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Twentieth century warming of the tropical Atlantic captured by Sr-U paleothermometry

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Key Points:
- Sr-U is strongly correlated with average growth temperature for multiple coral genera over the temperature range 23.15–30.12°C
- Application of the Sr-U calibration captures absolute SST, timing of multiyear variability, and twentieth century warming trend
- Sr/Ca records from the same corals do not capture absolute SST, multiyear variability, or the twentieth century warming trend

Supporting Information:
- Supporting Information S1

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Abstract Coral skeletons are valuable archives of past ocean conditions. However, interpretation of coral paleotemperature records is confounded by uncertainties associated with single-element ratio thermometers, including Sr/Ca. A new approach, Sr-U, uses U/Ca to constrain the influence of Rayleigh fractionation on Sr/Ca. Here we build on the initial Pacific *Porites* Sr-U calibration to include multiple Atlantic and Pacific coral genera from multiple coral reef locations spanning a temperature range of 23.15–30.12°C. Accounting for the wintertime growth cessation of one Bermuda coral, we show that Sr-U is strongly correlated with the average water temperature at each location ($r^2 = 0.91$, $P < 0.001$, $n = 19$). We applied the multispecies spatial calibration between Sr-U and temperature to reconstruct a 96 year long temperature record at Mona Island, Puerto Rico, using a coral not included in the calibration. Average Sr-U derived temperature for the period 1900–1996 is within 0.12°C of the average instrumental temperature at this site and captures the twentieth century warming trend of 0.06°C per decade. Sr-U also captures the timing of multiyear variability but with higher amplitude than implied by the instrumental data. Mean Sr-U temperatures and patterns of multiyear variability were replicated in a second coral in the same grid box. Conversely, Sr/Ca records from the same two corals were inconsistent with each other and failed to capture absolute sea temperatures, timing of multiyear variability, or the twentieth century warming trend. Our results suggest that coral Sr-U paleothermometry is a promising new tool for reconstruction of past ocean temperatures.

1. Introduction

The instrumental record of sea surface temperature (SST) extends back to ~1856 but is considered less reliable with increasingly sparse coverage prior to the 1950s [*Deser et al.*, 2010]. Sixty five years of reliable data is too short to enable robust assessment of shifts in mean temperature associated with anthropogenic activity [*Latif et al.*, 2006; *Enfield and Cid-Serrano*, 2010; *Solomon et al.*, 2011; *Otto et al.*, 2013; *Trenberth and Fasullo*, 2013] or to characterize multiyear variability and trends [*Johns et al.*, 2011; *Kilbourne et al.*, 2014]. The annually banded skeletons of massive, long-lived corals are a potentially valuable archive of ocean temperature conditions that can be used to extend the instrumental record across space and back in time. Sr/Ca [*Smith et al.*, 1979; *Beck et al.*, 1992] is currently the most widely used coral proxy thermometer [e.g., *Carilli et al.*, 2014; *DeLong et al.*, 2014; *Wu et al.*, 2014; *Thompson et al.*, 2015; *Toth et al.*, 2015]. Multiple abiogenic precipitation experiments show that Sr partitioning into the growing aragonite crystal is temperature dependent (summarized in *DeCarlo et al.* [2015]) and in several studies, coral Sr/Ca exhibits a strong correlation with SST on seasonal, interannual, and sometimes longer timescales [e.g., *Mitsuguchi et al.*, 2008; *Nurhati et al.*, 2009, 2011; *DeLong et al.*, 2014]. However, in others, coral Sr/Ca-derived temperatures did not consistently capture mean SST [e.g., *Goodkin et al.*, 2005; *Saenger et al.*, 2008; *Alpert et al.*, 2016], SST trends [e.g., *Smith et al.*, 2006; *Scott et al.*, 2010; *Grove et al.*, 2013; *Storz et al.*, 2013; *Carilli et al.*, 2014; *Alpert et al.*, 2016], or multidecadal variability [e.g., *Nurhati et al.*, 2011] evident in the post-1950 instrumental record. Indeed, differences in Sr/Ca-SST relationships among conspecifics on the same reef [e.g., *Cohen et al.*, 2002;

Alpert et al., 2016], between "bumps and valleys" on the same colony [*de Villiers et al.*, 1994; *Alibert and McCulloch*, 1997], and between skeletal elements within the same calyx [*Brahmi et al.*, 2012] indicate that processes other than temperature influence the Sr/Ca ratio of coral skeletons.

Multiple lines of evidence point to Rayleigh fractionation, associated with the precipitation of coral aragonite in a semi-isolated calcifying space, as an important source of intercolony and within-colony Sr/Ca "vital effects" [*Cohen et al.*, 2006; *Gaetani and Cohen*, 2006; *Gagnon et al.*, 2007]. Corals elevate the pH and carbonate ion concentration ([CO_3^{2-}]) [*Al-Horani et al.*, 2003; *Venn et al.*, 2011; *Cai et al.*, 2016] of the seawater-like fluid sequestered in the extracellular calcifying region located between the calicoblastic epithelial cells and existing skeleton or substrate [*Gagnon et al.*, 2012; *Tambutté et al.*, 2012], elevating its saturation state with respect to aragonite (Ω_{ar}) [*Cohen et al.*, 2009]. The higher (lower) the Ω_{ar} of the calcifying fluid, the more (less) aragonite precipitates, changing the Sr/Ca ratio of the fluid and consequently the Sr/Ca ratio of the precipitate. Because the Sr/Ca exchange coefficient is greater than 1 [*Gaetani and Cohen,* 2006], more (less) aragonite precipitating will decrease (increase) the Sr/Ca of that aragonite. These changes can occur independently of temperature. For example, in laboratory culture, coral Sr/Ca increased with decreasing seawater pH at constant temperature, because the amount of aragonite the corals were able to precipitate from each batch of calcifying fluid decreased with seawater acidification [*Cohen et al.*, 2009; *Gagnon et al.*, 2013; *Cole et al.*, 2016]. For corals out on a reef, these processes are superimposed on, and may even override, the fundamental temperature dependence of Sr/Ca in aragonite.

A novel coral paleothermometer, Sr-U [*DeCarlo et al.*, 2016], builds on the concept of Rayleigh-based multielement thermometry first proposed in *Cohen and Gaetani* [2010] and *Gaetani et al.* [2011]. *DeCarlo et al.* [2015] showed that the U/Ca ratio of aragonite precipitated abiogenically from an infinite seawater reservoir depends on [CO_3^{2-}] because uranyl complexes compete with CO_3^{2-} for space in the aragonite lattice. In abiogenic experiments, Sr/Ca does not respond to [CO_3^{2-}] directly, and Sr/Ca and U/Ca do not correlate [*DeCarlo et al*., 2015]. Coral biomineralization, however, differs from abiogenic precipitation in that corals grow aragonite in an isolated or semiisolated space in which the reservoir is finite. Here both Sr/Ca and U/Ca are influenced by Rayleigh fractionation linked to calcifying fluid [CO_3^{2-}], with the additional direct effect of [CO_3^{2-}] on U/Ca [*DeCarlo et al.*, 2015]. Consequently, a correlation between Sr/Ca and U/Ca is consistently observed in coral skeletons [*Cardinal et al.*, 2001; *Hendy et al.*, 2002; *Quinn and Sampson,* 2002; *Fallon et al.*, 2003; *Sinclair et al.*, 2006; *Felis et al.*, 2009, 2012; *Jones et al.*, 2015]. Sr-U exploits this correlation to isolate the influence of temperature from that of Rayleigh fractionation on coral Sr/Ca [*DeCarlo et al.*, 2016].

DeCarlo et al. [2016] showed that Sr-U of *Porites* corals from the Pacific Ocean and the Red Sea is strongly correlated with SST over a temperature range from 25.7 to 30.1°C. Here we test the applicability of the *Porites* Sr-U calibration to four additional coral genera used in paleooceanographic reconstructions and over a wider temperature range. We then performed a critical test of any thermometer, applying it down-core to construct SST time series from two Atlantic corals over a time period for which instrumental data are available. Neither coral used in the down-core reconstruction was included in the calibration. We compared the Sr-U-derived SST mean, variability, and trends with the instrumental SST record and with Sr/Ca-derived SST generated from the same corals.

2. Material and Methods

2.1. Corals

2.1.1. Determination of Sr-U for the Sr-U SST Calibration

Sr/Ca and U/Ca data generated from seven corals representing four coral genera (*Pocillopora*, *Diploria*, *Orbicella*, *Diploria*) from five coral reef sites (Figure 1, Table 1, and supporting information Figures S1, S2, S3, and S4) were combined with previously published data from Pacific *Porites* spp., [*DeCarlo et al.*, 2016] to construct a multispecies Sr-U SST calibration. All colonies analyzed in this study were sampled live and computerized tomography (CT) scanned using the Siemens Volume Zoom Spiral Computerized Tomography (CT) scanner at the Woods Hole Oceanographic Institution [*Saenger et al.*, 2009; *Vásquez-Bedoya et al.*, 2012] (supporting information Figure S6). Cores and branches were slabbed along the axis of maximum growth using a water-cooled IsoMet 1000 Precision saw. Subsamples milled from faster-growing corals (*Pocillopora damicornis* PAN-PD-014, *Orbicella annularis* STX-OA-001 and PR-OA-003, and Curacao *Diploria labyrinthiformis* CUR-DL-882) were analyzed by solution inductively coupled plasma mass spectrometry (ICPMS) analysis following the

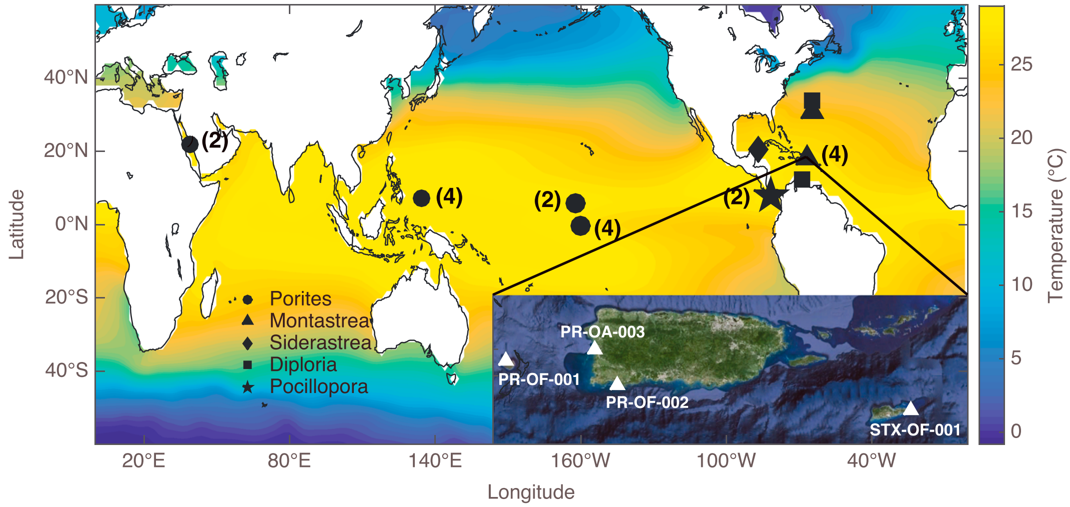

Figure 1. Map of 1981–2015 mean OISST [*Reynolds et al.*, 2002] showing locations of corals used in this study. Symbols are genera specific: massive *Porites* sp., (circles) *Orbicella* sp., (triangles) *Diploria labyrinthiformis* (squares), *Siderastrea siderea* (diamond), and *Pocillopora damicornis* (star). *Porites* data were previously published in *DeCarlo et al.* [2016].

procedure described in *Alpert et al.* [2016]. First, the coral slabs were ultrasonicated in deionized water to remove coral dust generated by drilling and sawing. Using annual bands visible in the CT scans to guide sampling resolution, 50–80 μg coral powder was milled at 0.6–1.0 mm intervals (depending on growth rate) using a Minicraft MB170 drill fitted with an 0.3 mm diameter diamond bit. Thickened skeletal elements known as thecal walls, which surround each individual coral polyp, were targeted for subsampling.

PAN-PD-014 was collected from the Canales Norte in the Gulf of Chiriqui, Pacific Panama, in November 2011. Two lateral branches were removed, cut down the center, and subsampled along the axis of maximum growth (supporting information Figure S7) using a Minicraft MB170 drill fitted with a 0.3 mm diameter diamond bit. No annual bands were visible in CT scans of the branches, so we assumed a linear extension rate of ~1.5 cm/yr [*Guzmán and Cortés*, 1993] to guide our sampling resolution of 1 mm. As the Sr/Ca and U/Ca values from the two branches of PAN-PD-014 were closely matched (supporting information Figure S1), we combined data from the two branches into a single data set.

Slower-growing (<5 mm/yr) Bermuda *Diploria labyrinthiformis* (BER-DL-003) and *Orbicella franksi* (BER-OFR-001) and Yucatan *Siderastrea sideraea* (Jardin C [*Gonneea*, 2014]) were analyzed using laser ablation ICPMS. Sections of BER-DL-003 and BER-OF-001 had been previously mounted in a 25.4 mm diameter aluminum ring [*Cohen et al.*, 2004; *Cohen and Thorrold*, 2007] and were reanalyzed here. A thin section of Jardin C was epoxy mounted on a glass slide [*Gonneea*, 2014]. All samples were polished down to 0.3 μm alumina suspension.

BER-DL-003 and BER-OF-001 were previously stained *in situ* with sodium alizarin sulphonate dye in June 2000, on 18 September 2000, and on 24 January 2001 prior to collection on 1 June 2001 (supporting information Figure S5) [*Cohen et al.*, 2004]. The skeletal stain lines indicated complete cessation of growth by BER-OF-001 in the wintertime. We used this information to estimate the average temperature this coral could have recorded in its skeleton, which is different from the average annual temperature at the site.

2.1.2. Down-Core Reconstruction

Two cores extracted from live *O. faveolata* colonies were analyzed for the time series (down-core) reconstruction. PR-OF-001 was cored live on Mona Island, Puerto Rico in 2012 at 7 m water depth. PR-OF-002 was cored live at Pinacles Reef in 1991 at 7 m water depth. Both cores were CT scanned, slabbed, sonicated and dried in a 60°C oven before subsampling along the axis of maximum growth, targeting the thecal walls. Sr/Ca and U/Ca data were generated using solution ICPMS. CT scan images of both corals revealed clear annual growth bands (supporting information Figure S6) from which the age models were constructed. The CT images reveal anomalously high-density stress bands in the Mona coral PR-OF-001 in 1923 and from 1998 to 2011, the latter in response to known 1998 and 2005 bleaching events. Therefore, Sr/Ca and U/Ca data from PR-OF-001 spanning the time period 1897 to 1997 were utilized, excluding 1921–1923. Data were generated from the Pinacles coral PR-OF-002 from 1950 to 1991.

Table 1. Coral Collection and Geochemical Information

Coral	Species	Location	Depth (m)	Extension Rate (cm/yr)	Mean Temperature Over Years Covered by Coral (°C) (Years in Record)	r^2 of Sr/Ca to U/Ca	Mean Sr/Ca (mmol/mol)	Sr-U
Jarvis West W490	*Porites* sp.	0.3696°S–160.0083°W	7	1.3	25.67[e] (2007–2012)	0.81	9.15	9.19
Jarvis West W497	*Porites* sp.	0.3689°S–160.0081°W	16	1.9	25.67[e] (2007–2012)	0.64	9.25	9.16
Jarvis East 16	*Porites* sp.	0.3739°S–159.9834°W	5	1.3	26.79[e] (2007–2012)	0.86	9.29	9.17
Jarvis East E500	*Porites* sp.	0.3715°S–159.9823°W	5	1.5	26.79[e] (2007–2012)	0.49	9.12	9.12
Palmyra 2	*Porites* sp.	5.8664°N–162.1095°W	13	1.3	27.93[e] (2006–2009)	0.55	9.04	8.90
Palmyra 3	*Porites* sp.	5.8664°N–162.1095°W	13	1.2	28.29[e] (1998–2010)	0.44	8.89	8.99
Red Sea 1	*Porites* sp.	22.0314°N–38.8778°E	1	0.9	28.41[e] (1998–2009)	0.23	9.02	9.00
Red Sea 44	*Porites* sp.	22.0314°N–38.8778°E	5	0.9	28.41[e] (2005–2009)	0.46	8.97	8.92
Palau 23 (Airai)	*Porites* sp.	7.3321°N–134.5602°E	4	1.6	29.18[e] (1997–1999)	0.36	8.89	8.94
Palau 221 + 229 (Uchelbeluu)[a]	*Porites* sp.	7.267°N–134.521°E	5	0.7	29.26[e] (2008–2009)	0.79	8.75	8.92
Palau 180 (Nikko Bay)	*Porites* sp.	7.3248°N–134.4684°E	6	0.9	30.04[e] (1997–1999)	0.29	8.94	8.83
Palau 168 + 169 (Nikko Bay)[a]	*Porites* sp.	7.3248°N–134.4684°E	3	0.6	30.12[e] (2008–2009)	0.30	8.69	8.78
This study: calibration								
BER-DL-003 (Bermuda)	*Diploria labyrinthformis*	32.32°N–64.71°W	13	0.5	23.15[b] (1995–1998)	0.60	9.10	9.35
BER-OFR-001 (Bermuda)	*Orbicella franksi*	32.32°N–64.71°W	13	0.2	25.12[b,c] (1994–2000)	0.62	9.22	9.27
STX-OA-001 (St Croix)	*Orbicella annularis*	17.74°N–64.58°W	0.5	1.1	27.73[b] (1995–2000)	0.60	9.00	9.06
Jardin C (Yucatan)	*Siderastrea siderea*	20.8321°N–86.8789°W	3	0.3	27.76[d] (1997–2009)	0.32	8.99	8.93
CUR-DL-882 (Curacao)	*Diploria labyrinthformis*	12.2021°N–69.0829°W	4	0.7	27.79[b] (2012–2013)	0.04	9.07	9.07
PR-OA-003 (Mayguez Bay, PR)	*Orbicella annularis*	18.2°N–67.2°W	<10	0.8	27.84[d] (2000–2005)	0.77	9.02	9.02
PAN-PD-014 (Canales Norte, Panama)	*Pocillopora damicornis*	9.73°N–81.58°W	~3	0.7 (L) 0.9 (R)	28.23[d] (2007–2010)	0.11	8.85	8.99
This study: time series								
PR-OF-001 (Mona Island, PR)	*Orbicella faveolata*	18.1153°N–67.9374°W	7		1900–1996			
PR-OF-002 (Pinacles, PR)	*Orbicella faveolata*	17.93°N–67.01°W	7		1954–1990			

[a] Due to small number of samples, Palau 168 and Palau 169 (Nikko Bay site), and Palau 221 and Palau 229 (Uchelbeluu site) located side by side at each reef site were grouped together to calculate Sr-U and Sr/Ca.
[b] OISST temperature [*Smith et al.*, 2008], logger verified.
[c] Adjusted to reflect mean temperature over period that coral grew.
[d] OISST temperature.
[e] Logger temperature.

2.2. Inductively Coupled Plasma Mass Spectrometry

2.2.1. Solution Analyses

Solution analyses followed the procedure described in *Alpert et al.* [2016]. Counts of ^{88}Sr, ^{238}U, and ^{48}Ca were measured on a single-collector Element ICPMS at the Woods Hole Oceanographic Institution. Coral powders were dissolved in 5% trace metal grade nitric acid. Sr/Ca values were determined by calibration to a curve of standards derived from coral skeleton (JCp-1) [*Okai et al.*, 2002; *Hathorne et al.*, 2013], fish otoliths (FEBS-1, NIES) [*Yoshinaga et al.*, 2000; *Sturgeon et al.*, 2005], and limestone (NBS-19) [*Fernandez et al.*, 2011], and U/Ca values were standardized to JCp-1. Mean Sr/Ca and U/Ca values of replicate analyses of three separate aliquots of JCp-1 powder are 8.870 ± 0.028 (1σ) mmol/mol and 1.23 ± 0.01 μmol/mol [*Alpert et al.*, 2016] which are within uncertainty of the means for Sr/Ca and U/Ca among 21 different labs (8.838 ± 0.042 mmol/mol and 1.192 ± 0.045 μmol/mol, respectively) and similar to the typical within-laboratory precision (0.026 mmol/mol for Sr/Ca and 0.012 μmol/mol for U/Ca) [*Hathorne et al.*, 2013]. Additionally, repeated measurements over 2 years of an in-house secondary coral standard indicate an external precision of ±0.035 mmol/mol (1σ, n = 173, 0.4% relative) for Sr/Ca and 0.02 μmol/mol for U/Ca (1σ, n = 173, 1.9% relative).

2.2.2. Laser Ablation Analyses

Sr/Ca and U/Ca of BER-OFR-001, BER-DL-003, and Jardin C were measured using the UP-193 nm New Wave Research laser ablation system mounted to the Element 2 Inductively Coupled Plama Mass Spectrometer at WHOI [*Cohen and Thorrold*, 2007]. For analysis of the Yucatan coral Jardin C, the laser was operated at 10 Hz with a 100 μm diameter spot size centered 200 μm apart. Sr/Ca values were determined using a calibration curve of dissolved standards including JCp-1, FEBS-1, and NIES, bracketing every eight samples. U/Ca values were standardized to JCp-1 [*Gonneea*, 2014]. BER-DL-003 was also analyzed with 100 μm diameter spots, but they were spaced 250 μm apart and preablated at 6 Hz for 4 s, followed by elemental measurements made at 9 Hz over a 65 s dwell time. Sr/Ca and U/Ca were determined by standardizing to the United States Geological Survey carbonate reference material MACS-3 [*Jochum et al.*, 2012], which bracketed every 22 samples. BER-OFR-001 was analyzed with the same laser settings as for BER-DL-003 but with spots 75 μm in diameter, centered 150 μm apart, and with a 100 μm diameter preablation. A die-pressed pellet of JCp-1 coral powder and MACS-3 were analyzed every eight samples. Each standard was measured three times, and the average value of the three measurements was used to construct a standard curve forced through the origin. To assess the precision of our laser ablation measurements, we withheld one of the JCp-1 analyses from each standard curve and instead treated it as a sample. Relative standard deviations of Sr/Ca and U/Ca were 1.8% and 4.1%, respectively, for these JCp-1 pellet analyses. The lower precision of our laser ablation measurements relative to our solution measurements is consistent with previous comparisons between these two techniques [*Sinclair et al.*, 1998; *Fallon et al.*, 1999], and our reproducibility of JCp-1 is within range of typical precisions reported for laser ablation measurements of coral Sr/Ca (1–4%) and U/Ca (3–5%) [*Sinclair et al.*, 1998; *Fallon et al.*, 1999; *Fallon et al.*, 2003; *Cohen and Thorrold,* 2007; *Montagna et al.*, 2007; *Felis et al.*, 2012]. Sr/Ca and U/Ca data are provided in supporting information Figures S1, S2, S3, and S4.

2.3. Age Models

For corals used in the calibration, and with the exception of the *Pocillopora* coral, annual density bands visible in 3-D CT scans were used to construct a first order chronology with an estimated error of ±1 year (supporting information Figure S6). The CT-based age models were fine tuned using the seasonal correlation between Sr/Ca and temperature [e.g., *Guilderson et al.*, 2004], assuming minimum Sr/Ca corresponds with maximum SST in each year. The maximum "adjustment" of any point was 8 months. The age model for BER-DL-003 was based on growth rates estimated in *Cohen et al.* [2004]. The slab prepared from BER-OFR-001 for laser ablation analyses could not be CT scanned, and therefore, age control was based solely upon the Sr/Ca data, assuming Sr/Ca variability relates to the timing of annual temperature cycles. For the long cores PR-OF-001 and PR-OF-002, seasonally resolved Sr/Ca data showed no correlation to SST on seasonal or longer timescales. Therefore, the age models for these corals are based solely on annual bands identified in the CT images (Figure S6).

2.4. Temperature Data for the Calibration

The initial *Porites* Sr-U calibration [*DeCarlo et al.*, 2016] was constrained with satellite SSTs and in situ logged temperatures from the Pacific and Red Sea reefs where the corals were sampled [*Cantin et al.*, 2010; *Barkley et al.*, 2015; *Alpert et al.*, 2016; *DeCarlo et al.*, 2016]. Temperature logger data were also available for the reef locations sampled for this study, but in some locations the data were discontinuous or did not cover the same time period as the coral records. For those sites where monthly averaged logger temperatures were consistent with contemporaneous monthly optimal interpolation SSTs (OISST v.3b [*Reynolds et al.*, 2002]), OISSTs were used.

SST at John Smith's Bay, Bermuda, was recorded daily between 1996 and 2001 by in situ Onset Stowaway XTI temperature data loggers at 13 m depth. As this record does not cover the entire period corresponding to the coral analyses and is discontinuous, we compared monthly averaged logger data to OISST values for a 1° × 1° grid box centered on Bermuda Island. On average the OISST values are 0.17°C warmer than those of the logger. The correlation coefficient between the two records is 0.92, a student *t* test indicates that their means are statistically indistinguishable, and an *F* test indicates that their variances are statistically indistinguishable. Therefore, we used OISST for the local grid box to estimate temperature at the John Smith's Bay site for the period over which the coral skeleton was analyzed (1994 to 2000).

SST near Willemstad, Curaçao, 14 km east of the coral collection site was recorded daily between March 1999 and September 2000 by an in situ Onset HOBO TidbiT v2 temperature logger deployed at 5 m depth. On average the OISST values are 0.02°C cooler than those of the logger. The correlation coefficient between monthly averaged logger temperature and OISST for this location is 0.99; a student *t* test and an *F* test indicate that their means and variances, respectively, are statistically indistinguishable. Therefore, we used OISST for the local grid box to estimate temperature at the Curaçao coral collection site for the period over which the skeleton was analyzed (2012–2013).

Bihourly water temperature was measured at Buck Island National Monument, St Croix, 8 km away from the coral collection site between January 1992 and January 2007 by a Ryan TempMentor 1.0 at a depth of 10 m [*Saenger et al.*, 2008]. Logger temperatures were statistically indistinguishable from OISST from the grid box centered on St Croix [*Saenger et al.*, 2008], and we used OISST to estimate temperature for the period over which the skeleton was analyzed (1995–2000).

OISST from Yucatan is in good agreement with average monthly SST recorded at a site within the Puerto Morelos lagoon [*Rodríguez-Martínez et al.*, 2010; *Gonneea*, 2014], and we used OISST to estimate temperature at the collection site.

Logger data are not available for the Canales Norte Panama site (PAN-PD-014), so we used monthly OISST temperature as a best estimate for water temperature.

Temperature data available for La Parguera, Puerto Rico (PR-OA-003), are based on bucket measurements made at the dock and are warmer than temperatures at the reef location where the coral colony grew, so we used OISST temperatures for this site.

2.5. Estimating the Temperature Range Over Which Bermuda Coral Growth Occurs

Alizarin Red S stain lines previously emplaced in the two Bermuda corals indicate that BER-OFR-001 stops growing during the coldest months of the year and is therefore incapable of recording the average annual growth temperature. We have no evidence of similar growth cessation in any of the other corals used in the calibration. With the exception of Bermuda, all the corals are from low-latitude sites (≤22°) that experience low-amplitude seasonal temperature (<6°C) and light cycles. Thus, we assume that these corals accrete skeleton throughout the year, albeit not necessarily uniformly, and we used the mean annual recorded temperature as the mean annual growth temperature. However, at 32°N, Bermuda experiences large seasonality in temperature and light, and independent evidence suggests that coral calcification rates on Bermuda vary by more than a factor of three between summer maxima and winter minima [*Cohen et al.*, 2004; *Venti et al.*, 2014].

Both Bermuda corals analyzed in this study were stained three times (2 June 2000, 24 September 2000, and 24 January 2001) over the course of one year and harvested one year after the first staining (1 June 2001), such that the surface marks a fourth time point [*Cohen et al.*, 2004]. However, the *O. franksi* coral (BER-OFR-001) captured only two of three stain lines; the mid-January stain line (24 January 2001) was missing completely, indicating lack of growth during the coolest period of the year. To estimate the average temperature over which BER-OFR-001 skeletal growth occurred, we measured the distances between stains along the thecal growth axis, using a Nikon SMZ1500 microscope (see supporting information Figure S5 and *Cohen et al.* [2004] (Figure 4). The stain line analysis indicated that 50% of the annual extension occurred between June and September. Since the stain lines separate the growth only into two time periods (June–September and October–May), we estimated a smooth monthly growth curve by minimizing month-to-month differences in extension while meeting the constraint identified from the stain lines. We then weighted the OISST monthly temperature according to the proportion of annual growth estimated for each month. This resulted in an average growth temperature of 25.12°C, which is 1.97°C warmer than the average annual temperature (23.15°C) because growth was strongly biased toward warmer months of the year.

2.6. Temperature Reanalysis Product for Comparison With the Derived SST Time Series

We compared the derived multidecade long SST time series from PR-OF-001 and PR-OF-002 to the National Oceanic and Atmospheric Administration's (NOAA's) Extended Reconstructed Sea Surface Temperature (ERSST v.3b) product [*Smith et al.*, 2008]. ERSST applies global spatial correlation scales identified in post-1981 satellite data to spatially and temporally discontinuous observations made prior to the satellite era, producing a globally complete gridded data set spanning 1854 to present. Although the corals were collected on reefs 100 km apart, both collection locations fall within the same 2° × 2° grid box, so we compare both coral Sr-U-derived SST records to the same ERSST time series. We used the same ERSST time series to assess the long Sr/Ca time series from the two corals.

Logger data are available for the Mona Island site from 2009 and 2014 and can be compared with the ERSST data over the same time period. Three in situ Onset Stowaway XTI data loggers recorded temperatures every 15 min within 500 m of the PR-OF-001 core location at 12 and 13 m depth. The logger data indicate that the coral site, located on the narrow Mona shelf, experiences periodic cold-water intrusions linked to internal wave activity [*Giese et al.*, 1990] that cool the water at the coral site by up to 2.5°C for several hours. This high-frequency variability nearly averages out on monthly timescales, but the logger data do indicate that the Mona coral site is, on average, 0.20°C cooler than the ERSST mean (supporting information Figure S9). Therefore, we adjusted the ERSST temperatures for both Mona and Pinacles down by −0.20°C for a closer estimate of actual SST.

2.7. Calculation and Temporal Resolution of Sr-U

"Sr-U" is a geochemical quantity determined by the relationship of Sr/Ca and U/Ca values in coral skeleton, as defined by *DeCarlo et al.* [2016]. We calculated Sr-U as follows:

$$\text{Sr-U} = f(\text{U/Ca});\ \text{U/Ca} = 1.1\ \ \mu\text{mol/mol} \tag{1}$$

where *f*(U/Ca) is the slope and intercept resulting from ordinary least squares (OLS) regression of Sr/Ca on U/Ca for a specified time period generated from an individual coral colony (Figure 2a and Table 1). U/Ca is used to account for the effects of Rayleigh fractionation on Sr/Ca, and evaluating the regression at the 1.1 μmol/mol benchmark aims to isolate the temperature component of Sr/Ca variability [*DeCarlo et al.*, 2016]. While both Sr/Ca and U/Ca are influenced by Rayleigh fractionation such that neither is strictly an independent variable, we treat U/Ca as the independent variable in OLS regression because we use U/Ca as a proxy for the Rayleigh fractionation vital effect on Sr/Ca.

Since a distribution of Sr/Ca and U/Ca values is needed to determine their relationship, Sr/Ca and U/Ca data spanning at least 1 year is usually required to generate a single Sr-U value, constraining the temporal resolution of SST reconstructions using Sr-U in its current form. The temporal resolution that can be achieved with Sr-U depends on both the strength of the correlation between Sr/Ca and U/Ca and the desired precision of the derived temperature estimate. In general, but not always, using shorter time windows to calculate Sr-U values leads to greater uncertainties in Sr-U because fewer data are available to constrain the regression between Sr/Ca and U/Ca. Uncertainty in Sr-U propagates to uncertainty in derived temperature, and thus, greater temporal resolution comes at the cost of lower temperature precision. In practice, this balance also depends on the distribution of U/Ca values for each coral. Because the present definition of Sr-U is defined at a benchmark U/Ca of 1.1 μmol/mol, there will be larger uncertainty in Sr-U for corals with U/Ca values that do not overlap with this benchmark.

In an effort to maximize resolution while minimizing uncertainty, we calculated an Sr-U value for each coral in the calibration using 3 or more years of Sr/Ca and U/Ca data and regressed Sr-U against SST averaged over the same time period, and in the case of the Bermuda coral BER-OFR-001, the estimated temperature over

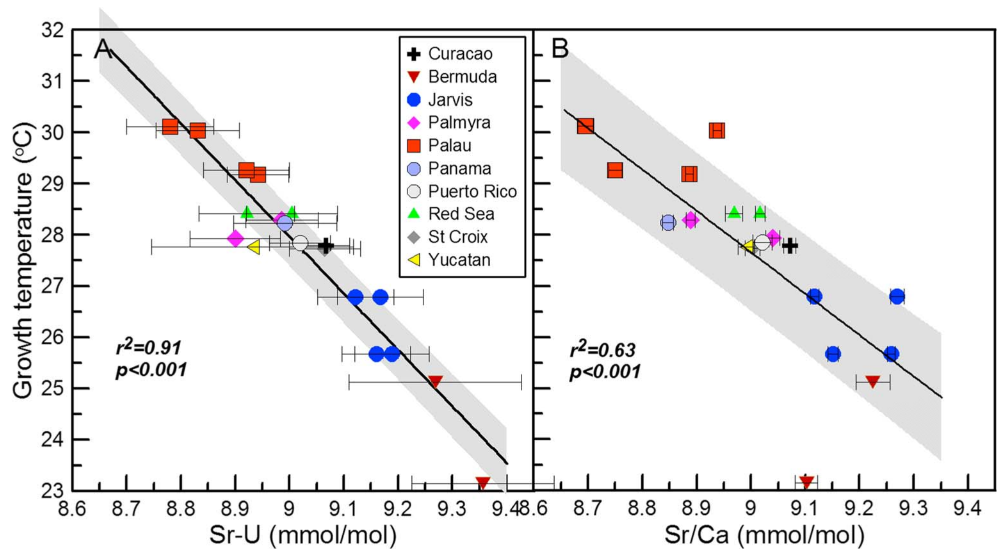

Figure 2. Regression of coral Sr-U and Sr/Ca onto average growth temperature from which equations (4) and (5), relating Sr-U and Sr/Ca to temperature, respectively, are derived. (a) Horizontal error bars represent the error on each Sr-U (1σ) value, i.e., the uncertainty of Sr/Ca at U/Ca = 1.1 µmol/mol for a given coral calculated using equation (2). Shading represents the uncertainty of the temperature prediction for a given Sr-U value, based on this regression (equation (3)). Average uncertainty is 0.62°C. (b) Horizontal error bars represent the standard error of the mean Sr/Ca (1σ) for each coral based on our external analytical precision of 0.035 mmol/mol. Shading represents the uncertainty of the temperature prediction for a given Sr/Ca value. Average uncertainty is 1.4°C.

which growth occurred. This exercise was repeated for Sr/Ca to construct a multispecies spatial Sr/Ca-SST calibration. The multispecies Sr-U and Sr/Ca calibrations were then applied to reconstruct multidecade long SST records from Mona Island (PR-OF-001) and Pinacles Reef (PR-OF-002), Puerto Rico. Neither coral was used in either the Sr-U or Sr/Ca calibrations.

For the time series reconstructions, we estimated Sr-U from 3 years of seasonally resolved Sr/Ca and U/Ca data in 1.5 year increments. The midpoint of each time period was assigned the year for that Sr-U value. For example, Sr-U calculated from Sr/Ca and U/Ca values spanning the period 1990–1993 was assigned the year 1991.5. The next Sr-U value was estimated for the 3 year window spanning 1988.5–1991.5 and assigned the year 1990. We continued to shift the beginning of the 3 year time window by 1.5 years to create a time series of Sr-U values with nominal resolution of 1.5 years (Figure 3). We also generated Sr/Ca time series, averaging Sr/Ca values for each core over the same time intervals used to calculate Sr-U values.

2.8. Coral Extension Rate

We quantified the annual upward linear extension rates of the Mona (PR-OF-001) and Pinacles (PR-0 F-002) corals and applied the *Saenger et al.* [2008] Sr/Ca-SST calibration equation that incorporates coral extension as a variable (Sr/Ca_{EXT}). Coral extension rates were quantified from the CT scan images as described in *Vásquez-Bedoya et al.* [2012] (supporting information Figure S10).

2.9. Statistics

Throughout this paper, we use the ordinary least squares method for regression and the analysis of variance function in MATLAB to compare trend slopes. We use a 95% confidence level ($\alpha = 0.05$) for statistical significance and uncertainty is reported as ±1σ.

3. Results

3.1. Sr-U Calibration

Data from 21 different corals are included in the calibration: 7 non-*Porites* corals analyzed for this study and 14 from the *Porites*-only study of *DeCarlo et al.* [2016]. Data from two pairs of Palau corals were grouped together to calculate Sr-U and mean Sr/Ca, resulting in effective n of 19 (Table 1). In Figure 2a, Sr-U calculated for all the corals are plotted against their estimated average annual growth temperature, which, in the

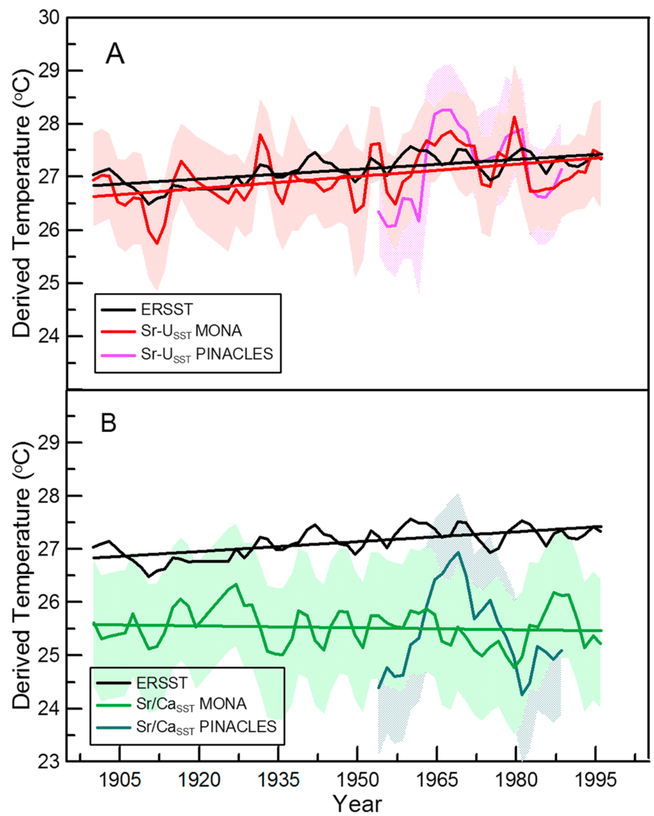

Figure 3. Temperatures derived using the spatial calibrations shown in Figure 2. (a) Sr-U derived temperatures from PR-OF-001 (red) and PR-OF-002 (purple) plotted with ERSST (black) [*Smith et al.*, 2008] adjusted for the 0.2°C difference between ERSST and the in situ logger data at this site. Neither coral was used in the calibration. Shading indicates the combined uncertainty of the spatial calibration and of Sr-U derived from Sr/Ca at U/Ca = 1.1 (1σ) (equation (4)). (b) Sr/Ca-derived temperatures from PR-OF-001 (light green) and PR-OF-002 (dark green) versus ERSST adjusted by −0.2°C (black). Shading indicates the combined uncertainty from the spatial Sr/Ca-SST calibration and the mean Sr/Ca value of each 3 year bin (1σ).

absence of evidence to the contrary, we assume is equivalent or close to the average annual site temperature except for BER-OFR-001, the Bermuda coral that ceased growth in midwinter. The 1σ uncertainty on each Sr-U value shown in Figure 2a was calculated from the standard error of prediction on the U/Ca versus Sr/Ca regression:

$$\sigma_{\text{Sr-U}} = \sqrt{\frac{1}{n-2}\times\sum_{i-1}^{n}\left(\text{Sr/Ca}_i - \text{Sr/Ca}_{\text{Pred}}\right)^2} \times \sqrt{1+\frac{1}{n}+\frac{\left(\text{U/Ca}-\overline{\text{U/Ca}}\right)^2}{\sum_{i=1}^{n}\left(\text{U/Ca}_i-\overline{\text{U/Ca}}\right)^2}} \quad (2)$$

where n is the number of points in the regression, $\text{Sr/Ca}_{\text{Pred}}$ is the Sr/Ca ratio predicted by the regression, and $\overline{\text{U/Ca}}$ is the mean U/Ca ratio. The standard error of prediction on the Sr-U SST regression was calculated as follows:

$$\sigma_{\text{SST}} = \sqrt{\frac{1}{n-2}\times\sum_{i-1}^{n}\left(\text{SST}_i - \text{SST}_{\text{Pred}}\right)^2} \times \sqrt{1+\frac{1}{n}+\frac{\left(\text{Sr-U}-\overline{\text{Sr-U}}\right)^2}{\sum_{i=1}^{n}\left(\text{Sr-U}_i-\overline{\text{Sr-U}}\right)^2}} \quad (3)$$

where SST_{Pred} is the SST predicted by the regression and $\overline{\text{Sr-U}}$ is the mean Sr-U ratio [e.g., *Chatterjee and Hadi*, 2012].

The correlation between Sr-U of multiple coral species and temperature is highly significant over the temperature range 23.15 to 30.12°C ($r^2 = 0.91$, $P < 0.001$, $n = 19$, Figure 2a and Table 1).

We derived an expression relating Sr-U to mean annual growth temperature, using ordinary least squares to regress temperature on Sr-U:

$$T_{\text{Sr-U}} = (-11.00 \pm 0.81)\,(\text{Sr-U}) + (126.98 \pm 7.29) \quad (4)$$

where Sr-U is the value resulting from equation (1) and $T_{\text{Sr-U}}$ is the resulting derived temperature. This expression was derived using corals from five Pacific and Atlantic coral genera distributed in both the Pacific and Atlantic Ocean is statistically indistinguishable from *DeCarlo et al.* [2016] derived using only Indo-Pacific *Porites* corals. The average standard error of prediction is 0.62°C (1σ, $n = 19$).

The average Sr/Ca ratio of each coral is shown in Figure 2b, also against average annual growth temperature. The expression relating mean Sr/Ca to temperature using ordinary least squares is:

$$T_{\text{Sr/Ca}} = (-8.084 \pm 1.42)\left(\overline{\text{Sr/Ca}}\right) + (100.41 \pm 12.74) \quad (5)$$

where $\overline{Sr/Ca}$ is mean Sr/Ca in mmol/mol and $T_{\text{Sr/Ca}}$ is the resulting derived temperature ($r^2 = 0.63$, $P < 0.001$, $n = 19$). The average standard error of prediction of this regression is 1.21°C (1σ, $n = 19$).

3.2. Time Series

Figure 3a shows SSTs derived from Mona (PR-OF-001) and Pinacles (PR-OF-0020 using the Sr-U calibration) compared against the ERSST record for the overlapping time period. ERSST values were calculated from 3 year bins staggered by 1.5 years, to match the temporal resolution of the Sr-U record. Neither coral was used to derive the calibration. Uncertainties on the predicted SST values were derived as follows. First, the uncertainty on Sr-U was calculated using equation (2). Next, uncertainty on the derived SST value was calculated by using a Monte Carlo algorithm to propagate $\sigma_{\text{Sr-U}}$ through the standard error of prediction of the Sr-U versus SST regression:

$$\sigma_{\text{SST}} = \sqrt{\frac{1}{n-2} \times \sum_{i-1}^{n} \left(\text{SST}_i - \text{SST}_{\text{Pred}}\right)^2} \times \sqrt{1 + \frac{1}{n} + \frac{\left(\text{Sr-U} \pm \sigma_{\text{Sr-U}} - \overline{\text{Sr-U}}\right)^2}{\sum_{i=1}^{n} \left(\text{Sr-U}_i - \overline{\text{Sr-U}}\right)^2}} \quad (6)$$

where SST_{Pred} is the SST predicted by the regression and $\overline{\text{Sr-U}}$ is the mean Sr-U ratio.

For corals not included in the calibration, there are two sources of uncertainty on SSTs derived from Sr-U: the uncertainty on the Sr-U SST calibration and the uncertainty on each Sr-U value generated from the coral in the time series. Consequently, each Sr-U derived temperature is associated with a unique uncertainty. On average $\sigma\text{SST}_{\text{Sr-U}}$ for the Mona coral (PR-OF-001) was 0.82°C and 0.95°C for the Pinacles coral.

Applying the multispecies spatial Sr-U SST calibration to the Mona coral that was not included in the calibration, captures the mean ERSST adjusted for the −0.2°C difference between the data product and the in situ logged SSTs at this site. The mean difference between Mona (PR-OF-001) Sr-U-derived SST and adjusted ERSST for a 2 × 2° grid square, over the full time period of the record, is 0.14°C. The mean difference between Pinacles (PR-OF-002) Sr-U-derived SST and −0.2°C adjusted ERSST for their overlapping time period is 0.12°C.

At Mona, Sr-U-derived SST indicates a twentieth century warming trend of 0.069°C/decade, which is statistically indistinguishable from the warming trend in the ERSST record over the same time period [*Smith et al.*, 2008]. The Sr-U-derived SST from the Mona coral also captures the timing of interannual variability evident in the instrumental record ($r^2 = 0.38$, $P < 0.01$, $n = 62$). However, the amplitude of interannual variability is greater in the coral-derived record than in the ERSST record. We compared the amplitude of variability in each time series by removing the mean and trend to describe the residual variability. The average amplitude (defined as the standard deviation of the detrended time series) for the 3 year window ERSST is 0.2°C and for the Sr-U SST is 0.4°C.

There are at least three possible, and not mutually exclusive, explanations for this discrepancy. First, the in situ record and the ERSST record agree on monthly timescales with a 0.2°C offset. However, the logger record is not long enough to assess in situ variability in temperature on multiyear timescales. Given the location of the coral site on the shelf edge, exposed to the effects of internal waves, it is possible that in situ multiyear

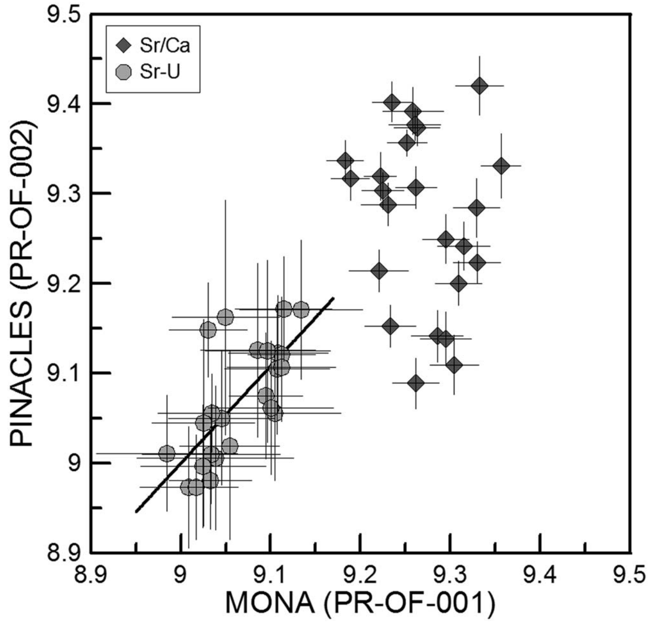

Figure 4. Sr-U (circles) and Sr/Ca (diamonds) from two corals, PR-OF-001 and PR-OF-002, for contemporaneous years. Sr-U values from each coral are highly correlated ($r^2 = 0.50$, $P < 0.01$, $n = 24$) whereas Sr/Ca values are not ($r^2 = 0.03$, $p = 0.43$, $n = 24$).

variability is larger than indicated by the satellite or ERSST records and that the coral is accurately recording its environment. It is instructive in this regard that the second coral PR-OF-002 also records higher multiyear variability than the ERSST and that the Sr-U SST mean and variability in the two corals from which time series were generated are significantly correlated ($r^2 = 0.50$, $P < 0.01$, $n = 24$, Figure 4), differing on average by 0.4°C (Figure 3a). Second, uncertainties in Sr-U values contribute variability to the temperature reconstructions and may account for some of the apparently exaggerated interannual amplitude. Since the standard deviation of detrended ERSST calculated in 3 year windows (0.19°C) is less than the average precision of SST derived from Mona Sr-U ($\sigma_{SST} = 0.82$°C), we would expect the SST reconstruction to be more variable than ERSST on interannual timescales.

Third, the greater amplitude of SST variability in the coral reconstruction appears to correlate with ERSST, meaning that the variability in Sr-U-derived SST—while expected based on propagating uncertainty of Sr-U ($\sigma_{Sr\text{-}U}$) to σ_{SST}—is not random in time. Thus, it is possible that the mechanisms of vital effects vary across temporal scales and the Sr-U calibration derived from data distributed in space overestimates the real multiyear variability. Finally, it is possible that skeletal growth biases such as that identified in BER-OFR-001 exist in other corals and have yet to be accounted for in the spatial calibration. Indeed, it is possible to reduce the amplitude of the reconstructed SST variability by using a temporal rather than a spatial calibration. Additional longer records are currently being used to test whether such a calibration derived from one coral applies universally.

The Sr/Ca variations in the records from Mona (PR-OF-001) and Pinacles (PR-OF-002) did not correlate with OISST or ERSST on annual or 3 year timescales, so it was not possible to construct an Sr/Ca-SST calibration specific to each coral. Thus, we took the same approach with Sr/Ca as we did with Sr-U, applying the spatial Sr/Ca-SST calibration (equation (5)) to the down-core Sr/Ca records from each colony (Figure 3b). We compared the Sr/Ca-derived SSTs with the ERSST reanalysis product. Unlike Sr-U, contemporaneous Sr/Ca values from the two colonies are not correlated ($r^2 = -0.03$, $p = 0.43$, $n = 24$, Figure 4). The mean Sr/Ca-derived temperature from Mona (PR-OF-001) is 1.6°C cooler than the −0.2°C adjusted ERSST and the mean Sr/Ca-derived temperature from Pinacles (PR-OF-002) is 1.9°C cooler. Interannual variability is significantly greater than the instrumental record and the Mona (PR-OF-001) Sr/Ca record fails to capture the twentieth century warming trend, instead showing a cooling trend, albeit not statistically significant.

3.2.1. Temporal Sr/Ca Calibration

Typically, temporal, not spatial calibrations are applied to reconstruct SST from Sr/Ca. As noted, the absence of a strong correlation between Sr/Ca and ERSST precluded deriving local or colony-specific Sr/Ca-SST calibrations for PR-OF-001 and PR-OF-002. *Kilbourne et al.* [2008, 2010] reported the same problem when working on an *Orbicella* core, also from Puerto Rico. Nevertheless, other Sr/Ca-SST calibrations exist for Caribbean *Orbicella* spp. including one derived from stained colonies using in situ logged temperatures [*Swart et al.*, 2002] and those of *Flannery and Poore* [2013] and *Saenger et al.* [2008]. Additionally, *Saenger et al.* [2008] used coral extension rate to correct for apparent growth-dependent Sr/Ca offsets among colonies. We applied these calibrations to the Sr/Ca time series derived from the Mona coral (PR-OF-001) (Figure 5). Regardless of calibration, none of the SST reconstructions derived using Sr/Ca only provides an accurate estimate of

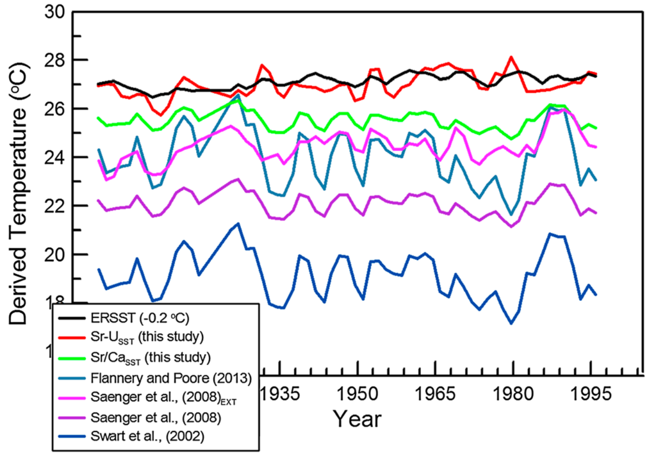

Figure 5. ERSST adjusted by 0.2°C (black) [*Smith et al.*, 2008] compared to Sr-U (red) and Sr/Ca (green)-based temperature estimates from the Mona coral (PR-OF-001) using the calibrations derived here (Figure 2), compared with temperatures derived using other published Sr/Ca-SST calibrations.

mean SST. This could, in part, be due to differences in absolute Sr/Ca values generated among different laboratories. However, neither do the Sr/Ca-SST calibrations, when applied to PR-OF-001, capture the timing or amplitude of SST variability or the long-term warming trend. Only one calibration, the one derived by *Saenger et al.* [2008], which attempts to correct for the vital effect using coral extension rate, yields a warming trend, but mean derived SSTs are too cold.

4. Discussion and Outlook

Extending the short instrumental record of SST across space and back in time is a high priority for paleoceanographic and climate studies. The skeletons of annually banded, long-lived corals are valuable archives of oceanographic variability, capable of recording information at subseasonal resolution, continuously for many centuries, and fossil corals can be accurately dated with U series. Single-element ratio thermometers, including Sr/Ca, have been applied to corals with mixed success, largely due to the distortion of the temperature dependence of element/Ca by "vital effects," although in older corals diagenetic alteration can also have significant effects on Sr/Ca and careful screening is important [e.g., *Cohen and Hart,* 2004; *Sayani et al*., 2011]. In response to the recognition that "vital effects" limit the interpretation of coral-based climate records even in young corals, new thermometers are actively being developed, based on fundamental understanding of element partitioning and the coral biomineralization process. These include Li/Mg [*Montagna et al*., 2014], Rayleigh-Based Multi-Element Thermometry [*Cohen and Gaetani*, 2010; *Gaetani et al*., 2011], and Sr-U [*DeCarlo et al*., 2016].

This study focused on validating the Sr-U thermometer in multiple coral genera commonly used in paleoclimate reconstructions and applying the spatially derived calibration in the temporal domain. We evaluated the ability of Sr-U to capture mean SST, multidecadal variability, and century-long trends in two corals from the tropical Atlantic. Our results show that Sr-U of multiple coral genera (*Porites*, *Siderastrea*, *Diploria*, *Orbicella*, and *Pocillopora*), calculated from seasonally resolved Sr/Ca and U/Ca data generated for each coral over several consecutive years, correlates with the mean SST of the same interval, over a temperature range spanning 23.15–30.12°C ($r^2 = 0.91$, $P < 0.001$, $n = 19$, Figure 2a and Table 1). In Bermuda corals that exhibit strongly seasonal growth or no growth for part of the year, Sr-U correlates with the estimated temperature over which growth occurs. It is likely that the difference we observed between mean annual temperature and mean growth temperature will have to be considered when conducting Sr-U thermometry (or any thermometry) using corals from high-latitude reef sites, or anywhere where coral skeletal growth shuts down for some period of the year, or during stressful events such as bleaching. One way to check whether growth occurs throughout the year would be by counting subannual growth increments such as dissepiments [e.g., *Winter and Sammarco*, 2010].

Application of the spatial calibration to Sr-U time series from two *O. faveolata* corals from Puerto Rico that were not included in the calibration yields average derived temperatures that are within 0.14°C of the ERSST data product when ERSST is adjusted for the in situ logger data at Mona (supporting information Figure S9). The longer derived temperature record replicates the twentieth century warming trend and the timing of multiyear variability inferred from ERSST. The amplitude of multiyear variability in the coral record is higher than that in the ERSST record.

Further refinement of Sr-U thermometry will focus on increasing the temporal resolution over which SST can be derived, refining the calibration and addressing the apparent overestimation on interannual variability. Sr-U paleothermometry does not currently resolve variability on seasonal or annual timescales but further development could address interpretation of temperatures on these scales. Whether the apparent exaggeration of multiyear variability in the Mona and Pinacles records is real, specific to this site, or a persistent characteristic of the thermometer as currently defined requires further testing as does the application of a temporal calibration. Results from our first application of Sr-U in the temporal domain shows it captures much of the key information needed for interpretation of climate variability and change: absolute SST, long-term trends, and timing of multiyear variability.

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