# Mid to late 20<sup>th</sup> century freshening of the western tropical South Atlantic triggered by southward migration of the Intertropical Convergence Zone

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#### 27 Abstract

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In the tropical Atlantic Ocean, the Intertropical Convergence Zone (ITCZ) is an 28 important climate feature controlled by the interhemispheric sea surface temperature 29 (SST) gradient, and greatly influences rainfall patterns over the adjacent continents. To 30 better understand ITCZ dynamics in the context of past and future climate change, long-31 term oceanic records are needed, but observational data are limited in temporal extent. 32 Shallow-water corals provide seasonally-resolved archives of climate variability over 33 the tropical ocean. Here we present seasonally-resolved records of stable oxygen ( $\delta^{18}$ O) 34 and carbon ( $\delta^{13}$ C) isotope values of a *Siderastrea stellata* coral from northeastern Brazil 35 (Maracajaú, ~5°S). We show that the long-term trends in the record of coral  $\delta^{18}$ O values 36 are not primarily driven by SST but by hydrological changes at the sea surface. 37 Combining the record of coral  $\delta^{18}$ O values with instrumental SST, we present the first 38 reconstruction of seawater  $\delta^{18}$ O changes ( $\delta^{18}$ O<sub>seawater</sub>) in the western tropical South 39 Atlantic back to the early 20<sup>th</sup> century, a parameter that is related to changes in sea 40 surface salinity. The reconstructed  $\delta^{18}O_{\text{seawater}}$  changes indicate a prominent freshening 41 between the mid-1940's and mid-1970's, which coincides with a weakening of the 42 Atlantic interhemispheric SST gradient during this time interval. Our results suggest 43 that the weakened Atlantic SST gradient resulted in a southward shift of the thermal 44 equator that was accompanied by a southward migration of the ITCZ, resulting in 45 freshening of the western tropical South Atlantic during the mid to late 20<sup>th</sup> century. 46

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#### 52 1. INTRODUCTION

The Intertropical Convergence Zone (ITCZ) is a well-defined zonally-oriented band of high precipitation, centered a few degrees to the north of the equator (Schneider et al. 2014). The ITCZ shows a marked seasonal meridional migration cycle, characterized by a northernmost position attained during boreal fall and a southernmost position during boreal spring (Waliser and Gautier 1993). The ITCZ strongly influences the distribution of rainfall over the tropical Americas, with substantial socio-economic impacts over northeastern Brazil (Nobre and Shukla 1996; Hastenrath 2012).

60 Observational and modeling studies of the ITCZ indicate that its position is controlled by the meridional sea-surface temperature (SST) gradient, that changes 61 62 seasonally with solar irradiance, as well as oceanic and atmospheric heat transport 63 (Schneider et al. 2014). However, observational data are extremely limited in temporal extent and many relevant climatic parameters (e.g., precipitation over the ocean) are 64 only available after the start of the satellite era. Ocean salinity records from the Atlantic 65 66 Ocean, with high enough resolution to resolve the ITCZ, are only available back to the 1970s (Reverdin et al. 2007), although some very sparse data, averaged over large areas 67 of the ocean, are now available for earlier time periods (Friedman et al. 2017). Land-68 based precipitation records can be longer, but few span the whole 20th century, 69 particularly over South America (e.g. Júnior and Lucena 2020). Thus, high temporal 70 71 resolution tropical marine paleoclimate records sensitive to ITCZ-related seawater salinity changes are needed to extend our understanding of ITCZ dynamics. 72

73 Shallow-water corals can be excellent tropical climate archives (e.g. Weber and 74 Woodhead 1970; Swart 1983; Swart and Grottoli 2003; Felis 2020). They have been used to reconstruct oceanographic and climatic changes in the Caribbean Sea (e.g., von 75 76 Reumont et al. 2008; Brocas et al. 2016; Fowell et al. 2016), Red Sea (e.g., ; Al-Rousan et al. 2003; Felis and Rimbu 2010; Murty et al. 2018), Pacific Ocean (e.g., Beck et al., 77 1992; Linsley et al. 2010; Carilli et al. 2014) and Indian Ocean (e.g., Gagan et al. 1996; 78 Lee et al. 2014). Those studies shed light on ocean-climate system phenomena like the 79 equatorial monsoon (Gagan et al. 1994; Charles et al. 1997; Klein et al. 1997), El Niño 80 81 Southern Oscillation (ENSO) (e.g. Fairbanks et al. 1997; Hereid et al. 2012; Cobb et al. 2013; Hetzinger et al. 2016) and the ITCZ (Saenger et al. 2008). 82

83 In contrast, only a few shallow-water coral records have been generated from corals sampled in the western tropical South Atlantic (Table 1) (Evangelista et al. 2007, 84 85 2018; Mayal et al. 2009; Pereira et al. 2016, 2017, 2018). These studies exclusively explored two sites (i.e., Rocas atoll and Abrolhos) from all available Brazilian reef 86 systems (Leão et al. 2016). The few existing coral records from the western tropical 87 South Atlantic provide valuable information, but do not extend long enough back in 88 time to facilitate a full understanding of the influence of climate modes and solar 89 90 forcing in this understudied region. Among the species with high potential for past climate reconstruction, the coral species *Siderastrea stellata* is one of the most 91 important Brazilian reef builders, with a spatial distribution ranging from the equator to 92 93 23°S (Lins-de-Barros and Pires 2007). Furthermore, this coral species may provide geochemical records up to 300 years or more in duration, substantially extending 94 instrumental climate records from the western tropical South Atlantic. 95

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Species	Location	Period	Proxy	Reference
Mussismilia braziliensis	Abrolhos reefs	1987 -2003	Sr/Ca, Ba/Ca, Mg/Ca	(Santedicola et al. 2008)
Mussismilia braziliensis	Abrolhos and Tinharé Reef	1998-2005	Growth rate	(Kikuchi et al. 2013)
Siderastrea stellata	Abrolhos	1883-2005	Growth rate	(Evangelista et al. 2015)
Porites astreoides	Rocas atoll	2002-2012	$\delta^{18}O,\delta^{13}C$	(Pereira et al. 2015)
Mussismilia leptophylla	Abrolhos	1939–1977	Growth rate	(Evangelista et al. 2007)
Siderastrea stellata, Porites astreoides and Montastrea cavernosa	Rocas atoll	-	δ <sup>53</sup> Cr, <sup>87</sup> Sr/ <sup>86</sup> Sr, δ <sup>13</sup> C	(Pereira et al. 2016)
Porites astreoides	Rocas atoll	2001-2013	$\delta^{18}$ O, $\delta^{13}$ C, Sr/Ca	(Pereira et al. 2017)
Siderastrea stellata	Rocas atoll	1948-2013	$\delta^{13}C$	(Pereira et al. 2018)
Siderastrea stellata	Rocas atoll	1970-2009	Sr/Ca, U/Ca	(Evangelista et al. 2018)
Mussismilia hispida	Rocas atoll	1943-1962	$\delta^{18}O,\delta^{13}C$	(Silva et al. 2019)
Siderastrea stellata	Maracajaú Reef	1927-2018	$\delta^{18}O,\delta^{13}C$	This study

## Table 1. Coral-based paleoclimate records from the western tropical South Atlantic, covered period and used paleoclimate proxy.

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Here we present records of stable oxygen ( $\delta^{18}$ O) and carbon ( $\delta^{13}$ C) isotope values 101 102 for a first Siderastrea Stellata coral sampled from the Maracajaú reef, situated off northeastern Brazil, covering the period from 1929 to 2018. These coral-based stable 103 isotope records are by far the longest (i.e., 90-years duration) and highest resolved (i.e., 104 105 ca. 8 data points per year) datasets from the western tropical South Atlantic. Previous work has reported a relatively low performance of the Sr/Ca-temperature proxy in 106 tracking SST at interannual and longer timescales off Brazil, when applied to a 107 108 Siderastrea stellata coral from Rocas Atoll (Evangelista et al. 2018). Consequently, we pair coral  $\delta^{18}$ O with available instrumental seawater temperatures, in order to provide 109 the first reconstruction of the oxygen-isotope composition of seawater ( $\delta^{18}O_{seawater}$ ) and 110 assess changes in sea surface salinity (SSS), and ITCZ position, in the western tropical 111

- South Atlantic throughout the 20<sup>th</sup> Century. We note that application of the Sr/Catemperature proxy to tropical North Atlantic corals of the same genus, *Siderastrea sideria*, has provided more promising results in terms of tracking long-term SST
  variability (Maupin 2008; DeLong 2014, 2016; Kuffner 2017; Weerabaddana 2021).
- 116 2. STUDY AREA

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Figure 1. Sea-surface temperature (A and B) and sea-surface salinity (C and D) of the
western tropical South Atlantic during austral winter (June-July-August) (A and C) and
summer (December-January-February) (B and D). Siderastrea stellata coral sampling
location at Maracajaú reef is represented by a white circle.

This study presents new records of stable carbon and oxygen isotope values for a *Siderastrea stellata* coral core collected from the shallow coastal reefs of Maracajaú, these spanning from approximately 5°21'12'' S to 5°25'30'' S and from 35° 14' 30'' W to 35°17'12'' W, off northeastern Brazil (Fig. 1). The regional climate is tropical, with warm humid conditions and a well-defined dry season from September to February, contrasting with a wet season from April to August, with peak precipitation during March–April, when the ITCZ is situated over northern northeastern Brazil (Chiessi et al. 2021). Wind speed peaks during the wet season, when SST reaches 26.5°C, and is weaker during the dry season, when seawater temperature reaches maxima of up to 29.0°C (Testa and Bosence 1999).

The Maracajaú reefs are part of an extensive reef complex (~30 km in length from 134 North to South), situated 5–7 km from the coastline and forming knolls and patch reefs 135 trending in a northwest-southeast direction, parallel to the coast (Santos et al., 2007). 136 137 The study location is situated within the largest coral patch within the Maracajaú reef complex, this being about 9 km in length and 3 km in width. Water depths in the 138 complex range from a maximum water depth ca. 5 m to partially exposed patches 139 140 during the lowest tides. Scleractinian corals comprise the reef structure, with S. stellata responsible for about 80% of reef construction, alongside calcareous algae (Laborel 141 1970). The Maracajaú reefs do harbor other scleractinian corals, such as Porites 142 143 astreoides, Favia gravida, Agaricia fragilis Agaricia agaricites, Porites branneri, 144 Meandrina braziliensis, Mussismilia hartii (Santos et al., 2007) and Mussismilia hispida 145 (rare, Roos et al. 2019), as well as the hydrocorals Millepora alcicornis, which form 146 crowns on the reef tops, and less abundant Millepora braziliensis (Santos et al., 2007).

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#### 3. MATERIAL AND METHODS

#### 148 3.1. Sea Surface Temperature

We used SST data from HadISST (Rayner et al. 2003) over the grid point 7° 30'
00'' S and 32° 30' 00'' W, available at the KNMI Climate Explorer
(https://climexp.knmi.nl). To evaluate possible differences between different SST data

products, we also assessed the ERSSTv5 reanalysis data product (Huang et al. 2017), and the high-resolution satellite Oiv2 SST data products (Reynolds et al. 2002), obtained from the same coordinates (see supplementary information). The high correlation between the SST data products indicates that there is no significant difference between them (Fig. S1) and we decide to use the HadISST data product for the further analyses developed in this work.

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#### 159 **3.2.** Coral sampling

A core of the coral species *Siderastrea stellata* (sample identification number 18SM-C2) was collected from the Maracajaú reef from a water depth ca. 1 m using a pneumatic drill, retrieving a 34 cm long core. Core 18SM-C2 then was cut into two halves, with one half cut into 5 mm thick slices, parallel to the growth axis of the whole *S. stellata* colony. After cutting, the coral slices were cleaned with deionized water, airdried and then X-rayed at 50 kV and 320 mA, with an exposure time of 3.2 s and a distance from equipment to the object of 108 cm.

A total of 870 carbonate powder samples were collected by continuous, progressive milling to 1 mm depth (using a Proxxon micro mill MF 70 coupled with a precision X-Y table) of the coral slab from the top towards the bottom of the colony, following the thecal wall, with samples taken in 0.4 mm intervals along the growth axis. This sampling resolution resulted in about 8 samples per year (using the mean growth rate as a reference).



175Figure 2. (A)  $^{230}$ Th ages versus sclerochronology ages (r² = 0.99, p < 0.0001). (B)</th>176Sclerochronology results from CoralXDS analysis. (C) Coral core 18SM-C2 X-ray image177with sampling track for stable carbon and oxygen isotope analyses (blue line), as well as178sampling locations for  $^{230}$ Th dating (white arrows).

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- 180 **3.3.** Geochemical analyses
- 181 *3.3.1.* <sup>230</sup>*Th dating*

Six 0.10–0.25 g subsamples were cut out from along the coral growth axis (Fig. 2) 182 for high precision <sup>230</sup>Th dating (Shen et al. 2008, 2012). These subsamples were gently 183 crushed, physically cleaned with ultrasonic methods, and dried for U-Th chemistry. 184 Chemistry was conducted in a class-10,000 metal-free clean room with class-100 185 benches at the High-Precision Mass Spectrometry and Environment Change Laboratory 186 (HISPEC), Geosciences Department, National Taiwan University (Shen et al., 2008). 187 U-Th isotopic compositions and concentrations were determined on a multi-collector 188 189 inductively-coupled plasma mass spectrometer (MC-ICP-MS) in the HISPEC (Shen et al., 2012). The half-lives of U–Th nuclides used for <sup>230</sup>Th age calculation are the given 190 half-lives reported in Cheng et al. (2013). Uncertainties in the U-Th isotopic data and 191 192 <sup>230</sup>Th dates are calculated at the  $2\sigma$  level (two standard deviations of the mean,  $2\sigma_m$ ), 193 unless otherwise noted.

#### 194 *3.3.2. Stable-isotope ratio records*

Values of  $\delta^{13}$ C and  $\delta^{18}$ O of 870 milled coral powder samples were determined at 195 the Paleoceanography and Paleoclimatology Laboratory, School of Arts, Sciences and 196 Humanities, University of São Paulo, using a Thermo Scientific<sup>™</sup> MAT253 isotope 197 ratio mass spectrometer coupled to a Thermo Scientific<sup>TM</sup> Kiel IV automated carbonate 198 preparation device. Stable isotope measurements were obtained by reaction of 35-100 199 mg of aragonite with 102% phosphoric acid at 70 °C and the results corrected to permil 200 201 units relative to Vienna Pee Dee Belemnite (VPDB) using a calcite-based correction (Kim et al., 2007). The SHP2L Solnhofen limestone was used as an internal working 202 203 standard, which has been calibrated against Vienna Pee Dee Belemnite (VPDB) using the NBS19 standard (Crivellari et al. 2021). Analytical precision was better than ±0.05 204 % for  $\delta^{13}$ C and ±0.07 % for  $\delta^{18}$ O (±1  $\sigma$ , *n* = 141). 205

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#### 207 **3.4.** Coral core chronology

An age model was developed by counting the density bands using the software CoralXDS (Helmle et al. 2002) and contrasted to radiometric <sup>230</sup>Th dating (Fig. 2). Then we compared these results with the number of  $\delta^{18}$ O cycles, assuming that the consecutive minima or maxima represent a single year and converted geochemical records from depth in the coral core to a timescale by pairing highest (and lowest)  $\delta^{18}$ O with minimum (and maximum) HadiSST data for the region using the software QanalySeries (Kotov and Pälike 2018).

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#### 3.5. Seawater $\delta^{18}$ O reconstruction

Values of  $\delta^{18}O_{seawater}$  were deconvoluted from paired coral  $\delta^{18}O$  and instrumental SST (HadISST), by adjusting the equation proposed by Cahyarini et al. (2008). To remove the SST signal from the coral  $\delta^{18}O$  record and retrieve the  $\delta^{18}O_{seawater}$  signal we subtracted the SST contribution, inferred by centered SST, from the centered coral  $\delta^{18}$ O signal, according to Equation 1 (for more details see Cahyarini et al. 2008 and Pfeiffer et al. 2019):

222 Eq (1) 
$$\delta^{18}O_{\text{seawater}} = \left(\delta^{18}O_{\text{coral}} - \overline{\delta^{18}O_{\text{coral}}}\right) - (\gamma 1) * (SST - \overline{\text{SST}})$$
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where  $\delta^{18}O_{coral}$  is the measured coral sample,  $\overline{\delta^{18}O_{coral}}$  is the mean value of measured  $\delta^{18}O_{coral}$  samples, SST is the monthly value obtained from HadISST and equivalent in time to the  $\delta^{18}O_{coral}$  measured on coral,  $\overline{SST}$  is the mean value of SST over the period studied.  $\Gamma_1$  is the regression slope of  $\delta^{18}O_{coral}$  versus SST retrieved from HadISST.

The correlation between  $\delta^{18}O_{coral}$  and SST (Fig. 3) yields a  $\delta^{18}O_{coral}\text{--}SST$ 228 relationship of -0.17% per 1°C (r = -0.55, p < 0.0001), a value slightly higher than the 229 slope of -0.138 % per 1°C reported by Maupin et al. (2008) for Siderastrea siderea in 230 231 the Atlantic Ocean, although those authors argue that their slope was likely flatter than it should have been because of their positive SSS (*i.e.* seawater  $\delta^{18}$ O contribution) and 232 SST relationship, which dampens the  $\delta^{18}O_{coral}$ -SST relationship. Although the 233 calibration slope for the Maracajaú S. stellata coral is very similar to the well-known 234 slopes for  $\delta^{18}O_{coral}$ -SST in the Indo-Pacific *Porites* corals (e.g., (Gagan et al., 1998; 235 Omata et al., 2006), the region of Maracajaú is marked by the co-variation of  $\delta^{18}$ O<sub>seawater</sub> 236 and SST, thus the slope obtained by the linear regression between SST and  $\delta^{18}$ O might 237 238 be biased (Cahyarini et al. 2008). In order to evaluate a possible bias, we tested the influence of different linear regression slopes on the reconstructed  $\delta^{18}O_{seawater}$ . Therefore 239 we compared the  $\delta^{18}O_{\text{seawater}}$  values obtained based on our linear regression (-0.17%) 240 241 per 1°C) to the values obtained based on the linear regressions from Maupin et al. (2008) (-0.138% per 1°C) and Juillet-Leclerc and Schmidt (2001) (-0.20% per 1°C) 242 243 (see supplementary material). We found no substantial difference on the long-term trend of the reconstructed  $\delta^{18}O_{seawater}$  when the three different slopes were applied. However, we prefer to use the linear regression slope value from the work of Juillet-Leclerc and Schmidt (2001) avoiding further issues concerning the influence of seawater isotopic composition on the  $\delta^{18}O_{coral}$  record and a possible bias generated by the covariation of SST and  $\delta^{18}O_{seawater}$ .



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Figure 3.  $\delta^{18}$ O–SST correlation for coral 18SM-C2 and HadISST (for grid point -7.5 (latitude) and -32.5 (longitude)). The linear regression of  $\delta^{18}$ O–SST is significant (r = -0.55 p < 0.0001) and the slope is -0.17‰ per 1°C (95% CI: -0.19, -0.15).

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254 3.5.1. Error propagation

The propagated error of reconstructed  $\delta^{18}O_{seawater}$  was determined using equation (2), modeled after Cahyarini et al., 2008, where  $\sigma_{\delta sw}$  is the error on reconstructed  $\delta^{18}O_{seawater}$ ,  $\sigma_{\delta c}$  is the error on measured  $\delta^{18}O_{coral}$ ,  $\gamma_1$  is the regression slope of  $\delta^{18}O_{coral}$ versus SST retrieved from HadISST and  $\sigma_{SST}$  is the error of the HadISST.

259 Eq (2) 
$$\sigma_{\delta sw}^2 = \sigma_{\delta c}^2 + (\gamma_1)^2 \sigma_{SST}^2$$

260 Combining the analytical error of  $\pm 0.07\%$  for coral  $\delta^{18}O$  and HadISST error 261 varying from 0.10 to 0.76 °C, and the slope value for the  $\delta^{18}O_{\text{coral}}$ -SST relationship ( $\gamma_1$ ) 262 of -0.17% per 1°C, resulted in an error varying from 0.07 to 0.147 % (Figure 6C), with 263 a mean error of  $\pm 0.085\%$  (1 $\sigma$ ) for  $\delta^{18}O_{\text{seawater}}$ .

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#### 4. RESULTS AND DISCUSSION

#### 266 **4.1.** Age model and *S. stellata* coral growth rates

Density-band counting revealed that coral core 18SM-C2 spans the interval from 1928 to 2018. U-Th isotopic compositions and <sup>230</sup>Th dates determined for this coral are listed in Table 2 and the geochronological approach agrees with density-band counting (Fig. 2).

Excluding the first (1928) and last (2018) years, which might not represent a complete year of coral growth, the Maracajaú reef *S. stellata* growth rate varied from 2.1 to 5.8 mm year<sup>-1</sup>, with a mean growth rate of  $3.8 \pm 0.7$  mm year<sup>-1</sup> (Fig. 2). The lowest growth rates were during 1987 (2.1 mm year<sup>-1</sup>), 1937 and 1962 (2.5 mm year<sup>-1</sup>), 1961 (2.5 mm year<sup>-1</sup>), 1938, 1941 and 1986 (2.7 mm year<sup>-1</sup>). The highest growth rates were during 2012 (5.2 mm year<sup>-1</sup>), 1993 (5.2 mm year<sup>-1</sup>), 1955 (5.4 mm year<sup>-1</sup>), 1959 (5.5 mm year<sup>-1</sup>) and 1944 (5.8 mm year<sup>-1</sup>).

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#### Table 2. U-Th isotopic compositions and <sup>230Th</sup> ages for subsamples of coral core 18SM-C2.

Analytical errors are  $2\sigma$  of the mean.

Sample ID	Weight g	<sup>238</sup> U 10 <sup>-6</sup> g/g <sup>a</sup>	<sup>232</sup> Th 10 <sup>-12</sup> g/g	$\delta^{234}$ U measured <sup>a</sup>	[ <sup>230</sup> Th/ <sup>238</sup> U] activity <sup>c</sup>	<sup>230</sup> Th/ <sup>232</sup> Th atomic (x 10 <sup>-6</sup> )	Age (yr ago) uncorrected	Age (yr ago) corrected <sup>c,d</sup>	Age (yr BP) relative to 1950 AD	$\delta^{234} U_{initial}$
18SD-1	0.2013	$2.3016 \pm 0.0020$	325.6 ± 2.5	146.0 ± 1.4	$0.0000421 \pm 0.0000045$	$4.90 \pm 0.53$	$4.00 \pm 0.43$	0.7 ± 1.7	-68.8 ± 1.7	146.02 ± 1.4
18SD-2	0.2122	2.3994 ± 0.0023	$225.6 \pm 2.2$	145.4 ± 1.5	0.0002094 ± 0.0000046	36.71 ± 0.89	19.93 ± 0.44	17.8 ± 1.2	$-51.8 \pm 1.2$	145.39 ± 1.5
18SD-3	0.2025	2.0174 ± 0.0017	286.7 ± 2.3	143.0 ± 1.3	0.0004106 ± 0.0000069	47.64 ± 0.89	39.18 ± 0.66	35.9 ± 1.8	$-33.8 \pm 1.8$	143.03 ± 1.3
18SD-4	0.2024	2.2394 ± 0.0019	311.8 ± 2.4	144.0 ± 1.3	0.0005929 ± 0.0000079	$70.2 \pm 1.1$	56.53 ± 0.76	53.3 ± 1.8	$-16.2 \pm 1.8$	144.03 ± 1.3
18SD-5	0.2440	2.3284 ± 0.0028	1113.6 ± 2.4	142.1 ± 1.5	0.000849 ± 0.000015	29.25 ± 0.53	81.0 ± 1.5	70.0 ± 5.7	$0.23 \pm 5.7$	142.17 ± 1.5

 ${}^{a}[{}^{238}U] = [{}^{235}U] \ge 137.77 \ (\pm 0.11\%) \ (\text{Hiess et al., } 2012); \\ \delta^{234}U = ([{}^{234}U/{}^{238}U]_{activity} - 1) \ge 1000.$ 

 ${}^{b}\delta^{234}$ U<sub>initial</sub> corrected was calculated based on  ${}^{230}$ Th age (*T*), i.e.,  $\delta^{234}$ U<sub>initial</sub> =  $\delta^{234}$ U<sub>measured</sub> *X* e<sup> $\lambda_{234*T}$ </sup>, and *T* is corrected age.

 ${}^{c}[{}^{230}\text{Th}/{}^{238}\text{U}]_{\text{activity}} = 1 - e^{-\lambda_{230}T} + (\delta^{234}\text{U}_{\text{measured}}/1000)[\lambda_{230}/(\lambda_{230} - \lambda_{234})](1 - e^{-(\lambda_{230} - \lambda_{234})T}), \text{ where } T \text{ is the age.}$ 

Decay constants are 9.1705 x 10<sup>-6</sup> yr<sup>-1</sup> for <sup>230</sup>Th, 2.8221 x 10<sup>-6</sup> yr<sup>-1</sup> for <sup>234</sup>U (Cheng et al., 2013), and 1.55125 x 10<sup>-10</sup> yr<sup>-1</sup> for <sup>238</sup>U (Jaffey et al., 1971).

<sup>d</sup>Age corrections, relative to chemistry date on July 15<sup>th</sup> and September 26<sup>th</sup>, 2019, were calculated using an estimated atomic <sup>230</sup>Th/<sup>232</sup>Th ratio of 4 (± 2) x 10<sup>-6</sup> (Shen et al., 2008).

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Before interpretations of the records of  $\delta^{18}$ O and  $\delta^{13}$ C values can be made, it is 282 important to assess possible growth-rate related kinetic effects (McConnaughey 1989; 283 Cohen and McConnaughey 2003), which could compromise coral geochemistry-based 284 reconstructions of SST, and ultimately SSS. For the coral S. stellata from Maracajaú 285 reef, there was no statistically significant relationship between annual growth rates and 286 annual mean  $\delta^{18}$ O and  $\delta^{13}$ C values (p>0.05; Fig. 4). Consequently, coral core 18SM-C2 287  $\delta^{18}O_{coral}$  variability seems to be independent of growth rate related kinetic effects 288 (McConnaughey 1989; Cohen and McConnaughey 2003). 289



Figure 4. Comparison between Maracajaú reef *S. stellata* coral colony 18SM-C2 annual growth rate and mean annual  $\delta^{18}$ O (p = 0.06) (A); and  $\delta^{13}$ C (p = 0.08) (B) values.

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294 **4.2.** *S. stellata*  $\delta^{13}$ C record

295 Data records of HadISST SST and Maracajaú reef *S. stellata*  $\delta^{13}$ C and  $\delta^{18}$ O 296 records and growth rate are shown in Figure 5.

297 S. stellata  $\delta^{13}C_{coral}$  values (Fig.5) vary from -2.64 to 0.44‰ and the  $\delta^{13}C_{coral}$ 298 record exhibits short-term (i.e., seasonal or intra-annual) variations, which are usually 299 interpreted as a result of seasonal changes in cloud cover and the availability of light. 300 Light availability influences coral zooxanthellae symbiont uptake of <sup>12</sup>C, seasonally

changing the carbon-isotope composition of the internal dissolved inorganic carbon 301 pool, from which the coral skeleton is precipitated (Fairbanks and Dodge 1979; Pätzold 302 1984; Grottoli and Wellington 1999). The  $\delta^{13}C_{coral}$  record also shows a general long-303 term trend to lower values, after the 1940s. This pattern is consistent with the Suess 304 Effect (Revelle and Suess 1957; Keeling 1979) which describes the release of <sup>13</sup>C-305 depleted  $CO_2$  into the atmosphere via the burning of fossil fuels, and the subsequent 306 dissolution of such CO<sub>2</sub> into the oceans. Similar  $\delta^{13}C_{coral}$  trends have been observed in 307 308 corals from other Atlantic Ocean sites (Swart et al. 2010) and the Maracajaú reef S. stellata decreasing trend of -0.019% per year is consistent with those values reported 309 by Pereira et al. (2018b) for other Brazilian Siderastrea corals sampled from 1948 and 310 311 2013 (Table 1).

312



Figure 5. A) Sea surface temperatures (SST) for the Maracajaú reef obtained from the HadISST product (Kennedy et al. 2019) retrieved from https://climexp.knmi.nl for the grid point -7.5 (latitude) and -32.5 (longitude). Maracajaú reef *S. stellata* coral core 18SM-C2  $\delta^{18}$ O and  $\delta^{13}$ C time series (B and C, respectively). (D) Coral core 18SM-C2 annual growth rate. Thick black lines in A-C are running (37-point window) averages.

319

#### 320 **4.3.** *S. stellata* $\delta^{18}$ O record

Gridded instrumental SST for the Maracajaú reef region varied from 25.47 to 29.30°C, with a mean of 27.26  $\pm$  0.84°C and maximum range of 3.83°C, for the period from 1928 to 2018 (Fig. 5). The SST time series exhibits an overall general increasing trend of 0.007°C per year, corresponding to an SST increase of 0.63°C through the 90year study interval.

S. stellata  $\delta^{18}O_{coral}$  varied from -4.00 to -2.43‰; the  $\delta^{18}O_{coral}$  record shows clear 326 lower magnitude seasonal (intra-annual) cycles contrasting with larger magnitude 327 interannual variability. The complete Maracajaú reef S. stellata  $\delta^{18}$ O record does not 328 correlate strongly with the independent HadISST SST record for the study region; only 329 30% of  $\delta^{18}$ O variance is explained by SST (r<sup>2</sup> = 0.30, p<0.001), such that 70% of the 330  $\delta^{18}$ O variance must be explained by other forcing variables. Since S. stellata growth rate 331 does not exhibit any strong correlation with  $\delta^{18}$ O (r<sup>2</sup> = 0.04, p = 0.06), the next most 332 plausible explanation is that  $\delta^{18}O_{coral}$  variability has been influenced by changing 333 surface seawater salinity (SSS). Nevertheless, it is important to recognize that, under 334 specific conditions (e.g. enclosed reef pools),  $\delta^{18}O_{coral}$  records may be better recorders 335 of more localized, reef-scale, SST conditions (Huang et al. 2017; Pfeiffer et al. 2019) 336 than available wider-scale SST datasets retrieved from combined satellite and in-situ 337 338 measurements, the latter also often comprising scarce data measurements that have been averaged across large spatial scales. 339

An important feature within the  $\delta^{18}O_{coral}$  record is an overall decreasing trend, to 340 lower  $\delta^{18}$ O values, from the mid-1940s and to the mid-1970s, that is decoupled from the 341 342 long-term variability evident in the HadISST SST record. Over this time interval,  $\delta^{18}$ O<sub>coral</sub> has an overall decreasing trend of -0.0056% per year, with a total decrease of 343 -0.50%. Conversion of  $\delta^{18}O_{coral}$  into SST (assuming the  $\delta^{18}O$ -SST relationship of 344 0.17%/°C), produces a trend with an annual SST increase of 0.033°C and a total rise of 345 2.96°C for this time interval, more than 4 times the HadISST SST increase for the 346 region. Even when assuming a 'traditional'  $\delta^{18}$ O-SST relationship of 0.22%/°C slope 347 (Juillet-Leclerc and Schmidt 2001), the observed -0.50% decrease in  $\delta^{18}O_{coral}$ 348 represents more than 2°C increase in SST, indicating that the identified long-term 349  $\delta^{18}$ O<sub>coral</sub> trend must be influenced by another factor, the most likely one being a change 350

in  $\delta^{18}O_{seawater}$ . Possible causes of which include the addition of freshwater into the marine system and/or decreased evaporation from the ocean surface.

As detailed above, the largest magnitude Maracajaú reef *S. stellata*  $\delta^{18}O_{coral}$ decreasing trend occurs from the mid-1940s to the mid-1970s (Fig. 4), with a subsequent lower magnitude general decrease between the mid-1970s and ca. 2010. The change in coral  $\delta^{18}O$  from 1945 to 1975 is -0.55‰, whereas the change in SST over the same period is +0.09°C. Thus, the SST trend can only explain a small fraction of the  $\delta^{18}O$  change. We suggest that the decreasing trend in coral  $\delta^{18}O$  from 1945 to 1975 was caused by a freshening in the upper western tropical South Atlantic.

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### 4.4. $\delta^{18}O_{seawater}$ reconstruction

Decoupling the HadISST SST signal from the Maracajaú reef S. stellata  $\delta^{18}O_{coral}$ 361 record (section 3.3.1) results in a  $\delta^{18}O_{\text{seawater}}$  reconstruction with a total range of 1.20% 362 across the study time interval (Fig. 6). One of the main features of this  $\delta^{18}O_{\text{seawater}}$ 363 reconstruction is an overall decreasing trend, to more negative  $\delta^{18}$ O values, from ca. 364 1947 to ca. 1975. Prior to 1947, average  $\delta^{18}O_{seawater}$  values were 0.17 ± 0.15% and 365 between 1975 and ca. 2012 average  $\delta^{18}O_{\text{seawater}}$  values were  $-0.16 \pm 0.16\%$ ; both time 366 intervals also exhibit some variability in reconstructed  $\delta^{18}O_{seawater}$ . After the 1980s, the 367  $\delta^{18}$ O<sub>seawater</sub> trend stabilized with substantial interannual variations, from ca. 2012, 368 reconstructed  $\delta^{18}O_{\text{seawater}}$  increases in magnitude, to more positive values, coincident 369 with the most intense drought in northeastern Brazil in recent decades (Brito et al. 370 2018). 371

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Figure 6. (A) Atlantic interhemispheric sea surface temperature (SST) difference from the 374 HadISST product (Kennedy et al. 2019) (red line) calculated from area-integrated SST of 375 376 the North Atlantic (Arctic Circle to equator) and the South Atlantic (Antarctic Circle to 377 equator) between 68°W and 20°E. The black line is a 37-point running average. (B) 378 Maracajaú reef seawater  $\delta^{18}$ O reconstruction (blue line) according to equation (1) with  $\gamma_1$ 379 = -0.20%/°C (Juillet-Leclerc and Schmidt 2001). Grey shadow is the error propagation according to equation 2 (separate errors are plotted for each data point), and the black 380 381 line is a 37-point running average. A freshening trend (vertical blue shading) is evident from ca. 1945 to ca. 1975 and coincides with a decrease in the interhemispheric SST 382 difference between North and South Atlantic (A). 383

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Further evidence supporting a robust regional climate signal in our  $\delta^{18}O_{\text{seawater}}$ 385 reconstruction can be found in the comparison of our record with a recent SSS data 386 compilation (Friedman et al. 2017). When comparing the two datasets for the time 387 interval 1975 to 2018, which is a period of abundant instrumental SSS data, the 388 Maracajaú reef S. stellata coral  $\delta^{18}O_{\text{seawater}}$  reconstruction is confirmed to be a very good 389 proxy for overall trends in regional SSS variability (Fig. 7). Statistical correlations can 390 be assessed for two grid boxes included in Friedman et al. (2017); for grid-box 3, 391 covering the area 5°N-3°S, 34°W-45°W r is 0.7 (p=0.006, N\*=13, years 1975-2016), 392 and for nearby grid-box 5, spanning 4°N-5°S, 20°W-35°W r=0.59 (p=0.03, N\*=13, 393 years 1975-2016). Before 1975, SSS observations are sparse, during some years 394

observations are absent and values have been interpolated over several years (Friedman 395 396 et al., 2017, supplemental material). Hence, we do not consider SSS correlations to the coral-based  $\delta^{18}O_{\text{seawater}}$  reconstruction prior to 1975 to be a valid exercise. Readers are 397 further cautioned that direct calibration between the spatially averaged open-ocean SSS 398 data and the single coastal observation from the coral  $\delta^{18}$ O data is not appropriate 399 400 because of the different amount of averaging inherent in each data source. However, the significant amount of shared variance between the datasets demonstrates that the 401 402  $\delta^{18}O_{\text{seawater}}$  reconstruction based on the Maracajaú reef S. stellata coral, which extends back to 1928, provides a substantial improvement to existing regional SSS observation. 403 In summary, the compelling similarity between the coral-based  $\delta^{18}O_{\text{seawater}}$ 404 reconstruction and the instrumental SSS data compilation for this region, during the 405 time interval 1975 to 2015 (Friedman et al., 2017), strongly suggests that the coral 406 407 proxy record captures large-scale hydrological signals in the surface ocean of the 408 tropical western South Atlantic.



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Figure 7. Instrumental sea surface salinity (SSS) data for the grid-box 5°N-3°S, 34°W-411 45°W (Friedman et al., 2017; crosses) and the Maracajaú reef S. stellata coral  $\delta^{18}O_{seawater}$ 412 reconstruction (circles) demonstrate the sensitivity of the coral  $\delta^{18}$ O proxy to capture 413

414regional salinity variations. The gridded SSS data are freely available from the French Sea415Surface Salinity Observation Service (www.legos.obs-mip.fr/observations/sss/). The416salinity data are available as annual March-February means that have been smoothed417with a [121] binomial filter; the δ <sup>18</sup>O<sub>seawater</sub> data have been treated the same to enable418comparison.

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#### 420

#### 4.5. Role of the ITCZ in tropical Brazilian Atlantic Ocean change

By examining the  $\delta^{18}O_{seawater}$  trend, we observed that most changes to the isotopic record occurred from 1945 to 1975, with a particularly steep decrease from the mid-1960s to the mid-1970s (Fig. 6C). The 1945–1975 drop represents a  $\delta^{18}O_{seawater}$  change of  $-0.33\%_{o}$  over a period of ca. 30 years. Using a  $\delta^{18}O_{seawater}$ -salinity relationship of  $-0.20 \pm 0.03 \%_{o}$  per psu, as reported by Watanabe et al (2001, 2002) for seawater collected in the Caribbean Sea, the observed SSS freshening represents a decrease in SSS of ca. 1.65 psu.

The ITCZ position is seasonally regulated by the thermal equator, promoting its meridional migration throughout the tropics (Schneider et al. 2014). Long-term changes in the interhemispheric SST gradient would affect the latitudinal displacement of the ITCZ, shifting its position further north or south according to the SST gradient (Mulitza et al. 2017; Chiessi et al. 2021), affecting SSS in the western tropical South Atlantic and, more specifically, in the Maracajaú reef. A southward (northward) migration of the thermal equator would trigger a decrease (increase) in SSS at the Maracajaú reef.

The period from the mid-1940s to the mid-1970s was indeed marked by a decrease in the SST gradient between the North and the South Atlantic (Fig. 6B). This change is expected to have shifted the thermal equator to the south, resulting in a southward migration of the ITCZ and thus in increased precipitation over northern northeastern Brazil. Such an ITCZ migration is entirely consistent with the reconstructed  $\delta^{18}O_{seawater}$  for the Maracajaú coral reef complex (Fig. 6C). We suggest

that the decrease in  $\delta^{18}O_{seawater}$  (and SSS) was produced by increased ITCZ-related 441 precipitation over the Maracajaú reef, western tropical South Atlantic. Furthermore, this 442 suggestion is consistent with a long-term instrumental precipitation record for Caicó 443 (6°27'35"S; 37°5'56"W), northeastern Brazil, an inland location about 230 km from 444 Maracajaú reef, which documents increasing precipitation over the period of 1957 to 445 1972 (Fig. 6A) (Júnior and Lucena, 2020). Although the exact timing of the SSS 446 freshening indicated by our Maracajaú reef S. stellata coral derived  $\delta^{18}O_{\text{seawater}}$ 447 448 reconstruction does not perfectly match the increase in continental precipitation for Caicó, the instrumental precipitation data clearly supports the notion that a substantial 449 450 input of freshwater into northern northeastern Brazil and the western tropical South Atlantic occurred at least from the late 1950s to the early 1970s as a result of changes in 451 the interhemispheric Atlantic Ocean temperature gradient. 452

453 Although our results indicated a freshening in the western South Atlantic Ocean, 454 instrumental records assessed by Curry et al. (2003) indicated a salinity adjustment at 455 the Atlantic Ocean, with the tropics becoming more saline between ca. 1950 and 1990. At the North Atlantic, the observation of Curry et al. (2003) was supported by 456 Rosenheim et al. (2005) which used sclerosponges records to reconstruct  $\delta^{18}O_{seawater}$  of 457 Salinity Maximum Water from the North Atlantic for the period of 1890-1990, where 458 they observed a consistent increase in  $\delta^{18}O_{seawater}$  from 1950 to 1990. Rosenheim et al 459 (2005) suggested that the change in salinity is related to recent intensification of the 460 North Atlantic Oscillation index, which is known to intensify the tradewinds in the 461 462 tropical and subtropical North Atlantic (Marshall et al. 2001). Stronger wind stress can consequently increase evaporation. It might be easy to conclude that these studies are in 463 contrast to the freshening trend in our data, but they are focused on the broader tropics 464 465 and subtropics, not specifically the ITCZ-related SSS minimum region as in our study.

An increasing salinity trend over a broad swath of the tropics is consistent with increased evaporation where E>P, but the water must go somewhere, and a concomitant decrease in salinity within the ITCZ-related SSS minimum would indicate that at least some of it is transported to the deep tropics.

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### 4.6. Connections to the Broader Climate System

471 The Maracajaú reef S. stellata coral derived  $\delta^{18}O_{\text{seawater}}$  reconstruction is clearly related to regional SSS (Figure 7), with western tropical South Atlantic SSS linked to 472 ITCZ-related precipitation (Tchilibou et al., 2015). The SSS changes reconstructed for 473 the western tropical South Atlantic could represent either a migration of the ITCZ or a 474 change in the strength of ITCZ-related precipitation, both potentially contributing to the 475 observed  $\delta^{18}O_{seawater}$  signal since 1928. The regional pattern of decreasing SSS in the 476 477 western tropical South Atlantic over the last 90 years, with a strong decrease in the 478 1960s-1970s and a strong recovery in the 2010s, evidenced by our coral record, is 479 consistent with the broader basin-wide pattern of climate variability over this period.

The strong 1960s-1970s SSS decrease shown by the Maracajaú reef S. stellata 480 record is consistent with a southward ITCZ migration, in response to a sharp decrease in 481 482 the Atlantic interhemispheric SST gradient (Fig. 6) (Thompson et al. 2010). The timing of the change coincides with the great salinity anomaly observed in the North Atlantic 483 Subpolar Gyre (Friedman et al., 2017), and a decrease in Sahel rainfall (Hodson et al., 484 485 2014), that is inverse to the SSS freshening trend off northern northeastern Brazil, all 486 these observations being consistent with a reduction in AMOC strength (Dima and Lohmann 2010, Zhang and Delworth 2005). Attempts to explore the causes of the 487 488 1960s-1970s interhemispheric temperature shift have ruled out volcanic, ENSO, and wintertime atmospheric advection forcings (Thompson et al., 2010). Modeling efforts 489 indicate the change is likely unforced variability and may be related to changes in 490

AMOC, although aerosol forcing cannot be completely discounted (Friedman 2020). 491 492 Indeed, AMOC variability is well known to impact ITCZ location across different time scales (Schneider et al. 2014; Mulitza et al. 2017; Liu et al. 2020), and if this cause is 493 the major driver of the long-term trend in the Maracajaú reef S. stellata  $\delta^{18}$ O<sub>seawater</sub> 494 reconstruction, then our new long-term record would be consistent with recent, though 495 controversial, claims that the AMOC has slowed down over the 20<sup>th</sup> Century 496 (Rahmstorf et al., 2015, Caesar et al., 2018, Thornalley et al., 2018, Caesar et al., 2021, 497 498 Kilbourne et al., *in press*).

The length of the new Maracajaú reef S. stellata record also puts the well-499 recognized 1960s-1970s shift into a broader context, by showing a long-term decrease 500 of  $\delta^{18}O_{\text{seawater}}$  from 1928 to 2010, albeit punctuated by substantial interannual- to 501 502 multidecadal-scale variations that culminate with an unprecedented trend to increased SSS from 2010–2018. This most recent trend and the large reconstructed  $\delta^{18}O_{seawater}$ 503 variations during the 1930s-1940s are not associated with a similar change in the 504 505 interhemispheric temperature gradient (Fig. 6), unlike the 1960s-1970s shift. This observation highlights other processes besides the mean location of the ITCZ that can 506 507 also influence regional SSS in the western tropical South Atlantic.

For instance, the intensity of the Hadley or the Walker Circulation could change the intensity of ITCZ-related rainfall. Servain et al. (2014) found evidence for intensification of the Hadley Circulation from 1960-2012. They found no significant trend in ITCZ location, as calculated by pseudo-windstress curl over those years, but instead documented warming temperatures centered under the ITCZ and intensification of the winds, consistent with an intensification of the Hadley cell. The Maracajaú reef *S. stellata* data are consistent with their study, showing no significant trend from

1960-2012 because the 1960s-1970s drop in SSS is balanced by the 2010s increase. 515 Changes in the Walker circulation are thus likely to impact the Maracajaú reef  $\delta^{18}$ O 516 517 record. This is not surprising given the strong connection between Atlantic ITCZrainfall, especially during March-April-May, and the Pacific Walker circulation 518 519 (Saravanan and Chang 2000, Sasaki et al., 2015). The interaction between the Atlantic and Pacific can go both ways (McGregor et al., 2014), thus highlighting the potential for 520 feedbacks between the tropical basins to be impacting the Maracajaú reef S. stellata 521 522 records.

523 5. CONCLUSIONS

The first Brazilian S. stellata coral  $\delta^{18}O_{seawater}$  reconstruction for the Maracajaú 524 reef complex as presented herewith clearly shows that these corals are promising 525 archives to understand key western tropical South Atlantic climate features, including 526 changes in the ITCZ position and the related SSS variability. The new records of  $\delta^{13}$ C 527 and  $\delta^{18}$ O values presented here are the longest reconstructions for the western tropical 528 South Atlantic. The Maracajaú coral  $\delta^{18}$ O values primarily records SST and the  $\delta^{18}$ O of 529 seawater, with no significant growth-related kinetic effects.  $\Delta^{18}O_{seawater}$  was 530 reconstructed by removing the SST contribution to the coral  $\delta^{18}$ O record using a gridded 531 instrumental SST product. The reconstructed  $\delta^{18}O_{seawater}$  record is marked by a 532 533 freshening trend from the 1940s to the 1970s, in agreement with a change in the interhemispheric temperature gradient during the same period, which also was 534 coincident with the mid-20<sup>th</sup> Century hiatus in global warming. 535

Since ITCZ location is influenced by the interhemispheric temperature gradient, a 536 537 decrease in the SST gradient between the North and the South Atlantic would have shifted the thermal equator to the south, resulting in southward migration of the ITCZ 538 and increasing precipitation over northeastern Brazil. Such an ITCZ migration could be 539

related to multidecadal- to centennial-scale variations in AMOC, although definitive 540 541 reconstructions of AMOC history are required to test further this relationship. Besides changes in the latitudinal position of the ITCZ, some of the reconstructed  $\delta^{18}O_{\text{seawater}}$ 542 variability featured by our Maracajaú reef complex record, could also represent changes 543 in Hadley and/or Walker cell intensity, which would influence ITCZ-related 544 precipitation and thus the western tropical South Atlantic SSS. A network of tropical 545 546 South Atlantic coral-based SSS records, paired with similar records in the northern 547 tropics, would facilitate distinguishing between intensity and latitudinal changes in the ITCZ, thus exploring in greater detail those processes that govern global heat 548 549 distribution in the ocean-atmosphere system over decades to centuries, timescales that are difficult to interrogate with the short-duration instrumental data sets that are 550 available. 551

In summary, the new Brazilian Maracajaú reef *S. stellata* geochemical records are an important step towards building a trans-hemispheric network and highlights the critical importance of tropical South Atlantic coral paleoclimate archives for improving our understanding of key global climate-system processes.

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