Ecological and temperature controls on Mg/Ca ratios of *Globigerina bulloides* from the southwest Pacific Ocean

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[1]   We present Mg/Ca data for *Globigerina bulloides* from 10 core top sites in the southwest Pacific Ocean analyzed by laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS). Mg/Ca values in *G. bulloides* correlate with observed ocean temperatures (7°C–19°C), and when combined with previously published data, an integrated Mg/Ca–temperature calibration for 7°C–31°C is derived where Mg/Ca (mmol/mol) = $0.955 \times e^{0.068 \times T}$ ($r^2 = 0.95$). Significant variability of Mg/Ca values (20%–30%) was found for the four visible chambers of *G. bulloides*, with the final chamber consistently recording the lowest Mg/Ca and is interpreted, in part, to reflect changes in the depth habitat with ontogeny. Incipient and variable dissolution of the thin and fragile final chamber, and outermost layer concomitantly added to all chambers, caused by different cleaning techniques prior to solution-based ICPMS analyses, may explain the minor differences in previously published Mg/Ca–temperature calibrations for this species. If the lower Mg/Ca of the final chamber reflects changes in depth habitat, then LA-ICPMS of the penultimate (or older) chambers will most sensitively record past changes in near-surface ocean temperatures. Mean size-normalized *G. bulloides* test weights correlate negatively with ocean temperature (T = $31.8 \times e^{-30.5 \times wtN}$; $r^2 = 0.90$), suggesting that in the southwest Pacific Ocean, temperature is a prominent control on shell weight in addition to carbonate ion levels.

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1.   Introduction

[2]   Trace elements in foraminiferal calcite are increasingly being used to reconstruct the thermal and chemical state of Earth's oceans (and atmosphere) during past climate fluctuations [e.g., *Nürnberg et al.*, 1996; *Lea et al.*, 1999, 2000; *Elderfield and Ganssen*, 2000; *Billups and Schrag*, 2003; *Elderfield et al.*, 2006; *Rosenthal*, 2007; *Tripati et al.*, 2009]. Geochemical studies of foraminifera have traditionally utilized bulk analytical methods in which up to ~100 foraminifera are crushed and dissolved [e.g., *Rosenthal et al.*, 1997; *Lea et al.*, 1999, 2000; *Elderfield and Ganssen*, 2000; *Barker et al.*, 2003]. Significant advances in microanalytical techniques, such as laser ablation induc-

tively coupled plasma mass spectrometry (LA-ICPMS) and secondary ionization mass spectrometry, now make it possible for the individual chambers of a foraminifera to be analyzed at a spatial resolution of tens of microns yielding in situ data for a range of trace elements and isotopes [*Eggins et al.*, 2003, 2004; *Bice et al.*, 2005; *Rollion-Bard et al.*, 2008; *Sadekov et al.*, 2008, 2009; *Kasemann et al.*, 2008, 2009].

[3]   Studies using LA-ICPMS and electron microprobe techniques have revealed the existence of significant trace element heterogeneity (up to 50%) between different chambers of an individual foraminifer [*Anand and Elderfield*, 2005; *Sadekov et al.*, 2008] and also from individual-to-individual in the same sample. Variable thicknesses and numbers of bands of calcite with high and low Mg/Ca have been identified to explain differences between individual chambers, whereas repeated analysis within the same chambers has generally demonstrated good reproducibility [e.g., *Eggins et al.*, 2003, 2004; *Sadekov et al.*, 2005, 2008]. The complexities of trace element incorporation in foraminiferal calcite revealed by in situ techniques provide a significant challenge to the use of this method as a robust proxy for past ocean temperatures.

[4]   Here we present LA-ICPMS trace element analyses of *Globigerina bulloides* (Figure 1) from the southwest Pacific

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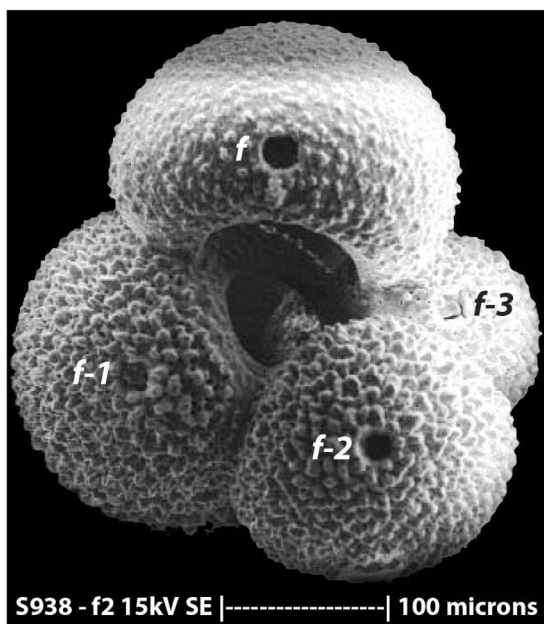
Figure 1. Scanning electron microprobe image of *Globigerina bulloides* (site S938; Figure 2). Each chamber is labeled, where *f* represents the youngest chamber with each subsequently older chamber labeled *f*-1 (penultimate chamber), *f*-2 (antepenultimate chamber), and *f*-3. Foraminiferal test sizes are taken from the measurement of the longest axis between chambers *f* and *f*-2. Laser ablation pits measuring 25 μm in diameter are visible.

Ocean, east and south of New Zealand, in order to evaluate the ecological, temperature and oceanographic controls on the incorporation of Mg into this species of planktic foraminifera. *G. bulloides* is a spinose, nonsymbiont species that inhabits a range of water temperatures found in tropical to subantarctic waters with highest abundances associated with eutrophic conditions [*Hemleben et al.*, 1989; *Sen Gupta*, 1999; *Crundwell et al.*, 2008]. Foraminifera were sampled from 10 core tops between latitudes 33.86°S and 53.63°S within two major surface water masses: subtropical and subantarctic waters (Figure 2 and Table 1). Geochemical data are complemented by detailed weight and size measurements of all individual foraminiferal tests analyzed.

[5] This data set is used to: (1) Develop LA-ICPMS–based Mg/Ca–temperature calibrations for *G. bulloides* suitable for near-surface water reconstructions in the southwest Pacific Ocean. This is important as LA-ICPMS based calibrations are not necessarily compatible with solution-ICPMS–based calibrations where chemical cleaning of samples prior to solution-ICPMS analysis differs from the protocols used in LA-ICPMS analysis to remove contaminants from the foraminiferal tests. (2) Evaluate any ecological controls on foraminiferal trace element chemistry and potential implications for paleoenvironmental reconstructions.

2. Environmental Setting

[6] The surface waters off eastern New Zealand feature markedly different water masses whose introduction to the region is dominated by the Subtropical Gyre from the north and the Antarctic Circumpolar Current from the south (Figure 2). The Subtropical Front (STF) is situated approximately along the east-west crest of Chatham Rise and separates warm, saline, macronutrient-poor, micronutrient-rich Subtropical Water (STW) in the north from cold, less saline, macronutrient-rich, micronutrient-poor Subantarctic Water (SAW) to the south [*Heath*, 1985; *Carter et al.*, 1998; *Sutton and Roemmich*, 2001].

[7] Knowledge of general foraminiferal habitats and life cycles of individual species is required to test or verify interpretations of foraminifera test chemistry, for example, depth habitat in the water column and timing of reproduction. Foraminiferal depth habitats can seldom be defined precisely because they undergo diurnal, ontogenetic, seasonal and long-term variations [*Hemleben et al.*, 1989; *Arnold and Parker*, 1999]. Superimposed on depth distribution patterns is the effect of water mass mixing, which is locally significant in water masses around New Zealand [e.g., *Weaver et al.*, 1997; *Murphy et al.*, 2001].

[8] In this study, the water column depth for *G. bulloides* is taken to be ~125 m for all sites except P69 and S938. This is based on surface mixed layer depths, peak fluorescence depths and plankton tow information where ~85% of *G. bulloides* were located at depths of 100–150 m near Campbell Plateau (NIWA, Cruise Report TAN 9814, unpublished report, 1998) [*Sutton and Roemmich*, 2001; *Mortyn and Charles*, 2003; *Condie and Dunn*, 2006; *Wilke et al.*, 2009]. *G. bulloides* numbers in a plankton tow were sparse for sites P69 and S938, but most (~70%) occurred at a depth of 50–100 m (L. Northcote, personal communication, 2008). This shallower depth for *G. bulloides* is consistent with the impact of the local warm core Wairarapa Eddy on regional oceanography (Figure 2) [*Roemmich and Sutton*, 1998; *Tilburg et al.*, 2001; *Chiswell*, 2005]. The movement of the Wairarapa Eddy results in nutrient entrainment and mixing; a feature supported by the shallow fluorescence peak depth [*Weaver et al.*, 1997; *Murphy et al.*, 2001; L. Carter, Voyage report East Coast Margin past and present TAN 0005, unpublished NIWA report, 2000]. Therefore, for sites P69 and S938 we use observed ocean temperatures from 75 m depth for *G. bulloides*.

[9] Many of the core top sites are dominated by seasonal productivity cycles [*King and Howard*, 2001; *Murphy et al.*, 2001; *Northcote and Neil*, 2005]. Thus, foraminiferal abundance from a single core top is likely to be dominated by the season or seasons that have peak productivity. However, as the seasonal variability of water column temperature is significantly reduced with depth, this effect is not particularly significant for *G. bulloides* in this study. For example, offshore of New Zealand the difference between summer and winter sea surface temperatures can vary by up to 5°C but below 75 m the seasonal difference is generally <1.5°C (e.g., data of *Sutton and Roemmich* [2001], *Northcote and Neil* [2005], and *Locarnini et al.* [2006]).

3. Materials and Methods

[10] *G. bulloides* was recovered from sediment (300–355 μm sieved fraction) at 10 core top sites situated within water masses of the southwest Pacific (Table 1). Sea surface temperatures at depth (SST) during the summer months range from 21°C at the northernmost site (P71) to 8.8°C
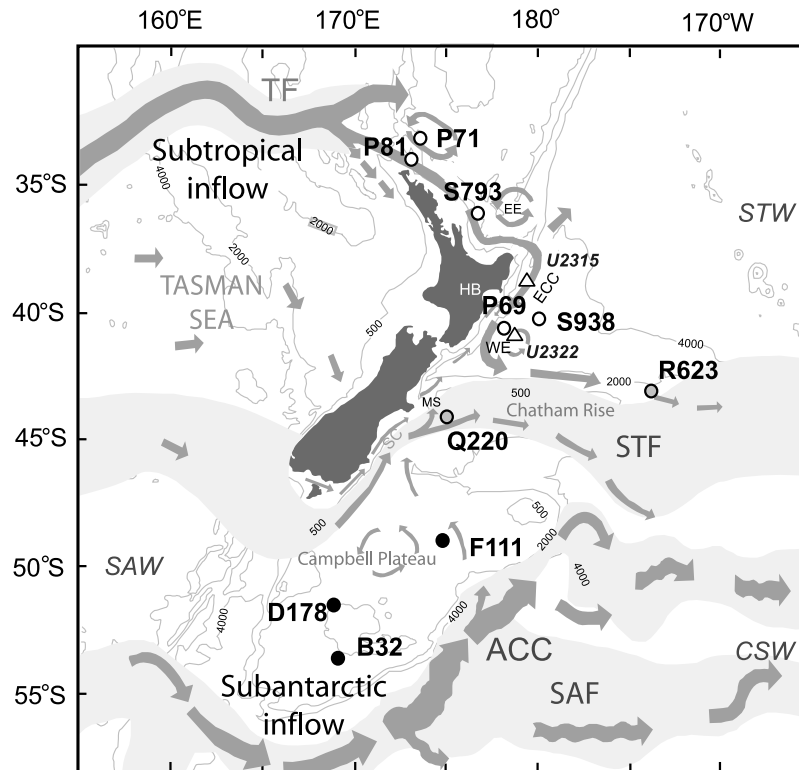
Figure 2. Generalized modern surface ocean currents and bathymetric features surrounding New Zealand (modified from *Carter et al.* [1998]). Circles mark the core top sites: white are subtropical, gray are mixed, and black are subantarctic sites. Triangles mark stratified plankton tow sites (Northcote, personal communication, 2008). TF is Tasman Front, STF is Subtropical Front, SAF is Subantarctic Front, SC is Southland Current, ECC is East Cape Current, ACC is Antarctic Circumpolar Current, STW is Subtropical Water, SAW is Subantarctic Water, CSW is Circumpolar Surface Water, MS is Mernoo Saddle, WE is Wairarapa Eddy, EE is East Cape Eddy, and HB is Hawke's Bay. Isobaths are in meters.

at the southernmost site (B32). Water depths of the core top samples range from 580 to 3003 m, with the deepest sites being close to the calcite saturation zone, which east of New Zealand lies between 2800 and 3200 m depth [*Feely et al.*, 2002, 2004; *Regenberg et al.*, 2006]. Foraminifera from these deeper sites (>2000 m) were accordingly examined and discarded if any sign of dissolution was identified.

[11] In order to obtain the necessary temperature gradient for calibration of Mg/Ca verse SST a suite of 10 core top samples from the southwest Pacific Ocean was selected. These samples span a SST range of ~7°C–19°C. The use of core top samples as an approximation of "modern" conditions is well established in the paleoceanographic literature [e.g., *CLIMAP Project Members*, 1976; *Bé et al.*, 1977; *Elderfield and Ganssen*, 2000; *Cléroux et al.*, 2008; *Sadekov et al.*, 2009; *Regenberg et al.*, 2009]. In part, core tops are used out of necessity; there are few long-term sediment trap and plankton tow samples from the southwest Pacific Ocean with (1) sufficient sample sizes of modern foraminifera and (2) suitable preservation media that allow trace elements to be analyzed [e.g., *Northcote and Neil*, 2005].

[12] Core top samples represent "average" conditions over a period of time determined by sedimentation and bioturbation rates and may vary from site to site. Holocene sedimentation rates have been calculated from (1) published core stratigraphies (Q220 [*Griggs et al.*, 1983], P69 [*Carter*

et al., 2000], and S938 [*Carter et al.*, 1995]), (2) unpublished lithological and isotope data (P71, P81) (G. Dunbar, 2010), and (3) estimated from nearby cores where the Holocene sedimentation rate is regionally uniform (F111 [*Carter et al.*, 2000], S749 [*Pillans and Wright*, 1992], and D178 [*Neil et al.*, 2004]). The implication of variable sedimentation and bioturbation rates is that the effect of past temperature changes will be incorporated into "core top" samples, potentially providing bias to the relationship between Mg/Ca and SST since the latter is determined from modern CTD and satellite measurements. However, these are not major factors for our samples as the lowest calculated sedimentation rate for a core top in this study is 0.9 cm/kyr (P81, P71) and it has been estimated that up to the upper 10 cm of surface sediment may be affected by bioturbational mixing [*Boyle*, 1995]. This would imply that there is potential for this sample suite to contain foraminifera up to 10 kyr in age. However, from the five radiocarbon ages obtained for these core tops, the maximum radiocarbon age is 6.22 kyr (Table 1). Over the last 6 kyr we assume that seawater temperature did not vary by more than 3°C based on previous studies in the region including alkenones (0.5°C–2.8°C variation [*Pahnke et al.*, 2003; *Pahnke and Sachs*, 2006]), foraminifera assemblage data (<1°C variation [*Barrows et al.*, 2007; *Crundwell*

Table 1. Details of Core Top Locations, Sedimentation Rates, Radiocarbon Ages, and Relevant Ocean Temperatures for the Sites Examined in This Study[a]

Core Top	Water Depth (m)	Latitude (°S)	Longitude (°E)	Water Mass[b]	Corer	Salinity (psu)	Sedimentation Rate (cm/kyr)	Radiocarbon Age (years)	Depth of Temperature (m)	Temperature (°C)
P71	1919	33.86	174.69	STW	Piston	35.64	0.9	5715 ± 40	125	18.1[c]
P81	2036	34.02	173.51	STW	Kullenberg Piston	35.47	0.9		125	17.1[c]
S793	2411	36.32	176.80	STW	Kasten	35.42	7.1		125	15.8[d]
P69	2195	40.40	178.00	STW	Piston	35.57	13.6		75	19.1[e]
S938	3003	40.03	180.00	STW	Kasten	35.53	8.3	570 ± 35	75	18.8[e]
R623	1128	43.20	186.00	SAW/STW	Kasten	35.04		6220 ± 35	125	12.2[d]
Q220	580	44.29	174.98	SAW/STW	Piston	34.55	2.8		125	9.7[f]
F111	704	48.95	174.98	SAW	Dredge	34.45	1.0	4582 ± 36	125	7.3[g]
D178	629	51.72	169.83	SAW	Dredge	34.40	1.7	4835 ± 33	125	7.4[g]
B32	799	53.63	169.87	SAW	Grab	34.40			125	7.8[h]

[a]Observed temperature and salinity values were used where available.
[b]STW is Subtropical Water, and SAW is Subantarctic Water.
[c]*Stanton* [1973].
[d]*Locarnini et al.* [2006].
[e]Carter (unpublished report, 2000).
[f]*Heath* [1975].
[g]*Neil et al.* [2004].
[h]*Morris et al.* [2001].

et al., 2008]) and Mg/Ca records (<2°C variation [*Barrows et al.*, 2007]).

[13] The trace element variability of individual foraminifera may also be a potential issue with core top samples, possibly resulting from effects of bioturbation. Variability in geochemical values has been observed by *Boyle* [1995] for Cd/Ca, individual oxygen isotopes [*Koutavas et al.*, 2006] and Mg/Ca [*Sadekov et al.*, 2008; *Hathorne et al.*, 2009]. This individual-to-individual heterogeneity within a single sample is normally homogenized by the use of bulk sampling techniques, but clearly persists after sediment trap and plankton tow sampling [*Sadekov et al.*, 2008] and therefore is an issue which extends beyond the use of core top material.

[14] Despite the aforementioned limitations, in order to achieve the spatial coverage necessary to produce a robust calibration core top material must be used. However, calibrations based on core tops that have undergone bioturbation may have an advantage of smoothing smaller-scale climatic oscillations, including the effects of transient events and therefore offer a more representative reflection of average conditions. Core tops therefore have value as they mimic the cored sedimentary record to which the Mg/Ca–temperature calibrations developed in this study are to be applied [e.g., *Lea*, 2003; *Barker et al.*, 2005].

[15] Prior to weighing and LA-ICPMS analysis, all foraminiferal tests were cleaned to remove loosely adhering clay and other detrital material by 2–3 ultrasonication rinses for 2–3 s in >18.2 MΩ ultraclean water and two AR-grade methanol rinses. Samples were subsequently examined under a binocular microscope and any individuals with visible adhering contaminants were treated to a further one or two ultrasonic cycles. Ultrasonic cleaning for longer than 3 s often fragmented foraminiferal tests, particularly for foraminifera from the more northerly (and warmer) sites. The foraminifera were then dried in an oven at 40°C for 24 h. All *G. bulloides* tests were then individually weighed using a Mettler Toledo (MX/UMX 2) ultra-microbalance. Analytical uncertainty on these weight measurements are <±5% (2 sd relative) based upon 15 replicate measurements of a single *G. bulloides* test weighing 8.7 μg.

[16] Trace element/Ca ratios of foraminifera tests were measured using a New Wave deep-UV (193 nm) solid-state laser ablation system coupled to an Agilent 7500CS ICP-MS at Victoria University of Wellington, New Zealand (see Table S1 in the auxiliary material for analytical conditions).[1] Laser ablation analyses were 120 or 180 s long, which consistently penetrated the *G. bulloides* test wall. Preablation cleaning of the outer test surface was not employed in this study, but each profile was screened for contaminants in the outer part of the test. After subtraction of background signals, trace element/Ca ratios of foraminifera were calculated by reference to ratios obtained from bracketing analyses of NIST610, and normalization to the known trace element values in this standard: Mg = 465, Al = 10798, Ca = 82191, Mn = 485, Zn = 456, Sr = 516, Ba = 435 ppm [*Pearce et al.*, 1997]. NIST610 was analyzed prior to and after every 5–20 analyses of foraminifera and showed no significant drift in trace element/Ca ratios beyond the internal precision of the analyses. The internal precisions (% 2 se relative) of trace element/Ca ratios for a single foraminiferal analysis are typically: Mg/Ca ± 1.0 to 2.5%, Al/Ca ± 5 to 20%, Mn/Ca ± 3 to 15%, Zn/Ca ± 6 to 23%, Sr/Ca ± 1.0 to 2.0%, Ba/Ca ± 3 to 30%. Multiple analyses (n = 3 to 5) of single chambers of an individual foraminifera show that the external reproducibility (2 sd) of trace element/Ca ratios for a single foraminifera analysis are less than: Mg/Ca ± 3.0 to 12% (±0.10 to 0.26 mmol/mol), Al/Ca ± 45 to 106% (±0.17 to 0.69 mmol/mol), Mn/Ca ± 15 to 60% (±0.009 to 0.033 mmol/mol), Zn/Ca ± 14 to 52% (±0.004 to 0.019 mmol/mol), Sr/Ca ± 3.3 to 7.6% (±0.04 to 0.08 mmol/mol), Ba/Ca ± 27 to 157% (±0.002 to 0.014 mmol/mol). However, some of this variability represents real heterogeneity within the foraminiferal test chambers and overestimates the true external reproducibility. In this paper only Mg/Ca and Sr/Ca ratios will be discussed. The significance of other trace element ratios measured are currently under investigation.
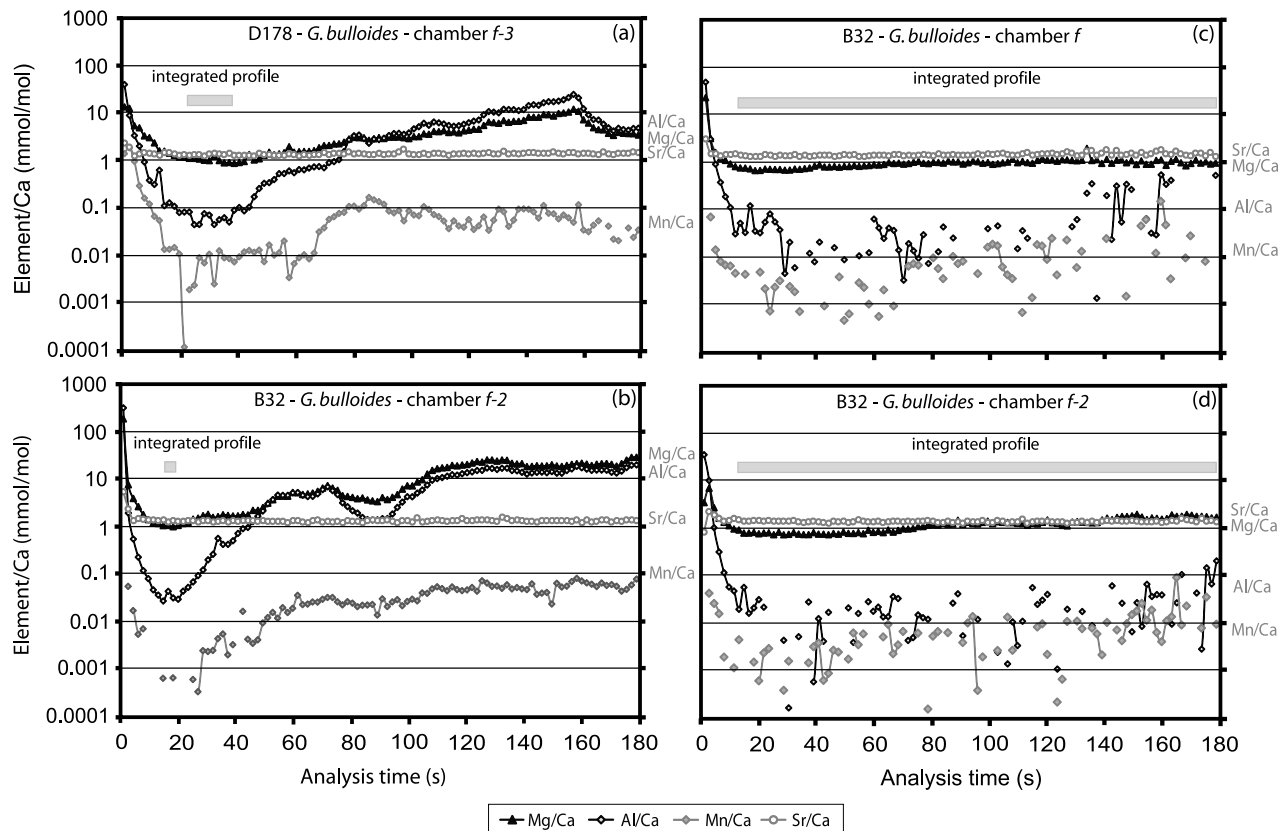
Figure 3. (a, b) Atypical and (c, d) typical time-resolved trace element depth profiles from 180 s laser ablation analyses of test chambers of three *G. bulloides* individuals (Figures 3c and 3d are analyses of different chambers from the same individual). These profiles were used to screen geochemical data for abnormal sections of a test profile resulting from contaminants. The "integrated profile" indicates the section of the analysis profile that is integrated when calculating the final trace element/Ca ratios. Note the marked stepwise change in Mg/Ca through the chamber *f*-2 of the individual from site B32 despite the low Al and Mn throughout the profile in Figure 3c.

[17] As discussed in more detail later, the outermost calcite of all tests shows significant enrichment in Mg and other trace elements, consistent with observations reported in previous in situ geochemical investigations of foraminifera [e.g., *Eggins et al.*, 2003; *Pena et al.*, 2005; *Sadekov et al.*, 2008] (Figure 3). This high-Mg "surface veneer" on the test was not integrated in the "final" trace element ratios presented in Table 2 and Table S3. *G. bulloides* is a porous,

spinose species of foraminifera [*Hemleben et al.*, 1989; *Sen Gupta*, 1999] and as such do not present an homogeneous ablation surface. Therefore, ablation takes place simultaneously in the internal and external part of the pores. Part of the surface enrichment observed in foraminiferal profiles may result from detrital material within the pores that is integrated into the final analytical signal, particularly during the first few seconds of the ablation. The data were screened

Table 2. Summary of the Size, Weight, and Trace Element Chemistry for *G. bulloides* From 10 Core Top Sites in the Southwest Pacific Ocean, East of New Zealand[a]

| Core Top	n	Water Temperature (°C)	Length Range (μm)	Length Mean (μm)	Weight Range (μg)	Weight Mean (μg)	Normalized Weight (g/m)	Mg/Ca (mmol/mol)	Al/Ca (mmol/mol)	Mn/Ca (mmol/mol)	Sr/Ca (mmol/mol)
P71	20	18.1	321–422	352 (*54.1*)	4.10–15.5	8.91 (*10.4*)	0.0253	2.671	0.125	0.024	1.22
P81	7	17.1	257–355	302 (*67.9*)	4.70–8.50	6.92 (*1.80*)	0.0229	2.412	0.101	0.165	1.39
S793	18	15.8	239–310	272 (*32.0*)	2.10–7.60	4.47 (*5.66*)	0.0164	2.714	0.152	0.069	1.33
P69	14	19.1	196–302	236 (*31.4*)	2.70–7.00	4.68 (*7.15*)	0.0198	3.328	0.062	0.039	1.41
S938	14	18.8	228–334	279 (*62.7*)	1.10–9.00	5.17 (*9.74*)	0.0185	3.475	0.024	0.153	1.39
R623	19	12.2	273–385	318 (*79.0*)	3.60–13.1	8.41 (*13.5*)	0.0264	2.210	0.298	0.040	1.38
Q220	11	9.7	252–369	312 (*67.8*)	5.60–23.1	12.1 (*28.1*)	0.0388	1.459	0.089	0.005	1.21
F111	26	7.3	314–429	391 (*100*)	6.90–32.6	19.7 (*32.6*)	0.0503	1.419	0.020	0.003	1.31
D178	19	7.4	295–482	397 (*93.6*)	11.4–55.3	17.0 (*16.5*)	0.0429	1.666	0.021	0.001	1.38
B32	25	7.8	326–498	407 (*54.1*)	7.30–38.5	19.3 (*10.4*)	0.0473	1.645	0.044	0.008	1.38

[a]Data presented are sample means. Values in italics are the 2 standard deviation of the mean. See also Figure 2.
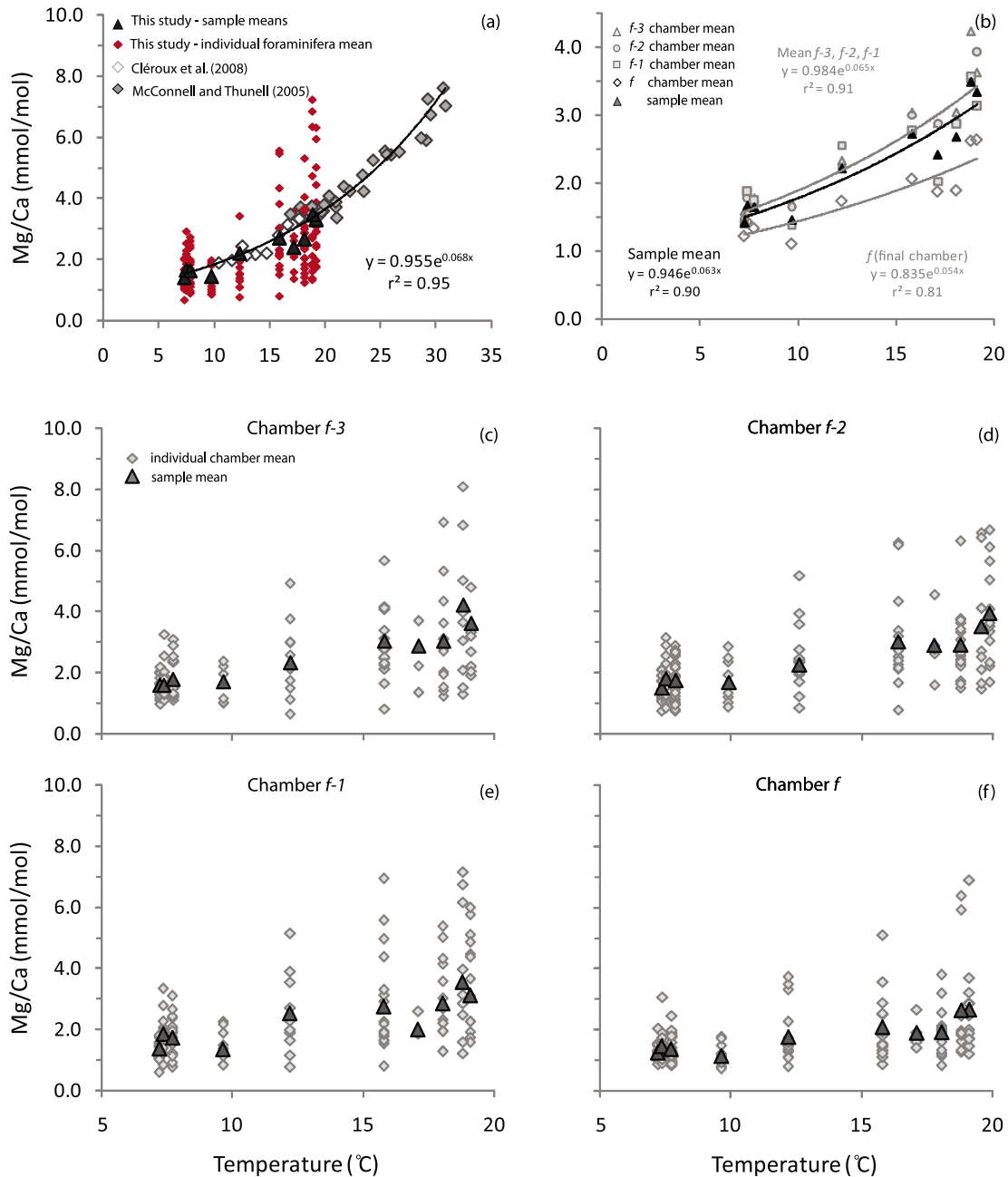
Figure 4. (a) Sample mean Mg/Ca values versus temperature for *G. bulloides*. The individual foraminifera mean analyses are also shown (small red symbols) and highlight the range in Mg/Ca values from single core top sites. Data from this study are compared with those of *Cléroux et al.* [2008] (calcification temperature) and *McConnell and Thunell* [2005] (observed sea surface temperature). Mg/Ca–temperature calibration for *G. bulloides* in this plot is a regression through all three sets of data. (b) Chamber and sample mean Mg/Ca values versus temperature for *G. bulloides*. The three curves represent Mg/Ca–temperature calibration lines based on all measurements from the three oldest visible chambers (*f*-1, *f*-2, and *f*-3), the final chamber (chamber *f*) and the sample means (all chambers) as annotated. Chamber *f* consistently yields the lowest Mg/Ca values at any given site. (c–f) All individual chamber mean Mg/Ca values (diamonds) plotted from each site highlight that the range in Mg/Ca values persists on a chamber-to-chamber level, with no single chamber exhibiting a significantly different range in Mg/Ca values.

by examining the trace element depth profiles (Figure 3), paying particular attention to Al/Ca and Mn/Ca (indicators of clay and Fe-Mn oxyhydroxide contamination). Final trace element/Ca ratios were averaged from "integrated

profiles," including those sections of the foraminiferal test that were deemed to be uncontaminated (Figure 3), whereas sections of a test with contamination on either the inner or outer test wall or through most of the wall (in rare cases;
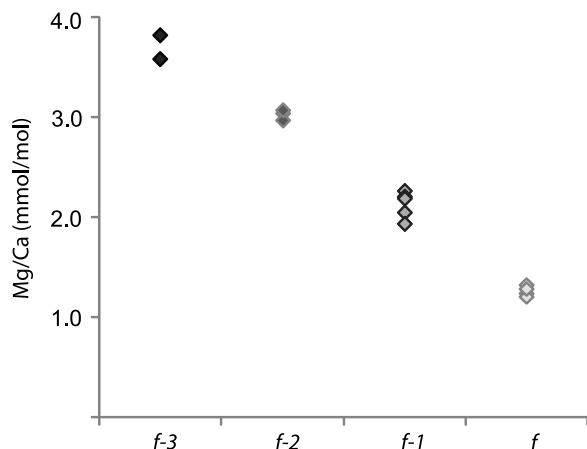
Figure 5. Individual chamber mean values and profiles for *G. bulloides*. Mg/Ca values of a *G. bulloides* individual from site S793 analyzed multiple times (*f*-3, n = 2; *f*-2, n = 4; *f*-1, n = 4; *f*, n = 3).

Figures 3a and 3b) were excluded from the average profile. The final four chambers of *G. bulloides* were analyzed separately for all individuals so as to compare geochemical data from the different chambers.

[18] A 25 nm thick carbon coating was applied to the foraminifera after LA-ICPMS analysis, and the foraminifera were then imaged by scanning electron microscopy (SEM). The SEM images of *G. bulloides* were used to measure test sizes from chamber *f* to *f*-2 (Figure 1) and assess the state of test preservation. Where appropriate, weight is normalized for individual foraminifera by dividing weight by length (normalized weight).

4. Results

[19] The trace element chemistry of foraminifera is presented and discussed as two different data sets that include (1) mean (screened) data of all visible chambers (*f*, *f*-1, *f*-2 and *f*-3) for all individuals from a core top sample and (2) mean (screened) data of one visible chamber (*f*, *f*-1, *f*-2 or *f*-3) for all individuals from a core top sample. We use the terms *sample mean* and *chamber mean*, respectively, to describe these data sets. Where reference is made to specific foraminifera the term *individual chamber mean* (mean data for a specific visible chamber (*f*, *f*-1, *f*-2 or *f*-3) or *individual foraminifera mean* (mean data of all visible chambers (*f*, *f*-1, *f*-2 and *f*-3) for a specific individual will be used.

4.1. Trace Element Depth Profiles Through Foraminifera Test Walls

[20] Figure 3 illustrates atypical and typical trace element depth profiles as the laser progressively penetrates the test wall from outside to inside. All depth profiles show a characteristic enrichment of up to 2 orders of magnitude in trace elements (except Sr) in the outermost part of the test, which is ablated in the first ~10 s of the analysis, comprising an enriched layer that we estimate to be significantly <1 μm thick. Approximately 4%–5% of foraminiferal profiles are atypical, where there is contamination throughout a signif-

icant portion of the test wall. Typical trace element depth profiles are then marked by low values of Al/Ca (<0.1 mmol/mol) and Mn/Ca (<0.1 mmol/mol). Despite low and uniform ratios of these elements, Mg/Ca may show small, but significant, systematic changes in Mg/Ca through test walls (Figures 3c and 3d). In particular, the older chambers (*f*-1, *f*-2 and *f*-3) may show 2 or 3 "plateaux" of Mg/Ca values with Mg/Ca progressively increasing by 20–40% from the outer to the inner test. In such cases, the Mg/Ca values of the outermost, low Mg/Ca, section of these older chambers generally has similar values to the average Mg/Ca of the final chamber (*f*) of the same individual (Figures 3c and 3d). One explanation for this observation is that this may reflect addition of a calcite layer to the outer part of all chambers of an individual during growth of the final chamber.

[21] Some trace element depth profiles reveal the presence of contaminants throughout most of the analysis (referred to as atypical profiles; Figures 3a and 3b). In such cases, Al, and Mn/Ca values may exceed 0.1 mmol/mol and, in particular, Al/Ca values often attain values >1–10 mmol/mol suggesting contamination by terrigenous sediment, for example, clay. Mg/Ca values show positive covariation with the other trace element ratios and only small parts of the profile can be used to extract reliable calcite Mg/Ca values. However, these parts of the profile still yield Mg/Ca values consistent with those from analyses that have typical profiles (Figures 3b, 3c, and 3d). The screened data (Table 2 and Table S2) presented in the subsequent sections excludes that for individual analyses where Al/Ca > 1 mmol/mol and results in sample means that generally have Al/Ca ≤ 0.1 mmol/mol. Screened individual analyses for each site show no correlation between Mg/Ca and Al/Ca.

4.2. Sample Mean Trace Element Variation

[22] In this study, we compare our trace element data for *G. bulloides* with that of *McConnell and Thunell* [2005] (Gulf of California) and *Cléroux et al.* [2008] (North Atlantic). The methods of data acquisition in these studies differ in two aspects to those of our study. First, they employed traditional solution-based analytical methods whereas we used the in situ LA-ICPMS method. Second, the temperatures used to construct the Mg/Ca–temperature calibration by *Cléroux et al.* [2008] were derived from oxygen isotope measurements of the foraminifera, which yielded calcification temperatures, as compared to the observed ocean temperature measurements used by *McConnell and Thunell* [2005] and in this study. However, *McConnell and Thunell* [2005] used observed sea surface temperatures rather than temperatures at 125/75 m depth and their samples were from a sediment trap placed at 485 m depth.

[23] Sample mean Mg/Ca ratios for *G. bulloides* exhibit a positive correlation with the observed water column temperature (for their assumed depth) and data from our study overlap and extend the range of temperatures for *G. bulloides* reported by *Cléroux et al.* [2008] and *McConnell and Thunell* [2005] (Figure 4). An exponential regression through our data yields (Figure 4)

$$\text{Mg/Ca (mmol/mol)} = 0.946 \times e^{0.063 \times T}$$
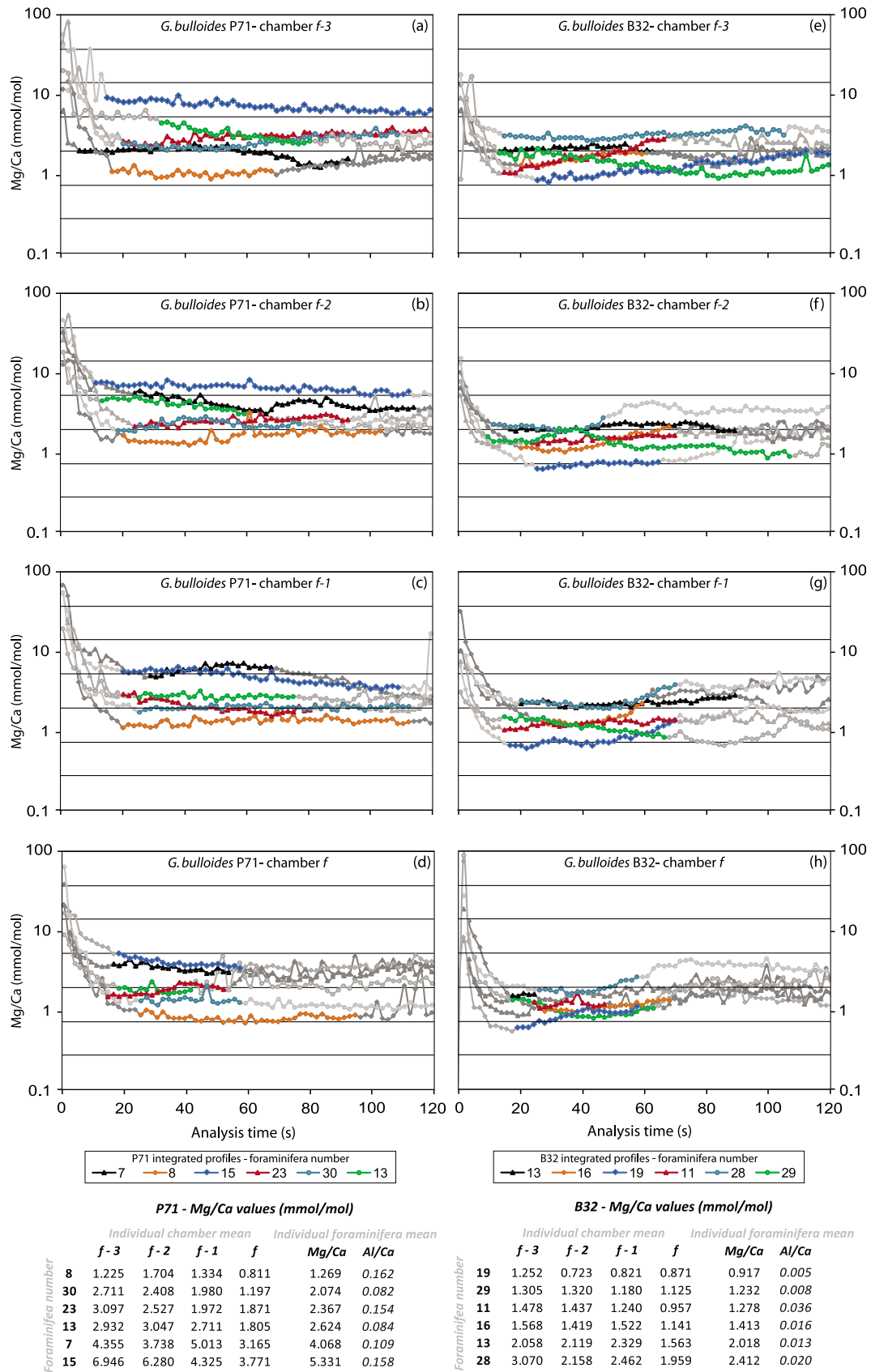$$\times (r^2 = 0.90; \text{T} = 7 \text{ to } 19°C)$$

Figure 6

and for this and the previously published studies yields (Figure 4)

$$Mg/Ca \text{ (mmol/mol)} = 0.955 \times e^{0.068 \times T}$$
$$\times (r^2 = 0.95; \ T = 7 \text{ to } 31°C).$$

Despite the agreement of the Mg/Ca–temperature relationship of this study with that of *Cléroux et al.* [2008] and *McConnell and Thunell* [2005], additional complexity is revealed when Mg/Ca values of individuals at a single site are considered or chamber-to-chamber variability is considered for each site. Sr/Ca ratios vary from 1.2 to 1.4 mmol/mol (Table 2) similar to data reported by *Cléroux et al.* [2008] with no clear relationship between Sr/Ca and temperature.

4.3. Individual-to-Individual and Chamber-to-Chamber Mg/Ca Variability

[24] At any given site, individual foraminifera show a wide range of Mg/Ca when the four chambers for each individual are averaged. For example, at sites P69 and S938 (T = 19.1°C and 18.8°C, respectively) Mg/Ca values of individual *G. bulloides* range from ~1.5 to 7 mmol/mol (Figure 4). This variability not only reflects the local influence of the warm core, Wairarapa Eddy (Figure 2), but is also observed at the cold southernmost sites (F111, D178, B32; T = 7.2°C to 7.5°C), which exhibit Mg/Ca values ranging from ~0.5 to 3 mmol/mol (Figure 4). If temperature was the main factor in driving the individual-to-individual Mg/Ca variability and using the calibrations developed above, this would correspond to temperature ranges for *individual* foraminifera of 6.5°C–29.3°C (P69, S938) and −9.5°C–16.8°C (F111, D178, B32). These calculated "temperatures" greatly exceed the range of seasonal temperatures in the near-surface ocean at these sites [e.g., *Neil et al.*, 2004], and require an important secondary control on Mg incorporation into individual foraminifera. No other trace element/Ca values in *G. bulloides* were found to consistently covary with Mg/Ca of individuals at each site.

[25] The chamber mean Mg/Ca values of *G. bulloides* also reveal a further feature that has been previously recognized by an electron microprobe study of *Anand and Elderfield* [2005] and LA-ICPMS study of *Bolton et al.* [2011]. At all sites, the lowest chamber mean Mg/Ca values characterize the final chamber (*f*) (Figure 4b and Table S2). The relationship between the chamber mean Mg/Ca for chamber *f* and water temperature is (Figure 4b)

$$Mg/Ca \text{ (mmol/mol)} = 0.835 \times e^{0.054 \times T} (r^2 = 0.81),$$

whereas the other, older, visible chambers (*f*-1, *f*-2 and *f*-3) define a relationship between the chamber mean Mg/Ca and water temperature of (Figure 4b)

$$Mg/Ca = 0.984 \times e^{0.065 \times T} (r^2 = 0.91).$$

[26] To investigate this further, a single *G. bulloides* individual from subtropical site S793 was analyzed multiple times by LA-ICPMS, including replicate analyses from each of the four visible chambers (Figure 5). Comparison of the analyses from different chambers of the individual, revealed measurable intrachamber and, in particular, interchamber trace element heterogeneity. The four analyses of chamber *f*-1 in the individual yielded a mean Mg/Ca of 2.13 ± 0.26 mmol/mol (2 sd) (Figure 5). The observed variation in Mg/Ca in this chamber (~12%) slightly exceeds analytical uncertainties (±5%–10%) and demonstrates that chemical heterogeneity persists even at the single chamber scale. However, chambers of the same individual gave average Mg/Ca values that were 74% higher in chamber *f*-3 than chamber *f* (Figure 6). The interchamber findings on this *G. bulloides* individual are consistent with large (~50%) interchamber differences in Mg/Ca in *Globigerinoides ruber* individuals [*Sadekov et al.*, 2008; *Hoogakker et al.*, 2009]. If we eliminate the final chamber from the calibration and use only the older 3 chambers for an integrated calibration this shifts the above given calibration equation of Mg/Ca (mmol/mol) = 0.955 × e$^{0.068 \times T}$ (r^2 = 0.95) to Mg/Ca = 1.008 × e$^{0.065 \times T}$ (r^2 = 0.96). This would lead to a decrease of estimated temperature from 19.1°C to 19.03°C for Mg/Ca values of 3.5 mmol/mol and a decrease from 6.64°C to 6.08°C for Mg/Ca values of 1.5 mmol/mol. The differences in temperature estimates hence are insignificant when compared to the calibration uncertainty of the temperature estimates (including analytical and calibration errors) and also in the view of the potential impact of seasonality and the range in Mg/Ca values from foraminifera from a single core top site.

4.4. Foraminiferal Test Weights and Sizes

[27] Foraminiferal test weights have been used to indicate the significance of postdepositional dissolution of foraminiferal calcite, which has been linked to the preferential loss of Mg (relative to Ca) during dissolution [e.g., *Brown and Elderfield*, 1996; *Dekens et al.*, 2002; *Rosenthal and Lohmann*, 2002; *Barker et al.*, 2004]. Foraminifera with the lowest measured test weights in this study were typically characterized by high Mg/Ca ratios (Table 2) and therefore dissolution is unlikely to be important in controlling either test weight or Mg/Ca. This is also supported by the robust Mg/Ca–temperature correlation observed for *G. bulloides*.

[28] Site mean data for *G. bulloides* exhibit a correlation between weight or size-normalized test weight and Mg/Ca or temperature (Figure 7). Samples from subtropical and subantarctic sites are clearly distinguished when mean Mg/Ca is plotted against weight. Both mean sample test weight and mean size-normalized test weight exhibit a negative correlation with the observed water column temperature (for the assumed depth). An exponential regression fitted through the

Figure 6. Time-resolved trace element depth profiles from 120 s laser ablation analyses of the four visible test chambers of 12 *G. bulloides* individuals from two sites (P71 and B32). Colored portions of the graphs represent the section of the profile integrated into the final chamber means. Relatively elevated or lower Mg/Ca values generally persist between all chambers in a particular individual; for example, elevated Mg/Ca values are foraminifera 15 for P71 and 28 for B32, and lower values are foraminifera 8 for P71 and 19 for B32. The final chamber (*f*) typically has lower Mg/Ca values. Contamination does not account for the variability in Mg/Ca values as indicated by the individual foraminifera mean Al/Ca values presented in the table at bottom.
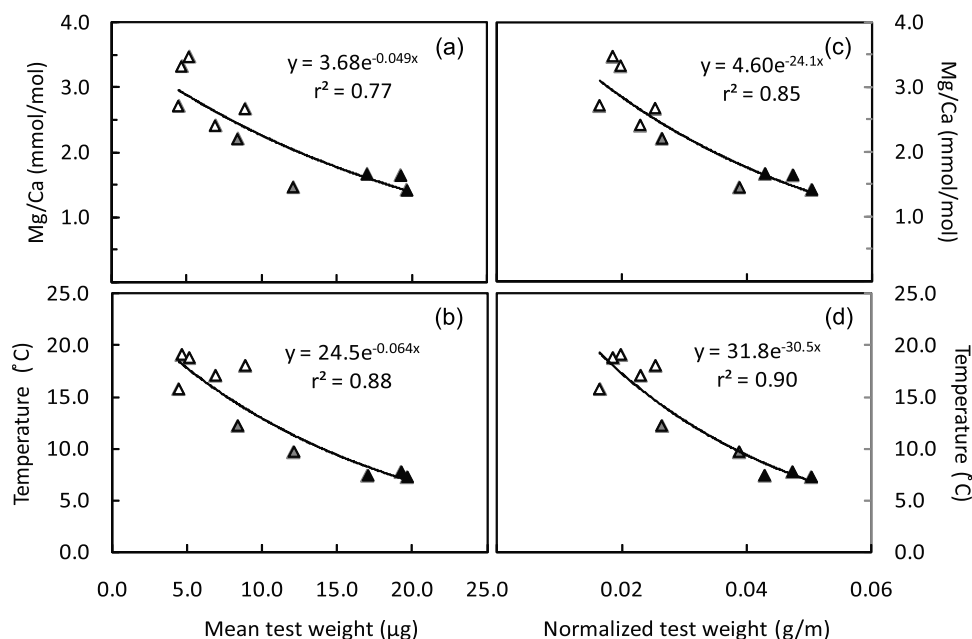
Figure 7. (a, b) *Globigerina bulloides* sample mean test weights and (c, d) mean size-normalized test weights plotted against temperature and mean Mg/Ca. Symbols are shaded according to the core top site location: subtropical sites are white, mixed sites are light gray, and subantarctic sites are dark gray.

sample mean test weights (normalized to test size) (Figure 7) is defined by the equation

$$T(°C) = 31.8 \times e^{-30.5 \times wtN} (r^2 = 0.90).$$

5. Discussion

### 5.1. Mg/Ca–Temperature Relationships in *G. bulloides*

[29] The effect of variable salinity on Mg incorporation into foraminiferal calcite has been highlighted as a potential issue for Mg/Ca paleothermometry [*Lea et al.*, 1999; *Kisakürek et al.*, 2008; *Ferguson et al.*, 2008; *Sadekov et al.*, 2009]. The salinity difference for the sites in this study is 1.2 psu (Table 1) with seasonality variability below 25 m water depth <0.5 psu [*Locarnini et al.*, 2006]. The *Lea et al.* [1999] or *Kisakürek et al.* [2008] Mg/Ca–salinity dependence of ~4 ± 3%/psu for cultured *Orbulina universa* and ~6 ± 2%/psu for cultured *G. ruber* is much smaller than Mg/Ca variations between *G. bulloides* individuals and their chambers from one core top site and between sample means from different core top sites (Table 2 and Table S2). Therefore salinity is unlikely to be a primary or significant secondary control on the incorporation of Mg into the calcite lattice in this study.

[30] Mg/Ca data for *G. bulloides* in this study correlate with observed water temperatures and are generally in good agreement with previously published core top/sediment trap calibrations and limited culturing experiments (Figure 8) [*Mashiotta et al.*, 1999; *Elderfield and Ganssen*, 2000; *McConnell and Thunell*, 2005; *Cléroux et al.*, 2008]. The lack of offset between the *McConnell and Thunell* [2005] SST-based data and that of this study (depth 125/75 m), reflects the facts that (1) *G. bulloides* dwells at greater depths in the southwest Pacific Ocean consistent with surface water mixing in northern sites of this study [*Weaver et al.*, 1997; *Lavín and Marinone*, 2003] and (2) *G. bulloides* dwells at deeper depths with increased latitude [*Mortyn and Charles*, 2003; *Wilke et al.*, 2009].

[31] While published Mg/Ca–temperature calibrations for *G. bulloides* are in good agreement with this study (Figure 8), there are some minor variations between the different Mg/Ca–temperature calibrations. Some of these differences may be explained by different carbonate ion concentrations, salinities and pH at the different sites used in these studies or minor effects of partial dissolution of foraminiferal tests at deep sites. However, we suggest that the heterogeneous Mg/Ca values observed between the final chamber (f) and older chambers (f-1, f-2, f-3) may explain some of this variability. The final chamber consistently has lower Mg/Ca than the older chambers by 20%–30% (Figure 6). Furthermore, a layer of calcite with comparable composition to the final chamber is typically found covering the outer part of the older chambers.

[32] The cause(s) of the chamber-to-chamber and individual-to-individual variability in Mg/Ca values observed in *G. bulloides* at a given core top site also requires explanation. The significant difference between the mean Mg/Ca values of the older three visible chambers (older chamber mean; f-1, f-2, f-3) (Mg/Ca (mmol/mol) = $0.984 \times e^{0.065 \times T}$) and the final visible chamber (f) (Mg/Ca (mmol/mol) = $0.835 \times e^{0.054 \times T}$) (Figure 4) can largely be explained by foraminiferal migration to greater depths (and lower temperatures) late in its life cycle. As *G. bulloides* is strongly associated with phytoplankton blooms in the ocean [*Mortyn and Charles*, 2003], it might have been expected that chambers f-3 to f-1 would have no consistent trends in their Mg/Ca test chemistry because of assumed migration related to food source. However, spinose foraminifera are general thought to migrate down through the water column late in
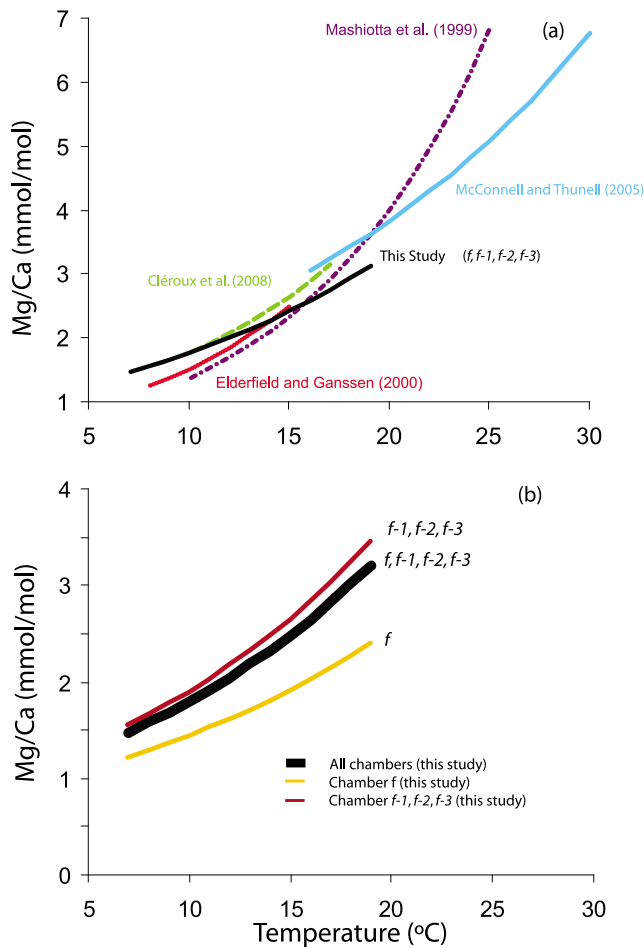
Figure 8. (a) Comparison of Mg/Ca–temperature calibrations for *Globigerina bulloides* as determined herein (thick black line) and solution ICPMS methods (dashed and solid colored lines). The length of each calibration line indicates the temperature range over which each calibration was established. (b) Three calibration lines are shown from this study corresponding to chambers *f*, *f*-1, *f*-2, and *f*-3 ("4 chamber calibration," thick black line); chambers *f*-1, *f*-2, and *f*-3 ("3 chamber calibration," red line) and chamber *f* only (orange line).

their life cycle as they shed their spines either to reproduce or soon after producing gametes [*Hemleben et al.*, 1989; *Hilbrecht and Thierstein*, 1996]. We interpret the lower values of Mg/Ca in the final chamber (*f*) to reflect this ontogeny. If *G. bulloides* migrates to a specific depth or oceanic boundary in the water column, for example, the bottom of the thermocline, this marked change in Mg/Ca values might be used in future studies to track the depth of these oceanographic features through time. This hypothesis has implications for reconstructing records of past ocean temperature by the LA-ICPMS technique as it implies that analysis of the earlier formed chambers when *G. bulloides* resides at shallowest ocean depth will most sensitively record changes in near-surface ocean temperature.

[33] While the primary control on the Mg/Ca values of each of the final four chambers is interpreted to reflect the conditions of calcification, which respond to effects such as

depth habitat varying with ontogeny, the observed variability of mean Mg/Ca values for single individuals cannot be easily explained. Mg/Ca values of individual foraminifera at a given site(s) of similar temperature vary markedly. Individuals with relatively low or high Mg/Ca from a particular site consistently record low or high Mg/Ca in all their chambers. Individuals from a specific site can record "apparent" temperatures that vary by as much as 30°C. The $\delta^{18}O$ values from individual *G. ruber* from single samples in the eastern Pacific also show a wide range in values (∼−0.4– −2.9‰) [*Koutavas et al.*, 2006]. This apparent temperature variability cannot be explained by differential vertical migration of foraminifera through the water column or seasonal changes in ocean temperatures as these observed variations are much less than the range in "apparent" temperatures. Mean Mg/Ca values of individual foraminifera from a given site show no correlation with foraminiferal size, weight or other trace element abundances. The process responsible for this effect must reflect some unidentified biological mechanism that affects Mg incorporation in foraminifera.

5.2. Comparison of Solution and Laser Ablation ICPMS Analysis Methods for Foraminiferal Mg/Ca Paleothermometry

[34] It is clear from the preceding discussion that both solution and LA-ICPMS techniques yield comparable Mg/Ca–temperature calibrations for *G. bulloides* and that both techniques are suitable to extract past ocean temperatures from this species. It is, however, pertinent to consider the relative advantages and disadvantages of these two techniques in foraminiferal Mg/Ca paleothermometry.

[35] The use of oxidative and/or reductive cleaning protocols prior to solution ICPMS analysis of foraminifera [e.g., *Martin and Lea*, 2002; *Barker et al.*, 2003] removes contaminant phases of bulk samples through a series of physical and chemical treatments, some of which are known to cause partial dissolution of test calcite [*Barker et al.*, 2003; *Sadekov et al.*, 2010]. Trace element depth profiles from LA-ICPMS analyses in this study revealed that most foraminifera only had relatively minor contamination on the inner and outer test wall. These zones of contamination were typically removed during the first and last ∼20 s of ablation for each 2–3 min analysis. Data corresponding to these contaminated zones were not used when calculating the trace element/Ca ratios reported here (Table 2 and Figures 3–9). However, despite the fact that all samples were treated with the same cleaning technique, some specimens were considerably more affected by contamination. In these cases, trace element/Ca ratios were an order of magnitude or more higher than the mean values of primary calcite. For example, Al/Ca, Mg/Ca, and Mn/Ca are observed to increase by up to 3 orders of magnitude throughout a significant portion (∼30%–80%) or even throughout the entire test wall (Figure 3).

[36] In some cases, this type of contamination affected only a single chamber in an individual, whereas in other individuals all chambers were affected. These *G. bulloides* individuals had no obvious visual or physical characteristics of contamination, including size, weight, or imaging with a binocular microscope or SEM. If one of these contaminated individuals was included in the data used to calculate sample mean trace element/Ca ratios, substantial bias would be introduced into the data set. For example, if the entire profile
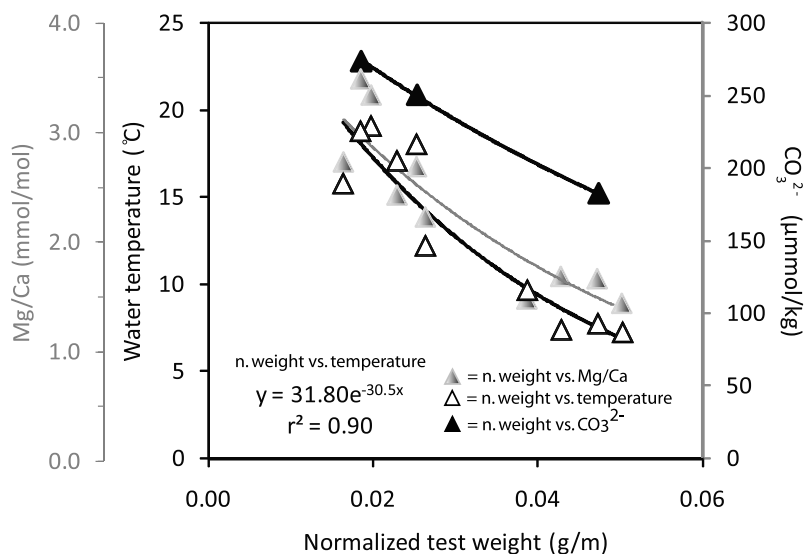
Figure 9. Comparison of the relationships between temperature, surface water carbonate ion concentration, Mg/Ca, and normalized test weights (n. weight). The negative exponential relationship between normalized test weight and Mg/Ca values is very similar to the relationship between normalized test weights and temperature, highlighting the temperature dependence of both. Carbonate ion values, although sparse, also have a negative relationship with normalized test weights. Data for the surface water carbonate ion concentrations were taken from the GLODAP Atlas (available at http://cdiac.ornl.gov/oceans/glodap/Glodap_home.htm) [*Key et al.*, 2004].

(from the integrated primary calcite to inner test wall) for a single chamber (Figure 3) from site B32 had been integrated into the sample chamber *f*-2 mean (n = 22), mean Mg/Ca would have been elevated from 1.75 to ~2.3 mmol/mol. The effect of integrating contaminated test parts in a sample mean is particularly severe for sites where only relatively small numbers of foraminifera are available for analysis. Although screening for Al/Ca would potentially have identified this bias using solution ICPMS techniques, the foraminiferal sample would have been homogenized and therefore a solution ICPMS analysis from this site would probably have been rejected. One of the principal advantages of the LA-ICPMS in situ technique is to be able to confidently exclude analyses of foraminiferal calcite where anomalous trace element signatures identify parts of foraminiferal tests that do not record primary Mg/Ca values. Further advantages of the LA-ICPMS technique compared to solution ICPMS include (1) the limited sample preparation required prior to analysis and (2) the ability to identify within-sample and within-individual trace element variations that are obscured during bulk analysis. However, solution ICPMS techniques have the advantage of greater element sensitivity, which permits better precision on the ultratrace elements such as Zn and Ba (and Li, B, U, etc.).

5.3. Test Weight-Temperature Relationships in *G. bulloides*?

[37] Early research into the relationship between test size and ocean temperature found a negative correlation defined by [*Malmgren and Kennett*, 1978]

$$\text{SST}(^{\circ}\text{C}) = -0.1184 \times (\text{mean } G. \text{ } bulloides \text{ width in } \mu\text{m}) + 45.06$$

for the southern Indian Ocean. The size-normalized test weights of planktonic foraminifera may also reflect the

intensity of downcore dissolution of foraminiferal tests [*Lohmann*, 1995; *Broecker and Clark*, 2001; *de Villiers*, 2005]. The variability in *G. bulloides* test weights from core top and downcore observations has also been linked to changes in surface ocean carbonate ion levels [e.g., *Bemis et al.*, 2000; *Barker and Elderfield*, 2002; *Russell et al.*, 2004; *Gonzalez-Mora et al.*, 2008; *Moy et al.*, 2009]. This observation is significant as an established proxy for carbonate system parameters would allow reconstruction of changes to the distribution of carbonate species in the surface and deep ocean and provide constraints necessary to evaluate the different mechanisms proposed to account for atmospheric CO_2 fluctuations [*Russell et al.*, 2004].

[38] *Barker and Elderfield* [2002] explained that size-normalized test weights are heavier during glacial periods due to the primary control on shell weight being surface water carbonate ion concentration, an observation supported by laboratory studies of *Bemis et al.* [2002] which found that increased CO_2 resulted in reduced calcification rates in planktonic foraminifera. The correlation of test weight to CO_2 variability was based on the assumption that if foraminiferal calcification is sensitive to atmospheric CO_2 concentrations, then test weight record should be affected by glacial-interglacial variations in CO_2 pressure [*Gonzalez-Mora et al.*, 2008]. *Moy et al.* [2009] used this observation to infer that recent anthropogenic increases in atmospheric CO_2, and an accompanying decrease in surface ocean carbonate ion concentration, have produced the observed 30%–35% reduction in weight of modern *G. bulloides* from sediment traps in the Australian sector of the Southern Ocean as compared to the weight of Holocene foraminifera from core tops in the same locations.

[39] However, CO_2 and carbonate ion concentrations in surface water also depend on sea surface temperature that

controls CO_2 solubility in seawater. Hence, temperature can also potentially change the calcification rate of planktonic foraminifera [*Gonzalez-Mora et al.*, 2008]. Temperature and carbonate ion levels can control foraminifera calcification rates in opposite directions depending on the foraminiferal species, and additional complexity is introduced by other variables such as salinity and nutrient availability [e.g., *de Villiers*, 2004; *Russell et al.*, 2004; *Gonzalez-Mora et al.*, 2008].

[40] Optimum growth conditions, especially temperature, offset calcification rates and thus the test size and weight of *G. bulloides* increase as species-specific optimal environmental conditions are approached [*Malmgren and Kennett*, 1978; *Bemis et al.*, 2000; *Schmidt et al.*, 2003; *de Villiers*, 2004]. The results from this study suggest that significant variability observed in mean test weight and size-normalized test weight for *G. bulloides* correlates with ocean temperature, and also lower carbonate ion levels (Figures 7 and 9). These results agree with that of *Gonzalez-Mora et al.* [2008]. We conclude that temperature in addition to ambient carbonate ion levels appears to control the normalized test weight of *G. bulloides*, where species-specific effects at lower water temperatures act to provide the conditions for the optimal growth. These results imply that caution should be exercised when solely attributing changes in mean test weights of *G. bulloides* to surface water carbonate ion concentrations, particularly in oceanographically complex regions where shifts in ocean fronts can produce marked changes in temperature, salinity and nutrient levels that can affect optimal environmental conditions for *G. bulloides* growth. To further resolve the relevant impact of temperature and carbonate ion levels on the species-specific normalized test weight further culturing experiments are required.

6. Conclusions

[41] A LA-ICPMS study of the trace element chemistry of *G. bulloides* from 10 core top sites in the southwest Pacific Ocean, east of New Zealand, has shown the following.

[42] (1) Core top Mg/Ca–temperature calibrations by LA-ICPMS can reliably reconstruct past ocean temperatures over the range 7°C–19°C.

[43] (2) These data, when combined with published data, yield an integrated calibration,

$$\text{Mg/Ca (mmol/mol)} = 0.955 \times e^{0.068 \times T} (r^2 = 0.95),$$

for a temperature range of 7°C–31°C. However, the final chamber has significantly lower Mg/Ca values (Mg/Ca (mmol/mol) = $0.835 \times e^{0.054 \times T}$, $r^2 = 0.81$) than older chambers, which may in part reflect the ecology of the species, for example, moving to deeper depths with ontogeny.

[44] (3) Subtle differences in published Mg/Ca–temperature calibrations for this species may result from incipient and variable dissolution of the thin final chamber, layers added to the outermost part of all chambers with lower Mg/Ca, and slightly different chemical cleaning procedures used prior to trace element analysis.

[45] (4) Large variations in mean Mg/Ca values of individual foraminifera from each core top site reflect some unidentified secondary control on Mg incorporation into foraminifera calcite.

[46] (5) Mean weight and size-normalized weight of *G. bulloides* correlate with ocean temperature (T (°C) = $31.8 \times e^{-30.5 \times wtN}$, $r^2 = 0.90$) and might be used to reconstruct past temperatures of surface waters, as well as highlighting potential pitfalls in attributing changes in foraminifera shell weights to one process, such as changes in surface ocean carbonate ion concentrations.

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