

All referee comments are in bold and our answers in normal font.
Changes in the manuscript text are kept in italics.

Referee Toby Tyrrell

This paper presents experimental evidence showing that calcification (converting calcium ions inside the cell into solid CaCO₃ coccoliths extruded onto the outside of the cell) may have helped coccolithophores outcompete competitors in the high-calcium oceans of the Cretaceous. The rate of growth of non-calcifiers is found to be considerably depressed by high calcium, whereas calcifiers are not greatly affected. This result is intriguing because it potentially offers an explanation for why so much chalk (fossil coccoliths) was deposited on the floor of the ocean during the Cretaceous, although, as the authors note, other explanations are also possible.

This paper is well written, is based on what seem to be reasonable experimental procedures, and makes a valuable and interesting contribution to the attempt to better understand the function of calcification in coccolithophores. I am happy to recommend it for publication.

We thank T. Tyrrell for his positive review and also acknowledge the interesting and helpful literature suggestions given below.

A few fairly minor changes should be made:

1. The novel finding here is not that coccolithophores are tolerant of high [Ca]. This has been shown in several previous studies, albeit usually just for *E. huxleyi*. The key finding here is that there is a difference between calcifiers and non-calcifiers because non-calcifying phytoplankton are, conversely, not tolerant of high [Ca]. It would be useful if the authors could compare their results to the results of any earlier work growing non-calcifying phytoplankton at high [Ca], or state that they are not aware of any.

Indeed, we are not aware of any other study investigating the effects of elevated seawater Ca concentrations on the physiological performance of non-calcifying phytoplankton.

We will make this statement in the revised manuscript:

"The results presented here demonstrate the influence of seawater Ca²⁺ concentrations on marine calcifying and non-calcifying phytoplankton physiology (in terms of growth and particulate organic carbon production). Whereas previous studies already investigated the effects of elevated seawater Ca²⁺ concentrations on calcifying coccolithophore physiology and coccolith formation (Herfort et al., 2004; Langer et al., 2007; Muller et al. 2011), this study is to our knowledge the first to investigate the Ca²⁺ sensitivity of non-calcifying phytoplankton in the laboratory."

2. It should be acknowledged that the highly-controlled, highly-ornate physical architectures of most coccoliths make it unlikely that excretion of calcium is the sole function.

The various shapes of coccoliths are indeed fascinating and one could easily suggest that these different coccolith architectures might even have different functions (as often stated in architecture "form follows function").

We clearly agree that removal of calcium is not the sole function of calcification in coccolithophores and will state this more clearly in the abstract (see comments to J. Young).

3. The authors may wish to compare their results to those of Maldonado et al (1999), who also obtained interesting results from growing present-day organisms in ancient seawater chemistry.

This is indeed a very interesting publication and we were not aware of it. However, we would not like to compare the processes of calcification and silicification as this might cause confusion for non-expert readers.

4. The paragraph (pg 12703, lines 15-20) on past ocean acidification events needs revising. Catastrophic ocean acidification appears not to have taken place at either the end of the Cretaceous (Tyrrell et al., 2015) or the PETM (Gibbs et al., 2011). And even if it did, the very long (>1 My) residence time of calcium in the ocean precludes the possibility of large and rapid changes to its concentration (see for example Merico et al., 2008).

This paragraph was also mentioned by the J. Young and will be removed in the revised manuscript.

5. It is unlikely (Conclusions) that there is a similar primary function of calcification in calcified cyanobacteria and coccolithophores, or at least not if the primary function is to excrete calcium. Calcification in the former group takes place at or near to the surface of the cell, using calcium from the surrounding seawater, and hence does not help the cell to rid itself of internal calcium (Riding, 2011)

The intracellular calcification process of coccolithophores is clearly different from the extracellular mechanism of cyanobacteria. However, theoretically it might be possible that cyanobacteria mats establish (due to calcification) a microenvironment with a decreasing Ca gradient from the seawater to the interior of the mats. Thus, creating a "low" calcium environment which might prevent Ca poisoning.

Therefore, we would like to keep the statement of the relation of cyanobacteria and seawater Ca concentration but will remove the statement regarding the "primary function" of calcification.

"The occurrence of calcified cyanobacteria in the geological record during the Phanerozoic also appears to be connected to elevated seawater Ca²⁺ concentrations (Arp et al., 2001), suggesting similarities in the benefits of calcification in fossil cyanobacteria and coccolithophores."

Maldonado, Manuel, et al. "Decline in Mesozoic reef-building sponges explained by silicon limitation." *Nature* 401.6755 (1999): 785-788.

Tyrrell, Toby, Agostino Merico, and David Ian Armstrong McKay. "Severity of ocean acidification following the end-Cretaceous asteroid impact." *Proceedings of the National Academy of Sciences* 112.21 (2015): 6556-6561.

Gibbs, Samantha J., et al. "Nannoplankton extinction and origination across the Paleocene-Eocene thermal maximum." *Science* 314.5806 (2006): 1770-1773.

Merico, Agostino, Toby Tyrrell, and Paul A. Wilson. "Eocene/Oligocene ocean deacidification linked to Antarctic glaciation by sea-level fall." *Nature* 452.7190 (2008): 979-982.

Riding, Robert. "Calcified cyanobacteria." *Encyclopedia of Geobiology*. Springer Netherlands, 2011. 211-223.