1 Three distinct Holocene intervals of stalagmite deposition and non-

deposition revealed in NW Madagascar, and their paleoclimate

implications

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ABSTRACT

Petrographic features, mineralogy, and stable isotopes from two stalagmites, ANJB-2 and MAJ-5, respectively from Anjohibe and Anjokipoty Caves, allow distinction of three intervals of the Holocene in NW Madagascar. The Malagasy *early* Holocene (between c. 9.8 and 7.8 ka) and *late* Holocene (after c. 1.6 ka) intervals (MEHI and MLHI, respectively) record evidence of stalagmite deposition. The Malagasy *middle* Holocene interval (MMHI, between c. 7.8 ka and 1.6 ka) is marked by a depositional hiatus of c. 6500 years.

Deposition of these stalagmites indicates that the two caves were sufficiently supplied with water to allow stalagmite formation. This suggests that the MEHI and MLHI intervals may have been comparatively wet in NW Madagascar. In contrast, the long-term depositional hiatus during the MMHI implies it was relatively drier than the MEHI and the MLHI.

The alternating "wet/dry/wet" conditions during the Holocene may have been linked to the long-term migrations of the Inter-Tropical Convergence Zone (ITCZ). When the ITCZ's mean position is farther south, NW Madagascar experiences wetter conditions, such as during the MEHI

and MLHI, and when it moves north, NW Madagascar climate becomes drier, such as during the MMHI. A similar wet/dry/wet succession during the Holocene has been reported in neighboring locations, such as southeastern Africa. Beyond these three subdivisions, the records also suggest wet conditions around the cold 8.2 ka event, suggesting a causal relationship. However, additional Southern Hemisphere high—resolution data will be needed to confirm this.

1. Introduction

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Although much is known about Holocene climate change worldwide (Mayewski et al., 2004; Wanner and Ritz, 2011; Wanner et al., 2011; 2015), high-resolution climate data for the Holocene period is still regionally limited in the Southern Hemisphere (SH) (e.g., Wanner et al., 2008; Marcott et al., 2013; Wanner et al., 2015), including Madagascar. This uneven distribution of data hinders our understanding of the spatio-temporal characteristics of Holocene climate change, and the forcings involved. For example, some forcings would have influenced the behavior of the Inter-Tropical Convergence Zone (ITCZ) as well as monsoonal responses in low- to mid-latitude regions (e.g., Wanner et al., 2015; Talento and Barreiro, 2016). In fact, Madagascar is ideally located to provide data on SH Holocene climate changes because of its location in Southwestern Indian Ocean and because it is seasonally visited by the ITCZ (Fig. 1a). Furthermore, a karst belt with caves extends from the north to the south of the island (Fig. 1c), crossing latitudinal climate belts, and this could potentially be a source of stalagmite data. Thus, Madagascar is a natural laboratory to study ITCZ dynamics over time. New records from Madagascar could fill gaps in paleoclimate datasets for the SH that might help refine paleoclimate simulations, and thus provide a better understanding of global circulation and land-atmosphere-ocean interactions during the Holocene.

In this paper, we present records of stable isotopes, petrography, mineralogy, variability of layer-specific width (or LSW) from stalagmites from Anjohibe and Anjohipoty caves. Stalagmites are used because of their potential to store significant climatic information (e.g., Fairchild and Baker, 2012, p. 9–10), and in Anjohibe Cave, recent studies have shown the replicability of paleoclimate records from stalagmites (e.g., Burns et al., 2016).

Two stalagmites were investigated, and these allowed us to characterize Holocene climate change in NW Madagascar. First, we developed a record of climate change using the stalagmite

proxy data. With a better understanding of Madagascar's paleoclimate, we then investigated possible drivers of tropical climate change to isolate the major factors controlling the hydrological cycle in NW Madagascar and surrounding regions during the Holocene.

2. Setting

2.1. Stalagmites and their setting

Stalagmites are secondary cave deposits that are $CaCO_3$ precipitates from cave dripwater. Calcium carbonate precipitation occurs mainly by CO_2 degassing, which increases the pH of the dripwater and thus increases the concentration of CO_3^{2-} . In some cases, evaporation may also contribute to increased Ca^{2+} and/or CO_3^{2-} concentrations in dripwater. CO_2 degassing occurs when high- PCO_2 water from the epikarst encounters low- PCO_2 cave air. Evaporation occurs when humidity inside the cave is relatively low. The fundamental equation for stalagmite deposition is:

$$Ca_{(aq)}^{2+} + 2HCO_{3(aq)}^{-} \rightleftharpoons CaCO_{3(s)} + CO_{2(g)} + H_2O_{(l)}$$
 (Eq. 1)

Growth and non-growth of stalagmites depends on conditions that affect Eq. 1. An increase in Ca^{2+} drives the equation to the right (towards precipitation) and an increase in CO_2 of the cave air and/or H_2O drives it to the left (towards dissolution). All components of the equation are influenced by the supply of water to the cave, which is generally climate-dependent. More water enters the cave during warm/rainy seasons than during cold/dry seasons. Stalagmites will form when cave dripwater is saturated with respect to calcite and/or aragonite. If the water passes through the bedrock too quickly to dissolve significant carbonate rock, and/or enters the cave and reaches the stalagmite too quickly to degas significant CO_2 , it will not be saturated with respect to $CaCO_3$, inhibiting stalagmite formation. Stalagmite growth will slow as dripwater declines and will stop entirely if flow ceases. Vegetation provides CO_2 to the soil via root respiration so the vegetation cover above the cave and the type of vegetation can promote or limit stalagmite growth. Overall, the karst hydrological system plays a crucial role in the deposition and non-deposition of stalagmites, and this is closely linked to changes in local and regional environment and climate.

2.2. Regional environmental setting

Stalagmites ANJB-2 and MAJ-5 were collected from Anjohibe and Anjokipoty caves, respectively, in the Majunga region of NW Madagascar (Fig. 1). Sediments and fossils from these caves have already provided many insights about the paleoenvironmental and archaeological history of NW Madagascar (e.g., Burney et al., 1997, 2004; Brook et al., 1999; Gommery et al., 2011; Jungers et al., 2008; Vasey et al., 2013; Burns et al., 2016; Voarintsoa et al., 2017b).

Anjohibe (S15° 32' 33.3"; E046° 53' 07.4") and Anjokipoty (S15° 34' 42.2"; E046° 44' 03.7") are about 16.5 km apart (Fig. 1c). Their location in the zone visited by the ITCZ (e.g., Nassor and Jury, 1998) makes them ideal sites to test the hypothesis that latitudinal migration of the ITCZ influenced the Holocene climate of NW Madagascar (e.g., Chiang and Bitz, 2005; Broccoli et al., 2006; Chiang and Friedman, 2012; Schneider et al., 2014). Majunga has a tropical savanna climate (Aw) according to the Köppen-Geiger climate classification, with a distinct wet summer (from October to April) and dry winter (May-September). The mean annual rainfall is around 1160 mm. The mean maximum temperature in November, the hottest month in the summer, is about 32°C. The mean minimum temperature in July, the coldest month of the dry winter, is about 18°C (Fig. 1b).

2.3. Climate of Madagascar

The climate of Madagascar is unique because of its varied topography and its position in the Indian Ocean (Figs. S1–S2; also see for e.g., Jury, 2003; DGM, 2008, Douglas and Zinke, 2015, p. 281-299; Voarintsoa et al., 2017b, p.138-139; Scroxton et al., 2017). Regionally distinct rainfall gradients from east to west and from north to south are evident across the country (Jury, 2003; Dewar and Richard, 2007), and these are linked to easterly trade winds in winter (May-October) and northwesterly tropical storms in summer, respectively. In NW Madagascar, summer rainfall is monsoonal and it is in phase with the seasonal southward migration of the ITCZ. Chiang and Bitz' (2005) and Broccoli et al.'s (2006)'s have suggested that cooler/warmer intervals bring the ITCZ south/north, thus regions in tropical SH are wet/dry. Generally, the ITCZ migrates towards the Earth's warmer hemisphere (Frierson and Hwang, 2012; Kang et al., 2008; McGee et al., 2014; Sachs et al., 2009). In fact, longer-term ITCZ migration appears to have affected climate in NW

Madagascar between c. 370 CE and 800 CE (see Fig, 8 of Voarintsoa et al., 2017b). This relationship was inferred from changes in global climate conditions.

The climate of Madagascar is also influenced by changes in Indian Ocean sea surface temperature (SST) (Zinke et al., 2004; see also Kunhert et al., 2014) and changes in SST the Aghulas Current off southwestern Madagascar (Lutjeharms, 2006; Beal et al., 2011; Zinke et al., 2014). The most immediate signal is the Indian Ocean Dipole (IOD), or Indian Ocean Zonal Mode (Li et al., 2003; Zinke et al., 2004), but El-Niño Southern Oscillation (ENSO) may also influence its climate (e.g., Brook et al., 1999). IOD has been linked to Holocene climate variability in tropical Indian Ocean (Abram et al., 2009; Tierney et al., 2013). However, its linkages to ENSO is still debated (e.g., Saji et al., 1999; Li et al., 2003; Lee et al., 2008; Brown et al., 2009; Schott et al., 2009; Shinoda et al., 2004; Venzke et al., 2000; Abram et al., 2008; Saji and Yagamata, 2003; Meyers et al., 2007). The complex interactions between these inter-annual climatic factors make them an ideal topic for further investigation using high-resolution records, and thus they will not be the focus of this paper. However, their possible effects are referred to briefly in Supplementary text no. 4.

2.4. The Holocene in NW Madagascar

Little is hitherto known about Holocene climate change in NW Madagascar nor about the major drivers of long-term climatic changes there. Most paleoclimate information from this region covers the last two millennia with more focus on the anthropogenic effects on the Malagasy ecosystems (e.g., Crowley and Samonds, 2013; Burns et al., 2016; Voarintsoa et al., 2017b). This is because several studies show that megafaunal extinctions in Madagascar coincide with the arrival of humans around 2-3 thousand (ka) BP (e.g., see Table 1 of Virah-Sawmy et al., 2010; MacPhee and Burney, 1991; Burney et al., 1997; Crowley, 2010). There are even fewer long-term paleoclimate records for the NW region, with only sediments from Lake Mitsinjo (3.5 ka BP; Matsumoto and Burney, 1994) and stalagmites from Anjohibe Cave (40 ka BP; Burney et al. 1997) providing records of more than 3 ka. Even though these records provide useful information about paleoenvironmental changes in NW Madagascar, links to global climatic changes, particularly the links to changes in ITCZ, are not yet fully understood.

3. Methods

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3.1. Radiometric dating

A total of 22 samples were drilled from Stalagmite ANJB-2 and 9 samples for Stalagmite MAJ-5 for U-series dating (Table S1 and S2). Each sample is a long (~5 to 20 mm), narrow (~1-2mm), and shallow (~1 mm) trench, allowing us to extract 50-250 mg of CaCO₃ powder. We followed the chemical procedures described in Edwards et al. (1987) and Shen et al. (2002) when separating uranium and thorium. U/Th measurements were performed on the multi-collector ICP-MS of the University of Minnesota, USA and on a similar instrument in the Stable Isotopes Laboratory of Xi'an, in Jiaotong, China. Instrument details are provided in Cheng et al. (2013). Corrected ²³⁰Th ages assume an initial ²³⁰Th/²³²Th atomic ratio of $4.4 \pm 2.2 \times 10^{-6}$. This is the ratio for "bulk earth" or crustal material at secular equilibrium with a 232Th/238U value of 3.8. The uncertainty in the "bulk earth" value is assumed to be ±50% (see footnotes to Tables S1 and S2). The error in the final "corrected age" incorporates this uncertainty. The radiometric data are reported as years BP, where BP is Before Present, and "Present" is A.D. 1950. Stalagmite chronologies were constructed using the StalAge1.0 algorithm of Scholz and Hoffman (2011) and Scholz et al. (2012), an algorithm using a Monte-Carlo simulation. The algorithm can identify major and minor outliers and age inversions. The StalAge scripts were run on the statistics program R version 3.2.2 (2015-08-14). The age models were adjusted considering hiatal surfaces identified in the samples, using the approach of Railsback et al. (2013; see their Fig. 9).

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3.2. Petrography and mineralogy

Petrography and mineralogy of the two stalagmites were investigated 1) by examining both the polished surfaces and the scanned images of the sectioned stalagmites, and by identifying any diagenetic fabrics (e.g., Zhang et al., 2014) that could potentially affect stable isotope values, 2) by observing eleven oversized thin sections (3x2 in) under the Leitz Laborlux 12 Pol microscope and the Leica DMLP equipped with QCapture in the Sedimentary Geochemistry Lab at the University of Georgia, 3) by using scanning electron microscopy (SEM) to better understand the mineralogical fabrics at locations of interest (Fig. S13), and 4) by analyzing about 30–100 mg of powdered spelean layers (n=15) on a Bruker D8 X-ray Diffractometer in the Department of

Geology, University of Georgia. For calcite and aragonite identification, we used $CoK\alpha$ radiation at a 2θ angle between 20° and 60° .

Layer-specific width (LSW) of clearly-defined layers was measured at selected locations on the stalagmite polished surfaces (Fig. S4; Sletten et al., 2013; Railsback et al., 2014; Voarintsoa et al., 2017b). LSW is the horizontal distance between two points on the flanks of the stalagmite where convexity is greatest. It is the width near the top of the stalagmite when the layer being examined was deposited. LSW is measured at right angles to the growth axis of the stalagmite; it is the horizontal distance between two points on the layer growth surface, at which a virtual line inclined at 35° to the growth axis becomes tangent to the layer growth surface as shown in Fig. S4. LSW may vary along the length of the stalagmite, with smaller values suggesting drier conditions and larger values wetter conditions.

3.3. Stable isotopes

Samples of 50–100 μg were drilled along the stalagmite's growth axis for stable isotope analysis. The trench size is very small (1.5 x 0.5 x 0.5 mm). Since a small mixture of calcite and aragonite could potentially change the $\delta^{18}O$ and $\delta^{13}C$ of the measured spelean layers (see for example Frisia et al., 2002), drilling and sample extraction were carefully done on individually discrete layers using the smallest drill-bit head (SSW-HP-1/4) to avoid potential mixing between calcite and aragonite. The polished surface of the two stalagmites were examined to see if features of diagenetic alteration are present (see for example fig. 2 of Zhang et al., 2014), but none was found. During sampling, the mineralogy at the crest, where stable isotope samples were extracted, was recorded for future mineralogical correction.

Aragonite oxygen and carbon isotopic corrections were performed to compensate for aragonite's inherent fractionation of heavier isotopes (e.g., Romanek et al., 1992; Kim et al., 2007; McMillan et al., 2005) and to remove the mineralogical bias in isotopic interpretation between calcite and aragonite. The correction consists of subtracting 0.8‰ for δ^{18} O (Kim and O'Neil, 1997; Tarutani et al., 1969; Kim et al., 2007; Zhang et al., 2014) and 1.7 ‰ for δ^{13} C (Rubinson and Clayton, 1969; Romanek et al., 1992) for the aragonite, as has been done previously (e.g., Holmgren et al.,

2003; Sletten et al., 2013; Liang et al., 2015; Railsback et al., 2016; Voarintsoa et al., 2017a), as shown in equations 2 and 3 below (where $R_{A/C}$ is the aragonite percentage if not 100%).

 $\delta^{18}O_{corr.}$ (%, VPDB) = $\delta^{18}O_{uncorr.}$ (%, VPDB) – [R_{A/C} x 0.8 (%, VPDB)] (Eq. 2)

 $\delta^{13}C_{corr.}$ (%, VPDB) = $\delta^{13}C_{uncorr.}$ (%, VPDB) – [R_{A/C} x 1.7 (%, VPDB)] (Eq. 3)

Supplementary Figures S6–S8 show both the corrected and uncorrected isotopic records.

For the analytical methods, oxygen and carbon isotope ratios were measured using the Finnigan MAT-253 mass spectrometer fitted with the Kiel IV Carbonate Device of the Xi'an Stable Isotope Laboratory in China (ANJB-2; n=654) and using the Delta V Plus at 50°C fitted with the GasBench-IRMS machine of the Alabama Stable Isotope Laboratory in USA (MAJ-5; n=286). Analytical procedures using the MAT 253 are identical to those described in Dykoski et al. (2005), with isotopic measurement errors of less than 0.1 % for both δ^{13} C and δ^{18} O. Analytical methods and procedures using the GasBench-IRMS machine are identical to those described in Skrzypek and Paul (2006), Paul and Skrzypek (2007), and Lambert and Aharon (2011), with ±0.1 % errors for both δ^{13} C and δ^{18} O. In both techniques, the results are reported relative to Vienna PeeDee Belemnite (VPDB) and with standardization relative to NBS19. An inter-lab comparison of the isotopic results was conducted, and it involved replicating every tenth sample of Stalagmite MAJ-5 at both labs. This exercise showed a strong correlation between the lab results (Fig. S5).

4. Results

4.1. Radiometric data

Results from radiometric analyses of the two stalagmites are presented in Tables S1 and S2. Corrected 230 Th ages suggest that Stalagmite ANJB-2 was deposited between c. 8977±50 and c. 161±64 years BP, and Stalagmite MAJ-5 was deposited between c. 9796±64 and c. 150±24 years BP. These ages collectively indicate stalagmite deposition at the beginning (between c. 9.8 and 7.8 ka BP) and at the end of the Holocene (after c. 1.6 ka BP). In both stalagmites, the older ages have small 2σ errors and they generally fall in correct stratigraphic order, except sample ANJB-2-120 and its replicate ANJB-2-120R, which were not used because of the sample's high porosity and high detritals content. In contrast, many of the younger ages have larger uncertainties. This is mainly because many of the younger samples have very low uranium concentration and the

detrital thorium concentration is also high, similar to what Dorale et al. (2004) reported. We also understand that the value for initial 230 Th correction, i.e. the initial 230 Th/ 232 Th atomic ratio of 4.4 \pm 2.2 \times 10⁻⁶ for a bulk earth with a 232 Th/ 238 U value of 3.8, in these samples could have slightly altered the 230 Th age of these younger samples, leading to larger uncertainties (such as discussed in Lachniet et al., 2012). We encountered similar problems while working on other younger samples from the same cave, but we compared the stable isotope profile with other published records using isochron dating methods, and results did not differ significantly (see Fig. 9 of Voarintsoa et al., 2017b). Since this work does not focus on decadal or centennial interpretation of the Late Holocene stable isotope data, additional chronology adjustment has not been made, and we used the chronology from StalAge to construct the time series. However, in Figures 5 and 6, age uncertainties are given below the stable isotope profiles so that comparisons with other records can accommodate these uncertainties.

The key finding from our age and petrographic data for the two stalagmites is that they indicate three distinct intervals of growth and non-growth during the Holocene (Figs. 2–4, 7). The evidence for this includes: (1) CaCO₃ deposition between c. 9.8 and 7.8 ka BP, (2) a long depositional hiatus between c. 7.8 and 1.6 ka BP, and (3) resumption of CaCO₃ deposition after c. 1.6 ka BP. In the rest of the paper, we will refer to these intervals as the Malagasy Early Holocene Interval (MEHI), Malagasy Mid-Holocene Interval (MMHI), and Malagasy Late Holocene Interval (MLHI), respectively.

4.2. Stable isotopes

Raw values of δ^{18} O and δ^{13} C for Stalagmite ANJB-2 range from -8.9 to -2.3% (mean = -5.0%), and from -11.0 to +5.2% (mean = -4.2%), respectively, relative to VPDB. Raw values of δ^{18} O and δ^{13} C for Stalagmite MAJ-5 range from -8.8 to -0.9% (mean = -4.9%), and from -9.4 to +2.6% (mean = -4.4%), respectively, relative to VPDB. Mean δ^{18} O and δ^{13} C values are distinguishable between the MEHI and the MLHI. In both stalagmites, the amplitude of δ^{18} O fluctuations was fairly constant throughout the Holocene; whereas the δ^{13} C records show a dramatic shift toward higher values (i.e. from -10.9% to +3.8%, VPDB) at c. 1.5 ka BP.

The MEHI and MLHI are isotopically distinct (Fig. 4). The MEHI is characterized by statistically correlated δ^{18} O and δ^{13} C (r^2 =0.65 and 0.53), and much depleted δ^{13} C values (c –11.0 to –4.0 ‰). A prominent isotopic excursion is evident between c. 8.1 and c. 8.3 ka BP (Fig. 5), when stalagmite δ^{18} O and δ^{13} C ratios reach their lowest values of –6.8 and –10.9‰, respectively. In contrast to the MEHI, the values of δ^{18} O and δ^{13} C during the MLHI are poorly correlated (r^2 =0.25 and 0.17), and δ^{13} C values are more enriched (Figs. 4, 6). Since Stalagmites ANJB-2 and MAJ-5 were collected from two different caves, 16 km apart, it is not surprising to see discrepancies between the stable isotopes during similar intervals, suggesting that local karst conditions could be one of the discrepancy factors. Another potential source for the discrepancy is the larger uncertainty of the younger ages due to low uranium and high detrital concentrations. This U-Th aspect has been a challenge for several young stalagmites (e.g., Dorale et al., 2004; Lachniet et al., 2012) including samples from NW Madagascar (this study). While the utility of speleothems as a climate proxy largely depends on replication of stable isotope values, this study specifically highlights the replication of stalagmite deposition and non-deposition and the isotopic characteristics of each depositional intervals of the Holocene.

4.3. Mineralogy, petrography, and layer-specific width

In both stalagmites ANJB-2 and MAJ-5, the hiatus of deposition is characterized by a well-developed Type L surface (Figs. 2, 3, S15). Petrography and mineralogy are distinct before and after this hiatus (Figs. 3, 5–6). Below the hiatus, laminations are well preserved in both stalagmites. Above the hiatus, laminations are not well-preserved, although noted in some intervals.

In Stalagmite ANJB-2, LSW varies from 37 to 26.5 mm with a mean of 30 mm. It decreases to 28 mm at the hiatus (Fig. 3). The mineralogy is dominated by aragonite below the hiatus, although there are also a few thick layers of primary calcite. A thin (~2-3 mm) layer of white, very soft, and porous aragonite is identified just below the hiatus (Fig. S15). This layer is also calcite and aragonite, with calcite dominant, and the calcite layers contain macro-cavities that are mostly off-axis macroholes (Shtober-Zisu et al., 2012).

As noted in previous section (4.2), there is a prominent isotopic excursion at c. 8.2 ka BP, and this excursion is in the calcite layer in Stalagmite ANJB-2 at 195–202 mm from its top. X-ray

diffraction spectrum from this layer suggests that the mineralogy is 100% calcite (Figs. S14, S16—S17). We believe the calcite to be primary and not a diagenetic product of aragonite for three reasons. First, the laminations in the thick layer of calcite were not altered (Figs. S16—S17). Second, the polished surface of the stalagmite shows no evidence of fiber relicts and textural ghosts such as observed in Juxtlahuaca Cave in southwestern Mexico (Lachniet et al., 2012) and in Shennong Cave in southeastern China (Zhang et al., 2014). Third, petrographic comparison with known examples of primary and secondary calcite observed under microscope (e.g., Railsback, 2000; Perrin et al., 2014) suggests that there is no strong evidence of aragonite—to—calcite transformation.

In Stalagmite MAJ-5, LSW varies from 50 to 22 mm with a mean of 35.5 mm. It decreases to 22 mm at the hiatus (Fig. 3). Below the hiatus, mineralogy is an even mixture between calcite and aragonite. Above the hiatus, mineralogy is mainly calcite, except the uppermost 2 mm where mineralogy is 75% calcite and 25% aragonite. Macro-cavities are also present throughout this upper part of Stalagmite MAJ-5.

4.4. Summary of results

The various proxy climate records from Stalagmites ANJB-2 and MAJ-5 suggest three distinct climate/hydrological intervals during the Holocene. The MEHI (c. 9.8 to 7.8 ka BP), with evidence of stalagmite deposition, is characterized by statistically correlated δ^{18} O and δ^{13} C (r^2 =0.65 and 0.53) and more negative δ^{13} C values (c. –11.0 to –4.0 %). The MMHI (c. 7.8 to 1.6 ka BP) is marked by a long-term hiatus in deposition, which is preceded by a well-developed Type L surface in both Stalagmite ANJB-2 and MAJ-5 (Figs. 3, S15). The Type L surface is observed as an upward narrowing of stalagmite width and layer thickness. It is best developed in Stalagmite MAJ-5 (Fig. S15). In Stalagmite ANJB-2, the hiatus at the Type L surface is additionally preceded by a c. 3 mm thick layer of highly porous, very soft, and fibrous white crystals of aragonite (the only aragonite with such properties). This aragonite is topped by a thin and well-defined layer of detrital materials (Fig. S15), further evidence of a hiatus. Finally, the MLHI (after c. 1.6 ka BP) is characterized by poorly correlated δ^{18} O and δ^{13} C (r^2 =0.25–0.17), and by a marked shift toward higher δ^{13} C values (Figs. 4, 6).

5. Discussion

5.1. Paleoclimate significance of stalagmite growth and non-growth: implications for paleohydrology

Growth and non-growth of stalagmites depends on several factors linked to water availability, which is largely determined by climate (more water during warm/rainy seasons and less water during cold/dry seasons). Water is the main dissolution and transporting agent for most chemicals in speleothems. Cave hydrology varies significantly over time in response to climate, and this variability influences the formation or dissolution of CaCO₃. In this regard, calcium carbonate does not form if there is little or no water entering the cave, or if there is too much (see Sect. 2.1). Absence of groundwater recharge most typically occurs during extremely dry conditions, whereas excessive water input to the cave occurs during extremely wet conditions. In the latter scenario, water is undersaturated and flow rates are too fast to allow degassing. Often, water availability is reflected in the extent of vegetation above and around the cave, as plants require soil moisture or shallow groundwater to survive and propagate, and this contributes to the stalagmites' processes of formation. The link between stalagmite growth/non-growth and cave dripwater and soil CO₂ is broadly influenced by changes in climate.

Major hiatuses in stalagmite deposition could be marked by a variety of features, including the presence of erosional surfaces, chalkification, dirt bands/detrital layers, offsetting of the growth axis, and/or sometimes by color changes (e.g., Holmgren et al., 1995; Dutton et al., 2009; Railsback et al., 2013; Railsback et al., 2015; Voarintsoa et al., 2017a). Railsback et al. (2013) were able to identify significant features in stalagmites that allow distinction between non-deposition during extremely wet (Type E surfaces) and non-deposition during extremely dry conditions (Type L surfaces; Fig. 3). Physical properties of stalagmites that are evidence of extreme dry and wet events are summarized in Table 1 of Railsback et al. (2013) and the mechanisms are explained in their Figure 5.

Type E surfaces are layer-bounding surfaces between two spelean layers when the underlying layers show evidence of truncation. The truncation results from dissolution or erosion (thus the name "E") of previously-formed stalagmites layers by abundant undersaturated water.

Type E surfaces are commonly capped with a layer of calcite (Railsback et al., 2013). This mineralogical trend is not surprising as calcite commonly forms under wetter conditions (e.g., Murray, 1954; Pobeguin, 1965; Siegel, 1965; Thrailkill, 1971; Cabrol and Coudray, 1982; Railsback et al. 1994; Frisia et al., 2002). Additionally, non-carbonate detrital materials are commonly abundant with varying grain size (i.e., from silt- to sand-size; Railsback et al., 2013).

Type L surfaces, on the other hand, are layer-bounding surfaces where the layers become narrower upward and thinner towards the flanks of the stalagmite. Decreases in layer thickness and stalagmites width upward are indications of lessening deposition (thus the name "L"; Railsback et al., 2013). Aragonite is a very common mineralogy below a Type L surface, especially in warmer settings. Layers of aragonite commonly form under drier conditions (Murray, 1954; Pobeguin, 1965; Siegel, 1965; Thrailkill, 1971; Cabrol and Coudray, 1982; Railsback et al., 1994; Frisia et al., 2002). Non-carbonate detrital materials are scarce, and if present, they tend to form a very thin horizon of very fine dust material (Railsback et al., 2013). Identification of Type L surfaces is aided by measuring the LSW (e.g., Sletten et al., 2013; Railsback et al., 2014; Fig. S4).

5.2. Holocene climate in NW Madagascar

The age models and petrographic features of Stalagmites ANJB-2 and MAJ-5 suggest three distinct Holocene climate intervals (MEHI, MMHI, and MLHI; see Sect. 4.1) in NW Madagascar. Possible conditions during these intervals are illustrated in the sketches of Figure 4.

5.2.1. Malagasy early Holocene interval (c. 9.8 –7.8 ka BP)

Stalagmite deposition during the early Holocene suggests that the chambers where stalagmites ANJB-2 and MAJ-5 were collected were sufficiently supplied with water to allow CaCO₃ precipitation, in accord with Eq.1. This in turn implies relatively wet conditions that could indicate longer summer rainy seasons relative to modern climate, or wet years in NW Madagascar. The correlative δ^{13} C and δ^{18} O values further suggest that vegetation consistently responded to changes in moisture availability, which in turn was dependent on climate.

The prominent negative δ^{18} O and δ^{13} C excursions in Stalagmite ANJB-2 (Sect. 4.2; Figs. 5 and 10) are parallel to the δ^{18} O excursion of the Greenland ice core records at c. 8.2 ka BP (e.g.

Alley et al., 1997). The decrease in δ^{18} O and δ^{13} C values and the presence of calcite mineralogy at the same interval combine to suggest a wet 8.2 ka BP event in NW Madagascar. The 8.2 ka BP event was triggered by a release of freshwater from the melting Laurentide Ice Sheet into the North Atlantic basin, bringing cooler conditions in several NH regions (e.g., Alley et al., 1997; Barber et al., 1999), and via global teleconnections, this may have affected climate in NW Madagascar (see Sect. 5.5).

The MEHI terminated when conditions became much drier, as suggested by increasing δ^{18} O and δ^{13} C values in Stalagmite ANJB-2, by decreasing LSW in both stalagmites, and by the presence of a major Type L surfaces in both stalagmites. The thin (c. 3 mm), porous, and white aragonite layer in Stalagmite ANJB-2, a very similar deposit to that described in Niggemann et al. (2003), suggests that the terminal drought was at times severe. Aragonite is a CaCO₃ polymorph that forms preferentially under drier conditions (Murray, 1954; Pobeguin, 1965; Siegel, 1965; Thrailkill, 1971; Cabrol and Coudray, 1982; Railsback et al. 1994; Frisia et al., 2002). The porous aragonite layer in Stalagmite ANJB-2 is capped by a very thin layer of non-carbonate, brown detritus, which may have been transported to the stalagmite as an aerosol and accumulated on the dry stalagmite surface over time. Accumulation of the detritus must take place in the absence of dripwater (e.g., Railsback et al., 2013). A shift to drier conditions is also supported by isotopic data from Stalagmite ANJ94-5 from Anjohibe Cave (Wang and Brook, 2013; Wang, 2016) in which relatively low δ^{13} C and δ^{18} O values prior to 7.6 ka BP give way to episodically greater values thereafter.

5.2.2. Malagasy mid-Holocene interval (c. 7.8–1.6 ka BP)

The MMHI was a long (~6.5 ka) depositional hiatus in both stalagmites (Figs. 2–3), potentially suggesting dry conditions. The question is why did neither stalagmite grow during the MMHI? Here, we try to explain the factors and the climatic conditions that may have been responsible.

The documented severe dry conditions at the end of the MEHI (see Sect. 5.2.1) could have had a significant influence (1) on the cave hydrological system (e.g., Fig. 5 of Asrat et al., 2007; Bosák, 2010), such as the water conduits (primary or secondary porosity) to the chambers, and (2) on the vegetation cover above the caves, particularly above the chambers where Stalagmites

ANJB-2 and MAJ-5 were collected. On one hand, it is possible that the dry conditions late in the MEHI could not only bring lesser water recharge to the cave, but also lowered the hydraulic head, and increased the rate of evapo-transpiration in the vadose zone. This condition possibly allowed more air to penetrate the aquifer, perhaps enhancing prior carbonate precipitation (PCP) in pores and conduits above the caves (e.g., Fairchild and McMillan, 2007; Fairchild et al., 2000; Johnson et al., 2006; Karmann et al., 2007; McDonald et al., 2007). This process must have blocked water moving towards Stalagmites ANJB-2 and MAJ-5. On the other hand, the late MEHI drying trend (Sect. 5.2.1) could have challenged vegetation to grow, and we assume that some areas above Anjohibe and Anjokipoty caves must have been devoid of vegetation. Consequently, biomass activities could have been reduced. Because vegetation contributes CO₂ to the carbonic acid dissolving CaCO₃, its absence in certain areas above the cave could decrease the pH of the percolating water, and perhaps dissolution did not occur. Under these conditions, even if water reached the stalagmites, it may not have precipitated carbonate.

Whatever factors were responsible for the long term-depositional hiatus in Stalagmite ANJB-2 and MAJ-5, we believe that the hiatus was caused by disturbances to water catchments that feed the chambers at Anjohibe and Anjokipoty caves. The disturbances could be inherited from the very dry conditions at the end of the MEHI, and/or due to the lack of water supply, perhaps associated with an increase in epikarst ventilation, and/or by the absence of vegetation. Water and vegetation are two components of the karst system that play an important role in CaCO₃ dissolution and precipitation (see Eq. 1). Their disturbance may have limited limestone dissolution in the epikarst and then carbonate precipitation in the cave zone.

Other evidence supports the idea of at least episodic dryness during the MMHI. A work on a 2-meter long stalagmite (ANJ94-5) from Anjohibe Cave suggests episodic dryness during the MMHI and a depositional hiatus around the time when Stalagmites ANJB-2 and MAJ-5 stopped growing (Wang and Brook, 2013; Wang, 2016). For regional comparison, dry spells were also felt in Central and Southeastern Madagascar (e.g., Gasse and Van Campo, 1998; Virah-Sawmy et al., 2009).

In summary, several lines of evidence suggest relatively drier climate in NW Madagascar during the MMHI compared to the MEHI. Drier intervals generally imply drier summer seasons

with less rainfall (Fig. 8), perhaps reflecting shorter visits by the ITCZ. In this regard, even though the region received rainfall, the necessary conditions could not have been attained to activate the growth of Stalagmites ANJB-2 and MAJ-5, thus the hiatuses.

5.2.3. Malagasy Late Holocene Interval (c. 1.6 ka BP–present)

Resumption of stalagmite deposition after c. 1.6 ka BP suggests a wetter climate in NW Madagascar with reactivation of the previous epikarst hydrologic system. Climatic conditions must have been similar to those of the early Holocene. The sudden beginning of stalagmite growth during the MLHI and the large δ^{13} C shift from depleted to enriched values at c. 1.5 ka BP (Fig. 6), after such long hiatuses may have been associated with changes in vegetation cover above the cave linked to human activities (e.g., Burns et al., 2016; Crowley and Samonds, 2013; Crowther et al., 2016; Voarintsoa et al., 2017b). Lower δ^{13} C values in Stalagmite MAJ-5 after 0.8 ka BP (Fig. 3), compared to higher values in Stalagmite ANJB-2, may suggest different local karst conditions, either natural, human-induced, or something else, at each site. Further investigations will be necessary to better understand this.

5.3. Holocene climate in NW Madagascar: implications for ITCZ dynamics

In NW Madagascar, stalagmite deposition during the MEHI and the MLHI suggests there was sufficient dripwater for stalagmite growth and therefore wetter conditions. This may indicate a more southerly mean position of the ITCZ. Factors that could influence the mean position of the ITCZ include changes in insolation (e.g., Haug et al., 2001; Wang et al., 2005; Cruz et al., 2005; Fleitmann et al., 2003, 2007; Schefuß et al., 2005; Suziki, 2011; Kutzbach and Liu, 1997; Partridge et al., 1997; Verschuren et al., 2009; Voarintsoa et al., 2017a) and difference in temperature between the two hemispheres (e.g., Chiang and Bitz, 2005; Broccoli et al., 2006; Chiang and Friedman, 2012; Kang et al., 2008; McGee et al., 2014; Talento and Barreiro, 2016).

In contrast, the depositional hiatuses during the MMHI could suggest overall drier conditions, and thus a northward migration of the mean ITCZ. It may agree with the paleoclimate simulation of Braconnot et al. (2007), although the simulation is of shorter term than the MMHI hiatus, but additional paleoclimate records are needed to improve its spatial and temporal

resolution. A northward shift in the mean position of the ITCZ is consistent with drier conditions in the southern tropics, e.g., weaker South American Summer Monsoon (Cruz et al., 2005; Seltzer et al., 2000; Wang et al., 2007; but see also Fig. 9 of Zhang et al., 2013), and with wetter conditions in the northern tropics (e.g., Dykoski et al., 2005; Fleitmann et al., 2007; Gasse, 2000; Haug et al., 2001; Weldeab et al., 2007; Zhang et al., 2013).

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5.4. Regional comparisons

Records from neighboring locations (Figs. 8–9; Table S3) show that the Holocene wet/dry/wet succession reported here for NW Madagascar also affected other locations. For example, hydrogen isotope compositions of the n-C31 alkane in GeoB9307-3 from a 6.51 m long marine sediment core retrieved about 100 km off the Zambezi delta show a similar wet/dry/wet climate during Early, Middle, and Late Holocene respectively (Schefuß et al., 2011). These changes correspond to changes in temperature from ~26.5° to 27.25° to 27°C, respectively, in the Mozambique Channel, as suggested by alkenone SST records from sediment cores MD79257 (Bard et al., 1997; Sonzogni et al., 1998). The Zambezi catchment is specifically relevant here because it is located at the southern boundary of the modern ITCZ, and so has a similar climatic setting as NW Madagascar, and its sensitivity to the latitudinal migration of the ITCZ could parallel that of Madagascar. Likewise, temperature reconstruction from the Mozambique Channel could be used to link regional changes in paleorainfall with regional changes in temperature. A general overview of the Holocene climate in the African neighboring locations to Madagascar suggests a roughly consistent wetter and drier climate during the early and middle Holocene, respectively (Fig. 9, Table S3, also see Gasse, 2000; Singarayer and Burrough, 2015). However, Late Holocene paleoclimate reconstructions vary. A simple explanation to this Late Holocene variability is unlikely, but several interacting factors, including the latitudinal migration of the ITCZ, changes in ocean oscillations and sea surface temperatures, volcanic aerosols, and anthropogenic influences may have played a role (e.g., Nicholson, 1996; Gasse, 2000; Tierney et al., 2008; Truc et al., 2013). Assessing these factors is beyond the scope of this study.

5.5. The 8.2 ka BP event in Madagascar: linkage to ITCZ and AMOC

The 8.2 ka BP event, a widespread cold event in the NH (e.g., Alley et al., 1997), is apparent in the stalagmite records (Figs. 5, 10). Stalagmite ANJB-2 δ^{18} O and δ^{13} C ratios reach their lowest values of –6.8 and –10.9‰, respectively during that interval, and mineralogy is primary calcite. These proxies suggest wet interval in NW Madagascar.

The 8.2 ka event was triggered by an abrupt freshwater influx from the melting Laurentide Ice Sheet into the North Atlantic (Alley et al., 1997; Barber et al., 1999; Kleiven et al., 2008; Carlson et al., 2008; Renssen et al, 2010; Wiersma et al., 2011; Wanner et al., 2015). This influx of meltwater altered the density and salinity of the NADW (e.g., Thornalley et al., 2009), weakening the Atlantic Meridional Overturning Circulation (AMOC, e.g., Barber et al., 1999; Clark et al., 2001; Daley et al., 2011; Vellinga and Wood 2002; Dong and Sutton 2002, 2007; Dahl et al. 2005; Zhang and Delworth 2005; Daley et al., 2011; Renssen et al., 2001). Weakening of the AMOC would cause a widespread cooling in the NH regions (e.g., Clark et al., 2001; Thomas et al., 2007) but warming in the SH regions (Wiersma et al., 2011; Wiersma and Renssen, 2006), creating a "bipolar seesaw" effect (e.g., Crowley, 1992; Broecker, 1998). The interhemispheric temperature difference between the NH and SH from this effect may be the driver of the southward displacement of the mean position of the ITCZ during the 8.2 ka abrupt cooling event. This may have intensified the Malagasy monsoon in NW Madagascar during austral summers, similar to what happened to the South American Summer Monsoon in Brazil (e.g., Cheng et al., 2009). In contrast, regions in the NH monsoon regions became drier at 8.2 ka BP as the Asian Monsoon and the East Asian Monsoon weakened (e.g., Wang et al., 2005; Dykoski et al., 2005; Cheng et al., 2009; Liu et al., 2013). The cold NH climate conditions and the wet climate conditions in NW Madagascar at 8.2 ka BP (Fig. 10) could suggest causal relationships. However, further research and data will be needed to confirm this possibility.

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6. Conclusions

Petrography, mineralogy, and stable isotope records from Stalagmite ANJB-2, from Anjohibe Cave, and Stalagmite MAJ-5, from Anjokipoty Cave, combine to suggest three distinct intervals of changing climate in Madagascar during the Holocene: relatively wet conditions during the MEHI,

relatively drier conditions, possibly due to episodic dry intervals, during the MMHI, and relatively wet conditions during the MLHI. The timing of stalagmite deposition during the MEHI and the MLHI in NW Madagascar could be attributed to a more southward migration and/or an expanded ITCZ, increasing the duration of the summer rainy seasons, perhaps linked to a stronger Malagasy monsoon. This could have been tied to the temperature gradient between the two hemispheres and weakening of the AMOC. In contrast, the c. 6500—year depositional hiatus during the MMHI could indicate a northward migration of the ITCZ, leading to relatively drier conditions in NW Madagascar. The evidence of the 8.2 ka event in the Malagasy records may further suggest a close link between paleoenvironmental changes in Madagascar and abrupt climatic events in the NH, suggesting that during the MEHI Madagascar's climate was very sensitive to abrupt ocean-atmosphere events in the NH.

Although the ITCZ is unquestionably one of the climatic drivers influencing climate in Madagascar and surrounding locations, several climatic factors need to be investigated in more detail. For example, we do not fully understand if the latitudinal migration is paired with the expansion and/or contraction of the ITCZ, which would affect the strengths of the associated monsoon systems. In addition, the interplay between ITCZ and other factors involving changes in sea surface temperatures, particularly IOD-ENSO, needs to be investigated in details. Data-model comparison (for example at the 8.2 ka event) and improved spatial and temporal resolution of paleoclimate datasets could be an approach to address this challenge.

Author Contribution

N.R.G.V. conceived the research and experiments. N.R.G.V, G.K, A.F.M.R, and M.O.M.R did the fieldwork and collected the samples. X.L., G.K., H.C., R.L.E, and N.R.G.V contributed to the ²³⁰Th dating analyses. N.R.G.V provided detailed investigation of the two stalagmites, provided stable isotope measurements, prepared thin sections, and conducted X-ray diffraction and SEM analyses on the samples. G.K. also assisted with the isotopic measurements on Stalagmite ANJB-2. N.R.G.V. wrote the first draft of the manuscript and led the writing. L.B.R. and G.A.B. provided a thorough review of the draft. N.R.G.V. and L.B.R. discussed and revised the manuscript, with additional comments from L.W. N.R.G.V revised the paper with input from all authors, reviewers, and editors.

Data availability: Data for this paper are available at xxxxxxxxx

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Competing Interests. The authors declare no conflict of interest.

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1041 Figures

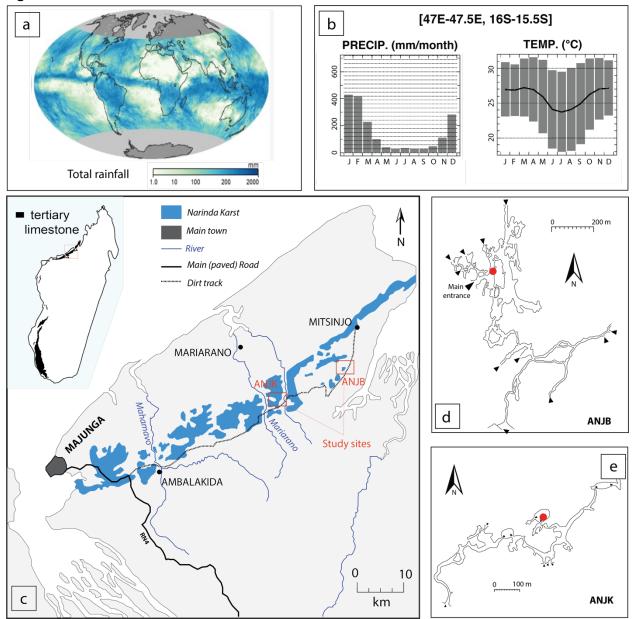


Figure 1: Climatological and geographic setting of Madagascar and the study area. (a) Global rainfall maps recorded by NASA's Tropical Rainfall Measuring Mission (TRMM) satellite showing the total monthly rainfall in millimeters and the overall position of the ITCZ during November, 2006. Darker shades of blue indicate regions of higher rainfall (source: NASA Earth Observatory, 2016). (b) Barplots of monthly precipitation, and monthly average of daily maximum, minimum, and mean temperature in NW Madagascar, based on 1971–2000 climate data. Source: http://iridl.ldeo.columbia.edu/ (accessed August 31, 2016). (c) Simplified map showing the

southwest part of the Narinda karst and the location of the study areas. Inset figure is a map of Madagascar showing the extent of the Tertiary limestone outcrop that makes up the Narinda karst. (d-e) Maps of Anjohibe (ANJB) and Anjokipoty (ANJK) caves (St-Ours, 1959; Middleton and Middleton, 2002), with approximate location for sample collection (red dots). See Figs. S1–S3 for additional information about the study locations.



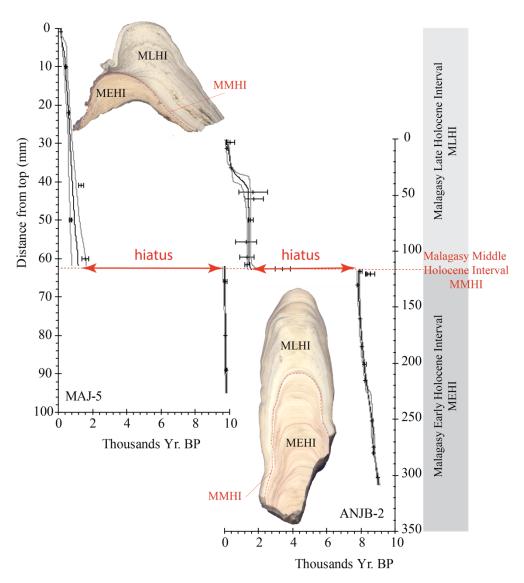


Figure 2: Age model of Stalagmite MAJ-5 (left) and ANJB-2 (right) using the StalAge1.0 algorithm of Scholz and Hoffman (2011) and Scholz et al. (2012). Scanned image of the two samples are shown for reference and to indicate the three distinct Holocene intervals.

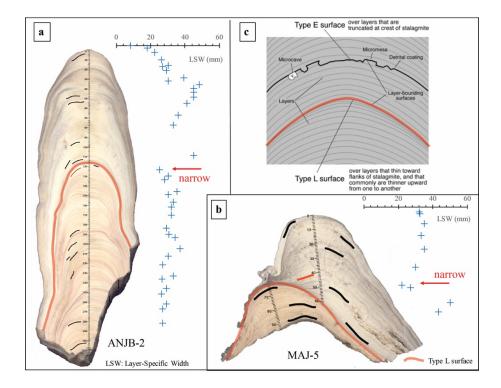


Figure 3: a) Scanned image of Stalagmite ANJB-2 and the corresponding variations in layer-specific width (LSW). b) Scanned image of Stalagmite MAJ-5 and the corresponding layer-specific width (LSW). c) Sketches of typical layer-bounding surfaces (Type E and Type L) of Railsback et al. (2013). Close-up photographs of the hiatuses are shown in Fig S6.

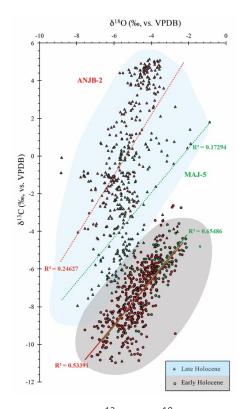


Figure 4: **Stable isotope data**. Scatterplots of δ^{13} C and δ^{18} O for Stalagmite MAJ-5 (green) and ANJB-2 (red) during the Malagasy early Holocene interval (circle) and the Malagasy late Holocene interval (triangle). The plot shows distinctive early and late Holocene conditions (roughly highlighted in gray and light blue, respectively).

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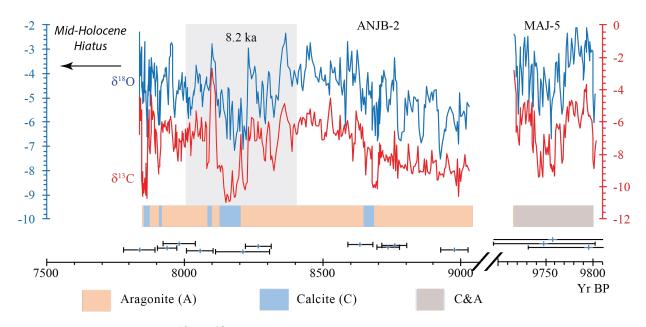


Figure 5: Variations in δ^{13} C, δ^{18} O, and mineralogy in Stalagmite ANJB-2 and Stalagmite MAJ-5 during the Malagasy Early Holocene Interval. Supplementary Fig. S6 shows both the corrected and uncorrected isotope values.

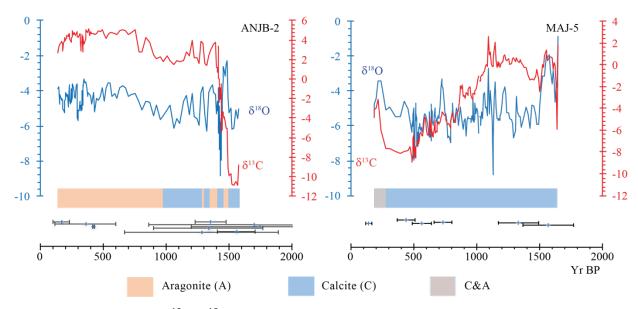


Figure 6: Variations in δ^{13} C, δ^{18} O, and mineralogy in Stalagmite ANJB-2 and Stalagmite MAJ-5 during the Malagasy Late Holocene Interval. Supplementary Fig. S7 shows both the corrected and uncorrected isotope values, and Fig. S8 compares the corrected δ^{18} O values for both stalagmites.

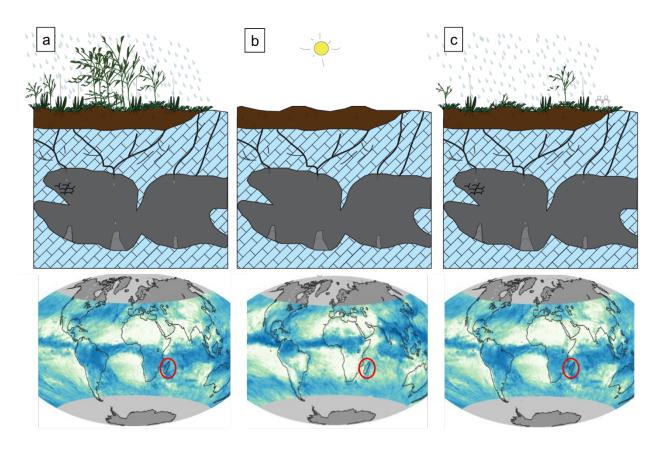


Figure 7: Simplified models portraying Holocene climate change in NW Madagascar and the possible climatic conditions linked to the ITCZ. a) Wetter conditions during the early Holocene with the ITCZ further south (prior to c 7.8 ka BP), favorable for stalagmite deposition. b) Periodic dry conditions during the mid-Holocene (between c. 7.8 and 1.6 ka BP) with the ITCZ further north leading to no stalagmite formation (refer to Sect. 5.2.2). c) Wetter conditions during the late Holocene (after c. 1.6 ka BP) with the ITCZ further south, favorable for stalagmite deposition. Drawings are not to scale. The bottom figures are from the same source as Fig. 1a, and they are only used here to give a perspective of the possible position of the ITCZ during the early, mid, and late Holocene. Madagascar is indicated with a red ellipse.



Figure 8: Regional comparison. Google Earth image showing the location of sites reported in Table S3 and in Figure 9. Most site records are from lake sediments, except for GeoB9307-3 (onshore off delta sediments), MD79257 (alkenone from marine sediment core), and Cold Air, Anjohibe, and Anjokipoty caves (stalagmites δ^{18} O).

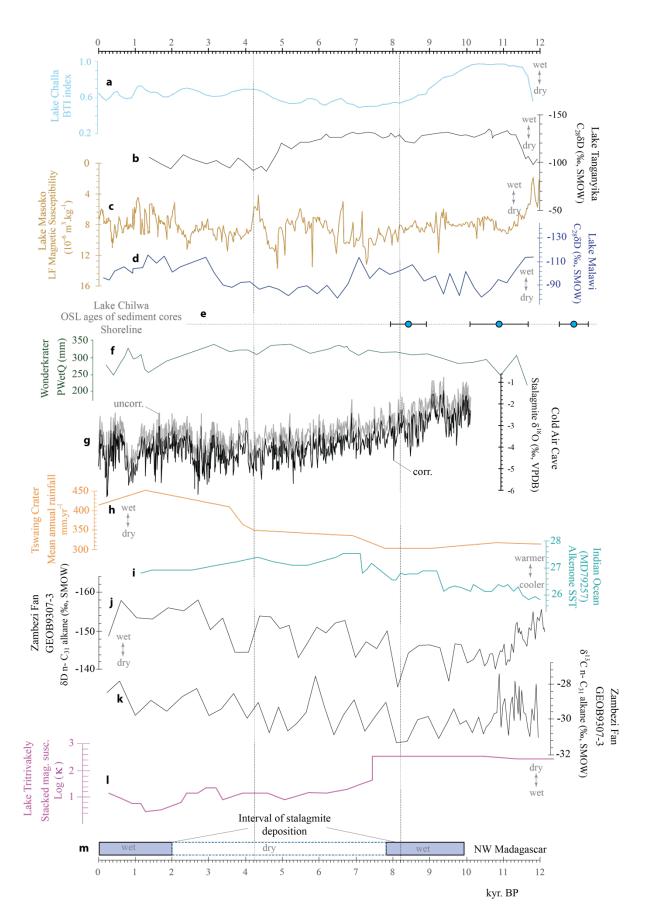


Figure 9: Regional comparison. a) Lake Challa BTI index (Verschuren et al., 2009). b) Lake Tanganyika C_{28} δD (Tierney et al., 2008, 2010). c) Lake Masoko low field magnetic susceptibility (10^{-6} .m 3 kg $^{-1}$) (Garcin et al., 2006). d) Lake Malawi C_{28} δD (Konecky et al., 2011). e) Lake Chilwa OSL dates of shoreline (Thomas et al., 2009). f) Wonderkrater reconstructed paleoprecipitation, PWetQ (Precipitation of the Wettest Quarter; Truc et al., 2013). g) Cold Air Cave corrected (corr.) and uncorrected (uncorr.) $\delta^{18}O$ profiles from Stalagmite T8 (Holmgren et al., 2003). h) Tswaing Crater paleo-rainfall derived from sediment composition (Partridge et al., 1997). i) Indian Ocean SST records from alkenone (Bard et al., 1997; Sonzogni et al., 1998). j-k) Zambezi δD n- C_{31} alkane $\delta^{13}C$ n- C_{31} alkane (Schefuß et al., 2011). I) Lake Tritrivakely stacked magnetic susceptibility (Williamson et al., 1998). m) NW Madagascar (Anjohibe and Anjokipoty) interval of deposition of Stalagmite ANJB-2 and Stalagmite MAJ-5 (this study). The two vertical dashed lines indicate the boundary of the Early, Middle, and Late Holocene by Walker et al. (2012) and Head and Gibbard (2015).

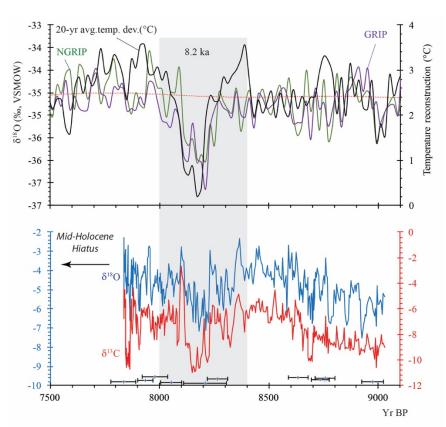


Figure 10: The 8.2 ka BP event in Madagascar. Oxygen isotope record from Greenland (GRIP and NGRIP) ice cores (Vinther et al., 2009) compared with Stalagmite ANJB-2 δ^{18} O and δ^{13} C.