Biogeosciences Discuss., 9, C1254–C1269, 2012
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Biogeosciences Discussions

Interactive comment on "Relationships between bottom water carbonate saturation and element/Ca ratios in coretop samples of the benthic foraminifera *Oridorsalis umbonatus*" *by* C. F. Dawber and A. Tripati

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Received and published: 15 May 2012

Author comments on "Relationships between bottom water carbonate saturation and element/Ca ratios in coretop samples of the benthic foraminifera Oridorsalis umbonatus" by C.F Dawber & A. Tripati.

Reply to Anonymous Referee 1

Referee Comment:- "This manuscript discusses new trace metal data from the benthic foraminifer O. umbatonus and discusses two possible biomineralization mechanisms

(the surface entrapment model and the Rayleigh fractionation model) governing trace metal incorporation into foraminiferal calcite. The results suggest both mechanisms could be responsible for some, but not all of the trace metal variability. I found the discussions thorough and the figures interesting (though not that straight forward to interpret). However, the author's point out that neither mechanism could be responsible for all four metals investigated. This left me feeling unsatisfied with their findings. Rather than focus on the two mechanisms for all four metals, I think it would have been better to discuss each of the metals separately – e.g. establish which biomineralization model can best explain the Sr data, which can best explain the Mg data. Then synthesize these findings".

Author Response:- We appreciate Reviewer 1's comments and acknowledge that examining each element ratio separately and then synthesizing the findings is a valid alternative of presenting the results, but one that requires more background knowledge for each element. However, two of the element ratios examined, (Li and B), have not previously been the subject of discussion for many of the proposed biomineralisation mechanisms. Therefore our approach has been to examine the mechanisms in the context of multiple element data.

Referee Comment:- "The paper also illustrates a positive, but weak, relationship between the trace metals and bottom water carbonate ion concentration (Δ CO32-). The paper only correlates the trace metal variability to bottom water carbonate ion concentration. Temperature does not vary enough for the author's to assess any possible control temperature may have on the trace metal variability of this species. The author's do not attempt to correlate their data with any other indicators of preservation (i.e. percent-calcite, shell fragmentation). The author's are quick to point out that "it is clear that other parameters must also influence some/all of the X/Ca ratios, and that the sensitivity of individual X/Ca ratios to these additional parameters may be different." Clearly, the R2 values are not very high (Figure 1), thus while Δ CO32- likely plays a role in the trace metal incorporation, 'other parameters' controlling the trace

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metal variability in this species ought to be investigated. For example, even though the temperature range is small, plots detailing the temperature and trace metals could be included or the dataset expanded to include a wider range of temperatures. Correlating the relationships between the trace metals and other indicators of preservation (fragmentation, shell weights, etc.) would strengthen their findings."

Author Response:- Reviewer 1 makes an important point that many factors potentially contribute to element incorporation in foraminifera calcite, including Δ CO32-, temperature, percent-calcite and shell fragmentation. The majority of these parameters co-vary in the oceans, therefore from an empirical basis, it may not be clear whether the regression relationships established for any one of these parameters reflects a true mechanistic cause, or simply arise due to the co-variation of these parameters. So the question becomes, how does one best assess the influence of parameters? Other studies have looked at sample sets where both temperature and saturation state have varied, which poses a set of challenges when trying to separate out the effects of each of these parameters. Our strategy has been to design our sample set solely to focus on just one parameter, Δ CO32- with no variability in temperature, in order to (1) determine if there is support for a strong or weak sensitivity to saturation state in each of these elemental ratios; (2) examine the evidence for an underlying mechanistic cause, which might give us both better understanding of biomineralisation mechanisms, and (3) contribute to developing the empirical regression relationships as proxies. However, it is clear that this manuscript represents only one step towards understanding the relative influence of Δ CO32- and temperature (and indeed other parameters) on foraminiferal element ratios. But in isolating the influence of one parameter (Δ CO32-) and assessing proposed biomineralisation mechanisms in this context, future studies can begin to factor in additional parameters (temperature, shell fragmentation) and examine the consistency of such data with the inferences from our ΔCO32- data.

We also note that, at present, there is no consensus within the community as to how shell fragmentation relates quantitatively to bulk foraminiferal element ratios, or how

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shell fragmentation can be quantified in a precise and consistent manner, i.e. what is defined as a fragment? Is it sufficient to assess using a light microscope or should we consider test surface evidence under SEM, and if so what is a sufficient sample size? Indeed, the heterogeneity in the foraminiferal test of some of the element ratios examined (Li, B) has not been established for Oridorsalis umbontus. These factors need to be investigated before a thorough assessment can be made of the influence on foraminiferal element ratios.

Referee Comment:- "Major concern with this study is the use of the calibration equations established here in the other study the author's have in review (Climate of the Past). The author's state in their paper that another strategy to examine the cause of the X/Ca variability would be to compare the X/Ca data down core and compare those results with other proxy data, which they handle in a separate manuscript (in review in Climate of the Past). I disagree that this is an appropriate method for testing the validity of the X/Ca to Δ CO32- relationship. The X/Ca data from the core tops should be correlated with other information gleaned from the core top samples and other hydrographic data (e.g. temperature) in order to establish which of these elements/Ca ratios could possibly be used as proxies for carbonate ion concentration. The calibration equations established in this study should not be applied down core until the comparisons between the trace metals and other environmental parameters or indicators of preservation (in addition to _CO32-) are established. The Author's did not convince me that _CO32- should be used as a proxy down core yet."

Author Response:- We appreciate the comment regarding the exploration of the influence of ΔCO32- on downcore element ratios, but this comment is not really applicable for the current Biogeosciences Discussions manuscript. Nonetheless, we would like to add that we have a different opinion to Reviewer 1's comment that "The author's state in their paper that another strategy to examine the cause of the X/Ca variability would be to compare the X/Ca data down core and compare those results with other proxy data, which they handle in a separate manuscript (in review in Climate of the Past).

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I disagree that this is an appropriate method for testing the validity of the X/Ca to Δ CO32- relationship" At present there are a number of studies in the literature that have reported empirical relationships between benthic foraminifera element ratios and bottom water carbonate ion, and have suggested that these relationships can be used as proxies for past variations in bottom water carbonate ion. These inferences have been reached prior to such a study as presented here, were we take an in-depth examination of mechanistic explanations for the empirical data. What we show in the Climate of the Past downcore element ratio manuscript, using several different methods, i.e. comparison with other carbonate ion proxies and carbon cycle indices, global minimization, is that you must exercise extreme caution when applying empirical carbonate ion-element ratio relationships to other time periods, as the influence of other parameters is clearly observable in our data. Therefore on this point, we reach the same opinion as reviewer 1 that further work is required to assess the influence of other environmental parameters (i.e. preservation) on element ratios. However, as discussed above, quantitatively determining the influence of preservation on foraminiferal element ratios, in a manner that is consistent in samples from different periods, locations and is species specific, requires extensive study. In the interim period, we defend our approach of applying the empirical regression relationships down core to Middle Eocene samples at ODP Site1209, as we have highlighted the nature and magnitude of discrepancies between element-specific bottom water carbonate ion reconstructions using a multi-proxy approach. A similar assessment may not have been possible for certain time periods, when the auxiliary proxy data is lacking.

Referee Comment:- "In addition, the other paper the Author's have in review in Climate of the Past shows that the metals are not, in any way, correlated in deep time. It was interesting to see the metal/Ca ratios detailed in a downcore record (in the other paper in Climate of the Past), however, the results of the Climate of the Past paper illustrate that despite the efforts in establishing a mechanistic cause for the controls on the metal/Ca ratios of this species, there is much to be learned. As stated above a larger core-top calibration dataset that correlates the X/Ca ratios to other MODERN hydrographic

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data and other preservation proxies (fragmentation, percent calcite, etc.) would better establish which X/Ca ratios are truly correlated to carbonate ion concentration. In summary, though this paper attempts to place the focus solely on a mechanistic control on metal incorporation in O. umbatonus, too much effort is spent discussing the positive correlation between the all of the X/Ca data and _CO32-. This positive relationship is not corroborated with their down core data presented in their CPD paper and this is a major concern that should be properly dealt with prior to publication of this (or both) manuscripts. This is the reason for the Fair rating in the scientific quality section."

Author Response:- We agree with Reviewer 1's comments in so much that there is much still to be learned and that additional work investing the influence of other environmental parameters is necessary. One contribution would be to expand the modern hydrographic empirical dataset and develop multiple-factor regression relationships, however as stressed previously the intrinsic co-variation of hydrographic parameters, specifically ΔCO32-, temperature and preservation will be difficult to untangle and it may not be possible to distinguish the relative influence of individual parameters or a mechanistic cause. In addition, from a statistical point of view, the co-variation of hydrographic parameters will lead to spatial correlations within the data set, leading to additional uncertainty in the multiple regression analysis. An alternative contribution to this subject is to examine down core variations, which reflect the additional, long-term seawater chemistry and post depositional processes that cannot be properly explored in modern hydrographic empirical data sets. Therefore for this reason, we defend the approach examined in our Climate of the Past manuscript and stress that this study highlights in a quantitative manner the extent to which other environmental parameters may influence foraminiferal element ratios – an observation that probably wasn't possible by examining modern hydrographic data alone.

Referee Comment:- "Minor concerns: Figure 1: The Pacific data appears to have a different slope in figures A, B, and C. In Figure A (Li/Ca vs. _CO32-) the slope may be steeper than the Atlantic and Indian Ocean. In Figure C, there is NO correlation

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between B/Ca and _CO32- in the Pacific. Perhaps the B/Ca – _CO32- relationship asymptotes at low carbonate ion concentrations. These relationships could be better established with a larger dataset." Author Response:- We agree with these comments.

Referee Comment:- "Figure 2 isn't really discussed at all. Again, these cross plots illustrate the X/Ca data in the Pacific samples may have a different slope in comparison to the Atlantic and Indian Ocean samples." Author Response:- We agree with these comments and have added more discussion on this point.

Referee Comment:- "The author's should include a map of the core locations" Author Response:- A map has been added.

Referee Comment:-"Dawber and Tripati, 2011 is cited in the first paragraph of the introduction, but is not in the reference list. Please check this and other references for any other omissions" Author Response:- This reference is now in the reference list.

Reply to Anonymous Referee 2

Referee Comment:- "This manuscript presents new trace element data from the benthic foraminifera Oridorsalis umbonatus and discusses how these data may be controlled by various biomineralisation processes. The dataset is extensive and thoughtfully discussed, and the manuscript is well written. However there are several issues with the analysis and the manuscript that should be addressed prior to final publication. This paper is set-up as investigating the relationship between DCO3= and X/Ca. DCO3= is implied to be the major control on X/Ca for the range of sites chosen (which have a relatively small range in temperature) and biomineralisation mechanisms are discussed within this framework. Although I wanted to see more discussion of the potential effect of other factors (such as temperature - see below) I was happy to accept that the main focus of this manuscript was the DCO3= vs. X/Ca relationship with respect to biomineralisation. However a potential pitfall of this approach is that future studies might take the given X/Ca vs. DCO3= relationships as full proxy calibrations. These relationships might be applied these back in time at sites with a broader range of en-

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vironmental conditions, where the “other” parameters which have not been examined, such as temperature, might exert a more important control. So I was thus surprised to find that these authors do just that in another manuscript under interactive discussion, Dawber and Tripati, 2011. Element/Calcium ratios in middle Eocene samples, CPD, 7, 3795-3821. In this manuscript Dawber and Tripati show that the X/Ca ratios examined here do not show coherent covariations throughout their record, in contrast to what is implied in this manuscript! It thus seems important that the lack of suitability of these relationships as downcore bottom water DCO= proxies is spelt out. The authors have significantly advanced understanding of these X/Ca ratios in this species between their two papers. As the implications of both of these papers are available, they should both be taken into account, and as mentioned in a comment on the CPD manuscript, several of the analyses and parts of the discussion in that paper might be more appropriately placed in this one."

Author Response:- We acknowledge Reviewer 2’s comments, and as with our Climate of the Past manuscript, we note that some re-structuring of the two manuscripts is necessary to clarify our discussion and also to link the two studies. Additional text will be added to this extent. However, the main focus of the current manuscript was to examine the possible mechanistic relationships between element incorporation in foraminiferal calcite and bottom water carbonate ion in the framework of biomineralisation and our sample set, i.e. it focuses on carbonate ion changes and spans a narrow temperature range. The observation that Middle Eocene reconstructions at Site 1209 contain discrepancies between different element ratios does not necessarily contradict or oppose the biomineralisation mechanisms discussed here, but highlight the need to look at multiple environmental parameters as a whole system rather than just one individual parameter. Similar studies examining the effect of temperature on biomineralisation mechanisms and which integrate our observations on bottom water carbonate ion concentration should better define the nature of the relationships between foraminifera element ratios and seawater chemistry/temperature.

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Referee Comment:- "Dx PLOTS AND DISCUSSION: Much of the discussion hinges on the relationship between DCO3- and empirical partition coefficients (Dx) or fraction of Ca used (F). However the plots provided make this very difficult to assess, as these parameters are not clearly plotted against one another. Instead DCO3= is shown in colour shading, which makes its relationship to Dx or F very hard to assess. âĂćFigure 3 would be improved by making a plot for each element with DCO3= on the x axis and Dx on the y, with the different species shown as different symbols. Some species could be cut for clarity if necessary, as not many of them are discussed. A condensed version of the current figure (but omitting the DCO3= shading) could be used in another panel if comparison of the different Dx for the different elements is important. âĂćFigures 4 and 5 would also be improved by plotting DCO3= on the x axis. As F is just a function of D, there seems little point in plotting these separately each time; instead these parameters could both be shown by showing two y axes, one D, one F. âĂćTo repeat my main point, as we are asked in the text to compare D (and/or F) to DCO3=, this relationship really needs to be shown. Expressions defining D and F should also be given at the start of this point in the discussion, and a reference for the seawater X/Ca values used."

Author Response:- Reviewer 2 makes valid comments, and we have changed the figures to implement these suggestions.

Referee Comment:- "DISCUSSION OF OTHER POSSIBLE PARAMETERS - i.e. TEMPERATURE Temperature could have a control on the X/Ca data within many of the mechanisms discussed, through changing diffusion and metabolic rates. This merits more discussion, even though the T range is small, especially given the correlations with temperature shown for Mg/Ca by previous authors (e.g. Lear 2002). I'd be interested to see plots of X/Ca vs. temperature, or at least to hear how they compare to the relationships with DCO3=, even if the focus remains DCO3=."

Author Response:- We agree that temperature could have a control on the element ratios through the discussed mechanisms, and we will add a few sentences to this extent. However, we feel that given the very narrow temperature range of our dataset (1.1 to

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3.6°C) discussion on the specifics of any temperature influence on the biomineralisation mechanisms would be imprudent.

Referee Comment:- "PREVIOUS B/CA DATA AND PORE WATER INFLUENCE Previous B/Ca data have been published for Oridorsalis umbonatus by Rae et al. 2011 and Brown et al. 2011. This should be included in Figure 1. A rough version of this compilation is attached (made in Illustrator as the data were not tabulated – please include a data table or supplement in the final version). Inclusion of all available B/Ca data significantly decreases the correlation of B/Ca with DCO3=. Previous studies attribute this to the pore-water environment of O. umbonatus, which may have different DCO3= and B/Ca to that in bottom water. This influence of pore water environment may also be important for the other trace elements (again, due to altered DCO3= or X/Ca) and should be discussed."

Author Response:- The data will be archived publicly. Will we include some discussion of potential reasons for the differences between our data and published studies. For further details, see response to E. Hathorne (below) and author response to the Climate of the Past manuscript (http://www.climpast-discuss.net/7/C2740/2012/cpd-7-C2740-2012.pdf).

Referee Comment:- Specific comments: Materials and Methods: - what morphotype of Oridorsalis umbonatus was used? There are two types. Would be great to include a photo. Author Response:- We cannot provide any photos as all the samples have been analysed. We did not differentiate the morphotypes.

Referee Comment:- how many tests were run? Author Response: Between 10 and 25 individuals were run per sample.

Referee Comment:- what concentrations were samples run at? Author Response: 100ppm Ca

Referee Comment:- would be good to re-state the exclusion ratios used. Author Re-

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sponse: We will include.

Referee Comment:- is reproducibility really that similar for B/Ca and Mg/Ca? I would be interested to see the actual numbers for each X/Ca ratio, and would also prefer to see 2 s.d. given. Also how many replicates is this based on? Author Repsonse:- Yes, the reproducibility is that similar – see Yu et al. methods paper; Tripati et al., 2009, 2011 where in that case, several dozen samples have been replicated.

Referee Comment:- as mentioned above, doesn't include data of Rae 2011 or Brown 2011. – Author Response:- These data have been included.

Referee Comment:-1490, 19: I don't think sensitivity is the right word to use here - it links the data too much with the supposed mechanistic relationship to DCO3=. For instance, the range of Mg/Ca is low, but this is typical for this element in hyaline benthic foraminifera - Mg/Ca is not very sensitive in general. Maybe instead discuss ranges and correlations. Author Response:- This is a valid comment that we will take on board.

Referee Comment:-1492, 5: after [CO3=] would be good to have "(and DCO3=)" as this is the important parameter in this study, where CO3= will vary significantly with depth. Author Response:- Yes this is a good point.

Referee Comment:-1493, 27: again may be interesting to discuss with reference to pore water conditions. Author Response:- Yes this is a good point, but it will involve making some first order assumptions about the pore water carbonate ion concentration at all sites.

Referee Comment:- 1494, 1-3: again, although I can accept the different sense of these relationships, it is very hard to see if they are "well defined" or not without them being properly plotted. Autor Response:- We will implement the suggested changes to the plots so that the trends are easier to observe.

Referee Comment:- 1495, 9: interesting hypothesis - are there any other data or studies which support this? Author Response:- To our knowledge there is no supporting

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evidence, but this is something that could potentially be investigated in culture, although deep-water species are notoriously difficult to keep in the lab.

Referee Comment:- 1495, 26: another sentence here re-stating hypothesised relationship between calcite phases and DCO3= would make this more clear. Author Response:- OK.

Referee Comment:- 1496, 16: wouldn't use "similar" relationship - really just that they all show an increase with increasing DCO3=. Author Response:- OK, we will clarify.

Referee Comment:- 1497, 25: see first general comment about applicability of these relationships back in time. Author Response:- OK.

Referee Comment Figures:Provide a map of core locations. Author Response:- OK

Referee Comment: Show R2in Figure 1 Author Response:- OK,

Referee Comment:Little discussion of Figure 2 in text Author Response:- Additional text will be added.

Referee Comment:- As previously mentioned, this MS really needs a table of the data and the locations used. Author Response:- the core locations are already provided in Table 1, and will be added to a map. As mentioned the data will be archived publicaly.

Referee Comment:- Technical corrections:1487, 4: be more clear about what the Fraction Ca model is alternative to (presumably SEMO, not vacuolisation as described in the previous paragraph). Author Response:- OK, this will be clarified.

Reply to E. Hathorne Comments

Reader Comment:-"This discussion paper presents a very interesting data set and is well written. However, there are some comparisons with existing data that would improve the paper. One reviewer already pointed out the omission of the B/Ca data from Rae et al. (2011) EPSL 302, 403, and Brown et al. (2011), EPSL 310, 360. In the response to the reviews of the Climate of the Past discussion paper

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(http://www.climpast- discuss.net/7/C2740/2012/cpd-7-C2740-2012.pdf), Dawber and Tripati suggest the Brown dataset may not be accurate because of variable Ca concentrations in their samples. Although having constant Ca concentrations in samples and standards is clearly the most precise method, variable Ca concentrations are unlikely to account for the differences between the Brown dataset and the data presented here. Comparison of the Brown data for other species with those of Yu and Elderfield (2007) suggests the Brown data are reliable and not offset from previous work."

Author Response:- This manuscript was submitted prior to the publication of the Brown et al., 2011 paper, and therefore it was not possible to include a comparison of these data sets. In the author comments to the Climate of the Past manuscript (details above), we stated that one possible reason for differences between the Brown et al., 2011 Oridorsalis umbonatus data set and our own data was due to the difference is the method of data acquisition. The data presented here followed the method of Yu et al., (2005) method of using matrix-matched Ca concentrations, whereas Brown et al., (2011) used variable Ca concentrations. The reader is correct in their observation that the C. wuellerstorfi data of Brown et al., (2011) is consistent with the C. wuellerstorfi data of Yu and Elderfield, despite the different methods of data acquisition. However, the C. wuellerstorfi B/Ca ratios presented by Brown et al., (2011) range from approx. 140-210 μmol/mol – which are significantly greater than the B/Ca of O. umbonatus (20-60 μmol/mol). Therefore the relative effect of potential inaccuracies due to variable Ca concentration will be significantly smaller for C. wuellerstorfi compared to O. umbonatus, because of the larger absolute ratio. We showed in our Author response to hte Climate of the Past manuscript that the accuracy of standards with B/Ca ratios similar to O. umbonatus (31 and 60 μmol/mol), ranged from +5 to +25% for variable calcium concentrations in the range of 105 to 50 ppm Ca. Therefore, given these observations and the small absolute B/Ca of O. umbonatus, there could be significant differences in B/Ca measured by matrix-match and variable Ca concentration methods. An inter-lab calibration study could help better define differences in the methods. Also we note, that the Brown et al., (2011) study contains B/Ca data for 9 samples spanning a range

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of approx. -20 to 40 μmol/kg, whereas our data set contains 37 samples spanning a range of -23 to 44 μmol/kg.

Reader Comment:-"1485 line 23: Co is missing the +" Author Response:- Noted, and this will be corrected

Reader Comment:- "1491 line 3: The cross plots shown in Figure 2 are interesting but not discussed. Given the previous investigation of the temperature dependence of Mg/Li ratios by Bryan and Marchitto (2008) it would be nice to see what the Mg/Li ratios look like for these samples. In the bottom two plots of figure 2 the equation has Li/Ca instead of Sr/Ca as the x term."

Author Response:- Noted, and additional discussion will be provided. However, as the sample set only spans 1.1 to 3.6°C, we do not think the dataset is suitable to comment on Mg/Li ratios with respect to temperature.

Reader Comment:- "1492 line 21-24: It is true that more inorganic partition experiments are required but the work of Marriott et al. (2004a) EPSL 222, 615, has not been cited. Those authors conducted inorganic calcite precipitation experiments with a “pH stat” type apparatus and found Li incorporation to be strongly temperature dependent, especially at low temperatures approaching those of the deep-sea."

Author Response:- This is a valid comment, and this reference will be added.

Reader Comment:-"1492 line 24-27: Like the reviewers I found Figures 3-5 not straightforward. If like one reviewer suggested the discussion is focussed on each element/Ca ratio separately then all the previous benthic foraminiferal Li/Ca data could be clearly compared with the new data. The findings of Bryan and Marchitto (2008) are in direct contrast to those of Lear and Rosenthal (2006) and the present study and this could be shown more clearly. Bryan and Marchitto (2008) found Li/Ca in 5 benthic species decreases with DCO32- (and temperature) but this is difficult to see from figures 3 and 5. Can the incorporation mechanisms be so different for those species and O. umbon-

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atus or are there other important environmental controls? Given the core locations and Li/Ca data are in the papers, the data for Uvigerina from Marriott et al. (2004b) Chem. Geol. 212, 5, and the C. wuellerstorfi data from Hall and Chan (2004) could also be compared"

Author Response:- We take on board the comment that some species appear to show the opposite relationship between DLi and $\Delta CO32-$ and this may be due to different incorporation mechanisms. We note that one of these species is an aragonitic species, and therefore a different Li incorporation mechanism is not unfeasible. At present, we do not have an explanation as to why O. umbonatus exhibits the opposite relationship to C. pachyderma and U. perigrina. A larger data set is required to investigate this further.

Reader Comment:- "1496 line 2-4: Hathorne et al. (2009) Paleocenography 24, PA4204, show the Li and B content is higher in the higher Mg/Ca layers of Globorotalia shells from a sediment trap. Raitzsch et al. (2011) Geology 39, 1039, show opposite ontogenic trends for B/Ca and Mg/Ca in P. wuellerstorfi, but the processes controlling trace element heterogeneity between different calcite layers and between different chambers could be different."

Author Response:- Yes, this is a very important point, and one that needs to be extended to benthic foraminifera. It also illustrates that within the literature, there is evidence for different species exhibiting different element content to the layers of the foraminifera test and underlines the importance of comparing data on a species level.

Reader Comment:- "1496: line 6-8: This is a very interesting observation but perhaps the mechanism behind this could be discussed? Relatively little Sr/Ca variability has been observed within the shells of low Mg calcite foraminifera (e.g. Anand and Elderfield, 2005)."

Author Response:- This is an interesting point, and certainly the relative homogeneity of Sr/Ca in planktonic foraminifera (e.g. Anand and Elderfield, 2005) might suggest that

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if similar homogeneity exists between layers in benthic foraminifera, the alternating low- and high-Mg calcite layering may not be the mechanism controlling Sr and perhaps other element ratios.

Interactive comment on Biogeosciences Discuss., 9, 1483, 2012.