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Reconstruction of Holocene coupling between the South America Monsoon System and local moisture variability from speleothem $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ records

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ABSTRACT

Investigating controls on past variability of South American hydroclimate is critical to assessing its response to future warming scenarios. $\delta^{18}\text{O}$ records from South America offer insight into past variability of the South American Monsoon System (SAMS). The controls, however, on precipitation $\delta^{18}\text{O}$ values can be decoupled from precipitation amount at a given site and, thereby, limit local moisture condition reconstructions. Here we use a principal components analysis to assess the coherence of speleothem and lake core Holocene $\delta^{18}\text{O}$ records in tropical and subtropical South America to evaluate the extent to which $\delta^{18}\text{O}$ variability reflects changes in SAMS intensity at different sites across the region. The main mode of variability across Holocene $\delta^{18}\text{O}$ records (PC1) closely tracks austral summertime insolation, consistent with existing work. Sites towards the periphery of the continent are heavily weighted on PC1, whereas interior sites are not. Further $\delta^{18}\text{O}$ variability at interior sites bear little similarity to each other and implicate controls, beyond regional monsoon intensity, on these $\delta^{18}\text{O}$ records. Further, we develop speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records spanning the Holocene from Tamboril Cave (Brazilian Highlands), Paraíso Cave (eastern Amazon Basin), Jaraguá Cave (Mato Grosso do Sul Plateau), and Botuverá Cave (Atlantic coastal plain) to investigate coupling between reconstructed monsoon variability (reflected by PC1) and local moisture conditions (interpreted from $^{87}\text{Sr}/^{86}\text{Sr}$ records). We interpret speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ variability as a proxy of local moisture conditions, reflecting the degree of water-rock interaction with the cave host rock as driven by variations in water residence time. Speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records from all the sites, except Botuverá cave, do not co-vary with PC1, suggesting that local moisture conditions do not necessarily follow variations in regional monsoon intensity at these interior sites. These speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records generally suggest dry mid-Holocene conditions relative to the early- and late-Holocene, consistent with interpretations of other paleo-moisture records in the region, but timing of wet-dry transitions varies between sites. These results highlight that controls, in addition to SAMS variability, might influence $\delta^{18}\text{O}$ variability. Further, our

results suggest spatially variable local moisture conditions at interior sites that do not uniformly respond to regional monsoon intensity, and stress the need for $\delta^{18}\text{O}$ -independent reconstructions of moisture conditions.

INTRODUCTION

The South American Monsoon System (SAMS) is a major source of water for municipal, industrial, and agricultural uses in tropical and subtropical South America, and supports the rich biodiversity of the Amazon rainforest. Investigating the response of the SAMS to changes in past climate provides an avenue for understanding how the monsoon domain's hydroclimate might respond to future climate change. Much attention has been focused on the reconstruction and dynamical understanding of past variability in the intensity of the SAMS, with a critical focus on the oxygen-isotope ($\delta^{18}\text{O}$) values preserved in a wide range of environmental archives, including ice cores, tree rings, lake sediments, and speleothems (e.g., Thompson et al., 2000; Ballantyne, 2011; Bird et al., 2011; Cruz et al., 2005). However, little attention has been paid to spatiotemporal connections between regional-scale variability in the SAMS and local moisture conditions (Wortham et al., 2017). Investigation of regional and local hydroclimate dynamics during the Holocene is of particular relevance to understanding the role of climate on the latitudinally dependent timing of shifts from savanna- to forest-dominated landscapes observed across South America from the mid to late-Holocene (Silva, 2014).

Variations in South American $\delta^{18}\text{O}$ records are commonly interpreted to reflect changes in the intensity of convective activity within the monsoon's core region of convection ($5 - 17.5^\circ\text{S}$, $72.5 - 47.5^\circ\text{W}$ as defined by Vuille et al., 2012). Modelling and modern monitoring studies have demonstrated that modern precipitation $\delta^{18}\text{O}$ ($\delta^{18}\text{O}_{\text{precip}}$) values across much of the monsoon domain are negatively correlated with convective activity and precipitation amount within the monsoon's core convective

region (i.e., more negative $\delta^{18}\text{O}$ values are correlated increases convection and precipitation amount and vice versa) (Vuille and Werner, 2005; Vuille et al., 2012). Consequently, $\delta^{18}\text{O}$ records from across the monsoon domain act as a fingerprint of convective strength within the core convective region, and thereby are commonly interpreted to reflect variations in regional SAMS intensity. This framework defines the paradigm for reconstructing SAMS activity from $\delta^{18}\text{O}$ variability preserved in a wide range of archives from the area, including ice cores (Thompson et al., 2000; Thompson et al., 1995), lake sediments (Bird et al., 2011), tree rings (Ballantyne et al., 2011) and speleothems (e.g., Burns et al., 2015; Strikis et al., 2018). Several studies have found striking co-variance in $\delta^{18}\text{O}$ variability from records across the monsoon domain (Vuille et al., 2012; Bird et al., 2011; Kanner et al., 2013), including a dipole in $\delta^{18}\text{O}$ variability between the western Amazon Basin and eastern Amazon Basin, (e.g., Cruz et al., 2009; Cheng et al., 2013; Lee et al., 2012; Lui and Battisti, 2015). There has been, however, limited recognition of differences among records within in the SAMS domain and discussion of the implications of such differences.

A compilation of $\delta^{18}\text{O}$ records from across the SAMS domain demonstrates both similarities and differences in Holocene $\delta^{18}\text{O}$ variability (Figure 1; Figure 2). In light of the interpretive framework discussed above (i.e., $\delta^{18}\text{O}$ records across the SAMS domain reflect monsoon core convective strength and regional monsoon intensity), clear differences among the records lead to two questions: i) where and when is $\delta^{18}\text{O}$ variability a proxy of SAMS intensity on the regional-scale, and ii) what additional processes influence the interpreted manifestations of regional climate as local $\delta^{18}\text{O}$ variability? It has long been recognized that the strength of $\delta^{18}\text{O}_{\text{precip}}$ as a proxy of regional monsoon intensity limits the extent to which $\delta^{18}\text{O}_{\text{precip}}$ reflects local precipitation amount (i.e., moisture conditions) (e.g., Vuille et al., 2012; Wortham et al., 2017). Although it is commonly, and sometimes implicitly, assumed that regional SAMS intensity uniformly drives local moisture conditions across the monsoon domain, previous work has documented decoupling between proxy records of SAMS intensity and local hydroclimate (Wortham

et al., 2017). This work highlights the need to use paired, or multiple, independent and distinct hydroclimate proxies to investigate variations in the nature of coupling between regional monsoon intensity and local moisture conditions (Wortham et al., 2017). More specifically, this coupling can be explored using a common regional monsoon signal identified from a compilation of $\delta^{18}\text{O}$ records from across the SAMS region as a proxy for monsoon intensity, and speleothem Sr-isotope ($^{87}\text{Sr}/^{86}\text{Sr}$) variability as a proxy of local moisture conditions (e.g., Banner et al., 1996; Frumkin and Stein, 2004; Oster et al., 2010; Zhou et al., 2009; Vaks et al., 2013; Oster et al., 2014).

Here, we leverage existing $\delta^{18}\text{O}$ records to assess the coherence of Holocene $\delta^{18}\text{O}$ records throughout the monsoon domain, with sites spanning the northern Andean foothills, eastern Amazon Basin, Mato Grosso do Sul Plateau, Brazilian Highlands, and Atlantic coastal plain, to understand where and when $\delta^{18}\text{O}$ reflects regional monsoon intensity, and explore additional processes that drive discordance among sites. We also develop corresponding speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records from several of these sites to investigate spatial and temporal variations in the coupling between regional monsoon variability and local moisture conditions.

HYDROCLIMATE SETTING

The hydroclimate of tropical and subtropical South America consists of distinct wet and dry seasons, driven primarily by the SAMS. The seasonal southward migration of the insolation maximum during the austral summer leads to differential heating and a thermal gradient between land and the Atlantic Ocean that drives mid-level atmospheric ascent and precipitation occurring over land (Garreaud et al., 2009). This atmospheric dynamic results in the formation of the Chaco Low and South American low-level jet that brings easterly winds traversing the Amazon Basin, and blocked by the Andes, to the south (Vera et al., 2006; Berbery and Barros, 2002). Additionally, the South Atlantic Convergence Zone

(SACZ) occurs as a NW-SE trending convection band as westerly flowing air masses coming off the continent intersect with easterly winds from the Atlantic (Carvalho et al., 2004). The Intertropical Convergence Zone (ITCZ), while notably distinct and separate from the SAMS, is an important conduit for moisture to the SAMS. The ITCZ follows the warmest tropical SSTs and its southward migration enhances SAMS convection in the modern system (Marengo et al., 2012; Vuille et al., 2012).

The Amazon Basin receives moisture from the tropical Atlantic Ocean year-round via prevailing easterlies (Vera et al., 2006). Annual precipitation, in the region where Paraíso Cave is located, is greater than 2,400 mm, with ~30% of total annual precipitation occurring during the mature phase of the monsoon (DJF), and ~70% occurring during the entirety of the wet season (November – June) (Figure 1). The more southerly Mato Grosso do Sul Plateau (Jaraguá Cave), Brazilian Highlands (Tamboril Cave), and Atlantic coastal plain (Grutas de Botuvera) have semi-humid climates, and receive ~1400 mm of annual precipitation. Moisture from these regions is sourced from both the Amazon Basin and the subtropical Atlantic via the SACZ (Vera et al., 2006). Precipitation over the eastern Amazon Basin and Brazilian Highlands is highly seasonal, primarily occurring when the SAMS is active (November – June), whereas the seasonality of precipitation is relatively less pronounced over the Mato Grosso do Sul Plateau and absent over the Atlantic coastal plain (Figure 1). Despite differences in the seasonality of precipitation amount, the seasonality of $\delta^{18}\text{O}_{\text{precip}}$ values is similar across the regions, reflecting i) the isotopic distillation of moisture when the SAMS is active, and ii) the advection of isotopically depleted moisture to regions downstream of the convection core (Vuille et al., 2012).

$^{87}\text{Sr}/^{86}\text{Sr}$ as proxy of water residence time

Sr-isotope ($^{87}\text{Sr}/^{86}\text{Sr}$) values in cave dripwater and speleothems can be used as a proxy of local moisture conditions, reflecting the length of water residence time within the vadose zone above the

cave. Infiltrating water obtains its initial $^{87}\text{Sr}/^{86}\text{Sr}$ signature from soil, as the amount of Sr in rainwater is negligible relative to that acquired from the soil (Figure 3). As water infiltrates through the host bedrock, the $^{87}\text{Sr}/^{86}\text{Sr}$ signature of the water evolves towards that of the bedrock via water-rock interaction (WRI) (i.e., calcite or dolomite recrystallization). The degree of WRI is dictated largely by water residence time within the subsurface above the cave, where increased water residence time enables increased WRI. Drier conditions promote longer water residence time and results in dripwater (or speleothem) $^{87}\text{Sr}/^{86}\text{Sr}$ values closer to that of the bedrock than the surface soil, as more Sr is progressively acquired from the dissolution of bedrock, and vice versa under wet conditions. Thus, speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ variability can potentially reflect relative changes in local wet/dry conditions (Figure 3). This conceptual model has been applied in settings that include arid (Vaks et al., 2013), semi-arid (Musgrove and Banner, 2004; Wong et al., 2011), Mediterranean (Oster et al., 2010), tropical (Banner et al., 1996), and monsoon (Wortham et al., 2017) climates.

Characterization of $^{87}\text{Sr}/^{86}\text{Sr}$ compositions of soil, bedrock, cave dripwater, and speleothems at individual cave sites can provide insight into the feasibility of using speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values as a proxy of water residence time associated with variability in local moisture conditions. Ideal settings are those in which the $^{87}\text{Sr}/^{86}\text{Sr}$ signatures of soil and bedrock are distinct and which the signatures of the endmembers vary within a narrow range relative to the difference between the endmembers. The occurrence of cave dripwater and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values between those of the soil and bedrock is consistent with the conceptual model that the $^{87}\text{Sr}/^{86}\text{Sr}$ value of infiltrating water is constrained by those of the soil and bedrock. Modelling of the evolution of water geochemical and isotopic compositions due to WRI can provide further means to assess the conceptual model. WRI is a strong control on cave dripwater and speleothem trace element concentrations relative to calcium (e.g., Sr/Ca), as well as $^{87}\text{Sr}/^{86}\text{Sr}$ values (Banner et al., 1996; Musgrove and Banner, 2004; Wong et al., 2011). The dissolution of bedrock with higher Sr/Ca ratios than the corresponding calcite that is re-precipitated increases Sr/Ca

values of the water as bedrock is recrystallized. Calcite precipitation from infiltrating water (i.e., prior calcite precipitation, PCP) results in an increase in Sr/Ca values. Specifically, Ca is preferentially removed from dripwater relative to Sr when calcite precipitates in voids above the cave or on the cave ceiling, thereby increasing the ratio of Sr relative to Ca in the remaining dripwater (Fairchild et al., 2007). Importantly, PCP does not affect $^{87}\text{Sr}/^{86}\text{Sr}$ compositions as isotopic fractionation of Sr is negligible during calcite precipitation. Therefore, PCP will result in the decoupling of the evolution of Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ values, making the co-variation of Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ values a useful delineator of WRI vs. PCP processes (Wong et al., 2011). Modelling results that can account for observed dripwater compositions indicate that water-rock interaction could be a strong control on cave dripwater and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values at modelled sites.

It is pertinent, however, to note that dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values and the magnitude of $^{87}\text{Sr}/^{86}\text{Sr}$ variability across sampling events can be different between sites within the same cave. Such differences might reflect spatial heterogeneity in soil and/or bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ values or differences in the dominant type of flow path supplying each site. With respect to the latter, diffuse flow paths through the matrix pore space of the host rock will reflect longer water residence time, more water-rock interaction, and $^{87}\text{Sr}/^{86}\text{Sr}$ values that are more similar to those of the bedrock relative to conduit flow paths. Conduit flow paths occur along solution-widened fractures that enable a more direct flow path between the surface and subsurface. Variability in flow rate and isotopic and geochemical compositions of dripwater at sites supplied by conduit flow paths is higher relative to sites supplied by diffuse flow paths that are buffered from changes at the surface. The spatial variability in $^{87}\text{Sr}/^{86}\text{Sr}$ compositions between sites from the same cave, therefore, requires that caution be taken when considering temporal variability in speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values compiled from distinct, non-overlapping speleothem samples. That is, offsets in values and differences in the magnitude of variability in $^{87}\text{Sr}/^{86}\text{Sr}$ values in speleothems from the same cave should be expected and taken into consideration when interpreting changes in moisture

conditions over time. Ideally, speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ would be based on a relative variability within a single speleothem sample. This limitation is less relevant with speleothem $\delta^{18}\text{O}$ records, which can more commonly be integrated across distinct speleothem samples with overlapping growth intervals. While differences in the nature of flow paths supplying different sites may still be relevant, spatial and temporal dripwater $\delta^{18}\text{O}$ variability can be relatively homogenous due to mixing of infiltrating water in the subsurface above the cave (Ayalon et al., 1998; Lachniet, 2009).

METHODS

This study generated $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ records for a speleothem from Tamboril Cave (TM6), and $^{87}\text{Sr}/^{86}\text{Sr}$ records for three additional cave sites in Brazil with existing $\delta^{18}\text{O}$ records, Paraíso (PAR01, PAR03, and PAR16), Jaraguá (JAR02, JAR04, and JAR07), and Botuverá (BTV21a) Caves. Each cave is hosted by carbonate bedrock within a unique geologic and vegetative setting (Table 1).

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

TM6 is a columnar stalagmite that was found, already broken, ~500 m into the cave in 2016 (Supplemental Figure 1). Age constraints ($n=12$) for TM6 were developed using U-series dating methods (Supplemental Figure 1). Sample preparation and chemical separation of U- and Th-fractions were carried out at Boston College Center for Isotope Geochemistry (BC-CIG) (following Edwards et al., 1987) and instrumental analyses were carried out at the Massachusetts Institute for Technology Paleoclimate and Geochronology Lab. Powder samples were dissolved and spiked with a ^{236}U - ^{233}U - ^{229}Th tracer prepared at MIT based on sample weight, age, and ^{238}U concentration estimates. Spiked samples were prepared for analyses using Fe co-precipitation purification and U and Th ion exchange chromatography (following Edwards et al., 1987). Prepared U and Th samples were analyzed on a Nu Plasma II-ES MC-ICP-MS with a static analysis, with unknowns bracketed by U and Th standard solutions. ^{234}U and ^{230}Th were

measured via ion counter, while remaining masses were measured on Faraday cups. Total procedural blank measurements are $< 2.0 \text{ pg } ^{238}\text{U}$, $< 0.8 \text{ fg } ^{234}\text{U}$, and undetectable ^{230}Th . For more information about analytical techniques, see Burn et al. (2016).

Assessing coherence of $\delta^{18}\text{O}$ records

Coherence of Holocene $\delta^{18}\text{O}$ variability from the region was assessed using a Principal Component Analysis (PCA). Records for which raw age-depth and proxy-depth data were accessible were included in the analysis. The age range of 2000 – 9600 years BP was chosen to allow for the largest number of records to be included in the analysis and inclusion of at least one record from each region to be included. Age-depth models were recalculated for existing records to quantify uncertainty associated with age-depth models and enable equitable comparison across all records (Breitenbach et al., 2012). The COPRA algorithm was used to generate 2,000 realizations of each age-model, and therefore, unique $\delta^{18}\text{O}$ time series, for each site for all records used in the PCA (Breitenbach et al., 2012). Raw proxy-depth information were not available for $\delta^{18}\text{O}$ records for LG3 and LG11 from Lapa Grande (Strikis et al., 2011) and BTV21a from Botuverá records (Bernal et al., 2016). For these speleothems, proxy-depths were estimated using a polynomial regression of the assessable age-depth and proxy-age data, and results were input into COPRA. Each $\delta^{18}\text{O}$ record was standardized individually and all records were interpolated to the same even time-step. Unique composite $\delta^{18}\text{O}$ records were made for sites for which there were multiple speleothems by averaging $\delta^{18}\text{O}$ values at each time-step in the overlapping interval using the 2,000 unique $\delta^{18}\text{O}$ time series for each speleothem sample output from COPRA. The PCA was conducted for each set of $\delta^{18}\text{O}$ time series, and the median outcome for each time-step is plotted.

Measuring $^{87}\text{Sr}/^{86}\text{Sr}$ values of speleothems, soil, bedrock, and cave dripwater

Soil, bedrock, and dripwater samples were collected at each cave site to characterize their $^{87}\text{Sr}/^{86}\text{Sr}$ compositions. Soils were collected at the cave entrance and multiple locations above the cave

when accessible. Bedrock samples were collected at the cave entrance and from several locations within the cave. Dripwater samples were collected from several distinct, actively dripping sites. Dripwater and bedrock leachates from Paraíso, Tamboril, Jaraguá, and Botuverá caves were analyzed for cation concentrations (Ca and Sr). Dripwater was analyzed using in the Quadrupole ICP-MS Lab in the Department of Geological Sciences at the University of Texas at Austin. Bedrock samples were analyzed as pressed powder pellets using x-ray fluorescence on a Philips PW2400 sequential spectrometer in the Ronald B. Gilmore XRF Lab, University of Massachusetts, Amherst (following the methods of Rhodes, 1996).

Analyses of $^{87}\text{Sr}/^{86}\text{Sr}$ values for soil and rock leachates, dripwater, and speleothems were conducted at BC-CIG. Speleothem powder (2 mg) was hand-drilled using a dental drill at discrete 1 cm increments along growth bands (~200 yr resolution). Dried, disaggregated soils (0.5 g) and crushed bedrock (0.01 g) were leached in 1 M ammonium acetate. All samples were processed using ion exchange methods as detailed by Montañez et al. (2002) and loaded onto outgassed single Re filaments in a TaF loading solution for analyses on an Isotopx Phoenix Thermal Ionization Mass Spectrometer (TIMS) at BC-CIG (Wortham et al., 2017). Samples were analyzed using a dynamic analysis. Uncertainty for $^{87}\text{Sr}/^{86}\text{Sr}$ values measured at BC-CIG is 0.000007 (2σ), based on the standard deviation of SRM987 measurements (mean = 0.710243, $n=42$). The $^{87}\text{Sr}/^{86}\text{Sr}$ of a subset of dripwater samples from Tamboril cave ($n=13$) was measured at University of Brasília.

Geochemical modeling of dripwater compositions

The evolution of dripwater Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ values due to WRI was modelled following Banner and Hanson (1990). Here we consider WRI to include calcite or dolomite recrystallization. Iterative mass balance calculations simulate increments of water passing through a given volume of bedrock (measured in moles), assuming each increment of water comes to elemental and isotopic equilibrium

with the bedrock it is in contact with based on the partition coefficients. Initial dripwater Sr/Ca values were assigned based on the lowest dripwater Sr/Ca (i.e., least evolved) measured at each cave. Initial dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values were assumed to have the $^{87}\text{Sr}/^{86}\text{Sr}$ values of soil above each cave (e.g., Banner et al., 1996; Musgrove and Banner, 2004; Wong et al., 2011; Wortham et al., 2017). Two sets of WRI trends are calculated for each cave system using the most and least radiogenic soil and bedrock values measured, which captures the range of possible dripwater values that could evolve under WRI.

RESULTS

$\delta^{18}\text{O}$ coherency across records

Oxygen-isotope records of sites toward the periphery of the continent (i.e., Botuverá, El Condor, Diamante, and Haupago Caves and Laguna Pumacocha; herein referred to as peripheral sites) exhibit a common decreasing trend that corresponds with increasing austral summer insolation throughout the Holocene (Figure 2) (Cruz et al., 2005a; Cruz et al., 2007; Wang et al., 2008; Kanner et al., 2013; Bird et al., 2011). This decreasing $\delta^{18}\text{O}$ trend is evident in the first principal component (PC1) delineated from the PCA, on which peripheral sites are all similarly and positively weighted (Figure 4). Sites toward the interior of the continent (Paraíso, Lapa Grande, Tamboril, and Jaraguá caves; here-in referred to as interior sites) do not exhibit a coherent decreasing trend from the early to late Holocene and are not heavily positively weighted on PC1. Additionally, $\delta^{18}\text{O}$ records of interior sites exhibit distinct variability that is not consistent across the region as evidenced by disparate, un-clustered weightings on PC1 and PC2. Sensitivity of the PCA results to inclusion of multiple sites from the same region were evaluated by including only one record from each region in the PCA (Supplemental Figure 5).

Just as interior sites exhibit variability dissimilar to the peripheral sites and regional signal, PC1, interior sites are also broadly dissimilar to one another. The Paraíso Cave $\delta^{18}\text{O}$ record becomes more

depleted from the early-Holocene until ~5ka, where values begin to increase until the present. While notably punctuated by growth hiatuses during much of the early- and mid-Holocene, Tamboril Cave exhibits increasing $\delta^{18}\text{O}$ values until the last 2,000 years, where there is then relatively less variability. Jaraguá cave, conversely, exhibits little variability throughout much of the early- and mid-Holocene, followed by broadly decreasing values from ~4ka to present. The aforementioned lack of variability in Jaraguá cave is matched by Lapa Grande cave in central Brazil.

Assessing $^{87}\text{Sr}/^{86}\text{Sr}$ values as proxy of moisture conditions

Soil and bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ values vary widely between sites. $^{87}\text{Sr}/^{86}\text{Sr}$ values of soil and bedrock at Tamboril are more radiogenic than those at Paraíso, Jaraguá, and Botuverá, and the difference between soil and bedrock values is greatest at Tamboril. Each site has a similar standard deviation of soil (± 0.0008) and bedrock (± 0.003) $^{87}\text{Sr}/^{86}\text{Sr}$ values, except Paraíso, where soil values have a relatively large range (± 0.004), but the tightest grouping of bedrock values (± 0.0002). Dripwater and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ are bracketed by those of soil and bedrock at each site. Tamboril dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values are more variable across drip sites than in other caves (± 0.0068 vs ± 0.001) (Figure 5), although relatively invariant across sampling events at a given site.

Soil $^{87}\text{Sr}/^{86}\text{Sr}$ values are distinct from those of the bedrock at Paraíso, Tamboril, Jaraguá, and Botuverá Cave sites, and modern dripwater and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values are bracketed by soil and bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ values at each site as well (Figure 5). Observed Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ values of dripwater from Paraíso, Tamboril, and Jaraguá caves could largely be accounted for by model WRI curves (Figure 5, blue shading between model lines). Dripwater with higher Sr/Ca values, for a given $^{87}\text{Sr}/^{86}\text{Sr}$ value, than model WRI curves, can be accounted for by prior calcite precipitation (PCP) (Wong et al., 2011). The consistency with which soils and bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ bracket those of dripwater and speleothems across all sites and the coincidence of modelled WRI (and, in some cases, PCP) curves with dripwater at each site

support the use of speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ variability as a proxy of variability in local moisture conditions at each site.

Differences among speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records

The speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records presented here do not co-vary i) with each other, ii) with speleothem $\delta^{18}\text{O}$ from the respective sites, nor iii) with PC1 delineated from the compilation of $\delta^{18}\text{O}$ records from the region, except for Botuverá cave. Botuverá is the only site at which speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{18}\text{O}$ records exhibit co-variability that is consistent with an interpretation that local moisture conditions became wetter with the increase in regional monsoon intensity over the Holocene (Figure 6; Figure 7). Further, Botuverá speleothem Sr/Ca, $\delta^{234}\text{U}$, and growth rate records previously developed by Bernal et al. (2016) are all consistent with this finding, further supporting the use of speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values as a proxy of moisture conditions. Importantly, however, is the distinct variability and up to a ~1‰ offset in Holocene $\delta^{18}\text{O}$ values over the last 4 ka in the three Botuverá speleothem records (BTV21a, Bernal et al., 2016; BTV2, Cruz et al., 2005; BT3, Wang et al., 2007) (Figure 6; Supplemental Figure 2). This observation highlights the potential for differential preservation of the same climate signal in different samples from the same cave (or from different caves in the same area) due to the influence of the cave system (e.g., Baker et al., 2010; Wong and Breecker, 2015).

The speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ record developed from Paraíso contains a negative excursion from 6 to 8 ka, inferred as a dry interval within the $^{87}\text{Sr}/^{86}\text{Sr}$ proxy framework. This interval is coincident with slower speleothem growth (Figure 2) but is not reflected in $\delta^{18}\text{O}$ values. We also note that this period coincides with abrupt drying in the Sahara present in several paleoclimate and archaeological archives (Stager et al., 2003; Tierney et al., 2017). The Paraíso $^{87}\text{Sr}/^{86}\text{Sr}$ values increase following the excursion, suggesting recovery from the dry interval by 6 ka. The decrease of $^{87}\text{Sr}/^{86}\text{Sr}$ values over the mid-to-late Holocene (2 to 5 ka) suggests the onset of prolonged dry conditions (Figure 7).

In the Tamboril cave records, the distinct nature of variability between TM0 and TM6 (i.e., not a one-to-one relationship between TM0 and TM6 $^{87}\text{Sr}/^{86}\text{Sr}$ values) might reflect differences in sensitivity of the two speleothems (Supplemental Figure 3) The speleothems were collected from two different sites within the cave system, which is formed in dolostone and limestone bedrock interbedded with silt and clay layers (Wortham et al., 2017). When considering a standardize TM6 record, a stepped change is evident from wet conditions inferred from high $^{87}\text{Sr}/^{86}\text{Sr}$ values during the early-Holocene (8 to 10 ka) to dry conditions inferred from lower values during the mid and late Holocene (2 to 6 ka) (Figure 7). During the last 2 ka at Tamboril cave $^{87}\text{Sr}/^{86}\text{Sr}$ values increase, suggesting wetter conditions in response to increased monsoon intensity over this interval as previously discussed (Wortham et al., 2017). Of note, $^{87}\text{Sr}/^{86}\text{Sr}$ values of the last millennia are anti-phased between Tamboril (i.e., increasing values reflecting wetter conditions) and both Paraíso and Botuverá caves (i.e., decreasing values reflecting drier conditions).

The Jaraguá cave $^{87}\text{Sr}/^{86}\text{Sr}$ record increases from 8 to 10 ka and decreases from 5 ka to present, marking the onset of a mid-Holocene dry interval coincident with mid/late-Holocene (~2 to 5 ka) drying at Paraíso and Tamboril (Figure 7; Supplemental Figure 3). Similar to Tamboril, the nature of variability in $^{87}\text{Sr}/^{86}\text{Sr}$ values varies between speleothems from Jaraguá cave. Standardized $^{87}\text{Sr}/^{86}\text{Sr}$ records more readily reveals that Jaraguá experienced wetter conditions at ~0.5 ka (Figure 7). This shift occurs 1.5 kyr later than the onset of wetter conditions at Tamboril, and is anti-phased with the occurrence of drying conditions at both Paraíso and Botuverá caves (Figure 7).

DISCUSSION

Discordance among Holocene $\delta^{18}\text{O}$ records

We document distinct variability among speleothem $\delta^{18}\text{O}$ record across tropical and subtropical South America, and suggest that the extent to which $\delta^{18}\text{O}$ variability reflects variability in SAMS intensity on the regional-scale may vary in space and time. Our analysis has demonstrated that Holocene $\delta^{18}\text{O}$ records from peripheral sites (i.e., northern Andes and Atlantic coastal plain) exhibit a gradual shift from higher to lower $\delta^{18}\text{O}$ values from early to late-Holocene (Figures 2 and 4). This trend is consistent with i) increasing austral summer insolation (Figure 4) driving an enhanced land-sea temperature gradient and more intense convection (Bird et al., 2011; Kanner et al., 2013), and ii) changes in Ti concentrations in the sediments of the Cariaco Basin (Figure 8) interpreted to reflect a southward shift of the ITCZ (Haug et al., 2001), which is a major moisture flux conduit that fuels convective activity in the SAMS region (Vuille et al., 2012). The lack of this trend, however, in $\delta^{18}\text{O}$ records at the other sites demonstrates that the inferred Holocene increase in monsoon intensity is not preserved in all records from the monsoon region (Figures 2 and 4). This indicates that precipitation $\delta^{18}\text{O}$ values at these sites are not sensitive to Holocene variations in insolation or strength and position of the ITCZ, and/or that other processes affecting precipitation $\delta^{18}\text{O}$ are also influential at these sites.

The occurrence of a dipole in $\delta^{18}\text{O}$ variability between records from the northern Andes and eastern Amazon Basin and Nordeste is not constant throughout the Holocene. The dipole is associated with the linked Bolivian High-Nordeste Low system (Lenters and Cook, 1997), and has long been recognized in the paleoclimate record (e.g., Cruz et al., 2009; Cheng et al., 2013). Our analysis documents the anti-phased trends from the mid to late-Holocene (3 to 6 ka) with $\delta^{18}\text{O}$ values increasing in records from Paraíso and Rio Grande do Norte and decreasing in records from the northern Andes (Figure 8). During the early Holocene (8 to 10 ka), however, the dipole is absent as all the records from the region exhibit decreasing $\delta^{18}\text{O}$ values (Figure 8). Variability in the extent to which the Bolivian High-Nordeste Low system influences $\delta^{18}\text{O}$ values in the Nordeste region, which has also been documented during the last millennium (Novello et al., 2012; Novello et al., 2018), suggests an additional control on

$\delta^{18}\text{O}$ variability during the early Holocene. Notably, the $\delta^{18}\text{O}$ records at Paraíso and Rio Grande do Norte correspond with Holocene changes in Ti concentrations in the sediments of the Cariaco Basin over this interval, as well as throughout much of the entirety of the Holocene (Figure 8). There is, however, seeming discordance in the dynamical interpretation linking the Cariaco Basin record with records from Paraíso and Rio Grande do Norte, as i) changes in the geochemistry of sediments of the Cariaco Basin reflect latitudinal shifts in the ITCZ with greater Ti concentrations corresponding with more *northerly* position of the ITCZ (Haug et al., 2001) and ii) precipitation $\delta^{18}\text{O}$ values over northeast Brazil correspond to the strength and position of the ITCZ, with lower $\delta^{18}\text{O}$ values associated with a more intense and *southerly* position of the ITCZ (Cruz et al., 2009). The coincidence of decreasing $\delta^{18}\text{O}$ values in speleothem records from Paraíso and Rio Grande do Norte caves when the ITCZ is inferred to be in a more northerly position based on higher Ti percentages in Cariaco Basin sediments, and vice versa, (Figure 8), highlights that shifts in the ITCZ cannot account for Holocene variations present in both regions. The presence of the east-west dipole in $\delta^{18}\text{O}$ variability during the mid to late Holocene might account for increasing $\delta^{18}\text{O}$ values at Paraíso and Rio Grande do Norte caves despite the more southerly position of the ITCZ inferred from the Cariaco Basin sediments. The absence, however, of the dipole during the early Holocene requires another explanation for decreasing $\delta^{18}\text{O}$ values at Paraíso and Rio Grande do Norte caves despite an increasingly more northward position of the ITCZ.

The distinct Holocene $\delta^{18}\text{O}$ variability at Jaraguá and Tamboril caves suggests that these sites may be influenced by hydroclimate dynamics in addition to regional-scale variability in the SAMS intensity. Existing work in the region has suggested $\delta^{18}\text{O}$ variability is linked to the Atlantic Ocean, for example, linked to the Atlantic Multidecadal Oscillation or perturbations in the Atlantic Meridional Overturning Circulation and resulting displacement of the ITCZ (Strikis et al., 2011, Strikis et al., 2018, Novello et al., 2012, Novello et al., 2018, Cheng et al., 2009, Bird et al., 2011). Additionally, the proximity of these sites to the South Atlantic Convergence Zone (SACZ) might indicate that climate dynamics

related to the strength and position of the SACZ might be an important influence on the $\delta^{18}\text{O}$ variability in these records (e.g., Carvalho et al., 2004; Novello et al., 2018). The lack of common variability, however, does not support the idea that there is a common process, such as teleconnections with the Atlantic, that dominantly controls $\delta^{18}\text{O}$ variability at these sites. Furthermore, speleothem $\delta^{18}\text{O}$ records developed from Botuverá, which is also located within the SACZ, follow records from the Andes and complicates the hypothesis that records from the SACZ region might be dominantly influenced by unique dynamics associated with the SACZ.

Decoupling of Local Moisture Conditions and Regional Monsoon Intensity

Of the four $^{87}\text{Sr}/^{86}\text{Sr}$ records developed in this study, only the record from Botuverá cave illustrates coupling with regional monsoon variability over the Holocene, suggesting moisture availability in the Atlantic coastal plain region was linked with increasing monsoon intensity consistent with the literature (e.g., Cruz et al., 2006; Bernal et al., 2016) (Figures 6 and 7). Lack of consistency between variability in speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records and regional-scale monsoon intensity at Paraíso, Tamboril, and Jaraguá Caves may suggest that $^{87}\text{Sr}/^{86}\text{Sr}$ variability is not a good proxy of local moisture conditions or the response of local moisture conditions to regional monsoon variability is not uniform across the SAMS region, and rather, spatially and temporally variable. We now consider the feasibility of alternative interpretations of $^{87}\text{Sr}/^{86}\text{Sr}$ variability as well as the possible implications of local moisture conditions that are decoupled from regional-scale variations in the monsoon system.

Evolution of the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of the soil could potentially be a critical control on speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ compositions because infiltrating water acquires its initial isotope signature from the soil. Contributions to the soil from volcanism or atmospheric dust are potential controls on soil $^{87}\text{Sr}/^{86}\text{Sr}$ compositions. Volcanic inputs, however, would produce short-lived, temporally asymmetrical perturbations in $^{87}\text{Sr}/^{86}\text{Sr}$ variability due to episodic fluxes of a readily weatherable Sr source (Banner et

al., 1996), and this type of variability is not seen in the records. Previous work has demonstrated that, despite the transport of African dust to South America during the austral summer (e.g., Koren et al., 2006; Ben-Ami et al., 2010), Saharan dust does not contribute substantial amounts of Sr to regional soils (Abouchami et al., 2013; Pourmand et al., 2014). Alternatively, physical thickening (or thinning) of the soil layer might lead to a weaker (or stronger) presence of the underlying bedrock in the $^{87}\text{Sr}/^{86}\text{Sr}$ signature of the soil horizon (Cooke et al., 2003). Variations in soil thickness might also be reflected in speleothem $\delta^{13}\text{C}$ values, as thicker soils (higher $^{87}\text{Sr}/^{86}\text{Sr}$ values) and lower $\delta^{13}\text{C}$ values are likely to occur under warmer, wetter conditions. Such conditions would favor C3 vegetation over C4 vegetation as well as enhanced subsurface CO_2 production that increases the proportion of vegetation-sourced (vs. atmosphere- or bedrock-sourced) CO_2 in speleothem forming waters (Breecker, 2015; Novello et al., in review). The broad correspondence between corresponding speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{13}\text{C}$ records at Paraíso and Jaraguá Caves is consistent with this interpretation (Supplemental Figure 6). The inverse relationship, however, between $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{13}\text{C}$ values is also expected from the proposed interpretative framework in which water-rock interaction is the dominant control on $^{87}\text{Sr}/^{86}\text{Sr}$ variability. That is, the processes that promote higher $\delta^{13}\text{C}$ values (e.g., water-rock interaction, prior calcite precipitation, shifts from C3 to C4 vegetation; Breecker, 2015) are enhanced under drier conditions under which more water-rock interaction and lower, more bedrock-like $^{87}\text{Sr}/^{86}\text{Sr}$ values are expected (e.g., Banner et al., 1996; Musgrave and Banner, 2004; Wortham et al., 2017). Whether $^{87}\text{Sr}/^{86}\text{Sr}$ variability is dominantly influenced by soil thickening or water-rock interaction, both processes lead to similar interpretations with lower $^{87}\text{Sr}/^{86}\text{Sr}$ values associated with drier conditions. The geochemical modeling conducted in this study, which is not commonly conducted as a part of most paleoclimate reconstruction studies, demonstrates the ability of water-rock interaction to explain the $^{87}\text{Sr}/^{86}\text{Sr}$ variability in each cave setting (Figure 5) and provides support for this interpretation.

Assuming that speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ variations reflect changes in local moisture conditions, stepped decreases in $^{87}\text{Sr}/^{86}\text{Sr}$ records from Paraíso (6 ka), Tamboril (between 6 and 7 ka), and Jaraguá (5 ka) within individual speleothems suggest a shift from wetter early to Holocene conditions to drier conditions during the mid-Holocene, followed by a transition to wetter conditions in the late-Holocene (~ 2 to 3 ka) at each site (Figure 7 and Supplemental Figure 3). The occurrence of a drier mid-Holocene interval is consistent with i) lake sediments in the Puna of Argentina (19 - 27°S) suggesting a transition period from humid to dry local moisture conditions from 5 to 6 ka (Morales et al., 2018), and ii) archeological studies in the Rio de la Plata Basin region (14 - 27°S, 43 - 67°W) that suggests movements of people out of increasingly more arid environments at ~5 ka (Iriarte et al., 2017). The onset of drying, though poorly constrained by the $^{87}\text{Sr}/^{86}\text{Sr}$ records in this study, is broadly coherent with a period of mid-Holocene drying documented in records of leaf wax δD and $\delta^{13}\text{C}$ from the more westerly located Laguna la Gaiba (18°S, 58°W, lowland Bolivia), which indicates onset to dry conditions at ~8 ka (Fornace et al., 2016). Onset of dry conditions between 7 and 8 ka is also found in Andean lake records, Lake Titicaca, Paco Cocha, and Taypi Chaka (e.g., Cross et al., 2001; Baker et al., 2001; Abbott et al., 2003, Tapia et al., 2003; Fritz et al., 2007; Pollisar et al., 2013) and in the onset of Andean glacier retreat (e.g., Cordillera Blanca (9°S) and Huaguruncho (10°S)) inferred from cosmogenic radionuclide ages and proglacial lake sediment flux records (Stansell et al., 2017; Stansell et al., 2014). With respect to the Amazon Basin, a pollen record from the southeastern part of the basin (6°S, 50°W) suggests a dry transition at ~10 ka, and aridity from 9 ka to the present (Hermanowski et al., 2012). In contrast, a pollen record from the western part of the basin (3°N, 73°W) suggests drier conditions between ~6 and 8.7 ka, with onset of wetter conditions at 6 ka (Behling and Hooghiemstra, 2000). The large range in the timing and duration of mid-Holocene aridity across the paleo-moisture reconstructions discussed here highlights the need for additional reconstructions that are temporally well-constrained.

The occurrence of dry mid-Holocene conditions in moisture reconstructions from across the region implicates a mechanism that has the potential to drive hydroclimate patterns across the region. A common climate dynamic that can account for the observed mid-Holocene aridity, which is temporally offset between sites, is not readily obvious. Teleconnections resulting from the patterns of tropical Pacific SSTs have been proposed as the control on Holocene variability in the Andean hydroclimate based on the pattern of spatial variability in mid-Holocene wetting and drying across Latin America (20°N to 20°S) being similar to that resulting from modern day ENSO variations (Pollisar et al., 2013). The occurrence, however, of mid-Holocene drying that spans the eastern Amazon Basin, Mato Grosso do Sul Plateau, Brazilian Highlands, and Andes is not consistent with the modern response of hydroclimate to ENSO variations. More specifically, ENSO variability induces a dipole response between the regions of the eastern Amazon Basin/Andes and Mato Grosso do Sul Plateau and Brazilian Highlands (Garreaud et al., 2009). Previous studies have also suggested a time-transgressive latitudinally-dependent shift in proxy records in the Andes back to wet conditions in the late-Holocene (Rodbell et al., 2008; Mayle and Powers 2008). This pattern, however, is not readily apparent in the records discussed here. Understanding the dynamical coherence of these mid-Holocene drying records and potential driving mechanisms is beyond the scope of this study and points to a need for further investigation.

Reconstructions of moisture conditions that document mid-Holocene drying are decoupled from regional-scale variability in monsoon intensity. This implies that variations in regional monsoon intensity may not be good indicators of local moisture conditions across the region, i.e., the convective intensity over the core monsoon region does not dictate the extent to which downstream locations receive more or less annual precipitation. This work implies that moisture reconstructions from tropical and subtropical South America that indicate wetter or drier conditions need not be in contradiction to reconstructions that indicate a weaker or stronger monsoon, respectively. Further, this highlights the potential importance of climate processes, independent of the monsoon system, to changes in water

availability pertinent to past and future environments, and the need to broaden the focus beyond monsoon variability in understanding past hydroclimate.

SUMMARY AND CONCLUSIONS

We assessed the coherence of existing Holocene speleothem and lake sediment $\delta^{18}\text{O}$ records and the relationship between $\delta^{18}\text{O}$ records and local moisture reconstructions. A new Holocene $\delta^{18}\text{O}$ record is developed for the Brazilian Highlands region, and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records are developed for the eastern Amazon Basin, Brazilian Highlands, Mato Grosso do Sul Plateau, and Atlantic Coastal Plain regions. Our results suggest the main mode of variability in Holocene tropical South America $\delta^{18}\text{O}$ records is one that closely tracks increasing austral summertime insolation, consistent with existing literature that argues that enhanced insolation drives increased monsoon convection. However, only $\delta^{18}\text{O}$ records from the tropical Andean foothills and Atlantic Coastal Plain are positively correlated with this mode, suggesting the other $\delta^{18}\text{O}$ records from more interior sites in the eastern Amazon Basin, Brazilian Highlands, and Mato Grosso do Sul Plateau are influenced by additional mechanisms beyond regional monsoon intensity. Delineation of such mechanisms is beyond the scope of this study and necessitates further research. We demonstrate that speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values are a feasible proxy of local moisture conditions. Holocene $^{87}\text{Sr}/^{86}\text{Sr}$ records exhibit variability that is decoupled with the main monsoon signal, suggesting that local moisture conditions are not tightly linked to regional monsoon variability, and rather, the local moisture response is spatially and temporally heterogeneous across the monsoon domain. Botuverá cave, however, does exhibit co-variability of speleothem $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ values throughout the Holocene, consistent with the existing literature that suggests moisture availability in the Atlantic Coastal Plain region is linked to regional monsoon intensity (Bernal et al, 2016). Speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ records are broadly coherent in the preservation of dry mid-Holocene

conditions, which is consistent with existing paleo-moisture reconstructions from the Andes and Mato Grosso do Sul Plateau. This widespread mid-Holocene dry interval, despite monotonic increases in regional monsoon intensity over this interval suggests a mechanism, independent of monsoon intensity, that modulates local moisture availability. It is clear that Holocene variability in hydroclimate over tropical and subtropical South America is complex with multiple mechanisms at play. This realization marks the importance of investigating potential controls on $\delta^{18}\text{O}$ variability at sites where regional-scale monsoon intensity does not appear to be the dominant control and the use of a multi-proxy approach to reconstructing hydroclimate across the SAMS region.

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References Cited

- Abbott, M.B., Wolfe, B.B., Wolfe, A.P., et al., 2003, Holocene paleohydrology and glacial history of the central Andes using multiproxy lake sediment studies: *Palaeogeography, Palaeoclimatology, Palaeoecology*, v. 194, no. 1-3, p. 123-138.
- Abouchami, W., Nthe, K., Kumar, A., et al., 2013, Geochemical and isotopic characterization of the Bodl Depression dust source and implications for transatlantic dust transport to the Amazon Basin: *Earth and Planetary Science Letters*, v. 380, p. 112-123.

533 Ayalon, A., Bar-Matthews, M., and Sass, E., 1998, Rainfall-recharge relationships within a karstic
534 terrain in the Eastern Mediterranean semi-arid region, Israel: $\delta^{18}\text{O}$ and δD characteristics:
535 *Journal of Hydrology*, v. 207, no. 1-2, p. 18-31.

536 Baker, A., and Bradley, C., 2010, Modern stalagmite $\delta^{18}\text{O}$: Instrumental calibration and forward
537 modelling: *Global and Planetary Change*, v. 71, no. 3-4, p. 201-206.

538 Baker, P.A., Seltzer, G.O., Fritz, S.C., et al., 2001, The history of South American tropical
539 precipitation for the past 25,000 years: *Science*, v. 291, no. 5504, p. 640-643.

540 Ballantyne, A.P., Baker, P.A., Chambers, J.Q., Villalba, R., and Argollo, J., 2011, Regional
541 differences in South American monsoon precipitation inferred from the growth and isotopic
542 composition of tropical trees: *Earth Interactions*, v. 15, no. 5, p. 1-35.

543 Banner, J.L., and Hanson, G.N., 1990, Calculation of simultaneous isotopic and trace element
544 variations during water-rock interaction with applications to carbonate diagenesis: *Geochimica*
545 *et Cosmochimica Acta*, v. 54, no. 11, p. 3123-3137.

546 Banner, J.L., Musgrove, M., Asmerom, Y., Edwards, R.L., and Hoff, J.A., 1996, High-resolution
547 temporal record of Holocene ground-water chemistry: Tracing links between climate and
548 hydrology: *Geology*, v. 24, no. 11, p. 1049-1053.

549 Behling, H., and Hooghiemstra, H., 2000, Holocene Amazon rainforest-savanna dynamics and
550 climatic implications: high-resolution pollen record from Laguna Loma Linda in eastern
551 Colombia: *Journal of Quaternary Science: Published for the Quaternary Research Association*,
552 v. 15, no. 7, p. 687-695.

553 Ben-Ami, Y., Koren, I., Rudich, Y., Artaxo, P., Martin, S.T., and Andreae, M.O., 2010, Transport of
554 North African dust from the Bodélé depression to the Amazon Basin: a case study: *Atmospheric*
555 *Chemistry and Physics*, v. 10, no. 16, p. 7533-7544.

556 Berbery, E.H., and Barros, V.R., 2002, The hydrologic cycle of the La Plata basin in South America:
557 *Journal of Hydrometeorology*, v. 3, no. 6, p. 630-645.

558 Bernal, J.P., Cruz, F.W., Strömberg, N.M., et al., 2016, High-resolution Holocene South American
559 monsoon history recorded by a speleothem from Botuverá Cave, Brazil: *Earth and Planetary*
560 *Science Letters*, v. 450, p. 186-196.

561 Bird, B.W., Abbott, M.B., Vuille, M., Rodbell, D.T., Stansell, N.D., and Rosenmeier, M.F., 2011, A
562 2,300-year-long annually resolved record of the South American summer monsoon from the
563 Peruvian Andes: *Proceedings of the National Academy of Sciences*, v. 108, no. 21, p. 8583-
564 8588.

565 Breecker, D.O., 2017, Atmospheric pCO_2 control on speleothem stable carbon isotope
566 compositions: *Earth and Planetary Science Letters*, v. 458, p. 58-68.

567 Breitenbach, S.F., Rehfeld, K., Goswami, B., et al., 2012, Constructing proxy records from age
568 models (COPRA): *Climate of the Past*, v. 8, no. 5, p. 1765-1779.

569 Burn, M., 2016, LA-ICP-QMS Th-U/Pb allanite dating: methods and applications: PhD, University of
570 Bern.

571 Burns, S.J., Kanner, L.C., Cheng, H., and Edwards, R.L., 2015, A tropical speleothem record of
572 glacial inception, the South American Summer Monsoon from 125 to 115 ka.: *Climate of the*
573 *Past*, v. 11, no. 6.

574 Carvalho, L.M., Jones, C., and Liebmann, B., 2004, The South Atlantic convergence zone: Intensity,
575 form, persistence, and relationships with intraseasonal to interannual activity and extreme
576 rainfall: *Journal of Climate*, v. 17, no. 1, p. 88-108.

577 Cheng, H., Fleitmann, D., Edwards, R.L., et al., 2009, Timing and structure of the 8.2 kyr BP event
578 inferred from $\delta^{18}\text{O}$ records of stalagmites from China, Oman, and Brazil: *Geology*, v. 37, no.
579 11, p. 1007-1010.

580 Cheng, H., Sinha, A., Cruz, F.W., et al., 2013, Climate change patterns in Amazonia and
581 biodiversity: *Nature communications*, v. 4, p. 1411.

582 Cooke, M.J., Stern, L.A., Banner, J.L., Mack, L.E., Stafford Jr, T.W., and Toomey III, R.S., 2003,
583 Precise timing and rate of massive late Quaternary soil denudation: *Geology*, v. 31, no. 10, p.
584 853-856.

585 Cross, S.L., Baker, P.A., Seltzer, G.O., Fritz, S.C., and Dunbar, R.B., 2001, Late Quaternary climate
586 and hydrology of tropical South America inferred from an isotopic and chemical model of Lake
587 Titicaca, Bolivia and Peru: *Quaternary Research*, v. 56, no. 1, p. 1-9.

588 Cruz Jr, F.W., Burns, S.J., Jercinovic, M., Karmann, I., Sharp, W.D., and Vuille, M., 2007, Evidence
589 of rainfall variations in Southern Brazil from trace element ratios (Mg/Ca and Sr/Ca) in a Late
590 Pleistocene stalagmite: *Geochimica et Cosmochimica Acta*, v. 71, no. 9, p. 2250-2263.

591 Cruz Jr, F.W., Burns, S.J., Karmann, I., et al., 2005, Insolation-driven changes in atmospheric
592 circulation over the past 116,000 years in subtropical Brazil: *Nature*, v. 434, no. 7029, p. 63.

593 Cruz, F.W., Vuille, M., Burns, S.J., et al., 2009, Orbitally driven east–west antiphasing of South
594 American precipitation: *Nature Geoscience*, v. 2, no. 3, p. 210.

595 Edwards, R.L., Chen, J.H., and Wasserburg, G.J., 1987, ^{238}U / ^{234}U / ^{230}Th / ^{232}Th systematics and
596 the precise measurement of time over the past 500,000 years: *Earth and Planetary Science*
597 *Letters*, v. 81, no. 2-3, p. 175-192.

598 Fairchild, I.J., and McMillan, E.A., 2007, Speleothems as indicators of wet and dry periods:
599 *International Journal of Speleology*, v. 36, no. 2, p. 2.

600 Fornace, K.L., Whitney, B.S., Galy, V., Hughen, K.A., and Mayle, F.E., 2016, Late Quaternary
601 environmental change in the interior South American tropics: new insight from leaf wax stable
602 isotopes: *Earth and Planetary Science Letters*, v. 438, p. 75-85.

603 Fritz, S.C., Baker, P.A., Seltzer, G.O., Ballantyne, A., Tapia, P., Cheng, H., and Edwards, R.L.,
604 2007, Quaternary glaciation and hydrologic variation in the South American tropics as
605 reconstructed from the Lake Titicaca drilling project: *Quaternary Research*, v. 68, no. 3, p. 410-
606 420.

607 Frumkin, A., and Stein, M., 2004, The Sahara–East Mediterranean dust and climate connection
608 revealed by strontium and uranium isotopes in a Jerusalem speleothem: *Earth and Planetary*
609 *Science Letters*, v. 217, no. 3-4, p. 451-464.

610 Garreaud, R.D., Vuille, M., Compagnucci, R., and Marengo, J., 2009, Present-day south american
611 climate: *Palaeogeography, Palaeoclimatology, Palaeoecology*, v. 281, no. 3-4, p. 180-195.

612 Haug, G.H., Hughen, K.A., Sigman, D.M., Peterson, L.C., and Röhhl, U., 2001, Southward migration
613 of the intertropical convergence zone through the Holocene: *Science*, v. 293, no. 5533, p. 1304-
614 1308.

615 Hermanowski, B., da Costa, M.L., Carvalho, A.T., and Behling, H., 2012, Palaeoenvironmental
616 dynamics and underlying climatic changes in southeast Amazonia (Serra Sul dos Carajás,
617 Brazil) during the late Pleistocene and Holocene: *Palaeogeography, Palaeoclimatology,*
618 *Palaeoecology*, v. 365, p. 227-246.

619 Iriarte, J., DeBlasis, P., De Souza, J.G., and Corteletti, R., 2017, Emergent complexity, changing
620 landscapes, and spheres of interaction in southeastern South America during the middle and
621 late Holocene: *Journal of Archaeological Research*, v. 25, no. 3, p. 251-313.

622 Kanner, L.C., Burns, S.J., Cheng, H., Edwards, R.L., and Vuille, M., 2013, High-resolution variability
623 of the South American summer monsoon over the last seven millennia: insights from a
624 speleothem record from the central Peruvian Andes: *Quaternary Science Reviews*, v. 75, p. 1-
625 10.

626 Koren, I., Kaufman, Y.J., Washington, R., Todd, M.C., Rudich, Y., Martins, J.V., and Rosenfeld, D.,
627 2006, The Bodélé depression: a single spot in the Sahara that provides most of the mineral dust
628 to the Amazon forest: *Environmental Research Letters*, v. 1, no. 1, p. 014005.

629 Lachniet, M.S., 2009, Climatic and environmental controls on speleothem oxygen-isotope values:
630 *Quaternary Science Reviews*, v. 28, no. 5-6, p. 412-432.

631 Lee, J., Johnson, K., and Fung, I., 2009, Precipitation over South America during the Last Glacial
632 Maximum: an analysis of the “amount effect” with a water isotope-enabled general circulation
633 model: *Geophysical Research Letters*, v. 36, no. 19.

634 Lenters, J.D., and Cook, K.H., 1997, On the origin of the Bolivian high and related circulation
635 features of the South American climate: *Journal of the Atmospheric Sciences*, v. 54, no. 5, p.
636 656-678.

637 Liu, X., and Battisti, D.S., 2015, The influence of orbital forcing of tropical insolation on the climate
638 and isotopic composition of precipitation in South America: *Journal of Climate*, v. 28, no. 12, p.
639 4841-4862.

640 Marengo, J.A., Chou, S.C., Kay, G., et al., 2012, Development of regional future climate change
641 scenarios in South America using the Eta CPTec/HadCM3 climate change projections:
642 climatology and regional analyses for the Amazon, São Francisco and the Paraná River basins:
643 *Climate Dynamics*, v. 38, no. 9-10, p. 1829-1848.

644 Mayle, F.E., and Power, M.J., 2008, Impact of a drier Early–Mid-Holocene climate upon Amazonian
645 forests: *Philosophical Transactions of the Royal Society B: Biological Sciences*, v. 363, no.
646 1498, p. 1829-1838.

647 Morales, M.R., Bustos, S., Oxman, B.I., Pirola, M., Tchilinguirian, P., Orgeira, M.J., and Yacobaccio,
648 H.D., 2018, Exploring habitat diversity of mid-holocene hunter-gatherers in the South-Central
649 Andes: Multi-proxy analysis of Cruces Core 1 (TC1), Dry Puna of Jujuy, Argentina: *Journal of*
650 *Archaeological Science: Reports*, v. 18, p. 708-721.

651 Musgrove, M., and Banner, J.L., 2004a, Controls on the spatial and temporal variability of vadose
652 dripwater geochemistry: Edwards aquifer, central Texas1: *Geochimica et Cosmochimica Acta*,
653 v. 68, no. 5, p. 1007-1020.

654 Musgrove, M., and Banner, J.L., 2004b, Controls on the spatial and temporal variability of vadose
655 dripwater geochemistry: Edwards aquifer, central Texas1: *Geochimica et Cosmochimica Acta*,
656 v. 68, no. 5, p. 1007-1020.

657 Novello, V.F., Cruz, F.W., Moquet, J.S., et al., 2018, Two millennia of South Atlantic Convergence
658 Zone variability reconstructed from isotopic proxies: *Geophysical Research Letters*.

659 Novello, V.F., Cruz, F.W., Karmann, I., et al., 2012, Multidecadal climate variability in Brazil's
660 Nordeste during the last 3000 years based on speleothem isotope records: *Geophysical*
661 *Research Letters*, v. 39, no. 23.

662 Oster, J.L., Montanez, I.P., Guilderson, T.P., Sharp, W.D., and Banner, J.L., 2010, Modeling
663 speleothem $\delta^{13}\text{C}$ variability in a central Sierra Nevada cave using ^{14}C and $^{87}\text{Sr}/^{86}\text{Sr}$:
664 *Geochimica et Cosmochimica Acta*, v. 74, no. 18, p. 5228-5242.

665 Oster, J.L., Montañez, I.P., Mertz-Kraus, R., et al., 2014, Millennial-scale variations in western Sierra
666 Nevada precipitation during the last glacial cycle MIS 4/3 transition: *Quaternary Research*, v.
667 82, no. 1, p. 236-248.

668 Polissar, P.J., Abbott, M.B., Wolfe, A.P., Vuille, M., and Bezada, M., 2013, Synchronous
669 interhemispheric Holocene climate trends in the tropical Andes: *Proceedings of the National*
670 *Academy of Sciences*, p. 201219681.

671 Pourmand, A., Prospero, J.M., and Sharifi, A., 2014, Geochemical fingerprinting of trans-Atlantic
672 African dust based on radiogenic Sr-Nd-Hf isotopes and rare earth element anomalies:
673 *Geology*, v. 42, no. 8, p. 675-678.

674 Rhodes, J.M., 1996, Geochemical stratigraphy of lava flows sampled by the Hawaii Scientific Drilling
675 Project: *Journal of Geophysical Research: Solid Earth*, v. 101, no. B5, p. 11729-11746.

676 Rodbell, D.T., Seltzer, G.O., Mark, B.G., Smith, J.A., and Abbott, M.B., 2008, Clastic sediment flux to
677 tropical Andean lakes: records of glaciation and soil erosion: *Quaternary Science Reviews*, v.
678 27, no. 15-16, p. 1612-1626.

679 Silva, L.C., 2014, Importance of climate-driven forest–savanna biome shifts in anthropological and
680 ecological research: *Proceedings of the National Academy of Sciences*, v. 111, no. 37, p.
681 E3832.

682 Stager, J.C., Cumming, B.F., and Meeker, L.D., 2003, A 10,000-year high-resolution diatom record
683 from Pilkington Bay, Lake Victoria, East Africa: *Quaternary Research*, v. 59, no. 2, p. 172-181.

684 Stansell, N.D., Licciardi, J.M., Rodbell, D.T., and Mark, B.G., 2017, Tropical ocean-atmospheric
685 forcing of Late Glacial and Holocene glacier fluctuations in the Cordillera Blanca, Peru:
686 *Geophysical Research Letters*, v. 44, no. 9, p. 4176-4185.

687 Stansell, N.D., Polissar, P.J., Abbott, M.B., Bezada, M., Steinman, B.A., and Braun, C., 2014,
688 Proglacial lake sediment records reveal Holocene climate changes in the Venezuelan Andes:
689 *Quaternary Science Reviews*, v. 89, p. 44-55.

690 Stríkis, N.M., Cruz, F.W., Cheng, H., et al., 2011, Abrupt variations in South American monsoon
691 rainfall during the Holocene based on a speleothem record from central-eastern Brazil:
692 *Geology*, v. 39, no. 11, p. 1075-1078.

693 Stríkis, N.M., Cruz, F.W., Barreto, E.A., et al., 2018, South American monsoon response to iceberg
694 discharge in the North Atlantic: *Proceedings of the National Academy of Sciences*, p.
695 201717784.

696 Tapia, P.M., Fritz, S.C., Baker, P.A., Seltzer, G.O., and Dunbar, R.B., 2003, A Late Quaternary
697 diatom record of tropical climatic history from Lake Titicaca (Peru and Bolivia):
698 *Palaeogeography, Palaeoclimatology, Palaeoecology*, v. 194, no. 1-3, p. 139-164.

699 Thompson, L.G., Mosley-Thompson, E., and Henderson, K.A., 2000, Ice-core palaeoclimate records
700 in tropical South America since the Last Glacial Maximum: *Journal of Quaternary Science*, v.
701 15, no. 4, p. 377-394.

702 Thompson, L.G., Mosley-Thompson, E., Davis, M.E., et al., 1995, Late glacial stage and Holocene
703 tropical ice core records from Huascarán, Peru: *Science*, v. 269, no. 5220, p. 46-50.

704 Tierney, J.E., and Pausata, F.S., 2017, Rainfall regimes of the Green Sahara: *Science advances*, v.
705 3, no. 1, p. e1601503.

706 Vaks, A., Woodhead, J., Bar-Matthews, M., et al., 2013, Pliocene–Pleistocene climate of the
707 northern margin of Saharan–Arabian Desert recorded in speleothems from the Negev Desert,
708 Israel: *Earth and Planetary Science Letters*, v. 368, p. 88-100.

709 Vera, C., Silvestri, G., Liebmann, B., and González, P., 2006, Climate change scenarios for
710 seasonal precipitation in South America from IPCC-AR4 models: *Geophysical Research*
711 *Letters*, v. 33, no. 13.

712 Vuille, M., Bradley, R.S., Werner, M., Healy, R., and Keimig, F., 2003, Modeling $\delta^{18}\text{O}$ in
713 precipitation over the tropical Americas: 1. Interannual variability and climatic controls: *Journal*
714 *of Geophysical Research: Atmospheres*, v. 108, no. D6.

715 Vuille, M., Burns, S.J., Taylor, B.L., et al., 2012, A review of the South American monsoon history as
716 recorded in stable isotopic proxies over the past two millennia: *Climate of the Past*, v. 8, no. 4,
717 p. 1309-1321.

718 Vuille, M., and Werner, M., 2005, Stable isotopes in precipitation recording South American summer
719 monsoon and ENSO variability: observations and model results: *Climate Dynamics*, v. 25, no. 4,
720 p. 401-413.

721 Wang, X., Auler, A.S., Edwards, R.L., et al., 2007, Millennial-scale precipitation changes in southern
722 Brazil over the past 90,000 years: *Geophysical Research Letters*, v. 34, no. 23.

723 Wang, X., Edwards, R.L., Cruz Jr, F.W., Cheng, H., and Auler, A.S., 2008, Millennial-scale climate
724 variability recorded in Brazilian speleothems.

725 Wong, C.I., Banner, J.L., and Musgrove, M., 2011, Seasonal dripwater Mg/Ca and Sr/Ca variations
726 driven by cave ventilation: Implications for and modeling of speleothem paleoclimate records:
727 *Geochimica et Cosmochimica Acta*, v. 75, no. 12, p. 3514-3529.

728 Wong, C.I., and Breecker, D.O., 2015, Advancements in the use of speleothems as climate archives:
729 *Quaternary Science Reviews*, v. 127, p. 1-18.

730 Wortham, B.E., Wong, C.I., Silva, L.C., et al., 2017, Assessing response of local moisture conditions
731 in central Brazil to variability in regional monsoon intensity using speleothem $^{87}\text{Sr}/^{86}\text{Sr}$
732 values: *Earth and Planetary Science Letters*, v. 463, p. 310-322.

733 Zhou, H., Feng, Y., Zhao, J., Shen, C., You, C., and Lin, Y., 2009, Deglacial variations of Sr and
734 $^{87}\text{Sr}/^{86}\text{Sr}$ ratio recorded by a stalagmite from Central China and their association with past
735 climate and environment: *Chemical Geology*, v. 268, no. 3-4, p. 233-247.

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Figures

Figure 1: Hydroclimate setting of study region

Maps of $\delta^{18}\text{O}$ records from cave (stars) and lake (triangle) sites and Global Network of Isotopes in Precipitation (GNIP) sites with percent of annual precipitation delivered during austral spring (SON, top), summer (DJF, middle), and fall (MAM, (bottom) and monthly mean precipitation amount (bottom) and

746 precipitation $\delta^{18}\text{O}$ (top) at GNIP sites (circles). PAR – Paraíso, RN – Rio Grande do Norte, COD – El
747 Condor, DIA – Diamante, PUMA – Laguna Pumacocha, HUG – Huagapo, LG – Lapa Grande, TM –
748 Tamboril, JAR – Jaraguá, BTV – Botuverá. Data from the Global Precipitation Climatology Centre
749 (GPCC) Full Data Reanalysis Version 7 0.5x0.5 Monthly Total Averaged from 1981 to 2010. Pink filled
750 site markers indicate localities where Sr isotope records were developed for this study. GNIP sites include
751 Manuas (1), Brasilia (2), Campo Grande (3), Porto Alegre (4), and Marcapomacocha (5). Monthly mean
752 precipitation data was calculated using the CPC Merged Analysis of Precipitation (includes NCEP
753 Reanalysis) using the closest grid points to GNIP stations.

Figure 2: Compilation of SAMS domain speleothem and lake core $\delta^{18}\text{O}$ records

Different shading in each record indicates a different speleothem from cave site. El Condor Cave (Cheng et al., 2013), Diamante Cave (Cheng et al., 2013), Huagapo Cave (Kanner et al., 2013), Laguna Pumacocha (Bird et al., 2011), Rio Grande do Norte (Cruz et al., 2009), Botuverá Cave (maroon curve - BTV21a, Bernal et al., 2016; yellow curve - BTV2, Cruz et al., 2005; pink curve - BT3, Wang et al., 2007), Paraíso Cave (Wang et al., 2017), Tamboril Cave (pink curve – TM0, Wortham et al., 2017; periwinkle curve – TM2, Wortham et al., 2017; purple curve – TM6, this study), Lapa Grande Cave (Strikis et al., 2011), Jaraguá Cave (dark green curve – JAR07, Novello et al., 2017, mustard green curve – JAR04, Novello et al., 2018, lime green curve – JAR02, Novello et al., 2018b submitted).

Figure 3: Conceptual model of $^{87}\text{Sr}/^{86}\text{Sr}$ proxy

$^{87}\text{Sr}/^{86}\text{Sr}$ variability as a proxy of water rock interaction associated with differences in water residence time under wet and dry conditions. Under dry conditions (left panel), flow through the matrix dominates, enabling greater water rock interaction and greater extent of evolution from the initial $^{87}\text{Sr}/^{86}\text{Sr}$ composition acquired from the soil toward that of the bedrock. Under wet conditions (right panel), flow through conduit flow paths occurs, limiting water rock interaction and preserving the $^{87}\text{Sr}/^{86}\text{Sr}$ signature acquired from the soil. Figure modified from Wortham et al., 2017.

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785 **Figure 4: Principal Components Analysis**786 First principal component from a PCA (pink curve) of compiled $\delta^{18}\text{O}$ records from the region, shown

787 relative to austral summer (January) insolation at 20°S (blue curve) (top panel). Weighting of each site on

788 PC1 and PC2 (bottom panel).

Figure 5: Water-rock interaction modelling

Sr-isotope ($^{87}\text{Sr}/^{86}\text{Sr}$) values of soil (triangles), bedrock (squares), dripwater (circles), and speleothems (asterisks) from Paraíso, Tamboril, Jaraguá, and Botuverá caves (narrow rectangular panels) shown with WRI modelling for Paraíso, Tamboril, and Jaraguá caves (large square panels). Distinct drip sites within each cave are represented by different colors. Grey triangles indicate the starting composition of dripwater. The evolution of dripwater compositions is modelled from the most (solid line) and least (dashed line) radiogenic soil and bedrock values and lowest Sr/Ca value (i.e., least evolved) measured in dripwater assuming a Sr K_D value of 0.03 (Tesorriero and Pankow, 1996; Terakado and Taniguchi, 2006). Dot-dashed grey line in Paraíso cave represents an idealistic WRI scenario fit to data by changing starting parameters. The blue vertical arrow indicates the evolution of dripwater compositions due to PCP. Shaded area within WRI lines indicates range of possible dripwater values that WRI can account for.

Figure 6: Botuverá Cave proxy records

Comparison of BTV21a Sr/Ca, $\delta^{18}\text{O}$, (Bernal et al., 2016) and $^{87}\text{Sr}/^{86}\text{Sr}$ (this study), and BTV2 (Cruz et al., 2005) and BTV3 (Wang et al., 2007) $\delta^{18}\text{O}$ records.

Figure 7: Normalized $^{87}\text{Sr}/^{86}\text{Sr}$ records

Axes flipped to denote “wet” down interpretation. Yellow bar indicates onset towards dry conditions and sustained drying in individual speleothem samples during mid-Holocene; dashed portion of line indicates a growth hiatus where precise onset of drying conditions cannot be assigned.

Figure 8: Comparison of similar records

Comparison of PC1, January Insolation at 20°S, Jaraguá $^{87}\text{Sr}/^{86}\text{Sr}$, Paraíso and Rio Grande do Norte $\delta^{18}\text{O}$, and Cariaco Basin %Ti. Axes are flipped for PC1, insolation, $^{87}\text{Sr}/^{86}\text{Sr}$, and Caricao Basin to readily compare similar trends.

818 **Supplemental Figures**

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821 **S1: TM6 Age Model**

822 COPRA developed age-depth model for TM6 from Tamboril Cave (upper panel). The distinct

823 hiatuses are delineated by near-zero slopes and marked as gaps in model. TM6 image with

locations of U/Th ages (years BP) marked (lower panel). Greyed age is omitted from TM6 age model.

S2: Botuverá $\delta^{18}\text{O}$ records

Comparison of BTV2 (yellow), BTV3a (pink), and BTV21a (maroon) speleothem $\delta^{18}\text{O}$ records.

Diamonds of respective colors indicate U/Th ages and associated error.

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837 S3: Raw $^{87}\text{Sr}/^{86}\text{Sr}$ records

838 Time series of PC1 and Paraíso, Tamboril, Jaraguá, and Botuverá $^{87}\text{Sr}/^{86}\text{Sr}$ records. $^{87}\text{Sr}/^{86}\text{Sr}$ axes are
 839 flipped so that inferred “wet” conditions are oriented downward.

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843 **S4: First Three PC's of PCA**

844 First three PC's of PCA utilized in this study. PC is marked on y-axis, with percent of variability in all
845 records explained by each PC.

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850 S5: PCA Sensitivity Testing

851 First three PC's of PCAs (A and B panels) utilizing varied combinations of cave sites. PC is marked on y-

852 axis, with percent of variability in all records explained by each PC.

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855 **S6: $\delta^{13}\text{C}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ Comparison**

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Tables

Table 1

Cave	Location	Host Geology	Primary Lithology	Bedrock Overburden (m)	Soil Thickness (cm)	Vegetative Cover	References
Paraíso	4°S, 55°W (60 m asl)	Itaituba Fm / Tapajós Group (Paleozoic)	Limestone	15	> 100	Thick tropical forest cover year round	Wang et al., 2017; this study
Jaraguá	21°S, 56°W (570 m asl)	Bocaina Fm / Corumbá Group (Neoproterozoic)	Dolomite	~5	20	Semi-deciduous cover and Cerrado surrounding	Novello et al., 2017
Tamboril	16°S, 47°W (600 m asl)	Sete Lagos Fm / Bambuí Group (Neoproterozoic)	Dolomite (w/ interbedded marls and siltstone)	~10 - 50	~10 - 20	Semi-deciduous cover and Cerrado surrounding	Silva et al., 2001; Wortham et al., 2017; this study
Botuverá	27°S, 49°W (230 m asl)	Botuvera Fm / Brusque Group (Neoproterozoic)	Dolomite and carbonate sediments within metavolcanoclastic sequence	~ 70	>100	Thick tropical Atlantic rainforest	Cruz et al., 2007; Bernal et al., 2016

Table 2

Sample ID	Depth	²³⁸ U	± (2σ)	²³² Th	± (2σ)	δ ²³⁴ U	± (2σ)	(²³⁰ Th/ ²³⁸ U)	± (2σ)	²³⁰ Th/ ²³² Th	± (2σ)	Age (yr)	± (2σ)	Age (yr)	± (2σ)	δ ²³⁴ U initial	± (2σ)	Age
	(mm)	(ng/g) ^a		(pg/g) ^a		(per mil) ^b		activity		ppm atomic		(uncorr) ^c		(corr) ^d		(per mil) ^e		(yr B.P)
TM6-T	2	8380	170	944	19	832.4	0.6	0.01718	0.00008	2420	11	1026	5	1025	5	834.8	0.6	958
TM6-16	16	6840	140	2230	50	840.5	0.7	0.02069	0.00014	1006	12	1232	8	1226	9	843.4	0.7	1159
TM6-36	36	5070	100	300	20	874.8	1.2	0.02385	0.00017	6400	400	1395	10	1394	10	878.3	1.2	1327
TM6-55	55	6560	130	186	19	852	0.6	0.0253	0.00015	14200	1400	1498	9	1498	9	855.6	0.6	1431
TM6-80	80	6950	140	710	30	815.3	1.6	0.02767	0.00013	4290	130	1673	8	1672	8	819.1	1.6	1605
TM6-93	93	3660	70	2770	60	849.9	0.8	0.0587	0.0002	1233	9	3509	15	3496	16	858.4	0.8	3429
TM6-115	115	3900	80	1270	30	870.7	0.8	0.0641	0.0002	3130	40	3794	13	3788	14	880	0.8	3721
TM6-117	117	17300	300	870	30	742.7	0.8	0.0887	0.0002	28000	600	5676	15	5675	15	754.7	0.8	5608
TM6-138	138	7050	140	3820	80	727.3	1.7	0.0904	0.0003	2646	13	5839	19	5830	20	739.3	1.8	5763
TM6-153	153	3040	60	160	20	582.5	0.7	0.1043	0.0004	32000	4000	7400	30	7400	30	594.7	0.7	7333
TM6-173	173	2570	50	210	20	601.6	1	0.1131	0.0004	22000	2000	7950	30	7950	30	615.3	1	7883
TM6-B	209	4080	80	370	7	607.3	1	0.1346	0.0004	23530	70	9490	30	9490	30	623.8	1	9423
TM6-191	191	2570	50	18300	400	607.9	0.8	0.1127	0.0004	252.2	0.9	7890	30	7760	70	621.3	0.8	7693

Notes:
^aReported errors for ²³⁸U and ²³²Th concentrations are estimated to be ±1% due to uncertainties in spike concentration; analytical uncertainties are smaller.
^bδ²³⁴U = ((²³⁴U/²³⁸U)_{activity} - 1) x 1000.
^c[²³⁰Th/²³⁸U]_{activity} = 1 - e^{-λ²³⁰T} + (δ²³⁴U_{measured}/1000)[λ₂₃₀/(λ₂₃₀ - λ₂₃₄)](1 - e^{-(λ²³⁰ - λ²³⁴)T}), where T is the age. "Uncorrected" indicates that no correction has been made for initial ²³⁰Th.
^dAges are corrected for detrital ²³⁰Th assuming an initial ²³⁰Th/²³²Th of (XX±XX) x 10⁻⁶.
^eδ²³⁴U_{initial} corrected was calculated based on ²³⁰Th age (T), i.e., δ²³⁴U_{initial} = δ²³⁴U_{measured} X e^{λ²³⁴T}, and T is corrected age.
^fB.P. stands for "Before Present" where the "Present" is defined as the January 1, 1950 C.E.
Decay constants for ²³⁰Th and ²³⁴U are from Cheng et al. (2013); decay constant for ²³⁸U is 1.55125 x 10⁻¹⁰ yr⁻¹ (Jaffey et al., 1971).
TM6-191 was not included in the age model due to due non-monotonic nature