Assessing response of local moisture conditions in central Brazil to variability in regional monsoon intensity using speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values

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Abstract

Delineating the controls on hydroclimate throughout Brazil is essential to assessing potential impact of global climate change on water resources and biogeography. An increasing number of monsoon reconstructions from $\delta^{18}\text{O}$ records provide insight into variations in regional monsoon intensity over the last millennium. The strength, however, of $\delta^{18}\text{O}$ as a proxy of regional climate limits its ability to reflect local conditions, highlighting the need for comparable reconstructions of local moisture conditions. Here, speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values are developed as a paleo-moisture proxy in central Brazil to complement existing $\delta^{18}\text{O}$ -based reconstructions of regional monsoon intensity. Speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values are resolved using laser ablation and conventional solution mass spectrometry at high resolution relative to existing (non- $\delta^{18}\text{O}$ -based) paleo-moisture reconstructions to allow comparisons of centennial variability in paleo-monsoon intensity and paleo-moisture conditions. Variations in speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values from Tamboril Cave are interpreted to reflect varying extents of water interaction with the carbonate host rock, with more interaction resulting in greater evolution of water isotope values from those initially acquired from the soil to those of the carbonate bedrock. Increasing speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values over the last millennium suggest progressively less interaction with the carbonate host rock likely resulting from higher infiltration rates, expected under wetter conditions. Increasingly wetter conditions over the last millennium are consistent with an overall trend of increasing monsoon intensity (decreasing $\delta^{18}\text{O}$ values) preserved in many existing $\delta^{18}\text{O}$ records from the region. Such a trend, however, is absent in $\delta^{18}\text{O}$ records from our site (central Brazil) and Cristal Cave (southeast Brazil), suggesting the existence of divergent (relevant to $\delta^{18}\text{O}_{\text{precip}}$) shifts in the climate patterns within and outside the core monsoon region.

Keywords: stable isotopes, strontium isotopes, Holocene, South American Summer Monsoon

1. Introduction

Brazil is a region with rich biodiversity and a society leveraging water resources to drive electrical production and an agricultural economy, making it particularly sensitive to changes in climate and water availability. Much effort has gone into reconstructing past variations in hydroclimate, with a large effort to characterize past variations in the intensity of the South American Monsoon System (SAMS). The SAMS is an important source of precipitation across much of tropical and sub-tropical South America (Fig. 1), and is predominantly driven by seasonal variations in insolation-driven convection centered over the southern Amazon Basin (herein referred to as the monsoon core region) (Zhou and Lau, 1998; Vuille et al., 2012). Past variations in moisture amount are typically attributed to changes in the intensity of the SAMS because the SAMS is such a critical climate phenomena in the region (e.g., Bird et al., 2011; Vuille et al., 2012; Kanner et al., 2013). Evaluation, however, of the assumption that past variations in local moisture conditions are dominantly dictated by past variations in monsoon intensity is typically absent in many studies.

Figure 1: SAMS precipitation patterns over study region (a) Shading reflects average austral summer (DJF) precipitation as a percentage of the annual precipitation (data spanning 1981-2010 from Global Precipitation Climatology Project). Black arrows represent the near-surface (850

hPa) winds that guide easterly transport of moisture across the Amazon Basin, and subsequent deflection of winds to the south by the Andean Mountain range creating the South American low-level jet. (b) Locations of existing late Holocene paleo-SASM reconstructions with decadal to subdecadal resolution indicated by circles. HG-Huagapo Cave (Kanner et al. 2013), CS-Cascayunga Cave (Reuter et al. 2009), LP-Lake Pumacocha (Bird et al. 2011), QC-Quelccaya Ice Cap (Thompson et al. 2013), CC-Cristal Cave (Vuille et al. 2012), DV-Diva de Maura record (Novello et al. 2012), ALCR – Pau d’Alho and Curupira Cave (Novello et al., 2016). The mean positions of the ITCZ and the SACZ during DJF are shown as black dashed lines. Star indicates the location of the study site, Tamboril Cave (TM).

Records of $\delta^{18}\text{O}$ variability preserved in ice cores, lake sediments, and speleothems provide the basis for much of our understanding of monsoon variability during the Holocene (Fig. 1). These records are well constrained (e.g., counting of annual lamina or varves and U - series dating) and sampled at fine (e.g., sub-millimeter) increments to yield highly resolved time series. Records on both the western and eastern side of the continent provide the means to assess the regional signal in monsoon variability. $\delta^{18}\text{O}$ variability acts as a proxy of regional monsoon intensity, reflecting the degree of water vapor ^{18}O depletion (with respect to ^{16}O) associated with convective activity over the Amazon Basin (i.e., monsoon intensity) (Vuille and Werner, 2005; Vuille et al., 2012). Vapor over the Amazon Basin is then exported to downstream regions, such as the Andes and the La Plata Basin, carrying with it the signature of upstream convective activity. It is well established that $\delta^{18}\text{O}_{\text{precip}}$ from across the region is strongly influenced by monsoon intensity based on the high correlation between precipitation amount over the monsoon core region and $\delta^{18}\text{O}_{\text{precip}}$ occurring across the region, making $\delta^{18}\text{O}$ values an undisputed proxy of regional monsoon strength (Vuille and Werner 2005; Vuille et al., 2012). On the other hand, the strength of $\delta^{18}\text{O}$ as a proxy of regional climate limits its ability to reflect local conditions at some (Vuille et al., 2012), but not all (Moquet et al., 2016), sites outside of the monsoon core region. Figure 2 further investigates the potential for local precipitation amount to be decoupled from monsoon strength by documenting the spatial variability in the sign (i.e., positive/negative), strength, and significance of the correlation between precipitation amount and various indices of monsoon strength. It is apparent that the nature of the relationship between monsoon intensity and precipitation is not homogenous across the region (Fig. 2).

Figure 2: Correlation between monsoon strength and precipitation amount. A, B, and C) Correlation between Monsoon Index I, II, and III and gridded precipitation from the Climate Prediction Center Merged Analysis of Precipitation (CMAP). D, E, and F) Correlation between Monsoon Index I, II, and III and precipitation gauge data. All correlations calculated for the interval of 1979-2013. Positive values indicate positive correlation between monsoon intensity and precipitation amount. Monsoon Index I calculated as the average DJF CMAP precipitation

amount over the monsoon core region (-2.5 to -17.5°N, -47.5 to -72.5°E, black-dashed box) after Vuille et al. (2012). Monsoon Index II calculated as the negative anomaly from climatological average of outgoing longwave radiation (OLR) spatially averaged over the monsoon core region following Vuille and Werner (2005). Monsoon Index III calculated as vertical wind shear ($u_{850}-u_{250}$) spatially averaged over the west tropical Atlantic Ocean (7.5 to -2.5°N, -20 to -45°E, blue-dashed box) following Vuille and Werner (2005). OLR and wind data are from the Modern Era Retrospective analysis for Research and Applications. Shading represents statistically significant ($p < 0.05$) regions of correlation. Representative precipitation gauge stations with close proximity to existing monsoon reconstructions (Fig. 1) and continuous coverage over the time period 1979-2013 were selected from the Global Historical Climatology Network (stations: PEM00084673 [1], PEM00084686 [2], VE000080438 [3], BR038983570 [4], BR00E3-0520 [5]) and the Bank of Meteorological Data for Education and Research (station 83377 [6]; GHCN, 2015).

Existing $\delta^{18}\text{O}$ -based reconstructions of monsoon intensity from across the region document a coherent signal of monsoon variability from the early to late Holocene and over the last millennium. Decreasing $\delta^{18}\text{O}$ values from the early to late Holocene suggest monsoon intensification in response to increasing summer insolation in the Southern Hemisphere (Bird et al., 2011; Kanner et al., 2013). Other, non- $\delta^{18}\text{O}$ -based, reconstructions of Holocene moisture conditions derived from vegetation and sediment records generally document increasingly wetter conditions from the early to late Holocene, which indicates monsoon strength is likely a strong driver of millennial variations in local moisture conditions (e.g., Rodrigues et al., 2016). During the last millennium, the offset in $\delta^{18}\text{O}$ values between the Little Ice Age (stronger monsoon) and Medieval Climate Anomaly (weaker monsoon) is commonly replicated finding records from the region (e.g., Vuille et al., 2012). Also present is the prevalence of a decreasing trend in $\delta^{18}\text{O}$ values over the last millennium (Fig. 3). The resolution of non- $\delta^{18}\text{O}$ -based paleo-moisture records (e.g., vegetation and sediment records) is generally insufficient to assess sub-millennial variability in moisture conditions. A review, however, of high quality pollen records document a diverse pattern of vegetation response to climate change in the region over the last 2 millennia, highlighting complexity at the local-scale (Sup. Fig. S1; Flantua et al., 2016). These findings emphasize the need for higher resolution reconstructions of last millennial moisture conditions from moisture-sensitive proxies, other than $\delta^{18}\text{O}$ records, to enable assessment of the coupling between local moisture conditions and regional monsoon intensity on time scales of decades to millennia.

Figure 3. Last millennial trends in $\delta^{18}\text{O}$ records. Standardized $\delta^{18}\text{O}$ variability (black) is shown with trend (dashed red) over the last 1 ky for each record (Sup. Table S1) along with standardized variability and trend in %Ti from Cariaco Basin sediments and changes in solar forcing (red) resulting from reconstructed solar variability (blue; Delague and Bard, 2010) and evolution of January insolation at 5°S (black; Lasker et al., 2004). Trend slope and h and p values reported for Mann-Kendall tests of trend significance. h=0 indicates accept null hypothesis (i.e., trend is absent); h=1 reject null hypothesis (i.e., trend is present).

In this study, we develop a multi-proxy record of the last millennium hydroclimate from Tamboril Cave located within the Amazon-Cerrado transition zone of central Brazil. Analysis of speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values at a resolution (multi-decadal) higher than most existing non- $\delta^{18}\text{O}$ -based moisture reconstructions in the area enables reconstruction of local moisture conditions at a more comparable resolution to $\delta^{18}\text{O}$ records. Comparison of TM0 $^{87}\text{Sr}/^{86}\text{Sr}$ variability to $\delta^{18}\text{O}$

values in the same stalagmite and to existing $\delta^{18}\text{O}$ records enables assessment of the coupling between monsoon intensity and paleo-moisture conditions over the last millennium.

2. Setting

Tamboril Cave (16°S, 47°W) is located in the northern part of Minas Gerais, Brazil. The cave consists of large hallways and high ceilings (up to 30 m), and is extensively decorated. Tamboril Cave is ~700 m in length and 55 m wide at the widest part (Silva et al. 2001), and bedrock overburden is roughly estimated to range between 10 and 50 m. The cave is formed in the Sete Lagoas Formation (Fm) of the Neoproterozoic Bambui Group in the Brasilia Belt (Fig. 4; Silva et al., 2001). The Sete Lagoas Fm consists of dolostone, limestone, and marls interbedded with silt and clay layers (e.g., Alvarenga et al., 2014). The back portion of the cave, beyond where any sampling was conducted, intersects a thrust fault that puts the underlying Paranoa Group in contact with the Sete Lagoas Fm (Silva et al., 2001). The Bambui and Paranoa Groups in the Brasilia Belt and the nature of the contact has been described stratigraphically and isotopically by Alvarenga et al. (2014). Soils in the general area result from the weathering of one of three erosional surfaces: South American, Velhas I, or Velhas II (Marques et al., 2004). The Cretaceous-age South American surface is comprised of clayey sediments derived from igneous and metamorphic material transported from a region near Brasilia, to the northwest of the cave site. Tectonic uplift and sea level regression during the Tertiary exposed underlying sedimentary and basaltic bedrock, in which Velhas I and Velhas II formed during prolonged arid intervals (Marques et al., 2004).

Figure 4: Tamboril Cave Setting A.) Geologic map from the Ministry of Mines and Energy, Geological Service of Brazil. Unit C described as siltstone with interbedded sandstone and mudstone (ar) and bodies of calcareous limestone (cc). B. Google Earth Image (oriented north) of carbonate mound in which the cave is found. Coarse overlay of cave indicated by yellow line (~100 m). C) Schematic stratigraphy column of Bambuí-Paranoá contact in the area after Alvarenga et al. (2014). D) Photo of 70 cm soil pit sampled above the cave. E) Cave map courtesy of a cave group from Brasília. F) Panoramic photo of mine labeled in C, person for scale on right.

The region is semi-humid with a mean temperature at the site of 22.5°C and annual rainfall ranging from 1200 to 1600 mm. Rainfall is mostly distributed from November to March (Sup.Fig. S2), and primarily controlled by the SASM and South Atlantic Convergence Zone (Vera et al., 2006; Garreaud et al., 2009). Native semi-deciduous forests cover the soils over this cave. This is the typical ecosystem associated with limestone and dolomite outcrops in the region (Felfili et al. 2007; Silva et al. 2010), forming a continuous tree cover during the rainy summer season but becoming partly leafless in the dry winter season. There is pronounced seasonal variability in $\delta^{18}\text{O}_{\text{precip}}$ values obtained from the nearby Global Isotopes in Precipitation station in Brasilia, with lowest values occurring during monsoon months (Supp. Fig. S2). The relationship between monthly average precipitation amount and $\delta^{18}\text{O}_{\text{precip}}$ values is seasonally variable, with low $\delta^{18}\text{O}_{\text{precip}}$ values occurring in the waning monsoon months (e.g., March and April) despite low precipitation amounts relative to the early monsoon months (e.g., November and December). This observation is consistent with moisture derived from the Amazon Basin that evolves over the monsoon seasons toward more depleted values. The correlation between monthly precipitation amount and $\delta^{18}\text{O}_{\text{precip}}$ values is weak ($r^2=0.2$), and there is no relationship between year-to-year variation in precipitation accumulated during monsoon months (November to March) and precipitation-weighted average $\delta^{18}\text{O}_{\text{precip}}$ values (Supp.Fig. S2). These observations suggest that $\delta^{18}\text{O}_{\text{precip}}$ values track monsoon intensity, but may not be the best proxy of interannual variations in local precipitation amount.

3. Methods

A speleothem (TM0) was collected ~220 m from cave entrance in 2011 in one of the largest and best-preserved rooms in the cave (Fig. 4). TM0 is 80 cm long, generally symmetrical around a central growth axis, and consists of white aragonite needles. Analyses of X-ray diffraction and laser Raman spectroscopy indicate the sample is pure aragonite as no trace amounts of calcite were detected. Petrographic evaluation of thin sections indicates the lack of clear boundaries or corrosive horizons throughout the stalagmite, indicating the absence of any major hiatuses.

Figure 5: Age-depth model. Age-depth relationship of 15 dated horizons. Red circles indicate data on which the age-depth model was calculated, whereas gray circles indicate data not included in the age model. In regions where there were several ages for closely spaced horizons, a representative date was selected for inclusion in the age model that minimized the distance from a linear fit through data from adjacent regions. Uncertainty on ages is smaller than the marker size.

The age model for TM0 is based on U/Th dating of 12 distinct horizons conducted at MIT, UC Davis (UCD) and Berkeley Geochronology Center (BGC) (Fig. 5) (Sup. Tables S2 and S3). Duplicate measurements of the same depth horizon were averaged and the error was propagated. Depth-age relationships were calculated using the COPRA algorithm (Breitenbach et al., 2012). Age models for all previously published records were re-calculated from raw age-depth and depth-proxy data using the COPRA algorithm to enable objective comparison of TM0 $\delta^{18}\text{O}$ values with existing records (Supplemental Material). Median values of statistics calculated for each of 100 realizations of the age model are reported. Such incorporation of age uncertainties into construction of the proxy time series tends to dampen variability in the records, reflecting the variability that can be constrained when taking into consideration age uncertainty (Breitenbach et al., 2012). Thus, there are differences between time series plotted here vs. in their original publication (Supplemental Material).

For stable isotope (O and C) analyses, approximately 80 μg of powder was micro-milled continuously over 1.5 mm intervals (~ 3 yr. resolution) along the growth axis. Powders were analyzed for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ using a GVI Optima IRMS within the Dept. of Earth and Planetary Sciences at the University of California, Davis (UCD) and a Thermo Finnegan MAT 253 with a Kiel Device within the Dept. of Geosciences at The University of Texas at Austin (UT). Uncertainty for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ is 0.07‰ and 0.04‰, respectively, for samples analyzed at UCD based on the standard deviation of 64 replicate analyses of an internal standard. For samples analyzed at UT, uncertainty for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ is 0.06‰ and 0.03‰, respectively, based on the standard deviation of 11 replicate analyses of an internal standard.

For solution-mode $^{87}\text{Sr}/^{86}\text{Sr}$ analyses, approximately 3-4 mg of powder was hand-milled using a dental drill from along the growth axis of TM0 at discrete intervals spaced 0.5-1 cm (~ 20 year resolution). Speleothem powders were processed using ion exchange chemistry following the methods of Montañez et al. (2000). Solution-mode Multiple Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS) analyses of strontium isotopic compositions were performed using a Nu Plasma HR instrument at the Interdisciplinary Center for Plasma Mass Spectrometry at University of California, Davis (UCD/ICPMS, $n=10$) and a Nu Plasma II instrument at the Facility for Isotope Research and Student Training (FIRST) at Stony Brook University ($n=84$). Both facilities were equipped with a DSN-100 desolvating nebulizer. Follow up analyses were made using an Isotopx Phoenix Thermal Ionization Mass Spectrometer in the Center for Isotope Geochemistry (CIG) Boston College (BC, $n = 16$). Uncertainty for analyses performed at UCD/ICPMS, FIRST, and CIG is 3.3×10^{-5} ($n=32$), 2.6×10^{-5} ($n=22$), and 0.6×10^{-5} ($n=24$), respectively, which is the 2SD of replicate analyses of NIST SRM987. Laser ablation (LA-MC-ICPMS) was additionally used to analyze speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values at millimeter resolution at UCD/ICPMS, with an uncertainty of 20×10^{-5} based on the 2SD of replicate analysis of a marine calcium carbonate internal standard. See Supplemental Material for additional analytical details.

Soils, bedrock, and dripwater were collected from Tamboril Cave during subsequent trips to characterize the potential $^{87}\text{Sr}/^{86}\text{Sr}$ endmembers in the cave setting. A 50 cm pit was dug above the cave ~ 50 m south-southwest from the cave entrance, and soils were sampled every 10-12 cm. Water was collected from three drip sites during a single dry season. Bedrock was collected at the cave entrance and near the lake. Soil samples were leached with 1 M ammonium

acetate buffered to a pH of 8 to determine the isotopic value of the exchangeable Sr (Wong and Banner, 2010; Oster et al., 2010; Wong et al., 2011). Bedrock samples were progressively leached with 1 M ammonium acetate, 4% acetic acid, and 6N HCl. Leachates were processed using ion exchange chemistry and analyzed for $^{87}\text{Sr}/^{86}\text{Sr}$ values as described above.

Soil mineralogy was assessed using x-ray diffraction (XRD) at the Soil Sciences Lab at the University of California, Davis. All XRD work was done with a Rigaku Ultima diffractometer operated at 40 kV and 40 mA to produce Cu Ka (alpha) radiation (Supplemental Material). Soil and bedrock samples were analyzed for trace element (Nb, Zr, Y, Sr, Rb, Th, Pb, Ga, Zn, Ni, Cr, V, Ba, Ce, La) concentrations using x-ray fluorescence (XRF) at the Ronald B. Gilmore XRF Lab, University of Massachusetts, Amherst. Concentrations were obtained using pressed powder pellets on a Philips PW2400 sequential spectrometer following the methods of Rhodes et al. (1996) (Supplemental Material). Bedrock samples were also analyzed for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values at the Stable Isotope Laboratory of Lamont-Doherty Earth Observatory at Columbia University.

4. Results

TM0 $\delta^{18}\text{O}$ values fluctuate between -6.0 and -4.5‰ with rare excursions reaching values as low as -7‰ (Fig. 3, Sup. Table S4), while $\delta^{13}\text{C}$ values fluctuate between -11.5 and -8‰ (Fig. 6). There is a weak correlation between $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values along the growth axis ($r^2=0.25$), and $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ measured along the same horizontal horizon ($n=3$) are within analytical uncertainty. The $\delta^{18}\text{O}$ value of aragonite that would precipitate from modern monsoon rainfall under equilibrium conditions is -5‰, which is consistent with the range of TM0 $\delta^{18}\text{O}$ values (Supplemental Material). Further, TM0 $\delta^{18}\text{O}$ values tentatively replicate a small overlapping portion of another speleothem sample collected from Tamboril cave (Supp. Fig. S3). These data suggest that kinetic fractionation is not a dominant control on speleothem isotope composition. A trend in $\delta^{18}\text{O}$ values over the last millennium is absent in our TM0 record and the speleothem record from Cristal Cave in southeast Brazil, despite the presence of a significant trend in many records from the monsoon region (Fig. 3). Trends of decreasing $\delta^{18}\text{O}$ values in many of the other records are consistent with the trend of i) increasing summer insolation across the last millennium, and ii) %Ti in sediments from the Cariaco Basin, commonly interpreted to reflect meridional migration of the Intertropical Convergence Zone (ITCZ) (Fig. 3).

Figure 6: $^{87}\text{Sr}/^{86}\text{Sr}$ of Tamboril Cave setting and TM0 time series. A) $^{87}\text{Sr}/^{86}\text{Sr}$ values are shown for leaches of soils collected above Tamboril Cave (n=4) and cave dripwater (n=3) and leaches of cave bedrock (n=2). Also plotted are literature values measured in unaltered rocks from the Bambui and Paranoa Group limestone (Alvarenga et al., 2014) and Saharan dust collected in Barbados (Pourmand et al., 2014). B) TM0 $\delta^{18}\text{O}$, $\delta^{13}\text{C}$, $^{87}\text{Sr}/^{86}\text{Sr}$ (discrete measurements as circles; laser ablation measurements as curves) and $\delta^{234}\text{U}$ values are shown along with growth rate (black). Individual transects of laser ablation $^{87}\text{Sr}/^{86}\text{Sr}$, shown as different colors beneath the interpolated $^{87}\text{Sr}/^{86}\text{Sr}$ laser data (red line) and solution-mode $^{87}\text{Sr}/^{86}\text{Sr}$ values (black dots). Curves for $\delta^{18}\text{O}$, $\delta^{13}\text{C}$, and $^{87}\text{Sr}/^{86}\text{Sr}$ values are based on an interpolation to a common 10-year time step. Age constraints (magenta) are shown with symbol width reflecting uncertainty. BP = years before 1950 C.E.

Bedrock carbonate $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values are consistent with those previously measured in the Lower Sete Lagoas Fm (Sup. Fig. S4). Bedrock leachate $^{87}\text{Sr}/^{86}\text{Sr}$ values range from 0.712 to 0.719 (Table 1), which are high relative to primary $^{87}\text{Sr}/^{86}\text{Sr}$ values (0.707-0.708) measured in rocks from the Bambui and Paranoa Group in the area (Alvarenga et al., 2014). High $^{87}\text{Sr}/^{86}\text{Sr}$ values coupled with low Sr concentrations (~50 ppm) indicated by the XRF results are indicative of post-depositional alteration of the carbonate-dominated bedrock as previously documented in the area (Alvarenga et al., 2014). Progressively stronger leaching agents generally resulted in lower $^{87}\text{Sr}/^{86}\text{Sr}$ values (Table 1), which was counter to the expectation of increasing values with increasing dissolution of more radiogenic and difficult to dissolve phases. An explanation for lower $^{87}\text{Sr}/^{86}\text{Sr}$ values with stronger leaching agents is not obvious, but could perhaps reflect i) greater proportion of carbonate mineral dissolution with increasing strength of the leaching agent, or ii) dissolution of a different diagenetic phases by each leaching agent (e.g., different generations and solubility of dolomites vs. calcite). Regardless of the origin of the $^{87}\text{Sr}/^{86}\text{Sr}$ leaching trend, these results suggest that a greater bedrock influence on dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values would result in lower $^{87}\text{Sr}/^{86}\text{Sr}$ values.

The bulk and sand-size fraction of the soils are mostly quartz (Supp. Fig. S5). The clay-size fraction is dominated by quartz, but illite, kaolinite and gibbsite are also present. The Th/Zr ratios of the soils collected above Tamboril (~0.05) are consistent with those measured in soils of the South American erosional surface, which were derived from igneous and metamorphic rocks (Supp. Fig. S5) (Marques et al., 2004). Th and Zr are refractory in soils, meaning that Th/Zr ratios will reflect the original parent material, although caution is taken in this interpretation as Th and Zr concentrations in soils can be grain size dependent (e.g. McGee et al., 2016). Soil leachate $^{87}\text{Sr}/^{86}\text{Sr}$ values range from 0.7309 to 0.7333 (n=4) and are consistent with soils

weathering from igneous parent material. Importantly, soil $^{87}\text{Sr}/^{86}\text{Sr}$ values are substantially higher than those of cave bedrock leachates.

Cave dripwater (0.718 to 0.723) and speleothem (0.718 to 0.725) $^{87}\text{Sr}/^{86}\text{Sr}$ values fall between the soil and bedrock end-members (Fig. 6 and Table 1). There is good agreement between solution mode $^{87}\text{Sr}/^{86}\text{Sr}$ values measured using TIMS and solution-mode MC-ICPMS (0.720 to 0.723, Sup. Table S5). LA-MC-ICPMS (0.718 to 0.725) agrees well with all solution mode data (TIMS and MC-ICPMS), and both sets of data document a trend of increasing values over the last millennium. $^{87}\text{Sr}/^{86}\text{Sr}$ values measured using exploratory LA-MC-ICPMS shows the potential for being resolved at higher resolution and, as expected, document greater variability. Extreme anomalies in LA-MC-ICPMS measurements were assessed as potential artifacts of the analytical method, and removed. This assessment included evaluation of spikes in ^{88}Sr signal intensity or ^{86}Kr and ^{87}Rb and identification of textural anomalies over which the laser might have traversed to a decadal scale. Reproducibility of our laser ablation methods was assessed using replicate analyses of overlapping transects along the growth axis, parallel transects collected during different analytical sections, and with high-resolution sampling and solution mode analysis (Fig. 6 and; Sup. Figs. S6 and S7).

Speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values increased over the last millennium (Fig. 6), gradually increasing from values closer to those of cave bedrock to values approaching those of soils overlying the cave (slope=0.0027, $p<0.001$ based on Mann-Kendall). The initial $\delta^{234}\text{U}$ values of TM0 range from 844 to 1111‰ (Sup. Table S2) and decreases from the early to the late part of the last millennium. TM0 growth rate ranges from 0.1 to 3.5 mm/yr, with greater occurrence of faster growth rate intervals in the late part of the last millennium (Fig 6). Evaluation of the covariation of multiple speleothem proxies can yield additional insight into the paleo-climate and environmental conditions (Wong and Breecker, 2015). However, the multiple, and potentially competing, controls on speleothem growth rate and $\delta^{13}\text{C}$ values make assessment of their covariation with $^{87}\text{Sr}/^{86}\text{Sr}$ values complicated (Supplemental Material).

5. Discussion

5.1 Assessing regional coherency in $\delta^{18}\text{O}$ records

The large number of highly resolved $\delta^{18}\text{O}$ records enables systematic assessment of the consistency in which the regional monsoon signal is preserved by $\delta^{18}\text{O}$ records from different

locations. This evaluation provides some constraints for delineating mechanisms driving last millennial trend in regional monsoon intensity, although a dynamical analysis is far beyond the scope of this study. $\delta^{18}\text{O}$ records from the Peruvian Andes, Andean foothills, and lowlands in western and northeastern Brazil all exhibit significant last millennium trends with similar slopes (Figs 3). The trend in the record from northeast Brazil is antithetical of the records from the west, which is consistent with the well-documented east-west anti-phasing of South American precipitation (Cruz et al., 2009; Novello et al., 2012). Increasing Southern Hemisphere summer insolation is a plausible driver of decreasing $\delta^{18}\text{O}$ values (increasing monsoon intensity) over the last millennium. An overall southward migration of the ITCZ over the last millennium might also be a mechanism contributing to the trend of increased monsoon intensity, although such a migration should drive lower $\delta^{18}\text{O}$ values over the Nordeste region and the opposite trend is documented (Novello et al., 2012). Further, meridional shifts in the ITCZ should have a dipole effect on Southern and Northern Hemisphere sites, as shifts in the ITCZ away from (toward) a respective hemisphere will cause enrichment (depletion) in $\delta^{18}\text{O}_{\text{precip}}$ in the Amazon Basin (Colose et al., 2016). A significant increasing trend in $\delta^{18}\text{O}$ values over the last millennium is present in only 2 of 5 $\delta^{18}\text{O}$ records from Central America (Fig. 3), and slopes of these trends are weak compared to the trends in $\delta^{18}\text{O}$ records from South America. These results tentatively suggests that the meridional migration of the ITCZ may not be the dominant process driving increasing SAMS intensity over the last millennium and/or factors in addition to shifts in the ITCZ may be governing variability in Central American $\delta^{18}\text{O}$ over the last millennium.

Records from our site (central Brazil) and Cristal Cave (southeastern Brazil) do not exhibit a significant trend over the last millennium in contrast with many records in the monsoon region. (Fig. 3). Such a contrast in the sensitivity of $\delta^{18}\text{O}$ records to variations in monsoon intensity might be expected with differences in elevation because Rayleigh fractionation is the dominant control on $\delta^{18}\text{O}_{\text{precip}}$ in the monsoon region. The consistency between the low elevation site (Pau D'Alho Cave in Mato Grosso, 600 m) in western Brazil and higher elevation Andean sites (Palestina and Huagapo Caves and Lake Pumacocha, 900 – 4,300 m), however, is not consistent with this explanation. Alternatively, it is possible that greater contributions of moisture sourced from the South Atlantic at sites more proximal to the Atlantic Ocean (Tamboril and Cristal Cave) dilute the monsoon signal inherited from upstream dynamics (Vuille et al., 2012). The absence of a last millennial trend in $\delta^{18}\text{O}$ records from Tamboril and Cristal Cave

suggests a process relevant to these sites operating to counter decreases in $\delta^{18}\text{O}_{\text{precip}}$ related to monsoon intensification over the last millennium. Such a process is not obvious, but we note that climate at these sites is influenced by the South Atlantic Convergence Zone (SACZ) - a convergence-driven convection in the region extending from the central Amazon Basin to the South Atlantic (Lenters and Cook, 1999; Vera et al., 2006). In the modern climate, the mean position and strength of the SACZ is dictated by variability in subtropical South Atlantic sea surface temperatures (e.g., Chaves and Nobre, 2004), pointing to a possible ocean-atmosphere mechanism. Notably, the SACZ is commonly considered a part of the SAMS, so any SACZ-related mechanisms that might account for a lack of trend in $\delta^{18}\text{O}$ values at Tamboril and Cristal Cave would also have to address the link between the SAMS and SACZ.

5.2 *Speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values as a moisture proxy*

Variations in TM0 $^{87}\text{Sr}/^{86}\text{Sr}$ values likely reflect changing amounts of water-rock interaction in response to variations in moisture conditions, similar to previous speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ time series that have been developed as proxies of paleoclimate conditions (e.g. Banner et al., 1996; Frumkin and Stein, 2004; Li et al., 2005; Oster et al. 2009; 2010; Zhou et al., 2009; Vaks et al., 2013). In regions where soil and bedrock have distinct $^{87}\text{Sr}/^{86}\text{Sr}$ signatures, dripwater and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values can reflect varying durations of contact with the host bedrock associated with varying water residence on time scales that range from sub-daily (Bullen et al., 1996) to sub-annual (Musgrove and Banner, 2004; Wong and Banner, 2010; Wong et al., 2011) to millennia and greater (e.g., Banner et al., 1996; Oster et al., 2009; Vaks et al., 2013). The amount of Sr in rainwater is negligible relative to the amount of Sr acquired from the soil (e.g., Bullen et al., 1997; Musgrove et al., 2004; Vaks et al., 2013), meaning that infiltrating water acquires an initial $^{87}\text{Sr}/^{86}\text{Sr}$ value from Sr leached from soils overlying the cave. Subsequently, the $^{87}\text{Sr}/^{86}\text{Sr}$ value of the water evolves toward the $^{87}\text{Sr}/^{86}\text{Sr}$ value of the bedrock as water infiltrates into and interacts (e.g., calcite and/or dolomite recrystallization) with the bedrock above the cave. The extent of the evolution of the water $^{87}\text{Sr}/^{86}\text{Sr}$ value from the signature of the soil to that of the bedrock reflects the amount of water-rock interaction that occurs, which is related to the duration of water transit (i.e., water residence time).

Water residence time is a function of flow path and moisture conditions. Flow paths can range between diffuse and conduit endmembers, where diffuse flow paths represent water

flowing through the matrix (i.e., pore space) of the bedrock and conduit flow paths represent water flowing through solution-widened fractures and conduits. Diffuse flow is more tortuous and allows for greater water residence time and water rock interaction, whereas conduit flow is more direct and limits water residence time and water rock interaction. Conduit flow is dominant during relatively wet periods as infiltrating water takes the most direct route, and diffuse flow is more dominant during dry periods as water drains from the bedrock matrix (Fig. 7). Modern cave monitoring demonstrates that drip sites dominantly supplied by conduit flow paths exhibit temporal (sub-annual) variability in $^{87}\text{Sr}/^{86}\text{Sr}$ values that correspond to changes in wet and dry conditions (Musgrove and Banner, 2004; Wong et al., 2010). Drip sites that are dominantly supplied by diffuse flow paths have $^{87}\text{Sr}/^{86}\text{Sr}$ values that are both less variable and more similar to values of the cave host rock relative to the conduit-supplied sites (e.g. Wong et al., 2010). Theoretically derived, numerical models of water-rock interaction can account for observed variations in dripwater and groundwater isotopic and geochemical compositions, further supporting water-rock interaction as a dominant control on dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values (Musgrove and Banner, 2004; Oster et al. 2010; Wong et al., 2011; Wong et al., 2013). That is, ground waters with relatively shorter water residence times exhibit less water rock interaction and more soil-like (vs. bedrock-like) $^{87}\text{Sr}/^{86}\text{Sr}$ values, and vice versa (Musgrove and Banner, 2004; Oster et al., 2010; Wong et al., 2011). This work demonstrates the potential sensitivity of speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values as proxies of the response of infiltration rate to changes in moisture conditions, and 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), and tropical (Banner et al., 1996) climates. Following this previously established logic, speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values will be closer to the $^{87}\text{Sr}/^{86}\text{Sr}$ signature of the soil under relatively wet conditions and closer to the $^{87}\text{Sr}/^{86}\text{Sr}$ signature of the bedrock under relatively dry conditions.

Figure 7: Conceptual diagram of water flow. Rainwater infiltrating through cave systems will initially acquire an $^{87}\text{Sr}/^{86}\text{Sr}$ value from Sr leached from the soil overlying the cave. During dry periods (left), infiltrating water will flow through the cave in diffuse flow paths (matrix porosity). Dry periods lead to high interaction between infiltrating water and bedrock and an evolution of the $^{87}\text{Sr}/^{86}\text{Sr}$ value of the waters to that of the bedrock. During wetter periods (right) the capacity of diffuse flow paths are exceeded and water infiltrates through conduit flow paths, and thus retains the $^{87}\text{Sr}/^{86}\text{Sr}$ of the soil to a greater degree.

5.3 Accounting for increasing speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values over the last millennium

We suggest that TM0 speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values reflect a trend of decreasing water residence time associated with greater infiltration rates due to increased water availability over the last millennium. Our interpretation is supported by the consistency of our results with an existing framework that delineates the link between variations in water residence time and $^{87}\text{Sr}/^{86}\text{Sr}$ variations. Furthermore, this interpretation is consistent with variations in speleothem $\delta^{234}\text{U}$ values, the regional trend in increasing monsoon intensity (Section 5.1), and a synthesis of pollen studies that indicate a pronounced increase in moisture availability in the last millennium in southeast Brazil (Sup. Fig. S1; Flantua et al., 2016). Alternative explanations, however, for an increasing trend in speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values are considered, including evolution of the soil $^{87}\text{Sr}/^{86}\text{Sr}$ composition and alternative mechanisms for decreasing water-rock interaction.

$^{87}\text{Sr}/^{86}\text{Sr}$ signatures of the soil, bedrock, and dripwater at Tamboril are consistent with the existing conceptual model that infiltrating water reflects interaction with soil and bedrock end-members. $^{87}\text{Sr}/^{86}\text{Sr}$ values of soil overlying the cave are distinctly higher than those of the underlying bedrock, despite the likely post-depositional alteration of bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ compositions at this site. Speleothem and dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values lie between those of soil and bedrock. The range of dripwater $^{87}\text{Sr}/^{86}\text{Sr}$ values, which is significantly lower than soil $^{87}\text{Sr}/^{86}\text{Sr}$ values, nearly spans the range of those of the speleothem. Variability in $^{87}\text{Sr}/^{86}\text{Sr}$ values between sites has been observed in other caves, and attributed to differences in the extent to which drip sites are supplied by conduit vs. diffuse flow paths (Musgrove and Banner, 2004; Wong et al., 2010, 2011). The $^{87}\text{Sr}/^{86}\text{Sr}$ values of dripwater collected in Tamboril Cave (during the dry season) likely represents water draining from the matrix of the bedrock (i.e., diffuse flow), and variability between sites may reflect differences in flow path characteristics (e.g., tortuosity, porosity, permeability) and/or variations in the relative extent to which the flow path interacts with inter-bedded silt and clay layers. It is possible that temporal variations in $^{87}\text{Sr}/^{86}\text{Sr}$ could be influenced by more/less interaction with more radiogenic silt and clay layers. This would, however, require a mechanism to account for progressively more interaction with silt and clay layers vs. carbonates with time, such as a greater contribution of flow from low porosity/permeability silicate layers during drier conditions. This mechanism would suggest increasing $^{87}\text{Sr}/^{86}\text{Sr}$ under drier conditions due to the radiogenic nature of the silicate material. On-going study of modern $^{87}\text{Sr}/^{86}\text{Sr}$ variations in dripwater as a function of climate variations will provide means to further elucidate the controls on and sensitivity of $^{87}\text{Sr}/^{86}\text{Sr}$ variability at this site. The consistency, however, of the setting at Tamboril Cave with an existing model provides strong validation for using speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ as a paleo-moisture proxy.

Decreasing TM0 $\delta^{234}\text{U}$ values over time are consistent with our interpretation of increasingly wetter conditions over the last millennium at Tamboril Cave (Fig. 6). Groundwater $\delta^{234}\text{U}$ values greater than 0‰ (secular equilibrium) reflect an enrichment of ^{234}U over ^{238}U generally attributed to the ejection of ^{234}Th (which decays to ^{234}U) from the bedrock into pore water following the energetic α -decay of ^{238}U and/or the subsequent, preferential leaching of ^{234}U from lattice sites within the bedrock damaged by α -decay (Richards and Dorale, 2003; Oster et al., 2012). Speleothem $\delta^{234}\text{U}$ values are commonly interpreted as a reflection of past moisture conditions mediating water residence time. That is, relatively dry conditions allow infiltrating

water to accumulate more ^{234}U (higher $\delta^{234}\text{U}$ value) due to leaching or α -recoil, and relatively wet conditions limit the extent to which this would occur, resulting in water with lower $\delta^{234}\text{U}$ values (Bar-Matthews et al., 1999; Frumkin and Stein, 2004; Zhou et al., 2005; Polyak et al., 2012; Oster et al., 2012). The decrease in TM0 $\delta^{234}\text{U}$ values from the early part to late part of the record likely reflects a decrease in water residence time associated with increasing moisture availability. Though the direction of change is the same in both the U and Sr isotope systems, the $\delta^{234}\text{U}$ record shows a step-like increase between 1100 and 700 yrs. BP and then plateaus, while the $^{87}\text{Sr}/^{86}\text{Sr}$ record increases more steadily. This apparent difference between the proxies could be an artifact of the differences in sampling resolution, as $\delta^{234}\text{U}$ was measured at lower resolution than $^{87}\text{Sr}/^{86}\text{Sr}$ values; thus the nature of the transition is less clearly resolved. Alternatively, the stepped nature of TM0 $\delta^{234}\text{U}$ values, relative to the more progressive evolution of $^{87}\text{Sr}/^{86}\text{Sr}$ values, suggests varying sensitivity of these two proxies to changing moisture conditions.

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. Mechanisms for changing the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of the soil include i) weathering of soils that results in a change in the amount and/or $^{87}\text{Sr}/^{86}\text{Sr}$ composition of available Sr, ii) physical erosional thinning of the soil layer resulting in greater contribution of fresh bedrock to soil, and iii) deposition of material with different $^{87}\text{Sr}/^{86}\text{Sr}$ composition. Consistent with previous studies of soil in the area, our results suggest the soils overlying the cave originate from the weathering of the South American erosional surface. It is likely that development of new soil over the 1 ky time interval of this study is negligible due to slow rates of soil formation. Further, the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of leachable Sr has been shown to remain constant after ~1 Ma of weathering (Bullen et al., 1997), so evolution of the isotopic signature of the soil from chemical weathering is likely negligible. Physical erosion and thinning of the soil layer might lead a stronger presence of underlying bedrock in the soil horizon (e.g., Cooke et al., 2003). This might result in higher soil pH and, consequently, cation exchange capacity and evolution of the soil horizon toward a $^{87}\text{Sr}/^{86}\text{Sr}$ signature of the underlying bedrock, both of which would result in a decrease of speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values. The presence of an increasing trend in $^{87}\text{Sr}/^{86}\text{Sr}$ values, therefore, indicates that soil erosion is likely not a dominant control on speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values in this setting. Lastly, variations in dust flux, reflecting changes in

atmospheric circulation (Li et al., 2005; Zhou et al., 2009) or aridity (Frumkin and Stein, 2004), can potentially influence speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ variability in settings where proximal dust sources have distinct isotopic signatures relative to the cave site. It is unlikely, however, that aerosol dust deposition played an influential role in $^{87}\text{Sr}/^{86}\text{Sr}$ variability of TM0, despite studies documenting transport of African dust across the Atlantic to South America during the austral summer (e.g., Koren et al., 2006; Ben-Ami et al., 2010). Although no studies specifically investigate dust deposition in the region of Tamboril Cave, several studies provide perspective on the relative contribution of Saharan dust to regions further north in the Amazon Basin that receive the largest fluxes (Koren et al., 2006; Ben-Ami et al., 2010). Saharan dust does not appear to contribute substantial amounts of Sr to regional soils as i) African dust and Amazonian deposits have distinct Pb-Nd-Sr isotopic signatures (Abouchami et al., 2013; Pourmand et al., 2014), ii) the composition of Amazonian deposits is consistent with *in-situ* weathering and erosion of the Precambrian Amazon craton (Abouchami et al., 2013), and iii) vertical erosion rates dwarf aeolian accumulation rates at the core of the Brazilian monsoon region (Abouchami et al., 2013). Additionally, recent work suggests that the boreal summer African dust plume has been stable over the last 2 ka (Hayes et al., in press), which may also suggest stability in the winter dust plume as the strength of summer and winter dust plumes tends to co-vary on decadal and longer timescales (Williams et al., in press). Given the limited influence of Saharan dust on the isotopic composition of surface material in the region of greatest dust flux and the stability of the dust flux over the last 2 ka, it is unlikely that Saharan dust played a role in evolving TM0 $^{87}\text{Sr}/^{86}\text{Sr}$ values over the last millennium. The authors are unaware of evidence that other regions may be a significant source of dust to the region.

Could additional factors, other than increasing infiltration rate, account for decreasing water-rock interaction? The extent to which infiltrating water dissolves carbonates in the cave host rock is largely governed by the saturation state of the water with respect to the carbonate minerals present. Therefore, pCO_2 –vadose water pCO_2 and the air–water pCO_2 gradient along the flow path is an important control on the extent of water-rock interaction. Changes in the pCO_2 of void spaces in the soil, epikarst, and vadose can dictate the extent to which infiltrating water dissolves (and re-precipitates) carbonate minerals. That is, CO_2 will dissolve into infiltrating water when pCO_2 –air is greater than pCO_2 –water, and water with greater pCO_2 will dissolve more carbonate. The pCO_2 of subsurface voids can be influenced by i) advective and diffusive

mixing with surface air (i.e., ventilation), and ii) production of CO₂ by respiration. Changes in cave air pCO₂ have been documented in response to diurnal and seasonal variations in airflow (e.g., Banner et al., 2007; Baldini et al., 2010; Breecker et al., 2012; Covington et al., 2015) and in response to disturbance of vegetation (Baldini et al., 2005; Wong et al., 2010). It is possible that long-term and progressive changes in ventilation leading to the steady decrease of pCO₂ of infiltrating water might account for decreased water-rock interaction, but a well-constrained mechanism to sustain such a change in ventilation over this time scale is not obvious. A steady reduction in subsurface respiration is also possible. Large shifts in soil carbon isotope compositions (~10‰) are typically observed when forest-savanna ecosystem transitions occur owing to metabolic differences between C₃ and C₄ plants (Dümig et al. 2008; Silva et al. 2008; Silva & Anand 2011). The absence, however, of large fluctuations in speleothem δ¹³C values suggests a stable forest cover over the cave throughout the entire record. Progressively drier conditions could reduce respiration, pCO₂-water, and carbonate dissolution, accounting for higher ⁸⁷Sr/⁸⁶Sr values. Drier conditions, however, would result in greater water residence time, which is inconsistent with decreasing δ²³⁴U values that reflect reduced water residence time. Unlike ⁸⁷Sr/⁸⁶Sr values, variations in δ²³⁴U do not require dissolution of bedrock carbonate. Instead, δ²³⁴U variations are driven by accumulation ejected ²³⁴Th during α-decay and/or preferential leaching of ²³⁴U from damaged lattice sites within the bedrock, both of which are sensitive to water residence time. Furthermore, increasingly drier conditions over the last millennium are not consistent with increasing monsoon intensity and pollen records that suggest progressively wetter conditions over the last millennium.

In summary, there is a trend in ⁸⁷Sr/⁸⁶Sr values over the last millennium that we interpret to reflect increasing moisture availability. Our interpretation is consistent with the established interpretive framework for using ⁸⁷Sr/⁸⁶Sr values as a moisture proxy, the trend of increasing monsoon intensity documented in δ¹⁸O records from the region, and a compilation of pollen records that suggest wet last millennium conditions. The trend in ⁸⁷Sr/⁸⁶Sr values could alternatively reflect an increasing influence of interbedded silicate material on dripwater isotopic compositions or reduced carbonate dissolution associated with vadose zone pCO₂ dynamics. Such mechanisms, however, would comprise a novel interpretation of paleo-environmental conditions from ⁸⁷Sr/⁸⁶Sr variability, and require a progressive drying over the last millennium, which is inconsistent with other paleoclimate reconstructions from the monsoon region.

Coincidence of increasingly wetter local conditions and intensification of monsoon strength over the last millennium would suggest a tight coupling between regional monsoon intensity and local moisture conditions at this site. That is, local moisture conditions may be dominantly dictated by monsoon intensity over the last millennium. We note that although a strong link between monsoon intensity and local moisture conditions through time has commonly been assumed, these results provide means for evaluating its validity. Importantly, the TM0 $^{87}\text{Sr}/^{86}\text{Sr}$ record is decoupled from both the TM0 and Cristal Cave $\delta^{18}\text{O}$ records over the last millennium. This decoupling suggests the influence of a process, in addition to those related to the monsoon, on speleothem $\delta^{18}\text{O}$ values in central and southeast Brazil as discussed in Section 5.1.

6. Conclusions and implications

Our current understanding of how and why moisture conditions varied over central Brazil over the last millennium relies on $\delta^{18}\text{O}$ -based reconstructions of monsoon intensity, for which the coupling to local moisture conditions has yet to be rigorously assessed due to the lack of high-resolution (non- $\delta^{18}\text{O}$ -based) paleomoisture records. We develop speleothem $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ records from a cave in central Brazil spanning the last millennium to assess the i) coherency of the monsoon signal across the region as preserved by $\delta^{18}\text{O}$ and ii) response of local moisture conditions to centennial variations in monsoon intensity. TM0 $^{87}\text{Sr}/^{86}\text{Sr}$ values evolve from the isotopic signature of the bedrock towards that of the soil, which is interpreted to reflect decreasing water-rock interaction due to increasing local moisture availability in central Brazil. This interpretation is consistent with more coarsely resolved decreasing speleothem $\delta^{234}\text{U}$ values, which also suggest increasingly wetter conditions over the last millennium. Increasing moisture availability is consistent with a trend of increasing monsoon intensity (decreasing $\delta^{18}\text{O}$ values) preserved in many existing $\delta^{18}\text{O}$ records from the monsoon region. Intriguingly, a trend in $\delta^{18}\text{O}$ values is absent in records from our site (central Brazil) and Cristal Cave (southeast Brazil) tentatively suggesting other relevant controls on $\delta^{18}\text{O}$ values in this region over the last millennium.

The recent synthesis of pollen records from the last 2 millennia documents the potential complexity in the response of local moisture conditions to variability in the SAMS, and highlights the need for non- $\delta^{18}\text{O}$ -based paleomoisture records. Our results demonstrate the

potential for speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values to address this need. Replication of solution-mode analyses of $^{87}\text{Sr}/^{86}\text{Sr}$ using laser ablation techniques leaves open the possibility of creating higher resolution paleomoisture records, especially in settings where the contrast between the cave host rock and overlying soils is large, speleothems have sufficient Sr ($>\sim 100\text{--}200$ ppm), and speleothem $^{87}\text{Sr}/^{86}\text{Sr}$ values are acutely sensitive to changes in infiltration rates. A network of highly resolved paleomoisture records that can complement the existing network of $\delta^{18}\text{O}$ -based records of monsoon intensity would enable a powerful assessment of spatial variability in the sensitivity of local-scale conditions to changes in regional climate, and place further constraints on identifying the climate processes driving variability in hydroclimate across the monsoon region.

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