Paleoceanography and Paleoclimatology

Supporting Information for

Drivers of late Miocene tropical sea surface cooling: a new perspective from the equatorial Indian Ocean

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Introduction

Supplemental Texts and Figures S1 to S5 provide more detailed information about the methods used to reconstruct SST from Mg/Ca. Texts S1 and S2 and Figures S1 and S2 assess Mg/Ca ratio sensitivity to cleaning method and foraminiferal test size. Texts S3 and S4 and Figures S3 to S5 describe and compare the effect of different calibrations, scenarios for seawater Mg/Ca variations, and associated corrections on reconstructed SST.

Supplemental Figures S6 to S9 provide further context to Mg/Ca-SST data presented in the main text. Figure S6 shows indicators of cleaning performance and test dissolution together with SEM images. Figure S7 compares time series analyses of Mg/Ca-SST record from Site U1443 (South Bay of Bengal) to the other existing tropical high-resolution Mg/Ca-SST records from Site 1146 (South China Sea) spanning the time interval from ~8 to 5 Ma. Figure S8 shows long term trends of other existing tropical low resolution SST records based on the TEX_{86} and $U^{K'}_{37}$ indices. Figure S9 is a map showing the effect of $p$$CO_2$ variations on modelled SSTs in tropical Indian Ocean during the late Miocene.

Text S1. Sensitivity of Mg/Ca_{foram} to cleaning protocol

In our samples from Site U1443, test fragments of *T. trilobus* for Mg/Ca analysis were cleaned to remove clay and organic matter, following the « Mg » protocol from Barker et al. (2003) without a reductive step (oxidative cleaning) whereas Dekens et al. (2002) used a « Cd » cleaning protocol from Boyle & Keigwin (1985) and Mashiotta et al. (1999) including a reductive step. It was reported that the « Cd » cleaning protocol often results in a lowering of the Mg/Ca ratio of 8 % compared to the « Mg » protocol by preferential dissolution of the Mg-rich parts of the test (Rosenthal et al. 2004). In order to confidently apply the Dekens et al. (2002) calibration, we tested the sensitivity of Mg/Ca to reductive cleaning in ten samples covering our late Miocene study interval. The samples were split after homogenization of crushed tests and one half was cleaned following the « Mg » protocol and the other half was cleaned following the « Cd » protocol. As observed in Figure S1, the difference in cleaning protocol does not result in a constant Mg/Ca offset, and there is no clear trend between the offset between the two cleaning protocol and the mean individual mass of *T.trilobus* tests. Therefore, we did not apply a correction to raw Mg/Ca ratios to account for the difference in cleaning protocol.

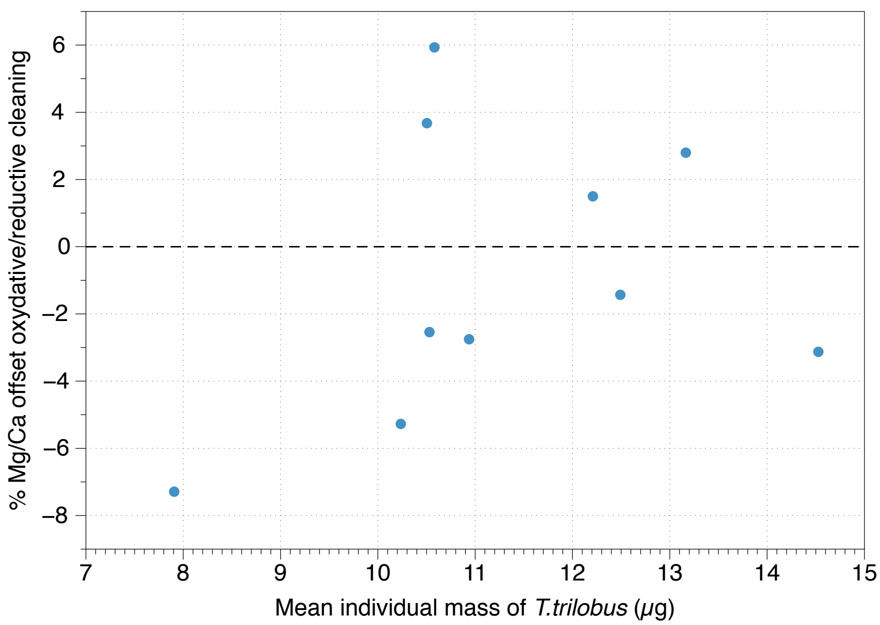

Figure S1. Cleaning protocol sensitivity test.
The % Mg/Ca offset between oxidative and reductive cleaning versus the mean individual mass of *T. trilobus* tests (total mass before crushing/number of individuals in population).

Text S2. Sensitivity of Mg/Ca_{foram} to test size

A test to determine Mg/Ca sensitivity to *T. trilobus* test size was performed for nine samples in two size fractions: 212-315 µm and 355-500 µm. The ΔMg/Ca (Mg/Ca 355-500 µm − Mg/Ca 212-315 µm) is represented as a function of the temperature calculated from Mg/Ca ratios in the 212-315 µm size fraction by applying the calibration of Anand et al. (2003). No correlation is observed between changes in ΔMg/Ca and temperature (Figure S2). For 7 of the 9 samples, Mg/Ca ratio increases with test size. For 6 samples the ΔMg/Ca is relatively constant: the tests from the 350-500 µm size fraction have a Mg/Ca ratio ~0.4 mmol/mol higher than the Mg/Ca of the smaller foraminifera (212-315 µm), representing an average variation of 13.8 % of the Mg/Ca ratio. However, for three of the samples this variation is not observed; for one sample the

ΔMg/Ca is 0.15 mmol/mol, for another sample it is negligible and for the last one it is negative. Therefore, the ΔMg/Ca is not constant over our whole record.

It should be noted that previous work that has investigated the relationship between Mg/Ca ratio and the size of foraminiferal tests shows contradictory trends. Elderfield et al. (2002) and Drury et al., (2018) observe a positive correlation (Mg/Ca ratio increases with test size). Elderfield et al. (2002) suggest that the signal would reflect a change in calcification rate, with smaller individuals calcifying faster and larger individuals forming calcite more in equilibrium with seawater composition. On the contrary, the general trend observed by Friedrich et al. (2012) is a negative correlation (Mg/Ca ratio decreases with test size); these authors interpret this signal as mainly environmentally controlled, and suggest it reflects a change in the habitat of foraminifera during their life cycle, with larger individuals migrating deeper in the water column. As in most of our selected samples, Mg/Ca and size test show a positive correlation, and foraminifera in the two size fractions were selected to exclude mature individuals with gametogenic calcite known to migrate deeper in the water column before reproduction, the second hypothesis from Friedrich et al. (2012) for our data is more unlikely.

However, because the observed positive correlation is not constant in all the samples, the same correction for our entire record is difficult to envisage, the decision was made to not apply a "size" correction to Mg/Ca values to compensate for the difference in size between our samples and those of certain calibrations (Anand et al., 2003), but rather to apply the calibration from Dekens et al. (2002) using a similar size fraction (250-355 µm).

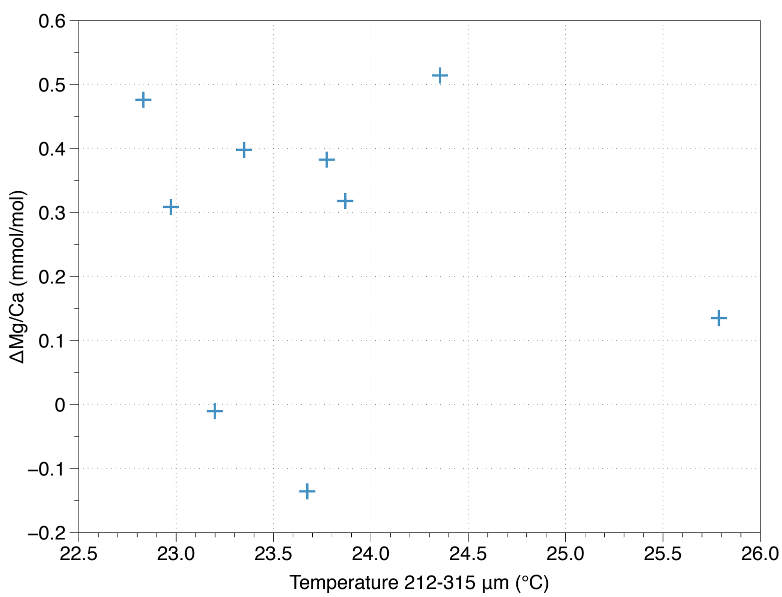

Figure S2. Sensitivity test of Mg/Ca ratio to foraminifera test size.
ΔMg/Ca, calculated as the Mg/Ca ratio (mmol/mol) of the 212- 315 µm size fraction minus the Mg/Ca ratio (mmol/mol) of the 355- 500 µm size fraction, plotted against temperature calculated for the 212-315 µm size fraction using the Anand et al. (2003) calibration equation.

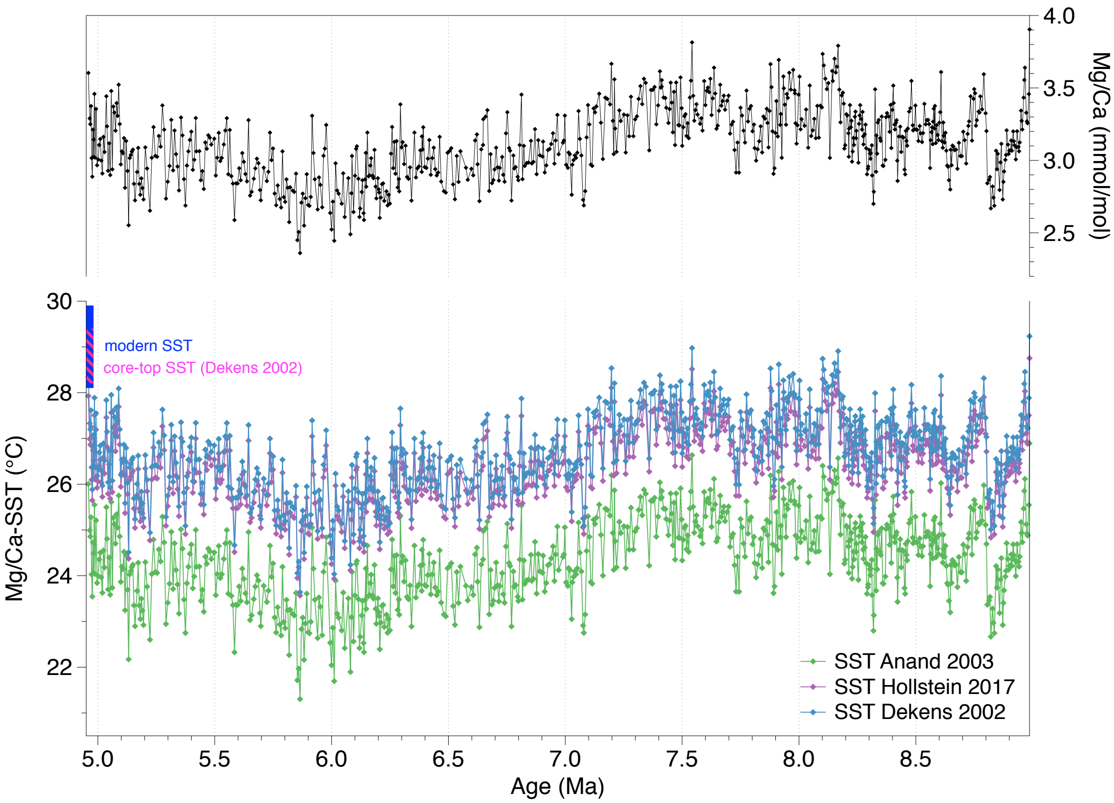

Figure S3. Effect of application of different Mg/Ca-T calibrations on reconstructed SSTs. Black curve: Mg/Ca ratio measured in *T. trilobus* tests, blue curve: SST calculated with the calibration of Dekens et al (2002), green curve: SST calculated with the calibration of Anand et al. (2003), purple curve: SST calculated with the calibration of Hollstein et al. (2017). The SST range from Site U1443 core-top Mg/Ca data is calculated with the calibration of Dekens et al. (2002).

Text S3. Scenarios for Late Miocene Mg/Ca$_{sw}$ reconstruction

In the history of the Phanerozoic ocean, changes in weathering input by rivers, hydrothermal activity, and carbonate deposition have led to changes in Mg and Ca concentration of seawater. The variation in Mg/Ca of seawater (Mg/Ca$_{sw}$) can in turn affect Mg/Ca ratio in foraminiferal tests (Mg/Ca$_{test}$) precipitated in this seawater. Therefore, for studies longer than ~1 Ma (residence time of Ca; Broecker & Peng, 1982) it is important to correct Mg/Ca$_{test}$ for Mg/Ca$_{sw}$ variations to accurately reconstruct Mg/Ca derived temperature. For example, previous studies (Medina-Elizalde et al., 2008; O'Brien et al., 2014) have demonstrated the importance of adjusting Mg/Ca-SST to Mg/Ca$_{sw}$ variations for understanding past climate dynamics and sensitivity.

Because past proxy reconstructions of Mg/Ca$_{sw}$ have low temporal resolution, wide errors bars and are not always in good agreement with each other, Mg/Ca$_{sw}$ reconstruction requires modeling studies. Among many existing scenarios (Berner 2004; Demicco et al., 2005; Fantle & DePaolo, 2006; Farkas et al., 2007; Higgins & Schrag, 2012; Stanley & Hardie, 1998, Wilkinson & Algeo, 1989) we selected three scenarios in agreement with Mg/Ca$_{sw}$ reconstruction derived from proxies and fitting their error bars. The SH98 scenario (Stanley & Hardie 1998) is a model based on seafloor spreading rate and scenarios FD06 (Fantle & DePaolo, 2006) and HS15 (Higgins & Schrag, 2015) are derived from chemical modelling of pore fluids and carbonate sediments (Mg, Ca and Sr) in the Ontong-Java Plateau (ODP Leg 130 Site 807). Works of Higgins & Schrag (2012 and 2015) follow the work of Fantle & DePaolo (2006), but they provide new constraints on the diagenetic effect on pore fluid chemistry. They use Mg isotopic profiles of sediment and pore fluid to determine the partition coefficient of Mg in recrystallized calcite allowing them to determine how much of Mg in pore fluid is due to recrystallization (and not linked to Mg/Ca_{sw} variation). Because the original FD06 scenario implies some unrealistic short-term Mg/Ca_{sw} variations given the residence time of Mg and Ca in the ocean, in Figure S4 a simple linear relationship was applied. Therefore, the HS15 scenario is our preferred scenario, but we compare the effect of the three scenarios (and the effect of H value) on reconstructed SST in the Figure S5.

In Figure S4, we also included the Mg/Ca_{sw} Monte Carlo simulation from Tierney et al. (2019) based on available proxy data (grey line and grey shading). This simulation includes data from corals (Gothmann et al., 2015), leading to considerable variability in terms of Mg/Ca_{sw} estimates. Given thatthe controls on Mg incorporation in modern corals are complex and not fully understood, we did not use this proposed Mg/Ca_{sw} reconstruction in our study.

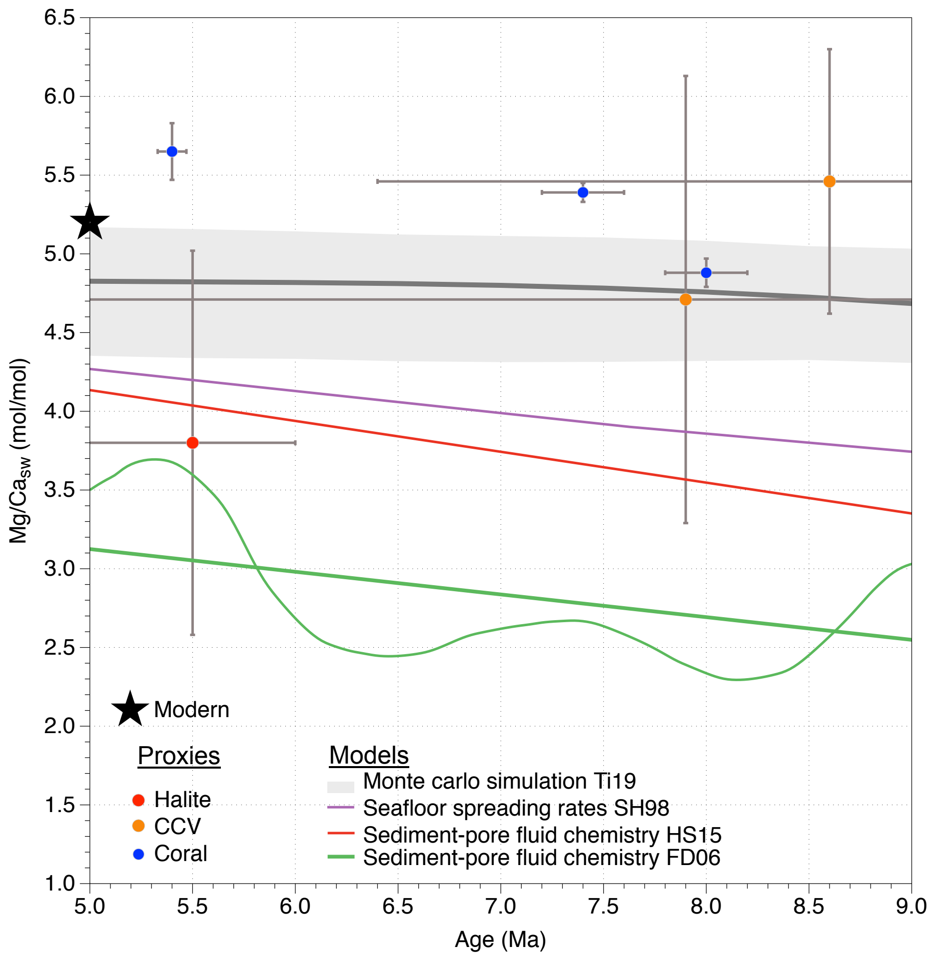

Figure S4. Mg/Ca_{sw} scenarios for the late Miocene (9-5 Ma).
Scenario SH98 in purple (Stanley & Hardie, 1998), scenario HS15 in red (Higgins & Schrag, 2015) and scenario FD06 in green (Fantle & DePaolo, 2006). For scenario FD06 a simple linear

relation was applied instead of the initial scenario. Mg/Ca_{sw} data are from halite fluid inclusions (Horita et al., 2001), fossil corals (Gothmann et al., 2015) and calcium carbonate veins (CCV; Coggon et al.,2010). The Monte Carlo simulation is from Tierney et al. (2019) and is based on proxy data. The modern Mg/Ca_{sw} of 5.2 mol/mol is indicated by the star.

Text S4. Effect of Mg/Ca_{sw} correction and relation between Mg/Ca_{sw} and Mg/Ca_{test} on reconstructed SST

There is no agreement in the literature concerning the Mg/Ca_{sw}-Mg/Ca_{test} relation and whether it is a power law (Evans & Muller, 2012) or linear (Tierney et al., 2019) relation.
The work of Evans & Muller (2012) suggested that the incorporation of Mg into calcite varies non-linearly with $Mg/Ca_{sw,}$ necessitating a power law to correct Mg/Ca_{test} for Mg/Ca_{sw} variations in the following form:

$$Mg/Ca_{test} = \frac{F \times Mg/Ca_{sw}^{t=t\ H}}{F \times Mg/Ca_{sw}^{t=0\ H}} \times Bexp^{AT}$$

With F and H being species-specific constants (for *T. sacculifer* H=0.41; data based on culture experiment from Delaney et al., 1985) and modern Mg/Ca_{sw} = 5.2 mol/mol (Broecker & Peng, 1982). On the other hand, Tierney et al. (2019) found a H value close to 1 for many species of foraminifera including *T. sacculifer* (data also based on culture experiment from Delaney et al., 1985), and suggested that a simple linear relation more adequately defines the Mg/Ca_{sw}- Mg/Ca_{test} relation. We also note that the study of Evans et al. (2016) proposed that the temperature sensitivity of Mg/Ca in foraminifera changes with Mg/Ca_{sw} but this effect has only been detected in culture experiment on one species of planktic foraminifera, *Globigerinoides ruber*, and was not reported in studies of benthic foraminiferal species (De Nooijer et al., 2017).

In order to have a global view on the possible range of absolute SST and amplitude of long term cooling associated with Mg/Ca_{sw} correction and H value, a comparison is made in Figure S5. Mg/Ca_{sw} corrections increase absolute temperature and amplitude of long term variation of reconstructed SST compared to uncorrected Mg/Ca_{sw}-SST. With no correction for Mg/Ca_{sw} the long term gradual cooling recorded from 7.4 to 5.8 Ma is 2.2 °C (Figure S5, blue curve), for HS15 scenario with a linear relation between Mg/Ca_{sw} and Mg/Ca_{test} it is 3.2 °C and with a power law relationship and H value of 0.41 it is 2.6 °C (Figure S5, red curve and red dashed curve). As works of Evans & Muller (2012) and Tierney et al. (2019) are based on the same data derived from the *T. sacculifer* culture experiment from Delaney et al., (1985), we prefer to use the linear relationship in our study as it provides a more simplistic approach, but further culture experiments are needed to better constrain how Mg incorporates into calcite tests of *T. sacculifer* at varying Mg/Ca_{sw}. The two most extreme scenarios are scenario FD06 H=1 with SST values comprised between 35 and 32 °C and a long-term cooling of 3.2 °C (Figure S5 green curve), and scenario SH98 H=0.41 with SST values comprised between 29 and 26 °C and a long-term cooling of 2.5 °C (Figure S5 purple dashed curve). Our preferred scenario HS15 H=1 is in red, SST values are comprised between 32 and 29 °C and the amplitude of long term cooling is 3.2 °C.

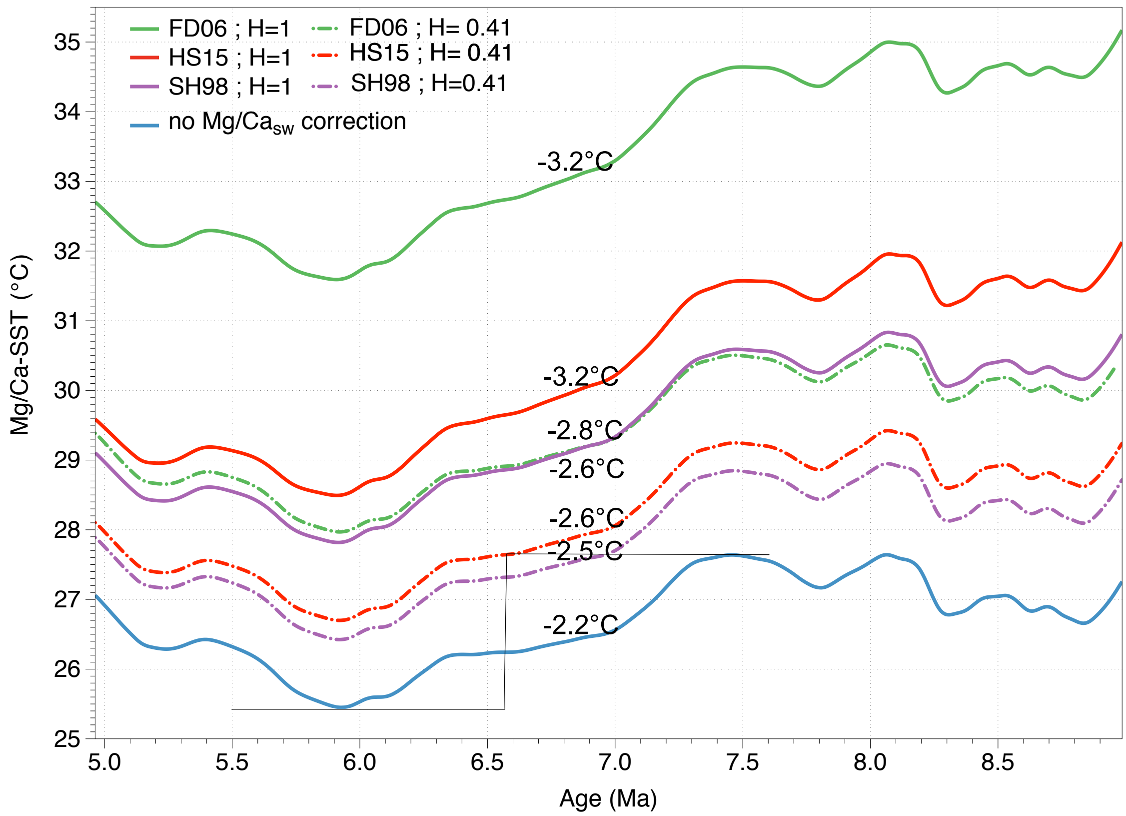

Figure S5. Effect of scenarios SH98, FD06 and HS15, and H value on absolute SST reconstruction and amplitude of variations.

For clarity only the long term SST trend is shown (calculated with a 10 % Lowess filter). Mg/Ca-SSTs were calculated using the Dekens et al. (2002) *T. sacculifer* calibration equation for the Pacific, without correction for Mg/Ca_{sw} (blue curve) and corrected for Mg/Ca_{sw} variability following HS15 scenario (red curves), FD06 scenario (green curves) and SH98 scenario (purple curves), with linear relation (solid lines) and power law relation with H=0.41 for *T. sacculifer* (dashed lines).

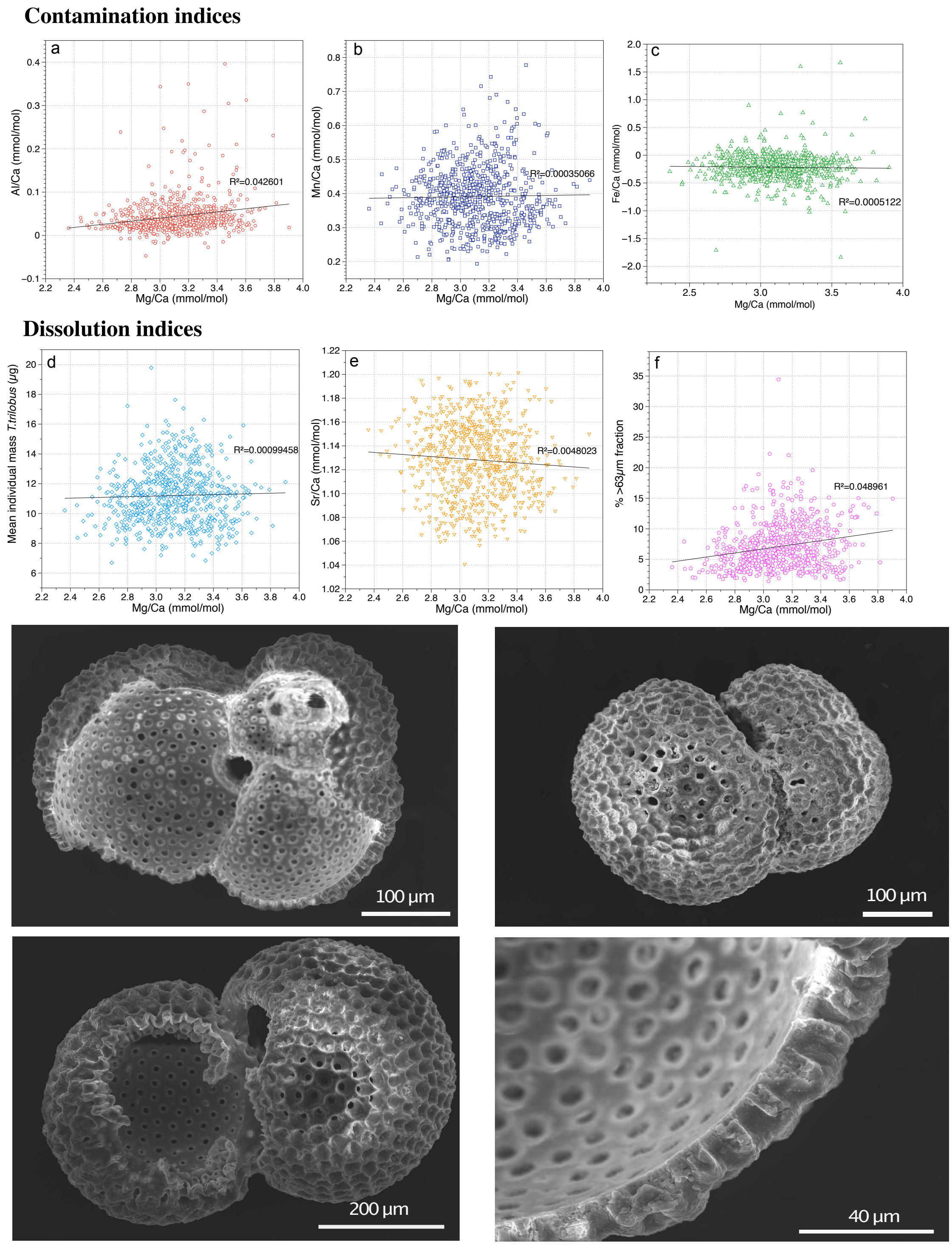

Figure S6. Contamination and dissolution indices and SEM images.
Contamination indices: (a) Al/Ca, (b) Mn/Ca, (c) Fe/Ca (mmol/mol), and dissolution indices: (d) mean individual mass of *T. trilobus* (µg) (e) Sr/Ca (mmol/mol) and (f) % >63 µm coarse fraction showing no significant correlation with Mg/Ca (mmol/mol), suggesting that Mg originates from foraminiferal tests and not from mineral contamination and that dissolution is not driving the Mg/Ca trend. Scanning Electron Microscopy (SEM) images of *T. trilobus* tests showing a good

apparent preservation of tests (inner and external walls, cross-section of wall and pore structure) and minor effect of diagenesis.

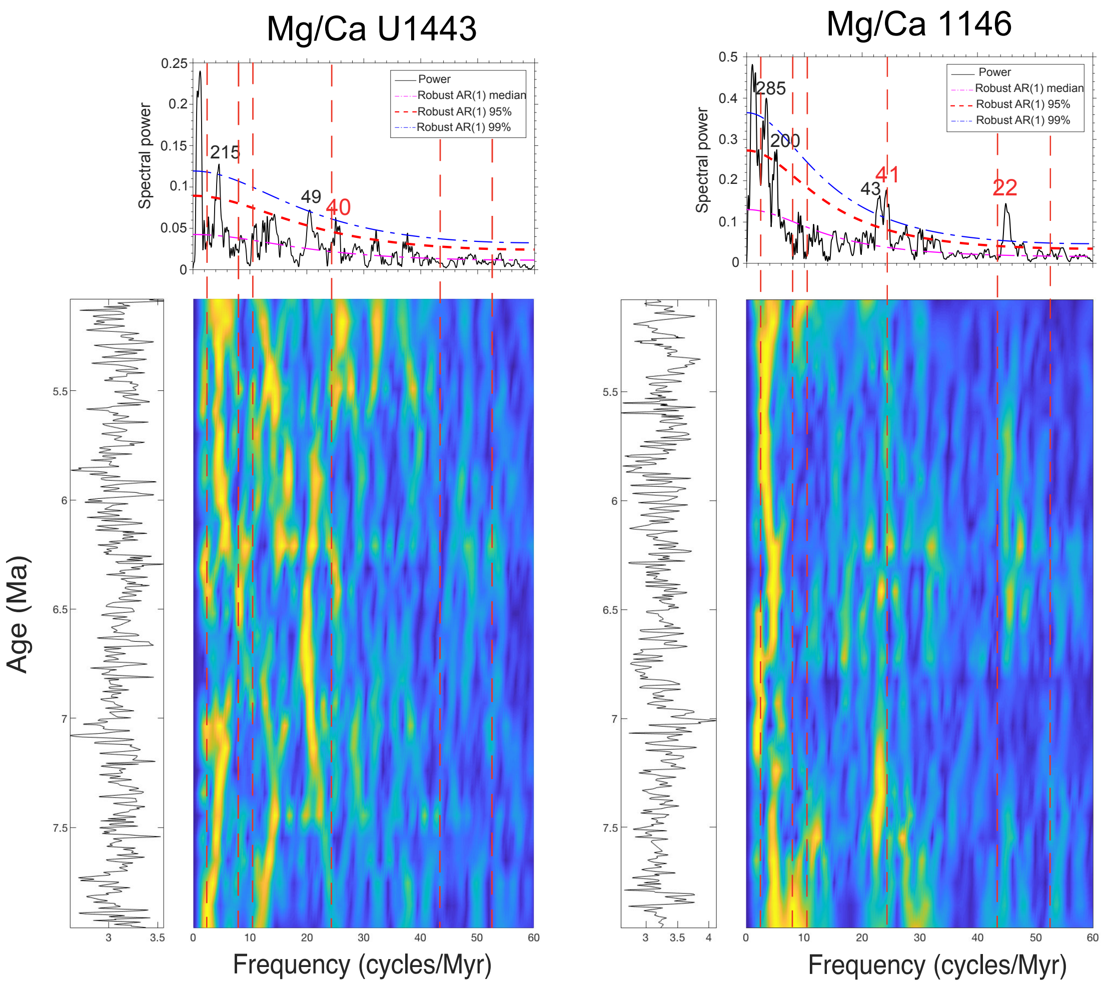

Figure S7. Time series analyses of Mg/Ca from Site U1443 (South Bay of Bengal) and Site 1146 (South China Sea).
Spectral analysis (Multi Taper Method, MTM) and evolutive spectral analysis (Fast Fourier Transform, LAH) (see Methods), left: Mg/Ca at Site U1443 in Southern Bay of Bengal, right: Mg/Ca at Site 1146 in South China Sea (Holbourn et al., 2018). For the two data sets MTM (top) and LAH (right) are performed on filtered records (left) (see Methods). Primary periods are in red and heterodynes are in black, red dotted line indicate periods of 404, 124, 95, 41, 24, 22 and 19 kyr resulting from Earth's orbital periods. Sampling resolution is similar in Site U1443 and Site 1146 indicating that the absence of significant power in the precession band after ~8 Ma and the lower power in the obliquity band at Site U1443 compared to Site 1146 are real features and are not an artifact of lower sampling resolution.

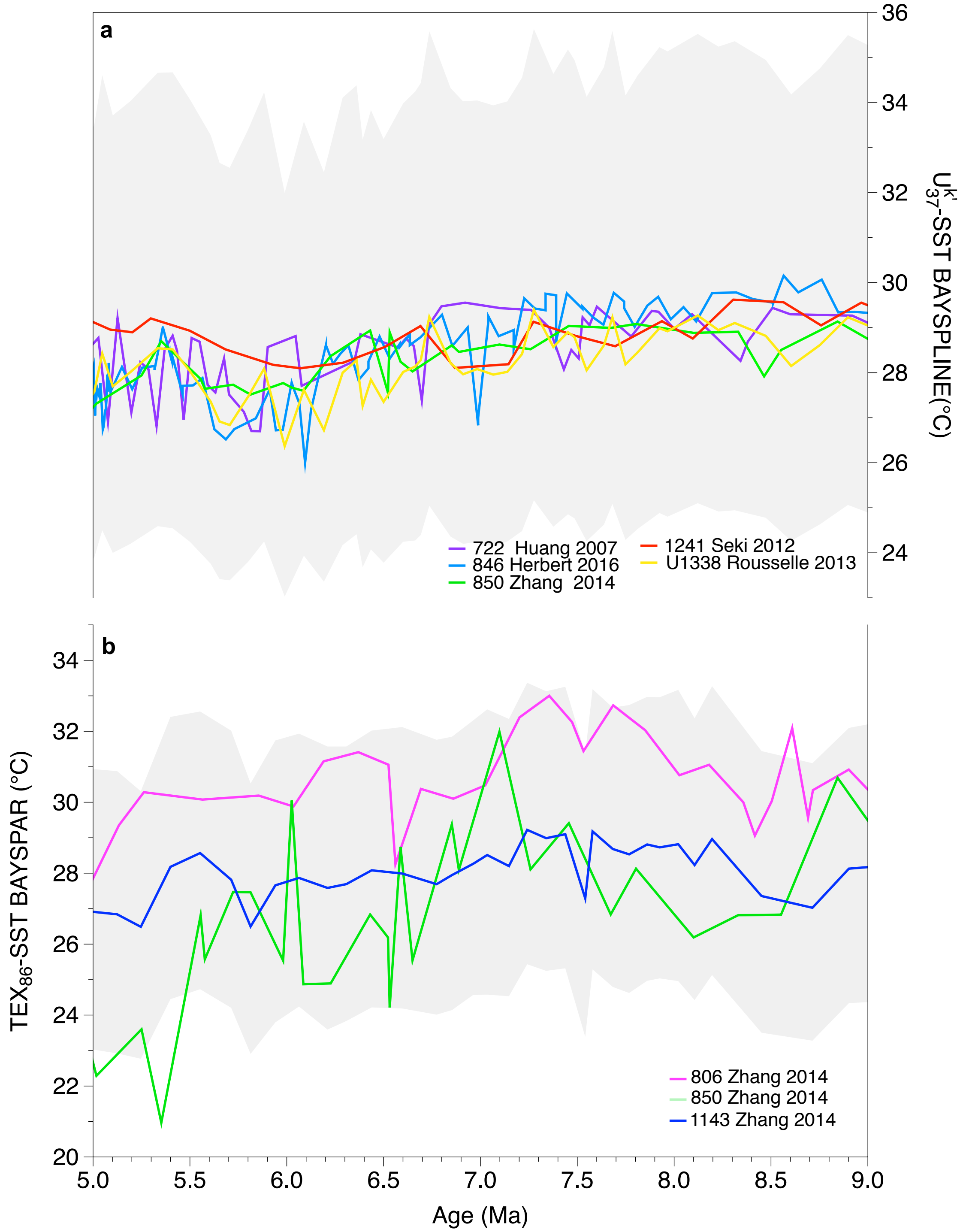

Figure S8. Tropical low resolution temperature records based on $U^{K'}_{37}$ and TEX_{86} indices. (a) Tropical $U^{K'}_{37}$ SST records recalculated using the BAYSPLINE calibration (Tierney & Tingley, 2018). For clarity the 1.5σ uncertainty estimate is only shown for one record (U1338); all records have a similar amplitude error envelope due to similar estimated SSTs. (b) Tropical TEX_{86} SST records recalculated using the BAYSPAR calibration Analog mode (Tierney & Tingley, 2015).

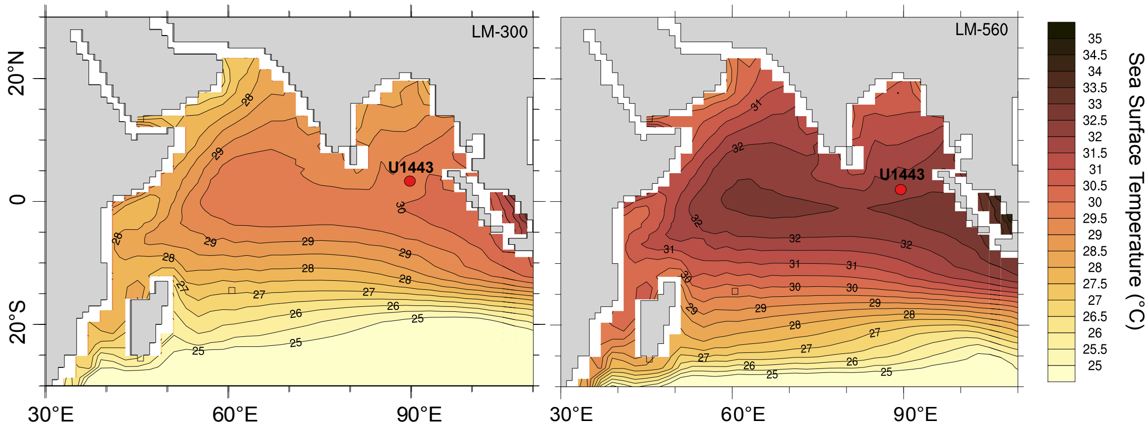

Figure S9. The effect of pCO_2 variations on modelled SSTs in the intertropical Indian Ocean during the late Miocene.

Right: Modelled SSTs with late Miocene boundary conditions and 2X preindustrial pCO_2 (560 ppm), representing condition at ~8 Ma. Left: Modelled SSTs with late Miocene boundary condition and pCO_2 concentrations at 300 ppm, representing conditions at ~6 Ma. In the two maps the paleolocation of Site U1443 at 6 and 8 Ma is shown (left and right, respectively).