The manuscript submitted by Blättler and co-authors entitled "Identifying vital effects in *Halimeda* algae with Ca isotopes" presents new and interesting results on the Ca-isotopic composition of naturally grown (Bahamas) and laboratory cultured green algae *Halimeda discoidea* (Hawaii), in order (1) to identify vital effects, (2) to evaluate the major carbonate sink of the marine Ca cycle and (3) to constrain the Ca isotope composition of seawater. Blättler and co-authors have chosen an experimental approach, in which they have collected the generally aragonite-precipitating algae *Halimeda* from its natural environment offshore Hawaii. From this collection they transplanted eight individuals into an aquarium tank, which was filled with artificial seawater with an Mg/Ca ratio mirroring Cretaceous-Eocene seawater conditions to stimulate the precipitation of both aragonite and calcite (as already observed for another *Halimeda* species (Stanley et al., 2010)). After ca 6 weeks of laboratory-controlled growth these individuals were harvested and pieces of their newly grown carbonate skeletons were prepared for XRD and Ca isotope analysis. Only small differences in mineralogy were found between one fully naturally grown and seven laboratory cultured samples, with aragonite being the dominating phase (> 91 %) and minor contributions of calcite. Only one sample revealed pure calcite instead of any aragonite. This difference in overall mineralogy is accompanied by a considerable difference in Ca isotopic compositions (ca. 0.7 – 1.0 ‰), which match the offset found between the Ca isotopic compositions of inorganic aragonite and calcite (Gussone et al., 2005; Mariott et al., 2005). The fact that aragonitic *Halimeda* samples were approx. 0.25 ‰ heavier in isotopic composition, than inorganic aragonite was explained to reflect a specific vital effect due to Rayleigh fractionation processes within the algal intercellular space. Based on these two findings the authors suggest that (1) vital effects are species-specific, (2) a mechanistic understanding of biogenic carbonate formation and vital effects is essential to develop environmental proxies, (3) mineralogy mainly determines the Ca-isotope budget of the carbonate sink and thus the Ca isotopic composition of seawater.

My biggest concern about the overall story of the manuscript is that it was stated that the single calcitic sample appeared particularly malformed. For somebody who has some experience in lab- and field-culturing experiments of marine calcifiers and proxy development this statement immediately questions the representativeness of this sample. A couple of studies (e.g. van der Putten et al., 2000) have shown that transplantation of certain calcifying species may disturb their bio-performance (e.g. precipitation rate, elemental pathways) likely affecting carbonate precipitation as well as the incorporation of trace elements and isotopes. However, a big part of the manuscript's story as well as the interpretations derived from Fig. 3 are based on the exclusiveness or exceptional position of this sample. I would argue that this finding needs to be reproduced (if it should maintain the current focus) or that the discussion is re-written, either by putting less emphasis on the calcitic sample or by discussing the new results of the aragonitic samples and their implications for the marine Ca budget and Ca isotopic seawater composition e.g. in the context of Blättler et al. (2012) Geology 40, 843-846.

As I have already mentioned, I think that the manuscript presents interesting new results but various shortcomings should be addressed before considering it for publication. I think that the topic of the manuscript fits in the scope of themes of *Biogeosciences* and thus should be published after major revisions. I have listed a number of comments below, which should be considered in a final version.

Specific comments that should be addressed in a revised version:

Page 3561-3562: The contradiction in wording concerning the reference of Stanley et al. (2010) and line 1-3 of the new page concerning the co-precipitation of calcite and aragonite.

Page 3562, 4-6: It is stated that the two expected carbonate polymorphs precipitate from the same fluid. It is not clear on which assumptions this statement is based. Does the chemical composition of the fluid that is involved in the biomineralization process really remain constant? What about slight changes in a boundary region or microenvironment? Maybe this issue becomes clearer if the paragraph on biomineralization in *Halimeda* is improved or if it is illustrated with a figure.

Page 3562, 22-26: Because of the T-dependence of Ca isotope fractionation and for comparability, the paragraph could be improved by adding the experimental growth temperatures for the respective carbonate polymorph (ACC, ikaiite and vaterite; if given in the publication).

Page 3564, 8-20: Introducing Ca isotope studies on vital effects, the systematic studies of Rollion-Bard et al. (2007) and Kasemann et al. (2008) on single shells of planktonic foraminifera using high-resolution SIMS analysis should be mentioned. Rollion-Bard et al. (2007) found a Ca isotope variability of 1.7 ‰ within one *G. inflata* test, as well as distinct differences (1.6 and 3.7 ‰) were found between ontogenetic and gametogenic foraminiferal calcite).

Page 3565-3566 Samples and method section: Because of several shortcomings this section would benefit from a number of modifications.

- Short paragraph on *Halimeda* and what is known about their calcification strategy (cf. Introduction). An illustration might additionally increase clarity.
- A brief summary on the sampling sites (Bahamas vs. Hawaii), e.g. coordinates, SST, SAL, water depth, distance from coast
- Some sentences on the culturing design and what was monitored during the 6 weeks of the experiment: collection by scuba diving (?), transfer time to the aquarium, acclimatization time, temperature, pressure, SAL, light supply, nutrition. *Halimeda discoidea* is a hard substrate dweller – How did the authors achieve the preferred life style after transplantation or did they not account for that?
- Sample preparation: Does the bleeching treatment cause any fractionation (cf. the methodological approach of Böhm et al., 2006)? Has this been tested or considered? How long were the samples pre-treated? What about ultrasonication and rinsing? Drying at what temperature?
- XRD: Information on the instrumental and analysis parameters is missing in the text (briefly given in Fig. 2). Concerning the physical mixtures to quantify the amount of calcite vs. aragonite it would have been advantageous to use biogenic calcite instead of a mineral spar. What is the error on XRD raw and on the quantified data? Why was only the newly grown carbonate sampled for XRD and not the carbonate of the basal sample? It would be interesting to know how variable the portion of calcite would be. For the natural samples from the Bahamas this information is only given in the text, but not in the table.
- Ca isotope analysis: Internal vs. long-term precision? Add "sample-standard-bracketing". Reference (Eisenhauer et al., 2004) should be given for the conversion of NIST-related values to seawater and $\delta^{44/42Ca}$ to $\delta^{44/40}$Ca.

Page 3567 Results:

I would suggest to re-order the results' section, starting with the findings on the mineralogy (XRD), then the Ca isotope data and in the end the combined findings from both methods. Moreover, the result section suffers from a mix of method descriptions and results, e.g. What is meant with skeletal mass fraction of the total mass? How is this defined? The section would further benefit from a clear assignment of the findings to the respective table, to some showcase X-ray diffractograms and a new figure on the Ca isotope composition of *Halimeda* and the measured solutions.

Page 3568-3571 Discussion:

- As mentioned earlier this paragraph would benefit from an illustration (cf. Introduction) on algae or *Halimeda* biomineralization.
- The Ca isotope composition is slightly different between the basal Hawaiian samples and the natural samples from Bahamas: Could this be because of the different water masses these individuals lived in? Specific vital effects? Different growth rates or precipitation rates? What about environmental parameters (SST, SAL) of the two sites (cf. Methods)?
- This study attempts to simulate Cretaceous-Eocene seawater conditions in respect to Mg/Ca ratios. How can the results be evaluated in comparison to the study of Steuber and Buhl (2006) who suggested that Cretaceous seawater was 0.3-0.4 ‰ lower than modern oceans.
- Page 3671, 8-12: This sentence states the importance of *Halimeda* for the late Cenozoic as one of the main aragonitic carbonate sinks. However, the discussion how this finding fits into the picture of δ^{44}Ca-changes in the Cenozoic remains insufficient. Multiple lines of evidence (Griffith et al., 2008; Schmitt et al., 2003; Heuser et al., 2005; Fantle and dePaolo, 2005, 2007) have shown that the isotopic composition of Ca changed during the Cenozoic. The changes are proposed to be associated with changes in seawater Ca concentration and changes in the input isotope ratio.

Page 3571-3572: The conclusion might probably be revised according to the proposed modifications.

Page 3577, Table 1:

- Ca isotope data for natural (Bahamas and Hawaii) and experimentally grown or laboratory cultured Halimeda algae …
- The explanations should be removed and placed in the method section.
- What about the variability in d44Ca between aragonitic Halimeda HAL-1 and HAL-8 of 0.32 ‰, between HAL-yX samples of 0.26 ‰, and the wild Halimeda of 0.24 ‰? Wild Bahamas samples are 0.23 ‰ lower than basal Hawaiian samples. Can you comment on these findings? What about the mineralogy of the wild and the basal Halimeda samples?

Page 3578, Fig. 1:

- The labelling of the axis should be completed, particularly for the insets. Here, the higher intensities (in counts) of calcite result from the fact that calcite is trigonal and aragonite orthorhombic.
- The explanations should be removed and placed in the method section.

Page 3578, Fig. 2:

- The photographs are too small to see any potential differences or deformities and the information given within the figure and in the caption is not clear enough. To my opinion the cultured ones do not really resemble the naturally grown ones. The calcified kidney-shaped segments look not that good developed. How representative are the lab-cultured samples?

Page 3580, Fig. 3:

- This figure should be replaced.

Minor issues and comments:

Page 3560, 11-14: The clarity of the sentence might be improved introducing the term "marine Ca isotope cycle".

Page 3560, 18 see above

Page 3562, 11: The references are missing.

Page 3563, 9: What samples were exactly measured by Jacobson & Holmden (2008)? This could be specified.

Page 3563, 14-15 References are missing for the precipitation experiments.

Page 3563, 20: The references are missing.

Page 3563, 23: Additional references are missing for completeness: Hippler et al. (2013), Kasemann et al. (2008) and Sime et al. (2005) or add e.g.

Page 3563, 24-25: Not only *G. sacculifer* expresses a steep slope, but also *N. pachyderma* (Hippler et al., 2009), which is in the same range 0.17 – 0.24 ‰ per °C.

Page 3564, 2: When you look at the respective figure of the publication than "significant deviations" would correspond to huge/considerable scatter in δ^{44}Ca-values spanning approx. 0.6 ‰.

Page 3568, 25: Mg/Ca ratios