

Dear Dr. Evans,

Thank you for the constructive comments we received on our manuscript. Below, all comments are listed in italics. We try to answer all specific comments raised by your review (in bold).

*Trans-membrane-transport is not required to explain these results, therefore a discussion of how they would be interpreted in terms of seawater vacuolisation should be included.*

**We agree with the reviewer that our results are not providing conclusive evidence for either one of the current biomineralization concepts, seawater endocytosis and TMT mixing. Still, the different observed correlations between the incorporation of trace elements in hyaline and porcelaneous species are best explained by primarily vacuolization in porcelaneous and a mixed signal in hyaline species. To accommodate the reviewers concern we now included a paragraph in which we evaluate the effect of the potential contribution of vacuolized seawater using our observations. The discussion now includes a comparison of the relative low (hyaline) and high (porcelaneous) contribution of vacuolized seawater on overall EI/Ca.**

*• Although to my knowledge it remains to be tested, it is likely that the extent to which foraminifera raise the pH of seawater vacuoles is dependent on the ambient seawater pH (also by analogy to the ECF in corals). Therefore, seawater pH can be expected to influence (e.g.) Zn and Ba speciation in the seawater vacuole. Because a certain species is probably preferentially incorporated during crystal growth, there is no need to invoke poorly selective channels or pumps (lines 311-315).*

**How (if at all) the internal pH of foraminifera depends on seawater pH outside the foraminifer, is currently not known. If it does change in concert, the internal pH at the site of calcification (which is  $\geq 9$ ; De Nooijer et al, 2009) would vary between  $>9$  and  $>8.6$  in our study. In theory such a change in internal pH from  $>9$  to  $>8.6$  changes  $[\text{CO}_3^{2-}]$  and thus the speciation of e.g. Zn and Ba at the site of calcification. However, over this range the change in  $[\text{CO}_3^{2-}]$  will be rather limited. Hence such an effect of differential speciation within the calcifying fluid does not suffice to explain the observed sensitivity in Zn and Ba to  $\text{pCO}_2$  in our study. This is in line with recent evidence on Zn/Ca in foraminifera, which suggests Zn incorporation is not governed by changes in seawater pH, but by carbonate ion concentration which does not change very much anymore at these high pH's (van Dijk et al., 2017, figure 5d).**

*• Given that the Mg distribution coefficient for some species is greater than that of inorganic calcite, it is difficult to see how TMT helps explain the geochemistry of these foraminifera. In fact, it causes problems. If these species are still sourcing a portion of the Ca through channels or pumps (lines 309-311 and 362-364), then presumably the Mg/Ca ratio of the calcifying fluid is lower than that of seawater, yet in some cases they precipitate calcite with a Mg/Ca ratio  $\sim 3$  times that of inorganic calcite. There are three species shown in Figure 3 with a Mg/Ca ratio twice that of the highest Mg/Ca species of this study. A different mechanism is required here, and it is unclear how TMT could fit into this given it would require pumping Ca out of seawater to raise the Mg/Ca ratio before precipitation. In contrast to what is stated on C2 lines 357-359, the highest Mg species are equally (or even more) different from inorganic calcite as the low-Mg species.*

**If the D of the foraminifera would be higher, this would indeed be true. However, we apologize for erroneously plotting the wrong inorganic D for Mg, which should have been ~150-200 (Mucci and Morse, 1983; Morse et al., 2007), indicating that all known foraminiferal partition coefficients are well below the inorganic D. We refrain from replotting this based on the next comment of this reviewer. Still, to accommodate this reviewers' concerns we extended our discussion, as mentioned above, to now also include other mechanisms potentially contributing to transport of ions to the SOC.**

2. Figure 3. The inorganic calcite distribution coefficients should only be displayed if they were characterised from calcite precipitated from seawater. For example, the sodium distribution coefficient is based on solutions with a chemistry very different from that of seawater, most notably the Mg concentration was much lower. It is coincidence that it is roughly the same as the miliolids and does not suggest that they precipitate shells with a similar  $D_{Na}$  to that of inorganic calcite in seawater (lines 360-361). It is well known that Mg exerts a control on trace element distribution coefficients (see below), therefore it is not representative to compare these results to inorganic precipitation where these are carbonates precipitated from non-seawater solutions.

**We have removed the inorganic D's from figure 3, since the inorganically precipitated calcites are usually not derived from (natural) seawater. Most inorganic precipitation experiments do not utilize seawater as a source for carbonate precipitation, as it complicates the design of the experiment. This makes a direct comparison with foraminiferal calcite impossible.**

3. Section 4.1 and Figure 3. The reason that trace element distribution coefficients are strongly positively correlated with  $DMg$  is because the incorporation of Mg into calcite modifies the incorporation of other elements through the associated lattice distortion. For example, this has been shown in inorganic calcite in the case of  $DSr$  [Mucci & Morse, 1983] and  $DNa$  [Okumura & Kitano, 1986], and we confirmed that this is also the case in foraminifera through cultures in variable seawater Mg/Ca [Evans et al., 2015]. The point is that this effect is not a consequence of ion transport, but has a basis in crystallography, especially given that hyaline foraminifera lie on the same  $DX-Mg/Ca$  calcite line as inorganic precipitates [see Evans et al., 2015 Fig. 7]. Furthermore, the trace element distribution coefficients shown in Figure 3 would be better expressed as a function of the calcite Mg/Ca ratio rather than  $DMg$ . It will not make much difference as most of these data are from foraminifera grown in seawater with a Mg/Ca ratio close to that of modern, but mechanistically it is the Mg concentration of calcite that is important.

**We agree that Mg incorporation might distort the calcite crystal lattice, allowing for a higher incorporation of for instance Sr and Na. Some of us were actually actively involved in studies specifically targeting this (e.g. Mewes et al., 2015). In the new version of our manuscript we now emphasized this issue by add a paragraph (in 4.2), mentioning that this interdependency of element incorporation on Mg has been observed in both inorganic and culture experiments, and might partly stem from crystallography. However, although this mechanism might explain some of the observed species specific element incorporation in hyaline foraminifera, this does not explain the difference between hyaline and porcelaneous foraminifera. Porcelaneous foraminifera have in general high Mg/Ca, but we actually observe lower incorporation of Na and Sr compared to hyaline species with similar  $DMg$  (Fig 3, upper right and left panel:  $DMg$  versus  $DNa$  and  $DSr$ ). When including porcelaneous species from other studies we also observe no increase in  $DSr$  over**

a larger range in DMg (Fig 3, upper left panel: DMg versus DSr). This indicates that the mechanisms (or mechanisms) might be very different for hyaline and porcelaneous species, which is discussed in more detail at the end of our revised discussion.

We plotted partitioning coefficients rather than element to calcium ratios since a couple of the published studies changed the Mg/Ca of the culture media, and this makes it easier for readers to evaluate their own or other data.

3. Lines 268-270. It is true that some benthic species show little response of Mg/Ca and Sr/Ca to the carbonate system, but the  $[\text{CO}_2-3]$  effect on some deep benthic foraminifera Mg/Ca is well known. These are also low-Mg so this statement is not accurate.

For deep sea species benthic foraminifera, observed response to changes in carbonation ion concentration are mainly due to calcification in undersaturated seawater, as described by ‘the carbonate ion saturation hypothesis’ (Elderfield et al., 2006) and also observed for Zn by Marchitto et al., (2000, 2005) and Cd and Ba by McCorkle et al. (1995). We now added this references and a few sentences on the carbonate ion effect in undersaturated water to the revised discussion (4.1). However, there is some evidence that Sr incorporation is directly influenced by the carbonate system (also shown by Keul et al., in press). Still, sensitivity of Sr/Ca to e.g.  $[\text{CO}_3^{2-}]$  is rather low (Keul et al., in press; van Dijk et al., 2017). In our experiment the change in  $[\text{CO}_3^{2-}]$  between the highest and lowest pCO<sub>2</sub> treatment is rather limited (90-220 μmol/kg), resulting in no observed correlation of Sr/Ca with pCO<sub>2</sub>

4. Lines 277-279. There is no significant correlation between Mg/Ca and either DIC or alkalinity in these studies.

**We removed this statement from the discussion section and state there is a correlation between Mg/Ca and the carbonate system for planktonic foraminifera.**

5. Lines 283-285. It is not really the case that there is a trend between Ba/Ca and the carbonate system in the planktonic cultures of Honisch et al. [2011]. Only the lowest pH cultures suggest any trend, but there are no replicates of these. How does the difference between these results and those presented here fit into the authors preferred biomineralisation model?

**If incorporation of Ba depends on speciation due to differences in  $[\text{CO}_3^{2-}]$ , we expect plateauing of free  $\text{Ba}^{2+}$  when we model speciation for lower pCO<sub>2</sub> conditions (higher  $[\text{CO}_3^{2-}]$ ). This is in line with the reviewers comments.**

7. Lines 334-335. I don’t understand this statement. Do you mean that the selectivity of these channels depends on the amount of ions transported? Is there any evidence for this? It is more intuitive that selectivity is not changing.

**We have re-written this section of our discussion. We agree with the referee that the second explanation is more logical/intuitive, with the amount of  $\text{Ca}^{2+}$  in the vicinity of the foraminifer decreasing, the relative element to calcium ratio of the seawater around the foraminifera increases. This would result in a higher transport of elements other than  $\text{Ca}^{2+}$ .**

8. Figure 3. There is a plotting mistake in the DNa panel. *P. acervali* and *H. antillarum* are shown with different sodium distribution coefficients but the data in Table 3 indicate that they it is the same in both species.

**This is actually a mistake in the table. *P. acervali* has a DMg of 0.46 (also reflected in the higher Mg/Ca<sub>CALCITE</sub>)**

*9. Figure 5. I understand the logic for plotting this as a function of pCO<sub>2</sub>, but given that pH is what we are able to reconstruct with boron isotopes I suggest adding a second set of x-axes to enable the two to be easily related. It would also be interesting to extend this plot to include the pH at the calcification site.*

**We added a second x-axis to this figure, with the corresponding pH from our culture study. We are not able to plot pH at the calcification site, since it is unknown how internal pH responds to changes in ambient pH.**

10. Figure 6. Half of this figure could be cut as both panels essentially show the same thing. Or, panel B could be replaced with a schematic showing how these results would fit into a biomineralisation model wherein the ions are sourced through seawater vacuolisation.

**We adapted this figure to our revised discussion, and use it to show the contribution of both mechanisms to fit our observations.**