



The effects of temperature, salinity, and the carbonate system on Mg/Ca in *Globigerinoides ruber* (white): A global sediment trap calibration

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ABSTRACT

The Mg/Ca of planktic foraminifera *Globigerinoides ruber* (white) is a widely applied proxy for tropical and sub-tropical sea-surface temperature. The accuracy with which temperature can be reconstructed depends on how accurately relationships between Mg/Ca and temperature and the multiple secondary controls on Mg/Ca are known; however, these relationships remain poorly quantified under oceanic conditions. Here, we present new calibrations based on 440 sediment trap/plankton tow samples from the Atlantic, Pacific and Indian Oceans, including 130 new samples from the Bay of Bengal/Arabian Sea and the tropical Atlantic Ocean. Our results indicate temperature, salinity and the carbonate system all significantly influence Mg/Ca in *G. ruber* (white). We propose two calibration models: The first model assumes pH is the controlling carbonate system parameter. In this model, Mg/Ca has a temperature sensitivity of $6.0 \pm 0.8\%/^{\circ}\text{C}$ (2σ), a salinity sensitivity of $3.3 \pm 2.2\text{‰}$ and a pH sensitivity of $-8.3 \pm 7.7\%/0.1$ pH units; The second model assumes carbonate ion concentration ($[\text{CO}_3^{2-}]$) is the controlling carbonate system parameter. In this model, Mg/Ca has a temperature sensitivity of $6.7 \pm 0.8\%/^{\circ}\text{C}$, a salinity sensitivity of $5.0 \pm 3.0\text{‰}$ and a $[\text{CO}_3^{2-}]$ sensitivity of $-0.24 \pm 0.11\text{‰}/\mu\text{mol kg}^{-1}$. In both models, the temperature sensitivity is significantly lower than the widely-applied sensitivity of $9.0 \pm 0.6\%/^{\circ}\text{C}$. Application of our new calibrations to down-core data from the Last Glacial Maximum, considering whole ocean changes in salinity and carbonate chemistry, indicate a cooling of $2.4 \pm 1.6^{\circ}\text{C}$ in the tropical oceans if pH is the controlling parameter and $1.5 \pm 1.4^{\circ}\text{C}$ if $[\text{CO}_3^{2-}]$ is the controlling parameter.

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1. Introduction

Planktic foraminiferal Mg/Ca paleothermometry is one of the most widely applied techniques to reconstruct sea-surface temperature (SST) (e.g. Lea, 2000; Weldeab et al., 2007; Schmidt and Lynch-Stieglitz, 2011; Mohtadi et al., 2014). Fundamental to robust SST reconstruction is accurate knowledge of the relationship between foraminiferal Mg/Ca and temperature, as well as the multiple secondary influences on Mg incorporation into foraminiferal calcite. The substantial biological mediation of Mg incorporation

into foraminiferal calcite necessitates species-specific calibrations (Nürnberg et al., 1996; Rosenthal et al., 1997; Lea et al., 1999; Erez, 2003).

Globigerinoides ruber (white) is the most widely used species for reconstructing tropical and subtropical SST due to its cosmopolitan nature, high abundance, and shallow habitat depth (0–50 m) (Hemleben et al., 1989; Schiebel and Hemleben, 2017). Estimates of the sensitivity of Mg/Ca in *G. ruber* (w) to temperature fall between 5–10% per $^{\circ}\text{C}$ (Table 1), and the most widely-applied calibration has a sensitivity of $9.0 \pm 0.6\%/^{\circ}\text{C}$ (2σ) (Dekens et al., 2002; Anand et al., 2003). Studies of *G. ruber* (w) grown in laboratory culture, where calcification temperature is well constrained, yield a temperature sensitivity of $8 \pm 3\%/^{\circ}\text{C}$ (Kisakürek et al., 2008).

Since the early development of Mg/Ca paleothermometry, salinity and carbonate chemistry have been known to exert a secondary influence on Mg/Ca (Nürnberg et al., 1996; Lea et al., 1999). Esti-

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Table 1
Published temperature sensitivities^a for *G. ruber* (white)^b.

Reference	Method	Temperature sensitivity (%) ^c
Lea (2000)	Coretops, equatorial Pacific	8.9 ± 1.4
Dekens et al. (2002)	Coretops, multivariate fit accounting for core depth	9 ± 1.5
Anand et al. (2003)	Multi-species, sediment trap, Sargasso Sea, $\delta^{18}\text{O}$ temperature	9.0 ± 0.6
Anand et al. (2003)	250–350 μm , sediment trap, Sargasso Sea, $\delta^{18}\text{O}$ temperature	10 ± 2
Anand et al. (2003)	350–500 μm , sediment trap, Sargasso Sea, $\delta^{18}\text{O}$ temperature	8.5 ± 1.2
McConnell and Thunell (2005)	Sediment trap, Gulf of California, satellite temperature	6.8
Kisakürek et al. (2008)	Laboratory cultures	8 ± 3
Mohtadi et al. (2009)	Sediment trap, Java, satellite temperature	8.4
Mohtadi et al. (2009)	Sediment trap, Java, $\delta^{18}\text{O}$ temperature	6.6
Khider et al. (2015)	Coretops, Bayesian multivariate fit accounting for salinity, and bottom water carbonate ion saturation	8.7 ± 0.9

^a Excluding studies where the temperature sensitivity was assumed.

^b Including one study combining multiple species of planktonic foraminifera.

^c 95% confidence interval.

mates of the sensitivity of Mg/Ca in *G. ruber* to salinity vary widely. Laboratory culture studies indicate a salinity sensitivity of 3–5% per salinity unit (Kisakürek et al., 2008; Hönisch et al., 2013), whereas several core-top studies suggest a significantly higher sensitivity of 15–29%/PSU (Ferguson et al., 2008; Arbuszewski et al., 2010; Mathien-Blard and Bassinot, 2009), although subsequent work has revised down these core-top estimates (Bousetta et al., 2011; Hertzberg and Schmidt, 2013; Hönisch et al., 2013). More recently, a Bayesian multivariate fit to core-top data suggested a salinity sensitivity of $3.9 \pm 1.2\%$ /PSU (Khider et al., 2015), similar to the results of the culture studies.

Laboratory culture studies show that the carbonate chemistry of seawater, as expressed by pH or carbonate ion concentration ($[\text{CO}_3^{2-}]$), significantly effects planktic foraminiferal Mg/Ca. Mg/Ca decreases as pH (and $[\text{CO}_3^{2-}]$) increases, with a sensitivity of ~5–10% per 0.1 pH unit (Lea et al., 1999; Russell et al., 2004; Kisakürek et al., 2008; Evans et al., 2016). Despite the significant effect of carbonate chemistry observed within culture studies, the influence of carbonate chemistry on Mg/Ca in planktic foraminifera that have calcified under oceanic conditions (i.e. none laboratory-grown) has not been previously investigated.

Here, we assess the relationship between Mg/Ca in *G. ruber* (w) and temperature, salinity, and the carbonate system using samples collected by sediment trap and plankton tow. Sediment trap and plankton tow samples minimise the influence of post-depositional calcite dissolution and/or secondary overgrowth deposition that hinder core-top studies, whilst allowing the analysis of foraminifera that have calcified under oceanic conditions within a well constrained time-period. We have generated new data from a transect of sediment traps located along an isotherm with a strong salinity gradient in the Bay of Bengal and Arabian Sea, which we use to assess the effects of salinity. We have also generated new data from a sediment trap in the tropical Atlantic Ocean, off Mauritania; in these samples, we separated foraminifera by morphotype and into narrow size fractions to assess the effects of biological variability on Mg/Ca. Our newly generated data are combined with all previously published *G. ruber* (w) Mg/Ca data from foraminifera collected by sediment trap and plankton tow. We use climatological data to calculate temperature, salinity, pH and $[\text{CO}_3^{2-}]$ at the depth habitat range of *G. ruber* (w) for each of the newly generated and previously published trap/tow samples, and constrain the individual effects of temperature and salinity, pH and $[\text{CO}_3^{2-}]$ on Mg/Ca using subsets of data where covariance between environmental variables is negligible.

2. Materials and methods

2.1. Arabian Sea and Bay of Bengal sediment trap samples

We utilised 51 samples from sediment trap deployments NBBT09 (17.383°N, 89.700°E; 1450 m water depth), CBBT06 (11.033°N, 84.433°E; 899 m water depth), SBBT09 (5.400°N, 86.767°E; 886 m water depth), and JGOFS AS02-M5 (10.003°N, 65.005°E; 2363 m water depth) (Unger et al., 2003; Honjo et al., 1999) (see supplementary material (SM) and Table S1). These sediment trap deployments form a transect spanning a large salinity gradient (~4 units), with ~no change in mean annual temperature between the sites (Fig. 1). Foraminifera were picked from the 200 to 400 μm size fraction (see SM), and between 5–30 individuals of *G. ruber* (w) *sensu stricto* (Wang, 2000) were used per sample for trace element analysis.

Foraminifera were cleaned following a modified version of the method of Pak et al. (2004) and the acidified samples were analysed at Rutgers University using magnetic-sector ICP-MS (Thermo Element XR) (SM). Reproducibility of consistency standards with a Mg/Ca similar to *G. ruber* (w) was 0.03 mmol/mol (0.6%) (2σ). Six of the foraminiferal samples (~12%) were split into replicates and cleaned/analysed separately, with a reproducibility of 0.12 mmol/mol (2.2%) (2σ). The resulting Mg/Ca data are given in Table S2.

2.2. Tropical Atlantic sediment trap samples

We measured Mg/Ca ratios in foraminifera collected by a sediment trap located off Mauritania, in the eastern tropical Atlantic (Fig. 1). The samples were collected in trap deployments CB-7, CB-9, CB-10, CB-12 and CB-13 located at 21.27°N, 20.75°W between 705–3610 m water depth (Fischer et al., 2016) (SM and Table S1).

We separated the foraminifera from each trap sample by morphotype (Wang, 2000), and into narrow size fractions to assess the potential impact of morphotype and test size on Mg/Ca, resulting in a total of 79 foraminiferal samples. 15–25 individual foraminifera were cleaned following a modified version of the method of Pak et al. (2004) and the acidified samples were analysed by ICP-OES (Perkin Elmer Optima 3300 R) at the University of Bremen (SM and Table S2). Reproducibility of our consistency standard was 0.14 Mg/Ca mmol/mol (~2%) (2σ).

2.3. Calculating calcification temperature and salinity

As the vast majority of sediment trap deployments do not have associated continuous in-situ CTD measurements of temperature and salinity, previous studies have used either $\delta^{18}\text{O}_{\text{calcite-water}}$

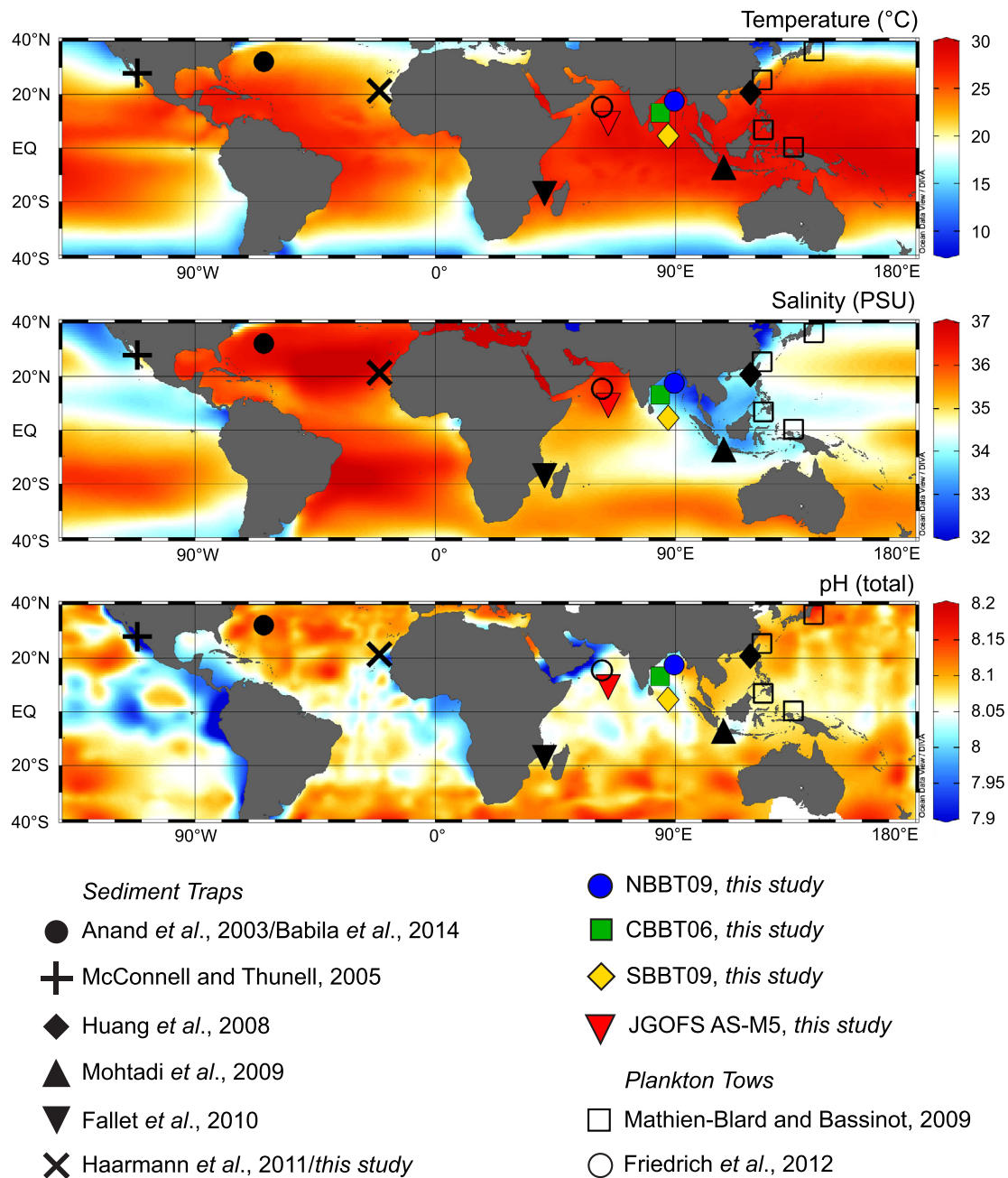


Fig. 1. Location of sediment traps (solid symbols) and plankton tows (open symbols) used in this study, overlaid on mean annual SST and SSS at 25 m depth from WOA13 (Boyer et al., 2013), and pH at 25 m depth from GLODAPv2 (Key et al., 2015; Lauvset et al., 2016). Note, the tropical Atlantic trap site used in this study is located ~120 nm further offshore than that of Haarmann et al. (2011). Published Mg/Ca data are from Anand et al. (2003), McConnell and Thunell (2005), Huang et al. (2008), Mathien-Blard and Bassinot (2009), Mohtadi et al. (2009), Fallet et al. (2010), Haarmann et al. (2011), Friedrich et al. (2012) and Babila et al. (2014).

or satellite temperatures (see SM). Here, we calculate temperature/salinity at the depth habitat of *G. ruber* (w) for the time-period represented by each trap/tow sample using WOA13 monthly climatologies (Boyer et al., 2013) (Fig. 2). We use Monte Carlo simulation to fully propagate the uncertainty in temperature and salinity relating to habitat depth range, error associated with the climatological mean (a combination of short-term and inter-annual variability), and change in temperature/salinity during the sampling period (SM). Following this approach, samples from locations/time-periods with significant temperature and salinity variation within upper water-column or substantial short term and/or inter-annual variability are associated with a wide distribution of temperature and salinity; samples from locations/time-periods with a more homogeneous upper water-column or less short term and/or inter-

annual variability are associated with a narrower distribution of temperature and salinity. These distributions of temperature and salinity are fully propagated as uncertainty in our regressions (section 2.6). All temperature and salinity values reported are for the 0–50 m depth range (Hemleben et al., 1989; Schiebel and Hemleben, 2017), unless otherwise stated. Hydrographic data for all samples are shown on Fig. S1 and given in Table S2.

To test the accuracy of the temperature estimates generated by this approach we used the WOA13 monthly climatological temperature to predict the temperatures at the only trap site with continuous CTD coverage, located within the Sargasso Sea (SM and Fig. S2). The result demonstrates that the differences between the CTD temperatures and climatological temperatures are normally distributed around a mean of ~0 ($p > 0.88$) (Fig. S2). Thus, while

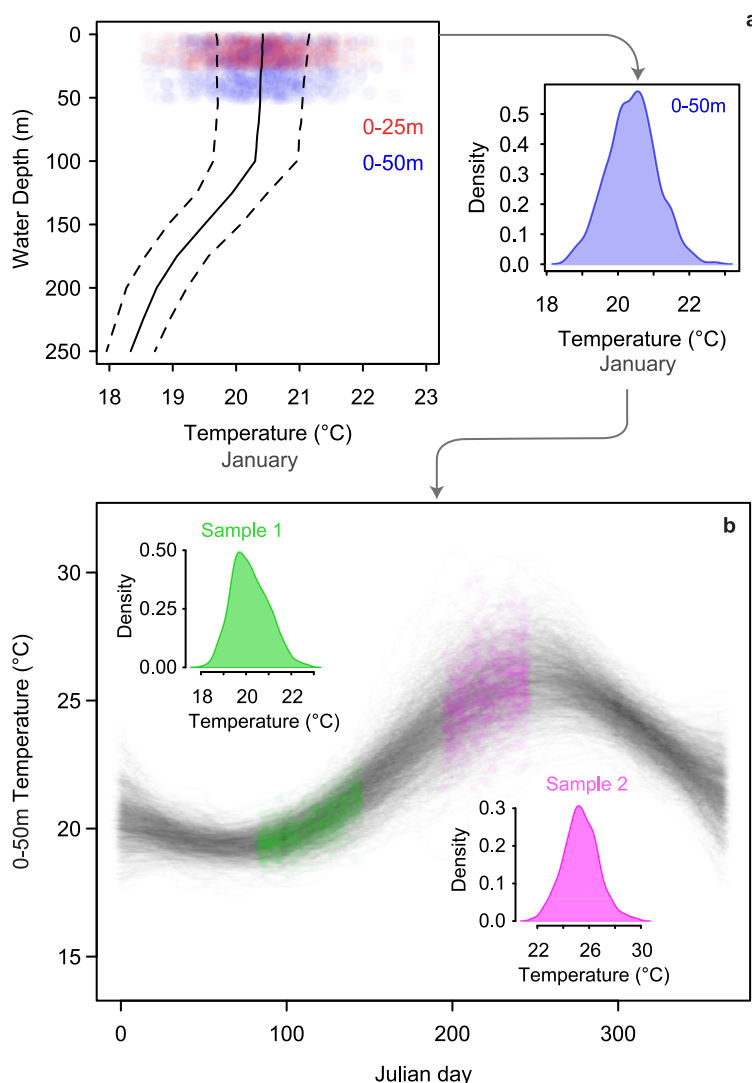


Fig. 2. Calculation of temperature and salinity for time-period represented by each trap sample at habitat depth of *G. ruber* (white) using WOA13 monthly climatologies at each trap site and a Monte-Carlo approach; (a) for each month, a random value is drawn from the population of temperatures/salinities from within the specified depth habitat range, accounting for the uncertainty associated with the climatological mean (blue dots = 0–50 m, red dots = 0–25 m) (b) a random sample is then drawn from these habitat-depth temperature/salinity populations for each month, and the data is fitted with a GAM (versus Julian day). A day is then randomly drawn from between the open/close date of each trap sample (after adjustment for sinking and calcification time), and the temperature/salinity for that day is calculated based on the GAM fit. This process is repeated 10,000 times to fully explore the range in possible calcification temperatures and salinities for each trap sample, accounting for the depth habitat, error associated with the WOA13 monthly climatologies (short term and inter-annual variability), and the period of time represented by each trap sample. The example shown is temperature data for the Sargasso Sea (Anand et al., 2003; Babila et al., 2014). After first generating monthly carbonate chemistry climatologies for each site (section 2.4), the same process is used to calculate carbonate chemistry estimates for each trap/tow sample. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the use of climatological temperature may result in random error (noise) in the calibration, it does not lead to systematic error (inaccuracy) in the calibration. In section 3.9, we discuss how much of the noise results from the use of climatological temperature.

2.4. Estimation of carbonate system parameters

As there is no carbonate chemistry database with a sufficient temporal, spatial and depth resolution to provide carbonate system estimates in the depth habitat of *G. ruber* (w), we first generated monthly carbonate system parameter estimates for each trap/tow site throughout the upper water-column. This is achieved by combining the gridded GLODAPv2 dissolved inorganic carbon (DIC) and alkalinity (Alk) data for the interior ocean (Key et al., 2015; Lauvset et al., 2016) with monthly DIC and Alk estimates for the surface ocean, calculated using the monthly $p\text{CO}_2$ climatologies

from the Landschützer et al. (2014a, 2014b) gridded database and estimates of alkalinity from the algorithms of Lee et al. (2006). Monthly values for DIC and Alk throughout the upper ocean were then obtained by interpolating between the surface value and the seasonally invariant ocean interior value using density. pH and $[\text{CO}_3^{2-}]$ were calculated from DIC and Alk using the OCMIP2 routines (SM).

To obtain pH and $[\text{CO}_3^{2-}]$ for the time interval represented by each sample at the habitat depth of *G. ruber* (w), the monthly gridded values were interpolated using the method described above for temperature and salinity, with Monte-Carlo simulation to account for the uncertainty in the estimates relating to habitat depth and sampling interval (SM and Fig. 2). All pH and $[\text{CO}_3^{2-}]$ values reported are for the 0–50 m depth range, unless otherwise stated. pH and $[\text{CO}_3^{2-}]$ estimates for all samples are shown in Fig. S2, and are given in Table S2.

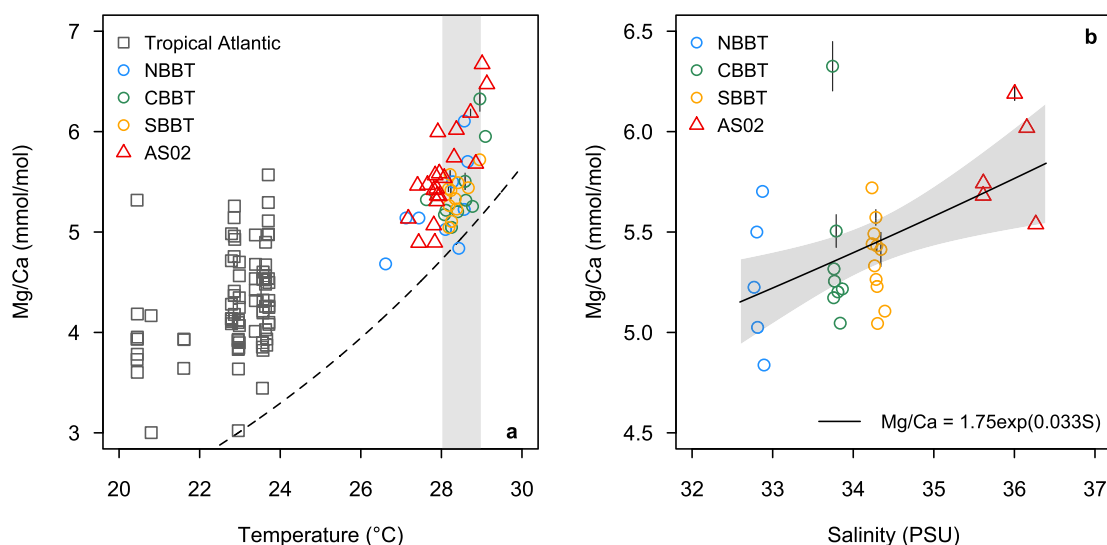


Fig. 3. (a) New Mg/Ca data from the Arabian Sea/Bay of Bengal and the tropical Atlantic, analysed as part of this study, plotted versus climatological temperature (0–50 m, Methods). The calibration of Dekens et al. (2002) and Anand et al. (2003) is shown by the dashed line. The grey box represents data between 28 and 29 °C shown in panel b. Error bars are $\pm 1\sigma$ (b) Mg/Ca versus salinity in the Arabian Sea/Bay of Bengal. Only samples with a temperature between 28–29 °C (grey box in a), and a pH of >8.00 are plotted. The best fit to this subset of data is shown by the solid line, with the 95% confidence interval shown in grey. The slightly higher spread in the NBBT data is likely due to the fewer number of individual foraminifera comprising each sample analysed.

2.5. Compilation of sediment trap/plankton tow data

The compilation of previously published data comprises 310 data points from 16 sediment trap/plankton tow sites in the Atlantic, Pacific and Indian oceans (Fig. 1, SM and Table S1). All Mg/Ca data, along with the original reference for the Mg/Ca data, size fraction, and morphotype are given in Table S2. When combined, our newly generated Mg/Ca data (130 samples) and the compilation of previously-published data comprise 440 sediment trap/plankton tow samples from 20 sites in Atlantic, Pacific and Indian basins. Within the dataset temperature, salinity, pH (seawater scale) and $[CO_3^{2-}]$ range from 18.4 to 29.1 °C, 32.6 to 36.7 PSU, 7.99 to 8.12 pH units, and 191 to 252 $\mu\text{mol/kg}$, respectively (Fig. S1). The variation in temperature within the dataset is mainly driven by seasonal variability at each site, whereas the variation in salinity is mainly driven by the location of the sites in differing oceanic regions. Variations in pH and $[CO_3^{2-}]$ are driven by both seasonal variability and regional differences; this essentially decouples the temperature, salinity and pH/ $[CO_3^{2-}]$ variation within the dataset. There is, however, covariance between temperature and salinity ($r = -0.63$), temperature and pH ($r = -0.68$), salinity and pH ($r = -0.67$), salinity and $[CO_3^{2-}]$ ($r = -0.52$), and pH and $[CO_3^{2-}]$ ($r = -0.55$) (Fig. S1).

2.6. Regression analysis

We first analyse the global dataset before taking subsets of this dataset to circumvent covariance between predictor variables, and better constrain the sensitivity of Mg/Ca to temperature, salinity and the carbonate system. Coefficients are from linear and non-linear least-squares regression. Confidence intervals are derived from bootstrapping (Efron, 1979), and we account for the uncertainty in predictor variables (T, S, pH, $[CO_3^{2-}]$) with Monte-Carlo simulation. Following this approach, the uncertainty associated with the T, S, pH and $[CO_3^{2-}]$ estimate of each sample is fully propagated through to the uncertainty associated with the derived sensitivities. All confidence intervals reported are $\pm 2\sigma$, unless otherwise stated. Additional regression results are given in Table S3.

3. Results and discussion

3.1. Arabian Sea/Bay of Bengal and Tropical Atlantic data

All newly generated Mg/Ca data are shown on Fig. 3. Within the Arabian Sea/Bay of Bengal dataset temperature, salinity, pH and $[CO_3^{2-}]$ range from 26.6 to 29.1 °C, 32.58 to 36.49 PSU, 7.99 to 8.05 pH units, and 191 to 246 $\mu\text{mol/kg}$, respectively (Fig. S1); Mg/Ca ranges from 4.68 to 6.67 mmol/mol (Fig. 3). These data are discussed in detail in section 3.4, where we use the data to constrain the effect of salinity on Mg/Ca.

Within the newly generated tropical Atlantic dataset temperature, salinity, pH and $[CO_3^{2-}]$ range from 20.4 to 23.7 °C, 36.47 to 36.70 PSU, 8.06 to 8.07 pH units, 217 to 232 $\mu\text{mol/kg}$ (Fig. S1); Mg/Ca ranges from 3.00 to 5.57 mmol/mol (Fig. 3).

3.2. Biological variability

Before investigating the environmental controls on Mg/Ca, we first assess the potential influence of morphotype and test size on Mg/Ca. To explore whether there is a significant difference in Mg/Ca between the *sensu stricto* and *sensu lato* morphotypes of *G. ruber* (w) (Wang, 2000) we focus on the newly generated data from the tropical Atlantic, where Mg/Ca was analysed on both the *sensu stricto* and *sensu lato* morphotypes from the same sediment trap cup. A Student's T test shows no significant difference in mean Mg/Ca of the two morphotypes ($p = 0.21$; $n = 32$) (Fig. S3). A combined analysis our or newly generated data with previously published data from the Indian Ocean (Mohtadi et al., 2009) demonstrates there is no significant difference in the mean, or variance, of Mg/Ca in the two morphotypes across the Atlantic and Indian basins (Fig. 4a).

Test size is also known to influence Mg/Ca in *G. ruber* (w) (Friedrich et al., 2012). To assess the possible effects of test size we utilise our newly generated dataset from the tropical Atlantic, in which foraminifera were separated into narrow size fractions. While Mg/Ca generally increases with increasing test size, $\sim 33\%$ of the samples do not show an increasing trend with test size (Fig. 4). Overall, there is no significant difference in mean Mg/Ca in the 150 to 425 μm size range, however the variance in the 300–425 μm

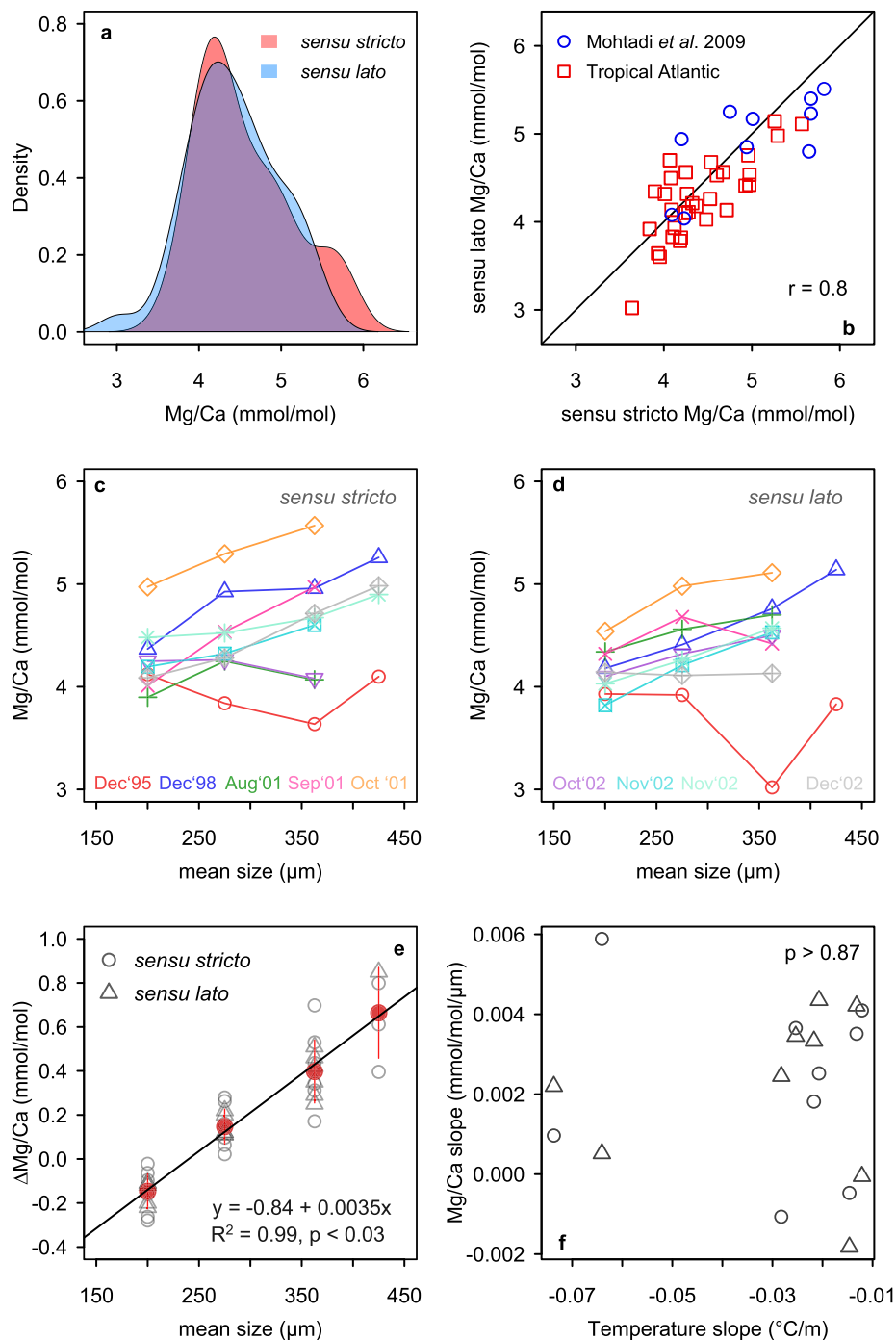


Fig. 4. (a) probability density of Mg/Ca in *G. ruber* (w) *sensu stricto* and *G. ruber* (w) *sensu lato* (all size fractions) from paired samples (b) Mg/Ca in *G. ruber* (w) *sensu stricto* versus Mg/Ca in *G. ruber* (w) *sensu lato* in paired samples. There is no significant difference in mean Mg/Ca or variance between the two morphotypes. Mg/Ca versus mean test size in (c) *G. ruber* (w) *sensu stricto* and (d) *G. ruber* (w) *sensu lato*, with different colours/symbols representing different trap samples (e) Δ Mg/Ca (the difference relative to the mean Mg/Ca of the 150–250 μ m and 250–300 μ m size fraction for each sample) plotted against mean test size, excluding the 33% of data that do not show an increasing trend. Red circles show the mean Δ Mg/Ca of both morphotypes for each size fraction. Error bars are $\pm 1\sigma$ (f) slope of the temperature change in the upper 50 m of water column during the time-period represented by each sample (Δ Temperature/ Δ depth) versus slope of Mg/Ca-size relationship in each sample (Δ Mg/Ca/ Δ size) (see SM). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

size fraction is significantly higher than the 150–250 μ m size fraction (Fig. S4). Excluding the 33% of data that do not show a Mg/Ca increase with test size, the data appear to fall on similar trend line of Δ Mg/Ca, with a slope of $0.35 \text{ mmol mol}^{-1}/100 \mu\text{m}$ (Fig. 4e). The samples showing no trend are not related to a particular season or year, and within the same cup sample the *sensu stricto* and *sensu lato* morphotypes often show differing trends. Comparing the slope of the relationship between Mg/Ca and test size and the slope of

temperature change with depth in the water-column during the interval of time represented by the trap sample shows no significant relationship (Fig. 4f); this likely indicates the relationship between Mg/Ca and test size is not due to changing environmental conditions within the water-column and differing habitat-depths of each size fraction. A recent study suggested that the size of planktic foraminifera may influence the biomineralisation response to changing carbonate chemistry (Henehan et al., 2017). While it

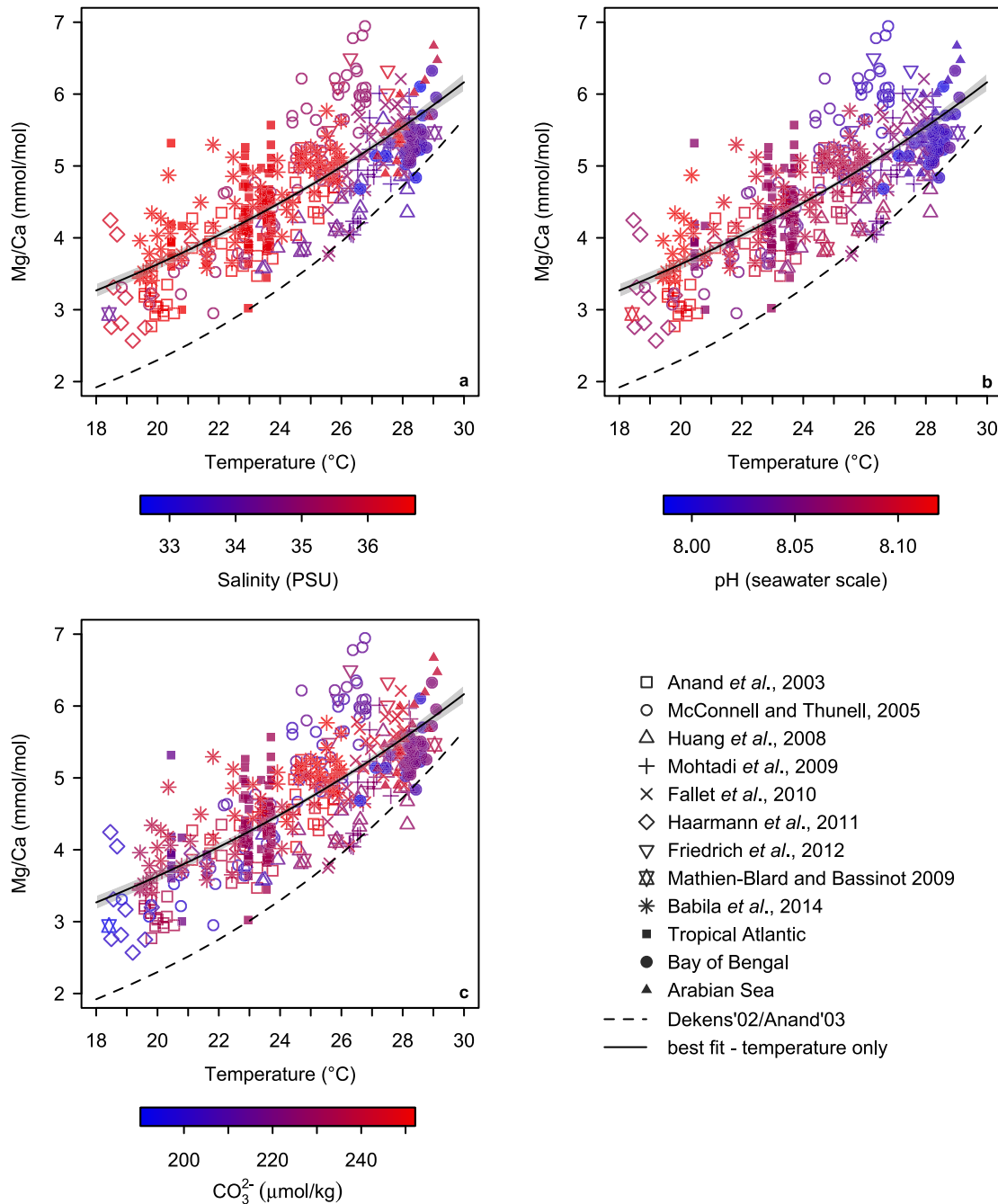


Fig. 5. Mg/Ca plotted against climatological temperature (0–50 m, Methods) (a) salinity represented by colour (b) pH represented by colour, and (c) $[\text{CO}_3^{2-}]$ represented by colour. The best fit (accounting for only temperature) is shown by the solid line (Eq. (S1)), with the 95% confidence interval shown in grey. The calibration of Dekens et al. (2002) and Anand et al. (2003) ('multi-species') is shown by the dashed line. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

is possible that the sensitivity with which Mg/Ca responds to an environmental variable (T , S , pH , $[\text{CO}_3^{2-}]$) may also be affected by size, it is not possible to test this hypothesis with the newly generated tropical Atlantic data due to the limited range in environmental variables at this site (Fig. S4). Our results suggest that while test size can clearly influence Mg/Ca, the relationship between test size and Mg/Ca is likely to vary not only temporally at sites (Fig. 4c and 4d), but also between sites, and cannot be systematically corrected for. We will return to test size in section 3.9 in a discussion of potential sources of noise within the calibration.

3.3. Global calibration

When our newly generated Mg/Ca data are combined with all previously published Mg/Ca data from sediment traps/plankton tows (section 2.5), Mg/Ca within the dataset ranges from 2.57 to 6.94 mmol/mol and generally increases with temperature (Fig. 5). Regressing Mg/Ca against only temperature results in a temperature sensitivity of $5.3 \pm 0.4\%/^{\circ}\text{C}$ ($p < 10^{-16}$) (Eq. (S1)). This is significantly lower than the widely-applied $9.0 \pm 0.6\%/^{\circ}\text{C}$ sensitivity of the calibration of Dekens et al. (2002) and Anand et al. (2003) (Fig. 5). Throughout the text, we refer to the 'multi-species' equation of Anand et al. (2003), as this is by far the most widely

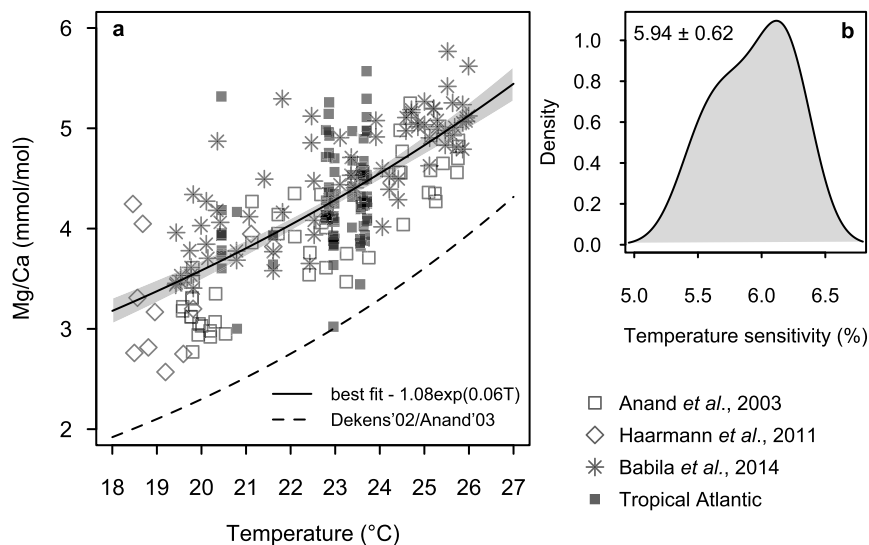


Fig. 6. (a) Mg/Ca versus climatological temperature (0–50 m, Methods) from a subset of data with a narrow range in salinity and significantly reduced covariance between temperature and salinity/pH. The best fit to this subset of data is shown by the solid line, with the 95% confidence interval shown in grey. The calibration line of Dekens et al. (2002) and Anand et al. (2003) is shown by the dashed line. Note the calibration line of Dekens et al. (2002) and Anand et al. (2003) does not fit the data of Anand et al. (2003) when climatological temperature, rather than the $\delta^{18}\text{O}_{\text{calcite}} - \delta^{18}\text{O}_{\text{water}}$ temperature, is used. Although the ‘multi-species’ equation of Anand et al. (2003) is plotted here, the same point stands for the *G. ruber* (w) calibrations of Anand et al. (2003) (b) ‘extreme jackknife’ of the dataset; each of the constituent datasets was successively removed from the subset of data and the temperature regression was re-run. The results demonstrate that no individual dataset is significantly biasing the results.

applied calibration. Incorporating salinity into the regression model results in a slightly higher temperature sensitivity of $6.2 \pm 0.5\%/^{\circ}\text{C}$ ($p < 10^{-16}$) and a salinity sensitivity of $3.3 \pm 1.2\%/\text{PSU}$ ($p < 10^{-7}$) (Eq. (S2)). The slight increase in the temperature sensitivity is due to the negative covariance of temperature and salinity within the dataset.

Next, we add the carbonate system parameters into the regression model, assuming it is either pH or $[\text{CO}_3^{2-}]$ (and not both) that influences Mg/Ca. Incorporating pH (seawater scale) into the regression model results in a temperature sensitivity of $5.3 \pm 0.6\%/^{\circ}\text{C}$ ($p < 10^{-16}$), a salinity sensitivity of $4.7 \pm 1.2\%/\text{PSU}$ ($p < 10^{-12}$), and a pH sensitivity of $-15.2 \pm 5.4\%/0.1$ pH units ($p < 10^{-9}$) (Eq. (S3)). The slight change in temperature and salinity sensitivity is due to the covariance of pH with temperature and salinity within the dataset. Substituting $[\text{CO}_3^{2-}]$ for pH results in a temperature sensitivity of $7.5 \pm 0.6\%/^{\circ}\text{C}$ ($p < 10^{-15}$), a salinity sensitivity of $7.8 \pm 1.6\%/\text{PSU}$ ($p < 10^{-16}$), and a $[\text{CO}_3^{2-}]$ sensitivity of $-0.35 \pm 0.12\%/\mu\text{mol kg}^{-1}$ ($p < 10^{-9}$) (Eq. (S4)). Again, the increase in temperature and salinity sensitivities from is due to the covariance of $[\text{CO}_3^{2-}]$ with salinity (and salinity with temperature).

Several key features stand out from these regressions. Firstly, temperature, salinity and pH/ $[\text{CO}_3^{2-}]$ all have a significant influence on Mg/Ca, with temperature exerting the greatest influence. Secondly, the widely-applied calibration of Dekens et al. (2002) and Anand et al. (2003) does not accurately describe the sediment trap/plankton tow data (Fig. 5). Thirdly, the sensitivity of Mg/Ca to salinity is significantly lower than the estimates derived from coretop studies (Ferguson et al., 2008; Arbuszewski et al., 2010; Mathien-Blard and Bassinot, 2009). Finally, the covariance between predictor variables (temperature, salinity, pH, $[\text{CO}_3^{2-}]$) is influencing the results of the regressions. In most instances this influence is relatively minor; however, a more significant difference is seen when $[\text{CO}_3^{2-}]$ is incorporated into the regression model.

In the next three sections, we address the issue of covariance to more accurately constrain the sensitivity of Mg/Ca in *G. ruber* (w) to temperature, salinity, and the carbonate system. To constrain the sensitivity of Mg/Ca to salinity (section 3.4) and temperature (section 3.5), we take subsets of the global dataset where covariance between predictor variables is reduced. We test the derived sensitivities from these subsets of data for the influence of covari-

ance by re-running the regression models, each time including in an additional predictor variable; when the inclusion of additional predictor variables does not change the resulting sensitivity, the influence of covariance is demonstrated to be negligible.

3.4. Sensitivity of Mg/Ca to salinity

To assess the sensitivity of Mg/Ca to salinity we focus on our new data from the Arabian Sea and Bay of Bengal. Regressions of the entire Arabian Sea/Bay of Bengal dataset demonstrate that covariance of predictor variables is influencing the results (SM). To better constrain the sensitivity of Mg/Ca to salinity, we take a subset of the Arabian Sea/Bay of Bengal dataset from a more limited temperature range (28 to 29°C ; Fig. 3) and exclude two samples with pH < 8.00 to reduce the variation in pH ($n = 29$, $\sim 60\%$ of Arabian Sea/Bay of Bengal dataset). We opt for this temperature range as the mean annual temperature at all four sites is $\sim 28.5^{\circ}\text{C}$, thus maximising amount of data within the subset. Within the subset there is very little variation in temperature ($< 0.9^{\circ}\text{C}$) or pH (0.005 units), and a large range in salinity (3.50 PSU), making it ideally suited to assess the Mg/Ca sensitivity to salinity (if pH is the controlling carbonate system parameter). Using this subset of data and regressing Mg/Ca against salinity only results in a salinity sensitivity of $3.3 \pm 2.2\%/\text{PSU}$ ($p < 0.01$) (Fig. 3),

$$\text{Mg/Ca} = 1.75 \pm 1.81 \times \exp(0.033 \pm 0.022 \times S) \quad (1)$$

(RSE = 0.30); incorporating both temperature and pH into the regression model results in an identical salinity sensitivity of $3.3 \pm 2.2\%/\text{PSU}$ ($p < 0.001$), indicating no effect of covariance between predictor variables on the results. This result agrees well with the culture study of Hönisch et al. (2013), which suggested a salinity sensitivity of $3.3 \pm 1.7\%/\text{PSU}$ for *G. ruber*, and is significantly lower than the coretop based estimates of Ferguson et al. (2008), Arbuszewski et al. (2010), and Mathien-Blard and Bassinot (2009).

Due to the strong covariance of salinity and $[\text{CO}_3^{2-}]$ within the Arabian Sea/Bay of Bengal dataset ($r = 0.96$), it is not possible to constrain the effect of salinity if $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter unless some assumption of the sensitivity of

Mg/Ca to $[\text{CO}_3^{2-}]$ is made (see section 3.6); substituting $[\text{CO}_3^{2-}]$ for pH within the regression model results in insignificant terms for both salinity and $[\text{CO}_3^{2-}]$.

3.5. Sensitivity of Mg/Ca to temperature

To constrain the sensitivity of Mg/Ca to temperature, we take a subset of the global dataset with significantly lower variation in salinity (<0.6 PSU) and where the covariance between temperature and salinity ($r = -0.09$), and temperature and pH ($r = -0.34$) is considerably reduced. We opt for this salinity/pH range (36.1 to 37.0 PSU/8.06 to 8.12 pH units) as it gives the greatest number of data points ($n = 215$, $\sim 50\%$ of the global dataset), while ensuring the maximum range in temperature (7.5°C) and the least covariance between predictor variables. The subset of data comprises three previously published datasets (Anand et al., 2003; Haarmann et al., 2011; Babila et al., 2014), and the newly generated data from the tropical Atlantic (Fig. 6).

Regressing Mg/Ca against temperature results in a temperature sensitivity of $6.0 \pm 0.8\%/^\circ\text{C}$ ($p < 10^{-15}$),

$$\text{Mg/Ca} = 1.08 \pm 0.25 \times \exp(0.060 \pm 0.008 \times T) \quad (2)$$

(RSE = 0.45) (Fig. 6). Adding salinity and pH into the regression model results in an identical temperature sensitivity of $6.0 \pm 0.7\%/^\circ\text{C}$ ($p < 10^{-15}$), indicating no influence of covariance between temperature and salinity, and temperature and pH, on the results. Substituting pH for $[\text{CO}_3^{2-}]$ results in a lower sensitivity of $5.4 \pm 0.9\%/^\circ\text{C}$ ($p < 10^{-15}$); however, temperature and $[\text{CO}_3^{2-}]$ covary within the subset of data ($r = 0.67$). Consequently, this subset of data cannot be used to constrain the sensitivity of Mg/Ca to temperature if $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter unless a sensitivity to $[\text{CO}_3^{2-}]$ can be assumed (see section 3.6).

We performed an ‘extreme jackknife’ of the temperature regression to assess whether the resulting temperature sensitivity is being significantly biased by any one of the constituent datasets (SM). This test assesses whether the temperature sensitivity given in Eq. (2) is biased by: (a) the Mg/Ca of a particular dataset being significantly influenced by another factor that is not accounted for in the regression model, or (b) a systematic offset between the habitat depth temperature in the time-period covered by the trap samples at a particular site and the 0–50 m depth climatological temperature at that site; the results demonstrate that no one particular dataset is significantly biasing the results (Fig. 6b).

A key finding of our study is that the temperature sensitivity of $6.0 \pm 0.8\%/^\circ\text{C}$ given in Eq. (2) is significantly lower than the widely-applied $9.0 \pm 0.6\%/^\circ\text{C}$ sensitivity of Dekens et al. (2002) and Anand et al. (2003); it is clear from Fig. 6 that the calibration of Dekens et al. (2002) and Anand et al. (2003) does not accurately describe the dataset. Assuming a 0–25 m habitat depth for this subset results in a lower temperature sensitivity of $5.2 \pm 0.8\%/^\circ\text{C}$ (Eq. (S5)). The residual standard error of this fit is, however, substantially higher than using a habitat depth of 0–50 m. The lower temperature sensitivity in Eq. (2) compared to the $9.0 \pm 0.6\%/^\circ\text{C}$ sensitivity in the calibration of Dekens et al. (2002) and Anand et al. (2003) is therefore not dependent on the assumed habitat depth of *G. ruber* (w). Forcing the temperature sensitivity to a higher value (i.e. $9\%/^\circ\text{C}$) increases the residual standard error of fit (Fig. S7). Our results do not support the relationship between Mg/Ca and temperature suggested by Dekens et al. (2002) and Anand et al. (2003).

Regressing only the data from Anand et al. (2003) against climatological temperature results in a temperature sensitivity of $7.0 \pm 1.2\%/^\circ\text{C}$ (Eq. (S6)). Therefore, the data of Anand et al. (2003) do not support a temperature sensitivity of $9\%/^\circ\text{C}$ when climatological temperature, rather than $\delta^{18}\text{O}_{\text{calcite-water}}$ temperature, is

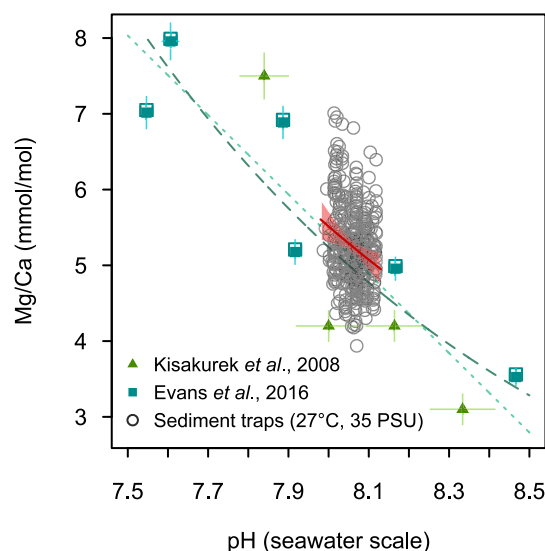


Fig. 7. Mg/Ca versus pH (seawater scale), showing both the sediment trap data from this study (normalised to 27°C and 35 PSU using a temperature sensitivity of $6.0\%/^\circ\text{C}$ and a salinity sensitivity of $3.3\%/ \text{PSU}$) and the data from the culture studies of Kisakurek et al. (2008) and Evans et al. (2016). The data of Evans et al. (2016) were normalised to 27°C and 35 PSU using a temperature sensitivity of $6.0\%/^\circ\text{C}$ and a salinity sensitivity of $3.3\%/ \text{PSU}$, and only data from foraminifera cultured at modern seawater Mg/Ca are included. The uncorrected data are shown by the open squares. The data of Kisakurek et al. (2008) were cultured at $27^\circ\text{C}/35$ PSU. The best fit to the sediment trap data (exponential) is shown by the solid red line, with the 95% confidence interval shown in red. The best fit to the cultured data is shown by the dashed line (exponential, Eq. (S11)), and the dotted line (linear). The pH data of Kisakurek et al. (2008) and Evans et al. (2016) were converted to the seawater scale. Error bars are $\pm 1\sigma$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

used. Note that temperature and pH covary almost perfectly in the dataset of Anand et al. (2003) ($r = -0.98$), which may be the cause of the slightly higher apparent temperature sensitivity in this dataset (see section 3.7 for further discussion on temperature–pH covariance). Anand et al. (2003) pointed out that the $\delta^{18}\text{O}_{\text{calcite-water}}$ temperatures used in their study were $\sim 3^\circ\text{C}$ warmer than the measured CTD temperatures of the upper 1 m of the water column during the winter months of the study period. The authors suggested this may have been due to a sampling bias associated with warm-core eddies. Our compilation demonstrates the data of Anand et al. (2003) are not exceptional.

Now we will revisit the possible cause of this discrepancy by assessing the application of different $\delta^{18}\text{O}_{\text{calcite-water}}$ calibrations, looking at the effects of $[\text{CO}_3^{2-}]$ on $\delta^{18}\text{O}_{\text{calcite}}$ (Spero et al., 1997), and exploring the potential for variability in the salinity– $\delta^{18}\text{O}_{\text{water}}$ relationship (SM). As Fig. S9 shows, the choice of $\delta^{18}\text{O}_{\text{calcite-water}}$ calibration makes little difference to the problem, with the seasonal range predicted in $\delta^{18}\text{O}_{\text{calcite}}$ always $\sim 0.5\%$ greater than the range observed in foraminiferal $\delta^{18}\text{O}_{\text{calcite}}$. Applying the $[\text{CO}_3^{2-}]$ – $\delta^{18}\text{O}$ relationship of Spero et al. (1997), the seasonal variability of $[\text{CO}_3^{2-}]$ in the Sargasso Sea can only explain 10% ($\sim 0.05\%$) of this discrepancy (Fig. S10).

With choice of $\delta^{18}\text{O}_{\text{calcite-water}}$ calibration and carbonate chemistry changes unable to adequately account for the discrepancy, we now assess the potential for a systematic seasonal change in the salinity– $\delta^{18}\text{O}_{\text{water}}$ relationship; this could result in a substantially larger change in $\delta^{18}\text{O}_{\text{water}}$ than suggested by the modest seasonal change in salinity. LeGrande and Schmidt (2006) state that current salinity– $\delta^{18}\text{O}_{\text{water}}$ relationships are only applicable at a regional level, and these relationships may change seasonally. Although no obvious seasonal cycle is apparent in the very limited number of $\delta^{18}\text{O}_{\text{water}}$ measurements from the region, the data do display variability of the magnitude required to explain the

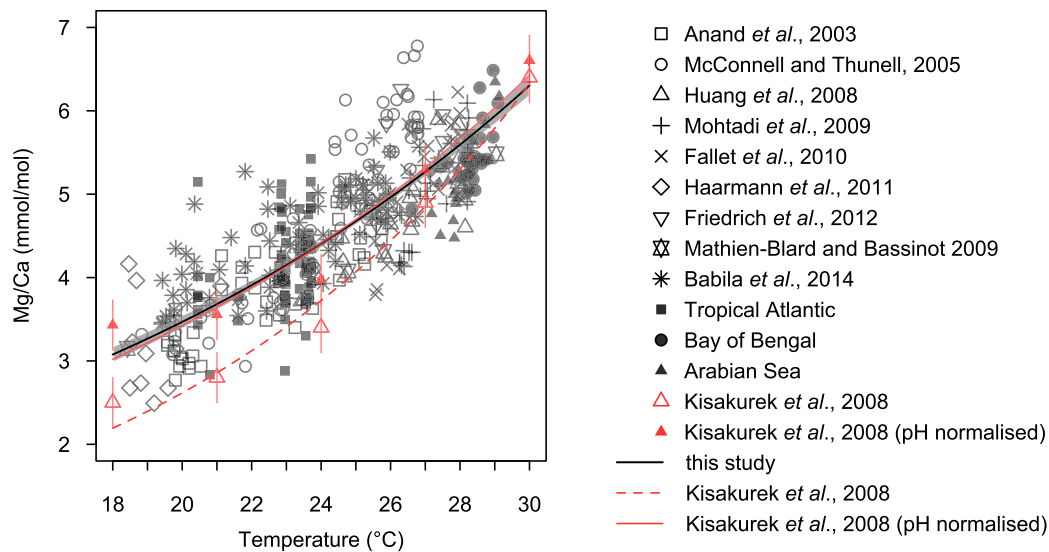


Fig. 8. Mg/Ca versus temperature, showing both the sediment trap data from this study and cultured *G. ruber* (white) data of Kisakürek et al. (2008). The sediment trap data are plotted versus climatological temperature (0–50 m, Methods). The sediment trap data have been normalised to a salinity of 35 PSU and a pH of 8.05 units using a 3.3%/PSU salinity sensitivity (Eq. (1)) and a $-8.3\%/0.1$ pH unit sensitivity (Eq. (3)), respectively. The data of Kisakürek et al. (2008) are shown both uncorrected (open triangles) and corrected (filled triangles) for the effect of pH on Mg/Ca. The calibration line suggested by this study if pH is the controlling parameter (Eq. (3)) is shown by the black line (for a salinity of 35 PSU and a pH of 8.05 units), with the 95% confidence interval shown in grey. The dashed red line indicates the best fit to the culture data without correcting for the effect of pH on Mg/Ca. The solid red line indicates the best fit to the culture data after correction for the effect of pH on Mg/Ca (the pH data of Kisakürek et al. (2008) were first converted to the seawater scale). Error bars are $\pm 1\sigma$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

discrepancy ($\pm 0.7\%$) (Fig. S11). A systematic seasonal change in $\delta^{18}\text{O}_{\text{water}}$ would therefore seem the most likely cause of the discrepancy in $\delta^{18}\text{O}_{\text{calcite}}$, and explain why it is observed in multiple planktic foraminiferal species simultaneously (Anand et al., 2003).

3.6. Sensitivity of Mg/Ca to the carbonate system

The limited range in pH and $[\text{CO}_3^{2-}]$ within the dataset does not allow us to use the same approach used to constrain the sensitivity to salinity and temperature. Instead, to assess the sensitivity of Mg/Ca to pH we perform a multivariate regression using the entire global dataset and prescribe the sensitivity of Mg/Ca to temperature and salinity as $6.0 \pm 0.8\%/^\circ\text{C}$ and $3.3 \pm 2.2\%/PSU$ (see sections 3.5 and 3.6). We account for the uncertainty in temperature and salinity sensitivity with Monte-Carlo simulation. This approach results in a pH sensitivity of $-8.3 \pm 7.3\%/0.1$ pH units ($p < 10^{-5}$) (Fig. 7),

$$\text{Mg/Ca} = \exp(0.060 \pm 0.008 \times T + 0.033 \pm 0.022 \times S - 0.83 \pm 0.73 \times (\text{pH} - 8) - 1.07 \pm 0.80) \quad (3)$$

(RSE = 0.50). The large uncertainty associated with the pH sensitivity in Eq. (3) is primarily due to the very small range in pH within the dataset (~ 0.13 units). This result is in very good agreement with an exponential fit to the *G. ruber* (w) culture experiments of Kisakürek et al. (2008) and Evans et al. (2016), which yields a pH sensitivity of $-9.3 \pm 4.1\%/0.1$ pH units (Eq. (S11)) (Fig. 7). Our estimate of the sensitivity of Mg/Ca to pH is entirely consistent with the results of the culture data of Kisakürek et al. (2008) and Evans et al. (2016).

If we consider that $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter we must take a different approach than that used to estimate the sensitivity to pH. This is because the salinity sensitivity used to account for the effects of salinity in the regression model is derived from the Arabian Sea/Bengal dataset, in which salinity and $[\text{CO}_3^{2-}]$ covary strongly. Furthermore, there is also covariance between $[\text{CO}_3^{2-}]$ and temperature in the subset of data used to

derive the sensitivity to temperature in Eq. (2). Instead we must assume a sensitivity to $[\text{CO}_3^{2-}]$ and use this assumed sensitivity to $[\text{CO}_3^{2-}]$ to calculate a sensitivity to salinity; we can then use the assumed sensitivity to $[\text{CO}_3^{2-}]$ and calculated sensitivity to salinity to derive the sensitivity of Mg/Ca to temperature if $[\text{CO}_3^{2-}]$ is the controlling parameter.

Regressing the data from the culture experiments of Kisakürek et al. (2008) and Evans et al. (2016) against $[\text{CO}_3^{2-}]$ results in a sensitivity of $-0.24 \pm 0.12\%/ \mu\text{mol kg}^{-1}$ (Eq. (S12); Fig. S12), within error of the $-0.35 \pm 0.12\%/ \mu\text{mol kg}^{-1}$ suggested by multivariate regression of the global dataset (Eq. (S4)). Taking a subset of data from the Arabian Sea/Bay of Bengal dataset from the same temperature range used to calculate the sensitivity to salinity in Eq. (1) (28–29 $^\circ\text{C}$), and regressing the data against salinity with a prescribed sensitivity to $[\text{CO}_3^{2-}]$ of $-0.24 \pm 0.12\%/ \mu\text{mol kg}^{-1}$ results in a salinity sensitivity of $5.0 \pm 3.0\%/PSU$ ($p < 0.001$). Incorporating temperature within the regression model results in a very similar salinity sensitivity of $5.1 \pm 2.8\%/PSU$ ($p < 10^{-5}$). Prescribing the salinity sensitivity of $5.0 \pm 3.0\%/PSU$, and the $[\text{CO}_3^{2-}]$ sensitivity of $-0.24 \pm 0.12\%/ \mu\text{mol kg}^{-1}$ in a multivariate regression of the entire dataset results in a temperature sensitivity of $6.7 \pm 0.8\%/^\circ\text{C}$ ($p < 10^{-15}$),

$$\text{Mg/Ca} = \exp(0.067 \pm 0.008 \times T + 0.050 \pm 0.030 \times S - 0.0024 \pm 0.0012 \times [\text{CO}_3^{2-}] - 1.37 \pm 1.31) \quad (4)$$

(RSE = 0.50), similar to the temperature sensitivity if pH is the controlling parameter (Eq. (3)). Regressing the subset of data with very little variation in salinity (< 0.6 PSU) used to calculate the temperature sensitivity in section 3.5 against temperature with a prescribed sensitivity to $[\text{CO}_3^{2-}]$ of $-0.24 \pm 0.12\%/ \mu\text{mol kg}^{-1}$ results in an identical (within error) temperature sensitivity of $7.0 \pm 0.9\%/^\circ\text{C}$. This confirms the temperature sensitivity given in Eq. (4), without necessitating an assumption of the sensitivity to salinity. Application of a lower $[\text{CO}_3^{2-}]$ sensitivity, as suggested for *G. ruber* (pink) (Allen et al., 2016), has very little effect on the resulting temperature and salinity sensitivities (SM).

3.7. Reconciling sediment trap and culture temperature sensitivity estimates

The temperature sensitivities indicated by the sediment trap data, i.e. $6.0 \pm 0.8\%/^{\circ}\text{C}$ if pH is the controlling parameter, and $6.7 \pm 0.8\%/^{\circ}\text{C}$ if $[\text{CO}_3^{2-}]$ is the controlling parameter, are lower than the temperature sensitivity of $8 \pm 3\%/^{\circ}\text{C}$ inferred from the culturing experiment of Kisakürek et al. (2008) (Fig. 8). One way by which the sediment trap and culture data can be reconciled is if the influence of temperature on the dissociation constant of water ($K_w = [\text{H}^+][\text{OH}^-]$) is considered. As K_w changes as a function of temperature, the pH of water decreases with increasing temperature by ~ -0.015 pH units/ $^{\circ}\text{C}$, without changing the ratio of Alk/DIC (Millero, 1979). Hence, in the culture experiments of Kisakürek et al. (2008) temperature and pH covary perfectly ($r > 0.99$), with a 0.18 pH unit decrease from coldest to warmest sample. If the culture data of Kisakürek et al. (2008) are normalised to a pH of 8.05 (the mean value within the sediment trap dataset) to remove the internal shift of pH due to temperature, using a pH sensitivity of $-8.3\%/0.1$ pH units derived from the sediment trap dataset (Eq. (3)), the Kisakürek et al. (2008) data suggest a lower temperature sensitivity of $6.3 \pm 1.9\%/^{\circ}\text{C}$ ($p < 0.01$) (Fig. 8),

$$\text{Mg/Ca} = 0.97 \pm 0.50 \times \exp(0.063 \pm 0.019 \times T) \quad (5)$$

(RSE = 0.36; $n = 5$), in excellent agreement with the temperature sensitivity indicated by the sediment trap dataset if pH is the controlling carbonate system parameter (Eq. (3); Fig. 8). Using the pH sensitivity of $-9.3 \pm 4.6\%/0.1$ pH units suggested by the pH experiments to normalise the culture data instead, results in an identical (within error) temperature sensitivity of $6.2 \pm 1.9\%/^{\circ}\text{C}$ ($p < 0.01$). Accounting for the influence of temperature on the dissociation constant of water (K_w) within the culturing experiments thus allows the culture and sediment trap datasets to be reconciled if pH is the controlling carbonate system parameter. If $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter, the sediment trap and culture data cannot be reconciled as the ratio of Alk/DIC (and thus $[\text{CO}_3^{2-}]$) is not changing in the temperature experiment of Kisakürek et al. (2008). Based on our current understanding of foraminiferal calcification mechanisms (e.g. Erez, 2003; de Nooijer et al., 2009), Evans et al. (2016) argued that pH is the carbonate system parameter that influences foraminiferal Mg/Ca (SM), and recent work has revealed foraminifera actively pump protons during calcification (Toyofuku et al., 2017). Our results provisionally support this inference, as this would allow the sediment trap and culture data to be reconciled.

3.8. Proposed calibrations

We propose the following calibration for the conversion of *G. ruber* (w) Mg/Ca to temperature if pH is the controlling carbonate system parameter,

$$\text{Mg/Ca} = \exp(0.060 \pm 0.008 \times T + 0.033 \pm 0.022 \times S - 0.83 \pm 0.73 \times (\text{pH} - 8) - 1.07 \pm 0.80) \quad (3)$$

(RSE = 0.50), and the following calibration if $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter,

$$\text{Mg/Ca} = \exp(0.067 \pm 0.008 \times T + 0.050 \pm 0.030 \times S - 0.0024 \pm 0.0012 \times [\text{CO}_3^{2-}] - 1.37 \pm 1.31) \quad (4)$$

(RSE = 0.50). As discussed above, we suggest that pH is most likely to be the controlling carbonate system parameter as this allows the reconciliation of the culture experiment and sediment trap datasets, but further culturing work is needed to test this assertion.

3.9. Sources of noise within the calibration

Significant noise can be seen in the sediment trap Mg/Ca dataset (Fig. 8), with $\sim 30\%$ of the variance unaccounted for by temperature, salinity and the carbonate system. Here we assess how much of this noise relates to (i) the use of climatological temperature and (ii) different size fractions within the dataset.

To assess how much noise may be explained by the use of climatological temperatures we simulate synthetic foraminiferal samples using both CTD temperature and WOA13 temperature for the same period of time, and examine the difference expected in Mg/Ca between these samples (SM). We utilise data from the Sargasso Sea, which is the only trap site with continuous CTD data for the time interval covered by the sediment samples, and simulate samples broadly representative of the actual samples analysed by Anand et al. (2003) and Babila et al. (2014). We then calculate the residual Mg/Ca based on the difference in temperature between the WOA-synthetic foraminiferal samples and the CTD-synthetic foraminiferal samples and compare this to the residual of the measured Mg/Ca data from the Sargasso Sea sediment trap site. The results show that the residual in measured Mg/Ca data is significantly higher than expected based on the uncertainty introduced by using climatological temperature alone ($p < 10^{-7}$) (Fig. S15); however, $\sim 40\%$ of noise can be explained by the use of climatological temperature. If the Sargasso Sea dataset is representative of the wider trap/tow dataset, substantial improvements in the precision of the calibration could be made if in-situ temperatures were collected along with foraminiferal samples.

Another source of noise may relate to ‘biological’ variability. One aspect of biological variability that might be constrained is the influence of size fractions within the global dataset. The mean size of foraminiferal sample within the dataset is 300 μm , and 95% of samples fall into the 200 to 400 μm range of mean test size. To test if it is possible to systematically correct for variations in test size, we use the relationship between test size and Mg/Ca given in Fig. 4e ($0.35 \text{ mmol mol}^{-1}/100 \mu\text{m}$) to normalise all sediment trap and plankton tow data within the dataset to the same size (300 μm). Regressing the size normalised Mg/Ca against temperature, salinity and $\text{pH}/[\text{CO}_3^{2-}]$ increases the residual standard error of fit compared to regressions of the uncorrected data. Incorporating test size as a predictor variable within the regression model returns an insignificant coefficient for test size. Thus, while test size variability might be causing much of the noise, such effects cannot easily be corrected.

4. Implications for tropical SST during the LGM

We apply our new multivariate calibration models to previously published *G. ruber* (w) Mg/Ca data from multiple core sites located in the tropical Pacific, Indian and Atlantic Oceans covering the Last Glacial Maximum (LGM) (SM). Applying the calibration given in Eq. (3), which assumes pH is the controlling carbonate system parameter, results in a tropical ocean cooling of $3.5 \pm 1.6^{\circ}\text{C}$ (2σ , accounting for Mg/Ca variability only) if no change in salinity or pH is accounted for (Fig. 9). If the 1.15 PSU whole ocean increase in salinity during the LGM (Adkins et al., 2002) is included, the magnitude of cooling increases to $4.2 \pm 1.6^{\circ}\text{C}$. However, if a 0.13 unit surface ocean pH increase (SM) is also accounted for, the magnitude of cooling is reduced to $2.4 \pm 1.6^{\circ}\text{C}$. This calculation demonstrates that the combined pH and salinity influence in Eq. (3) reduces the inferred temperature change by about a third. We note that while this value is similar to the value ($2.3 \pm 1.3^{\circ}\text{C}$) obtained using the calibration of Dekens et al. (2002) and Anand et al. (2003) (Fig. 9), this similarity is not a confirmation of the calibration of Dekens et al. (2002) and Anand et al. (2003), which has

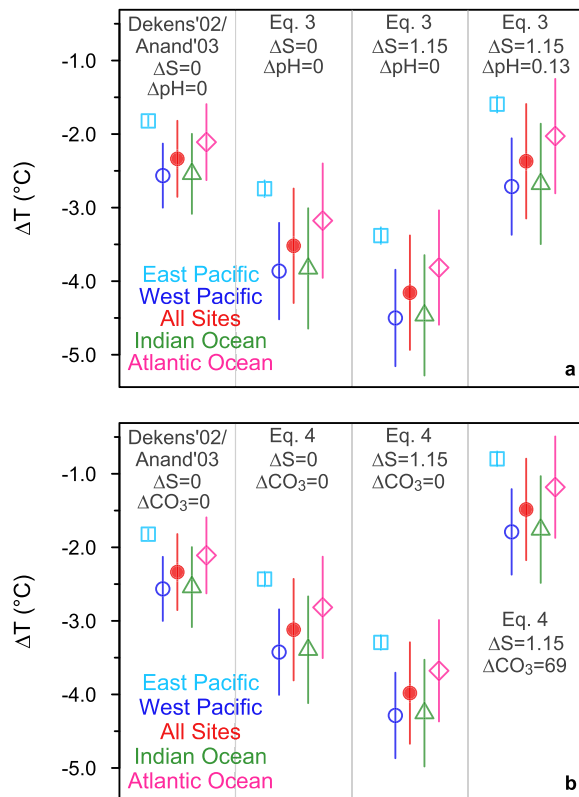


Fig. 9. Magnitude of LGM cooling in the tropical ocean using the calibration of Dekens et al. (2002) and Anand et al. (2003) and the calibrations given in this study if (a) pH is the controlling carbonate system parameter (Eq. (3)) or, (b) $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter (Eq. (4)). The magnitude of cooling is shown with and without accounting for the whole ocean increase in salinity (1.15 units) and pH (0.13 units)/ $[\text{CO}_3^{2-}]$ (69 $\mu\text{mol/kg}$) (supplementary material). Error bars ($\pm 1\sigma$) relate only to the standard deviation of the $\Delta\text{Mg/Ca}$ within each region. The true uncertainty of the temperature change is dependent on both the uncertainty of the sensitivities of Mg/Ca to T, S and pH/ $[\text{CO}_3^{2-}]$ within the calibration, and the uncertainty in regional changes in salinity, pH/ $[\text{CO}_3^{2-}]$, and dissolution during the LGM. Currently, we have very little constraint on regional changes in salinity and pH/ $[\text{CO}_3^{2-}]$ during the LGM, thus the uncertainty associated with these changes is essentially unquantifiable at present.

a higher temperature sensitivity and does not account for changes in salinity or carbonate chemistry.

Applying the calibration given in Eq. (4), which assumes $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter, results in a cooling of $3.1 \pm 1.4^\circ\text{C}$ (2σ) if no change in salinity or $[\text{CO}_3^{2-}]$ is considered (Fig. 9). If the 1.15 PSU whole ocean increase in salinity is accounted for, the magnitude of cooling during the LGM increases to $4.0 \pm 1.4^\circ\text{C}$. However, if a 69 $\mu\text{mol/kg}$ surface ocean $[\text{CO}_3^{2-}]$ increase is also included (SM), the magnitude of cooling is reduced to $1.5 \pm 1.4^\circ\text{C}$. The substantial effect of including the carbonate ion change on the temperature estimate reflects the strong influence of carbonate ion in Eq. (4), where it accounts for a significantly greater weighting than temperature for LGM conditions.

These scenarios are not intended as a thorough estimation of LGM temperature change, but instead serve only to highlight that salinity and carbonate chemistry have a substantial effect on reconstructed temperature, and the necessity of obtaining regional salinity and pH/ $[\text{CO}_3^{2-}]$ estimates for robust SST reconstruction using Mg/Ca. In addition to the secondary influences on Mg uptake into foraminiferal calcite, the effects of dissolution (usually expressed as a function of bottom-water carbonate ion saturation, ΔCO_3^{2-}) on Mg/Ca must also be accounted for in temperature reconstructions from fossil foraminifera (Regenberg et al., 2014). Uncertainty in reconstructed temperature change from Mg/Ca will therefore be dependent on the uncertainty of the sensitivities of

Mg/Ca to temperature, salinity and the carbonate system within the calibration, the uncertainty in past changes in salinity and pH/ $[\text{CO}_3^{2-}]$, as well as both the uncertainty in the relationship between Mg/Ca and dissolution, and the uncertainty in past changes in bottom- (or pore-) water carbonate chemistry.

5. Conclusions

This study utilises 440 sediment trap/plankton tow samples from 20 sites in Atlantic, Pacific and Indian basins to provide the most comprehensive field based calibration for Mg/Ca in *G. ruber* (white) to date. The temperature sensitivity within our calibration of 6.0% per $^\circ\text{C}$ (assuming pH is the controlling carbonate system parameter) or 6.7% per $^\circ\text{C}$ (assuming $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter) is significantly lower than the widely-applied temperature sensitivity of 9% per $^\circ\text{C}$. The significant effects of salinity (with a sensitivity of 3.3% or 5% per PSU, depending on the controlling carbonate system parameter) and carbonate chemistry (with a sensitivity of -8.3% per 0.1 pH units or -0.24% per $\mu\text{mol/kg}$ $[\text{CO}_3^{2-}]$) on Mg/Ca in *G. ruber* (white) complicate the use of Mg/Ca as a paleothermometer.

Applying our calibration model which assumes pH is the controlling carbonate system parameter (we provisionally suggest pH is most likely to be the controlling parameter), without considering changes in salinity and carbonate chemistry, results in a cooling of $\sim 3.5^\circ\text{C}$ during the LGM; if whole ocean changes in salinity and carbonate chemistry are accounted for the cooling is reduced to $\sim 2.3^\circ\text{C}$. Applying our calibration model which assumes $[\text{CO}_3^{2-}]$ is the controlling carbonate system parameter, without considering changes in salinity and carbonate chemistry, results in a cooling of $\sim 3.1^\circ\text{C}$ during the LGM; if whole ocean changes in salinity and carbonate chemistry are accounted for the cooling is reduced to $\sim 1.5^\circ\text{C}$. The substantial influence of salinity and carbonate chemistry on Mg/Ca necessitates independent estimates of salinity and carbonate chemistry for reliable temperature reconstruction. While boron isotopes may offer a viable tool to account for changes in carbonate chemistry (Henehan et al., 2013), the lack of a quantitative salinity proxy currently adds a currently-unquantifiable uncertainty into Mg/Ca-based temperature estimates.

Future efforts to further improve the Mg/Ca paleothermometer should include (i) culture experiments to assess whether it is pH or $[\text{CO}_3^{2-}]$ that controls Mg/Ca, and determine the exact form and sensitivity of this relationship (ii) studies to ascertain if the secondary influences on Mg/Ca in *G. ruber* (white) are as pronounced in other planktic foraminiferal species and, (iii) the development and refinement of an independent salinity proxy.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.epsl.2017.11.026>. Data are also available in the NOAA National Climatic Data Center (<https://www.ncdc.noaa.gov/paleo/study/23110>).

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