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Influence of seawater Sr content on coral Sr/Ca and Sr thermometry

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Abstract The Ca content of a *Porites* coral from Xisha, South China Sea is quite uniform along its 18-year growth axis. A comparison with previously published data shows that the Ca content of corals from different sites varies by only 0.4%. This is much smaller than the variation of Ca in seawater (2.2%), indicating that Ca variations in seawater do not significantly affect the Ca compositions of coral skeletons. The variation in skeletal Ca contents results in only $\pm 0.6^\circ\text{C}$ of uncertainty in SST calculations, which is much smaller than the large disparities observed for previously established coral Sr/Ca thermometers. In contrast, Sr in tropical seawater varies spatially by as much as 2.4%, corresponding to $\sim 4^\circ\text{C}$ offset for coral Sr/Ca calibrations. The effect of seawater Sr variations on coral Sr/Ca thermometers is evaluated and we demonstrate that the content of seawater Sr is the major factor responsible for disparities in these coral Sr/Ca thermometers. The disparities can be significantly reduced when seawater Sr contents are included in the Sr/Ca thermometers.

Keywords Coral · Sr/Ca thermometry · Sea-surface temperature · Coral Ca content · Seawater Sr concentration

Introduction

Massive corals incorporate a diversity of trace elements in their calcareous skeletons, and the contents of these elements vary with the environment in which the coral lives (Smith et al. 1979; Cross and Cross 1983; Shen and Boyle 1988; Shen and Dunbar 1995; Hart and Cohen 1996). Mg, Sr and U have relatively long residence times in seawater (Swart 1981; Swart and Hubbard 1982; Beck et al. 1992; Shen and Dunbar 1995), and their ratios to Ca in coral skeletons have been shown to be temperature dependent (Beck et al. 1992; Min et al. 1995; Mitsuguchi et al. 1996; Wei et al. 2000). As the Sr content in corals is relatively high, the Sr/Ca ratios have been studied in the greatest detail (Weber 1973; de Villiers et al. 1995; Gregor et al. 1997; Enmar et al. 2000; Allison et al. 2001; Cohen et al. 2001) and have been used to develop thermometers for reconstruction of sea-surface temperatures (SST) (Beck et al. 1992, 1997; Guilderson et al. 1994; Alibert and McCulloch 1997; McCulloch et al. 1996, 1999). However, significant disparities exist among coral Sr/Ca thermometers established for individual corals from different sites (de Villiers et al. 1994; Alibert and McCulloch 1997; Fairbanks et al. 1997; Gagan et al. 2000). Results typically differ by 2–3°C when different Sr/Ca thermometers are applied to a given coral (Stoll and Schrag 1998; Enmar et al. 2000; Crowsely 2000), and there is no universally accepted coral Sr/Ca–SST relation (Gagan et al. 2000). Previously published coral Sr/Ca thermometers can only be applied with caution to specific and limited areas (McCulloch and Esat 2000). Therefore, it is imperative to examine the causes for the disparity among coral Sr/Ca thermometers. The variation of Sr/Ca ratio in seawater was considered to be the major cause for the disparity and several workers (de Villiers et al. 1994; Shen et al. 1996) suggested using the distribution coefficient of the Sr/Ca ratio between corals and their ambient seawater [$D = (\text{Sr}/\text{Ca})^{\text{coral}}/(\text{Sr}/\text{Ca})^{\text{water}}$], instead of the coral Sr/Ca ratio alone, for SST calculations. The disparity, however, was not satisfactorily reduced by this

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method. After confirming that the Ca contents in corals are quite uniform, our study indicated that neither the Sr/Ca ratio nor the Ca content of seawater is the primary cause of the disparity. Rather, variability in the Sr content of seawater appeared to be the major factor.

Sampling and methodology

Sampling and analytical procedures

A living *Porites* coral growing at 20 m water depth near Xisha Island, South China Sea, was drilled along the dominant axis of growth in early June 1994 using a hydraulic drill with 6.0-cm inner diameter core barrel. The core was rinsed in ambient seawater before being transported to the laboratory where it was sliced into 7-mm-thick slabs parallel to its vertical axis and cleaned with deionized water. X-radiographs of the slabs were taken in order to determine the chronology of the specimen (Barnes and Lough 1993). Each couplet of low- and high-density bands represents the coral growth during 1 year. The actual chronology was determined by counting the density bands from the top (outmost) band, which corresponds to the time of collection.

A microsurgical machine, CHINA 2 (specially designed by the Institute of Earth Sciences, Academia Sinica in Taipei), was used to cut samples to 1 mm depth at 1 mm intervals perpendicular to the maximum extension of the coral slabs (Sun et al. 1999). Each subsample corresponds approximately to 1 month of the coral's growth. The saw was then rotated 90° to cut 1 mm grids, and then the 1 mm³ subsamples were collected using a surgical knife and a pair of surgical tweezers.

Each of the subsamples (~0.5 mg) was weighed with a microbalance, UMT-5 (sensitive to 0.1 µg), and was dissolved in 10 ml 0.5 mol/l HCl. Then a 1 ml aliquot of the solution was weighed and mixed with an appropriate amount of ⁴²Ca–⁴⁴Ca–⁸⁴Sr spike. The mixture was dried on a hot-plate and analyzed for Ca and Sr concentrations by VG 354 mass spectrometry at the Institute of Earth Sciences (Taipei), Academia Sinica, following the method of Shen (1996) and Shen et al. (1996).

Results and discussion

Ca contents in the coral and an Sr/Ca thermometer for the Xisha coral

The sampling technique commonly used in previous studies was to collect small amounts of coral powder by drilling (e.g., Beck et al. 1992; Alibert and McCulloch 1997), in order to obtain subsamples equivalent to 1 month or less of growth. The amounts thus collected were too small to be weighed accurately in most laboratories. As a result, these studies reported coral Sr/Ca ratios, instead of precise coral Sr and Ca concentrations.

In our study, the specially designed microsurgical machine allowed us to collect 1 mm³ subsamples that can be weighed accurately. The new subsampling technique enabled us to determine coral Ca and Sr contents, and Sr/Ca ratios, based on independent measurements of concentration for each element. Therefore, Ca and Sr variations in our coral sample were precisely evaluated.

A total of 60 subsamples spread along the growth record were analyzed for both Ca and Sr. The Ca data show that the Xisha coral has a very uniform Ca content of 9.52 mmol/g, with a very small variation [± 0.01 (1σ) mmol/g] (Fig. 1). This variation gives only $\pm 0.14^\circ\text{C}$ error in the SST calculation, which falls well within the range of the uncertainty for coral Sr/Ca-SST calibration ($\pm 0.5^\circ\text{C}$, e.g., Beck et al. 1992). Similar findings of uniform Ca content have been reported for specimens of *Porites* species from sites in the northern Red Sea (29.5°N, 34.9°E, Enmar et al. 2000), Tarawa Atoll (1°N, 172°E) in the Western Pacific (Allison et al. 2001) and Johnston Atoll (16°N, 169°W) in the North-Central Pacific (Cohen et al. 2001). Because Ca is a major constituent of coral skeletons, it should be relatively uniform in concentration, and this was assumed in some previous trace element studies (Weber 1973; Allison 1996). For example, Sholkovitz and Shen (1995) used a concentration of 10 mmol/g for Ca, a theoretical value for pure CaCO₃, to evaluate the distribution coefficients of rare earth elements between modern coral and seawater. Fallon et al. (2002) used a constant Ca content as an internal standard for determination of coral trace element contents by laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS). Since the Ca content in a coral is uniform, the coral Sr/Ca thermometer can be simplified to an Sr thermometer (Sun et al. 1999).

The temporal variation of Sr in the Xisha coral was reported by Sun et al. (2004). The data show annual cycles that correlate well with the instrument-measured SST. An Sr thermometer for this coral was established as $\text{SST} = -1.9658 \times \text{Sr} + 193.26$, with $r^2 = 0.96$ (Sun et al. 2004). Using the uniform Ca content (9.52 mmol/g) of the Xisha coral, the Sr thermometer referred above can be converted to the following Sr/Ca thermometer: $\text{SST} = -18.7144 \times \text{Sr/Ca} + 193.26$, where SST and

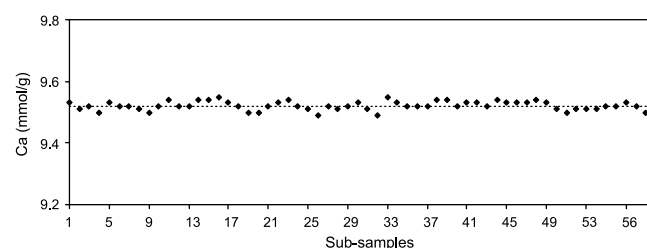


Fig. 1 Ca concentrations in the skeleton of the Xisha coral along sub-sampling sequence in an 18-year period from 1976 to 1994. The data fall within a range of 9.52 ± 0.01 (1σ) mmol/g, with a mean value of 9.52 mmol/g for the entire record (dashed line)

Sr/Ca are sea surface temperature in °C and coral Sr/Ca ratio in mmol/mol, respectively.

Disparities among previously established coral Sr/Ca thermometers

Using precise determination of Sr/Ca ratios by thermal ionization mass spectrometry (TIMS), several Sr/Ca thermometers were established for corals from the tropical Pacific, Indian and Atlantic Oceans (Beck et al. 1992; Shen et al. 1996; Alibert and McCulloch 1997; Cardinal et al. 2001). These thermometers show striking disparities on Sr/Ca versus SST diagrams (Fig. 2). This may be due to differences in the internal biological processes of the corals, or in external environmental conditions (de Villiers et al. 1995; McCulloch and Esat 2000; Cohen et al. 2001). The thermometers on the upper part of the diagram were derived for corals from Taiwan (Shen et al. 1996), Nusa Barung Island, Orpheus Island and Dampier Archipelago (Gagan et al. 1998). These areas are commonly associated with fresh water discharge (Shen et al. 1996; Gagan et al. 1998; McCulloch and Esat 2000). McCulloch and Esat (2000) also noted that coral Sr/Ca thermometers for inshore and mid-outer reef sites are substantially different. They reported that the calibrations of two corals from the mid-outer reef location are indistinguishable from each other, and gave a combined calibration of $Sr/Ca = 10.42 - 0.059 T$, significantly different (greater than 2.5°C) from the inshore coral calibration of $Sr/Ca = 10.73 - 0.0639 T$ (Gagan et al. 1998). The linear equation derived from the inshore coral data gives

systematically higher temperatures than the coral from the mid-outer reef site (McCulloch and Esat 2000).

These results imply that external environmental parameters, such as salinity, clarity and elemental contents of ambient seawater may be responsible for some of the disparities of coral Sr/Ca thermometers. Previous studies have shown higher variations in salinity at in-shore sites (Table 1) due to the influence of fresh water discharge (Shen et al. 1996; Gagan et al. 1998). Shen et al. (1996) found significant differences in Sr and Ca contents between the filtered and unfiltered seawater sampled near Taiwan (Table 2). The Sr and Ca concentrations in the unfiltered seawater were higher than those in the filtered seawater, suggesting that seawater from in-shore sites contains microparticles with higher contents of Sr and Ca. The question remains—which is the dominant element responsible for the above disparities, i.e., Ca or Sr, or Sr/Ca?

Influence of seawater Sr on coral Sr/Ca (or Sr) thermometers

A large range of measured concentrations of Sr and Ca, and a small range of ratios of Sr/Ca were reported for tropical seawater (Table 2). Their variations are 2.4, 2.2 and 0.3%, respectively. Seawater Sr and Ca have similar magnitudes and patterns of fluctuation (Fig. 3), which results in a small variation in the seawater Sr/Ca ratio of only 0.3% (Klein et al. 1996; Stoll and Schrag 1998). This small variation was considered to be an advantage for coral Sr/Ca thermometers (Beck et al. 1992; de Villiers et al. 1994; Shen et al. 1996), because it apparently

Fig. 2 Comparison of coral SST-Sr/Ca linear regression lines for data from different sites. Large disparities of $\leq 5^\circ\text{C}$ exist among established coral Sr/Ca thermometers

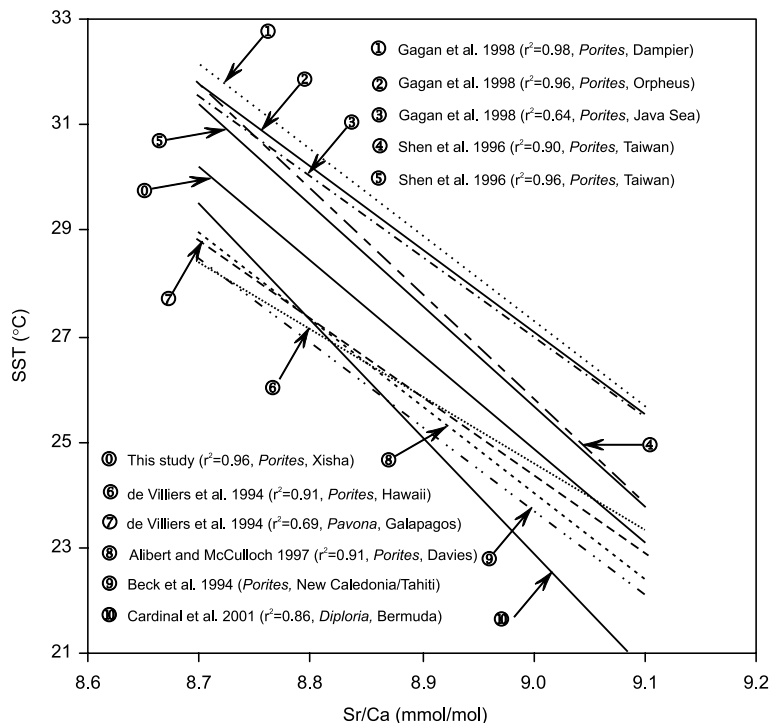


Table 1 Hydrographical conditions around collection sites of corals used to derive Sr/Ca thermometers

Site	Location	SSS (‰)	Environment	Sources
Xisha	16°50'N, 112°20'E	33.3 to 33.9	Monsoonal rainfall	This study
Taiwan	21°58'N, 120°42'E	Unavailable	Monsoonal rainfall, fresh water discharge	Shen et al. (1996)
Nusa Barung Island, Java Sea	8°31'S, 113°22'E	30 to 34	Monsoonal rainfall, coastal upwelling	Gagan et al. (1998)
Orpheus Island, Central Great Barrier Reef	18°45'S, 146°29'E	28 to 36	Monsoonal river runoff	Gagan et al. (1998)
Dampier Archipelago, eastern Indian Ocean	20°36'S, 116°45'E	35 to 39	Seasonal hypersalinity	Gagan et al. (1998)

SSS sea surface salinity

Table 2 Surface seawater Sr, Ca concentrations, and Sr/Ca ratios in tropical waters

Location	Sr $\mu\text{mol/kg}$	Ca mmol/kg	Sr/Ca mmol/mol	Source
Xisha	85.07	9.962	8.539	This study
05°S, 25°W	90.10	10.61	8.493	^a de Villiers (1999)
00°, 25°W	90.14	10.52	8.565	
05°N, 25°W	89.43	10.44	8.562	
10°N, 25°W	88.95	10.40	8.549	
15°N, 28°W	90.86	10.59	8.579	
20°N, 24°W	91.32	10.61	8.605	
25°N, 21°W	92.90	10.84	8.569	
30°N, 19°W	92.29	10.80	8.547	
10°S, 179°E	85.64	10.04	8.525	
05°S, 179°E	85.49	10.04	8.514	
00°, 179°E	85.13	9.987	8.524	
05°N, 179°E	84.70	9.911	8.546	
15°N, 179°E	86.67	10.17	8.524	
20°N, 179°E	87.30	10.27	8.501	
25°N, 179°E	87.76	10.34	8.489	
24°N, 154°W	89.52	10.52	8.506	
29°N, 150°W	87.94	10.34	8.501	
Galapagos (reef)	86.52	10.08	8.586	de Villiers et al. (1994)
Galapagos (station #1)	87.06	10.11	8.611	
Galapagos (station #2)	87.28	10.10	8.642	
Gulf of California (reef)	86.61	10.13	8.547	
Gulf of California (station)	86.44	10.07	8.581	
Taiwan (unfiltered)	88.63	10.33	8.578	^b Shen et al. (1996)
Taiwan (filtered, 5 μm)	86.29	10.03	8.603	
Taiwan (filtered, 0.45 μm)	82.52	9.626	8.572	

^aSr data are retrieved from normalized Sr concentrations by de Villiers (1999) at salinity of 35‰, and Ca data are calculated from these Sr data and Sr/Ca ratios

^bSr data are calculated from seawater Sr/Ca ratios and Ca concentrations near Taiwan (Shen et al. 1996)

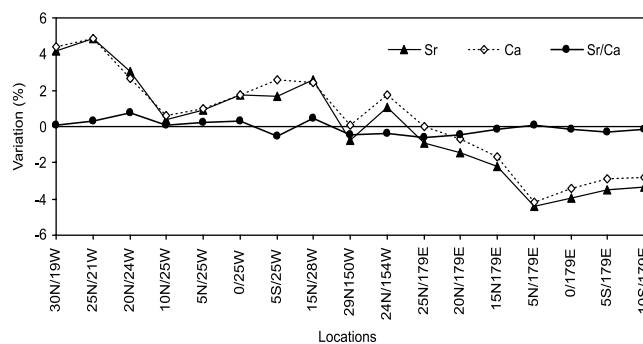


Fig. 3 Spatial variations of Sr, Ca contents and Sr/Ca ratios in tropical waters. The variations are calculated against the mean values (derived from data presented in Table 2)

allows application of a coral Sr/Ca thermometer to large regions and even to fossil corals. In order to eliminate the influence of seawater Sr/Ca variation, de Villiers et al. (1994) and Shen et al. (1996) suggested using the distri-

bution coefficient of Sr/Ca between the coral sample and ambient seawater [$D = (\text{Sr/Ca})^{\text{coral}} / (\text{Sr/Ca})^{\text{water}}$] to establish coral Sr/Ca thermometers. However, the disparities were not significantly reduced when this approach was taken. We believe that this is because corals take up Ca and Sr by different mechanisms, thus the small variation in the Sr/Ca ratio of seawater is not an advantage for coral Sr/Ca thermometers. Ca is the major element in coral skeletons, whereas Sr is a trace element admitted to the skeletons during calcification. As the two elements behave differently during coral growth, the disparities among coral Sr/Ca thermometers could be due to variations of seawater Sr or Ca contents, rather than the seawater Sr/Ca ratio.

The average variation of Ca in seawater is 2.2%, with a maximum of 9% between the two sites in the Atlantic (25°N, 21°W) and Pacific Oceans (5°N, 179°W) (Table 2 and Fig. 3). In contrast, Ca contents in corals fall within a much narrower range, suggesting that fluctuation of seawater Ca content does not significantly affect Ca

uptake by corals. Sun et al. (1999) reported a uniform value of 9.52 mmol/g Ca for a coral from Luta, southeastern Taiwan, and 9.53 mmol/g Ca for a 3,050-year-old coral from Kenting, southern Taiwan. Sinclair et al. (1998) reported a value of 9.56 mmol/g Ca for a coral from the Great Barrier Reef. The maximum Ca difference among these coral specimens is 0.04 mmol/g, i.e., 0.4% variation, which results in only 0.6°C offset for the SST calculation. Clearly, the variation in skeletal Sr/Ca ratios is mainly due to variations in coral Sr composition, as suggested by Enmar et al. (2000). Therefore, we conclude that neither the difference of Ca contents among individual corals nor seawater Ca variation can be the main cause of the disparities of coral Sr/Ca thermometers.

The influence of seawater Sr on coral Sr/Ca (or Sr) thermometry should not be underestimated. Shen and Dunbar (1995) reached a similar conclusion for coral U/Ca thermometry, i.e., that the coral U/Ca ratio was influenced primarily by the seawater U content and not by the seawater U/Ca ratio. The implications of seawater Sr for coral Sr/Ca (or Sr) thermometry can be assessed in terms of the distribution coefficient (K) of Sr between the coral and ambient seawater:

$$K = \frac{Sr^{coral}}{Sr^{water}} \quad (1)$$

where Sr^{coral} and Sr^{water} are coral Sr and seawater Sr contents, respectively.

Accordingly, the following relationship between the variations of coral Sr and seawater Sr is derived:

$$\frac{\Delta Sr^{coral}}{Sr^{coral}} = \frac{\Delta Sr^{water}}{Sr^{water}} \quad (2)$$

Based on the coral Sr/Ca thermometer, $SST = A \times (Sr/Ca)^{coral} + B$, and because Ca is uniform in the coral, the following equations can be derived for the offset of coral Sr/Ca-temperature (ΔSST):

$$\begin{aligned} \Delta SST &= \frac{A}{Ca^{coral}} \times \Delta Sr^{coral} \\ &= A \times \frac{Sr^{coral}}{Ca^{coral}} \times \frac{\Delta Sr^{coral}}{Sr^{coral}} \\ &= A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \frac{\Delta Sr^{coral}}{Sr^{coral}} \end{aligned} \quad (3)$$

Combining Eq. 2 and Eq. 3, the offset of coral Sr/Ca-SST arising from seawater Sr variation is expressed as:

$$\Delta SST = A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \frac{\Delta Sr^{water}}{Sr^{water}} \quad (4)$$

This shows that tropical seawater with 2.4% of variation in Sr concentration can result in $\sim 4^\circ C$ variation in the calculated Sr/Ca-SST. Therefore, the difference in ambient seawater Sr concentrations can account for the large observed disparities of coral Sr/Ca thermometers.

Normalization of coral Sr/Ca thermometers

Here, we suggest that the seawater Sr content in the coral be included in Sr/Ca (or Sr) thermometers. Firstly, the seawater Sr content in the study site needs to be measured, before an established Sr/Ca thermometer is used. Taking Xisha as an example, the ambient seawater Sr content of 85.07 mmol/kg is used to normalize a coral Sr/Ca thermometer established in a different site to get $SST_{normalized}$, thereby correcting the disparity caused by the difference in seawater Sr.

$$SST_{normalized} = SST_{established} - \Delta SST \quad (5)$$

The difference in seawater Sr (ΔSr^{water}) between the site used to establish the coral Sr/Ca thermometer and the Xisha site is:

$$\Delta Sr^{water} = Sr_{established}^{water} - Sr_{Xisha}^{water} \quad (6)$$

According to Eq. 4,

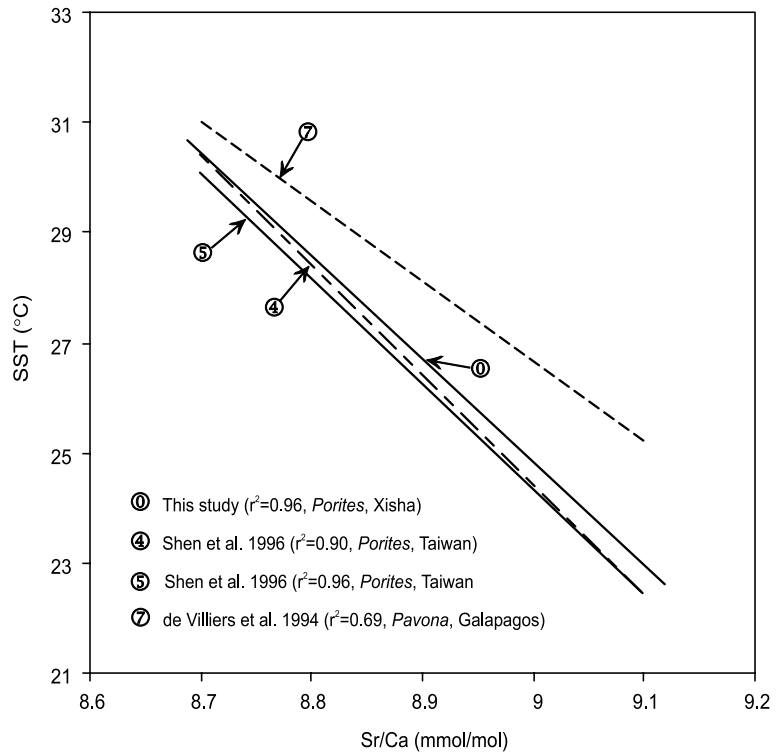
$$\Delta SST = A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \frac{\Delta Sr^{water}}{Sr_{Xisha}^{water}} \quad (7)$$

Because the established Sr/Ca thermometer has the form of $SST_{established} = A \times (Sr/Ca)^{coral} + B$, the normalized Eq. 5 can be derived as follows:

$$\begin{aligned} SST_{normalized} &= SST_{established} - \Delta SST \\ &= A \times \left(\frac{Sr}{Ca}\right)^{coral} + B - A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \frac{\Delta Sr^{water}}{Sr_{Xisha}^{water}} \\ &= A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \left(1 - \frac{\Delta Sr^{water}}{Sr_{Xisha}^{water}}\right) + B \\ &= A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \left(1 - \frac{Sr_{established}^{water} - Sr_{Xisha}^{water}}{Sr_{Xisha}^{water}}\right) + B \\ &= A \times \left(\frac{Sr}{Ca}\right)^{coral} \times \left(2 - \frac{Sr_{established}^{water}}{Sr_{Xisha}^{water}}\right) + B \end{aligned} \quad (8)$$

Among the previously published coral Sr/Ca thermometers, seawater Sr contents were only reported for the Galapagos and Taiwan areas, i.e., 86.52 $\mu mol/kg$ and 84.40 $\mu mol/kg$ (average of seawater filtered at 5 μm and 0.45 μm in Table 2) respectively (de Villiers et al. 1994; Shen et al. 1996). Comparison of the normalized coral Sr/Ca thermometers from these two sites with the Xisha coral Sr/Ca thermometer shows that the disparity between the linear equation for Xisha and Taiwan corals is significantly reduced, but the disparity between the equations for the Xisha and Galapagos corals still exists and is in the opposite direction (Fig. 4). The normalization of coral Sr/Ca thermometers in relation to seawater Sr contents seems to account for the disparity between one pair of sites, but not for the other. The reasons for this probably are (1) the seawater Sr data are not representative of the ambient seawater for the Galapagos site, because the seawater sample was 100 km

Fig. 4 Previously published coral Sr/Ca thermometers with known seawater Sr contents are normalized to the seawater Sr content of Xisha. Comparison with the Sr/Ca thermometer of Xisha coral in this study show that the disparity with the thermometer from Taiwan is significantly reduced. The lines of Shen et al. (1996) used 84.40 $\mu\text{mol/kg}$ of seawater Sr around Taiwan region; and the line of de Villiers et al. (1994) used 86.52 $\mu\text{mol/kg}$ of seawater Sr in the Galapagos region. The number labels of the lines are the same as those in Fig. 2



away from the coral collection site (de Villiers et al. 1994), and only one measurement is available, or (2) other factors such as sampling directions and species of studied corals also affect the coral Sr/Ca thermometers. The coral Sr/Ca thermometer for the Galapagos was developed from samples taken off the axis of maximum growth (de Villiers et al. 1994). However, sampling along the maximum growth extension is considered optimal for SST studies, because distinct differences in coral Sr/Ca ratios were observed between the fast- and slow-growth extensions of the same coral sample (de Villiers et al. 1994; Alibert and McCulloch 1997). In addition, the Galapagos coral is a *Pavona* species, which differs from the *Porites* species usually employed for coral Sr/Ca thermometry (Beck et al. 1992; Gagan et al. 1998).

In summary, our results demonstrate that Ca contents in coral skeletons are very uniform, and the variation of Ca in corals is much smaller (0.4%) than the variation of Ca in seawater, indicating that variations in seawater Ca do not significantly affect the contents of Ca in coral skeletons. The difference in skeletal Ca corresponds to $\pm 0.6^\circ\text{C}$ of uncertainty for coral Sr/Ca thermometers; therefore, it cannot be responsible for the large disparities of previously established coral Sr/Ca thermometers. In contrast, Sr concentrations at different sites exhibit 2.4% variation for tropical waters, and this can account for much of the disparity observed in Sr/Ca thermometers.

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