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Biogeosciences
Discussions
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Interactive comment on "Ba, B, and U element partitioning in magnesian calcite skeletons of Octocorallia corals" *by* T. Yoshimura et al.

T. Yoshimura et al.

yoshimurat@jamstec.go.jp

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Response to review comments

Dear Editor and Referees,

We thank two anonymous referee for the constructive reviews, and sincerely appreciate the comment which helped us to improve this manuscript. Please find our responses to the general and specific comments below.

Sincerely yours,

Toshihiro Yoshimura ************* Atmosphere and Ocean Research Institute, the University of Tokyo Department of Ocean Floor Geoscience, Submarine Geophysics Sec-

tion 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8564, Japan tel: +81-4-7136-6142 (fax: 6184), http://ofgs.aori.u-tokyo.ac.jp/member-j.html

Reviewer#1

My main concern with this manuscript is that it duplicates large portions of the companion isotope manuscript (sections 3.2, 4.2.1, 4.2.2) and those duplicated sections are some of the strongest of this report. The Ba/Ca data show a convincing linear correlation with seawater Ba concentrations, which is consistent with previous studies on octocorals, bamboo corals and aragonitic deep-sea corals. While not significantly novel, this result is worth publication, although I recommend some changes below. In contrast, the B/Ca and U/Ca data should be combined with the companion isotope manuscript, so that redundancies can be eliminated. Because both manuscripts are short and the Ba/Ca data merely confirm previous studies, I would recommend combining all data in one manuscript.

->We have split the results of trace element partitioning and isotopic fractionation of calcitic corals in two paper because the main topics discussed in O & C isotope ratios and Ba/Ca ratios were very different, and their research areas were ocean acidification and paleoceanography, respectively. But we combined all data in this manuscript (trace element paper) according to Reviewer #1 and #2 suggestions. We also added Supplementary materials.

The combined manuscript should reason why these specific elements are worthwhile to be studied, and the mechanism for U/Ca variations in response to carbonate chemistry changes needs to be reviewed in the introduction as well.

->We added the sentences below in Introduction. "In seawater, uranium exists in several different carbonate complexes, uranyl triscarbonate (UO2(CO3)34−), bicarbonate (UO2(CO3)22−) and monocarbonate (UO2CO30) complexes (Djogic et al., 1986). As pH decreases, a preferential uptake of UO2(CO3)22− and/or UO2CO30 can explain the inverse relationship between U/Ca in CaCO3 and seawater pH. Early studies show convincing annual U/Ca cycles in the reef corals (Min et al., 1995; Shen and Dunbar, 1995), and the primary objective of this paleotracer is to evaluate them as seawater pH or [CO32-] proxies (e.g., Anagnostou et al., 2011; Inoue et al., 2011; Raitzsch et al., 2011; Raddatz et al., 2014). Although the large discrepancy in pH dependence found between coral and foraminifera is due to the different CaCO3 polymorphs (Reeder et al., 2000) and species-specific calcification mechanisms, U/Ca ratios decrease as seawater [CO32-] increases (Russel et al., 2004; Anagnostou et al., 2011; Inoue et al., 2011; Raitzsch et al., 2011; Raddatz et al., 2014). In planktonic foraminifera calcite, the core-top empirical calibration shows that U/Ca is significantly affected by calcification temperature and preferential dissolution effect (Yu et al., 2008). The empirical calibration and intra-shell variation of U/Ca in calcitic corals also offers the possibility of examining the use of this proxy as an indicator of past ocean conditions (Sinclar et al., 2011)."

While validating proxies in living organisms from known chemical and physical conditions is a valuable and broadly applied approach, data interpretation is often challenging due to several environmental parameters varying simultaneously. The authors acknowledge that the current manuscript suffers from this difficulty, in that pH and temperature both decrease with water depth and thus preclude unequivocal association of decreasing B/Ca and U/Ca ratios with either one of these parameters. This complicates direct relation of oxygen isotopes to seawater acidity, for which B/Ca may be a proxy. However, the observed patterns are consistent with previous observations in foraminifera and corals (Spero et al. 1997, McConnaughey et al. 1989), where d18O decreases at lower pH (i.e. lower B/Ca ratios at greater depths, but also lower temperatures at depth). While the data shown in Figure 3 are consistent with this expectation, the text is erroneous. For instance, in the abstract (page 414, line 17) the authors say that "that d18O and d13C are enriched in light isotopes when conditions are less alkaline", page 426, line 24: "If B/Ca is assumed to be a function of the pH of the ECF, then light isotopes would be enriched in the calcifying fluid under less alkaline conditions, because B/Ca is positively correlated with d18O and d13C values. B/Ca versus d18O

regressions are shown as positive (Table 2)". These interpretations and correlations are erroneous because the relationship between B/Ca and d18O is inverse, as obvious in Figure 3. Such an inverse relationship agrees with theoretical studies on O and C isotope partitioning in seawater. The authors should read the studies of Zeebe (1999, 2001). This study still requires removal of the temperature effect on d18O before any pH effect can be evaluated, but I assume the companion manuscript deals with that. Plots of DIC, temperature and pH should be provided.

->As Reviewer #1 pointed out, pH proxies, B/Ca, and U/Ca ratios presented in this study are simultaneously affected by seawater carbonate chemistry and water temperature, i.e. pH and temperature both decrease with water depth, thus this precludes unequivocal association of decreasing B/Ca and U/Ca ratios with either one of these parameters. We changed related sentences in 4.2.2.

The Ba/Ca correlation with the seawater Ba concentration is convincing but the data presentation should be modified to include plots of these relationships in aragonitic cold-water scleractinian corals published by Anagnostou et al. (2011) and in calcitic planktic foraminifers by Hönisch et al. (2011). While the relationship of Anagnostou et al. (2011) appears similar to the ones presented in Figure 4, it falls above those relationships, and the foraminifer equation presented by Hönisch et al. (2011) falls below them, consistent with observations from inorganic studies presented in the text. Section 4.2.1 should be corrected accordingly.

->We added the data of cold-water corals and plankonic foraminifera in the Fig. 4 as suggested.

There are several redundancies in the text, some typos and some rephrasing is required in various sentences, however, given the substantial rewriting that this manuscript should undergo, I find it premature to dwell on such minor aspects. An aspect that the authors should focus on is a better presentation of the sampling strategy of the individual coral species. Which portion of the skeleton was sampled and how? This is well explained for the intra-skeletal transect but not for the other samples.

->The samples were cleaned with ultrapure H2O2 and distilled water in an ultrasonic bath to remove organic compounds. After the chemical treatment, the coral skeletons were cut along the growth axis and sliced into slabs with a diamond saw. Most of the skeleton was red or pink, with the core showing some white. The whitish core part was traceable along the growth direction. We separated the core from the outer part of the skeleton using the diamond saw, and then crushed it into sand-sized particles in an agate mortar. The coral skeletons were ground to powder in an agate mortar before ICP-MS analysis.

Furthermore, data of the same species should be plotted with the same symbol in Figure 3, so that species-specific patterns can be identified. It should be discussed how the intra-skeletal variations observed ion one specimen relate to octocorals in general. Is this one observation significant for all corals or could it be specific to this one species, or even just this specimen?

->We changed the symbol in Fig. 3 as suggested. The skeletal macro- and microstructures can greatly influence element partitioning during skeletal growth. We investigated the distributions and chemical forms of minor and major elements in the skeleton of precious corals (Paracorallium japonicum, Corallium elatius, C. rubrum) using synchrotron radiation micro-XRF (Tamenori et al., 2014, J. Struct. Biol.; Nguyen et al., 2014, Geochem. Cosmochim. Acta; and unpublished data). For example, as in the case of major elements, the core part of the precious coral skeletons are generally enriched in Mg and depleted in S (unpublished data). Similar phenomenon is observed in the aragonitic scleractinian cold-water coral Desmophyllum (Yoshimura et al., 2014, Geo-Mar. Lett.). Two major structural components in septa, centers of calcification (COC) and the surrounding fibrous region, were morphologically and compositionally different. The COCs were characterized by higher concentrations of P and Mg and lower concentrations of O and Sr. In this study, elevated Ba/Ca ratios were observed at the central axis of the skeleton, and similar Ba enrichment has been re-

ported previously in particular skeletal microstructures (please see 4.1.2.). Hasegawa et al. (2012) J. Exp. Mar. Biol. Ecol. reported that barium is homogeneously across the cross-sections of the skeleton of Japanese white coral (Corallium konojoi). Distinct distributional patterns are apparent for each element and for each genus. The variation in intra-skeletal Ba, U, and B distribution have yet to be investigated fully, but practically speaking, the inner part of the skeletons fibrous is less suitable for paleoceanographic reconstruction. This is ongoing topic in our lab.

Finally, the authors should read and cite Uchikawa et al. (2015), who performed inorganic precipitation experiments for B/Ca. The authors cite Sanyal et al. (2000) but that study did not measure B/Ca ratios but estimated them from B concentration experiments by isotope dilution. The Uchikawa data are more accurate and provide much deeper insight into B uptake into inorganic calcite.

->We changed Discussion in consideration of the latest paper by Uchikawa et al. (2015) regarding B incorporation into synthetic calcite.

Figure 6 is not discussed or introduced in the text and should be removed.

->Fig. 6 is introduced in the text (p. 426, Line 13). The relationship between water depth and previously reported skeletal growth rates of calcitic Octocorallia 10 coral taxa (Gri_n and Dru_el, 1989; Dru_el et al., 1990; Garrabou and Harmelin, 2002; Marschal et al., 2004; Andrews et al., 2005; Bramanti et al., 2005; Roark et al., 2006; Bruckner and Roberts, 2009; Gallmetzer et al., 2010; Nguyen et al., 2013; Vielzeuf et al., 2013) (Fig. 6) indicates a growth rate decrease per meter of depth. In corals living at intermediate and deep depths, differences in the availability of nutrients at habitat water depths may affect coral calcification rates.

Please not also that Yu and Elderfield (2007) studied benthic foraminifers, which follow different B incorporation patterns than planktic foraminifers and respond to Delta Carbonate Ion. The text should be corrected accordingly. Also, Allen and Hönisch (2012) argue against a temperature effect in planktic foraminifers, this study is cited in a somewhat misleading way. However, the observations made in planktic foraminifers are not necessarily true for corals, where B/Ca has been shown to be sensitive to temperature (e.g. Fallon et al. 2003). Discussion of environmental controls on B/Ca in corals needs to be improved.

->As pointed out by Reviewer #1, calcification physiology of foraminifera is very different from that of corals. As in the case of aragonitic corals, there is a growing number of boron paper (both B/Ca and δ11B). On contrary, there is still a few paper dealing with B/Ca partitioning in calcitic coral skeletons. Generally speaking, the extent of trace element uptake by CaCO3 is controlled primarily by the crystal lattice structure, so we considered foraminifera results in order to discuss possible controlling factors of B/Ca ratios in calcitic corals. We corrected the text according to Reviewer's comments.

Reviewer#2

Studies with octocorals are in their infancy and the results from this study will be an important contribution. However, the results as summarized in the abstract are unclear. Ba/Ca reflects seawater Ba/Ca, pH or carbonate ion, and is a nutrient proxy? I think they mean B/Ca is a pH proxy and Ba/Ca is a nutrient proxy, thus a typo in abstract. There is no mention of boron results in the abstract yet it warrants mention in the title? U is mentioned in the last sentence only in relation to Ba/Ca.

->We changed Abstract as suggested. Please see revised manuscript.

However, upon further review of the paper, I found a duplication of results between two papers in review with the same journal. The inclusion of _18O and _13C data were found for this paper (Table 1), this data is presented in another paper currently in discussion in the same journal (Mechanism of O and C isotope fractionation in magnesian calcite skeletons, Biogeosciences Discuss., 12, 389–412, doi:10.5194/bgd-12-389-2015, 2015). Table 1 is same in both papers and Table 2 is largely duplicated. Figure 2 presents the same data for _13Cdic as the Figure 1 in the other paper. The other paper is not cited in any of the relevant captions except Figure 3 nor in the meth-

ods and the first mention of the other paper is in the results section 3.2. Additionally, the other paper has a B/Ca vs. _18O figure that seems like it was in this paper at one point. The magnesium data in Table 1 is presented first in another paper by the authors (Yoshimura, T., Tanimizu, M., Inoue, M., Suzuki, A., Iwasaki, N., and Kawahata, H.: Mg isotope fractionation in biogenic carbonates of deep-sea coral, benthic foraminifera, and Hermatypic coral, Anal. Bioanal. Chem., 401, 2755–2769, 2011) but is not reference in the table caption but it is mentioned in the methods section. The authors probably did not mean to do anything egregious but they should clearly state there is a companion paper reporting on the same data at the start of this paper. I suggest either combining the papers, since _18O and _13C are central to their interpretations presented in this paper, or develop two papers as a part one and part two that it clearly show the two papers are related like "13C and 18O isotopic disequilibrium in biological carbonates: I. Patterns and II. In vitro simulation of kinetic isotope effects (McConnaughey, 1989a, b). The second option will clearly tie the two papers together in the same journal.

->We combined all data in this manuscript (trace element paper) according to Reviewer #1 and #2 suggestions. We also added reference of Mg/Ca data (Yoshimura et al., 2011) in the table caption.

Individual scientific questions/issues ("specific comments"): One specimen examined is a bamboo coral, Keratoisis sp. where as the others are precious corals of Corallium sp. These corals belong to the same subclass, Octocorallia but differ in families and morphologies. I would suggestion caution and/or additional support to include the bamboo coral in this study or include in the discussion the possibility of a species effect. Early work with isotopes in ahermatypic corals found differences between coral families and order (Weber, 1973a) and differences in trace elements in hermatypic corals has been found at the genus level between corals in close proximity and same reef environmental conditions (DeLong et al., 2011). Table 1 shows there are differences between oxygen and carbon isotopes and trace element ratios between C. konojoi and

P. japonicum at the same site and water depth. It is unclear if there is a species effect (Weber, 1973b) among deep sea corals at the same location and environmental conditions but the authors should consider this.

->As Reviewer #2 pointed out, the variations in the isotope ratios were greater at some depths than they were between the surface and the deepest depths. The variation in local habitat characteristics and individual coral physiology (species effect) can account for the large variation in growth rates, trace element partitioning and δ18O and δ13C at certain depths. We added sentences about species effect in Discussion 4.2.2. We also changed the colors of symbol of Fig. 3 in order to distinguish three different genera (Corallium, Paracorallium, Keratoisis).

Technical corrections: There are many technical issues to list but I withhold a detailed list until the paper structure of the two papers can be resolved. δ18O is sometimes referred to δ18 in the text and abstract, this may be an issue with special character but other occurrences are correct.

->We corrected typos in the text. Sorry for overlooking these in the proof.

Table 2 Should p-value for B/Ca and U/Ca be the same is both occurrences? One is 0.000 and the other is 0.6092.

->Sorry for the erratum. This table and Figures of the companion paper are shown as supplement material.

References Cited DeLong, K.L., Flannery, J.A., Maupin, C.R., Poore, R.Z., Quinn, T.M., 2011. A coral Sr/Ca calibration and replication study of two massive corals from the Gulf of Mexico. Palaeogeography Palaeoclimatology Palaeoecology 307, 117-128. McConnaughey, T., 1989a. 13C and 18O isotopic disequilibrium in biological carbonates: II. In vitro simulation of kinetic isotope effects. Geochimica et Cosmochimica Acta 53, 163-171. McConnaughey, T., 1989b. 13C and 18O isotopic disequilibrium in biological carbonates: I. Patterns. Geochimica et Cosmochimica Acta 53, 151-162. Weber, J.N.,

1973a. Deep-sea ahermatypic scleractinian corals - isotopic composition of skeleton. Deep-Sea Research 20, 901-909. Weber, J.N., 1973b. Incorporation of strontium into reef coral skeletal carbonate. Geochimica et Cosmochimica Acta 37, 2173-2190.

->Thank you for a list of suggested readings. We changed the text according to these references. âĂČ Interactive comments:

Anonymous Referee #1

This study presents Ba, B and U to Calcium ratios measured in 13 specimens of octocorals collected from various water depths and locations in the Pacific Ocean. The manuscript has been submitted in parallel to a companion manuscript on oxygen and carbon isotopes measured in the same specimens (Yoshimura, T., Suzuki, A., and Iwasaki, N.: Mechanism of O and C isotope fractionation in 25 magnesian calcite skeletons of Octocorallia corals and an implication on their calcification response to ocean acidification, Biogeosciences Discuss., 12, 389–412, doi:10.5194/bgd-12-389-2015), and Mg/Ca and Mg isotopes have been published earlier. My main concern with this manuscript is that it duplicates large portions of the companion isotope manuscript (sections 3.2, 4.2.1, 4.2.2) and those duplicated sections are some of the strongest of this report. The Ba/Ca data show a convincing linear correlation with seawater Ba concentrations, which is consistent with previous studies on octocorals, bamboo corals and aragonitic deep-sea corals. While not significantly novel, this result is worth publication, although I recommend some changes below. In contrast, the B/Ca and U/Ca data should be combined with the companion isotope manuscript, so that redundancies can be eliminated. Because both manuscripts are short and the Ba/Ca data merely confirm previous studies, I would recommend combining all data in one manuscript. This recommendation is further based on several inconsistencies in the data presentation and discussion, which will shorten the text upon revison. The combined manuscript should reason why these specific elements are worthwhile to be studied, and the mechanism for U/Ca variations in response to carbonate chemistry changes needs to be reviewed in the introduction as well. While validating proxies in living organisms from known chemical and physical conditions is a valuable and broadly applied approach, data interpretation is often challenging due to several environmental parameters varying simultaneously. The authors acknowledge that the current manuscript suffers from this difficulty, in that pH and temperature both decrease with water depth and thus preclude unequivocal association of decreasing B/Ca and U/Ca ratios with either one of these parameters. This complicates direct relation of oxygen isotopes to seawater acidity, for which B/Ca may be a proxy. However, the observed patterns are consistent with previous observations in foraminifera and corals (Spero et al. 1997, McConnaughey et al. 1989), where d18O decreases at lower pH (i.e. lower B/Ca ratios at greater depths, but also lower temperatures at depth). While the data shown in Figure 3 are consistent with this expectation, the text is erroneous. For instance, in the abstract (page 414, line 17) the authors say that "that d18O and d13C are enriched in light isotopes when conditions are less alkaline", page 426, line 24: "If B/Ca is assumed to be a function of the pH of the ECF, then light isotopes would be enriched in the calcifying fluid under less alkaline conditions, because B/Ca is positively correlated with d18O and d13C values. B/Ca versus d18O regressions are shown as positive (Table 2)". These interpretations and correlations are erroneous because the relationship between B/Ca and d18O is inverse, as obvious in Figure 3. Such an inverse relationship agrees with theoretical studies on O and C isotope partitioning in seawater. The authors should read the studies of Zeebe (1999, 2001). This study still requires removal of the temperature effect on d18O before any pH effect can be evaluated, but I assume the companion manuscript deals with that. Plots of DIC, temperature and pH should be provided. The Ba/Ca correlation with the seawater Ba concentration is convincing but the data presentation should be modified to include plots of these relationships in aragonitic cold-water scleractinian corals published by Anagnostou et al. (2011) and in calcitic planktic foraminifers by Hönisch et al. (2011). While the relationship of Anagnostou et al. (2011) appears similar to the ones presented in Figure 4, it falls above those relationships, and the foraminifer equation presented by Hönisch et al. (2011) falls below them, consistent with observations from inorganic studies presented in the

text. Section 4.2.1 should be corrected accordingly. There are several redundancies in the text, some typos and some rephrasing is required in various sentences, however, given the substantial rewriting that this manuscript should undergo, I find it premature to dwell on such minor aspects. An aspect that the authors should focus on is a better presentation of the sampling strategy of the individual coral species. Which portion of the skeleton was sampled and how? This is well explained for the intra-skeletal transect but not for the other samples. Furthermore, data of the same species should be plotted with the same symbol in Figure 3, so that species-specific patterns can be identified. It should be discussed how the intra-skeletal variations observed ion one specimen relate to octocorals in general. Is this one observation significant for all corals or could it be specific to this one species, or even just this specimen? Finally, the authors should read and cite Uchikawa et al. (2015), who performed inorganic precipitation experiments for B/Ca. The authors cite Sanyal et al. (2000) but that study did not measure B/Ca ratios but estimated them from B concentration experiments by isotope dilution. The Uchikawa data are more accurate and provide much deeper insight into B uptake into inorganic calcite. Figure 6 is not discussed or introduced in the text and should be removed. Please not also that Yu and Elderfield (2007) studied benthic foraminifers, which follow different B incorporation patterns than planktic foraminifers and respond to Delta Carbonate Ion. The text should be corrected accordingly. Also, Allen and Hönisch (2012) argue against a temperature effect in planktic foraminifers, this study is cited in a somewhat misleading way. However, the observations made in planktic foraminifers are not necessarily true for corals, where B/Ca has been shown to be sensitive to temperature (e.g. Fallon et al. 2003). Discussion of environmental controls on B/Ca in corals needs to be improved.

Referee #2

Overall quality of the discussion paper ("general comments") This study reports on trace elemental ratios (Ba/Ca, B/Ca, and U/Ca) in Octocorallia deep sea corals with high-magnesium calcite skeletal mineralogy, different from aragonite corals in the surface oceans commonly used to environmental reconstructions where Ba/Ca is generally considered an upwelling and/or terrigenous input proxy, B is a potential pH proxy, and U/Ca is a temperature proxy. Studies with octocorals are in their infancy and the results from this study will be an important contribution. However, the results as summarized in the abstract are unclear. Ba/Ca reflects seawater Ba/Ca, pH or carbonate ion, and is a nutrient proxy? I think they mean B/Ca is a pH proxy and Ba/Ca is a nutrient proxy, thus a typo in abstract. There is no mention of boron results in the abstract yet it warrants mention in the title? U is mentioned in the last sentence only in relation to Ba/Ca. The focus of the paper is clearly Ba/Ca, I suggest focusing on that trace However, upon further review of the paper, I found a duplication of results between two papers in review with the same journal. The inclusion of _18O and _13C data were found for this paper (Table 1), this data is presented in another paper currently in discussion in the same journal (Mechanism of O and C isotope fractionation in magnesian calcite skeletons, Biogeosciences Discuss., 12, 389–412, doi:10.5194/bgd-12-389-2015, 2015). Table 1 is same in both papers and Table 2 is largely duplicated. Figure 2 presents the same data for _13Cdic as the Figure 1 in the other paper. The other paper is not cited in any of the relevant captions except Figure 3 nor in the methods and the first mention of the other paper is in the results section 3.2. Additionally, the other paper has a B/Ca vs. _18O figure that seems like it was in this paper at one point. The magnesium data in Table 1 is presented first in another paper by the authors (Yoshimura, T., Tanimizu, M., Inoue, M., Suzuki, A., Iwasaki, N., and Kawahata, H.: Mg isotope fractionation in biogenic carbonates of deep-sea coral, benthic foraminifera, and Hermatypic coral, Anal. Bioanal. Chem., 401, 2755–2769, 2011) but is not reference in the table caption but it is mentioned in the methods section. The authors probably did not mean to do anything egregious but they should clearly state there is a companion paper reporting on the same data at the start of this paper. I suggest either combining the papers, since _18O and _13C are central to their interpretations presented in this paper, or develop two papers as a part one and part two that it clearly show the two papers are related like “13C and 18O isotopic disequilibrium

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Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/12/C2475/2015/bgd-12-C2475-2015-supplement.pdf

Interactive comment on Biogeosciences Discuss., 12, 413, 2015.

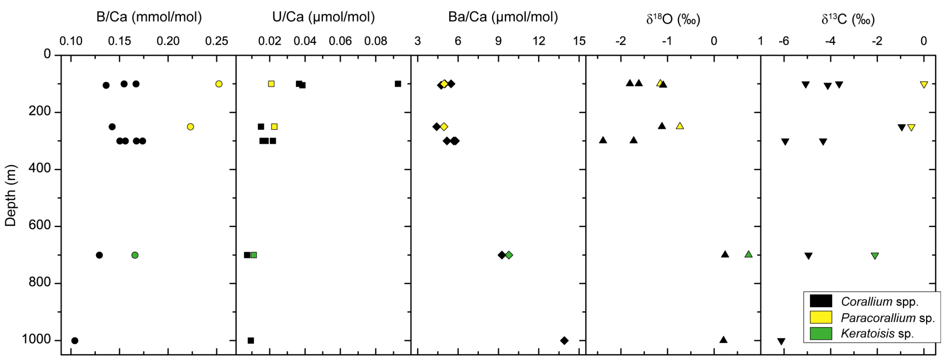

Fig. 1. Figure3_R1

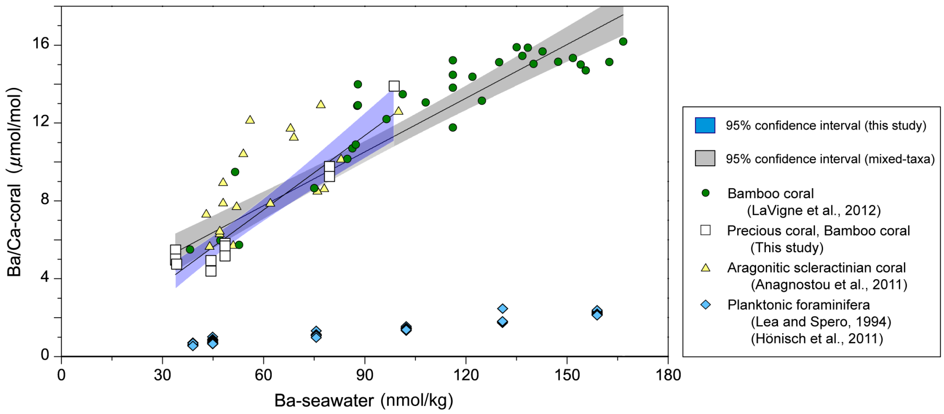

Fig. 2. Figure4_R1