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Holocene interaction of maritime and continental climate in Central Europe: new speleothem evidence from Central Germany

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4 Sebastian F.M. Breitenbach^{1,*}, Birgit Plessen², Sarah Waltgenbach³, Rik Tjallingii², Jens

5 Leonhardt⁴, Klaus Peter Jochum⁵, Hanno Meyer⁶, Bedartha Goswami⁷, Norbert Marwan⁷, Denis

6 7 Scholz³

8 ¹ Sediment- and Isotope Geology, Institute for Geology, Mineralogy and Geophysics, Ruhr-

9 Universität Bochum, Universitätsstr. 150, 44801 Bochum, Germany

- 10 * sebastian.breitenbach@rub.de, phone: +49 23432 22307
- ² GFZ-Potsdam, Section Climate Dynamics and Landscape Evolution, Potsdam, Germany
- 12 ³ Institut für Geowissenschaften, Johannes Gutenberg-Universität, Mainz, Germany
- 13 ⁴ Thüringer Höhlenverein, Erfurt, Germany
- ⁵ Climate Geochemistry Department, Max Planck Institute for Chemistry, Mainz, Germany

⁶ Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research, Periglacial Research

- 16 Section, Potsdam, Germany
- ⁷ Potsdam Institute for Climate Impact Research (PIK), Member of the Leibniz Association,
- 18 Potsdam, Germany
- 19

20 Abstract

21 Central European climate is strongly influenced by North Atlantic (Westerlies) and Siberian High 22 circulation patterns, which govern precipitation and temperature dynamics and induce 23 heterogeneous climatic conditions, with distinct boundaries between climate zones. These climate 24 boundaries are not stationary and shift geographically, depending on long-term atmospheric 25 conditions. So far, little is known about past shifts of these climate boundaries and the local to 26 regional environmental response prior to the instrumental era.

High resolution multi-proxy data (stable oxygen and carbon isotope ratios, S/Ca and Sr/Ca) from two Holocene stalagmites from Bleßberg Cave (Thuringia) are used here to differentiate local and pan-regional environmental and climatic conditions Central Germany through the Holocene. Carbon

30 isotope and S/Ca and Sr/Ca ratios inform us on local Holocene environmental changes in and

31 around the cave, while δ^{18} O (when combined with independent records) serves as proxy for (pan-

- 32)regional atmospheric conditions.
- 33 The stable carbon isotope record suggests repeated changes in vegetation density (open vs. dense
- 34 forest), and increasing forest cover in the late Holocene. Concurrently, decreasing S/Ca values
- 35 indicate more effective sulphur retention in better developed soils, with a stabilization in the mid-
- 36 Holocene. This goes in hand with changes in effective summer infiltration, reflected in the Sr/Ca
- 37 profile. Highest Sr/Ca values between 4 ka and 1 ka BP indicate intensified prior calcite precipitation
- 38 resulting from reduced effective moisture supply.

39 The region of Bleßberg Cave is sensitive to shifts of the boundary between maritime (Cfb) and 40 continental (Dfb) climate and ideally suited to reconstruct past meridional shifts of this divide. We 41 combined the Bleßberg Cave δ^{18} O time series with δ^{18} O data from Bunker Cave (western Germany) 42 and a North Atlantic Oscillation (NAO) record from lake SS1220 (SW Greenland) to reconstruct the 43 mean position of the Cfb-Dfb climate boundary. We further estimate the dynamic interplay of the 44 North Atlantic Oscillation and the Siberian High and their influence on Central European climate. 45 Repeated shifts of the Cfb-Dfb boundary over the last 4,000 years might explain previously observed 46 discrepancies between proxy records from Europe. Detailed correlation analyses reveal multi-47 centennial scale alternations of maritime and continental climate and, concurrently, waning and 48 waxing influences of Siberian High and NAO on Central Europe.

49

50 Keywords

51 Germany, Speleothem, Holocene, oxygen isotopes, carbon isotopes, S/Ca, Sr/Ca, palaeoclimate,

52 continental climate, maritime climate, climate boundary

53

54 **1. Introduction**

55 European climate is characterized by heterogeneous climate conditions, with distinct boundaries 56 that demarcate large-scale geographical regions with a coherent climatic pattern (Kottek et al. 57 2006). The coherence between the climatic characteristics within each such climate zone depends 58 on large scale atmospheric systems and can be detected using climate network techniques 59 (Rheinwalt et al. 2016). Central European climate is strongly influenced by intricately linked North 60 Atlantic Oscillation (NAO) and Siberian High (SH), which govern (winter) precipitation and 61 temperature over Europe. Both, NAO and SH, are expressions of (mainly winterly) semi-permanent, 62 guasi-stationary surface pressure features that reside over the North Atlantic and northern Eurasia 63 that closely interact with each other (Cohen et al. 2001). We will illuminate the non-stationary 64 interaction between NAO and SH and its relation to the boundary that separates maritime and 65 continental climates in the discussion below.

66 The boundary between the modern maritime Cfb and continental Dfb climatic zones according to 67 the Köppen-Geiger classification (Peel et al. 2007), is not stationary, but shift in space and time 68 (Kottek et al. 2006). Future shifts of the Cfb-Dfb climatic boundary mirror circulation changes that 69 result from global warming and might lead to more frequent extreme weather patterns like heat 70 waves and droughts, with significant repercussions for society (Cohen et al. 2014, Mann et al. 2018). 71 To delineate the mechanisms related to Central European climate, it is imperative to understand the 72 interaction of maritime and continental climates and the positioning of the border between the two. Climatic changes from multi-annual to centennial timescales can affect human society (Büntgen et 73 74 al. 2011, 2016; Kennett & Breitenbach et al. 2012; Ludlow et al. 2013; Tan et al. 2015). Pre-industrial 75 communities experienced spatio-temporal variations in regional precipitation and temperature

76 (droughts, floods, unusual cold spells) more severely due to the direct impact on agricultural yield,

in combination with high production and re-distribution costs. While multiple underlying causes for
temperature or precipitation changes have been invoked, including solar and volcanic forcing,
coupled with changes in atmospheric circulation patterns (Hurrell et al. 1995, Brönnimann et al.
2007, Ludlow et al. 2013, Ridley et al. 2015, Thieblemont et al. 2015, Büntgen et al. 2016), little is
known about the associated changes in seasonal climate hydrological and thermal conditions in

82 Central Europe for the Holocene (Simonis et al. 2012).

83 Temporally highly resolved (i.e. seasonal to decadal) multi-proxy climate reconstructions are the 84 most promising tool to put the local and regional environmental response into perspective to global 85 changes, including rising temperatures. Most importantly, there is an urgent need for palaeoclimate reconstructions, which are sensitive to seasonal aspects of regional and local climate as well as 86 87 environmental conditions (Wong & Breeker 2015, James et al. 2015, Rehfeld et al. 2016). Only with 88 sufficiently high resolved and spatially distributed reconstructions can we begin to quantify shifts in 89 climate boundaries (Seager et al. 2018). Holocene climatic and environmental variability in Central 90 Europe (and the responsible forcings) remains insufficiently understood, due to seasonal biases in 91 some palaeoclimate time series, and a lack of chronological control in others. Although tree ring 92 records give very detailed insights into changes in summer temperatures and precipitation, they 93 cover only the last ~2.5 ka (Büntgen et al. 2006, 2011). Available high-resolution lacustrine 94 reconstructions, based on robust varve counting and/or radiocarbon chronologies, often cover only 95 parts of the Holocene (e.g. von Grafenstein et al. 1999, Martin-Puertas et al. 2012, Czymcik et al. 96 2016, Malkiewicz et al. 2016).

97 Speleothems (secondary cave carbonates) constitute powerful palaeo-environmental archives that 98 can be dated very accurately radiometrically and provide a large number of environmental and 99 climate proxies (Dorale et al. 2007, Fairchild & Baker 2012). However, so far, only a few speleothem-100 based Holocene records are available from Europe, mainly from southern and western Europe 101 (Fohlmeister et al. 2012, Frisia et al. 2005a, McDermott et al. 2011, Mischel et al. 2017, Niggemann 102 et al. 2003, Smith et al. 2016, Verheyden et al. 2000). One exception is the well dated high-resolution 103 isotope record from Spannagel Cave, but due to its position in the Alps, it remains difficult to 104 separate the influencing factors involved (Mangini et al. 2005, Fohlmeister et al. 2013). Highly 105 resolved, well-dated time series are available, but limited to the last few thousand years (e.g. Proctor 106 et al. 2000, Boch et al. 2009, Mangini et al. 2005). While these time series have added important 107 details to our understanding of the factors that regulate European climate variability, additional 108 spatial coverage is required to fully comprehend local to regional variability.

Here we present a multi-decadally to decadally resolved Holocene multi-proxy reconstruction from two stalagmites from Bleßberg Cave, Thuringia, Germany, and investigate shifts of the mean position of the maritime-continental climate boundary through time. The location of Bleßberg Cave at the western limit of continental climate region makes this study site particularly well suited to investigate spatio-temporal changes of the Cfb-Dfb boundary. δ^{13} C and δ^{18} O are combined with S/Ca and Sr/Ca records to gain insights into past local and (pan-)regional environmental dynamics in Central Europe over the last 11,000 years. We attempt to single out environmental responses to climatic changes at a more continental setting and to reconcile apparently divergent palaeoclimate reconstructions from different European regions. We argue that east-west shifting of the maritime and continental climate boundary might explain observed differences between palaeoclimate reconstructions from western and eastern sites. Our results suggest that reconstructions from western sites might not be representative of conditions further east and *vice versa* and that time series networks are needed to better understand local responses to global climatic changes.

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123 **2. Geographic and geologic setting**

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2.1 Geology and geography of Bleßberg Cave

The NW-SE oriented Bleßberg Cave is located at 50°25'28" N and 11°01'13" E at ca. 500 m a.s.l. at the southern fringe of the Thuringian schist mountains ca. 7 km from the town of Eisfeld, Germany (Fig. 1). Bleßberg Cave developed in Triassic marly limestones (Lower Muschelkalk/Anisium) and is orientated parallel to the Franconian Line, a Hercynian (NW-SE) directed, deep-reaching Cretaceous reverse fault (Reicherter et al. 2008). The cave was discovered in 2008 during construction of the new high-speed rail line between Berlin and Munich. Blasting work opened the cave ~240 m from the south portal of the ~8 km long Bleßberg tunnel.

- 132The cave had no naturally accessible entrance prior to its discovery, but is drained by a small133stream. The lack of a natural entrance preserved the cave from human or animal disturbance before
- 134 2008. The cave's atmosphere is relatively stable, but the stream running through the main passage
- and airflow through breakdown could influence cave ventilation.

Bleßberg Cave is overlain by 12 to 50 m (35-40 m above the sampling site) of marly limestone (Lower and Middle Muschelkalk), which carries a relatively thin soil. The soil can be classified as agriculturally altered Leptosol (Rendzina, Ap-Ah-(T)-Cv-Cn, Boden AG 2006), possibly with clay enrichment below the Ah horizon. The vegetation directly above the cave is anthropogenically altered farmland, consisting mainly of weeds and crops, whereas uphill, mixed deciduous and pine forests take over.

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2.2 Climatic characterization of the study site

144 The cave site is located near the watersheds between Weser, Elbe and Rhine rivers. Modern climate 145 can be characterized as Cfb climate in the Köppen classification, with a mean annual air 146 temperature of ca. 6°C at Neuhaus am Rennweg, ca. 12 km NE from the cave (Kottek et al. 2006). 147 Rather high precipitation (862 mm/year, DWD Climate Data Center. 148 www.dwd.de/EN/ourservices/cdcftp/cdcftp.html) results from orographic precipitation on the 149 southwest-facing slope of the Thuringian mountains. Meteorological data from the station Neuhaus 150 am Rennweg shows no significant seasonality in precipitation, whereas air temperature is highest 151 in July (average $T_{Jul} = 14.6^{\circ}$ C) and coldest in January (average $T_{Jan} = -3.2^{\circ}$ C) (Fig. 2).

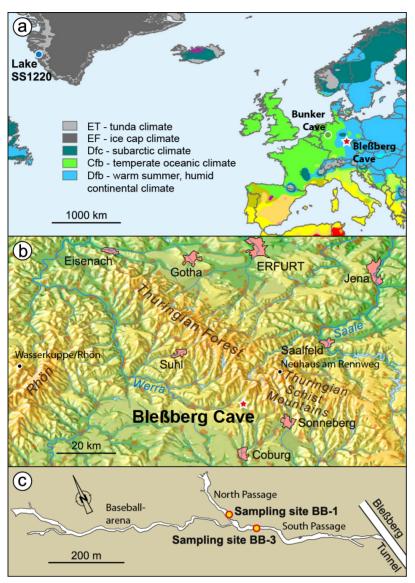


Figure 1: A) Location of Bleßberg Cave (red circle) in Central Europe. Bunker Cave and lake SS1220 in Greenland are
shown as green star and blue circle respectively. Color coding indicates climate regimes according to Peel et al. (2007).
The boundary between Cfb and Dfb climates crosses Central Europe meridionally. B) The study site is located at the
SW facing slope of the Thuringian Schist Mountains near Eisfeld. The 2.5D elevation panorama has kindly been provided
by mr-kartographie, Gotha, and is subject to copyright. C) Bleßberg Cave is oriented NW-SE, parallel to the Franconian
Line, and was discovered during construction of the Bleßberg railway tunnel. The sampling sites are roughly in the
middle of the poorly ventilated cave.

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162 The seasonality in temperature leads to significant variation in potential and real evapotranspiration,

163 which in turn leads to maximum effective infiltration between September and March (Fig. 2). Thus,

164 the isotopic composition of dripwater entering Bleßberg Cave should be slightly biased towards the

165 winter season, with implications for the interpretation of the $\delta^{18}O_{\text{speleothem}}$ signal, similar to other caves

166 in Germany (e.g. Wackerbarth et al. 2010, Mischel et al. 2015).

167 Observational rainfall and temperature data reveals that winter temperature at Neuhaus am 168 Rennweg correlates positively with the NAO index (data from ftp://ftp.cpc.ncep.noaa.gov), which is

169 in agreement with the results of Baldini et al. (2008). The modern location of the Cfb-Dfb boundary

east of Bleßberg Cave mirrors the oceanic influence in Central Europe (Kottek et al. 2006). In line with earlier results (Baldini et al. 2008, Riechelmann et al. 2017), we are confident that δ^{18} O in dripwater and speleothems can be used to study winter climate when the site is influenced by the Siberian High, which brings cold and dry air masses to Bleßberg Cave. The position of the cave near the western limit of the continental Dfb climate regime makes it highly sensitive to shifts of the boundary between the Cfb and the Dfb climate further west.

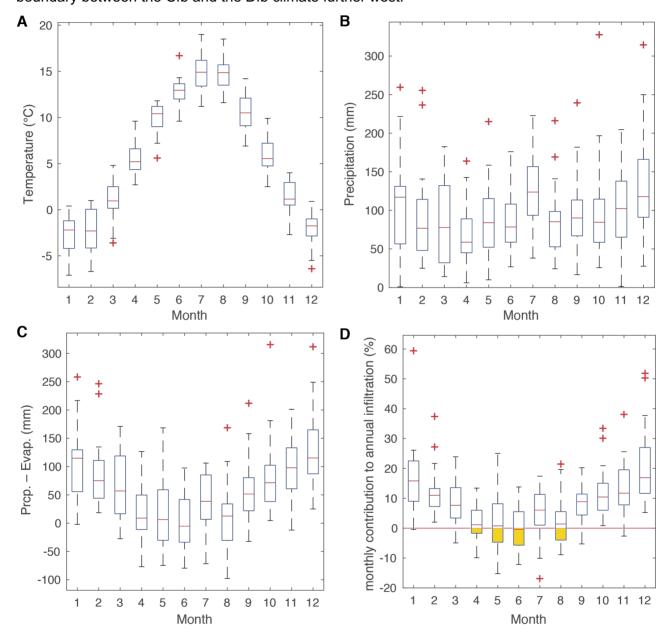




Figure 2: Boxplots of main meteorological characteristics at station Neuhaus am Rennweg. A) A clear seasonality,
with maximum in summer is found in monthly temperature, B) monthly precipitation shows no distinct seasonal
variability, C) monthly effective precipitation (precipitation minus potential evapotranspiration after Haude
1954) with maximum infiltration in winter, and D) monthly effective infiltration in percent of annual infiltration.
Negative interquartile ranges are shown in yellow. The dataset includes the interval 31.05.1989 to 31.12.2014.
Source: DWD climate data center.

184 This position however also means that any link between Bleßberg and the NAO system might be 185 lost if the climate boundary is located west of the cave and continental climate prevails (see 186 discussion).

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2.3 Samples

The stalagmite samples BB-1 and BB-3 were recovered following the discovery of the cave. Sample BB-1 is 17.7 cm long and ca. 7 cm wide, while BB-3 is ca. 15 cm long with a diameter of ca. 9 cm. BB-1 was broken before collection, and the original drip site feeding this sample is thus unknown. Sample BB-3 was collected from the main passage and showed a fresh and wet surface (Fig. 1c). The samples were cut along their growth axes and polished and show a very clear, pale-ochre color, with rarely visible lamination and very little detrital material.

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196 **3. Methodology**

197**3.1 Cave monitoring**

198 In order to gain insight into cave ventilation and infiltration dynamics, which influence the 199 geochemical proxies in Bleßberg Cave, microclimatic parameters are monitored. Between 2009 and 200 2012, monitoring was severely limited; since January 2013, data collection improved with renewed 201 access to the cave. A CORA device (Luetscher & Ziegler 2012) is used to log temperature, humidity, 202 air pressure and pCO₂ at 4-hour intervals. Dripwater was collected in airtight 5-12 mL vials during 203 our visits and stored in cool, dark conditions until analysis. Stable isotope values (δ^{18} O and δ D) of 204 dripwater were determined at the Alfred Wegener Institute (AWI) in Potsdam. 18 samples were 205 collected to cover the different seasons (Table 1, Fig. 3). A Finnigan MAT Delta-S mass 206 spectrometer equipped with two equilibration units was used for the online determination of 207 hydrogen and oxygen isotopic composition. The external errors for standard measurements of 208 hydrogen and oxygen are better than 0.8‰ and 0.1‰, respectively (Meyer et al. 2000).

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3.2 Mineralogy and geochemistry

211 Optical microscopy and fluorescence microscopy were performed on a Zeiss Jenalumar, SEM 212 imaging and X-ray diffraction (XRD) at the Helmholtz Centre Potsdam, Deutsches 213 GeoForschungsZentrum, Potsdam, Germany, to verify the mineral structure and lamination of the 214 stalagmite samples. BB-1 consists of calcite and has been tested with cathodoluminescence (CL) 215 microscopy to test for any diagenetic alterations. BB-3 grew on a flowstone in the middle of a ca. 8 216 m high passage and consists of calcite, too. Most of the stalagmite shows very clear parallel calcite 217 crystals and horizontal growth layers, making the stalagmite ideally suited for palaeoclimate studies. The lowermost centimeters of the sample are already part of the underlying flowstone and show 218 219 brownish clay layers and radial fan-structured crystals. Examination of thin sections under blue light 220 reveals growth intervals with bright green laminae, suggesting the presence of organic material (Fig. 221 4b).

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223 **3.3**²³⁰Th/U dating

224 For ²³⁰Th/U-dating, sub-samples with masses between 50 and 200 mg were cut from the growth 225 axes of the individual stalagmites using a precision diamond wire saw as well as a micro band saw. 226 In total. 41 230 Th/U-ages were determined (N_{BB-1} = 18, N_{BB-3} = 23, Table 2). Chemical preparation 227 and mass spectrometric analysis were performed at the Max Planck Institute for Chemistry, Mainz. For separation of Th and U, the samples were dissolved in 7N HNO₃, and a mixed ²²⁹Th-²³³U-²³⁶U 228 229 spike was added (see Gibert et al., 2016, for a detailed description of spike calibration). Potential 230 organic material was removed by addition of a mixture of concentrated HNO₃, HCl and H₂O₂. After 231 evaporation and re-dissolution in 6N HCI, U and Th were chemically separated using ion exchange 232 column chemistry (Yang et al., 2015). For mass spectrometric measurements, the separated 233 fractions of U and Th were dissolved in 2 ml of 0.8N HNO₃ and analysed with a Nu Plasma multi-234 collector inductively coupled plasma mass spectrometer (MC-ICPMS). Analytical details are 235 described in Obert et al. (2016). Ages were calculated using the half-lives of Cheng et al. (2000), and the correction for potential detrital contamination assumes an average ²³²Th/²³⁸U weight ratio 236 of the upper continental crust (3.8±1.9) and ²³⁰Th, ²³⁴U and ²³⁸U in secular equilibrium. 237

Age-depth models for stable isotope and XRF proxy profiles were established using COPRA (Breitenbach et al. 2012). In order to calculate 2.5% and 97.5% quantile confidence limits (ca. 2σ), 5000 Monte Carlo (MC) simulations were run using a piecewise cubic interpolating. COPRA transposes the chronological uncertainty to the proxy domain, so that the x-axis (time) is error-free (Breitenbach et al. 2012). This procedure allows statistical reanalysis with the entire ensemble of age models.

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3.4 Stable carbon and oxygen isotope analysis

Stable carbon and oxygen isotope ratios of the stalagmites (δ^{13} C and δ^{18} O) were determined at the 246 247 Helmholtz-Centre Potsdam (GFZ). Stable isotope samples have been obtained using a "Sherline 248 5410" vertical milling machine with an inhouse-built sample stage and a micrometer-drive to allow 249 precise movement of the speleothem block, for stepwise milling at a resolution of 0.1 mm. Carbonate aliquots were analysed for δ^{13} C and δ^{18} O values using a Finnigan MAT253 isotope ratio 250 251 mass spectrometer (ThermoFisher Scientific) connected to an automated carbonate-reaction device 252 (KIEL IV). Samples of around 20-60 µg were automatically dissolved with 103% H₃PO₄ at 72°C, and 253 the isotopic composition was measured on the released and cryogenically purified CO₂. Results are 254 reported relative to VPDB, and replicate analysis of international reference material (NBS19) yielded external standard deviations of 0.06 ‰ (1 σ) for both δ^{13} C and δ^{18} O. 255

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3.5 Micro-XRF analysis

The distribution of elements detectable with micro X-ray fluorescence (µXRF) has been determined in both samples, BB-1 and BB-3 (Figs. 4a, b). Both samples were scanned along the stable isotope track using an EAGLE III XL μ XRF spectrometer (Röntgenanalytik, Germany) at the GFZ. The μ XRF is equipped with a 50 W Rh X-ray source that was operated at 40kV and 300 μ A. Measurements were performed under vacuum every 40 μ m with a spot size of 50 μ m and a counting time of 60 s. The resulting intensities are given in counts per second (cps) and the reproducibility is proven with repeated measurements. Besides Ca, significant count variations were found for Sr and S. Element intensities have been normalized over Ca to minimize sample density effects and scaled by multiplying the intensity ratios by 1000.

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4. Results

4.1 Cave microclimate monitoring

Bleßberg Cave has a rather stable thermal regime, with a mean temperature of $8.7\pm0.1^{\circ}$ C and a relative humidity of 99.8±0.2 %. Cave air pCO₂ is slightly elevated relative to outside air and varies from 780 to 824 ppm, with a maximum in winter, which probably results from enhanced effective infiltration and introduction of soil CO₂ in winter. Although the level of the cave stream changes significantly in the eastern cave passages, it seems not to strongly alter the cave atmospheric conditions.

276 Stable oxygen and deuterium isotope values (Table 1) from 18 Bleßberg Cave dripwater samples 277 are relatively invariable, with $\delta D = -65.5 \pm 1.7\%$ and $\delta^{18}O = -9.5 \pm 0.2\%$ (Fig. 3a) and agree well with 278 the winter season (Sep-Mar) averages of precipitation from GNIP stations Wasserkuppe/Rhön 279 $(\delta D_{Sep-Mar} = -69.6\%, \delta^{18}O_{Sep-Mar} = -10.1 \pm 1\%, IAEA/WMO 2017), Hof (\delta D_{Sep-Mar} = -71.3\%, \delta^{18}O_{Sep-Mar})$ = -9.8±1.3‰) and Leipzig ($\delta D_{Sep-Mar}$ = -67.1‰, $\delta^{18}O_{Sep-Mar}$ = -9.6±1.4‰) (Figs. 3a, b, IAEA/WMO 280 281 2017; meteorological data cover 1983-2003 for Hof and Wasserkuppe/Rhön, and 1986-2003 for 282 Leipzig). The slightly lower mean winter value for Wasserkuppe/Rhön is due to the altitude effect 283 (the station's altitude is 921 m a.s.l., compared to 567 m in Hof and 125 m in Leipzig). All values fall 284 on the Global Meteoric Water Line (GMWL, Fig. 3b), which indicates that no secondary evaporation 285 effects influence our samples. The d excess is in the range of global average of precipitation. Temporal and spatial dripwater δ^{18} O variability is rather small, with only about 0.6 % variation 286 287 between different sampling days and sampling spots in different cave passages, but denser 288 sampling is needed to evaluate this aspect.

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4.2 ²³⁰Th/U dating and age modeling

The results of ²³⁰Th/U-dating are presented in Table 2. The ²³⁸U concentration of the stalagmites varies between 0.614±0.009 (BB-3) and $3.74\pm0.02 \ \mu g \ g^{-1}$ (BB-1). BB-1 has a substantially higher ²³⁸U-concentration compared to BB-3. The ²³²Th-concentration is generally low (<1 ng g⁻¹; Table 2) and even below the detection limit for some samples.

Only four samples show an elevated input of detrital material with a 232 Th-content between 1.09±0.01 ng g⁻¹ (NR-31 of BB-1) and 55.87±0.53 ng g⁻¹ (BB3-5.2 of BB-3), respectively. All four samples were taken at the base of the two stalagmites. The relatively high concentrations of uranium 298 (up to 3.4 μ g/g) and lack of detrital thorium (Table 2) result in mean dating errors of ±30 years in 299 BB-1 and ±86 years in BB-3 (2 σ).

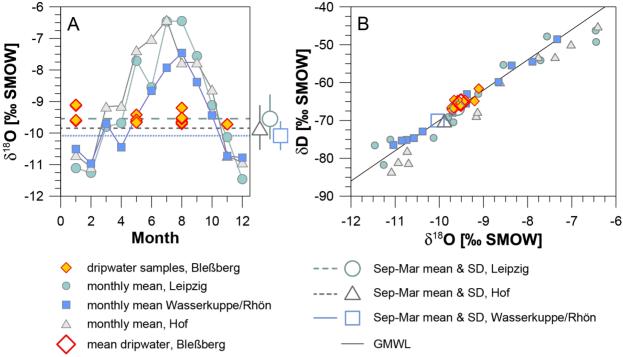
300 Stalagmite BB-1 grew between 5.6 and 0.6 ka BP (kilo years before present, i.e. 1950 CE), while 301 BB-3 grew between 11.9 and 5.4 ka BP (Fig. 5). Unfortunately, the last ca. 600 years are missing 302 and the reconstruction cannot be directly linked with meteorological data.

303 The age-depth slopes indicate that both stalagmites grew at different rates; slow growth prevailed 304 between ca. 11 and 7 ka BP and 3.7 and 0.2 ka BP, whereas faster growth occurred in the mid-305 Holocene between 6 and 4 ka BP (Fig. 5). BB-3 shows relatively steady growth in the early Holocene 306 and a slight increase after ca. 7 ka BP. A hiatus has been identified during ²³⁰Th/U-dating of BB-3. 307 located at 100.2 mm from top, which is characterized by yellow-brownish coloring of the stalagmite. 308 This hiatus covers a ca. 400-year long interval between 8.26 and 7.85 ka BP. BB-1 shows a drastic 309 reduction in growth rate after ca. 4.7 ka BP, approaching values more similar to those in the early 310 Holocene. While growth rates differ, the convergence in both stalagmites during the overlapping 311 period suggests common forcing(s) that adjust the growth pattern. The interval where both 312 stalagmites overlap (ca. 5.6 to 5.2 ka BP) reveals that BB-3 (the older of the two) has lower δ^{13} C and S/Ca, and higher Sr/Ca and δ^{18} O values compared to BB-3. BB-1 also shows higher high-313 314 frequency variability.

315

Sample ID	Sampling	Location	δ ¹⁸ Ο [‰	δ²Η [‰	d
	date		VSMOW]	VSMOW]	excess
BBH-NG-23.01.09	23.01.2009	N. Passage	-9.61	no data	no data
BBH-NG-23.01.09	23.01.2009	N. Passage	-9.62	-65.2	11.7
BBH-NG-23.01.09	23.01.2009	N. Passage	-9.59	-66.1	10.6
BBH-SG-24.01.09	24.01.2009	S. Passage	-9.11	-61.6	11.3
BBH-SG-24.01.09	24.01.2009	S. Passage	-9.13	NAN	NAN
BBH-SG-24.01.09	24.01.2009	S. Passage	-9.11	-61.6	11.3
BBH-MP1 24.11.2012	24.11.2012	Station 1	-9.72	-66.9	10.9
BBH-MP2 24.11.2012	24.11.2012	Station 1	-9.72	-66.6	11.2
dripwater 24.08.2013 BB-1	ter 24.08.2013 BB-1 24.08.2013		-9.71	-67.3	10.4
dripwater 24.08.2013 BB-1	ipwater 24.08.2013 BB-1 24.08.2013		-9.64	-66.7	10.4
dripwater 24.08.2013 86 24.08.20		S. Passage	-9.67	-66.7	10.7
dripwater 24.08.2013 2m 86	24.08.2013	S. Passage	-9.20	-64.9	8.7
drip water 24.08.2013 103	ip water 24.08.2013 103 24.08.2013		-9.56	-66.1	10.4
drip water 24.08.2013 2m 103	ip water 24.08.2013 2m 103 24.08.2013		-9.52	-66.0	10.2
rip water 27.05.2017, MP90 27.05.2017		Baseball Arena	-9.63	-64.9	12.2
drip water 27.05.2017, MP90	27.05.2017	Baseball Arena	-9.42	-64.6	10.8
drip water 27.05.2017, MP90	drip water 27.05.2017, MP90 27.05.2017		-9.58	-64.6	12.1
drip water 27.05.2017, MP90	27.05.2017	Baseball Arena	-9.67	-64.6	12.8

316 **Table 1** Dripwater samples collected in Bleßberg Cave for stable isotope analysis.



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Figure 3: Stable isotope data from dripwater and precipitation. A) Dripwater δ^{18} O data from Bleßberg Cave 319 compared with mean monthly δ^{18} O of precipitation at Wasserkuppe/Rhön (squares), Leipzig (circles) and Hof 320 (triangles) (IAEA/WMO 2017). B) Cross-plot of dripwater and monthly mean precipitation δ^{18} O and δ D values 321 from the same stations. Large empty symbols denote mean values for the winter season (September to March).

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323 Before an interpretation in terms of environmental changes can be attempted the observed offsets 324 must be explained as both stalagmites are apparently precipitated from different feeding systems. 325 BB-1 was likely fed from fracture flow, whereas BB-3 received seepage flow water from a low-326 permeability host rock matrix (Fairchild and McMillan 2007).

327 Seepage flow from the epikarst matrix goes in hand with prolonged residence time of infiltrating water and mixing of waters of different age. This ultimately leads to smoothing of the proxy signals, 328 329 which explains the reduced high-frequency variability in BB-3. Seepage flow from a less permeable 330 matrix forces longer interaction of the infiltrating water with soil and host rock, which can well explain 331 the observed lower δ^{13} C and S/Ca ratios, and higher Sr/Ca levels in BB-3. Lower δ^{13} C in BB-3 332 compared to BB-1 can be explained by less CO₂-degassing in a matrix with lower permeability, i.e. 333 seepage from a host rock matrix is less prone to open conditions and PCP, compared to fracture 334 flow where open conditions might occur faster and more frequently (Fairchild and McMillan 2007). 335 Lower S/Ca ratios in BB-3 might result from better sulfur retention in the host rock, compared to

- 336 larger fractures. Prolonged dissolution of host rock in a seepage flow system can also increase the
- 337 Sr/Ca baseline in BB-3 (which is subsequently modulated by PCP dynamics). On the other hand, a
- 338 fracture-flow fed BB-1 would reflect a faster response to changes in infiltration, higher S/Ca caused
- 339 by lower sulfur retention in the soil and epikarst, and a lower Sr/Ca baseline due to shorter water-
- rock interaction. Finally, the lower δ^{18} O values observed in BB-1 might result from a more direct 340
- 341 response to cold-season infiltration and/or spring snowmelt through the fractures feeding BB-1, and

342 less mixing with summer-season water in the epikarst. These factors must be kept in mind when 343 interpreting the proxy variability in both stalagmite records. We note however that millennial-scale 344 trends and centennial dynamics in both records are still interpretable in terms of environmental 345 conditions at the cave site."

346

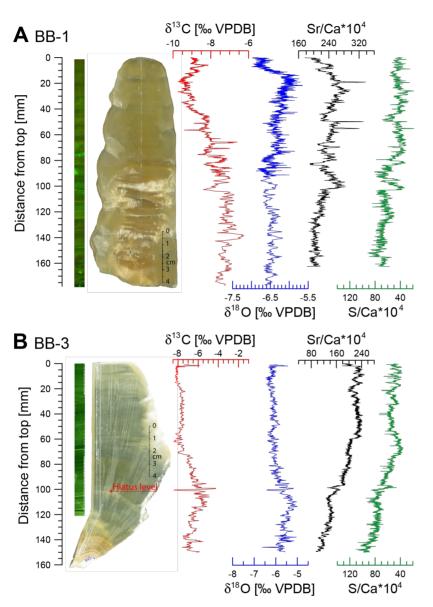




Figure 4: Images of stalagmites BB-1 (A) and BB-3 (B), together with fluorescence images, stable isotope profiles (δ^{13} C and δ^{18} O) and μ XRF Sr/Ca, and S/Ca ratios. The lower brownish section of BB-3 belongs genetically to the flowstone onto which BB-3 grew and the geochemical data from this interval is not discussed in the main text.

351

4.3 Stable isotopes

In total, about 2000 samples have been analysed for stable isotopes in both stalagmites (1451 in BB-1, and 538 in BB-3). The total amplitude in δ^{18} O across the entire Holocene is ~2‰, from -5‰ around 9 ka BP to -7‰ in the last millennium (Figs. 4a, 6). In the younger stalagmite BB-1, δ^{18} O values range from -7‰ to -5.7‰, with an increasing trend from ca. -6.5‰ in the lower part towards a maximum value of -5‰ at 1.3 ka BP. At the top, the BB-1 profile shows a large shift to values

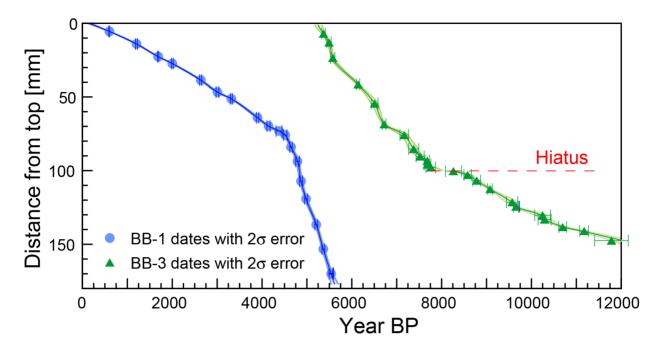
- around -6.8‰. Superimposed on the millennial to centennial changes, multi-decadal variability is detected with amplitudes of 0.3 to 0.5‰. In the older stalagmite BB-3, δ^{18} O values vary from -8‰ to -5‰. A trend to lower values is observed in the upper half of this stalagmite (since ~9 ka BP), which is only interrupted by two prominent excursions before and after the hiatus around 8 ka BP. A first notable shift of ~0.8‰ to lower values occurred just before the hiatus. A second shift is found after the hiatus, with a rapid change from values around -5.4‰ to -6‰ (Figs. 4b, 6). No clear trend is found between 7.2 and 5.3 ka BP, but multi-centennial scale variability of ~0.2-0.4‰ characterizes
- this section. Where BB-3 overlaps with BB-1, it is offset and less negative by ~0.5‰.
- Across both stalagmites, δ^{13} C values vary ~8.5‰, from -9.8‰ to -1‰ (Figs. 4, 6), with a general trend towards lower values through the Holocene (Fig. 6). A plateau, with values around -7‰, is
- found between 11 ka BP and 9 ka BP. This stable phase is interrupted at ~11 ka BP, when δ^{13} C
- 369 increased about 1‰ for 400 years. Around 8.7 ka BP, a rapid 2‰ shift to more negative values
- 370 occurred, followed by the hiatus (~8.3-7.8 ka BP). This hiatus lasted 400 years, after which growth
- 371 commenced again. The δ^{13} C profile restarted with values around -6‰, decreasing steadily to -8‰
- around 7 ka BP (Fig. 6). Variable δ^{13} C values are found during the mid-Holocene. After 3 ka BP, a further decrease occurred, with the lowest values of the entire profile at around 2 ka BP, and a last
- 374 shift to higher values since 1.5 ka BP.
- 375 In addition to the samples obtained for proxy records, ~250 samples were drilled for Hendy tests
- 376 (Hendy 1971). In BB-1, 5 Hendy test transects were performed, with 10 to 15 samples each. Neither
- 377 a significant positive correlation between δ^{13} C and δ^{18} O, nor an increase of δ^{18} O values with distance
- 378 from the apex is observed. From BB-3, no Hendy tests have been obtained.
- 379

Sample ID	Depth [mm]	²³⁸ U [μg g ⁻¹]	²³² Th [ng g ⁻¹]	(²³⁴ U/ ²³⁸ U)	(²³⁰ Th/ ²³⁸ U)	(²³⁴ U/ ²³⁸ U) _{initial}	age _{uncorrected} [ka BP]	age _{corrected} [ka BP]
BB-1 17	5.00	1.772 ± 0.011	0.0334 ± 0.0018	2.5544 ± 0.0031	0.01546 ± 0.00026	2.5573 ± 0.0030	0.600 ± 0.011	0.599 ± 0.011
BB-1 25	13.75	2.240 ± 0.012	< LoD	2.5606 ± 0.0033	0.02956 ± 0.00048	2.5662 ± 0.0033	1.204 ± 0.020	1.204 ± 0.020
BB-1 18	22.00	2.145 ± 0.013	0.0431 ± 0.0013	2.5601 ± 0.0036	0.04076 ± 0.00034	2.5678 ± 0.0036	1.687 ± 0.015	1.687 ± 0.015
BB-1 1	27.00	1.789 ± 0.011	0.02200 ± 0.00087	2.5363 ± 0.0039	0.04755 ± 0.00045	2.5453 ± 0.0038	2.002 ± 0.020	2.202 ± 0.020
BB-1 26	38.00	2.254 ± 0.012	< LoD	2.5598 ± 0.0034	0.06263 ± 0.00050	2.5717 ± 0.0034	2.636 ± 0.023	2.636 ± 0.022
BB-1 19	46.00	2.480 ± 0.016	0.0552 ± 0.0019	2.6334 ± 0.0047	0.07322 ± 0.00071	2.6477 ± 0.0047	3.008 ± 0.031	3.008 ± 0.032
BB-1 9	50.75	2.330 ± 0.015	0.0501 ± 0.0013	2.6158 ± 0.0037	0.07996 ± 0.00055	2.6313 ± 0.0037	3.317 ± 0.024	3.317 ± 0.020
BB-1 20	64.00	2.424 ± 0.017	0.0350 ± 0.0011	2.5585 ± 0.0084	0.09154 ± 0.00060	2.5761 ± 0.0083	3.902 ± 0.030	3.902 ± 0.030
BB-1 27	69.00	2.448 ± 0.013	< LoD	2.5770 ± 0.0035	0.09779 ± 0.00076	2.5958 ± 0.0036	4.146 ± 0.033	4.146 ± 0.034
BB-1 28	73.00	2.332 ± 0.012	< LoD	2.5883 ± 0.0031	0.1036 ± 0.0015	2.6083 ± 0.0032	4.379 ± 0.067	4.379 ± 0.068
BB-1 10	75.75	2.838 ± 0.019	0.0588 ± 0.0016	2.5686 ± 0.0033	0.10590 ± 0.00081	2.5891 ± 0.0032	4.516 ± 0.035	4.516 ± 0.040
BB-1 21	84.00	2.235 ± 0.014	0.0879 ± 0.0017	2.6106 ± 0.0035	0.11064 ± 0.00057	2.6321 ± 0.0035	4.647 ± 0.025	4.646 ± 0.025
BB-1 29	93.75	3.489 ± 0.019	0.3223 ± 0.0062	2.5826 ± 0.0036	0.11276 ± 0.00092	2.6045 ± 0.0037	4.792 ± 0.041	4.791 ± 0.041
BB-1 30	107.00	3.739 ± 0.020	0.6705 ± 0.0087	2.5768 ± 0.0034	0.11429 ± 0.00077	2.5989 ± 0.0034	4.872 ± 0.034	4.870 ± 0.035
BB-1 2	119.50	3.146 ± 0.022	0.0953 ± 0.0016	2.6056 ± 0.0062	0.11809 ± 0.00064	2.6286 ± 0.0062	4.981 ± 0.031	5.981 ± 0.030
BB-1 11	137.00	3.445 ± 0.022	0.4939 ± 0.0056	2.6072 ± 0.0036	0.12344 ± 0.00068	2.6314 ± 0.0035	5.210 ± 0.030	5.209 ± 0.030
BB-1 3	153.50	3.153 ± 0.020	0.6703 ± 0.0073	2.6034 ± 0.0036	0.12683 ± 0.00063	2.6282 ± 0.0036	5.367 ± 0.029	5.365 ± 0.030
BB-1 31	170.25	3.489 ± 0.020	1.0871 ± 0.0140	2.5966 ± 0.0053	0.1305 ± 0.0010	2.6221 ± 0.0054	5.542 ± 0.045	5.539 ± 0.045
BB3-1	7.00	1.0058 ± 0.0052	0.00249 ± 0.00055	2.6449 ± 0.0029	0.12900 ± 0.00090	2.6703 ± 0.0029	5.370 ± 0.040	5.370 ± 0.039
BB3-3.1	13.00	1.1394 ± 0.0073	0.01789 ± 0.00092	2.628 ± 0.010	0.1311 ± 0.0010	2.654 ± 0.010	5.493 ± 0.049	5.493 ± 0.049
BB3-2.1	23.50	1.3399 ± 0.0077	0.01428 ± 0.00039	2.6977 ± 00.0086	0.1365 ± 0.0011	2.7250 ± 0.0084	5.577 ± 0.048	5.576 ± 0.047
BB3-2	41.50	1.4212 ± 0.0074	< LoD	2.7119 ± 0.0026	0.15060 ± 0.00087	2.7421 ± 0.0027	6.140 ± 0.038	6.140 ± 0.038
BB3-3.2	54.50	1.263 ± 0.010	0.02225 ± 0.00098	2.645 ± 0.017	0.1554 ± 0.0015	2.676 ± 0.016	6.508 ± 0.077	6.507 ± 0.076
BB3-3	68.50	0.9372 ± 0.0049	0.01793 ± 0.00057	2.6959 ± 0.0031	0.1634 ± 0.0010	2.7288 ± 0.0032	6.721 ± 0.042	6.721 ± 0.042
BB3-4.1	75.8	0.9140 ± 0.0050	0.0369 ± 0.0010	2.7006 ± 0.0037	0.1742 ± 0.0022	2.7357 ± 0.0037	7.168 ± 0.094	7.168 ± 0.095
BB3-4.2	85.3	0.8869 ± 0.0049	0.01965 ± 0.00083	2.6871 ± 0.0036	0.1782 ± 0.0026	2.7229 ± 0.0036	7.38 ± 0.11	7.38 ± 0.11
BB3-4.3	90.4	1.0252 ± 0.0055	0.02138 ± 0.00070	2.6986 ± 0.0031	0.1823 ± 0.0021	2.7354 ± 0.0031	7.520 ± 0.090	7.520 ± 0.090
BB3-4	93.3	0.9824 ± 0.0051	0.01449 ± 0.00060	2.6941 ± 0.0027	0.1857 ± 0.0012	2.7316 ± 0.0027	7.681 ± 0.049	7.680 ± 0.049
BB3-4.4	95.9	0.9500 ± 0.0051	0.02093 ± 0.00061	2.7095 ± 0.0032	0.1869 ± 0.0017	2.7474 ± 0.0033	7.685 ± 0.075	7.684 ± 0.075

 Table 2: Results of the ²³⁰Th/U-dating. Activity ratios are indicated by parentheses.

BB3-4.5	97.8	0.6847 ± 0.0037	0.0813 ± 0.0019	2.7051 ± 0.0031	0.1883 ± 0.0025	2.7432 ± 0.0031	7.76 ± 0.10	7.76 ± 0.11
BB3-4.6	100.3	0.8475 ± 0.0046	0.0406 ± 0.0012	2.6870 ± 0.0032	0.2036 ± 0.0026	2.7282 ± 0.0033	8.54 ± 0.12	8.54 ± 0.12
BB3-4.7	102.9	0.8829 ± 0.0047	0.0458 ± 0.0011	2.6979 ± 0.0033	0.2069 ± 0.0019	2.7399 ± 0.0034	8.581 ± 0.083	8.580 ± 0.083
BB3-2.2	106.8	0.7950 ± 0.0047	0.03316 ± 0.00065	2.6958 ± 0.0092	0.2114 ± 0.0019	2.739 ± 0.010	8.781 ± 0.088	8.781 ± 0.087
BB3-5	112.50	0.6969 ± 0.0036	0.0794 ± 0.0011	2.7504 ± 0.0029	0.2227 ± 0.0012	2.7962 ± 0.0029	9.080 ± 0.052	9.079 ± 0.051
BB3-3.6	121.50	0.8424 ± 0.0064	0.0823 ± 0.0015	2.719 ± 0.015	0.2316 ± 0.0025	2.767 ± 0.015	9.57 ± 0.12	9.57 ± 0.13
BB3-6	124.50	0.7713 ± 0.0040	0.05218 ± 0.00092	2.7477 ± 0.0027	0.2361 ± 0.0014	2.7963 ± 0.0028	9.659 ± 0.058	9.658 ± 0.058
BB3-3.7	130.3	0.7511 ± 0.0085	0.1224 ± 0.0019	2.719 ± 0.027	0.2473 ± 0.0031	2.769 ± 0.028	10.25 ± 0.17	10.25 ± 0.17
BB3-2.3	133.3	0.7367 ± 0.0044	0.872 ± 0.013	2.758 ± 0.010	0.2519 ± 0.0032	2.810 ± 0.010	10.31 ± 0.14	10.29 ± 0.15
BB3-5.1	138.3	0.9239 ± 0.0053	13.45 ± 0.12	2.8869 ± 0.0081	0.2736 ± 0.0020	2.9451 ± 0.0081	10.839 ± 0.064	10.697 ± 0.087
BB3-7	141.00	0.7759 ± 0.0040	15.85 ± 0.15	2.9018 ± 0.0059	0.2867 ± 0.0023	2.9632 ± 0.0060	11.37 ± 0.056	11.17 ± 0.10
BB3-5.2	147.5	0.6155 ± 0.0036	55.87 ± 0.53	2.949 ± 0.025	0.3065 ± 0.0087	3.015 ± 0.025	12.665 ± 0.088	11.78 ± 0.37

LoD = Limit of Detection; BP = before present, i.e. 1950 CE



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Figure 5: Age models for stalagmites BB-1 (blue) and BB-3 (green). Median growth and the 2.5% and 97.5%
 confidence envelopes are shown together with the individual ²³⁰Th/U-ages and 2σ errors. The age models are based on 5000 Monte Carlo simulations and polynomial interpolation between the discrete dates.

388

389 **4.4 μXRF data**

390 In the older stalagmite BB-3, the Sr/Ca ratio varies from 65 and 160, while in BB-1, it ranges from 391 100 to 195 (Fig. 4). In BB-3, Sr/Ca increases slowly until ca. 6.8 ka BP, and reaches then a relatively 392 stable level at ca. 150, before the values fall back to ca. 120 at the top of the stalagmite. The general 393 trend varies only little. In BB-1, the Sr/Ca profile is characterized by a higher degree of variability. 394 Starting at ca. 120, it increases to ca. 160 until ca. 3.3 ka BP, when the values drop to ca. 130. 395 Afterwards, Sr/Ca values increase again until ca. 2 ka BP, before the trend reverses, and Sr/Ca 396 values are lowest values in BB-1 (ca. 100). High-frequency changes are found superimposed on 397 the long-term trends. Sharp multi-decadal increases are found at 4.8, 3.5, 3.2, and 1.5 ka BP, with 398 Sr values reaching 195.

S/Ca ratios show a long term trend opposite to Sr/Ca over the Holocene. In BB-3, a multi-millennial
trend to lower S/Ca values is found (Fig. 4c), from 80 in the early Holocene to 30 at around 6 ka BP.
Few significant deviations are observed, although centennial-scale changes are present. In BB-1,
S/Ca values further decline between 5.6 and 4.5 ka BP, but then stabilize at a level of 30 to 20 (Fig.
403 4a). Only minor fluctuations are observed at centennial scale, with one stronger shift to higher S/Ca
values at 3.3 ka BP and a second possibly at 1.7 ka BP.

405 406

4.5 Statistical analyses of spatio-temporal relationships between proxy records

407 To statistically test for periods of east- and westward Cfb/Dfb boundary migration, we performed a 408 rigorous correlation analysis on the δ^{18} O time series from BB-1 and BU-4 and the DSM record from

409 lake SS1220. As the sampling resolution of the proxy records is different and irregular, and the time 410 axes contain a certain amount of uncertainty, we use a special correlation analysis that considers 411 the chronological uncertainties of each record when calculating correlations. For all palaeoclimate 412 records, we consider a moving window of 500 years, moved in steps of 50 years over the time span 413 4 to 0.5 ka BP. At each window position the COPRA proxy record ensembles (with 5,000 members 414 each) for the two caves are taken and in a first step 5,000 pairs of proxy records are randomly 415 selected, one from each cave. For each pair, we extract the part of the proxy time series that falls 416 within the time window, and estimate the cross-correlation at lag zero between these extracted sub-417 time series using a Gaussian kernel-based correlation approach (NESTool, normalised kernel width 418 h = 0.75) (Rehfeld et al., 2011). This results in an empirical test distribution of 5,000 correlation 419 estimates for that particular window location. Repeating this for all other timings of the window, we 420 obtain an ensemble of 5,000 time series of correlation estimates. The statistical significance of this 421 'observed' correlation distribution is obtained on the basis of a 2-sample Kolmogorov-Smirnov (KS) 422 test, which tests the null hypothesis that the probability distribution of the correlation samples (from 423 the observed data) equals the probability distribution of correlation values of uncorrelated times 424 series of the same statistical properties than the observed data). The random correlation estimates 425 are obtained by repeating the correlation calculation as before, but with the difference that the 426 randomly chosen proxy time series are further uniform-randomly shuffled before estimating the 427 kernel-based cross correlation. The KS test statistic is based on the maximum difference between 428 the two empirical cumulative distribution functions obtained from the two correlation samples and is 429 not necessarily related to an overlap of their interquartile ranges. This test provides a *p*-value, the 430 statistical significance of which is obtained at a confidence level of 1%, after taking into account 431 multiple comparisons by using Holm's method combined with the Dunn-Šidák correction factor.

432

433 **5. Discussion**

434 Stable oxygen and carbon isotope data from speleothem carbonates have been studied for 435 decades, but ambiguities remain with regard to identification of the processes underlying changes 436 in these isotope proxy records (e.g. Gascoyne 1992, McDermott 2004, Mangini et al. 2005, Lachniet 437 2009, Fairchild & Baker 2012). To fully understand individual controls on environmentally sensitive 438 proxies, monitoring is required at the local level. Here, the combination of stable isotope and µXRF 439 elemental data allows detailed insights into seasonally-biased controls on the proxies in 440 speleothems from Bleßberg Cave and the reconstruction of local and (pan-)regional environmental 441 changes. Since the hydrological system in the epikarst acts as a low pass filter, the climatic signal 442 does not reach the cave immediately, thus imposing a lag of unclear but likely annual to multiannual 443 length to speleothem records from Bleßberg Cave

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445 **5.1 Interpretation of geochemical proxies**

447 5.1.1 δ^{13} C, S/Ca and Sr/Ca records as proxies for vegetation, soil development and

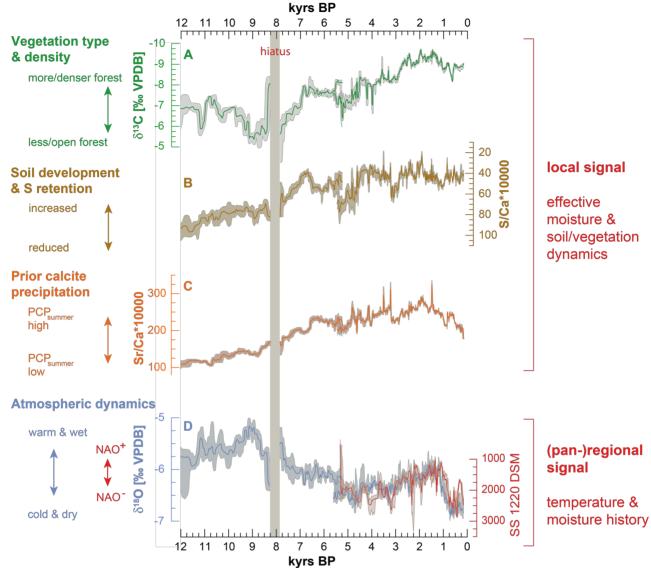
448 infiltration

- Both stalagmite δ^{13} C values and S/Ca ratios (Fig. 6a, b) can serve as proxy for changes in vegetation
- 450 composition and density, soil microbial activity as well as changes in infiltration (Lechleitner et al.
- 451 2017). In temperate locations, these factors contribute most to the carbon budget in the dripwater
- 452 and can be reflected in speleothem δ^{13} C values (Genty & Massault 1999; Genty et al. 2001; Fairchild
- 453 & Baker 2012). Speleothem Sr/Ca constitutes a sensitive proxy for changes in infiltration, with higher
- 454 values being recorded during times of reduced moisture availability (Fairchild & Treble 2009). Since
- 455 Bleßberg Cave had no natural entrance before its discovery during tunnel construction, ventilation-
- 456 induced in-cave CO₂ degassing can be regarded as subordinate process affecting the δ^{13} C values
- 457 of the dissolved inorganic carbon (DIC) and in stalagmites. CO₂ dynamics could potentially be 458 mediated by changing stream volume and/or flow velocity (Troester & White 1984). Unfortunately,
- 459 in Bleßberg Cave, this process cannot be tested as a consequence of the massive concrete injection
- 460 during the tunneling process (ILEK 2011).
- 461 The Bleßberg site is characterized by higher soil microbial and vegetation activity during spring and 462 summer, which, together with higher winterly effective infiltration induces elevated (relative to surface atmosphere) CO₂ levels with low soil δ^{13} C signature. In the epikarst, this δ^{13} C signal is 463 464 adjusted to more positive values, depending on the residence time of the percolating water, and 465 ultimately transferred to the cave (Lechleitner et al. 2017). The stalagmites then record a complex 466 signal of epikarst and in-cave degassing, vegetation composition and density as well as soil 467 microbial activity above the cave (McDermott 2004). This mechanistic explanation is corroborated by the observed low $\delta^{13}C$ values between -7‰ and -10‰ in the time series from Bleßberg Cave. If 468 469 δ^{13} C would solely reflect the composition of the host rock limestone much higher values (+1 to -2‰) 470 would be expected. Thus, we infer that Holocene vegetation and soil development left a 471 recognizable imprint on the δ^{13} C signal. Denser C₃ vegetation (temperate forest) cover or soil 472 development would lead to lower speleothem δ^{13} C, while more open conditions or thin soils would 473 shift δ^{13} C towards higher values (e.g. Scholz et al. 2012). A more open forest and/or thinner soil with 474 reduced microbial activity and reduced production of soil CO2 on the other hand would result in less 475 negative δ^{13} C values (Genty et al. 2001, 2003).
- 476 Further information on soil and vegetation dynamics can be gained from the S/Ca signal (Fig. 6b). 477 Generally, multiple sulfur sources have been identified, including sea spray, dust and volcanic 478 aerosols as well as anthropogenic pollution (Frisia et al. 2005b, Wynn et al. 2010; Wolff et al., 2017). 479 However, the observed long-term trend in S/Ca is most likely not caused by changes in volcanic 480 aerosol loadings, which vary randomly. Similarly, changes in sea spray are unlikely to cause for the 481 observed long-term trend because Bleßberg Cave is a continental site. Anthropogenic influence is 482 similarly unlikely, as a major impact would be expected with industrialization, which is not covered 483 by the youngest section of BB-1.
 - 18

484 Inorganic and organic S is also derived from weathering and organic matter decomposition 485 (Edwards 1998). Sulfur reduction and cycling also depends on redox potential Eh and pH conditions 486 in the soil, although this is generally more significant in water-logged soils (Connell et al. 1968, 487 Husson 2012). The Rendzina above Bleßberg Cave is characterized by high pH, low water holding 488 capacity and strong ventilation. The predominant inorganic form of sulfur in aerobic soils is easy-489 leachable sulfate in the aqueous solution and attached to minerals. This also means that plant 490 uptake of sulfate ions competes with direct leaching to the cave, with better developed soils resulting 491 in improved retention and reduced S-loss through leaching. Sulfur can be retained in the soil by two 492 main mechanisms, i.e. immobilization and adsorption, the relative importance of which can vary 493 (Anderson 1988). The retention times of S can be long, depending on adsorptive capacity and the 494 input of S (Edwards 1998). Importantly, increasing contact time between S and soil results in 495 reduced SO²⁻⁴ leaching due to adsorption and/or transformation of S to less mobile components 496 (Edwards 1998). The negative multi-millennial trend in S/Ca observed in BB-3, which is 497 anticorrelated with Sr/Ca (Fig. 6b, c), suggests that increasing prior calcite precipitation (PCP) (as 498 evidenced by higher Sr/Ca) results in enhanced S retention (lower S in speleothems), due to 499 reduced infiltration and prolonged contact between S components and soil. The decrease of S in 500 BB-3 through the early to mid-Holocene, thus, likely results from soil development, in tandem with 501 diminishing (summertime) infiltration.

502 Interestingly, visual inspection of the S/Ca and Sr/Ca profiles reveals a positive correlation on multi-503 decadal to centennial timescales independent from the long-term trend (Fig. 4), indicating that other 504 factors partake in forcing sulfur mobility on these timescales. Changes in weathering rates, aerosol 505 input or soil activity are all potential candidates for this high-frequency variability. Still, the 506 responsible mechanism must affect both S/Ca and Sr/Ca to cause the observed pattern. Enhanced 507 S immobilization and mineralization and low PCP under colder and wetter conditions, reflected in 508 lower S and Sr content in the stalagmites (Edwards 1998) is a viable explanation. In summary, both 509 proxies are interpreted as indicators of local conditions, which of course are embedded and 510 moderated by the larger-scale climatic milieu.

511 The climate in the study region shows no clear seasonality in precipitation (Fig. 2b), but 512 evapotranspiration, and thus effective infiltration, varies throughout the year, with a maximum during 513 the winter months (Fig. 2d). Infiltration-sensitive proxies should therefore be able to record infiltration 514 changes, with a bias towards the warm season and reduction in spring to autumn infiltration. Dry 515 conditions can lead to (more) open conditions in soil and epikarst, and associated CO₂-degassing 516 from infiltrating water into epikarst air pockets and the cave atmosphere, which in turn allows PCP 517 (Fairchild & McMillan 2007, Breitenbach et al. 2015). PCP can alter the elemental composition of 518 dripwater feeding the stalagmites and increases the Sr concentration in the aqueous solution 519 (Fairchild & Treble 2009).



520 kyrs BP
 521 Figure 6: Bleßberg Cave proxy records and their interpretation. A) δ¹³C values from BB-1 and BB-3 indicate
 522 changes in vegetation density. B) S/Ca records from BB-1 and BB-3 reflect soil development and sulfur retention.
 523 C) Sr/Ca shows long-term but opposite trends to S/Ca. D) δ¹⁸O values from both stalagmites, with lower values
 524 being observed during cold and dry periods, stronger Siberian High influence on the study site. Lower δ¹⁸O values
 525 (blue) are frequently observed during intervals of negative NAO (higher DSM values in lake SS1220, red curve).
 526 The DSM record is based on the updated age-depth model provided in this study.

527

528 The Sr/Ca profile across both stalagmites shows a positive trend throughout the early to mid-529 Holocene (11 ka BP to 6 ka BP, Fig. 6c). This trend is interpreted as resulting from increasing 530 summertime PCP with increasing insolation and a more negative moisture balance in summer (Kalis 531 et al. 2003, Russo & Cubasch 2016). Subsequent to the Holocene Thermal Maximum (Wanner et 532 al. 2011), Sr/Ca ratios remain on a relatively constant level (even if the offset between both 533 stalagmites is taken into account). The observed higher centennial-scale variability after ca. 5.5 ka 534 BP is likely due to the fact that this stalagmite received fracture flow with a faster response to surface 535 conditions. Several positive Sr peaks at 3.5 ka, 3.2 ka and 1.5 ka BP suggest intense, but short-536 lived periods of moisture reduction and intensified PCP. Since about 2 ka BP, Sr/Ca values 537 decrease again, possibly related to weakening summer insolation (Russo & Cubasch 2016) and

improved hydroclimatic conditions with higher effective infiltration. Strong PCP also results in increased degassing of CO₂ and precipitation of calcite (Fairchild & McMillan 2007) and can, thus, also be reflected in less negative δ^{13} C values. However, as outlined above, the δ^{13} C signal is largely unrelated to Sr and governed by vegetation dynamics.

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- 545
- 546

5.1.2.1 Regional factors controlling δ^{18} O in precipitation

5.1.2 Controls on δ^{18} O in precipitation and dripwater

547 Precipitation is delivered to Bleßberg Cave throughout the year and mainly from the North Atlantic, 548 without clear seasonal maximum (Fig. 2) but higher effective infiltration in the winter season (Fig. 549 3). The oxygen isotopic composition in precipitation ($\delta^{18}O_p$) is strongly linked to air temperature and 550 the NAO conditions (Baldini et al. 2008). These relationships are exemplified with δ^{18} O data from meteorological stations Hof, Leipzig and Wasserkuppe/Rhön in Fig. 3a; at all three stations $\delta^{18}O_{0}$ is 551 552 highest in summer, with a total annual amplitude of ca. 4 to 5%. Thus, under higher temperatures 553 and/or a positive NAO, $\delta^{18}O_p$ is expected to increase. The temperature control on $\delta^{18}O_p$ in the region 554 (using Hof, Leipzig and Wasserkuppe/Rhön as reference) is ca. +0.23^{\overlaweverlaw} 555 balancing the negative temperature effect on carbonate precipitation. This is different to Bunker 556 Cave, for which Wackerbarth and coworkers (2010) observed an additional effect of the amount of 557 winter precipitation on precipitation δ^{18} O, which we do not observe for the reference stations used 558 here.

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5.1.2.2 Local factors controlling δ^{18} O in infiltrating water

Local factors that influence δ^{18} O in soil, epikarst and cave include i) evaporation of soil water, ii) 562 563 evaporation of dripwater, iii) CO₂-degassing of dripwater in the cave, iv) temperature, and v) mixing 564 of older and younger water in the epikarst. Evaporation of soil water is more pronounced in the 565 summer season (Fig. 2), but is unlikely to contribute to changes in Bleßberg Cave dripwater on short 566 time scales because May and August samples are not significantly enriched relative to winter month 567 samples (Fig. 3a) and because no secondary evaporation trend, with values following a slope lower 568 than 8, is observed (Fig. 3b). Evaporation of dripwater can be excluded based on the same 569 observations and the fact that in cave relative humidity is stable >98 %. CO₂ degassing from 570 dripwater into the cave air during times of lower drip rates might affect δ^{18} O (Mühlinghaus et al. 2009). However, this effect is very small (i.e. $\Delta\delta^{18}$ O ~0.1‰/1000 sec. at T=10°C, Deininger et al. 571 2012) and would not explain δ^{18} O changes in the range of 0.5 to 2 permil over multi-decadal to 572 centennial time scales. Temperature effects on the dripwater are unlikely because the cave air 573 574 temperature does not vary appreciably ($T_{cave air} = 8.7 \pm 0.1^{\circ}C$). Finally, mixing of infiltration waters of different age is likely the main reason for the relatively stable dripwater δ^{18} O (Fig. 3a) and a very 575

576 narrow range along the GMWL (Fig. 3b). The small offset between the two individual stalagmites,
577 and the dampened variability found in BB-3 are evidence for mixing of waters in the epikarst, which
578 differ for the two stalagmite sites.

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5.1.2.3 δ^{18} O as proxy of atmospheric circulation

The Bleßberg Cave $\delta^{18}O(\delta^{18}O_{spel})$ record (Fig. 6d) integrates precipitation history and the 581 582 corresponding δ^{18} O signal in precipitation (δ^{18} O_p), seasonal changes in infiltration, and temperature 583 in a complex fashion and thus records longer-term pan-regional environmental dynamics. Given the 584 observed seasonal infiltration pattern (Fig. 2), it can be argued that both dripwater and speleothem 585 δ^{18} O values represent a mean signal that is slightly biased towards the winter season. This is similar 586 to other German caves (e.g. Mischel et al. 2015, Wackerbarth et al. 2010). Thermal conditions in 587 the cave are stable and cave air temperature changes over the course of the Holocene are unlikely 588 to have caused the observed 2‰ amplitude in $\delta^{18}O_{spel}$ values. Assuming a (cave) temperature 589 dependence of the isotopic composition during carbonate precipitation of ca. -0.24‰°C⁻¹ (McDermott 2004), counter-balanced by a positive $\delta^{18}O_p/T$ relation of ca. +0.23‰°C⁻¹ would imply 590 591 $\Delta T_{\text{Holocene}} \sim 8^{\circ}$ C to explain the observed 2‰, which seems unrealistically high for the last 9-10 ka BP. 592 Several lines of evidence suggest an important influence of atmospheric circulation, moisture source dynamics and $\delta^{18}O_p$ on $\delta^{18}O_{spel}$ in Bleßberg Cave. A significant positive correlation has been found 593 594 between the NAO and $\delta^{18}O_p$ over Central Europe (Hurrell 1995, McDermott et al. 2011, Baldini et 595 al. 2008, Comas-Bru et al. 2016). The strong positive impact of the winter NAO on $\delta^{18}O_p$ is caused 596 by more frequent inflow of cold, isotopically depleted precipitation during negative NAO phases 597 (Baldini et al. 2008). A tentative link between NAO and Bleßberg $\delta^{18}O_{spel}$ is found in the visual 598 similarity between BB-1 δ^{18} O and the updated detrital silicate mineral (DSM) record from lake 599 SS1220 (DSM_{SS1220} = $\Sigma_{ISr. Zn. Til}$, Olsen et al. 2012, Fig. 6d). For this study we updated the SS1220 600 chronology by using the Intcal13 calibration curve (Reimer et al. 2013) to re-calibrate the ¹⁴C 601 measurements originally reported in Olsen et al. (2012). We then sampled 5,000 stratigraphically 602 ordered age models from the multimodal re-calibrated ages, which resulted in a 5,000 member 603 DSM_{SS1220} proxy record ensemble. Warmer conditions with increased meridional flow of warm 604 maritime air over SW Greenland are normally associated with negative NAO, while positive NAO 605 conditions are characterized by cold, dry air masses over southern Greenland (Olsen et al. 2012). 606 In Central Europe, negative (positive) NAO conditions are linked to colder (warmer) and drier 607 (wetter) air under increased (decreased) Siberian High influence, reflected in more negative (positive) $\delta^{18}O_p$ and $\delta^{18}O_{spel}$ values (Baldini et al. 2008). 608

Thus, climatic conditions in Central Europe are not solely determined by maritime (NAO) influence from the Atlantic, but also by the SH. The winterly SH is another major player that influences atmospheric circulation, precipitation, and temperature (Berry & Chorley 2010, Tubi & Dayan 2012).

612 The interaction between the NAO and the SH results from complex tropospheric and stratospheric

613 linkages (Cohen et al. 2001, Cohen et al. 2014; Ambaum & Hoskins 2002). Both systems are 614 intimately linked with the Arctic Oscillation (AO), which has been suggested to lag NAO dynamics 615 by a few days (Ambaum & Hoskins 2002). A positive NAO leads to a stronger stratospheric vortex 616 and a deepened low pressure cell over the North Pole, and effectively a positive AO. This in turn 617 results in stronger Westerlies and fewer and smaller meanders of the zonal jet stream (Cohen et al. 618 2014). Under such conditions, the strong Westerlies transport more and warmer moisture from the 619 N Atlantic towards Europe and into Eurasia and the SH has rarely the chance to reach Central 620 Europe. In a negative NAO/AO mode on the other hand the Arctic low pressure system is weaker 621 and the zonal jet is characterized by increased meandering which allows frequent atmospheric 622 blocking and intrusion of cold air from the north/northeast. The Westerlies are weaker and frequently 623 redirected southward, thus delivering less moisture to Germany, while the intruding Arctic air is very 624 cold. Thus, the influence of the SH increases under negative NAO/AO conditions. Inflow of north-625 easterly continental air under strong SH conditions replaces warm maritime air from the west. The 626 increasing SH influence with distance from the Atlantic coast is directly expressed in the change 627 from a maritime Cfb climate to a continental Dfb climate at ca. 10°E (Peel et al. 2007). Thus, $\delta^{18}O_{spel}$ 628 from Bleßberg Cave is sensitive to shifts of the boundary between the two climate zones.

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5.2 Local and pan-regional environmental changes in Central Europe

The multi-proxy datasets from BB-1 and BB-3 allow detailed insights into both local and pan-regional environmental conditions throughout the Holocene. Below, we first discuss how the local environment evolved after the last deglaciation. Then, we compare the climatic conditions in Thuringia to those in Europe and the North Atlantic realm and discuss the role of a shifting Cfb-Dfb climate boundary and jet stream as explanation for the differences between sites.

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5.2.1 Local environmental changes

638 We use the δ^{13} C, S/Ca, and Sr/Ca time series to reconstruct local vegetation, soil and infiltration dynamics through the Holocene. High δ^{13} C values between the Late Glacial and the onset of the 639 640 Holocene (Fig. 6a) point to an open, and possibly grassland vegetation, typical for periglacial 641 conditions (Hahne 1991, Bebermeier et al. 2018). After a short-lived excursion to drier and/or colder 642 conditions shortly before 11 ka BP, the δ^{13} C record trended towards higher values for the next ca. 643 2400 years, potentially indicative of an early coniferous forest (Hahne 1991; Bebermeier et al. 2018). 644 Decreasing S/Ca and low, slowly increasing Sr/Ca values between 11 and 9 ka BP indicate 645 cumulative soil development and improved sulfur retention with sufficient effective infiltration to 646 minimize summerly PCP (Figs. 6b, c). The Preboreal to early Holocene development is comparable 647 to that in the southern Harz Mountains, ca. 100 km to the north of our study site (Bebermeier et al. 648 2018).

649 Elevated δ^{13} C values after ca. 9 ka BP (the onset of the Boreal, sensu Hahne 1991) point to a return 650 to a more open forest, or a change in vegetation composition. Pollen profiles from the Rhön (Hahne 651 1991), ca. 40 km west of Bleßberg Cave, and Eichsfeld (Bebermeier et al. 2018) suggest 652 replacement of pine forest by Corylus communities which prefer warmer conditions. A change to 653 more open forest is not mirrored in the S/Ca and Sr/Ca ratios, suggesting that it was not 654 accompanied by increased (summer) drought and related soil or infiltration changes. Less negative 655 δ^{18} O values at ca. 9 ka BP also suggest relatively warm/wet conditions (Fig. 6d). A drastic decrease in both δ^{13} C and δ^{18} O between ca. 8.7 ka BP and 8.4 ka BP, concurrent with a slight increase in 656 657 Sr/Ca might be explained by a post-glacial afforestation under drier summer conditions and 658 intensified PCP. A scenario where the vegetation composition changed from deciduous to 659 coniferous forest is less likely because one would expect a shift to more positive δ^{13} C values 660 (Amiotte-suchet et al. 2007), opposite to the observed trend. A concurrent trend towards more 661 negative δ^{18} O values suggests cooling and drying and/or increased winter precipitation prior to the 662 8.2 ka event.

663 The hiatus found in BB-3 covers ca. 400 years, starting 8.26 and ending 7.85 ka BP, with a minimum 664 and maximum length of 64 and 710 years, respectively, when considering the chronological 665 uncertainties in the BB-3 record. The hiatus corresponds closely with the 8.2 ka event (Mayewski et 666 al. 2004, McDermott 2004) and can only be explained by complete absence of dripwater, caused 667 either by flooding or infilling of the cave passage, re-routing of the water, prolonged drought, or 668 permafrost conditions (Vaks et al. 2010, 2013; Lechleitner et al. 2017). A return of permafrost during 669 the 8.2 ka event seems the most likely scenario because we have no evidence in support of infilling 670 of the cave with sediment or water, or drought with zero effective infiltration, which should be 671 reflected in increased Sr/Ca (increased PCP). The development of permafrost (i.e. at least two 672 consecutive years with ground temperature <0°C, Harris et al. 2009) during the 8.2 ka event would 673 require a decrease in mean annual air temperature of >6°C with respect to modern conditions, which 674 at first glance seems very high. A drastic, although not reaching 6°C, decrease in temperature during 675 the 8.2 ka event has been found in western and northern Europe (Seppä et al. 2009, Vincent et al. 676 2011). However, given the complex interaction between geomorphology, geology, precipitation, air 677 and ground temperatures (Harris et al. 2009) local permafrost development in hilly Thuringia seems 678 plausible. More detailed work is needed to reconstruct the climatic conditions surrounding the 8.2 679 ka event.

After ca. 7.8 ka BP, conditions improved and stalagmite growth commenced again, with rapid soil development and denser forest vegetation, as seen in lower S/Ca ratios and δ^{13} C values. This climatic amelioration was accompanied by slightly increased PCP, likely due to warmer summers (Fig. 6c). After ca. 6.7 ka BP, soil development and/or sulfur retention level out, with only two excursions around 5.5 and 3 ka BP, when conditions seem to have been comparatively drier. Vegetation, however, was likely more dynamic, as reflected in noticeable variations in δ^{13} C between

5.5 and 3.5 ka BP (Fig. 6a), sometimes together with increased PCP. A trend to more negative δ^{13} C values after 3 ka BP suggests increased forest cover, which was reversed only in the last ca. 1500 years, possibly related to deforestation activities of Bronze Age settlers (Bebermeier et al. 2018).

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5.2.2 Pan-regional climatic changes

691 Today, Central Europe is governed by maritime Cfb climate, while eastern Europe is characterized 692 by continental Dfb climate (Kottek et al. 2006, Peel et al. 2007). The meteorological border between 693 these two climatic provinces is located over Germany (Fig. 1) and has also been identified as a 694 significant shift in the orientation of precipitation isochron pattern (Rheinwalt et al. 2016). 695 Importantly, over time this climatic boundary is not stationary and shifts, depending on climatic 696 conditions (Kottek et al. 2006). It should, in principle, be possible to detect past changes of the 697 geographical position of this climatic divide if sufficient well dated and resolved proxy time series 698 were available from locations distributed across Europe. Links between reconstructions that are 699 located on each side of the boundary and influenced by factors that govern climate in the respective 700 zone can give insights how this boundary shifted in space and time. Using only one additional record 701 from the opposing climate zone would not suffice however, because one would find either positive, 702 negative or zero correlation. While the latter two scenarios could be interpreted to indicate a climate 703 boundary between the two sites, the first scenario could only suggest a common forcing factor, but 704 would not clarify whether the climate boundary is located east or west of both sites. Thus, at least 705 one third record is needed to identify the position of the boundary relative to the sites, and the 706 controls relevant at each location.

707 To estimate the geographical position of the Cfb-Dfb boundary, we use two time series from the 708 North Atlantic realm in addition to the Bleßberg Cave BB-1 record (see Figs. 1 and 7). The first of 709 these records is the δ^{18} O time series from stalagmite BU-4 ($\delta^{18}O_{BU4}$) from Bunker Cave in western 710 Germany (Fohlmeister et al. 2012). The $\delta^{18}O_{BU4}$ record has been interpreted as reflecting a complex 711 signal of temperature and moisture supply. While the δ^{18} O signal in Bunker Cave reflects multiple 712 processes, less negative δ^{18} O values indicate colder and/or drier conditions, while lower values 713 suggest warmer and/or wetter climate. Multi-annual variations in surface temperature and 714 precipitation amount have been linked to NAO dynamics (Wackerbarth et al. 2010, Riechelmann et 715 al. 2017).

The second reconstruction used to test the concept of a shifting climate boundary is a multi-element proxy record of DSM input from lake SS1220 (DSM_{SS1220}) in southwestern Greenland (Olsen et al. 2012). The DSM record is the sum of Sr, Zn and Ti counts observed in the lake sediment profile, in a similar fashion as proposed by Saarni et al. (2016), and is used here to infer NAO conditions. Higher DSM values indicate increased sediment transport associated with snowmelt and runoff into the lake. In southwestern Greenland increased runoff and sediment supply are expected in mild and/or short winters with longer thaw periods and prolonged runoff; conditions that have been associated with negative NAO phases (Olsen et al. 2012). High DSM values are thus interpreted as
 representative for negative NAO phases (see Fig. 6d).

These time series and the Bleßberg δ^{18} O record from BB-1 ($\delta^{18}O_{BB1}$) reflect relevant circulation 725 726 features with sufficient resolution and chronological control for the last ca. 4,000 years to allow a 727 first estimation of past boundary dynamics. To establish the Cfb-Dfb boundary, we estimate 728 correlations between the three records, which we then link with the information on the atmospheric 729 circulation patterns (NAO index and SH strength) assigned to the proxy records to estimate the position of the climate boundary. It must be kept in mind that the interpretation of δ^{18} O from Bleßberg 730 731 and Bunker caves is opposite in sign, with lower δ^{18} O values in Bleßberg Cave indicating colder and 732 drier conditions under a more pronounced SH and negative NAO influence, while reflecting wetter 733 and warmer (maritime) conditions above Bunker Cave. This relationship could change however due 734 to changes in seasonal precipitation pattern and resulting slopes of the $\delta^{18}O_{p}$ -T relationship. The 735 obtained correlation patterns allow the identification of the following Cfb-Dfb boundary position 736 scenarios (Fig. 7):

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Scenario 1 Cfb-Dfb boundary east of Bleßberg Cave

739 The first scenario is similar to modern conditions (Kottek et al. 2006), but with the Cfb-Dfb boundary 740 located shifted further east of Bleßberg Cave. The maritime influence reaches towards eastern 741 Central Europe while the Siberian High influence on western Europe is relatively weak. Under such 742 conditions, Bleßberg and Bunker caves would record the same maritime climate and a common 743 forcing with strongly positive NAO influence. The interpretation of $\delta^{18}O_{BB1}$ would be reversed, 744 because continentality, and with it the slope of the $\delta^{18}O_{p}$ -T relationship would be lowered (Bowen 745 2008). Consequently, Bleßberg Cave would react much like Bunker Cave, with a positive correlation between the two δ^{18} O records, while DSM_{SS1220} values should be lowered (Fig. 7). A boundary east 746 747 of Bleßberg Cave thus would result in a positive correlation between all three records. Modern 748 observational data show a stronger Atlantic influence on Central Europe and a significant positive 749 link between NAO index and precipitation δ^{18} O at Wasserkuppe/Rhön near Bleßberg Cave (Baldini 750 et al. 2008). Such conditions occur when the zonal jet is stronger and less meandering (Cohen et 751 al. 2014). Unfortunately, since the Bleßberg Cave record does not cover the last few hundred years, 752 we cannot test the inferred links with meteorological records.

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Scenario 2 Cfb-Dfb boundary between Bleßberg Cave and Bunker Cave

A second scenario would see the Cfb-Dfb boundary located between the two German caves, with Bunker Cave being governed by maritime climate and Bleßberg Cave by continental climate conditions. Under these conditions, $\delta^{18}O_{BB1}$ should decrease (due to increased continentality and resultant colder winters), while the $\delta^{18}O_{BU4}$ and DSM_{SS1220} records might remain unchanged or increase compared to scenario 1, with the cave records reflecting atmospheric dynamics related to the climatic regime on either side of the Cfb-Dfb boundary. As a result, negative correlation between $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ should be found, while the Greenland DSM_{SS1220} record would be expected to show negative to nil correlation to $\delta^{18}O_{BB1}$, but remain positively linked to $\delta^{18}O_{BU4}$, as long as the Westerlies exert some influence on Bunker Cave (Fig. 7). Scenario 2 would be characterized by frequent SH influence on Central Europe, a consistently changing NAO and stronger meandering of the jet stream, but relatively mild conditions.

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Scenario 3 Cfb-Dfb boundary east of Bleßberg Cave

768 A third scenario might occur where the climate boundary is positioned east of Bleßberg Cave, similar 769 to scenario 1, but with more stable climate conditions. In scenario 3, both speleothem δ^{18} O records 770 would be expected to be influenced by maritime climate, with rare incursions of cold Arctic air during winter and strongly positive NAO (Fig. 7b). The interpretation of $\delta^{18}O_{BB1}$ would be reversed (as in 771 772 scenario 1), because of strongly diminished continentality. Bleßberg and Bunker cave would be 773 more responsive to oceanic conditions, reflected in a positive correlation between both δ^{18} O records. A negative correlation between $\delta^{18}O_{spel}$ and DSM_{SS1220} would result, with lower $\delta^{18}O_{spel}$ and 774 775 increased DSM values reflecting negative NAO dynamics and vice versa.

776 This scenario differs from scenario 1 in that it includes a poleward displacement of the jet stream in 777 a generally warmer atmosphere (Archer and Caldeira 2008, Woolings and Blackburn 2012), so that 778 the maximum moisture transport would be redirected towards Scandinavia. Concurrently, the 779 Siberian High influence on Central Europe would be diminished, which in turn would lead to more 780 stable conditions, albeit with decreased winter moisture and higher PCP at Bleßberg Cave (Fig. 6). 781 An intensified and poleward jet and positive NAO result from a large pressure difference between a 782 cold Arctic and very warm mid-latitudes (Archer and Caldeira 2008). This interpretation would explain higher $\delta^{18}O_{BB1}$ values (Fig. 7b) as response to drier winter conditions in Thuringia, and lower 783 784 $\delta^{18}O_{BU4}$ values, due to warmer and/or wetter conditions at Bunker Cave. Higher winter precipitation 785 would be expected in Northern Europe.

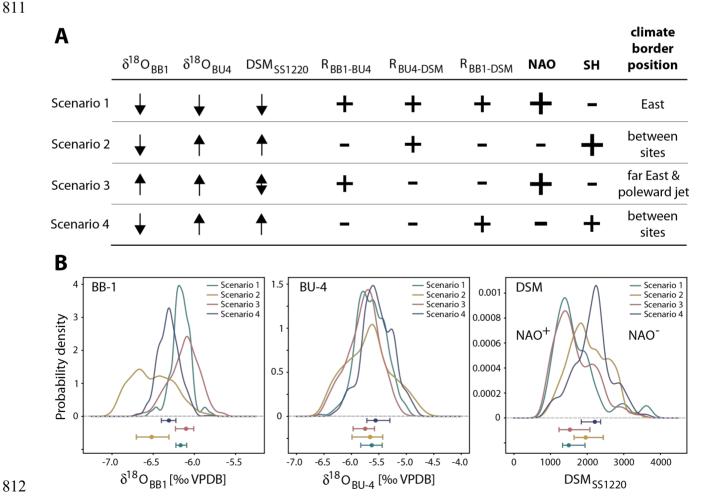
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Scenario 4 Cfb-Dfb boundary between both caves

788 A fourth scenario has been found in the correlation analyses whereby the $\delta^{18}O_{BU4}$ time series shows an increasing trend, while the $\delta^{18}O_{BB1}$ record shows an opposite trend (and vice versa), i.e. both 789 790 cave records are anticorrelated. The DSM record is positively correlated with the $\delta^{18}O_{BB1}$ time series. The $\delta^{18}O_{BB1}$ values are slightly more negative compared to scenario 1 (Fig. 7b) while the $\delta^{18}O_{BB1}$ 791 792 and DSM_{SS1220} values are higher. These observations suggest a moderate to strong influence of the 793 Siberian High on Bleßberg Cave and a pronounced negative NAO. The lower $\delta^{18}O_{BB1}$ values and 794 the positive correlation of $\delta^{18}O_{BB1}$ with DSM_{SS1220} suggest winter conditions with frequent SH influence on Bleßberg Cave and negative NAO (i.e. cold winters bringing ¹⁸O-depleted precipitation 795 796 sensu Baldini et al. 2008). We hypothesize that the climatic boundary is located between both caves. 797

798 hypothetical Scenario 5 Cfb-Dfb boundary west of Bunker Cave •

799 Theoretically, a fifth scenario could occur if very cold conditions would lead to a westward retreat of 800 the Cfb-Dfb boundary. Such conditions might have prevailed in periods with southward displaced 801 jet stream, e.g. the late glacial before northern hemisphere warming would allow the maritime 802 climate to reach Central Europe. This scenario is not observed in the discussed late Holocene 803 records. At Bunker Cave increased continentality would result in a positive slope of the $\delta^{18}O_{0}$ -T relationship and a reversed interpretation of $\delta^{18}O_{BU4}$ (higher $\delta^{18}O_{BU4}$ indicating warmer conditions). 804 805 Since continental conditions would prevail, a positive correlation between the two cave records is 806 expected. If the jet stream would be located far west of Bunker Cave, the latter would loose 807 sensitivity to the maritime climate. This would be reflected in a reduced or lacking correlation (in 808 either direction) between the cave proxies and the DSM_{SS1220} record, which would be expected to 809 still be governed by North Atlantic climate variability and indicative of the NAO state. Western 810 Europe would be expected to be more frequently affected by the winterly Siberian High.





813 Figure 7: A) Interpretation of proxy records, their correlations (R), and inferred state of the North Atlantic 814 Oscillation and the Siberian High in the four scenarios of the position of the Cfb-Dfb boundary over Europe. The 815 size of the signs indicates the relative strength of the parameter (i.e. the correlations are weaker). B) Probability 816 densities for the three records for each of the four observed scenarios.

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The correlations between the three sites Bleßberg Cave, Bunker Cave and Greenland lake SS1220 (Fig. 8d-f) allow us to estimate changes in the longitudinal position of the Cfb-Dfb boundary and the jet stream over the last ca. 4,000 years. NAO and SH pattern are inferred from the DSM_{SS1220} profile and the $\delta^{18}O_{BB1}$ time series, respectively. The correlations, the NAO index, relative strength of the Siberian High, and the inferred position of the climate boundary are summarized in Fig. 8g.

Prior to about 3.9 ka BP, concurrent positive correlation between $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ and negative correlation between the cave records and DSM_{SS1220}, suggest that the Cfb-Dfb boundary was located east of Bleßberg Cave (scenario 3) and that Central Europe was under influence of maritime climate, likely with a moderate winterly SH influence at Bleßberg Cave. With a climate boundary shifted east the influence of the NAO on Central Europe was likely increased. Relatively high DSM values suggest negative NAO conditions, which would go in hand with stronger meandering of the zonal jet and cold air inflow during winter.

After 3.9 ka BP we find a reversed $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ correlation (scenario 4), which we tentatively 830 831 interpret as a ca. 200-year long transition phase from scenario 3 to scenario 2. Insufficient data points in the DSM_{SS1220} record prevent computing the correlations with $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ between 832 833 3.7 and 3.3 ka BP (Fig. 8e, f) and statements regarding the NAO impact on European climate must 834 remain vague. Intermediate DSM values suggests variable NAO conditions. The negative correlation between $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ can be used however to infer a likely position for the Cfb-835 836 Dfb boundary between the two caves (scenario 2). This interval was followed by a shift back to 837 scenario 4. This period with a negative NAO and strong SH lasted for only about a century (~3.3-838 3.2 ka BP) and was then succeeded by a 500-year long period of the similar scenario 2, with negative correlations between $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ and $\delta^{18}O_{BB1}$ and DSM_{SS1220}. At this time the Cfb-839 840 Dfb boundary was located between the two caves; both the SH and the NAO exerted their influence 841 on their respective climate zones. A strong SH at that time has also been identified by Mayewski et 842 al. (2004) in the GISP2 ice core potassium record and frequent negative NAO conditions have been 843 noted between ca. 3.1 ka and 2.5 ka BP by Olsen et al. (2012).

Around 2.7 ka BP another transition occurred, found as a ca. 300-year interval with unclear but likely eastward boundary shift. Considering the available evidence, it is likely that this time was characterized by higher climate variability, with continued boundary shifts and variable SH and NAO strength. The low DSM_{SS1220} values suggest that the NAO shifted to a positive phase at the time. At 2.4 ka BP a prominent, 500-year long interval of near-modern climate set in, with the Cfb-Dfb boundary located between both caves. The SH had influenced winter conditions concurrent with a positive NAO, as indicated by the DSM values.

This maritime interplay was rapidly (within ca. 100 years) replaced by a more maritime climate that lasted from ~1.9 ka BP to ~1.1 ka BP (scenario 3). This period was characterized by a concurrent poleward displacement of the jet stream and more stable conditions in Central Europe. Winters at

- Bleßberg Cave were most likely drier due to this northward shift of the Westerlies which would bring increased winterly moisture to Scandinavia and Bunker Cave, while the Thuringian summers were seemingly warm and dry (as indicated by increased PCP, Fig. 6). The DSM_{SS1220} record suggests a positive NAO. Saarni et al. (2016) noted increased snow accumulation during positive NAO,
- 858 consistent with our interpretation.
- 859 A last change is recorded in our time series at around 1.1 ka BP, when the climate boundary shifted 860 westward, to remain between the two caves until at least 500 years ago when our Bleßberg record 861 ends. This last period was characterized by a strong SH and concurrently variable and frequently 862 negative NAO, which together left winterly Germany colder. The inferred strong SH is supported by 863 the GISP2 K record (Mayewski et al. 2004). The inferred negative NAO at the time (covering the 864 Medieval Climate Anomaly) is also supported by a tree ring-based reconstructions of a multi-865 centennial mega-drought in Scandinavia (Helama et al. 2009) and humid and mild summers in 866 Central Europe (Büntgen et al. 2011).
- 867 Above Bleßberg Cave summertime PCP diminished slowly, indicating increasing summer 868 infiltration. In western Germany (at Bunker Cave) higher climate variability can be inferred from the 869 δ^{18} O record (Fig. 8b), which can be understood if we assume less severe influence of the SH on 870 least conditions at its forward or limit
- 870 local conditions at its far western limit.
- 871 Because the last 500 years are not covered by the Bleßberg stalagmite, we cannot connect our
- 872 reconstruction with historical and meteorological data. The Cfb-Dfb climate boundary continued to
- 873 migrate eastward, is currently located near the eastern border of Poland, and expected to shift even
- 874 further east under global warming conditions (Kottek et al. 2006).
- In summary, the comparison of multiple sites allows the identification of shifts in the mean position of the Cfb-Dfb climate boundary over the last ca. 4,000 years, depending on the relative strength and dynamics of the Siberian High and the Westerlies and the position of the jet stream. To refine this reconstruction, a denser network of records would be required, possibly by exploiting the new Speleothem Isotope Synthesis and Analysis database (Atsawawaranunt et al. 2018). With improved spatio-temporal coverage it might also be possible to deduce directionality of forcings and determination of the speed of such climate transitions.

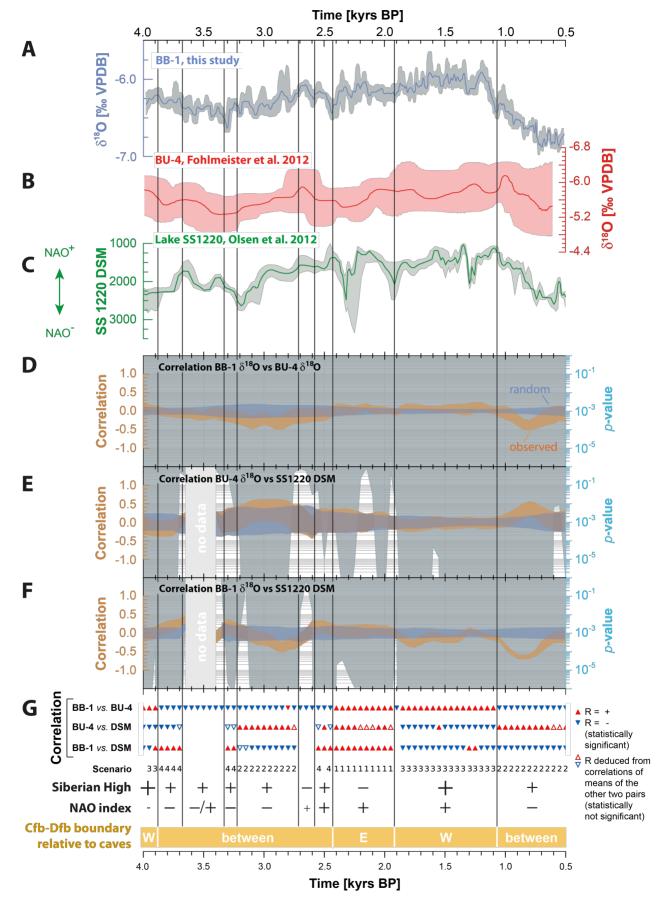




Figure 8: Proxy records from Bleßberg ($\delta^{18}O_{BB1}$, A), Bunker Cave ($\delta^{18}O_{BU4}$, B), and Greenland (DSM_{SS1220}, C)

884 covering the last 4,000 years are correlated to highlight non-stationary relationships. Solid lines in A-C denote the

885 median proxy value and shades represent the inter-quartile ranges as obtained from COPRA proxy record 886 ensembles composed of 5,000 age-depth simulations each. The DSM_{SS1220} profile is based on an updated 887 chronology in which the IntCal13 calibration curve was used to calibrate the radiocarbon ages. Randomly chosen 888 pairs proxy records from the COPRA ensembles are used to estimated the correlations (orange bands, D-F) 889 between each pair of records using the NESTool methodology that allows estimation of correlation for irregularly 890 sampled time series. The relationship between the records is compared to that obtained from an ensemble of 891 pairs of randomized time series (blue bands), and this is quantified with the *p*-value of a 2-sample Kolmogorov-892 Smirnov test (light blue shaded backgrounds, see section 4.5 for details). (G) Correlation summary and inferred 893 Siberian High strength and NAO index, as well as estimates of the position of the Cfb-Dfb climate boundary. Solid 894 triangles indicate statistically significant correlations, while empty triangles refer to statistically not significant 895 values that are deduced from the (significant) correlations between the median values of the other pairs. 896 Insufficient data points in the DSM_{SS1220} record prevent computing the correlations with $\delta^{18}O_{BB1}$ and $\delta^{18}O_{BU4}$ 897 between 3.7 and 3.3 ka BP.

898

899 6. Conclusions

900 Two U-series dated stalagmites (BB-1 and BB-3) from Bleßberg Cave, Thuringia, are used to 901 reconstruct local and (pan-)regional environmental conditions. The time series from stalagmites BB-902 1 and BB-3 cover the entire Holocene with the exception of a ca. 400-year long hiatus at 8.2 ka. All 903 proxies discussed above respond to local processes, which themselves are governed by regional 904 atmospheric dynamics. In this way, they all inform on individual aspects of environmental conditions 905 active at the time of speleothem deposition. δ^{13} C, S/Ca and Sr/Ca inform us about local 906 environmental changes, including vegetation and soil changes above the cave, as well as prior 907 calcite precipitation linked to effective infiltration through the course of the Holocene. Stalagmite 908 δ^{18} O is interpreted as recorder of changes in moisture source dynamics and temperature and thus 909 as (pan-)regional signal. We combine the Bleßberg Cave δ^{18} O record with distal proxy 910 reconstructions to infer changes in the relative importance of marine and continental conditions on 911 Central European environment.

For the last ca. 4,000 years we compared the BB-1 δ^{18} O record with the δ^{18} O profile from stalagmite 912 913 BU-4 from Bunker Cave in western Germany and a detrital silicate material record from lake SS1220 914 (SW Greenland) and estimate the changes in the Cfb-Dfb climate boundary, as well as the strength 915 of the Siberian High and dynamics of the North Atlantic Oscillation. We find a complex temporal 916 pattern with repeated multi-centennial scale E-W shifts of the Cfb-Dfb boundary. The local 917 environment at Bleßberg Cave reacts to an intensified Siberian High mainly with reduced soil 918 development and increased prior calcite precipitation during the warm season, due to reduced 919 winterly precipitation.

920 The interpretative value of the presented estimate of the dynamics of the climate boundary is
921 currently limited by the low number of well-dated and highly resolved reconstructions from Europe.
922 Using this approach to integrate available datasets from additional archives, including speleothem,

923 tree-ring and pollen data, will help refine and possibly quantify our insights in Holocene climate and 924 vegetation dynamics. We are confident that with improved spatial coverage the history of climate 925 boundaries will become recognizable at much finer scale. Discrepancies between spatially 926 distributed reconstructions might indicate competing environmental forcings and can be exploited 927 to resolve climatic regimes in different climate zones.

928

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940

941 Data availability

942 The datasets presented here will be available on the website of the corresponding author (SFMB)

and on public repository, including https://www.pangaea.de. Data can also be requested bycontacting SFMB directly.

945

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