

Reviewer: In their manuscript, Jurkowska and colleagues examine the factors governing the siliceous sponge dissolution and reprecipitation of silica polymorphs in the seabed mud. For this purpose, they analysed the mineralogy and micro-texture of siliceous (cherts) and carbonate-siliceous (opoka) rocks of the Late Cretaceous and compared those to rocks in which siliceous sponges were found but no silica polymorphs. They concluded that the formation of cherts and silica polymorphs is an open system with a dynamic diffusion between seawater, seabed mud/ pore water and sponge.

**Authors: Exactly. Thank you for the very insightful summary.**

Reviewer: The study also raises the question what information is recorded in the Si isotope data of these siliceous rocks, which is important for future interpretations of these signals.

**Authors: We agree and wish to emphasize that, with regard to the  $\delta^{30}\text{Si}$  isotope study, we are merely raising a question rather than questioning the method, as has been suggested by the Referee #1.**

Reviewer: Generally, the structure of the MS should be improved, as pointed out by anonymous referee #1. As proposed and planned by the authors, the “re-distribution” of section 5 and the incorporation of section 3 into section 4 “materials and methods” will be helpful. Anonymous referee #1 pointed out the manuscript tends to be too “confusing and wordy”. I agree that the authors can be more concise and provided examples out in the specific comment section.

**Authors: The ms will be reorganized according to the structure suggested by both Referees. Additionally, the terminology will be refined and more clearly defined.**

Reviewer: Overall, there are some other points to raise, which should be considered more carefully in the revised version. In my opinion the authors statement of “dSi as primary environmental factor” is an oversimplification. The authors don't explain what governs the dSi concentration, factors such as pH, temperature, ionic strength, dissolved Si polymers and complexation, seawater and more importantly porewater composition are mentioned to some extent in the manuscript, but I recommend to clearly state how these are related to the Si concentration and how they drive dissolution and precipitation of opal-A..

**Authors: The description of the factors driving opal-A dissolution and precipitation will be added to the text. We discuss the factors that drive dSi precipitation from porewater because these are the only parameters we are able to recognize based on geological/mineralogical studies of rocks. The subject of this study is the Cretaceous environment, and the estimations of dSi concentration are based on the study of silica polymorphs preserved in a rock. We are not able to address the factors governing dSi concentration in seawater as outlined by Referee #2 because we lack the knowledge**

(from the literature) of these factors and we cannot reconstruct them based on rock studies alone. The only material we have for study is the rock, which was formed through the sedimentation of material on the seabed followed by diagenesis. Based on previous studies (Siever, 1991) and our models, we estimate the dSi concentration in seawater, which was likely variable within the water column and exhibited seasonal changes. However, we cannot definitively determine this from geological studies alone. Unfortunately, geological studies of rocks that are 70 million years old face significant limitations, and we are unable to apply the same analyses used for modern or younger sediment studies. To clarify these differences we will add a chapter describing the limitations inherent in geological studies of Cretaceous and older rocks.

Reviewer: Lastly, I would recommend to also cite more studies from the recent years, as there are a few investigating the dissolution and precipitation behaviour of opal-A under various conditions. Even though these are often experimental studies, I believe this would strengthen the authors arguments. Further points to consider are, that not only Mg, but also Al and Fe influence the solubility of opal-A.

Authors: We will add a discussion on the variable patterns of opal-A dissolution and the factors controlling it.

Specific comments

Reviewer: L37-42: These sentences are almost identical. Here I recommend being more concise.

Authors: The sentence will be changed.

Reviewer: L78-93: The structure of the paragraph could be improved by e.g. starting with the description of the classical model and then point out that it is derived by mineralogical and paleontological analysis. Within this paragraph, L87: The authors state that the classical model is generally accepted and extended for other siliceous rocks like opoka. The literature given regarding the "other siliceous rocks (e.g. opoka...)" is dated significantly earlier than that of Maliva and Siever, 1989a,b., which would imply these studies developed the model. I am aware this is a detail, and just a matter of wording, but could lead to confusion.

Authors: Thank you for that comment. We will improve this chapter in line with the suggestions.

Reviewer: L97: Some of the "many studies" should be cited.

Authors: The reference will be added to the text.

Reviewer: L120: What do the authors mean with "geochemically dependent"?

Reviewer: L126-130: Impurities (e.g., Al) within the opal structure of the sponge could also affect its solubility.

Authors: This means that it is controlled by geochemical conditions. Following the general comment of the Referee, this section will be extended and explained in detail, including recent studies on the factors controlling opal-A dissolution.

Reviewer: L131-136: This sentence is long and hard to follow.

Authors: The sentence will be changed.

Reviewer: L166-171: These statements need a reference.

Authors: The sentence will be rewritten.

Reviewer: Another point to consider in this paragraph or later on in the MS (section 5.3 maybe) is the nature of OM. Amino acids significantly enhance the dissolution of amorphous silica (<https://doi.org/10.1346/CCMN.2009.0570203>), which could also affect preservation of the siliceous material.

Authors: In this chapter, we discuss our methodological approach, which includes the general environmental trophic conditions in terms of the presence of OM undergoing microbial decomposition, which generates geochemical zonation within the seabed mud, triggering silica precipitation. We did not analyze the nature of the OM, but in the article by Kawano et al. (2009), the authors postulate that in specific solutions containing various amino acids at pH 4, these acids can enhance the dissolution of opal-A. This situation could occur in the close environment of sponges in the Turonian marls we studied, and we will incorporate this suggestion and discuss other factors affecting the rate of opal-A dissolution. The scale of this process is difficult to estimate because, in the studied fossils preserved as pyrite/marcasite, the pH would not drop below 6, as the primary calcium carbonate is not dissolved.

Reviewer: L190-193: The dynamics of this process should be presented more precisely and by emphasizing that clay formation scavenges dSi out of the water and precipitates in the voids. dSi concentration is therefore low, which would slow down the reprecipitation process etc.

Authors: We will add a more detailed description of the process of dSi scavenging by clays from the porewaters.

Reviewer: L278: Section 5 contains many parts where the sentences are very long and complicated which makes it often hard to follow. This should be considered when incorporating this section into the discussion.

Authors: Section 5 will be incorporated into the discussion, and the lengthy sentences will be rewritten.

Reviewer: L348-535: This sentence is long and I have trouble following the authors argument here, and I am not sure how it is connected to the previous sentence.

Authors: The previous sentence has been rewritten as suggested by Referee #1. The long sentence has also been revised.

Reviewer: L394: If "Mg-rich clays" form or a precursor, there must be Al present which affects the solubility of amorphous Si significantly, which has so far not been discussed.

Authors: The Mg-rich clays are a very early phase that precipitates simultaneously with silica polymorphs (opal-CT) and is likely a precursor phase of opal-CT (Kastner et al., 1977). The presence of this early form does not indicate significantly high Al content, but is presence in an environment in casual amount (Jurkowska et al., 2019). During precipitation, early silica forms incorporate foreign ions into their structure, including naturally occurring ions in porewater such as Al and Mg.

Reviewer: L608-610: Please add the reference of this experimental study.

Authors: The reference will be added to the text.

Reviewer: L702-712: In various studies it has been shown that in the sediment the porewater dSi increases with depth. To what sediment depth do you assume the seawater influences the mud. What depth do you assume the diagenesis took place?

Authors: In modern sediments, dSi increases with depth due to the dissolution opal-A. As we highlighted in our response to Referee #1, the Cretaceous seabed mud was very distinctive in composition and characteristics. Although we are unable to estimate the exact depths within the sediment, we can reconstruct the geochemical zonation based on the presence of authigenic minerals and geochemical conditions required for their precipitation). The Cretaceous seabed mud was non-lithified, water-saturated, with a pudding-like consistency, and exhibited high porosity and permeability. Careful estimations suggest that the SRZ zone was located at a depth of 25cm-2m depth (Clayton, 1984).

Reviewer: Temperature would influence Si solubility levels and maybe responsible for the lack of silica polymorph precipitation. What is the role of temperature in their system? What depth did the diagenesis occur in, did the elevated seawater temperature of the Cretaceous influence the system?

Authors: The temperature could affect the transformation of silica polymorphs during late diagenesis (due to an increase temperature with burial), but this did not occur in

the studied sediment, which were only shallowly buried and did not reach the temperatures high enough to trigger the diagenetic changes in the silica polymorphs (Jurkowska and Świerczewska-Gładysz, 2020a). Had such changes occurred, we would have observed them not only through mineralogical and microtextural alternations of the silica polymorphs but also in other minerals. Another effect of temperature on silica polymorphs could be inducted by seawater temperature. Unfortunately, we have limited data on the temperature of the seawater near the seabed. Bojanowski et al. (2016) studied the Campanian chalk of Poland and estimated that the near-surface temperatures could range from approximately 16-19°C, while bottom and porewater were around 13-18°C. However, these data reflect only approximate temperatures for a specific time interval and cannot be interpreted as permanent. The studied area was influenced by both colder Atlantic waters and warmer Tethyan currents, but the detailed circulation pattern during the studied interval has only been briefly recognized (Remin et al., 2016). According to our model, the main source of dSi originates from the LIP sources in the Atlantic ocean and was delivered to the European Basin by inflows, suggesting that the siliceous rocks may have formed under the colder water inflows. However, to trace this further, we would need to perform  $\delta^{18}\text{O}$  analysis of the silica polymorphs. The potential influence of temperature on silica polymorph precipitation was discussed in our previous studies (Jurkowska and Świerczewska-Gładysz, 2020a; b).

Technical corrections

Reviewer: L80: Cenozoic

Authors: The sentence has been changed and do not include that word.

Reviewer: L206: Remove one comma after in contrast.

Authors: It will be corrected.

Reviewer: L236: has been investigated in previous studies

Authors: It will be corrected.

Reviewer: L242: voids of them

Authors: It will be corrected.

Reviewer: L357-359: I strongly recommend simplifying this part of the sentence for the reader

Authors: It will be corrected.

Reviewer: L358: "remarks" seems wrong here

Reviewer: L395: Do you mean dissolution marks?

Authors: Should be "marks". It will be corrected.