

1 Stalagmite evidence for Early Holocene multidecadal hydroclimate variability in Ethiopia

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35 **hydroclimate variability in Ethiopia**

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## 63 **ABSTRACT**

64

65 A multiproxy oxygen and carbon isotope ( $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ ), growth rate and trace element  
66 stalagmite paleoenvironmental record is presented for the Early Holocene from Achere Cave,  
67 Ethiopia. The annually laminated stalagmite grew from 10.6 to 10.4 ka, and from 9.7 to 9.0  
68 ka with a short hiatus at ~9.25 ka. Using oxygen and carbon isotopic, and cave monitoring  
69 data, we demonstrate that the stalagmite deposition is out of isotopic equilibrium, yet trace  
70 element and isotope geochemistry is sensitive to hydroclimate variability. Variogram analysis  
71 of annual growth rate data suggests that this proxy can only contain hydroclimate information  
72 over less than 28-year timescales. Statistically significant and coherent spectral frequencies in  
73  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  are observed at 15-25 and 19-23 years respectively. Combined with  
74 compelling evidence for deposition out of isotope equilibrium, the observed ~1 % amplitude  
75 variability in stalagmite  $\delta^{18}\text{O}$  is likely forced by non-equilibrium deposition, likely due to  
76 kinetic effects during the progressive degassing of  $\text{CO}_2$  from the water film during stalagmite  
77 formation. These frequencies are similar to the periodicity reported for Holocene stalagmite  
78 records from Ethiopian caves, suggesting that multidecadal variability in stalagmite  $\delta^{18}\text{O}$  is  
79 typical. We hypothesise that a hydroclimate forcing, such as runs of one or more years of low  
80 annual rainfall, is likely to be the primary control on the extent of the partial evaporation of  
81 soil and shallow epikarst water, which is subsequently modulated by karst hydrology, and the  
82 extent of in-cave non-equilibrium stalagmite deposition. Combined with possible recharge-

83 biases in drip water  $\delta^{18}\text{O}$ , modulated by karst hydrology, these processes can generate  
84 multidecadal  $\delta^{18}\text{O}$  variability which can operate with opposite signs. Comparison of Early  
85 Holocene  $\delta^{18}\text{O}$  stalagmite records from the monsoon regions of Ethiopia, Oman and central  
86 China show different multi-decadal  $\delta^{18}\text{O}$  signals, implying regional difference in climate  
87 forcing. Seismic activities due to the active tectonics in the region control the frequency of  
88 growth gaps (hiatuses) by changing the water flow paths to the stalagmite.

89

90 **Key Words:** Early Holocene, multi-decadal variability, eastern Africa, paleoclimate, Oxygen  
91 isotopes

92

### 93 **1. Introduction**

94

95 A number of major air streams and convergence zones influence the modern climate in  
96 Ethiopia and the larger Horn of Africa region (Nicholson, 2017). Rainfall amount and  
97 intensity in Ethiopia is determined by the annual migration of the African rain belt, which is  
98 associated with the movement of the Intertropical Convergence Zone (ITCZ). The annual  
99 migration of the ITCZ determines the onset, duration and termination of the East African  
100 monsoon, leading to a strongly bimodal annual cycle, resulting in two rainy seasons: the ‘big  
101 rains’ or summer rains (between June and September), which is dependable and whose  
102 maxima migrates with the position of the ITCZ, and a second rainy season, the ‘small rains’  
103 or spring rains, which is less consistent and occurs between March and May with maxima in  
104 April.

105 In addition, East-West adjustments in the zonal Walker circulation regulated by the El  
106 Niño-Southern Oscillation (ENSO) and the Indian Ocean Dipole (IOD) cause short-term  
107 (annual to decadal) fluctuations in the intensity of precipitation in Ethiopia. These are

108 possibly a direct response to sea-surface temperature (SST) variations in the Indian and  
109 Atlantic Oceans, which are in turn affected by the ENSO and the IOD (Nicholson, 2017;  
110 Taye et al., 2021). While the global-scale atmospheric circulation patterns determine the  
111 rainy seasons in Ethiopia, local rainfall distribution is modulated by the topographic features  
112 such as the highland barriers separated by a rift zone (Asrat et al., 2018).

113         Nearly 80 % of the >100 million people inhabiting Ethiopia depend on rain-fed  
114 agriculture for their subsistence. Both the summer and spring rains in most parts of the  
115 country are important for adequate and sustained harvest. However, the interannual  
116 variability of the spring rains is higher than the summer rains (e.g., Viste et al., 2013) and  
117 failure of the spring rains is common (Diro et al., 2008). Failure of the spring crop usually  
118 leads to a reduced annual productivity (McCann, 1990) and in most cases leads to famine, at  
119 least in some worst-hit parts of the country, such as in 1984 and 2009, the two driest years  
120 since 1971 (Viste et al., 2013). The southeastern Ethiopian lowlands were affected by failure  
121 of the spring rains as recently as the 2013/2014 and 2015/2016 growing seasons.

122         There has been a general decline in the reliability of the spring rains since 1979 (e.g.,  
123 Williams and Funk 2011; Viste et al., 2013), and data on the failure of the spring rains for the  
124 modern era suggests this occurs at a multidecadal frequency. For instance, within the 1995-  
125 2010 period, Viste et al. (2013) identified a cluster of dry spring seasons nationwide in 1999-  
126 2004 (except 2001), and in 2008-2011. The causes for the failure of the spring rains remain  
127 unclear. However, some studies (e.g., Segele et al., 2009; Williams and Funk, 2011; Viste et  
128 al., 2013) agreed that the failure is usually associated with deflections of the transport of  
129 moisture to Ethiopia due to atmospheric circulation anomalies. For instance, the 2009 spring  
130 drought was largely attributed to the deflection of the easterly flow bringing moisture from  
131 the Northern Indian Ocean and the southeasterly flow bringing moisture from the southern  
132 and equatorial Indian Ocean, by southwesterly anomalies (Viste et al., 2013).

133 Paleoclimate records provide a useful insight into the processes determining rainfall  
134 climate variability (Bar-Matthews et al., 1997; Hu et al., 2008), such as the multidecadal  
135 frequency of failure of the spring rains described earlier. For Ethiopia, annually-laminated  
136 records such as those widely present in stalagmites from the country have the necessary  
137 temporal resolution to investigate past multi-decadal climate variability (Asrat et al., 2007;  
138 2018; Baker et al., 2007; 2010). Previous research has shown that the strong seasonality of  
139 rainfall leads to the ubiquitous formation of annual growth laminae (Asrat et al., 2008). The  
140 warm climate leads to a fast stalagmite annual growth rate (100 to 500  $\mu\text{m}/\text{yr}$ ), permitting  
141 high-resolution geochemical analyses. Tectonic activity associated with the adjoining East  
142 African Rift System to the cave sites leads to discontinuous stalagmite deposition rarely  
143 lasting more than 1,000 years, with stalagmites often having distinctive cone-shaped  
144 morphologies indicative of a drainage of a water source (Asrat, 2012). Two discontinuously  
145 forming, Early to Middle Holocene stalagmite records from the Mechara caves (Ach-1 and  
146 Bero-1 stalagmites) have previously exhibited multidecadal variability in  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ , as  
147 well as growth rate (Asrat et al., 2007; Baker et al., 2010). However, multi-decadal variability  
148 in speleothems can be climatically forced, can derive from the inherent non-linear properties  
149 of karst hydrology, or can arise from a combination of the two; e.g. non-linear karst processes  
150 amplifying the signal from extreme climate events (Baker et al., 2012).

151 Multi-stalagmite and multi-proxy analyses is essential for investigating the  
152 reproducibility of paleoclimate records in speleothems (Hellstrom and McCulloch, 2000;  
153 Dorale and Liu, 2003). Here, we present a third high-resolution stalagmite paleoclimate  
154 record for the Holocene from Achere Cave, southeastern Ethiopia. Stalagmite Ach-3, which  
155 formed in the Early Holocene, is dated by U-Th series and annual laminae, and analysed for  
156  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  and trace elements. Combined with geostatistical analyses, we investigate the

157 multidecadal geochemical proxy signal in the stalagmite and compare this to other Middle  
158 and Late Holocene stalagmite records from the region.

159

## 160 **2. Methods**

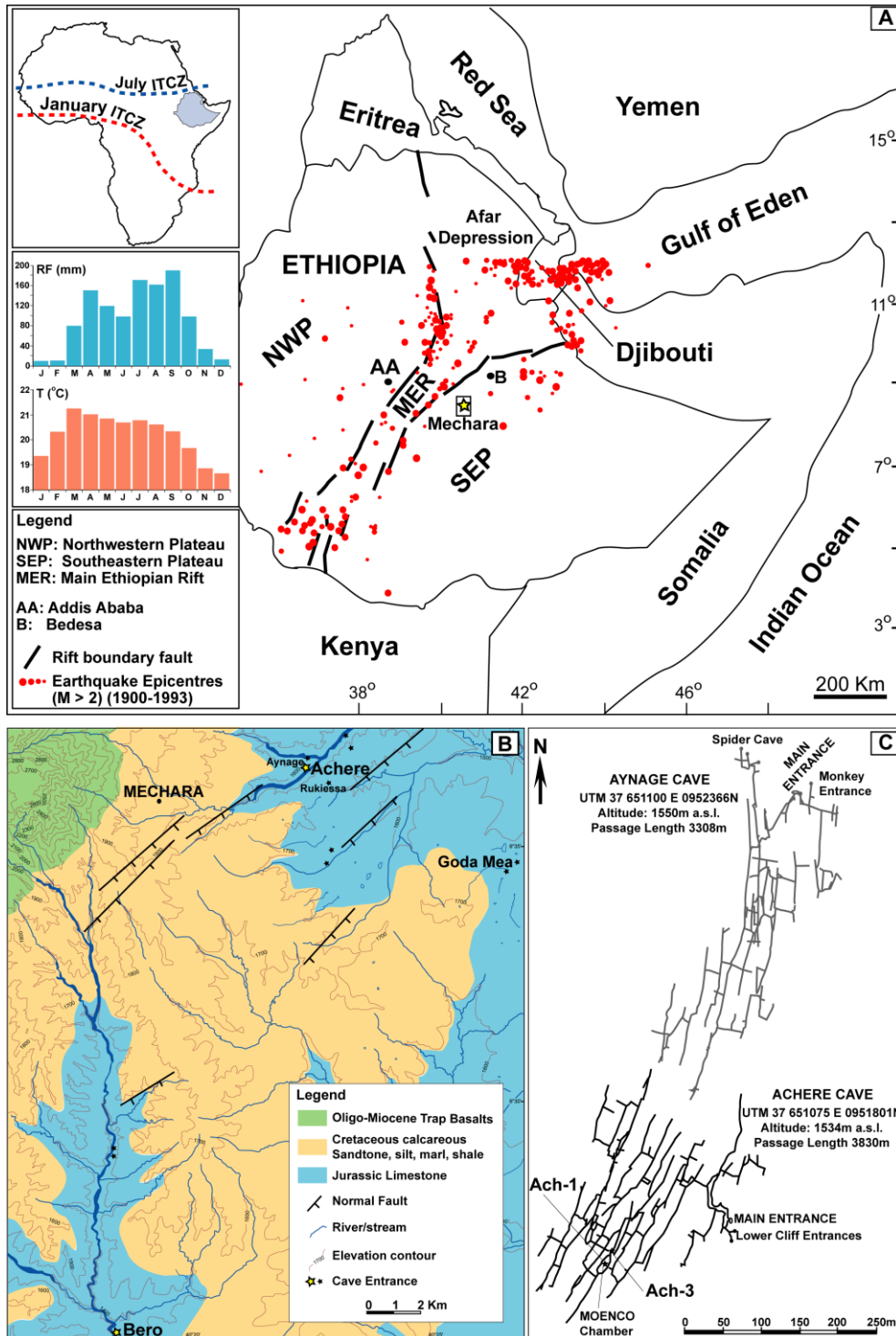
161

### 162 *2.1. Site Description*

163

164 The Achere cave forms part of the bigger Achere-Aynage cave system and has been  
165 previously described ([Asrat et al., 2007; 2008](#)). The Achere-Aynage cave system developed  
166 along numerous NE-SW oriented parallel rifts on the Southeastern Ethiopian highlands, close  
167 to the Main Ethiopian Rift (MER), indicating their development and modification through  
168 time in close association with rift forming processes ([Fig. 1](#)). The maze-like cave network  
169 developed within a narrow, 20-25 m vertical zone, parallel to the bedding of Jurassic  
170 limestone. A laterally extensive calcareous mudstone/marl horizon within the limestone  
171 currently marks the roof of the cave chambers ([Brown et al., 1998; Gunn and Brown, 1998;](#)  
172 [Asrat et al., 2007; 2008](#)).

173 The limestone terrain in the Mechara area including the top of the limestone beds  
174 forming the Achere-Aynage caves are overlain by very shallow (generally less than 50 cm  
175 deep) soils composed of lime-rich, soft calcareous layers overlain by dark organic rich humus  
176 layers, classified as rendzinas ([Bruggeman, 1986](#)). In the wider area, chromic cambisols  
177 develop over the sandstones and shales, which form low hills above the limestone sequence.  
178 These soils are in most parts strongly eroded ([Asrat et al., 2008](#)).



179

180 Figure 1. (A) Regional structural setting of Ethiopia showing the location of the Mechara caves. The  
 181 epicentres of the major earthquakes in the Main Ethiopian Rift and the adjoining highlands are marked  
 182 (Note that earthquake epicentres in the northern Afar depression are not represented). Insets show the  
 183 mean position of the ITCZ in July (summer) and January (winter) over Africa; and the mean monthly  
 184 rainfall (mm) and mean monthly temperature of the Mechara region, at the Bedesa Meteorological Station  
 185 (1994-2014 data from the Ethiopian Meteorological Agency). Location of Fig. 1(B) is marked by a solid  
 186 rectangle around the location of Mechara; (B) The topography, geology, structure and drainage system of  
 187 the Mechara karst area and locations of the entrances to the caves (including Aynage-Achere and Bero);  
 188 (C) Achere-Aynage cave survey showing the location of stalagmite Ach-3 and a previously published  
 189 stalagmite, Ach-1. Figures (A) and (B) modified from [Asrat et al. \(2008; 2018\)](#); Fig. (C) modified from  
 190 [Brown et al. \(1998\)](#).



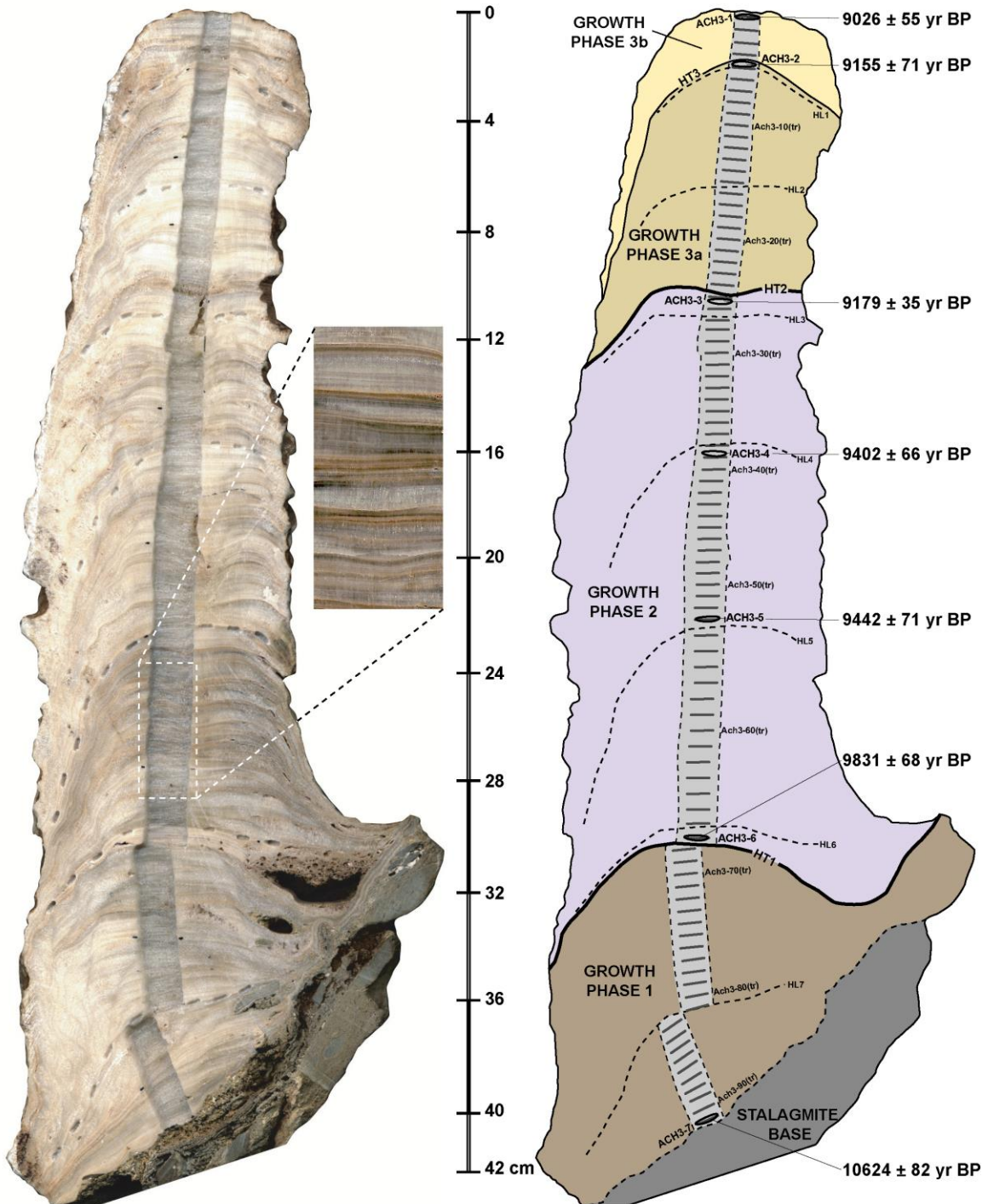
191 [Asrat et al. \(2008\)](#) reported cave monitoring data, which indicated that the Ach-3 stalagmite  
192 grew in a cave where modern relative humidity is  $87.5 \pm 11.5$  %, within cave  $p\text{CO}_2$  content is  
193  $745 \pm 365$  ppm, and has nearly constant within-cave temperature of  $\sim 20.5$  °C. Drip waters in  
194 the cave have  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  concentrations of  $3.13 \pm 1.88$  mmol/L and  $0.66 \pm 0.57$  mmol/L,  
195 respectively. Compared to the range of drip water  $\text{Ca}^{2+}$  concentration ( $= 2.63 \pm 2.36$  mmol/L)  
196 in all the monitored caves in Mechara, the Achere cave drip waters have distinctly higher  
197  $\text{Ca}^{2+}$  concentration implying “open system” evolution ([Baker et al., 2016](#)), where the  
198 calcareous (limestone, marl and carbonate rich mudstone) aquifer readily contributes  $\text{Ca}^{2+}$   
199 ions to the drip waters, and likely lead to rapid calcite formation which could be out of  
200 isotope equilibrium. Limited cave drip water oxygen isotope data from Achere cave  
201 demonstrate a limited range of  $\delta^{18}\text{O}$  composition from -1.6 to -0.5 ‰ (n=10) ([Asrat et al](#)  
202 [2008](#)).

203

## 204 *2.2. Sample description*

205

206 The Achere-Aynage cave system contains abundant speleothems. Ach-3 stalagmite was  
207 sampled in Achere cave in April 2004 from a narrow chamber leading to the bigger *Moenco*  
208 Chamber (where Ach-1 was sampled, [Asrat et al., 2007](#)), about 200 m from the cave  
209 entrance. Ach-3 developed on a low, narrow ledge 2 m beneath a roof marked by a mudstone  
210 layer. The chamber was dry and the speleothem was inactive at the time of sampling, though  
211 some soda straw stalactites in the vicinity of the chamber indicate recent seasonal dripping.  
212 Ach-3 is a 420 mm long, slender stalagmite, narrowing from the bottom (120 mm diameter)  
213 to the top (60 mm diameter; [Fig. 2](#)). The stalagmite was sectioned into two halves, and one  
214 half was polished and scanned at high resolution, on which lamina counting in triplicate has  
215 been conducted using Image analysis software. Continuous laminae were visible



- Sampling tracks for isotopes  
 — Sampling positions for trace elements  
 - - - HL1 - HL7: Hendy Test Lines  
 ○ ACH3-6 U-Th Dating sample positions  
 9026 ± 55 yr BP: U-Th ages (with 2 sigma errors) in yrs BP  
 — HT1 - HT2: Major Growth Hiatuses  
 - - - HT3: Minor Growth Hiatus

216  
 217  
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 222

Figure 2. Ach-3 hand-section in both scanned image (left) and sketch (right), showing the four growth phases, locations of the major and minor growth hiatuses, and sampling for isotopes, trace elements and U-Th analyses, and U-Th ages. The central panel is a sample of a high-resolution scan (not to scale) along the central growth axis showing the annual laminae of Ach-3.

223 throughout the sample marked by changes in calcite fabric, alternating between brownish  
224 dense and white porous calcite layers (Fig. 2). Some growth hiatuses are apparent marking  
225 slight shifts in the growth axis. The other half of Ach-3 was milled down its long-profile  
226 using a hand-held dental drill for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  analysis at  $\sim 0.51$  mm resolution (825  
227 samples), and trace element analysis at  $\sim 4.6$  mm resolution (91 samples). Additional samples  
228 for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  were also drilled following some individual growth layers in order to  
229 perform the “Hendy test”. The fast growth rate of individual lamina of Ach-3 (with lamina  
230 width ranging between 0.2 and 1.3 mm and average width of 0.45 mm), allows drilling of  
231 individual growth layers even at the flanks of the stalagmite. Seven samples for U-Th dating  
232 were similarly drilled using a dental drill, with samples located at the top and base of the  
233 stalagmite, on either side of possible growth hiatuses, and regularly spaced within growth  
234 phases (Fig. 2).

235

### 236 *2.3. Geochemical analyses*

237

238 Our methods follow those previously published in [Asrat et al. \(2007\)](#), [Baker et al. \(2010\)](#) and  
239 [Asrat et al. \(2018\)](#). A total of 825 samples for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  analysis were milled down the  
240 long-profile of the sample.  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  were analysed at the National Environmental  
241 Isotope Facility at Keyworth, UK. The calcite samples were reacted with phosphoric acid and  
242 cryogenically purified before mass spectrometry using an Isoprime plus multiprep dual inlet  
243 mass spectrometer. The “Hendy test” samples were analysed at the University of New South  
244 Wales (UNSW, Sydney) Analytical Centre using a MAT 253 mass spectrometer using a Kiel  
245 carbonate device. By comparison with a laboratory marble standards KCM (Keyworth) and  
246 IAEA603 (UNSW), the sample  $^{18}\text{O}/^{16}\text{O}$  and  $^{13}\text{C}/^{12}\text{C}$  ratios are reported as  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$

247 values in per mil (‰) versus VPDB. Analytical precisions are 0.07 ‰ for  $\delta^{18}\text{O}$  and 0.04 ‰  
248 for  $\delta^{13}\text{C}$  on the standard marble (KCM) and 0.05 ‰ for  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  (IAEA603).

249 Trace elements were analysed from 91 powders at UNSW, Sydney. Samples of  
250 approximately 0.05 g were dissolved in 1:1 hydrochloric acid, diluted, and analysed for Ca  
251 and Mg using the PerkinElmer Optima™ 7300DV ICP-OES. Ba, Sr, Al, Cu, Fe, K, Na, P,  
252 Pb, S, Zn and U were analysed by PerkinElmer NexION 300D ICP-MS.

253 Seven U-Th analyses were performed in the Uranium Series Chronology Laboratory,  
254 Institute of Geology and Geophysics, Chinese Academy of Sciences. The powdered sub-  
255 samples of approximately 0.1 g were totally dissolved and spiked with a mixed  $^{229}\text{Th}$ - $^{233}\text{U}$ -  
256  $^{236}\text{U}$ . Uranium and thorium fractions were separated on 2 ml anion exchange columns  
257 following standard techniques (Edwards et al., 1987). Then, the separated uranium and  
258 thorium solutions were measured on a multi-collector inductively coupled plasma mass  
259 spectrometer (MC-ICP-MS, Neptune plus). The procedures followed those described in  
260 Cheng et al. (2013).

261

#### 262 *2.4. Time series analysis*

263

264 Variogram analysis on the annual growth rate time series was undertaken to determine the  
265 flickering parameter ( $f$ ), information content (IC) and range ( $r$ ) (Mariethoz et al., 2012).

266 Flickering quantifies the growth rate acceleration from one year to the next through the lag-

267 one autocorrelation of the detrended growth rate series, where 0 is no flickering (regular

268 increases or decreases in growth rate) and -0.5 is the signal obtained from white noise. The

269 observed flickering parameter (Mariethoz et al., 2012; Asrat et al., 2018) typically ranges

270 between -0.5 and 0, the more negative  $f$  values indicating stronger flickering, interpreted as

271 large changes in growth rate from year to year, indicative of a karst store filling and draining.

272 To enable such a large inter-annual variability whilst maintaining continuous deposition over  
273 hundreds of years, a sufficiently large volume karst store is hypothesised. Other statistical  
274 measures of information contained in the growth rate data are the *IC* and the *r*. *IC* quantifies  
275 the proportion of correlated signal in the time series as opposed to noise, and varies between  
276 0% (pure noise) to 100% (noiseless correlated signal). Range (*r*) is the temporal range of the  
277 correlated part of the signal, i.e., the time over which useful information might be expected to  
278 be obtained from a growth rate time series.

279 Stable isotope and annual growth rate time series data were analysed for their spectral  
280 properties. Spectral analysis was performed using the SPECTRUM software for unevenly  
281 spaced paleoclimate timeseries (Schulz and Statteger, 1997). Lomb-Scargle Fourier  
282 transforms were conducted, with five windows used (Bartlett, Hanning, Rectangular, Welsh  
283 and Triangular) in order to undertake the spectral analysis of oxygen, carbon and growth rate  
284 time series, and the coherency between isotope time series. The autocorrelation of the stable  
285 isotope time series was investigated by determining the autocorrelation function.

286

### 287 **3. Results and Interpretation**

288

#### 289 *3.1. Chronology*

290

291 Stalagmite Ach-3 is laminated throughout with 925 laminae. In hand-section, likely growth  
292 hiatuses with dissolution features were visually identified at lamina number 675 (growth  
293 hiatus 1) and 243 (growth hiatus 2) from the top, separating the sample into three growth  
294 phases (Fig. 2): growth phase 1 (laminae 925-676); growth phase 2 (laminae 675-244), and  
295 growth phase 3 (laminae 243-1). A third possible growth hiatus was identified near the end of  
296 the third growth phase (growth phase 3a: laminae 243-28; growth phase 3b: laminae 27-1).

297           The results of seven U-Th analyses are provided in [Table 1](#). An age-depth model  
298    which confirms the three growth phases is given in [Figure 3](#). A basal date of  $10,624 \pm 82$   
299    years BP constrains the initial growth phase containing 243 laminae. The six other U-Th ages  
300    occurred in stratigraphic order from  $9,831 \pm 68$  years BP to  $9,026 \pm 55$  years BP. The  
301    stratigraphically youngest three ages are all very similar to one another, despite the presence  
302    of possible hiatuses, suggesting that they were of short duration. The close agreement  
303    between the duration of stalagmite formation after hiatus 1 as determined by U-Th (the  
304    difference between the corrected U-Th ages ACH3-1 and ACH3-6 of  $805 \pm 93$  years,  $1\sigma$ ) and  
305    the number of laminae (675 laminae) is indicative that the laminae of Ach-3 are annual in  
306    nature. This would agree with the widespread observation of annual laminae in other  
307    Ethiopian speleothems, which is due to the strong seasonality of rainfall with a distinct dry  
308    season ([Asrat et al., 2007; 2018; Baker et al., 2007; 2010](#)). Ach-3 lamina thickness has an  
309    average of 0.45 mm, and this is equivalent to the annual accumulation rate observed in  
310    Holocene and last interglacial Ethiopian stalagmites: Ach-1 (0.53 mm/yr); Bero-1 (0.45  
311    mm/yr), Merc-1 (0.29 mm/yr); Asfa-3 (0.32 mm/yr) and GM-1 (0.44 mm/yr) ([Asrat et al](#)  
312    [2007; 2019; Baker et al 2007; 2010](#)). We are therefore confident that the laminae are annual  
313    in nature.

314 Table 1.  $^{230}\text{Th}$  dating of stalagmite Ach-3. The error is  $2\sigma$ .

Sample	Distance from top	$^{238}\text{U}$	$^{232}\text{Th}$	$^{230}\text{Th}/^{232}\text{Th}$	$\delta^{234}\text{U}^*$	$^{230}\text{Th}/^{238}\text{U}$	$^{230}\text{Th}$ Age (yr)	$^{230}\text{Th}$ Age (yr)	$\delta^{234}\text{U}_{\text{initial}}^{**}$	$^{230}\text{Th}$ Age (yr BP) $^{***}$
Number	(mm)	(ppb)	(ppt)	(atomic $\times 10^{-6}$ )	(measured)	(activity)	(uncorrected)	(corrected)	(corrected)	(corrected)
ACH3-1	0.84	1467 $\pm 5$	1904 $\pm 38$	1338.3 $\pm 27.5$	315.6 $\pm 3.0$	0.1054 $\pm 0.0005$	9074 $\pm 51$	<b>9045 <math>\pm 55</math></b>	324 $\pm 3$	<b>9026 <math>\pm 55</math></b>
ACH3-2	25.09	1305 $\pm 3$	1781 $\pm 36$	1294.2 $\pm 27.3$	319.5 $\pm 2.4$	0.1071 $\pm 0.0007$	9203 $\pm 67$	<b>9174 <math>\pm 71</math></b>	328 $\pm 2$	<b>9155 <math>\pm 71</math></b>
ACH3-3	104.74	1010 $\pm 1$	1752 $\pm 35$	1021.5 $\pm 20.6$	318.8 $\pm 1.5$	0.1074 $\pm 0.0002$	9236 $\pm 23$	<b>9198 <math>\pm 35</math></b>	327 $\pm 2$	<b>9179 <math>\pm 35</math></b>
ACH3-4	162.93	1299 $\pm 2$	1953 $\pm 39$	1198.5 $\pm 25.1$	311.8 $\pm 1.6$	0.1093 $\pm 0.0007$	9454 $\pm 62$	<b>9421 <math>\pm 66</math></b>	320 $\pm 2$	<b>9402 <math>\pm 66</math></b>
ACH3-5	231.21	1344 $\pm 2$	1827 $\pm 37$	1329.1 $\pm 28.1$	310.6 $\pm 2.0$	0.1096 $\pm 0.0007$	9491 $\pm 68$	<b>9461 <math>\pm 71</math></b>	319 $\pm 2$	<b>9442 <math>\pm 71</math></b>
ACH3-6	301.02	1571 $\pm 2$	4158 $\pm 83$	707.3 $\pm 14.6$	302.8 $\pm 1.5$	0.1135 $\pm 0.0006$	9908 $\pm 54$	<b>9850 <math>\pm 68</math></b>	311 $\pm 2$	<b>9831 <math>\pm 68</math></b>
ACH3-7	401.74	1806 $\pm 4$	4551 $\pm 91$	825.7 $\pm 17.3$	345.5 $\pm 3.0$	0.1262 $\pm 0.0008$	10697 $\pm 73$	<b>10643 <math>\pm 82</math></b>	356 $\pm 3$	<b>10624 <math>\pm 82</math></b>

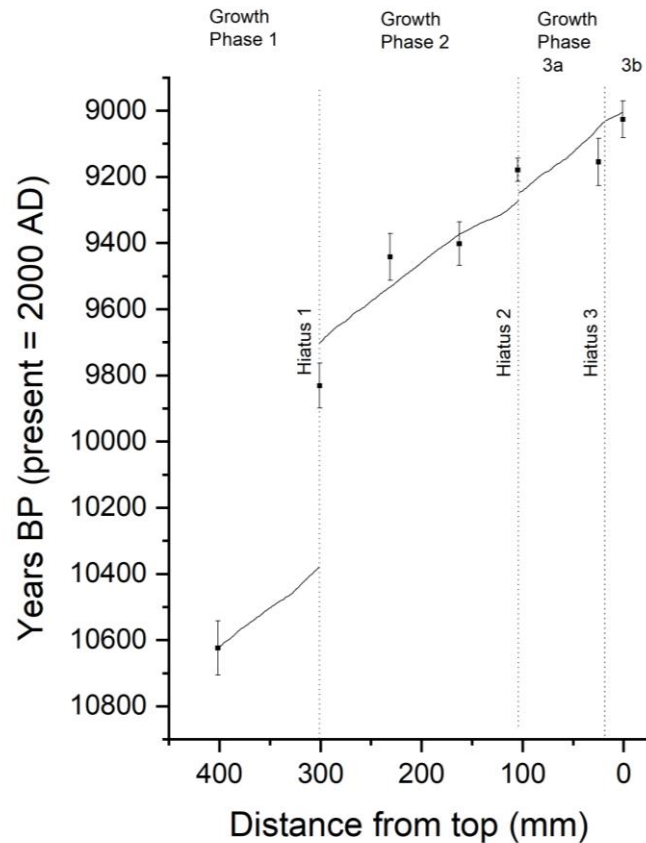
315

316  $^*\delta^{234}\text{U} = ([^{234}\text{U}/^{238}\text{U}]_{\text{activity}} - 1) \times 1000$ .  $^{**}\delta^{234}\text{U}_{\text{initial}}$  was calculated based on  $^{230}\text{Th}$  age (T), i.e.,  $\delta^{234}\text{U}_{\text{initial}} = \delta^{234}\text{U}_{\text{measured}} \times e^{\delta^{234}\text{U} \times T}$ .

317 Corrected  $^{230}\text{Th}$  ages assume the initial  $^{230}\text{Th}/^{232}\text{Th}$  atomic ratio of  $4.4 \pm 2.2 \times 10^{-6}$ .

318 Those are the values for a material at secular equilibrium, with the bulk earth  $^{232}\text{Th}/^{238}\text{U}$  value of 3.8. The errors are arbitrarily assumed to be 50%.

319  $^{***}\text{B.P.}$  stands for “Before Present” where the “Present” is defined as the year 2000 A.D.



320

321 Figure 3. An age depth model for Ach-3. Depth measured as distance (mm) from the top of the speleothem.  
 322 Locations of ages and hiatuses are marked.

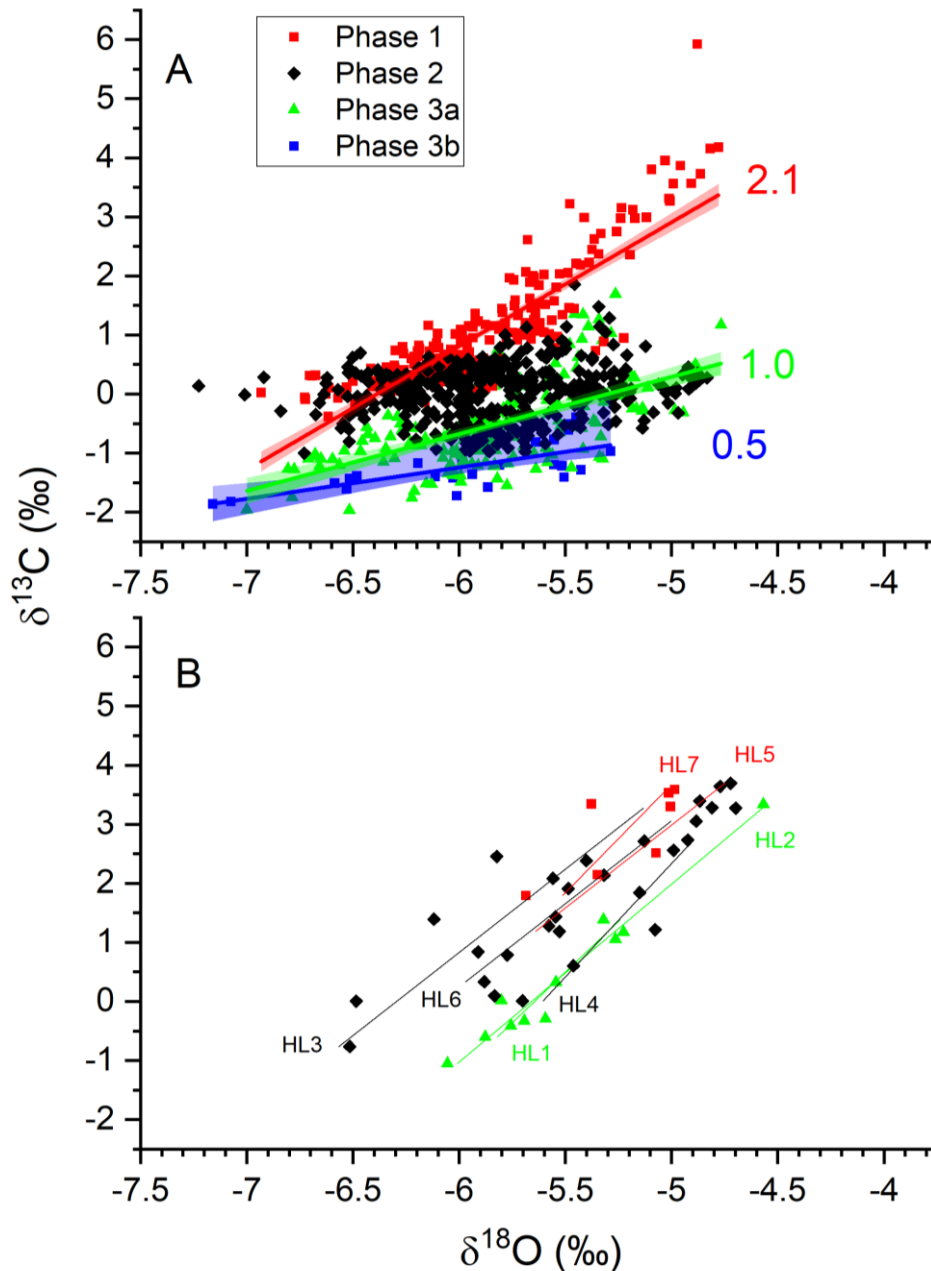
323

### 324 3.2. Geochemical proxies

325

326 The 825  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  analyses are presented in Figure 4A as scatter plots of oxygen vs  
 327 carbon isotopes down the growth axis, as well as for analyses made along six growth laminae  
 328 (Fig. 4B) equivalent to the classic ‘Hendy test’ (Hendy, 1971). Figure 4A shows that the two  
 329 isotopes are positively correlated along the growth axis in all growth phases except for  
 330 growth phase 2, and Figure 4B shows that the two isotopes are positively correlated along all  
 331 sampled growth laminae, including those in growth phase 2. This correlation between  $\delta^{13}\text{C}$   
 332 and  $\delta^{18}\text{O}$  is similar to other Ethiopian stalagmites (Asrat et al., 2007; 2018; Baker et al.,  
 333 2010), and demonstrates that deposition is not in equilibrium (Fantadis and Ehhalt, 1970;  
 334 Mickler et al., 2006; Wiedner et al., 2008). The gradient of  $\delta^{13}\text{C}/\delta^{18}\text{O}$  is between 3.0 and 3.5





335

336 Figure 4. Scatter plots of  $\delta^{18}\text{O}$  vs  $\delta^{13}\text{C}$  (A) for each growth phase (B) ‘Hendy’ tests along growth laminae in  
 337 stalagmite Ach-3. Note that similar dis-equilibrium deposition was observed in Ach-1 and Bero-1 (Asrat et al.,  
 338 2007; Baker et al., 2010).

339

340 along growth laminae, and for growth phases 1, 3a and 3b is 2.1, 1.0 and 0.5, respectively,

341 with no correlation between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  in growth phase 2. These gradients observed in

342 stalagmite Ach-3 are similar to the mean value of the gradient of  $\delta^{13}\text{C}/\delta^{18}\text{O}$  of 3.8 observed

343 along vertical transects and 3.9 observed spatially across calcite deposited on glass plates by

344 Mickler et al. (2006). These were attributed to kinetic fractionation during non-equilibrium

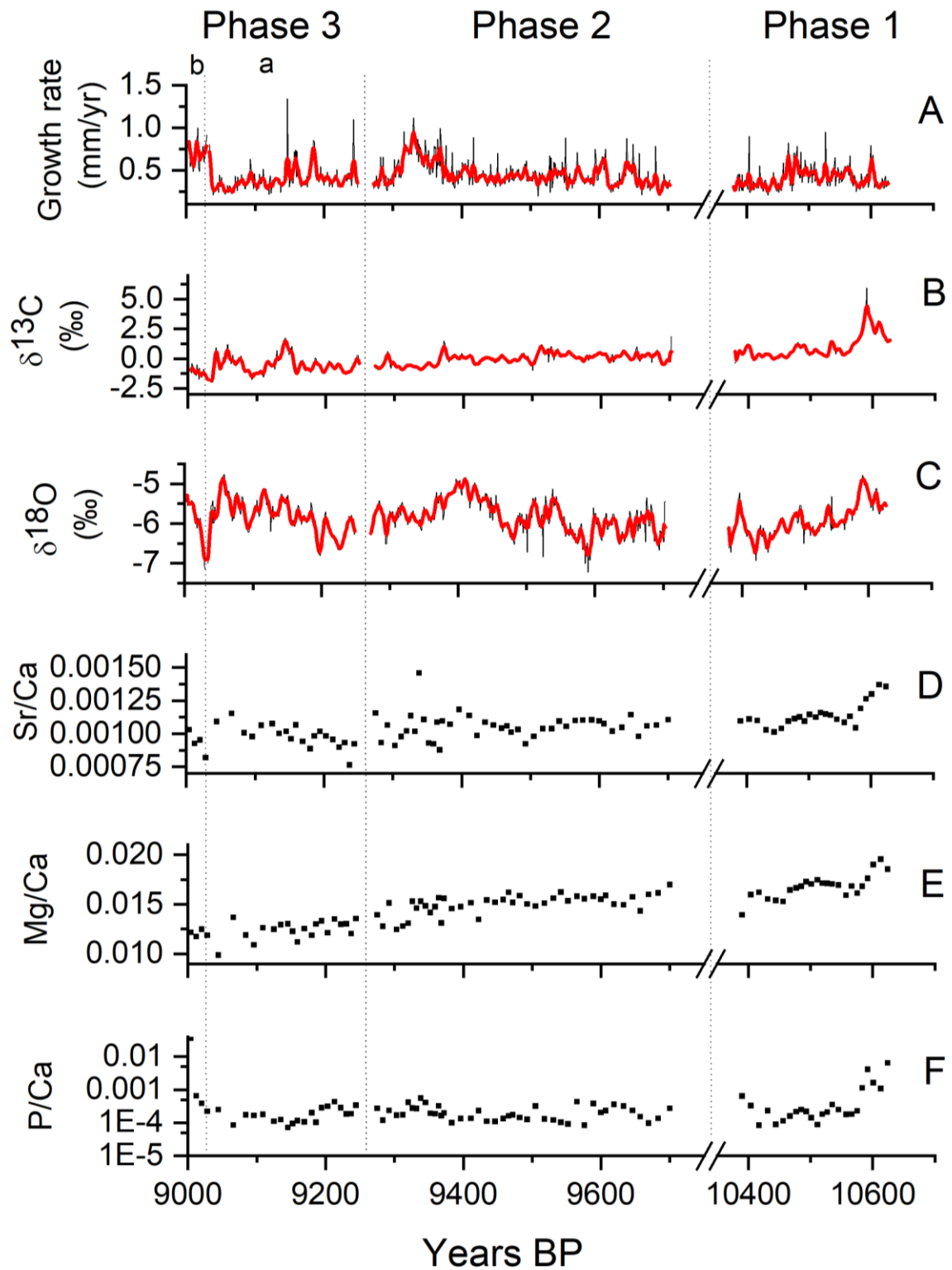
345 deposition due to  $^{18}\text{O}$  and  $^{13}\text{C}$  Rayleigh-distillation enrichment in the  $\text{HCO}_3^-$  reservoir during  
346 progressive  $\text{CO}_2$  degassing and calcite precipitation. They are also similar to the gradient of  
347  $\delta^{13}\text{C}/\delta^{18}\text{O}$  of  $1.4 \pm 0.6$  for the fast-degassing of  $\text{CO}_2$  in carbonate precipitation experiments  
348 (Wiedner et al., 2008). Though the classic “Hendy test” might not be conclusive in predicting  
349 the equilibrium or non-equilibrium deposition of calcite (e.g., Dorale and Liu, 2003), our  
350 cave monitoring and modern speleothem records from the Mechara caves further confirm that  
351 non-equilibrium deposition is likely for Ach-3. The lowest values of the predicted  
352 equilibrium calcite  $\delta^{18}\text{O}$  variations from measured modern drip water  $\delta^{18}\text{O}$  data in various  
353 caves in the region are not observed in speleothem  $\delta^{18}\text{O}$  records, indicating non-equilibrium  
354 deposition of calcite (Baker et al., 2007; Asrat et al., 2008). However, in Ach-3 we note a  
355 trend over time in the  $\delta^{13}\text{C}/\delta^{18}\text{O}$  gradient, and extent of non-equilibrium deposition. In the  
356 last years of deposition (Phase 3), the gradient is 1.0 (Phase 3a) and 0.5 (Phase 3b), which  
357 could indicate a change in the extent or type of isotope fractionation, for example additional  
358 evaporative fractionation due to slower drip rates, and / or increased kinetic fractionation due  
359 to increased drip water  $p\text{CO}_2$ .

360 Trace element data for the 91 samples is presented in Supplemental Table 1. Elements  
361 were normalised to calcium and analysed using PCA (Supplemental Figure 1). Three  
362 components explained 80 % of the variability in the data. PC1 (36 % of the variance  
363 explained) correlated with the elements P, Na, K and Zn; PC2 (22 % of the variance  
364 explained) correlated with Mg, Sr, and U; and PC3 (22 % of the variance explained)  
365 correlated with Fe, Al, Ba and Pb. We interpret PC1 as soil or cave sediment derived  
366 elements, given the presence of nutrients and organic-associated metals (Borsato et al., 2007;  
367 Hartland et al., 2012). PC2 is interpreted as bedrock-derived dissolution elements, and PC3 as  
368 elements derived from sediment, colloidal and particulate material (Borsato et al., 2007).  
369 Time series of the three principal components shows that all three components have high

370 scores at the start of growth, and decline over the first growth phase ([Supplemental Figure 2](#)).  
371 PC2 then has a long-term decrease over the rest of the period of deposition, indicative of a  
372 decrease in bedrock-derived metals over time ([Supplemental Figure 2](#)). PC1 increases to its  
373 highest value, and PC2 increases by a lesser amount, over the last years of deposition, while  
374 at the same time PC3 decreases to its lowest score.

375         The time series for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  are presented in [Figure 5](#), together with  
376 representative trace element data for PC1 (P/Ca) and PC2 (Sr/Ca, Mg/Ca) and annual growth  
377 rates. The 825 isotope analyses represent an approximately annually resolved record. In the  
378 first deposition phase, from ~10,600 – ~10,350 BP, there is a trend towards lower ratios in  
379 Mg/Ca, Sr/Ca, and more negative  $\delta^{18}\text{O}$ , indicative of generally increasingly wetter conditions  
380 or a shorter vadose zone water residence time. The observation of higher concentrations of  
381 soil or cave sediment derived elements such as P at the start of deposition agrees with the  
382 observation of detrital material in the bottom few growth laminae, likely derived from cave  
383 sediments, as well as the subsequent flushing of soluble elements from the soil and vadose  
384 zone at the start of the growth phase.

385         Stalagmite deposition from ~9,700 to ~9,000 years BP in growth phases 2 and 3a has  
386 a long-term trend to more negative  $\delta^{13}\text{C}$  and lower Sr/Ca and Mg/Ca. This could be indicative  
387 of the continuation of the trend to increasingly wetter conditions or a shorter vadose zone  
388 water residence time and decreasing prior calcite precipitation along the flow path over this  
389 period ([Fairchild et al., 2000](#)). Growth rates and oxygen isotope composition exhibit no long-  
390 term trend, instead have multi-decadal variability.



391

392 Figure 5. Time series of growth rate and geochemical proxies in Ach-3. (A) Annual growth rate, (B)  $\delta^{13}\text{C}$ , (C)  
 393  $\delta^{18}\text{O}$ , (D) Sr/Ca, (E) Mg/Ca, (F) P/Ca.

394

395 Geochemical and isotope data are unchanged over the possible short-duration growth phase

396 3b at ~9300 BP. However, over this last 28 years of deposition, geochemical trends reverse,

397 with increasing PC1 and decreasing PC3 in the last years of deposition ([Supplemental Figure](#)  
398 [2](#)), increases in Sr, Mg, and  $\delta^{18}\text{O}$ , and an increase in growth rate. A change in geochemical,  
399 growth rate and isotopic trends has been observed previously at the end of stalagmite  
400 deposition during Middle and Late Holocene ([Asrat et al., 2007; 2018](#)) and interpreted as a  
401 change in hydrological regime as the hydroclimate dries, e.g. disconnection from the soil  
402 water store or decrease in fracture flow component. In these records, the role of active  
403 tectonics in controlling speleothem growth duration by changing the flow regimes has been  
404 identified.

405 The mean  $\delta^{18}\text{O}$  composition of the Early Holocene Ach-3 ( $-5.86 \pm 0.42\text{‰}$ ) is more  
406 negative compared to all other modern (Merc-1:  $-1.22 \pm 0.31\text{‰}$ ; Asfa-3:  $-1.37 \pm 0.37\text{‰}$ ;  
407 Baker et al., 2007), and Middle to Late Holocene (Bero-1:  $-3.42 \pm 1.45\text{‰}$ , [Baker et al. 2010](#);  
408 Ach-1:  $-3.20 \pm 0.35\text{‰}$ , [Asrat et al., 2007](#)) samples from the region. All the published  
409 stalagmite records have evidence of non-equilibrium deposition. Assuming a similar extent of  
410 disequilibrium in all the stalagmites, including Ach-3, it indicates that drip water was  $\sim 2\text{‰}$   
411 more negative in the Early Holocene (Ach-3:  $-5.86 \pm 0.42\text{‰}$ ) compared to that of Middle  
412 Holocene (Ach-1:  $-3.20 \pm 0.35\text{‰}$ ; Bero-1:  $-3.42 \pm 1.45\text{‰}$ ).

413

### 414 *3.3. Time series analysis*

415

416 A summary of the results of spectral analysis on both stable isotopes and growth rate time  
417 series, and variogram analysis and flickering of growth rate time series, is presented in [Table](#)  
418 [2](#). Full spectral analysis results are presented in [Supplemental Table 2](#) and [Supplemental](#)  
419 [Figure 3](#), and autocorrelation plots in [Supplemental Figure 4](#).

420 Variogram analysis of growth rates time series suggests this is a stalagmite with a  
421 relatively short time period where there is useful climate information in the growth rate data.

422 Table 2. (A) variogram analysis for stalagmite Ach-3 for the three growth phases (phase 1 – oldest; phase 3 – youngest): range  $r$ ; information  
 423 content IC; flickering  $f$ . (B) Summary of geostatistical properties for Ach-3, Bero-1 and Ach-1: univariate spectral analysis, showing the dominant  
 424 and statistically significant periodicities in the oxygen isotope, carbon isotope and growth rate time series. Oxygen and carbon isotope time series  
 425 have coherent periodicities at 15-16 years (Ach-3), 16-17 and 25 years (Bero-1) and 16-17 years (Ach-1). Summary variogram statistics  $r$  and  $f$ .

(A) Variogram analysis for stalagmite Ach-3

Phase 3: 28-243	Phase 2: 244-675	Phase 1: 676-925
$r = 12.1$ years	$r = 28.4$ years	$r = 13.0$ years
IC = 66.7%	IC = 64.3%	IC = 50.5%
$f = -0.34$	$f = -0.38$	$f = -0.26$

(B) Geostatistical summary for Ach-3, Bero-1 and Ach-1 (period in years)

Statistically significant periodicities (years)					Variogram Analysis	
	$\delta^{18}\text{O}$	$\delta^{13}\text{C}$	Growth rate	$r$	$f$	
<u>Ach-3</u>						
~9.2 ka	18-25	19-21	$\geq r$	13	-0.26	
~9.4 ka	17-18	19-21	11	28	-0.38	
~10.4 ka	25-30	19-23	$\geq r$	12	-0.35	
<u>Bero-1</u>						
~4.3–4.2 ka	8, 13, 26-28	8, 13, 16-17, 26-28	11–15, 17–21	6	-0.36	
~5.4–5.2 ka	16-20	18-21, 26	10–13, 15–20	$>^*$	-0.74	
~7.8–7.5 ka	28-30	21, 27-30	11–13, 18–19	335	-0.51	
<u>Ach-1</u>						
~5.1–4.7 ka	18–21	18-19	18–21	80	-0.36	

427 These are the range,  $r$ , = 28 years in growth phase 2, and a much shorter range of  $r$  = 12-13  
428 years in growth phases 1 and 3 (Table 2A). The  $r$  values are low compared to a global  
429 analysis of the growth rates of laminated stalagmites in Mariethoz et al. (2012) and Baker et  
430 al (2021), but similar to other Ethiopian samples. The information content,  $IC$ , in the growth  
431 rate time series ranges from 0.50 to 0.67, highest and relatively similar in growth phases 2  
432 and 3. An  $IC$  over 50% means that the stalagmite growth rate data contains significant useful  
433 signal. An  $IC > 50\%$  and  $r < 150$  years classifies Ach-3 as a "Type A" stalagmite of  
434 Mariethoz et al. (2012), which is likely to be suitable for interpreting multi-decadal  
435 information, with the higher  $IC$  in phases 2 and 3 suggesting that these are less noisy. The  
436 presence of flickering,  $f$ , of -0.26 (phase 1), -0.37 (phase 2) and -0.34 (phase 3), is indicative  
437 of a water filled store supplying the stalagmite of sufficient volume to maintain continuous  
438 deposition for at least several decades, with hydrologically-controlled year-by-year variations  
439 in water level controlling inter-annual growth rate variations. Phase 1 of deposition has a  
440 lower  $IC$  and relatively short range, and suggests that the first growth phase contains the least  
441 climate information.

442 Inspection of the autocorrelation of  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  time series for each growth phase  
443 (Supplemental Figure 4) shows that the autocorrelation for both stable isotopes is similar to  
444 each other for growth phases 1 and 3. Between growth phases, there is a slight decrease in  
445 autocorrelation from growth phase 1 to growth phase 3, and a slight decoupling of the  $\delta^{13}\text{C}$   
446 and  $\delta^{18}\text{O}$  autocorrelation functions in growth phase 2. If soil processes were the dominant  
447 control on speleothem  $\delta^{13}\text{C}$ , a stronger autocorrelation in  $\delta^{13}\text{C}$  compared to  $\delta^{18}\text{O}$  might be  
448 expected. This is not observed in Ach-3. The lower autocorrelation of  $\delta^{13}\text{C}$  compared to  $\delta^{18}\text{O}$   
449 in growth phase 2 agrees with the observed lack of correlation between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$   
450 through time in growth phase 2, and a possible decrease in the extent or type of isotope  
451 fractionation in this phase. Overall, the similarity in the autocorrelation functions of  $\delta^{13}\text{C}$  and

452  $\delta^{18}\text{O}$ , combined with the evidence of isotope fractionation from the correlation between  $\delta^{13}\text{C}$   
453 and  $\delta^{18}\text{O}$  over time and along growth layers, suggests the dominant control of in-cave isotope  
454 fractionation processes on the composition of both  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ , strongest in growth phases  
455 1 and 3.

456 Spectral analysis on the  $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$  and growth rate time series is presented in [Table](#)  
457 [2B](#) and [Supplemental Figure 3](#). There are similar and consistent periodic components in the  
458  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  time series at around 15-25 years and 19-25 years in all three growth phases.  
459 Bivariate analysis of  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  demonstrates a coherency at 16-17 years. In growth  
460 phases 1 and 3, these periodic components in the stable isotope time series occur at time  
461 periods greater than the value of  $r$  obtained from the growth rate data, suggesting an  
462 independent forcing mechanism is dominant. Evidence that isotope fractionation is occurring  
463 during deposition, and that this is likely to be from within-cave fractionation processes,  
464 suggest that within-cave isotope fractionation processes are the dominant driver of the  
465 observed multi-decadal periodicity in the stable isotope time series. These within-cave  
466 isotope fractionation processes can be climatically forced, and we cautiously interpret these  
467 spectral frequencies as representative of an indirect hydroclimatic forcing affecting in-cave  
468 isotope fractionation processes. Spectral analysis on the growth rate timeseries demonstrates  
469 that there are no periodic signals shorter than the range,  $r$ , for all growth phases ([Table 2B](#)).  
470 [Table 2B](#) also presents the results of previously published spectral analyses on Holocene  
471 Ethiopian stalagmites, demonstrating a consistent multidecadal periodic signal in  $\delta^{18}\text{O}$  time  
472 series between different time periods and different caves.

473

## 474 **4. Discussion**

475

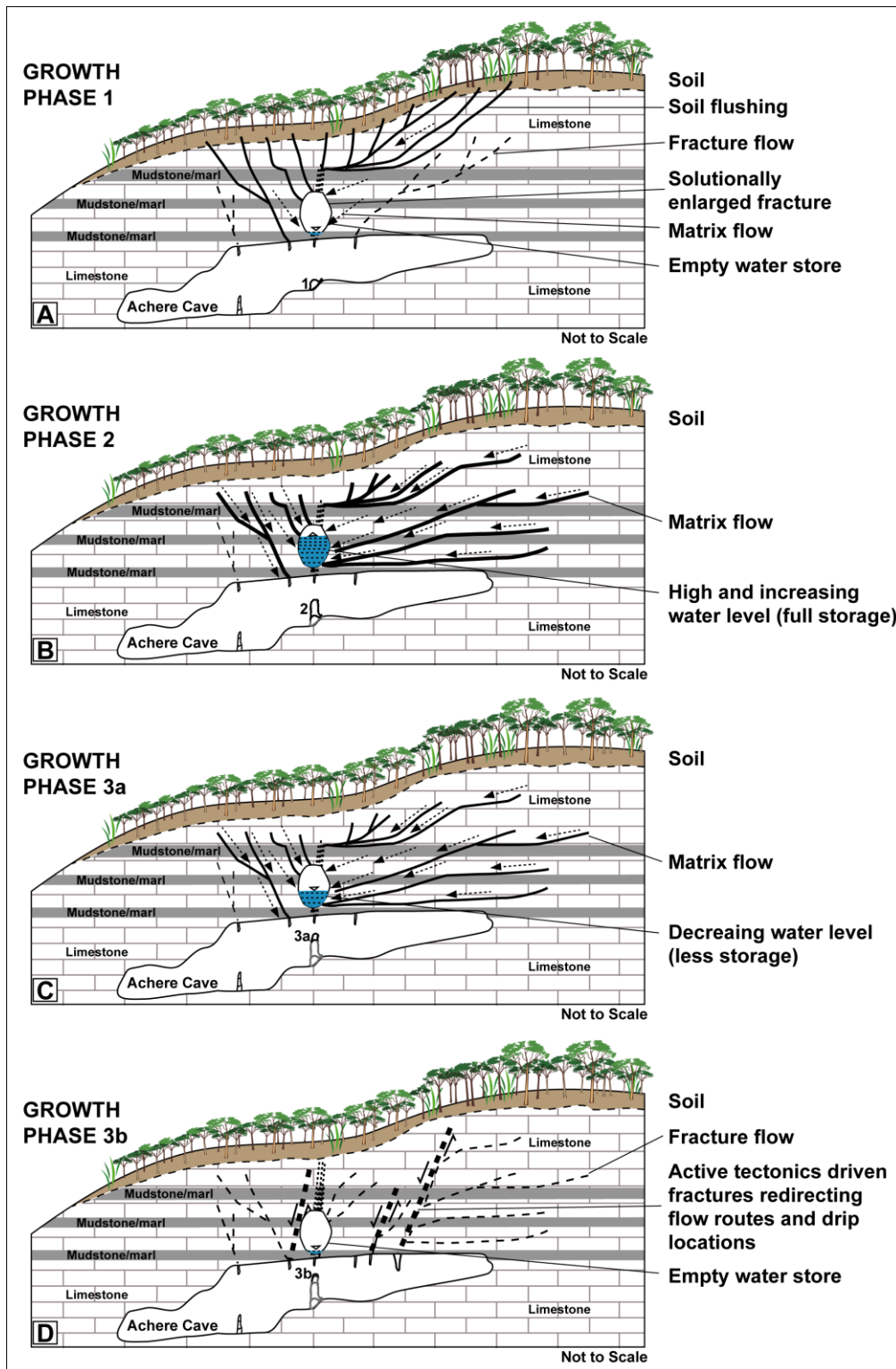
### 476 *4.1. Conceptual growth model*



477

478 Stable isotope and trace element geochemical data and geostatistical analyses, combined with  
479 our hydrogeological understanding of the unsaturated zone properties of the limestone (Asrat  
480 et al., 2007), suggest that stalagmite Ach-3 is fed by a mixture of diffuse flow, through  
481 porous limestone and calcareous mudstone, as well as solutionally enlarged fractures. The  
482 latter are relatively small in volume and more important than diffuse flow contributions, as  
483 indicated by the 28-year range in growth rate time series. This is indicative of a relatively  
484 small water store which controls growth rate variability through limits on the extent of prior  
485 calcite precipitation (PCP) in the fracture, and can determine drip rate. Considering the whole  
486 period of stalagmite formation, trace element data identifies an initial sediment or soil-  
487 derived elemental signal, potentially indicative of an initial flush of trace elements from the  
488 soil or interactions with cave sediments, and a loss of this elemental signal in the last decade  
489 of deposition. The duration of this last growth phase is the same as the range in the variogram  
490 analysis of growth rate and consistent with the inferred small water volume of the karst  
491 fracture.  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  have very similar autocorrelation functions, have coherent, periodic  
492 signals in the timeseries, and strongly correlate between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  along growth laminae  
493 and within growth phases. This indicates a common control on both isotopes of within-cave  
494 isotope fractionation. Multidecadal variability in stable isotopes is therefore due to changes in  
495 the extent of isotope fractionation, either through kinetic or disequilibrium fractionation  
496 processes, such as changes in drip rate or drip water calcite saturation that control the extent  
497 of  $^{18}\text{O}$  and  $^{13}\text{C}$  enrichment in the  $\text{HCO}_3^-$  water film during progressive  $\text{CO}_2$  degassing and  
498 stalagmite precipitation (Mickler et al., 2006; Scholz et al., 2009).

499 We present a conceptual model of the hydrogeochemistry associated stalagmite  
500 growth in Figure 6. In growth phase 1, there is an initial input of soil or sediment derived  
501 material.



502

503 Figure 6. Conceptual model for the deposition of stalagmite Ach-3: (A) Growth phase 1: initiation and flushing  
 504 from soil dominating the flow; (B) Growth phase 2: wet and continuous growth from full storage, with multi-  
 505 decadal variability due to within cave processes (such as drip rate or water saturation); (C) Growth phase 3a:  
 506 similar flow conditions to that of phase 2 but with less water storage; and (D) major tectonic process leading to  
 507 the redirecting of flow regimes and relocation of drip sources leading to rapid shutoff and growth cessation.  
 508 Cartoons modified from [Asrat et al. \(2007; 2018\)](#).

509

510           There is a low information content in the growth rate time series in this growth  
511 phase, indicating a relatively noisy signal due to the combination of growth rate controls from  
512 the initial flush of soil-derived material as well as a hydrological control. The periodic signal  
513 in the growth rate time series and range are identical, at ~12 years, suggesting relatively  
514 limited water storage to the stalagmite during this growth phase (indicated by an empty  
515 reservoir in Fig. 6A). In growth phase 2 the best information content and largest range is  
516 observed, which we interpret as the karst store relatively full of water (full storage reservoir  
517 in Fig. 6B) compared to other growth phases. Decreasing Sr/Ca and Mg/Ca ratios over this  
518 growth phase further indicate increasing water availability. In this growth phase the  $\delta^{13}\text{C}$  and  
519  $\delta^{18}\text{O}$  data show some evidence that isotope fractionation processes have less dominant  
520 control on isotopic composition than in the other phases. In growth phase 3a, the range in the  
521 growth rate time series analysis decreases, but all other proxies are identical to phase 2 and  
522 indicative of persisting high water availability (half storage reservoir in Fig. 6C). Throughout  
523 these growth phases there is a consistent multidecadal variability in  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ , which is  
524 interpreted as being forced by non-equilibrium deposition processes. Finally, in phase 3b, we  
525 have a 28-year period of deposition where trace element data indicates a decrease or loss of  
526 soil connectivity. This results in an increase in growth rate until growth cessation (an empty  
527 reservoir in Fig. 6D). Given the preceding growth indicated progressive increases in water  
528 availability, we infer that tectonic activity disrupted the water flow path to the stalagmite  
529 between growth phases 3a and 3b.

530

#### 531 *4.2. Non-equilibrium deposition and multidecadal variability in $\delta^{18}\text{O}$*

532

533 Spectral analyses on  $\delta^{18}\text{O}$  for the three stalagmites: Ach-3, and the previously published  
534 Bero-1, and Ach-1, demonstrate a multidecadal variability through the Holocene (Table 2 and

535 [Supplemental Figure 5](#)). The amplitude of this variability is ~1 ‰. The dominant statistically  
536 significant frequencies are between 13 and 30 years. We observe spectral frequencies in this  
537 range in stalagmites from different sites with different hydrogeology and flow paths. This  
538 multi-decadal variability in  $\delta^{18}\text{O}$  can (i) derive from water isotope fractionation process,  
539 including partial evaporation of soil and shallow epikarst water, and within-cave fractionation  
540 due to changes in the extent of isotopic dis-equilibrium during stalagmite formation (drier  
541 and increased drip water  $p\text{CO}_2$  = more positive isotopic composition), and (ii) in cases with  
542 limited water mixing and a fast flow component to the hydrology, a signal from  $\delta^{18}\text{O}$  of  
543 precipitation or recharge (wetter = more negative) ([Fig. 7](#)). Our evidence base is:

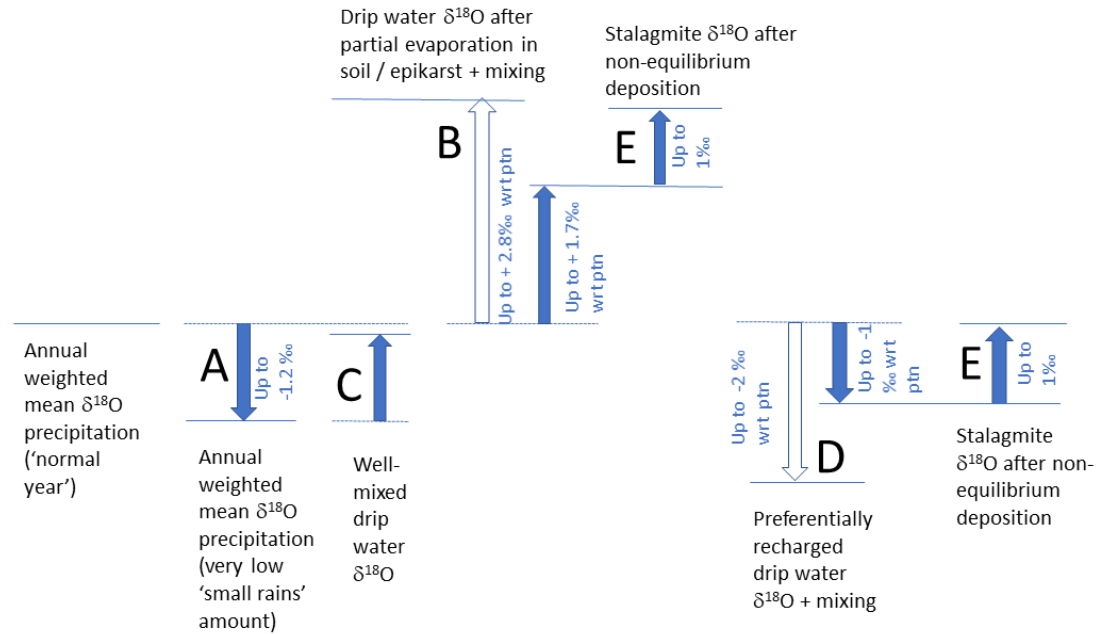
544

545 (1) there is a relatively small seasonal variability in the modern  $\delta^{18}\text{O}$  of precipitation.

546 [Baker et al. \(2010\)](#) presented IAEA data from Addis Ababa which show that the  
547 isotopic composition of precipitation in July and August, the peak of the summer  
548 ('big') rains, has  $\delta^{18}\text{O}$ , which is more negative than April 'small' rains by ~3 ‰. Low  
549 rainfall amounts during the small rains could therefore lead to more negative  
550 recharge water  $\delta^{18}\text{O}$ , but as the small rains typically represent just 25-35 % of total  
551 annual rainfall, with a total range of 15-43 % (data from 20 years of complete data  
552 since 1984, Bedessa meteorological station, see [Fig. 1](#)), any effect is expected to be  
553 less than ~1.2 ‰ in annual weighted mean isotopic composition of precipitation ([Fig.](#)  
554 [7](#), process 'A').

555 (2) Recharge waters will likely mix with water of different ages, depending on the flow  
556 path and the presence and volume of any subsurface karst water stores, such as  
557 solutionally enhanced fractures. Where well-mixed water from a single store is the  
558 source of drip water, and no soil or epikarst evaporation is significant, there will be a  
559 more negative  $\delta^{18}\text{O}$  signal deriving from the precipitation  $\delta^{18}\text{O}$ . Any changes in the

560 annual mean  $\delta^{18}\text{O}$  of precipitation due to changes in the relative proportion of small  
 561 and big rains (see point 1) will be decreased in amplitude due to the mixing of waters  
 562 (Fig. 7, process 'C').



563  
 564 Figure 7. Isotope composition conceptual diagram. The changes in oxygen isotope composition are based on  
 565 observed Addis Ababa IAEA monthly  $\delta^{18}\text{O}$  precipitation (Process A); observed global range of epikarst and soil  
 566 evaporative fractionation (open arrow) and range for P/PET = 0.9 (filled arrow) (Baker et al., 2019) (Process B);  
 567 well-mixed drip water  $\delta^{18}\text{O}$  (Process C); observed global range of recharge bias (open arrow) and range for P/PET  
 568 = 0.9 (filled arrow) (Baker et al., 2019) (Process D); and modelled non-equilibrium fractionation factors (Scholz  
 569 et al., 2011) (Process E).  
 570

571 (3) A single mixed store is a simplification of actual karst hydrology where multiple  
 572 water flow paths are more common (Tooth and Fairchild, 2003; Fairchild et al., 2006;  
 573 Hartman and Baker, 2017), e.g. an additional fracture or by-pass flow which allows a  
 574 fast flow, less mixed flow component (Fig. 7, process 'D'). In these instances, a  
 575 recharge-bias in the  $\delta^{18}\text{O}$  signal may be preserved in the drip water  $\delta^{18}\text{O}$ . In the  
 576 global meta-analysis of global dripwater  $\delta^{18}\text{O}$ , Baker et al. (2019) demonstrated drip  
 577 waters which were up to 2 ‰ more negative than the annual mean of precipitation,  
 578 most commonly observed in regions with very distinct wet seasons in otherwise  
 579 water-limited environments. P/PET in the Mechara region is calculated to be 0.86

580 (FAO New\_LocClim) or 0.88 (Wagari Furi, 2005), and for an equivalent P/PET in  
581 the global analysis of dripwater  $\delta^{18}\text{O}$ , cave drip waters in the Mechara region might  
582 be expected to be up to 1 ‰ more negative than the annual mean of precipitation due  
583 to selective recharge. In-cave fractionation processes could operate in the opposite  
584 direction to this effect (see point 5 below).

585 (4) Precipitation that contributes to the soil water store, and in some cases the shallow  
586 epikarst water, can undergo evaporation, leading to the remaining water  $\delta^{18}\text{O}$   
587 becoming increasingly isotopically positive (Cuthbert et al., 2014). Partially  
588 evaporated water may be subsequently recharged to the cave, having a more positive  
589  $\delta^{18}\text{O}$  than the original precipitation. In a global meta-analysis, Baker et al. (2019)  
590 identified the presence of drip water that was exceptionally up to +2.8 ‰ compared  
591 to weighted mean precipitation  $\delta^{18}\text{O}$ , and for water limited environments with P/PET  
592 similar to the Mechara region, up to + 1.7 ‰. Partially evaporated  $\delta^{18}\text{O}$  has  
593 previously been hypothesised as forming part of the  $\delta^{18}\text{O}$  in an Ethiopian stalagmite  
594 (Baker et al., 2010), where forward modelling for the modern growth phase of the  
595 Bero-1 stalagmite identified a positive isotope offset of 2.0 to 2.5 ‰, attributed to  
596 evaporative fractionation processes between rainfall and the stalagmite (Fig. 7,  
597 process 'B'). However, the effect of possible changes in the relative proportion of  
598 small and big rains on the partial evaporation of soil or epikarst waters is unclear. For  
599 example, if the small rains led to the recharge of more partially evaporated water than  
600 the big rains, due to relative low rainfall amounts in the former, then relatively dry  
601 small rain seasons could lead to more negative drip water  $\delta^{18}\text{O}$ .

602 (5) All stalagmites analysed in Ethiopia to date, demonstrate conclusive evidence of non-  
603 equilibrium deposition. In Ach-3, there is strong correlation between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$   
604 along growth laminae and over time, with  $\delta^{13}\text{C}/\delta^{18}\text{O}$  gradients  $< 3$ . Bero-1 and Ach-1

605 also had  $\delta^{13}\text{C}/\delta^{18}\text{O}$  gradients  $<3$ . The similar range in  $\delta^{13}\text{C}/\delta^{18}\text{O}$  gradients of the three  
606 stalagmites to laboratory experiments (Wiedner et al., 2008) and the meta-analysis  
607 and field observations of Mickler et al. (2006), combined with the strong correlations  
608 between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  for each stalagmite, and similar and coherent multidecadal  
609 spectral frequencies between  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ , suggests a dominant in-cave control.  
610 One such mechanism is a change in drip rate which controls disequilibrium isotope  
611 fractionation during the progressive degassing of  $\text{CO}_2$  from the water film during  
612 stalagmite formation (Fig. 7, Process 'E'). All three stalagmites have similar  
613 amplitude in multidecadal signal (up to  $\sim 1$  ‰). The iSOLUTION model of oxygen  
614 and carbon isotope composition of stalagmite calcite (Scholz et al., 2009; Deininger  
615 and Scholz, 2019) models disequilibrium isotope fractionation processes, and  
616 produces this magnitude of oxygen isotope fractionation for high  $p\text{CO}_2$  drip waters  
617 and relatively slow drip rates. Kinetic isotope fractionation due to rapid degassing  
618 from high  $p\text{CO}_2$  drip waters could also lead to this magnitude of isotope fractionation  
619 for faster drip rates (Mickler et al., 2006, Wiedner et al., 2008) and would be  
620 considered likely given the fast growth rates of Ethiopian stalagmites.

621

622 We provide multiple lines of evidence that the multidecadal variability in stalagmite  $\delta^{18}\text{O}$  in  
623 Ethiopian stalagmites is likely due to the karst hydrological processes on water mixing, with  
624 preferential recharge and isotope fractionation processes operating with opposite signs in  
625  $\delta^{18}\text{O}$  from the same climate forcing as visualised in Figure 7. We hypothesise that inter-  
626 annual variability in the relative amounts of small and big rains may have some influence on  
627 the mean annual precipitation  $\delta^{18}\text{O}$ . However, cave drip water  $\delta^{18}\text{O}$  is further altered by the  
628 effects of variable karst hydrology and water isotope fractionation processes, leading to a  
629 cave drip water isotopic signature that represents a combination of precipitation  $\delta^{18}\text{O}$ ,

630 preferential recharge  $\delta^{18}\text{O}$  and / or a  $\delta^{18}\text{O}$  that represents the extent of partial evaporation of  
631 the water. In years of decreased recharge, this in turn potentially leads to decreases in drip  
632 rate to the stalagmites, leading to the potential of increased isotope fractionation due to  
633 disequilibrium deposition. Decreases in drip rate do not necessarily have a linear relationship  
634 with surface hydroclimate forcing, due to the non-linear nature of karst hydrology and mixing  
635 of waters in karst stores and fractures. Our observation of multidecadal spectral frequency in  
636  $\delta^{18}\text{O}$  is therefore likely to be due to individual extremes of dry years, which determine the  
637 volume of recharge to these karst stores, and in turn the drip rate from the store, including  
638 both the mean annual drip rate and /or the duration of dripping in one year. Superimposed on  
639 this signal could also be evaporative fractionation of water in the soil, shallow vadose zone or  
640 in the cave, and / or a preferential recharge signal, both of which operate with the opposite  
641 sign. With drier conditions, in-cave isotope fractionation and evaporative fractionation effects  
642 operate with the same sign, increasing drip water  $\delta^{18}\text{O}$  due to increased evaporation at the  
643 same time as disequilibrium deposition increased with lower drip rates. However, for some  
644 samples with a fast-flow or bypass-flow component, preferential recharge could be  
645 significant in controlling drip water  $\delta^{18}\text{O}$ , and this signal could dominate over fractionation  
646 processes and generate a multi-decadal signal with the opposite sign. Superimposed on all  
647 flow types is the possibility of kinetic isotope fractionation due to high drip water  $p\text{CO}_2$ ,  
648 which is likely given the very fast growth rates of Ethiopian stalagmites.

649         The modern East African monsoon is connected to the larger Indo-Pacific-Asian  
650 monsoon, as the annual formation of the South Asian monsoon generates trade winds in the  
651 eastern Indian Ocean and eastern Africa, and associated moisture and rainfall in Ethiopia  
652 ([Vizy and Cook, 2003](#); [Funk et al. 2016](#)). These moisture patterns are further modulated by  
653 the strength of the Indo-Pacific warm pool heating, through multi-decadal climate phenomena  
654 including ENSO, IOD and the Pacific Decadal Oscillation (PDO). Hence, it is useful to



655 compare the multidecadal variability in  $\delta^{18}\text{O}$  in Early to mid-Holocene Ethiopian stalagmites  
656 to records of similar temporal resolution and growth periods in the broader Indo-Pacific-  
657 Asian monsoon region. For example, high-resolution north Chinese stalagmite  $\delta^{18}\text{O}$  records  
658 over the last 2000 years (Zhang et al., 2019) suggest a strong Indian Ocean influence on  
659 stalagmite multidecadal  $\delta^{18}\text{O}$  variability, with a more distal moisture source leading to more  
660 negative  $\delta^{18}\text{O}$  precipitation during La Niña events.

661 We identified three suitable high-resolution stalagmite records for comparison (Table  
662 3). In Southern Oman, the Qunf Cave Q5 record of Fleitmann et al (2003) deposited over the  
663 period 8.0 to 2.7 ka had spectral peaks at 10.9 and 10.2 years, noting the sampling resolution  
664 for  $\delta^{18}\text{O}$  was 2-15 yr. In Northern Oman, Neff et al (2001) sampled at 1.4 yr resolution from  
665 around 8.4 to 7.9 ka and identified multidecadal peaks in  $\delta^{18}\text{O}$  in their untuned record at 3.3,  
666 4.9, 7.3, 35 and 78 years. In southern inland China, Dykoski et al (2005) report an annual  
667 resolution record from 8250 – 8110 years BP with peaks at 2.3 – 2.4, 4.8, 5.1, 6.4, 6.6 and 44  
668 years. Disregarding spectral peaks that are close to the sampling resolution of each of these  
669 records, a comparison between Ethiopian, Oman and central China stalagmite  $\delta^{18}\text{O}$  in the  
670 Early Holocene suggests that there are different multi-decadal  $\delta^{18}\text{O}$  signals in the Early  
671 Holocene in Ethiopia, Oman and China. Additional annual or near-annual resolution  $\delta^{18}\text{O}$   
672 records would be of benefit to ascertain if this is due to regional climate differences in the  
673 multidecadal forcing of  $\delta^{18}\text{O}$  within the monsoon region, regional differences in the strength  
674 of climate influence on precipitation  $\delta^{18}\text{O}$ , and/or the relative importance of karst  
675 hydrological variability.

676 Furthermore, the precipitation  $\delta^{18}\text{O}$  record from the only long-term monitoring station  
677 at Addis Ababa shows that there is little seasonal variability in  $\delta^{18}\text{O}$  (e.g., Baker et al., 2010).  
678 A recent study by Bedaso et al. (2020) on  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  of precipitation samples collected at  
679 daily, weekly and monthly intervals in different parts of Ethiopia representing local climate

680 Table 3. Comparison of the  $\delta^{18}\text{O}$  record of Ach-3 with Holocene speleothems from Oman and China.

Speleothem	Location	Periodicity (yrs)*	Growth period	Reference
Qunf Cave, stalagmite Q5, 0.7 mm sampling resolution = 2 - 15 yr resolution	Southern Oman	<i>~220, ~140, ~107, 10.9, 10.2</i> (90% red noise)	8.0 – 2.7ka	Fleitmann et al. (2003)
Hoti Cave, stalagmite H5, high resolution section: 0.4 mm = 1.4 yrs	Northern Oman	78, 35, 7.3, 4.9, 3.3 (90% red noise)	~7.9 – 8.4ka	Neff et al. (2001)
Dongge Cave, high resolution section: ~1 yr /sample	Southern inland China	44, 6.6, 6.4, 5.1, 4.8, 2.4-2.3	8110 – 8250 yrs BP	Dykoski et al. (2005)

681 \*Italics indicates close to sampling resolution; significance level shown where reported.

682

683 regimes confirmed the weak correlation between rainfall amount and  $\delta^{18}\text{O}$  values of

684 precipitation. The same study further indicated the absence of discernible source region

685 variability among the different stations. The mean moisture back-trajectory paths show the

686 Mechara caves on the Southeastern Ethiopian highlands receive most of their moisture from

687 the southwestern and northern Indian Ocean, on southerly and easterly wind trajectories,

688 respectively. Hence, the different multi-decadal variabilities in the Ethiopian stalagmites

689 cannot be explained by moisture source variability alone.

690

#### 691 4.3. The role of tectonics

692

693 The MER and the adjacent highlands have been tectonically and seismically active since the

694 Mid-Miocene (Gouin, 1979; Ayele and Kulhánek, 2000; Corti, 2009; Ayele, 2017). The rift-

695 related extensional tectonics led to the initiation and further development of the Achere-

696 Aynage cave system where conduits developed following NE-SW oriented fractures, while

697 the zone of conduit development was restricted by the mudstone/marl horizons (Asrat et al.,  
698 2008). The aquifer architecture and hydrological flow regimes above the caves are therefore a  
699 strong reflection of this tectonic-lithological interaction, which has been changing through  
700 time, even within the time frame of a single speleothem growth. Active tectonics in many  
701 cases is responsible for developing and continuously modifying the fracture systems which  
702 usually refocused groundwater flow paths along newly formed or reactivated fractures and  
703 conduits, in many cases leading to the cessation of growth of speleothems, manifested in  
704 growth hiatuses (Asrat, 2012).

705 In addition to frequent growth hiatuses, the control of seismicity on speleothem  
706 growth in the Mechara caves is manifested in: (i) frequent anomalous laminae such as those  
707 containing higher proportion of impurities corresponding to known earthquake events; (ii)  
708 frequent deviations from vertical growth axis which can be attributed to shifting of the drip  
709 location; and (iii) upward thinning stalagmites which show in many cases abrupt change from  
710 regular ‘candlestick’ shape implying a regular flow regime to upward thinning shape which  
711 could indicate the release of water from a karst store after a seismic event (Asrat, 2012).

712 Though seismicity data for the region are scarcely available for the period prior to the  
713 20<sup>th</sup> century (Gouin, 1979), several studies in the MER show that tectonic activities  
714 particularly intensified during the late Quaternary towards the margins of the central MER  
715 (Abebe et al., 2007; Agostini et al., 2011). A study of the Plio – Pleistocene and Holocene  
716 fluvio-lacustrine sequence in the central MER further indicated that late Quaternary syn-  
717 depositional tectonic activities were frequent in the region (Le Turdu et al., 1999). The  
718 Mechara area is located on the plateau adjoining the eastern margin of the MER, and have  
719 been seismically active. For instance, Ayele and Kulhánek (2000) have relocated the  
720 epicenter of the August 25, 1906 earthquake sequence (maximum magnitude of 6.5) on the  
721 eastern shoulder of the MER, just 50km south of Mechara (Asrat, 2012).

722 A recent probabilistic seismic hazard analysis (PSHA) for Ethiopia and the Horn of  
723 Africa region has shown that not only the rift but also the adjoining rift margins (including  
724 the Mechara area) lie within a significantly important seismic hazard zone (Ayele, 2017). The  
725 analysis considered the whole range of earthquake magnitudes recorded in the region since  
726 1900, and calculated the Peak Ground Acceleration (PGA) corresponding to 10 % and 2 %  
727 probability of exceedance in 50 years, corresponding to return periods of 475 and 2475 years,  
728 respectively, with a spatial resolution of 0.5° x 0.5° grids. The results show PGAs ranging  
729 from 0.0 – 0.18 g and 0.0 – 0.35 g for the 475 and 2475 return periods, respectively,  
730 characterizing various parts of the Ethiopian rift and the highlands. The Mechara area lies  
731 within the zone of about 0.06 g for the 10 % and 0.13 g for 2 % probabilities, respectively.  
732 Such horizontal ground accelerations could significantly affect the aquifer architecture above  
733 the caves leading to the modification of groundwater flow paths and drip locations. The  
734 relatively short return periods of significant earthquake magnitudes closely match with the  
735 short growth phases of the Holocene stalagmites (Ach-1, Ach-3, Bero-1), and this could  
736 explain the ubiquity of discontinuous stalagmite deposition rarely lasting more than 1,000  
737 years. Furthermore, a global synthesis of annually laminated stalagmites indicated that  
738 Ethiopian stalagmites are unique in their relatively short growth phases (with median growth  
739 duration of 172 years) compared to the median growth duration of 447 years globally (Baker  
740 et al., 2021). This feature of Ethiopian stalagmites can be attributed to the location of the  
741 Mechara caves in close proximity to an active seismic zone of the MER (see Fig. 1).

742

## 743 **5. Conclusions**

744

745 We use trace element, growth rate,  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  of Early Holocene stalagmite Ach-3 to  
746 understand the processes occurring during its deposition. The trace element composition

747 identifies an initial growth period with a flush of soil-derived material, and a final growth  
748 period where there is a decoupling from the soil zone, indicative of drying conditions. We  
749 observe a multidecadal  $\delta^{18}\text{O}$  variability in the Early Holocene Ach-3 and other two Middle  
750 and Late Holocene Ethiopian stalagmites of amplitude  $\sim 1\%$ . Covariation of  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$   
751 demonstrates that all three stalagmites are dominated by isotope fractionation, likely due to  
752 both kinetic and disequilibrium effects during the progressive degassing of  $\text{CO}_2$  from drip  
753 waters with a high  $p\text{CO}_2$  during stalagmite formation. The amplitude of multidecadal  
754 variability in  $\delta^{18}\text{O}$  is similar to that modelled due to changes in drip rate. Rapid growth rates,  
755 fast drip rates, and isotope fractionation effects are likely the primary controls on the isotope  
756 geochemistry while active tectonics has played an important role in determining the growth  
757 duration of the three Ethiopian stalagmites, with additional influences possible from  
758 evaporative fractionation, and for samples with very short water residence time, a small  
759 primary precipitation seasonality signal. Despite the extent of non-equilibrium deposition,  
760 differences in mean stalagmite  $\delta^{18}\text{O}$  through the Holocene are larger in magnitude than the  
761 multi-decadal variability. Thus long-term (centennial and longer) trends in stalagmite  $\delta^{18}\text{O}$   
762 are likely to be good proxies for climate as they record long-term climatic forcing on  
763 precipitation  $\text{d}^{18}\text{O}$  and drip water  $\delta^{18}\text{O}$ .

764

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766

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775

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939 **List of Supplemental Materials**

940

941 Supplemental Table 1. Trace element analysis.

942 Supplemental Table 2. Full spectral analysis results on both stable isotopes and growth rate  
943 time series of Ach-3 stalagmite. The dominant spectral for the respective proxy is  
944 marked in Bold.

945

946 Supplemental Figure 1. Principle Component Analysis (PCA) of trace element data.

947 Supplemental Figure 2. Time series of the three Principle Components (PC1 to PC3).

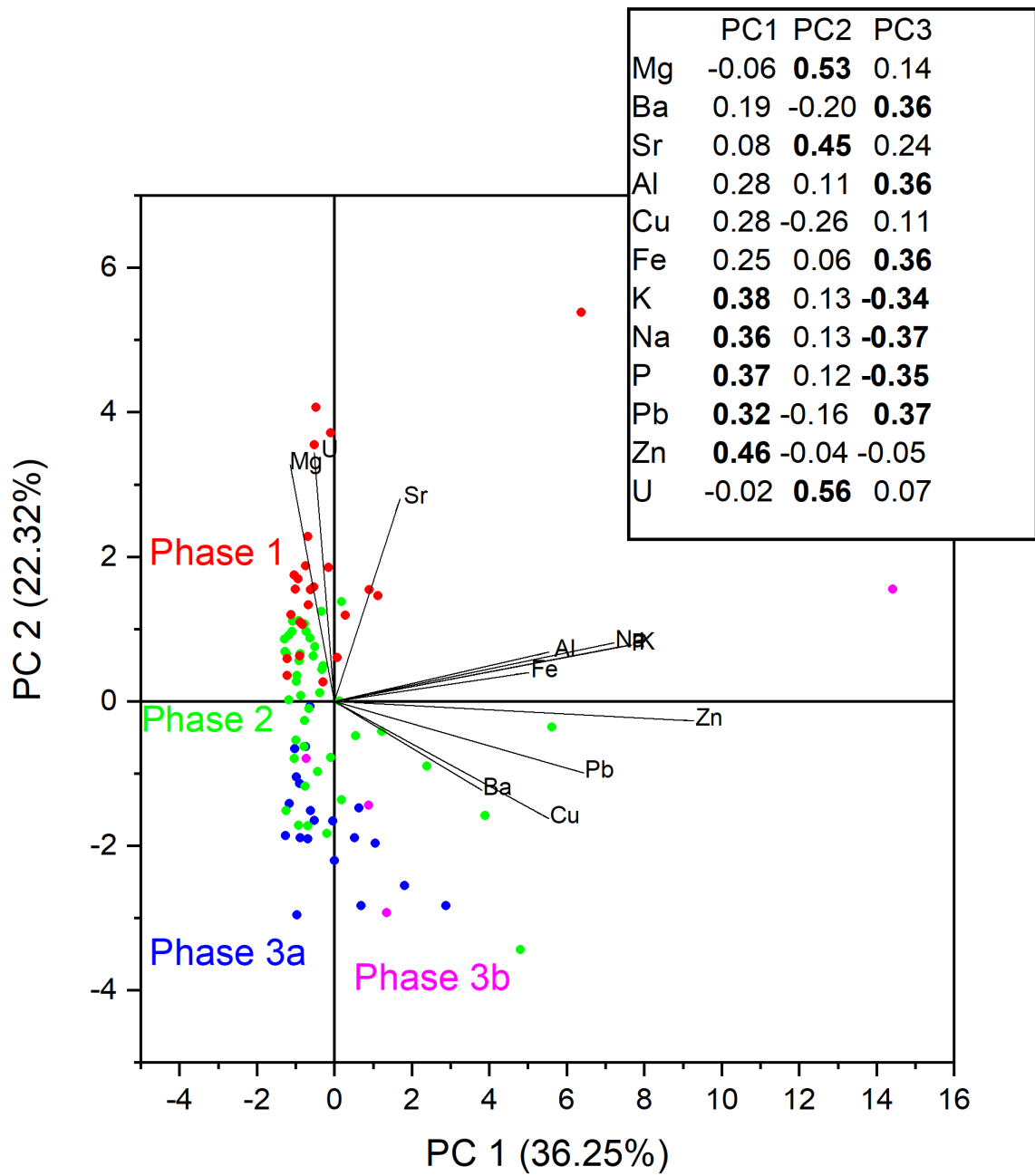
948 Supplemental Figure 3. Spectral analysis results on both stable isotopes and growth rate time  
949 series of Ach-3 stalagmite. Four spectral windows were applied (Rectangular, Welsh,  
950 Hanning and Blackman-Harris) using the SPECTRUM software ([Schulz and Stattegger,  
951 1997](#)). The horizontal line indicates the lower bound for statistically significant power  
952 e.g. distinguished from white noise.

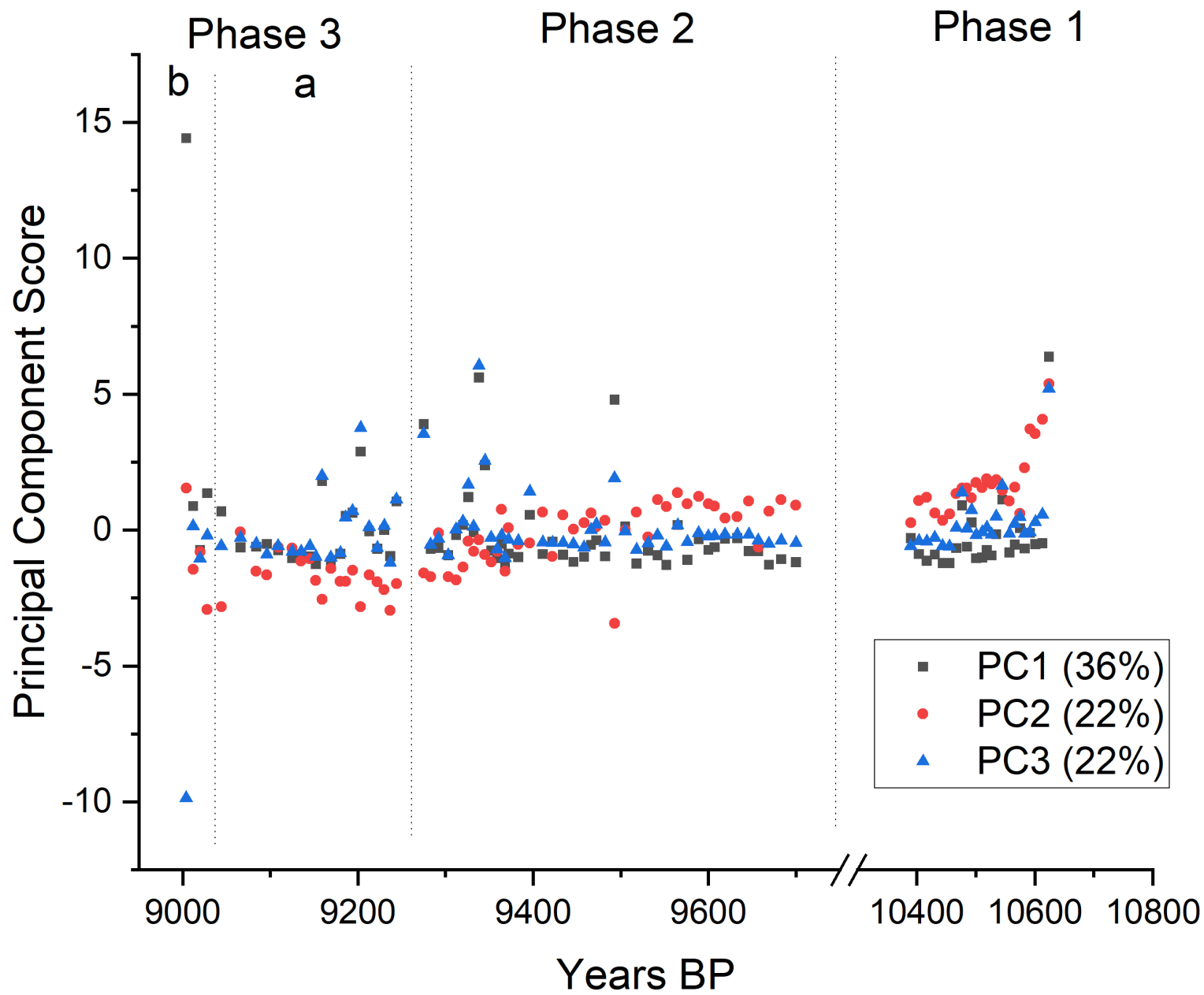
953 Supplemental Figure 4. Autocorrelation functions for  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ .

954 Supplemental Figure 5. Spectral analysis results on oxygen isotopes for stalagmites Ach-3,  
955 Ach-1 and Bero-1. Four spectral windows were applied (Rectangular, Welsh, Hanning  
956 and Blackman-Harris) using the SPECTRUM software ([Schulz and Stattegger, 1997](#)).  
957 The horizontal line indicates the lower bound for statistically significant power e.g.  
958 distinguished from white noise. Results are tabulated in [Table 2](#).

959

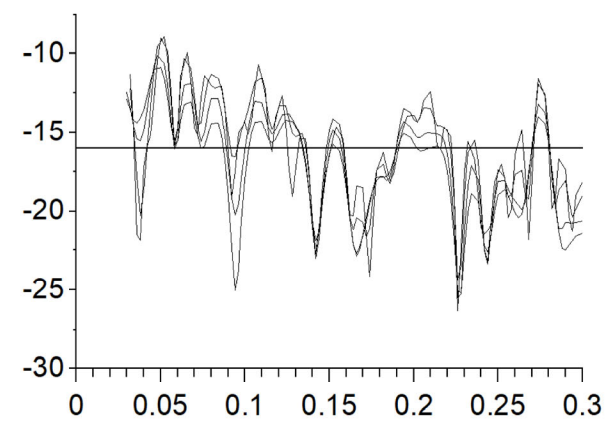
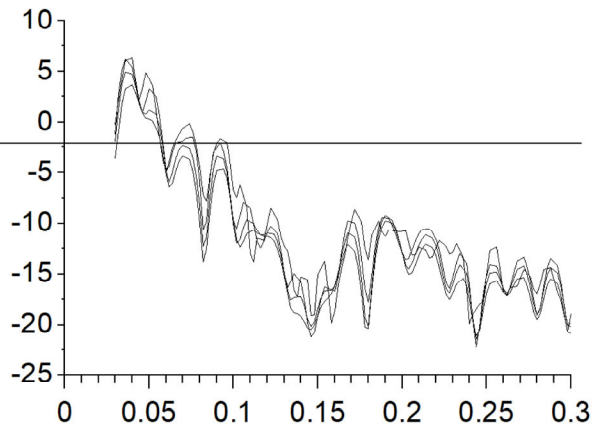
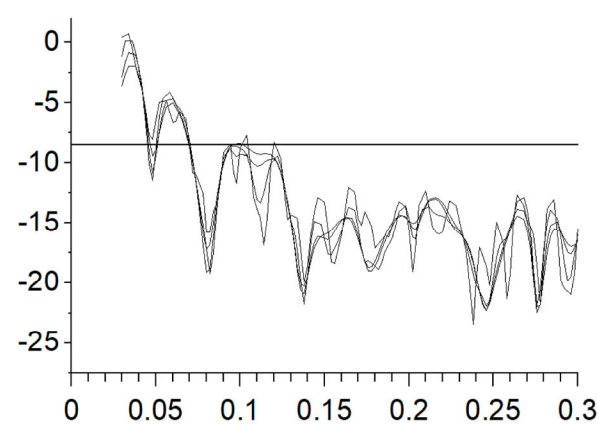
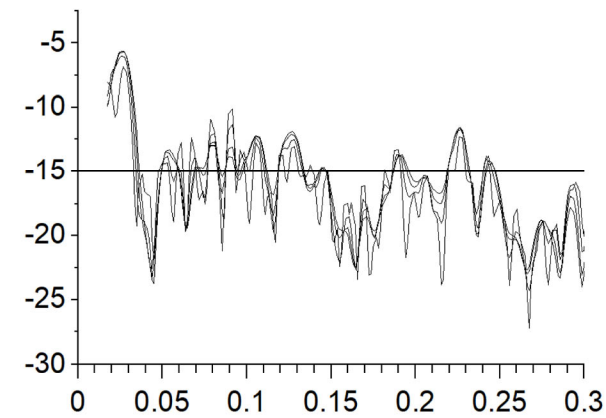
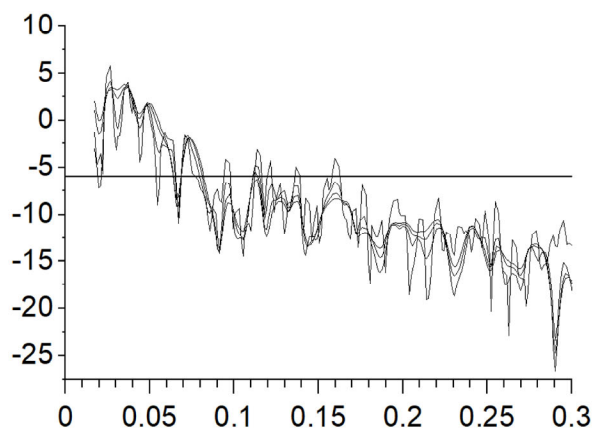
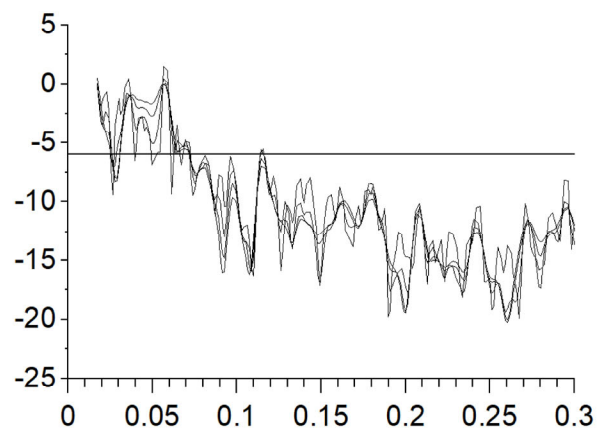
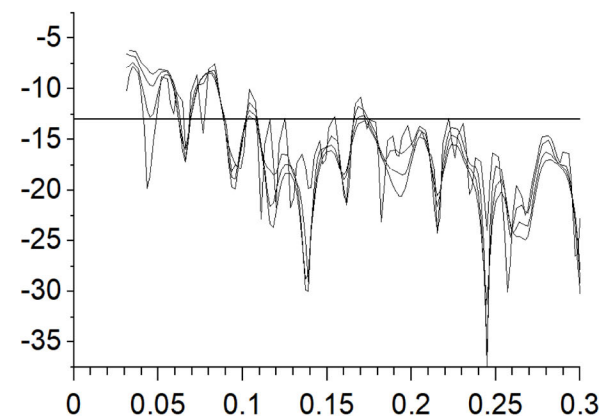
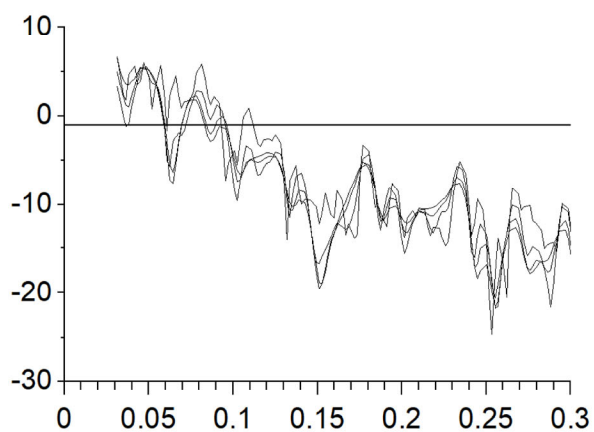
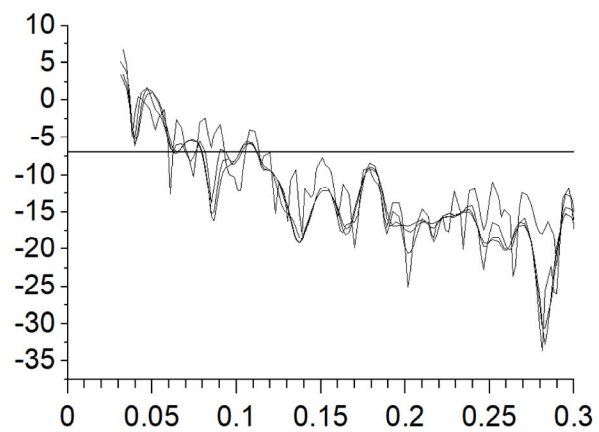
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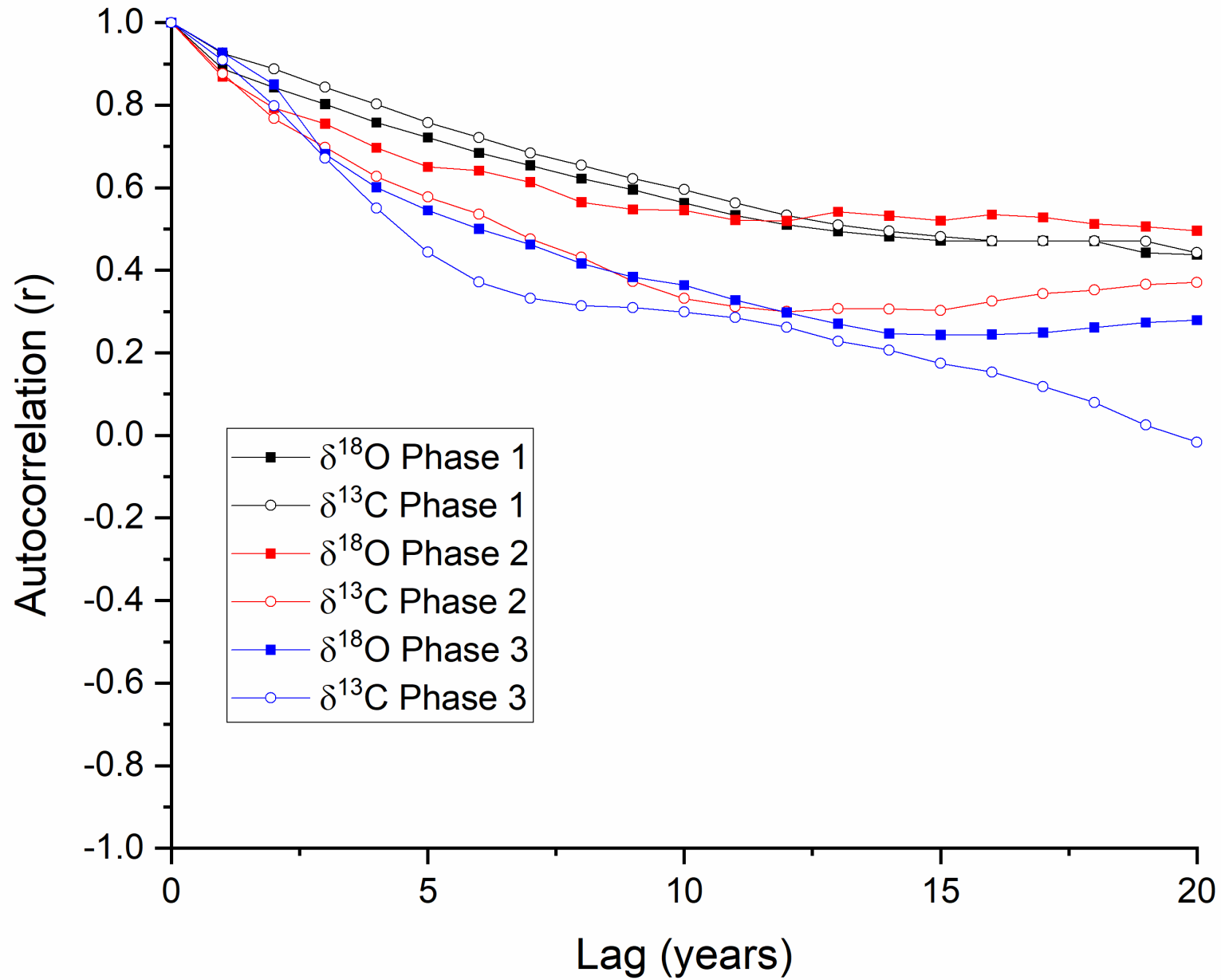
$\delta^{18}\text{O}$  $\delta^{13}\text{C}$ 

growth rate

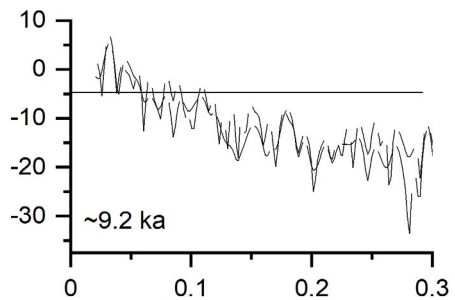


Frequency (1/yr)

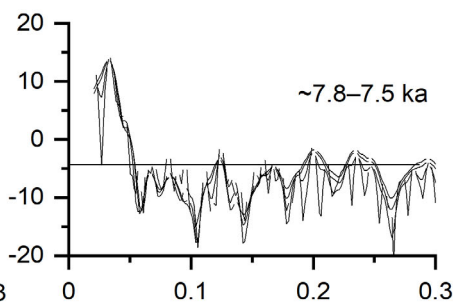
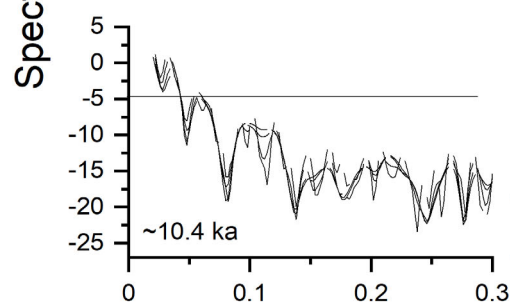
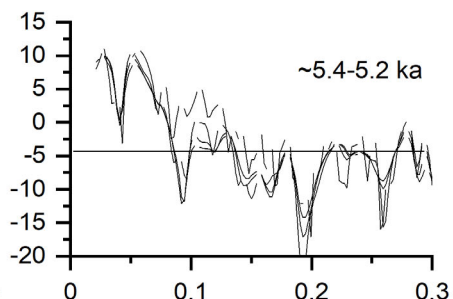
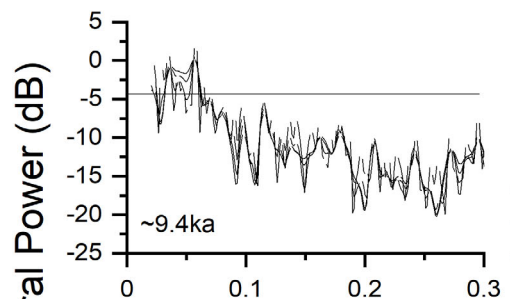
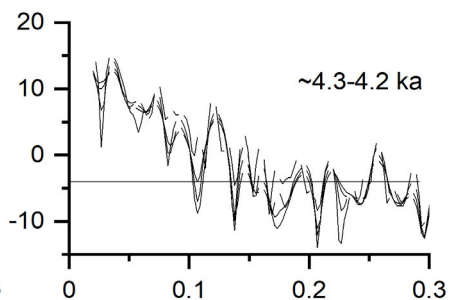




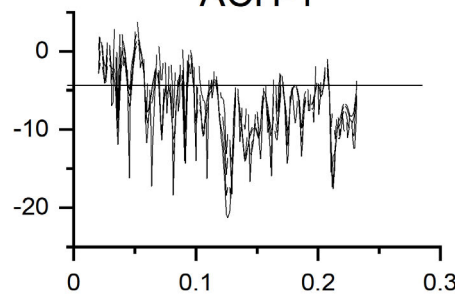
ACH-3



BERO-1



ACH-1



Frequency (1/yr)

Supplemental Table 1. Trace element analysis.

Sample Id	Ca	Mg	Ba	Sr	Al	Cu	Fe	K	Na	P	Pb	S	Zn	U
	wt %	wt %	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
ACH3-01	37.3	0.455	10.70	385	47.7	8.9	50.9	1239	759	13397	0.35	3869	23.3	1.6
ACH3-02	38.3	0.451	6.95	355	132.0	4.2	13.9	112	81	251.9	0.18	3014	3.27	1.66
ACH3-03	37.9	0.474	7.10	362	11.7	0.6	15.9	70.5	108	148.1	0.09	2966	0.88	1.79
ACH3-04	38.1	0.452	6.71	312	19.0	16.9	148.6	94	68	84.9	0.14	2526	5.42	1.67
ACH3-05	38.7	0.382	8.48	423	12.0	13.1	13.2	118	52	97	0.09	2866	4.29	1.41
ACH3-06	38.0	0.520	9.45	439	5.6	3.9	18.5	56.3	72	32.6	0.05	3305	1.40	1.75
ACH3-07	38.0	0.452	10.39	383	16.8	3.7	15.3	54.2	58.8	66.8	0.11	2964	1.21	1.61
ACH3-08	38.7	0.422	8.44	379	10.1	3.8	16.1	95	62.2	64.8	0.08	2759	1.76	1.73
ACH3-09	38.0	0.480	13.7	405	5.9	1.8	12.2	95	61.7	68.3	0.07	3200	1.03	1.75
ACH3-10	37.9	0.474	8.28	408	2.8	0.8	9.6	99	59.7	42.2	0.04	3019	0.35	1.66
ACH3-11	37.3	0.483	8.18	374	3.6	1.7	8.6	71	64.7	46.4	0.09	2723	0.91	1.56
ACH3-12	37.9	0.493	14.0	386	8.9	0.8	3.9	54.3	65.2	27.3	0.10	2567	0.61	1.58
ACH3-13	37.9	0.465	7.19	364	2.5	0.6	4.2	56.3	50.3	35.8	0.07	2437	0.28	1.40
ACH3-14	38.1	0.426	63.1	406	65.1	2.5	39.3	65.0	56.3	44.6	0.82	2436	4.67	1.48
ACH3-15	39.0	0.489	8.19	367	2.4	0.8	4.3	55.1	61.3	41.7	0.05	2774	1.08	1.64
ACH3-16	39.4	0.468	10.52	349	5.2	1.9	5.4	46.5	63.5	79.3	0.19	2703	1.18	1.71
ACH3-17	39.0	0.508	24.5	384	12.8	5.9	51.4	55.5	69	40.7	0.51	2802	3.07	1.64
ACH3-18	39.3	0.523	43.0	400	38.0	3.4	50.4	77	81	113.2	0.40	2859	2.85	1.63
ACH3-19	39.2	0.476	80.0	386	52.5	3.5	458.0	47.1	62	124.8	0.89	2429	3.85	1.42
ACH3-20	38.7	0.523	35.2	370	25.0	1.4	37.0	65.8	75	168.9	0.30	2585	1.81	1.52
ACH3-21	39.0	0.508	19.9	351	8.3	2.3	10.6	50.1	67	111.3	0.16	2521	1.51	1.56

ACH3-22	38.9	0.507	36.6	363	24.4	3.5	16.6	50.0	63	71.6	0.32	2531	2.16	1.49
ACH3-23	37.8	0.456	11.5	289	6.4	2.8	8.6	44.4	55.3	71.3	0.11	2286	1.30	1.40
ACH3-24	38.6	0.524	41.2	357	37.2	4.2	205.4	81	73	131.4	0.39	2541	2.86	1.51
ACH3-25	37.8	0.527	76.7	437	136.6	9.8	164.6	124	79	103.6	0.70	2449	7.52	1.47
ACH3-26	38.8	0.497	20.0	362	22.6	2.2	11.8	60	61	46.6	0.10	2537	1.33	1.56
ACH3-27	38.0	0.575	21.4	405	8.1	2.3	7.6	84	77	89.9	0.08	2883	1.03	1.70
ACH3-28	39.5	0.492	11.16	360	4.4	2.3	12.7	72	59.5	67.5	0.09	2556	1.11	1.65
ACH3-29	41.0	0.525	49.6	402	18.5	4.1	7.9	63	69	71.6	0.15	2663	2.00	1.69
ACH3-30	39.1	0.513	48.6	400	25.0	3.0	32.7	85	73	163.5	0.22	2744	2.31	1.67
ACH3-31	38.1	0.583	70.5	433	49.3	3.8	76.8	106	86	105.8	0.36	3070	2.90	1.66
ACH3-32	40.4	0.589	41.3	411	23.8	1.5	27.9	102	83	109.2	0.21	3144	1.88	1.72
ACH3-33	37.7	0.577	153.9	551	142.5	9.3	200.9	150	102	211.5	0.99	3045	8.24	1.62
ACH3-34	38.2	0.567	64.2	423	123.8	6.4	76.5	80	78	157.5	0.53	3070	5.44	1.61
ACH3-35	40.0	0.566	25.2	372	15.9	0.9	30.8	56.5	68	77.0	0.17	2739	0.81	1.68
ACH3-36	38.3	0.566	11.10	354	8.3	0.9	7.8	54.9	70	71.6	0.11	2813	0.94	1.67
ACH3-37	38.3	0.600	16.4	416	17.3	1.0	13.9	104	87	124.4	0.13	3457	1.31	1.92
ACH3-38	38.8	0.509	8.43	341	7.5	0.5	9.1	52.2	60	64.1	0.05	2825	0.68	1.62
ACH3-39	37.1	0.579	11.59	407	7.6	0.4	8.8	85	64	73.5	0.10	2600	0.97	1.56
ACH3-40	38.2	0.556	14.4	410	8.6	0.7	7.9	64.8	62	38.7	0.08	2646	0.59	1.53
ACH3-41	38.4	0.566	67.4	454	60.3	1.9	17.0	89	68	51.9	0.25	2671	1.95	1.55
ACH3-42	37.9	0.574	7.70	431	10.6	0.3	16.0	100	77	51.3	0.04	3128	0.70	1.76
ACH3-43	39.2	0.527	17.2	386	32.8	1.6	25.6	113	66	92.0	0.08	2818	1.64	1.65
ACH3-44	39.0	0.603	9.83	425	4.0	1.1	4.3	88	84	43.2	0.09	3319	0.71	1.89
ACH3-45	39.2	0.595	13.62	417	5.1	0.3	5.3	68	74	42.9	0.04	3200	0.08	1.71

ACH3-46	38.1	0.590	7.49	397	7.4	0.6	11.4	82.7	83	51.2	0.05	3196	0.80	1.76
ACH3-47	39.7	0.644	7.97	421	29.6	0.5	33.9	72	92	64.5	0.21	3537	1.13	1.88
ACH3-48	37.2	0.565	6.95	377	22.2	0.8	152.4	80	71	65.4	0.13	3024	0.79	1.72
ACH3-49	38.5	0.611	6.65	398	22.2	0.6	12.7	74	79	55.8	0.05	3170	0.68	1.77
ACH3-50	39.1	0.588	6.74	361	12.8	29.6	77.5	71	75	49.9	1.23	3104	15.27	1.71
ACH3-51	38.5	0.570	7.38	377	67.9	2.0	47.1	127	80	124.6	0.15	3116	2.39	1.82
ACH3-52	37.8	0.572	7.48	393	3.7	0.2	4.2	82	78	49.8	0.01	3563	0.30	1.95
ACH3-53	38.3	0.599	6.81	398	14.2	0.8	21.0	82	75	46.5	0.11	2630	1.34	1.54
ACH3-54	36.9	0.600	7.17	405	21.0	0.8	19.1	72	78	38.5	0.05	3374	0.46	1.87
ACH3-55	36.9	0.566	6.89	390	4.5	0.1	6.5	69	73	34.1	0.01	3253	0.18	1.93
ACH3-56	35.8	0.565	7.35	393	54.7	1.0	84	151	74	156.6	0.05	3367	2.46	1.88
ACH3-57	37.8	0.587	7.05	417	13.3	0.3	4.8	64.8	83	31.9	0.02	3416	0.49	1.90
ACH3-58	37.7	0.595	7.50	416	36.4	0.6	43.8	114	80	145.3	0.05	3607	2.22	1.95
ACH3-59	34.1	0.528	6.70	374	23.6	0.4	23.5	81	67	70.1	0.04	3162	1.15	1.72
ACH3-60	38.2	0.606	7.40	411	31.3	0.6	36.8	91	82	89.1	0.04	3575	1.41	1.87
ACH3-61	38.9	0.584	7.45	397	47.1	0.8	58.1	93	78	140.1	0.06	3553	2.36	1.91
ACH3-62	37.2	0.557	7.23	391	41.7	0.7	48.4	98	73	120.2	0.07	3375	2.44	1.82
ACH3-63	36.4	0.574	7.23	417	19.2	0.6	21.4	77	72	86.7	0.05	3290	1.08	1.80
ACH3-64	38.0	0.545	6.85	373	38.6	0.8	23.2	67	59	58.6	0.07	2967	1.17	1.60
ACH3-65	36.2	0.579	6.51	383	6.7	0.2	5.8	50.4	70	35.3	0.02	3168	0.46	1.75
ACH3-66	36.0	0.581	6.75	384	12.6	0.2	15.4	65.4	73	49.0	0.03	3216	0.77	1.87
ACH3-67	37.9	0.644	6.76	420	5.7	0.2	5.9	83	71	104.6	0.02	2941	0.39	1.72
ACH3-68	39.5	0.551	7.61	434	22.5	0.8	30.1	147	75	257.1	0.05	3549	2.63	1.88
ACH3-69	36.8	0.587	6.59	408	12.3	0.4	11.4	79	83	121.8	0.05	3223	0.98	1.85

ACH3-70	37.6	0.609	7.23	413	5.0	0.2	4.7	64.8	83	31.6	0.05	3648	0.55	1.94
ACH3-71	37.0	0.575	6.83	380	27.4	0.4	26.4	67.6	72	86.4	0.06	3238	0.64	1.85
ACH3-72	37.6	0.578	6.54	381	12.8	0.2	9.6	58.1	74	33.6	0.03	3325	0.36	1.82
ACH3-73	38.8	0.593	6.88	404	9.0	0.2	9.0	66	80	44.3	0.03	3478	0.30	1.94
ACH3-74	37.4	0.615	7.52	410	28.7	0.9	54.4	77	82	60.0	0.08	3587	0.61	1.95
ACH3-75	36.2	0.602	8.95	403	58.0	1.8	203.1	119	87	78.3	0.32	3586	2.18	1.92
ACH3-76	36.5	0.614	7.15	412	19.1	0.6	29.5	85	82	91.9	0.15	3647	0.88	1.92
ACH3-77	36.1	0.625	7.44	396	71.4	1.8	30.8	92	77	79.8	0.26	3322	2.49	1.77
ACH3-78	36.2	0.618	7.17	415	6.9	0.3	6.4	71	82	51.7	0.08	3584	0.33	1.93
ACH3-79	37.1	0.648	7.15	417	11.4	0.3	14.0	70	78	32.6	0.10	3537	0.45	1.88
ACH3-80	36.7	0.629	7.80	426	25.0	0.7	19.4	77	81	67.2	0.07	3699	0.84	1.96
ACH3-81	36.3	0.622	7.18	417	7.0	1.1	8.5	71	78	76.3	0.06	3564	0.82	1.92
ACH3-82	35.8	0.610	8.90	408	54.0	1.2	23.7	87	81	128.1	0.16	3735	1.47	1.95
ACH3-83	36.2	0.614	8.25	402	80.7	3.0	181.6	108	83	90.9	0.34	3655	2.61	1.89
ACH3-84	36.4	0.580	7.00	395	18.4	0.7	31.2	68	74	65.6	0.09	3563	0.76	1.89
ACH3-85	36.5	0.614	7.80	413	38.8	1.1	19.1	76	79	66.7	0.12	3763	0.96	1.92
ACH3-86	38.4	0.620	8.87	402	59.2	1.7	68.5	91	79	88.4	0.23	3649	2.09	1.85
ACH3-87	36.1	0.607	7.77	430	10.6	0.7	9.6	91	81	411.8	0.07	3882	1.00	2.08
ACH3-88	37.0	0.651	8.34	468	8.8	1.1	5.5	123	87	1525.4	0.15	3973	1.51	2.50
ACH3-89	36.6	0.695	8.00	476	9.6	0.6	8.7	91	87	596.9	0.12	3971	1.20	2.20
ACH3-90	35.7	0.698	8.00	489	6.5	0.6	7.6	88	96	388.7	0.17	4185	0.92	2.20
ACH3-91	36.3	0.674	9.40	493	240.0	3.7	577.6	241	108	2357.1	0.64	3879	9.8	2.6
<b>MDL</b>	<b>0.50</b>	<b>0.02</b>	<b>0.5</b>	<b>1.0</b>	<b>0.5</b>	<b>0.2</b>	<b>0.5</b>	<b>0.5</b>	<b>0.5</b>	<b>0.5</b>	<b>0.2</b>	<b>100.0</b>	<b>0.2</b>	<b>0.2</b>

Supplemental Table 2. Full spectral analysis results on both stable isotopes and growth rate time series of Ach-3 stalagmite. The dominant spectral for the respective proxy is marked in Bold.

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Phase 3a (laminae 28-243)

$\delta^{18}\text{O}$

Rectangular	34-29, <b>25-21, 18</b>
Welsh I	34-29, <b>23-20</b>
Hanning	36-29, <b>23-19</b>
Triangular	36-29, <b>22-18</b>
Blackman Harris	39-29, <b>22-18</b>

$\delta^{13}\text{C}$

Rectangular	41-36, 25, 21
Welsh I	41-32, <b>23-21</b>
Hanning	41-32, <b>23-20</b>
Triangular	38-34, <b>23-20</b>
Blackman Harris	<b>23-19</b>

Growth Rate

Rectangular	30-26, 18, 14, 12
Welsh I	32-26, 19-18
Hanning	36-27, 19-18, 13-12
Triangular	34-26, 19-18, 13-11
Blackman Harris	34-26, 21-18, 13-12

Phase 2 (laminae 244-675)

$\delta^{18}\text{O}$

Rectangular	17.3
Welsh I	<b>17.3-17.6</b>
Hanning	<b>18-17</b>
Triangular	<b>18-17</b>
Blackman Harris	18-17

$\delta^{13}\text{C}$

Rectangular	<b>20-21</b> , 16-17, 14 yr
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Welsh I	<b>20-21</b> , 14 yr
Hanning	<b>19-21</b> , 14 yr
Triangular	<b>20-21</b> , 14 yr
Blackman Harris	<b>20-21</b> , 14 yr

#### Growth Rate

Rectangular	39-33, 11.1
Welsh I	43-35
Hanning	43-34
Triangular	43-34
Blackman Harris	43-36

#### Phase 1 (laminae 676-925)

#### $\delta^{18}\text{O}$

Rectangular	37-26, <b>18-17</b>
Welsh I	33-26, <b>18-15</b>
Hanning	33-26, <b>18-15</b>
Triangular	33-26, <b>18-15</b>
Blackman Harris	33-26, <b>18-15</b>

#### $\delta^{13}\text{C}$

Rectangular	29-25, <b>21-19</b> , (15), (14)
Welsh I	29-25, <b>21-19</b>
Hanning	29-25, <b>21-19</b>
Triangular	29-25, <b>21-19</b>
Blackman Harris	30-26

#### Growth Rate

Rectangular	21-19, 15, 12
Welsh I	21-19, 15
Hanning	21-18, 13-11,
Triangular	21-19, 15, 12
Blackman Harris	21-19, 13-11