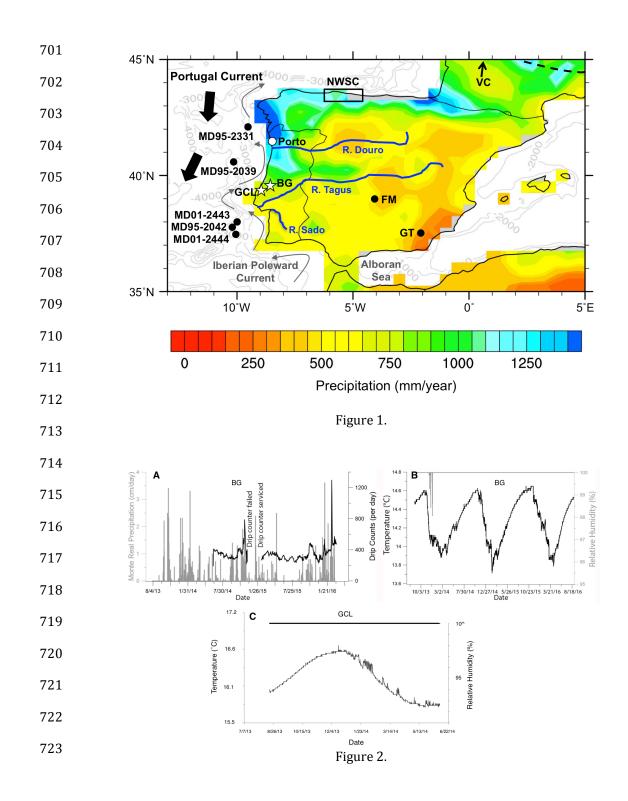




687 reconstruction for core MD01-2443 (Martrat et al., 2007; see Fig. 1). For ease of comparison to 688 SST, GCL6 is shifted by 2\% (original values in grey). Light blue vertical bands denote Heinrich 689 and Cold events (data from McManus et al., 1999 using Hulu cave time scale as presented in 690 Barker et al., 2011), some of which are labeled. Note correlation between hiatuses in stalagmite 691 growth and periods of depressed SST. Bottom Panel: Composite BG/GCL stalagmite  $\delta^{13}$ C time 692 series (as in top panel) and Iberian margin temperate pollen (Tzedakis et al., 2004; Sánchez Goñi 693 et al., 2008; Sánchez Goñi et al., 2013; Margari et al., 2014). 694 695 Figure 8. Comparison of select millennial events in the BG and Greenland ice records. Left: 696 Portion of the last deglaciation showing the BG stalagmite response (green) to cold events:  $\delta^{13}$ C 697 increase during the YD and a hiatus during HS1. Also shown is the NGRIP oxygen isotopic time 698 series (North Greenland Ice Core Project members, 2004) (blue); Right: GI/GS events 90-70 ka. 699 Stratigraphic nomenclature from Rasmussen et al. (2014). 700

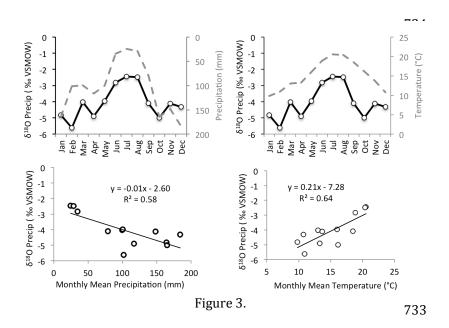


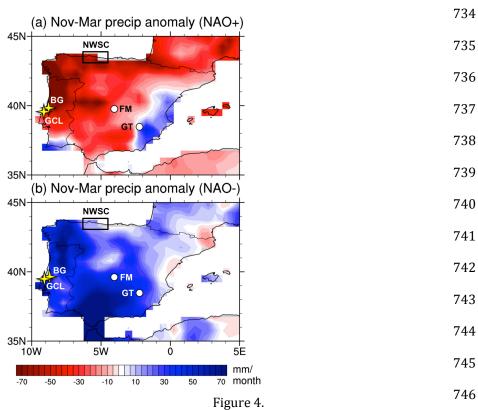






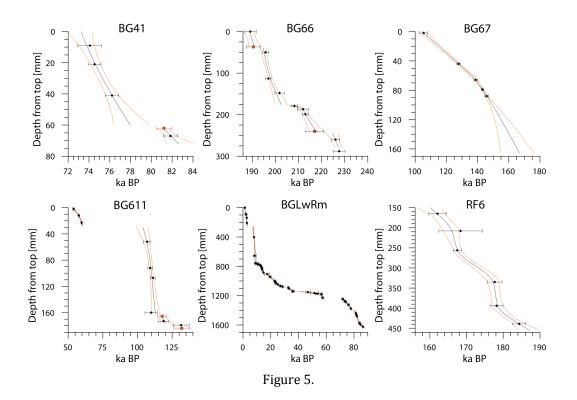






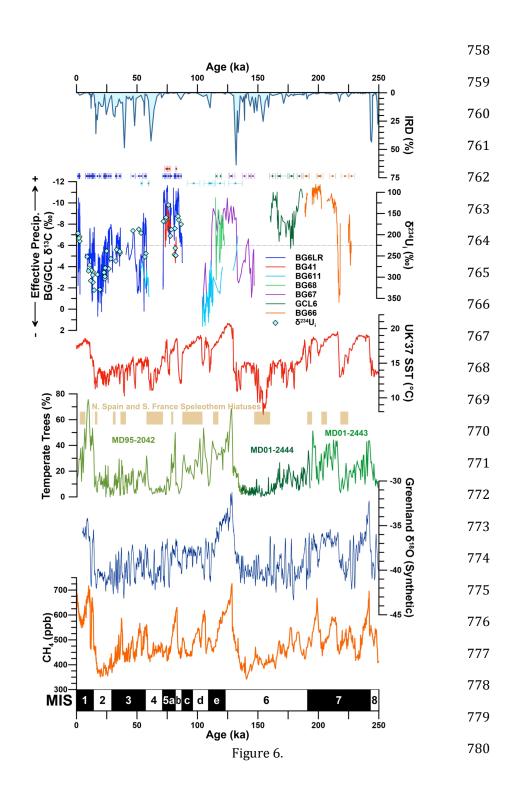






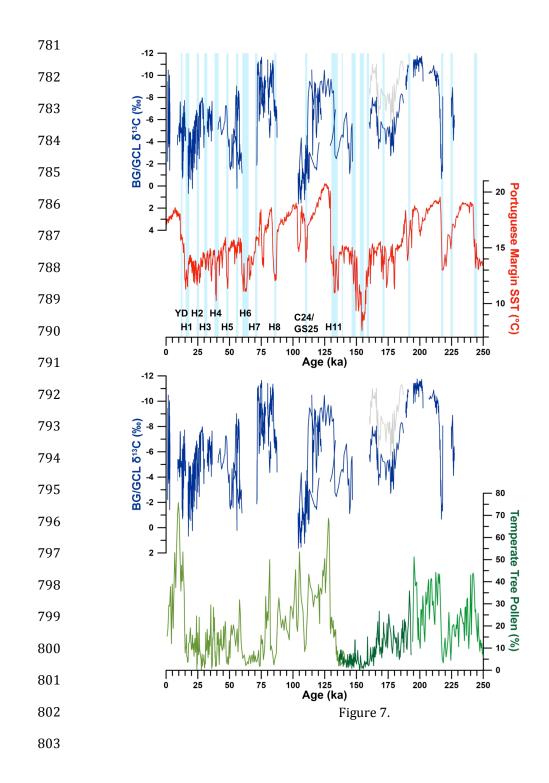










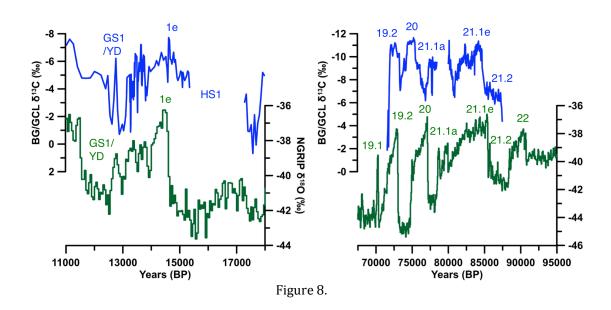


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Table 1. Isotopic Ratios and <sup>230</sup>Th Ages

Table 1. Isotopic Ratios and 250 Th Ages													
Stalagmite	Distance (mm) <sup>a</sup>	<sup>238</sup> U (ng/g)	<sup>232</sup> Th (pg/g)	<b>8</b> <sup>234</sup> U <sup>b</sup> (corr'd)	Error <sup>c</sup>	<sup>230</sup> Th/ <sup>238</sup> U (activity)	Error	<sup>230</sup> Th/ <sup>232</sup> Th (ppm)	Error	Uncorrected Age (yr BP) <sup>d</sup>	Error (yr)	Corrected Age (yr BP) <sup>e</sup>	Error (yr)
BG41	67	148	2,892	524.7	2.2	0.779	0.0023	657.7	18.6	82,926	389	82,553	538
BG41	41	293	4,635	522.8	2.2	0.742	0.0030	773.8	8.5	77,026	463	76,724	486
BG41	21	217	1,858	566.6	3.1	0.748	0.0039	1,440.0	40.6	74,906	567	74,746	588
BG41	9	271	2,088	610.8	9.8	0.764	0.0073	1,635.6	22.3	74,392	1,135	74,253	1,142
BG66	288	127	7,307	509.7	5.2	1.177	0.0025	338.3	2.5	231,038	1,960	227,590	2,562
BG66 <sup>r</sup>	280	85	6,980	698.6	9.3	1.283	0.0057	256.5	1.8	223,637	3,252	219,220	3,829
BG66 BG66	260 199	84	3,481	709.4	4.5	1.291	0.0025	514.1 623.6	4.2	228,224	1,503 1,052	226,040	1,836 1,379
BG66	187	101	3,132	532.4	3.1	1.174	0.0015	298.1	4.6	214,835	1,580	213,011 211,971	2,478
BG66	179	75 68	4,657 2,003	429.2 499.5	3.8	1.116 1.149	0.0025	644.2	1.7 7.4	215,891 210,002	1,175	208,236	1,456
BG66	148	95	4,336	379.4	3.1	1.073	0.0019	386.6	3.6	204,768	1,460	201,770	2,063
BG66	113	104	2,193	443.5	2.6	1.100	0.0025	864.2	11.9	198,297	930	196,990	1,128
BG66 <sup>r</sup>	68	82	1,476	293.1	3.9	1.015	0.0030	933.1	29.6	200,740	1,920	199,494	2,275
BG66	50	104	2,661	345.4	2.4	1.041	0.0016	672.5	8.4	197,507	994	195,798	1,298
BG66 <sup>r</sup>	24	91	6,080	389.7	5.1	1.061	0.0047	263.1	1.9	191,832	2,368	187,477	3,144
BG66	1	76	995	564.2	6.2	1.159	0.0057	1,453.3	64.3	189,936	2,538	189,182	2,549
BG67	88	320	2,153	617.8	2.9	1.095	0.0043	2,689.7	51.5	146,174	1,146	145,802	1,158
BG67	79	195	2,799	485.8	2.3	1.014	0.0022	1,164.3	18.2	144,037	695	143,171	814
BG67	66	250	4,187	610.3	4.9	1.072	0.0046	1,057.4	12.7	139,735	1,279	138,803	1,350
BG67	44	162	4,858	484.7	2.4	0.969	0.0023	531.9	5.3	129,620	608	127,800	1,087
BG67	2	216	5,542	401.5	2.6	0.837	0.0039	538.0	5.1	107,150	843	105,501	1,168
BG68	200	200	1,104	239.9	1.7	0.797	0.0017	2,382.0	66.9	119,376	509	119,247	525
BG68	80	199	317	273.0	2.2	0.802	0.0034	8,277.3	1,090.9	115,745	894	115,709	894
BG611 BG611	179 173	164 119	24,228 11.744	235.9 202.6	6.6 3.5	0.869 0.801	0.0046	97.1 133.9	0.5 0.8	142,696	1,841 1,253	131,493	5,716 3,908
BG611	160	110	12,829	230.9	4.6	0.792	0.0041	112.1	0.8	126,291 118,612	1,233	118,714 109,828	4,469
BG611	107	99	3,304	238.0	3.0	0.776	0.0038	383.6	5.6	113,518	1,028	111,086	1,574
BG611	30	122	16,801	251.3	5.0	0.762	0.0043	91.2	0.5	107,202	1,028	96,920	5,126
BG611	23	313	552	340.8	1.4	0.553	0.0024	5,168.2	353.7	59,726	345	59,608	350
BG611	2	250	4,109	376.7	1.8	0.533	0.0021	535.0	5.9	54,959	284	53,887	604
BG6LR	1,623	72	133	175.0	1.5	0.631	0.0015	5,665.9	1,162.4	86,532	342	86,392	350
BG6LR	1,593	98	140	165.3	1.4	0.618	0.0014	7,166.0	1,764.0	84,748	318	84,639	324
BG6LR	1,574	74	905	156.6	1.6	0.615	0.0016	824.8	25.3	84,848	360	83,894	596
BG6LR	1478	159	26	249.2	1.8	0.645	0.0021	63,745.2	114,069.8	82,068	428	82,056	428
BG6LR	1464	166	1,138	246.8	1.5	0.641	0.0009	1,542.3	35.8	81,475	214	80,983	325
BG6LR	1442	162	77	185.4	1.4	0.634	0.6339	21,885.5	13,014.6	81,442	396	81,407	396
BG6LR	1375	112	220	202.9	1.5	0.602	0.6016	5,064.2	652.0	77,823	234	77,677	246
BG6LR	1324	120	1,908	130.2	1.4	0.566	0.5660	585.8	15.3	77,213	330	75,946	712
BG6LR	1283	132	1,019	159.5	2.0	0.566	0.5659	1,213.9	71.1	74,623	422	74,029	515
BG6LR	1276 1246	105 83	353	167.8	2.1	0.564	0.5637	2,766.4	298.1 14.2	73,512	425 369	73,254	444
BG6LR BG6LR	1179	62	1,232 1,114	168.7 252.0	1.4 2.6	0.561 0.507	0.5613 0.5071	625.8 464.4	15.9	72,957 57,877	465	71,819 56,584	675 792
BG6LR	1179	77	2,544	196.0	2.0	0.307	0.4736	235.4	3.8	55,882	375	53,375	1,299
BG6LR		5	367	187.1	2.6	0.482	0.4730	100.4	1.6	57,644	524	51,517	3,066
BG6LR	1166 1153	81	3,460	190.7	2.2	0.433	0.4331	167.2	2.3	49,960	367	46,707	1,654
BG6LR	1141	52	1,159	242.6	2.8	0.359	0.3591	266.4	10.4	37,626	449	36,016	918
BG6LR	1138	55	750	239.5	1.8	0.352	0.3518	426.3	33.1	36,815	381	35,830	625
BG6LR	1101	71	283	235.2	2.0	0.323	0.3234	1,344.2	198.7	33,449	272	33,161	310
BG6LR	1093	70	472	262.1	2.1	0.327	0.3269	802.0	73.4	33,052	331	32,575	409
BG6LR	1077	101	595	256.6	1.8	0.290	0.2899	810.6	63.4	28,851	193	28,431	287
BG6LR	1068	85	1,034	280.0	1.4	0.285	0.2847	384.9	15.5	27,675	178	26,820	463
BG6LR	1046	56	705	238.2	2.2	0.260	0.2603	339.0	19.7	25,911	265	24,993	531
BG6LR	1026	123	2,093	304.1	1.9	0.262	0.2617	253.3	8.5	24,612	206	23,438	621
BG6LR	1025	123	493	296.4	1.4	0.253	0.0017	1,041.2	151.0	23,814	175	23,538	226
BG6LR	1019	80	377	298.5	2.1	0.252	0.2525	887.3	107.4	23,753	221	23,430	276
BG6LR	1001	68	1,464	288.7	1.5	0.256	0.2558	196.1	4.3	24,291	156	22,789	765
BG6LR	944	76	1,896	329.3	2.1	0.233	0.2330	154.8	3.9	21,131	196	19,450	861
BG6LR	899	79	4,209	294.0	3.4	0.227	0.2266	70.6	1.3	21,074	283	17,360	1,863
BG6LR	883	91	233	330.3	2.0	0.168	0.1684	1,082.0	213.7	14,806	165	14,633	189
BG6LR	843	100	1,409	287.7	4.0	0.162	0.1623	190.4	6.7	14,718	164	13,738	516
BG6LR	827	103	332	295.0	2.9	0.152	0.1521	783.5	116.9	13,645	154	13,424	192
BG6LR	819	75	491	311.6	1.4	0.158	0.1581	400.0	22.8	14,032	123	13,587	255
BG6LR BG6LR	783	95 107	525	283.8	2.2	0.141	0.1406 0.1304	418.7	35.3 5.7	12,661 11,795	150 119	12,275	246 463
BG6LR BG6LR	774		1,351 2,725	271.4 285.4	1.4	0.130 0.133	0.1304	169.8	5.7	11,795	141	10,901	1,031
BG6LR BG6LR	764 750	93 135	4,177	285.4 251.5	1.6 1.5	0.133	0.1334	75.3 64.7	1.6 1.0	11,942	141	9,892 8,846	1,031
BG6LR BG6LR	759 657	86	2,566	212.9	1.4	0.121	0.1210	62.1	0.9	10,540	96	8,846	1,113
BG6LR	657 139	172	323	204.2	1.7	0.112	0.0010	272.6	41.0	2,790	96 96	2,651	1,100
BG6LR	86	155	80	207.9	1.7	0.022	0.0007	720.9	312.3	1,987	62	1,949	67
BG6LR	1	122	43	196.7	18.9	0.014	0.0019	677.5	519.3	1,271	173	1,245	174
			1.0	0.,									
GCL6	439	91	2,815	76.3	2.3	0.862	0.0029	461.2	9.3	185,093	1,779	184,255	1,815

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GCL6	335	70	4,579	82.7	3.0	0.856	0.0029	214.9	2.3	179,406	1,794	177,624	1,977
GCL6	256	116	1,019	86.2	2.2	0.836	0.0020	1,574.3	71.8	167,617	1,102	167,382	1,105
GCL6 <sup>f</sup>	208	7	186	92.9	2.6	0.843	0.0124	522.0	99.0	169,018	6,007	168,307	5,988
GCL6	165	94	2,507	122.4	4.2	0.847	0.0049	526.3	13.4	162,712	2,368	162,022	2,550

<sup>&</sup>lt;sup>a</sup> Distances are relative to the top of each stalagmite. <sup>b</sup>  $\delta^{234}U_{meas'd} = [(^{24}U/^{238}U)_{meas'd}/(^{244}U/^{238}U)_{ea}] \times 10^{238}U_{ea}$  is secular equilibrium activity ratio:  $\lambda_{238}/\lambda_{234} = 1.0$ . Values are reported as permil.

<sup>&</sup>lt;sup>c</sup> Errors are 2σ.

e Trions at 20.0 defined as the year AD 1950.

Initial <sup>230</sup>Th/<sup>232</sup>Th atomic ratio of 13.5 x 10.6 ± 6.75 x 10.6 used to correct for unsupported <sup>230</sup>Th.

Not used in age model.