

Dear Dr. de Nooijer,

**Thank you for providing constructive comments and offering us the opportunity to revise our manuscript. We have revised the manuscript following your suggestions, as detailed below. We greatly appreciate your time invested in editing our manuscript, and we look forward to publishing in Biogeosciences.**

Sincerely,  
Thomas DeCarlo on behalf of all co-authors

Editor comments:

Dear Dr. DeCarlo and co-authors,

Thank you for uploading the revised version of your manuscript. In general, you have uploaded a considerably revised manuscript in which most issues raised by the reviewers have been addressed. Some minor issues remain that need to be corrected before this manuscript can be accepted for publication in Biogeosciences. In addition, I would like to ask you to have a critical look at your title: perhaps it does not (100%) reflect the content anymore.

Sincerely,

Lennart de Nooijer

**We followed the suggested minor revisions listed below. However, we prefer to retain the present title. We agree that our previous revisions highlight some additional investigations that will help to further understand the Raman-  $\Omega_{Ar}$  proxy, but nevertheless our study does suggest that calcifying fluid  $\Omega_{Ar}$  can be derived from Raman spectroscopy. Thus, we believe the present title reflects the main thrust of the paper.**

Page 18, line 16-18: “Finally, our analysis indicates that the Raman  $\nu_1$  FWHM is an accurate proxy of fluid  $\Omega_{Ar}$ , making it a complementary approach to B/Ca and  $\delta^{11}B$  because combining information from the two approaches allows calculating the full carbonate system via  $[CO_3^{2-}]$ , pH and now importantly  $[Ca^{2+}]$ .”. The authors should be careful when making these kind of statements. The results from the inorganic precipitation experiments do not justify this interpretation promoted by the authors. In my opinion, it is currently a potential proxy for fluid  $\Omega_{Ar}$  in inorganic aragonite, which can be used to develop a way to indirectly measure  $\Omega_{Ar}$  of the calcification fluid of aragonite producing organisms, like corals.

**We revised this statement to, “Finally, our analysis of JCp-1 suggests that the Raman  $\nu_1$  FWHM may be an accurate proxy of fluid  $\Omega_{Ar}$ , making it a complementary approach to B/Ca and  $\delta^{11}B$  because combining information from the two approaches enables calculating the full carbonate system via  $[CO_3^{2-}]$ , pH and possibly now  $[Ca^{2+}]_{cf}$ .” We believe this better reflects the findings of the study, while leaving room for potential uncertainties that remain.**

Fig. 4. The authors seem to neglect the distinct offset between their observations and the ones made by Alkhatib and Eisenhauer (2017, GCA). How can you explain this offset?

**The difference is potentially related to the fluid media used in the experiments. We added two sentences, “Although our  $K_D^{Mg/Ca}$  data show a similar trend to those of Alkhatib and Eisenhauer (2017), their  $K_D^{Mg/Ca}$  are systematically lower. This offset is potentially explained by the different media used in the experiments: filtered seawater in DeCarlo et al. (2015) and Holcomb et al. (2016) as opposed to ammonium carbonate solutions in Alkhatib and Eisenhauer (2017).”**

Minor comments:

Please make change solid Mg/Ca to  $Mg/Ca_{solid}$  or  $Mg/Ca_{aragonite}$  to avoid confusion with media/seawater Mg/Ca.

**We changed to  $Mg/Ca_{solid}$  throughout.**

Throughout the manuscript:  $pCO_2$  instead of  $pCO_2$ . (i.e. please italicize the “p”). Also in the references please check  $pCO_2$  and  $CO_2$  (example page 25, lines 16 and 24).

**We changed to  $pCO_2$  throughout, including the references.**