# Characterization and correction of evaporative artefacts in speleothem fluid inclusion isotope analyses as applied to a stalagmite from Borneo.

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#### Abstract

Fluid inclusion water isotope measurements in speleothems have great potential for paleoclimate studies, as they can be used to provide reconstructions of precipitation dynamics and land temperature. Several previous observations, however, suggest that inclusion waters do not always reflect the isotopic composition of surface precipitation. In such cases, dripwaters are thought to be modified by evaporation in the cave environment that result in more positive d2H and d18O values and shallow d2H/d18O slopes. Although evaporation can occur in cave systems, water can also be lost to evaporation during analysis but before water extraction. Here, we examine the likelihood of this possibility with a stalagmite from Borneo. We demonstrate that many samples loose water, and that water loss is controlled by the type and size of inclusions. With multiple replicate measurements of coeval samples, we calculate an evaporative d2H/d18O slope of  $1\pm0.6$  (2SE). This value is consistent with model predictions of evaporative fractionation at high analytical temperature at low humidity. Finally, we provide a robust and physically based correction method. We find that fluid–calcite d18O paleotemperatures calculated with corrected d18O data show excellent agreement with recent microthermometry temperature estimates for Borneo during the last deglaciation, suggesting minimal variations in stalagmite d18O disequilibrium over time. Similarly, corrected fluid inclusion d18O and d2H values follow the expected hydroclimate response of Borneo to periods of reduced Atlantic Ocean meridional overturning circulation. Our results suggest that careful petrographic examination and multiple replicate measurements are necessary for reliable paleoclimate reconstructions with speleothem fluid inclusion water isotopes.

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# Characterization and correction of evaporative artefacts in speleothem fluid inclusion isotope analyses as applied to a stalagmite from Borneo.

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- 13

## 14 Abstract

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16 Fluid inclusion water isotope measurements in speleothems have great potential for paleoclimate studies, as they can be used to provide reconstructions of precipitation dynamics 17 18 and land temperature. Several previous observations, however, suggest that inclusion waters 19 do not always reflect the isotopic composition of surface precipitation. In such cases, 20 dripwaters are thought to be modified by evaporation in the cave environment that result in more positive  $\delta^2 H$  and  $\delta^{18} O$  values and shallow  $\delta^2 H / \delta^{18} O$  slopes. Although evaporation can 21 22 occur in cave systems, water can also be lost to evaporation during analysis but before water extraction. Here, we examine the likelihood of this possibility with a stalagmite from Borneo. 23 24 We demonstrate that many samples loose water, and that water loss is controlled by the type 25 and size of inclusions. With multiple replicate measurements of coeval samples, we calculate an evaporative  $\delta^2 H/\delta^{18}O$  slope of 1±0.6 (2SE). This value is consistent with model predictions of 26 27 evaporative fractionation at high analytical temperature at low humidity. Finally, we provide a 28 robust and physically based correction method. We find that fluid–calcite  $\delta^{18}$ O paleotemperatures calculated with corrected  $\delta^{18}$ O data show excellent agreement with recent 29 microthermometry temperature estimates for Borneo during the last deglaciation, suggesting 30 31 minimal variations in stalagmite  $\delta^{18}$ O disequilibrium over time. Similarly, corrected fluid 32 inclusion  $\delta^{18}$ O and  $\delta^{2}$ H values follow the expected hydroclimate response of Borneo to periods 33 of reduced Atlantic Ocean meridional overturning circulation. Our results suggest that careful 34 petrographic examination and multiple replicate measurements are necessary for reliable 35 paleoclimate reconstructions with speleothem fluid inclusion water isotopes. 36 37 38 1.0 Introduction 39 40 The majority of stalagmite-derived paleoclimate reconstructions have traditionally been 41 based on the oxygen isotope composition of carbonate (e.g., Fleitmann et al., 2003; Wang et 42 al., 2001). More recently, thanks to recent technological improvements (e.g., Affolter et al.,

43 2014; Dassié et al., 2018; de Graaf et al., 2020), research has increasingly focused on

44 measurements of fluid inclusion water isotopes. Fluid inclusions in speleothems preserve relics of ancient dripwater. Measurements of  $\delta^2$ H and  $\delta^{18}$ O in fluid inclusion water offer unique 45 advantages relative to carbonate  $\delta^{18}$ O values alone. They provide a more direct constraint on 46 rainwater  $\delta^{18}$ O than carbonate  $\delta^{18}$ O. Thus, they can be used to reconstruct changes in 47 precipitation dynamics such as rainfall amounts (e.g., Arienzo et al., 2015; van Breukelen et al., 48 49 2008; Millo, 2017) and in the sources of moisture (e.g., Matthews et al., 2021; Rogerson et al., 2019), especially when supported by suitable moisture transport information (e.g., Baker et al., 50 51 2015). They also allow disentangling the respective contributions of temperature and dripwater  $\delta^{18}$ O variability on stalagmite carbonate  $\delta^{18}$ O (e.g., Arienzo et al., 2015; Meckler et al., 2015, 52 53 2021; Wortham et al., 2022), yielding temperature estimates from the difference of  $\delta^{18}$ O in 54 inclusion water and the surrounding calcite . Additionally, in settings where temperature is the 55 main control on rainfall isotope variability, fluid inclusion measurements have also been used directly to estimate air temperature at the site of precipitation, assuming that present-day 56 57 relationships hold true for the past (Affolter et al., 2019).

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59 Fluid inclusions often yield meaningful isotope values showing that the isotopic composition of the inclusion water is well preserved. Fluid inclusion isotope analyses thus contribute 60 significantly to our understanding of speleothem paleoclimate records. However, observations 61 62 from some stalagmites suggest that recovery of the original isotope signal of precipitation is not always possible. For instance, recently published fluid inclusion isotope data from five 63 stalagmites show large positive enrichments in  $\delta^2 H$  and  $\delta^{18} O$  values that in some cases plot far 64 65 to the right of local meteoric water lines (LMWLs) (Affolter et al., 2019; Matthews et al., 2021; Nehme et al., 2020; Warken et al., 2022; Wortham et al., 2022). These data are usually 66 67 obtained from samples with relatively low water yields, and follow typical evaporation lines in a  $\delta^2$ H/ $\delta^{18}$ O diagram. Such observations suggest that fluid inclusion waters can be modified to an 68 69 extent that they no longer reflect the isotopic composition of surface precipitation. Modification of fluid inclusion water  $\delta^{18}$ O through carbonate-water isotope exchange has been 70 proposed (Demény et al., 2016), but fails to explain the enrichment observed in both  $\delta^2$ H and 71  $\delta^{18}$ O values. Additionally, such O isotope exchange has been shown not to readily occur in 72 73 speleothems (Uemura et al., 2019).

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75 A common hypothesis to explain the mismatch between LMWLs and the fluid inclusion 76 isotopes is that modifications are the result of evaporative fractionation in the soil, epikarst, or cave environment. Evaporation results in heavier oxygen and hydrogen isotope compositions 77 relative to the initial liquid and in shallow  $\delta^2 H / \delta^{18} O$  slopes that intersect the steeper local 78 79 meteoric water lines (LMWLs) (e.g., Craig H, 1961; Horita et al., 2008). While this hypothesis 80 appears at first sight to explain the main observations outlined above, there are other possible 81 explanations. Several studies have raised the possibility that evaporation could also occur 82 during analysis when samples are placed in an analytical apparatus at high temperatures 83 (typically 120°C; Matthews et al., 2021; Warken et al., 2021). However, additional data from 84 experiments and measurements of natural samples are needed to determine which of the 85 hypotheses explains the observations.

87 Here, we test the reliability of fluid inclusion isotope analyses with a stalagmite from 88 Borneo that grew during the last deglaciation. We specifically examine the extent to which fluid 89 inclusion waters can evaporate during laboratory analyses before the crushing step that 90 releases water to the analyzer. Borneo is the ideal location for this work for several reasons. 91 Evaporation in Borneo caves is unlikely given the year-round 100% relative humidity of cave air 92 at this tropical location (Meckler et al., 2015). If isotopic enrichments occur in fluid inclusion 93 water isotopes, they are most likely the result of another process. Furthermore, the temperature and hydroclimate evolution of Borneo through the deglaciation are relatively well 94 understood. Changes in hydroclimate are constrained with carbonate  $\delta^{18}$ O measurements, 95 which reproduce well across four different stalagmites (Buckingham et al., 2022; Partin et al., 96 97 2007), suggesting minimal variations over time in isotopic disequilibrium during stalagmite formation. The interpretation of carbonate  $\delta^{18}$ O is corroborated by calculated drip-water  $\delta^{18}$ O 98 values (Løland et al., 2022), and is consistent with isotope-enabled transient climate simulations 99 100 (Buckingham et al., 2022; He et al., 2021). The regional temperature evolution is well-101 constrained with fluid inclusion microthermometry, a method that produces exceptionally 102 precise paleotemperatures (Krüger et al., 2011). These available constraints allow us to 103 compare fluid inclusion water isotope data to independently derived dripwater  $\delta^{18}$ O, and calculated fluid-calcite  $\delta^{18}$ O temperatures with available microthermometry estimates. 104 105 106 2.0 Methods 107 108 2.1 Site and sample description 109 110 We measured water isotopes in fluid inclusions (also referred to as fluid inclusion isotopes here) from a stalagmite collected in Northern Borneo (SSC01; Partin et al. 2007). 111 112 Borneo is located in the tropical West Pacific, the region with the warmest modern ocean temperatures. The site remains year-round within the center of tropical convection as the 113 Intertropical Convergence Zone (ITCZ) migrates north and south. The region receives an 114 115 average of 5000 mm annual rainfall with only weak seasonality. The stalagmite SSC01 was 116 collected in 2003 from Snail Shell Cave (4º 12' 20.8" N, 114 º 56'26.9" E), which is located in 117 Gunung Buda in the northwestern corner of Malaysian Borneo and has previously been dated

- and investigated for carbonate  $\delta^{18}$ O (Partin et al., 2007).
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121 We compare our data to calculated dripwater isotope values and fluid inclusion 122 microthermometry estimates from another stalagmite from an adjacent mountain, SC02 (Løland et al., 2022). Stalagmite SC02 was collected in 2006 from Secret Chamber in Gunung 123 124 Mulu National Park (Buckingham et al., 2022). Dripwater isotope values in Gunung Mulu caves have been shown to closely match rainfall  $\delta^2$ H and  $\delta^{18}$ O values with an average residence time 125 in the karst of 3 to 18 months, and regional rainfall amount is thought to exert the dominant 126 127 control on interannual rainfall isotope variability (Ellis et al., 2020; Moerman et al., 2014). The position of the local meteoric water line (LMWL) is well constrained with quasi biweekly 128 129 dripwater samples collected from 2006 to 2018 at three sites in two caves (see supplemental

130 1). A linear regression drawn through these data results in the following equation:  $\delta^2 H =$ 131 8.06 (±0.04) ×  $\delta^{18}O$  + 13.74 (±0.3). Long multi-year measurements of cave temperatures 132 exist for several caves in Gunung Mulu, and these show that cave temperatures remain close to 133 24°C with less than 0.2 °C variations throughout the year (Løland et al., 2022). Since long-term 134 temperature measurements do not exist for Snail Shell Cave, we assume that the nearby Mulu 135 temperature range is also representative for that site.

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The SSC01 stalagmite was originally split in half along the central growth axis by Partin 137 138 et al. (2007), who used one half for geochemical analyses, while the other half is used here. The 139 stalagmite half was embedded in plaster, and a diamond wire saw (Well 6500) was used to cut slabs of 7 mm thickness lengthwise. Blocks of 20 to 40 mm length and 20 mm width were then 140 141 cut from the slabs along the central axis. Before the blocks were sampled for fluid inclusion 142 isotope analyses, thin sections were obtained for petrographic inspection. Samples for fluid 143 inclusion analysis were then cut from the blocks with a smaller diamond wire saw (Well 3421). 144 To this end, the calcite blocks were first glued to glass plates on both sides and fixed on the 145 sample holder of the saw. The sample holder was attached to a translation stage, which enables 146 stepwise lateral displacement of the sample with respect to the cutting direction resulting in a 147 curvature of the cuts. The samples were cut along to the growth bands closely following their 148 three-dimensional orientation. This ensures that coeval samples are obtained. Two to five 149 samples were obtained from each block and placed in acetone to remove the glued glass plates. 150 These samples were then cut into three to four equally sized replicate samples of 151 approximately 0.2 to 0.8 g.

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## 153 **2.2 Calcite stable isotope measurements and age model**

To place the results in the revised chronology of (Partin, 2008), calcite  $\delta^{18}$ O was re-154 measured on the sample blocks and the results were compared to the published record 155 (Supplemental 2). Samples were obtained by continuous milling at 1 mm increments with a 156 157 Sherline 540 micromill. Measurements were performed in the Facility for Advanced Isotopic Research (FARLAB) at the Department of Earth Science at the University of Bergen using a MAT 158 159 253 isotope ratio mass spectrometer (IRMS-Thermo Fisher) coupled to a Kiel IV carbonate preparation device. A total of 347 calcite samples were measured. The average measurement 160 error is less than 0.05‰ (1 $\sigma$ ) for  $\delta^{13}$ C and 0.10‰ (1 $\sigma$ ) for  $\delta^{18}$ O. 161

## 162 **2.3 Fluid inclusion isotope measurements**

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164 Fluid inclusion water isotope measurements were conducted following the procedures 165 described by de Graff et al. (2020). The analytical line consists of a crusher device that is connected to a humidity generator and to a Picarro cavity ring-down spectrometer (CRDS) on 166 opposite ends. The crusher device is constructed in stainless steel and follows the design of de 167 168 Graff et al. (2020) with some modifications. Samples were measured with all analytical 169 components under constant humid conditions to avoid memory effects (Affolter et al., 2014; 170 Dassié et al., 2018; de Graaf et al., 2020). The humidity generator is composed of a modified 171 300 ml stainless-steel cylinder that serves as an evaporation chamber and a microdrop

172 dispenser head (MK-130, Microdrop GmbH, Germany) that ejects water droplets (<100 µm 173 diameter) into the evaporation chamber by piezoelectric stimulation. Specific details on the 174 design and performance of the humidity generator device are described in Sodemann et al. (in

- 175 prep). During analyses the evaporation chamber of the humidity generator was kept at a
- 176 constant temperature of 60°C. The crusher device and related stainless-steel lines were kept at a temperature of 120°C. A distilled water standard of known isotopic composition (DI2;  $\delta^{18}$ O = -177

7.63  $\pm$  0.06 and  $\delta^2$ H = -50.72  $\pm$  0.46) was used to generate the humid background.

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180 Depending on the expected water content, between 0.2 to 0.8 g of stalagmite sample 181 were used for analyses, and whenever possible samples were measured a minimum of three 182 times. A total of 119 replicate analyses from 40 samples were performed. Before analysis, 183 samples were loaded into the pre-heated crusher device and approximately 15-25 minutes were needed for background stabilization. After stabilization the sample was crushed, and the 184 resulting water was measured by the analyzer. Raw  $\delta^{18}$ O and  $\delta^{2}$ H data for sample and 185 background were corrected for the humidity dependency of the analyzer following the 186 procedures described in (Weng et al., 2019). Sample  $\delta^{18}$ O and  $\delta^{2}$ H values were then calculated 187 following the algorithm presented by Affolter et al. (2014). These were subsequently 188 normalized to the VSMOW scale with a set of three in-house water standards, which had 189 190 previously been calibrated against international standards. Standards were injected on a daily 191 basis through a port that is installed immediately before the crusher device. The analytical precision (1 $\sigma$ ) of replicate analyses for samples larger than 0.3  $\mu$ l, as judged by injected 192 calibration standards, are 0.3% and 1.3 % for  $\delta^{18}$ O and  $\delta^{2}$ H, respectively. For samples between 193 0.1 and 0.3  $\mu$ l, precisions are 0.4‰ and 1.4 ‰ for  $\delta^{18}$ O and  $\delta^{2}$ H. Final errors in fluid inclusion 194 data are presented as confidence intervals at 68 or 95%. These are calculated with the standard 195 196 deviation of replicate analyses or the long-term reproducibility from water standards (shown 197 above), whichever was greater.

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199 The accuracy of water isotope measurements with the crushing line was assessed with 200 standard waters sealed in borosilicate glass capillaries (following Weißbach, 2020), and with 201 aliquots of a travertine sample that was previously measured in another laboratory 202 (Semproniano travertine, de Graaf et al. 2020) The glass capillary samples were designed to 203 mimic real inclusions since they receive the same treatment as calcite samples (i.e., water is released in the same location as calcite samples by turning the thread of the crusher device). 204 205 The mean values obtained for both glass capillaries and the travertine are statistically indistinguishable from their 'known' values. Stalagmite water amounts ( $\mu$ l/g) were calculated 206 with a transfer function built with calculated water amounts (area under peaks) and water 207 amounts from injections of standard waters (0.05-1  $\mu$ l, r<sup>2</sup>=0.99, RMSE=0.02). The accuracy of 208 water amount measurements were determined to be  $\pm$  5% (1 $\sigma$ ) based on weighted glass 209 210 capillaries. 211

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216 3 Results

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#### 218 **3.1 Petrography – SSC01 Calcite Fabrics and Fluid Inclusions**

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220 Thick sections (200  $\mu$ m) of stalagmite SSC01 were investigated with an optical 221 microscope under plane polarized (PPL) and cross polarized light (XPL) to characterize the calcite fabrics and occurrence of fluid inclusions. The calcite fabrics and occurrence of fluid 222 223 inclusions can be classified into two general groups that are closely associated with age. For 224 instance, Holocene and late deglacial calcite fabrics are dominated by clear, transparent 225 columnar calcite with straight crystal boundaries and up to centimeter wide domains of uniform extinction under XPL (Figure 1A, 1B). The fabric is characterized by countless very small 226 227 fluid inclusions that are dispersed in the matrix and range in size between 1 and 10  $\mu$ m<sup>3</sup> (Figure 1C), and by some large up to 1 mm long inter-crystalline inclusions. Sporadically, the calcite 228 229 fabric is interrupted by distinct layers with numerous inclusions in the range of  $10^2$ -  $10^3 \mu m^3$ 230 (Fig. 1A, 1B). These inclusion-rich layers possibly formed onto former growth hiatuses. Early 231 deglacial and glacial samples, in contrast, consist of alternating layers of open columnar and 232 microcrystalline calcite fabrics (Figure 1C, 1D). The latter are rich in organic matter, as confirmed by confocal fluorescence microscopy. Fluid inclusions are abundant in the columnar 233 fabric and orders of magnitudes larger ( $10^3$  and  $10^5 \mu m^3$ ) than the tiny inclusions that dominate 234 the main part of the Holocene and late deglacial fabric of stalagmite SSC01 (Figure 1F). Large 235 236 inter-crystalline inclusions are also present in the early deglacial and glacial parts of the 237 stalagmite.

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A. Holocene (8-9.5 ka)



D. Early deglacial (16.7-17.2 ka)











F. Early deglacial - glacial



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245 Figure 1. Optical microphotographs of polished thick sections from representative portions of the glacial, 246 deglacial, and Holocene sections of SSC01. A and B) thin section images from Holocene and late deglacial 247 sections taken under cross-polarized light. C) microphotograph at higher magnification showing the very small (1 248 and 10  $\mu$ m<sup>3</sup>) inclusions present in the Holocene and late deglacial portions of the stalagmite. 1) columnar calcite 249 with countless tiny fluid inclusions and some large inter-crystalline inclusions indicated by yellow arrows. 2) dark 250 inclusions-rich layers possibly overgrowing a former hiatus surface enriched in organic material. D and E) thin 251 section images from the early deglacial and the glacial portion of the stalagmite taken under cross-polarized 252 light. F) microphotograph at higher magnification showing the large ( $10^3$  and  $10^5 \mu m^3$ ) inclusions present in the 253 early deglacial and glacial portions of the stalagmite. 3) example of light, transparent open columnar layers with 254 abundant large fluid inclusions. 4) examples of dark-colored layers composed of organic-rich microcrystalline

- 255 calcite. Yellow arrows indicate large elongated inter-crystalline fluid inclusions.
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#### 258 **3.2 Relationship of fluid inclusion isotope data to the LMWL**







Figure 2. Fluid inclusion water isotope composition. A) Plot of sample means. B) Plot of replicate analyses. Points with red borders are within ± 68% (CI) of the LMWL. Data points are color coded by age as shown in the colorbar. The red dashed line is the LWML and black dashed line is the global meteoric water line (GMWL). Error bars in A and B are 68% confidence intervals.

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- standard deviations in  $\delta^{18}$ O greater than 0.4 ‰ are shown. Blue lines are individual linear fits calculated with the algorithm of York et al. (2004). Shaded grey regions are regression 68% confidence intervals obtained by Monte Carlo simulations considering errors in both  $\delta^{18}$ O and  $\delta^{2}$ H values. B) Water content of individual replicate analyses vs.  $\Delta\delta^{18}$ O LMWL ( $\delta^{18}$ O deviation from LMWL). Black dashed lines are linear regressions with negative slopes, and red dashed lined are linear regressions with positive slopes.
- 298 slopes, and red dashed lined are linear r
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## 301 **3.2** – Relationships between fluid inclusions, water content and $\delta^{18}$ O deviations from the 302 LMWL

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There is a close association between water content,  $\delta^{18}$ O deviations from the LMWL 304  $(\Delta \delta^{18} O LMWL)$ , and the occurrence of fluid inclusions in the stalagmite. Higher water contents 305 306 are observed in samples from the early deglacial and glacial samples (Figure 4A); these samples have abundant large (i.e.,  $10^3$  and  $10^5 \mu m^3$ ) fluid inclusions in the open columnar fabric. Much 307 308 lower water amounts were observed in the Holocene and late deglacial samples (Figure 4A). 309 These samples have abundant but small inclusions (i.e., 1 and 10  $\mu$ m<sup>3</sup>) and very few larger (i.e., 310 up to 1 mm long) inter-crystalline inclusions. The abrupt change in water content that we 311 observe during the deglaciation is also mirrored in the proportions of samples and replicate 312 analyses that fall off the LWML (Figure 4B). For instance, most of early deglacial and LGM samples fall close to the LMWL, i.e., they have  $\delta^{18}$ O values that are close to what is expected 313 based on their  $\delta^2$ H composition ( $\Delta \delta^{18}$ O LMWL values are close to zero). The younger samples 314 from the late deglacial and Holocene, however, have  $\delta^{18}$ O values that, with few exceptions, fall 315 316 off the LMWL and are farther away from the LWML. 317



Figure 4. A) Water content ( $\mu$ l/g) of SSC01 stalagmite plotted against age. Yellow squares are mean values of replicate analyses, and circles are individual analyses. Red circles are replicates with  $\delta^{18}$ O and  $\delta^{2}$ H values that fall within 68% CI of the LMWL. B)  $\delta^{18}$ O deviations from the LMWL ( $\Delta\delta^{18}$ O LMWL). Circles are replicate analyses and blue triangles are sample means. Red data circles are replicates that have values that fall on the LMWL.

3.2 Comparison of inclusion data with independent temperature and dripwater  $\delta^{18}$ O proxies

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331 The time-dependence of observed offsets and their possible cause can be further investigated by comparing the fluid inclusion isotope results to available independent 332 information on temperature (from fluid inclusion microthermometry; Løland et al., 2022) and 333 dripwater  $\delta^{18}$ O from stalagmite SC02 collected from a cave in an adjacent mountain (Fig 5). 334 Dripwater  $\delta^{18}$ O values for SCO2 were calculated with microthermometry temperature estimates 335 and the corresponding calcite  $\delta^{18}$ O values using the empirical cave calibration for  $\delta^{18}$ O of 336 337 Tremaine et al. (2011) (see Løland et al., 2022). We observe that SSC01 fluid inclusion  $\delta^{18}$ O 338 values are generally more positive than calculated drip water values from SC02. However, values are closer to those in SCO2 in older parts of the SSCO1 record than in younger parts. 339 Moreover, we find closer agreement when only the SSC01  $\delta^{18}$ O values that fall on the LMWL are 340

341 considered (Figure 5A). The agreement is even more apparent when only individual replicate 342 analyses are considered; replicates that fall on the LMWL follow the same trends as in SCO2 343 throughout the length of the record (Supplementary Figure 4). In contrast to fluid inclusion 344  $\delta^{18}$ O,  $\delta^{2}$ H values are much closer to the calculated SCO2 drip water values, although still

- 345 showing larger deviations in the younger part of the record (Figure 5B).
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347 If we were to use our fluid inclusion water isotope results at face value, fluid-calcite  $\delta^{18}$ O 348 temperatures, calculated with the equation of Tremaine et al. (2011), yield mostly

- 349 unrealistically high temperatures and are almost always warmer than microthermometry
- estimates (Figure 5C). A closer agreement is observed during the LGM and the early deglacial
- 351 where temperatures are on average 2.3°C degrees warmer. In younger samples, on the other
- hand, temperatures are on average 7.3°C warmer than fluid inclusion homogenization
- temperatures. In a similar manner to SSC01 fluid inclusion  $\delta^{18}$ O values, we observe better agreement between both paleothermometers when only temperatures calculated with  $\delta^{18}$ O
- agreement between both paleothermometers when only temperatures calculated with  $\delta^{18}$ O values that fall on the LMWL are considered. This is true at the sample mean and the replicate
- 356 level (Figure 5C and Supplemental 4). Overall, the comparison with independent constraints
- 357 supports our interpretation that the offsets from the LMWL are likely artifacts and illustrates
- 358 their impact on the interpretation of the results.



Figure 5. A) Measured fluid inclusion  $\delta^{18}$ O values of stalagmite SSC01 (circles) and calculated drip water  $\delta^{18}$ O values of stalagmite SC02 from the same region (triangles; Løland et al., 2022). B) Measured fluid inclusion  $\delta^{2}$ H values of SSC01 (circles) and calculated drip water  $\delta^{2}$ H values of SC02, using the LMWL (triangles). C) fluid-calcite  $\delta^{18}$ O temperatures of SSC01 calculated with the temperature relationship of Tremaine et al. (2011) (circles) and fluid inclusion microthermometry temperatures of SC02 from Løland et al. (2021). Error bars are 68% confidence intervals. Open circles are samples that fall within 68% CI of the LMWL.

#### 370 4. Discussion

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#### 372 **4.1 Water loss through evaporation during analyses**

- We observe enrichments in fluid inclusion  $\delta^2 H$  and  $\delta^{18} O$  values, shallow  $\delta^2 H / \delta^{18} O$  slopes 374 375 in replicate analyses, and poor agreement between fluid inclusion data and independent 376 paleoclimate proxies. These observations could be interpreted as indication of evaporation in 377 the cave. For the cave site investigated here, however, we expect that evaporation inside the 378 cave is unlikely given a year-round relative humidity in Borneo caves of nearly 100% (Meckler et 379 al., 2015). For this reason, we now explore the alternative hypothesis that evaporation occurred 380 in the crusher device at 120 °C, immediately before the crushing step. The physical reasoning 381 behind this hypothesis is the increase of fluid pressure with increasing temperature in an isochoric system. At a certain threshold the fluid overpressure inevitably induces irreversible 382 383 volume changes of the inclusions, most likely due to the formation of cracks in the confining 384 calcite host, preferentially along the cleavage planes and, in case of inter-crystalline inclusions, 385 along crystal boundaries. The formation of cracks results in a volume increase of the inclusions, and thus, in a relaxation of the fluid overpressure. Upon further heating, the cracks propagate 386 387 and waters can then leak along crystal boundaries, internal porosity and primary fractures in 388 the stalagmite and evaporate. Partial evaporation of water from fluid inclusions would then lead to some degree of isotopic modifications and enrichments in  $\delta^2 H$  and  $\delta^{18} O$  values and thus 389 390 affect the isotope signal that is measured after crushing the sample. Inclusions that lose all their water in the course of the 15-20 minutes of background stabilization, on the other hand, have 391 392 no effect on the measured isotope signal.
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394 Ideally water loss can be observed as samples are heated and the evolving water is 395 detected by the analyzer. However, this is not possible with our analytical set-up. When the 396 crusher device is opened to load a sample, the water background measured by the analyzer 397 quickly decreases to match the humidity of the laboratory air. Once the crusher is closed, the 398 humidity slowly rises over the next 15-20 minutes to the level set by the humidity generator. 399 This makes it impossible to observe any water loss from the sample that may be occurring 400 during this time. For that reason, we designed an experiment to manometrically determine if 401 water is lost as a sample is heated in a vacuum line (see supplemental 5 for details). Briefly, a 402 sample was loaded into a glass tube, connected to a vacuum line and evacuated. The sample 403 was then heated to approximately 120°C with a heat gun, and any released water was collected 404 in a cold trap bathed in liquid N<sub>2</sub>. After 20 minutes, the liquid N<sub>2</sub> was removed, and the water 405 pressure was measured in a manometer. The manometer pressure was calibrated for water 406 amounts by adding known amounts of water to the same volume. The experiment was 407 performed with two different samples from the Holocene portion of the stalagmite; we 408 estimate that for both samples approximately 10% of the water content was lost (see 409 supplemental 5 for details). 410

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#### 414 4.2 Evaporation model

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416 We have demonstrated that in our samples, water loss can occur at temperatures of 417 120°C, and have argued that sample specific evaporative trends are suggestive of analytical 418 artifacts. In the next section, we use a Rayleigh model to simulate the isotopic composition of 419 water during fluid inclusion analysis, in comparison to those that would be expected for 420 evaporation in cave environments. These predictions are then used to determine if the  $\delta^2 H/\delta^{18}$ O slopes and the magnitudes of isotopic fractionations observed in the fluid inclusion 421 422 data are characteristic of the temperature and humidity conditions of the crusher device. 423 424 Isotope ratios during evaporation can be calculated with the Rayleigh equation:

425

$$R_l = R_{l-0} \times f^{\left(\alpha_{eff} - 1\right)} \tag{1}$$

426

427 where  $R_l$  is the isotope ratio (<sup>18</sup>R or <sup>2</sup>R) of the liquid at each step of an evaporative process, 428  $R_{l-0}$  is the isotope ratio of the initial liquid, f is the fraction of liquid remaining at each step, 429 and  $\alpha_{eff}$  is the effective vapor-liquid isotope fractionation factor during evaporation 430  $(R_v/R_l < 1)$ .  $\alpha_{eff}$  was calculated with the formulation of the Craig-Gordon model (Craig and 431 Gordon, 1965) for evaporation into an atmosphere of fixed humidity and isotopic composition: 432

$$\alpha_{eff} = \alpha_{diff} \left[ \frac{\alpha_{eq} - h(R_a/R_l)}{1 - h} \right]$$
(2)

434

435 where  $\alpha_{eq}$  is the temperature dependent equilibrium vapor-liquid isotope fractionation factor, 436 *h* is the relative humidity,  $R_a$  is the isotope ratio in air,  $R_l$  is the isotope ratio in the liquid, and 437  $\alpha_{diff}$  is the isotope fractionation during diffusion in air.  $\alpha_{diff}$  can be expressed as the ratio of 438 the molecular diffusivities of the heavy (HDO and H<sub>2</sub><sup>18</sup>O) and light (H<sub>2</sub>O and H<sub>2</sub><sup>16</sup>O) water 439 isotopologues: 440

- 440
- 441

 $\alpha_{diff} = \left(\frac{D_H}{D_L}\right)^n \tag{3}$ 

442

443  $D_H$  and  $D_L$  are the heavy and light isotopologues, respectively. The exponent n varies from zero 444 in flow under completely turbulent conditions to one in purely diffusional regimes (Gonfiantini 445 et al., 2018; Horita & Wesolowski, 1994).

446

447 For evaporation in the crusher device, we used equilibrium fractionation factors at 448 120°C calculated with the relationships presented by Horita & Wesolowski (1994), and a value 449 for relative humidity that corresponds to the typical humid backgrounds set by the humidity 450 generator ( $h \approx 1$  % for 10,000 to 14,000 ppmv of H<sub>2</sub>O at 120°C). The diffusion fractionation 451 factor was calculated with the molecular diffusivities of Merlivat (1978), and a value of n452 reflective of evaporation dominated by molecular diffusion (n=1), as expected for evaporation from porous materials like soil or leaf waters (Gonfiantini et al., 2018; Horita et al., 2008). 453 Isotope ratios of the water vapor in air and the initial liquid (<sup>18</sup>R and <sup>2</sup>R) were calculated with 454

455 the isotopic composition of the water background using a laboratory standard (DI2;  $\delta^{18}$ O = -7.63 456 ± 0.06 and  $\delta^2$ H = -50.72 ± 0.5) and with the average isotopic composition of the fluid inclusion 457 data ( $\delta^{18}$ O = -7.7 and  $\delta^2$ H = -48.1), respectively.

458

459 For evaporation in the cave system, we used equilibrium fractionation factors at 25°C 460 (Horita and Wesolowski 1994). The isotope ratios of the initial liquid were approximated with the average isotopic composition of the fluid inclusion data, and the isotope ratios of the cave 461 462 air vapor were assumed to be equal to the isotope ratios of vapor in isotopic equilibrium with 463 the initial liquid. Even though measured cave humidity in Borneo has always been close to 100%, we used a range in relative humidity of 60 to 90% to include the possibility of less humid 464 465 conditions. Appropriate values of the *n* parameter for typical cave conditions are difficult to 466 assign as they are not known. In laboratory experiments, however, high n values (close to 1) are obtained when evaporation rates are slow under quiet atmospheres (Gonfiantini et al., 2018). 467 These conditions are likely similar to those experienced inside caves with poor ventilation. 468 469 Similarly, n values close to 1 are expected if evaporation occurs in the epikarst where water 470 evaporates through fractures and/or internal porosity. On the other hand, lower n values (close 471 to 0.5) are observed in experiments when evaporation proceeds in open air conditions 472 (Gonfiantini et al., 2018). These values may be more appropriate for caves with strong 473 ventilation. Given these uncertainties, we use a range of n values of 0.5 to 1, which 474 encompasses the full range of possible conditions that may occur in a cave system. 475



476 477

478
479 Figure 6. Results of the Rayleigh models for an evaporation process. A) Slopes in δ<sup>18</sup>O vs. δ<sup>2</sup>H space for
480 evaporation in the crusher device (slope 1.4 - blue line) and range of slopes for evaporation in a cave (slopes 3.9
481 to 2.7 - green filled area). Dashed line is the LWML. B and C) Change in fluid inclusion δ<sup>18</sup>O and δ<sup>2</sup>H values,

482respectively, vs. total water lost to evaporation (f = fraction of initial water remaining). Blue and green circles483are  $\delta^{18}$ O and  $\delta^{2}$ H replicate analyses, respectively. F (initial) was calculated assuming that the initial water484content for each sample is equal to that observed in the replicate analyses that plot closer to the LWML. D and485E) Change in  $\delta^{18}$ O and  $\delta^{2}$ H values vs. water lost in an evaporative reservoir. Dashed lines indicate the trends486predicted given different amounts of inclusions that experience evaporation (i.e., 100% means that all inclusions487experience evaporation, and 25% means that only 1/4 of total inclusions loose water).

488

489 For evaporation in the crusher, the Rayleigh model predicts a slope of 1.4 (Fig. 6A, blue 490 line). This value is statistically indistinguishable to the mean evaporative slope calculated with 491 the replicate dataset (1 ± 0.6 2SE). This fact adds additional support to our interpretation that 492 isotopic enrichments are the result of partial evaporation during analyses. For evaporation in 493 the cave system, the model predicts slopes that are larger than the empirical estimate, ranging 494 between 2.7 to 3.9 (Fig. 6A, red area), and where magnitudes are primarily controlled by the value of the *n* parameter (Supplemental 6). These results are generally consistent with the 495 496 isotopic enrichments inferred for evaporation in cave systems. Values of evaporative slopes are 497 not always reported, but two estimates are available. Warken et al. (2021) report a slope of 2.4 498 for fluid inclusion data from a stalagmite from Puerto Rico, and interpret it as the result of 499 evaporation on top of the stalagmite. An additional slope of 3.9 was reported by Wortham et al. 500 (2022), who interpreted it as the result of evaporation in the soil, epikarst or inside the cave. At 501 face value, these slopes are consistent with the results of the model. Moreover, and in contrast 502 to our Borneo stalagmite, evaporation is actually possible in both previously analyzed caves, as 503 they experience seasonal changes in cave ventilation and humidity lower than 100 %. However, 504 we caution that evaporative slopes were calculated from samples that are not coeval. 505 Evaporative slopes may therefore be uncertain if the primary isotopic composition of the 506 unevaporated water was variable. Moreover, evaporative loss through analyses cannot be ruled 507 out, so results may incorporate modifications by both processes.

508

509 The results of both model scenarios predict larger fractionations – and thus larger 510 distances from the LMWL – in samples that experienced more evaporation (Fig. 6B and 6C). 511 Our observations from the replicate measurements are mainly consistent with these 512 predictions. For example, in the replicate dataset there is a general relationship between lower water contents and larger  $\delta^{\rm 18}{\rm O}$  offsets from the LMWL (Figure 3B). Note that this is only true in 513 514 9 out of the 11 samples that we tested. Two samples show either no trend or a trend in the 515 opposite direction (red dashed lines in Figure 3B), which cannot be explained by evaporative fractionation. However, we interpret these discrepancies as the result analytical errors in  $\delta^{18}$ O 516 517 variable water contents, difficulties in measuring strictly coeval samples, and variability in the 518 number of inclusions that experience partial vs. total leakage. These are further discussed 519 below (Sec. 4.3). 520

521 Although the model for the crusher device explains well the slopes of the evaporation 522 trends, and the general relationship observed between distance from the LMWL and water loss, 523 it does not correctly predict the magnitudes of the fractionations that we observe. For example, 524  $\delta^{18}$ O and  $\delta^{2}$ H enrichments in the model are more than 30 ‰ larger than our observations 525 (Figure 6B and 6C). Smaller observed fractionations, though, can be explained by mixtures of 526 water from pristine and partly leaked inclusions. While all inclusions become stretched upon 527 heating to 120 °C, only some of them start leaking, i.e. connect to the sample surface via cracks, 528 crystal boundaries and interconnected pores and lose all or part of their water through 529 evaporation. Inclusions that do not leak, preserve the original drip water isotopic composition. 530 When these are crushed, the pristine water mixes with waters from partially evaporated 531 reservoirs, and the magnitude of total observed fractionation is reduced. These mixing effects 532 can be easily incorporated into the evaporation model, but direct comparison between 533 predictions and our data are not straightforward. The main reason is that we do not know the 534 relative size of the pool of inclusions in our samples that are amenable to leakage. 535 Nevertheless, we can show that the range of observed fractionations are consistent with 536 mixtures of 95 to 75% of pristine inclusions and 5 to 25% evaporated reservoirs (Figure 6D and 537 6E). It is important to note that this does not necessarily mean that only a small percentage of 538 inclusions lost water; on the contrary, in some cases the majority of water was probably lost. For instance, we observe a range of more than 80% in the water content of some samples 539 540 (Figure 6B and 6C). Part of that range likely results from variable initial water contents, but a 541 considerable part of it is probably due to leakage and complete loss of water from inclusions 542 through evaporation. Since completely evaporated reservoirs do not affect the measured 543 isotopic composition, they can explain large variabilities in recovered water with only small 544 concomitant changes in delta values.

#### 545

#### 546 **4.3 Fluid inclusions, water contents, and isotopic modifications**

547

The majority of early deglacial and glacial samples yield  $\delta^{18}$ O values that fall close to the 548 LMWL. The opposite is true for Holocene and late deglacial samples; most of them yield  $\delta^{18}$ O 549 550 values that plot to the right of the LMWL. We have demonstrated that isotopic enrichments can be explained by evaporative fractionations during analyses. Since we observe a close 551 552 correlation between fractionations and the age of the samples, an open question is what 553 features make some samples more amenable to isotopic modifications in the crusher device? 554 The most striking differences between younger and older portions of the stalagmite are the size 555 distributions of the fluid inclusions. While very large and elongated inter-crystalline inclusions 556 are present throughout the stalagmite, the older parts have abundant and relatively large fluid 557 inclusions that are orders of magnitude larger than the very small fluid inclusions observed in 558 the Holocene and late deglacial portions of the stalagmite (see section 3.1). These observations 559 explain why more water was recovered from older samples but the water content of the 560 samples alone does not explain why the younger samples tend to show larger isotopic 561 modifications. 562

563 Our hypothesis to explain why that occurs is that the very large and elongated inter-564 crystalline fluid inclusions (present throughout the stalagmite) are more likely to leak and lose 565 water than other inclusions in the calcite fabric. A possible reason is the length of these 566 elongated inclusions and their position between crystal boundaries, which gives them a better 567 opportunity to be connected to the surfaces of the samples via cracks and interconnected 568 pores. These inclusions, thus, form a partially evaporated reservoir that mixes with a more 569 pristine reservoir from the other inclusions when the samples are crushed. Younger samples 570 show larger isotopic modifications because the very small inclusions (pristine) likely contribute 571 only small amounts of water, and a relatively large fraction of water that was measured in 572 these samples was provided by large, partially evaporated, inter-crystalline fluid inclusions. In 573 the older samples, in contrast, where isotopic modifications are significantly smaller or absent, 574 the inclusions that contribute to the pristine reservoir are orders of magnitude larger than 575 those in the Holocene and late deglacial samples and likely provide most of the water. 576 Although evaporation also occurs in these samples, the size of the partly evaporated reservoir 577 relative to the pristine reservoir is much smaller. Given these observations, we suggest avoiding 578 samples with low water contents and a large contrast of inclusion sizes. Figure 4B, however, 579 illustrates that isotope compositions close to the LMWL are also possible with samples that 580 contain only little water. That being said, we caution that additional observations from other 581 stalagmites are necessary to draw broad conclusions on the leakage susceptibility of different 582 fluid inclusions and on sampling strategies for fluid inclusion analyses.

583

585

#### 584 **4.4 Correction of evaporative trends**

586 In this section we describe an approach to correct fluid inclusion water isotope 587 measurements for evaporative fractionation during analysis, and we discuss the reliability of 588 the proposed correction algorithm. The most straightforward approach to correct the data is to use the  $\delta^2 H/\delta^{18}$ O slope of the evaporative trends. This is possible because all samples should 589 share the same slope, as they were subjected to the same conditions during analyses (e.g., 590 591 temperature, relative humidity, flow regimes). To correct the data, we constructed linear 592 models for each replicate measurement using the evaporative slope that was estimated with the Raleigh distillation model ( $\delta^2 H / \delta^{18} O = 1.4$ ; Figure 6A). The original (pristine) isotope 593 compositions were then determined by the points of intersection between the linear models 594 595 and the LMWL. Corrected data are presented in Figure 7.

596

597 Unlike the raw fluid inclusion data (Figure 5), the corrected data are in very close 598 agreement to the calculated SC02 dripwater isotope values (Figure 7A and 7B). For example, calculated and measured  $\delta^{\,\rm 18}{\rm O}$  and  $\delta^{\rm 2}{\rm H}$  values show the same trends throughout the length of 599 the record, and measured SSC01  $\delta^{18}$ O and  $\delta^{2}$ H values are within ± 95% CI of calculated values 600 601 from SC02. Moreover, as expected – given the shallow slopes of the evaporative trends – the correction has a larger relative effect on  $\delta^{18}$ O values than on  $\delta^{2}$ H values. Much closer 602 agreement is also found between the fluid-calcite  $\delta^{18}$ O temperatures and SCO2 603 604 microthermometry estimates when the former are calculated with the corrected  $\delta^{18}$ O values 605 (Figure 7C). Corrected values are all within ± 95% CI of the microthermometry estimates. 606



Figure 7. A) Corrected fluid inclusion  $\delta^{18}$ O values of the SSC01 stalagmite (circles) and calculated drip water  $\delta^{18}$ O

- 610 values of the SC02 stalagmite (triangles). B) Corrected fluid inclusion  $\delta^2$ H values of the SSC01 stalagmite (circles) 611 and calculated drip water  $\delta^2$ H values of the SC02 stalagmite with the LMWL triangles). C) fluid-calcite  $\delta^{18}$ O
- temperatures of SSC01 from corrected  $\delta^{18}$ O values (circles) and microthermometry temperatures of SC02 from
- 613 Løland et al. (2022) (triangles). Fluid-calcite  $\delta^{18}$ O temperatures were calculated with the Tremaine et al. (2011)
- relationship. Error bars are 68% confidence intervals. Open circles are samples that fell within 68% CI of the
- 615 LMWL before correction.

616 617 We have shown that the correction procedure removes the disagreement between 618 calculated and measured dripwater isotope values, and between fluid-calcite  $\delta^{18}$ O temperatures and microthermometry estimates. However, our correction routine relies 619 620 on independent knowledge of the LWML. We used the present-day LWML to correct data for 621 analytical artifacts; however, in some settings the LMWL may not be precisely known or it may 622 have been different in the past. For example, the LWML can vary because it depends on the 623 deuterium excess of local precipitation, which is primarily controlled by the moisture conditions 624 of the source regions (e.g., temperature, relative humidity and isotope composition; Pfahl & 625 Sodemann, 2014), and the presence/absence of non-equilibrium processes during precipitation 626 (e.g., re-moistening and rain drop evaporation; Lee & Fung, 2008; Martinelli et al., 1996). This 627 is the main disadvantage of our correction routine, and it can be an important source of bias in 628 paleoclimate reconstructions.

629

630 To examine the sensitivity of corrected values to possible changes in the LWML, we 631 looked at results from the iTRACE experiment (He et al., 2021), which is a transient (20-11 ka) 632 water isotope enabled simulation of global climate using the Community Earth System Model 633 version 1.3. In the closest grid cell to the Borneo site, the model predicts variations in LMWL 634 parameters of up to 3 ‰ in the intercept and 0.3 ‰ in the slope. The magnitude of these 635 changes, however, are insufficient to impact corrected fluid inclusion data outside of analytical 636 error (see Supplemental Figure 7). As an additional robustness check, we looked at LMWLs from 637 three Global Network of Isotopes in Precipitation (GNIP) sites in the Indo-Pacific region. These 638 sites are located at latitudes where seasonal migrations of the intertropical convergence zone 639 (ITCZ) cause large differences in seasonal precipitation amounts. We use changes in LMWLs at 640 these sites as proxies for hydrological conditions in Borneo during the deglaciation, where 641 regional arid conditions are thought to be caused by a southward shift in the ITCZ over the 642 Pacific (Buckingham et al., 2022; Carolin et al., 2013; Partin et al., 2007). We also find that 643 changes in the LMWLs at these sites do not have an impact on data correction outside of 644 analytical errors. These observations suggests that the correction is not very sensitive to 645 changes in the LWML parameters within a realistic range (see Supplemental Figure 7).

- 646
- 647 **4.5 Agreement between Borneo stalagmite paleothermometers**
- 648

649 Our corrected fluid inclusion data presents an opportunity to test the performance of the empirical fluid-calcite  $\delta^{18}$ O cave calibration of Tremaine et al. (2011). Empirical calibrations 650 651 such as the one from Tremaine et al. (2011) are needed because speleothem calcite rarely 652 forms in oxygen isotope equilibrium with dripwater (Guo & Zhou, 2019; Mickler et al., 2004). As previously discussed, fluid-calcite  $\delta^{18}$ O temperatures determined with this cave calibration 653 654 are statistically indistinguishable from independent microthermometry estimates over the 655 length of both records (Figure 7C). Thus, the cave calibration of Tremaine et al. (2011) appears 656 to perform very well in Borneo possibly because disequilibrium remained approximately 657 constant through time. This is surprising given the evidence for changes in temperature and 658 hydroclimate in Borneo through the deglaciation, which were likely accompanied by changes in 659 the environmental variables that drive disequilibrium (e.g., PCP, cave air pCO2, drip rate, water

660 film thickness, and growth rates; e.g., Carlson et al., 2020; Deininger et al., 2021; Johnston et 661 al., 2013). While counterintuitive at first sight, the observations can be explained if 662 disequilibrium – at the timescales captured by our samples– is not very sensitive to changes in 663 these variables. There is some evidence from theoretical models to support this idea. For 664 example, Guo & Zhou (2019) recently published results of a reaction diffusion model that tracks 665 the concentrations of isotopologues in the CaCO<sub>3</sub>-DIC-H<sub>2</sub>O system during speleothem formation. They found that their model is able to reproduce the temperature dependence of 666 <sup>18</sup>O fractionations observed by Tremaine et al. (2011). Moreover, when environmental variables 667 (PCP, cave air pCO<sub>2</sub>, and water film thickness) are randomly varied by 25% in their model, the 668  $\delta^{18}$ O value of calcite varies only by ± 0.23 ‰ (2 $\sigma$ ). This corresponds to only ± 1.2 °C, a 669 magnitude that cannot be detected given typical errors in fluid-calcite  $\delta^{18}$ O temperatures. Thus, 670 we hypothesize that disequilibrium in Borneo, despite changes in environmental variables that 671 672 drive it, remained close to the average magnitudes observed in modern speleothems and 673 within our error bars. 674 675 676 4.6 Deglacial hydroclimate evolution in Borneo 677 Corrected fluid inclusion data provide the first directly measured Borneo dripwater  $\delta^{18}$ O 678 and  $\delta^2 H$  values during the last deglaciation. These data agree well with previous estimates of 679 hydrological conditions inferred from calcite  $\delta^{18}$ O and with proxies for the strength of the 680 681 Atlantic Ocean meridional overturning circulation (AMOC) (Figure 8). For instance, we observe a shift to the most positive dripwater  $\delta^{18}$ O values during Heinrich event 1 (H-1), a time 682 683 interpreted as a period of weakened regional convection based on stalagmite  $\delta^{18}$ O 684 measurements (Partin et al., 2007; Buckingham et al., (2022) and calculated dripwater  $\delta^{18}$ O (Løland et al. 2022). Both the measured and calculated dripwater  $\delta^{18}$ O records show a gradual 685 decrease in dripwater  $\delta^{18}$ O values during H-1, a small decrease during the Bølling-Allerød (BA). 686 and a return to LGM values during the Early Holocene (Figure 8C and 8D). Especially for H-1, 687 our data add new details to the existing dripwater  $\delta^{18}$ O record, showing a late onset of the 688 hydroclimate response when compared to the AMOC record, and even suggesting a two-step 689 690 evolution. These features are worth further investigation.

- 691
- 692



Figure 8. Comparison of Borneo dripwater  $\delta^{18}$ O values with cave temperature and other paleoclimate records. A) 695 696 Greenland ice core (NGRIP)  $\delta^{18}$ O, a proxy for Greenland temperature (North Greenland Ice Core Project members, 2004). B) Changes in North Atlantic Ocean meridional overturing circulation based on the <sup>231</sup>Pa/<sup>230</sup>Th 697 698 of N. Atlantic sediment core OCE326-GGC5 (McManus et al., 2004). C) Measured SSC01 fluid inclusion  $\delta^{18}$ O 699 values after correction for analytical artifacts (blue circles, errors bars are ± 69% ci), and calculated SCO2 700 dripwater  $\delta^{18}$ O values from (Løland et al., 2022) (red dots). Both data series were corrected for ice volume related changes the  $\delta^{18}$ O value of the global ocean (see Løland et al., 2022 for details). The open blue symbol 701 702 shows a sample that was obtained across a hiatus in SSC01 stalagmite, implying uncertainty in its age. D) SC02 703 cave temperatures from fluid inclusion microthermometry corrected for sea-level induced changes in cave 704 altitude (Løland et al., 2022). E) Antarctic temperature anomaly (Parrenin et al., 2013).

707

## 706 **5. Conclusions and recommendations**

708 We measured the isotope composition of fluid inclusion waters in a stalagmite from 709 Borneo that grew during the last deglaciation. We find that many samples partially lose water due to evaporation during warm-up before the crushing step, and that water loss and the 710 magnitude of isotopic modifications are controlled by the type and size distribution of fluid 711 inclusions. Partial evaporation results in  $\delta^{18}$ O and  $\delta^{2}$ H enrichments that follow typical 712 evaporation lines with shallow  $\delta^{18}O/\delta^2H$  slopes. Through multiple replicate analyses of coeval 713 samples, we constrain the mean value of the evaporative  $\delta^{18}$ O/ $\delta^{2}$ H slope at 1 ± 0.6 (2SE). This 714 value is identical to the predictions of a Rayleigh distillation model that incorporates kinetic <sup>2</sup>H 715 and <sup>18</sup>O fractionations during evaporation in the hot crushing apparatus. 716 717

718We provide a robust and physically based method to correct for analytical modifications719and demonstrate that it can be used to reliably correct data to pristine dripwater isotopic720compositions. We find that corrected fluid-calcite  $\delta^{18}$ O and resulting paleotemperatures show721excellent agreement with independent dripwater  $\delta^{18}$ O and temperature estimates, which722implies only small variations in speleothem  ${}^{18}$ O disequilibrium. The corrected data provide the723first directly measured dripwater  $\delta^{18}$ O and  $\delta^2$ H estimates for Borneo across the last deglaciation724and support the previously inferred hydroclimate evolution.

#### 725

726 Our observations suggest that water loss during fluid inclusion water isotope analysis 727 readily occurs and that the magnitude of isotopic modifications can vary from sample to 728 sample. Future improvements to the analytical method should seek to remove these artifacts. 729 In lieu of such improvements, we suggest that multiple replicate measurements of coeval 730 samples and careful petrographic inspections be used to identify suitable samples, calcite 731 fabrics, and the type of fluid inclusions that are prone to evaporative artifacts. Special attention 732 should be given to samples with low water yields as these have been identified in this work and 733 by others as samples that more likely show isotopic modifications. Caution is warranted when interpreting speleothem fluid inclusion isotope data that is not checked for or corrected for 734 evaporation. This is especially true for  $\delta^{18}$ O data as these are affected by non-equilibrium 735 fractionation to a much greater extent than  $\delta^2 H$  relative to expected respective variations in 736 737 precipitation.

738 739

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741

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751	
752	7.0 Open Research
753	
754	Data availability statement. Data archiving is currently underway in the EARTHCHEM archive. A
755 756	temporary copy of the data is available as part of the supplementary information.
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