Comments for the Movie and the main text

Reviewer2's comment

The present article deals with the occurrence of Al in diatoms frustules. Data are presented that show that Al is also found in the inner parts of diatoms frustules. This underlines that Al is taken up and incorporated in the frustules and excludes that Al is from other sources like detrital aluminium silicates. These data are a valuable addition to our knowledge of Al in BSi and should be published.

However, the authors state that the "occurrence mechanism of Al in marine diatoms is unclear. In particular, whether or not Al is incorporated in the structure of BSi." This is in my view not correct: Gehlen et al. (2002, cited by the authors) already gave evidence that Al is incorporated in frustules. They also showed that Al is present in the Si structure of cultured diatoms exclusively is in a four-fold coordination. Additional work by Beck et al. (2002, not cited by the authors) show that Ca is present in proportion to Al^{3+} further supporting the structural incorporation of Al in the SiO₂ network of frustules.

In my view, this work has to be acknowledged and the research question of the paper should be adapted to this.

Authors' general reply:

We are very grateful for your comments and recognition of our work, which have been quite helpful in the improvement of our manuscript. Prof. Beck and Prof. Gehlen proposed Al occurrence in BSi based on the XANES analysis, showing tetrahedral coordination of Al in BSi, and we agree that the Al was attributed to the incorporated Al in BSi. However, the visible, evidence lacks and the XAS analysis can not showing the content and distribution of structural Al. Moreover, the Al-detection by XAS is difficult in avoiding the interference of adsorbed Al and Al-bearing minerals from the sediments which may contain some minerals with tetrahedral coordinated Al, such as illite and mica, as described in the paper (*Despite our best efforts to isolate clean diatom frustules, a contamination by adhering clay particles cannot be excluded at this stage. Gehlen et al, 2002*). Therefore, our study provided a visible evidence of Al in BSi, showed more information about incorporated Al in BSi, and proposed an effective method to detect the BSi by avoiding any interference of adsorbed Al and Al-contained minerals. Considering the contribution of Beck and Gehlen, we changed the expression, accordingly.

The replies to the major issues and the corresponding revisions are briefly summarized and listed below. Please keep us informed if any more questions are raised or further discussion is requested. Thank you very much.

Main comments

1. L20: "occurrence mechanism" should be replaced by a more clearer wording	The text has been amended, accordingly.
2. L37: Given the global perspective of the paper, the range of Al concentrations can be specified ranging from below 1 nM in the Southern Ocean (e.g., van Beusekom et al., 1997) to well above 100 nM in the Mediterranean (e.g., Chou & Wollast, 1997).	The text has been amended, accordingly.
3. L55: Here, the work by Gehlen et al. (2002) and Beck et al. (2002) showing that Al is a structural part of the BSi should be cited. Note that Gehlen et al. acknowledge that Al bearing minerals may interfere when analyzing field material, but they also report on cultured diatoms grown in mediums well below the concentrations used in the present study.	The references have been added.
4. L65 ff: These statements need to be precised regarding the work by Beck et al. and Gehlen et al. who actually showed (not proposed) the coordination of Al in BSi (and the compensation of charge by inclusion of Ca).	Yes, we have added the corresponding information about the research findings of Beck et al. and Gehlen et al in the text.

5. L74-75: This is an unclear sentence that needs rewording.	The text has been amended, accordingly.
6. L90. Figure 1: misspelling of "isolate"	Yes, we have corrected the wrong words in the picture.
7. L94 Light intensity of 100 μ mol photons (isn't the unit in μ mol?)	Yes, we have corrected the wrong unit.
8. L95: F/2 was used. This contains Fe-EDTA. A large part of the Fe may be released and precipitate, impacting the Al concentrations. Given the very high concentrations used (2000 nM (10-fold of max concentrations in the ocean) precipitates may occur. Please comment on this.	Yes, the F/2 medium contains Fe-EDTA, which is of bioavailability for diatoms (Rich and Morel, 1990). Alcan hardly form complexes with EDTA in such alkaline seawater (Zhou et al., 2016; Liu et al., 2019; Zhou et al., 2021). In this scenario, EDTA may cause little influence on the Al uptake of diatoms. Moreover, we checked the F/2 with 2 μ M Al before diatoms were added and Al-bearing precipitates were not found.
 9. L101: Please indicate the Al concentration of the medium without Al. These concentrations can be substantial. 	We measured the concentration of dissolved Al before Al was added by Ultraviolet-visible Spectrophotometer and the results showed the Al concentration of several nM.
10. L130: Are the authors referring to their own studies? This should be cited.	The references have been added, and the text has been amended, accordingly.
12. L225: Here, the work by van Cappellen et al. on the effect of Al on the dissolution rate would be appropriate.	Yes, the reference has been added to the main text.
13. L236: The conclusion that 2000 nM Al is not toxic should be discussed in the light of the possibility that part of the Al is coprecipitated with Fe from the F/2 Trace metal mix. Furthermore, it has to be clarified whether EDTA is able to build a complex with EDTA. This is also relevant for the cleaning procedure where EDTA is used to desorb impurities.	Al is confirmed not to form complexes significantly with EDTA in alkaline seawater. In addition, thus the high Al-tolerant of diatoms was not attributed to the influence of EDTA (Canterford, 1979). In addition, many previous studies also indicated the diatom has high Al-tolerant (Xie et al., 2015; Gillmore et al., 2016). As suggested by Reviewer 1, we deleted the section of Al-tolerant.
14. L237: The statement that structural Al in BSi is demonstrated for the first time is in my view not valid, as Gehlen et al. and Beck et al. this already demonstrated. The observation that Al is present throughout the BSi is a good addition to our knowledge of Al in BSi.	The text has been changed, accordingly.

References

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