

Clay alignment by sedimentation: new insights from 4D-synchrotron experiments

Corresponding Author: Dr Rebecca Kuehn

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Decision Letter:

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Dear Dr Kuehn,

Your manuscript titled "Clay alignment by sedimentation: new insights from 4D-synchrotron experiments" has now been seen by 2 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

Please find below our editorial threshold:

****Expand the theoretical background of your field of study, and discuss the novelty of your approach in advancing the current state of knowledge in your field.**

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Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Olivier Sulpis, PhD
Editorial Board Member
Communications Earth & Environment
0000-0002-6463-3320

Carolina Ortiz Guerrero, PhD
Associate Editor
Communications Earth & Environment

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript reports on a new approach to measuring clay-mineral alignment in subaqueous environments. In addition to quantifying the degree of alignment, the authors consider the time factor, cumulative overburden, and the mineral composition. The manuscript presents interesting and, to the best of my knowledge, new findings with interesting

conclusions regarding the extent of overburden's effect on clay sedimentation alignment. The introduction is well-written, and the results are presented in an informative fashion. I would suggest addressing and clarifying the following:

- While the experiment's sedimentation time is clearly dependent on the available synchrotron beam time, the method for determining this time itself is unclear. The mode of the particle size distribution, despite neglecting the shape factor, falls within the smaller range for clay minerals. This suggests a longer sedimentation time, as predicted by Stokes' Law (for instance).
- There could be more details provided on the quantification of CFO from the X-ray diffraction data analysis.
- Figure 3d suggests a nearly uniform distribution for illite, implying minimal contribution or even potential interference with the overall CPO? For clarity, this should be further addressed.
- The manuscript does not discuss the impact of clay-mineral type (e.g., kaolinite and illite) on platelet bond energy and surface charge. For instance, kaolinite platelets interact primarily via F-F hydrogen bonds, while illite relies on potassium coordination. These differences should be considered in the context of particle alignment. Additionally, increased ionic strength is known to compress the electrical double layer, potentially promoting better alignment. However, the authors' observations seem to contradict this expectation, making it an interesting point for discussion.
- There are some experimental details that could be provided for a more comprehensive description of the experiment such as: the ionic strength of the seawater, the chemical composition, solutions pH (which could also affect the edge charge), etc.

Reviewer #2 (Remarks to the Author):

1. Overall, this is an interesting short communication. The main weakness is that the authors do not really review the current state of knowledge at the beginning of the manuscript, and therefore they cannot point out exactly how their work advances the current state of knowledge. The experiments are promising but also seem rather preliminary. For example, the work would be more fundamental and generalizable if carried out using pure clay samples and systematically selected salinities or counterion types. The writing is good, but the manuscript would benefit from more work aimed at sharpening the narrative and, in particular, at identifying and highlighting the novel aspects of the work.
2. Lines 60-61, 65-66: I'm not sure I understand these sentences.
3. Figure 1: what is the basis of this figure? Is this well-established knowledge? Is it a summary of the literature (and if so, based on what references)? Is it a new framework proposed in this work? The classifications of different structures seems ambiguous and qualitative (e.g., what distinguishes "aggregate" from "non-aligned particles"?)
4. The synthesis of previous knowledge of the microstructural properties of kaolinite is very minimal. This synthesis is based on eight references (8-15), several of which focus on clay minerals other than kaolinite, with very different aggregation properties. I would recommend a more thorough review of the existing state of knowledge on clay (and particularly kaolinite and illite) microstructure and consolidation. This would help the authors highlight the new insight that is generated by their results. For example, work by Eric Ferrage, Carlos Santamarina, Etela Tombacz, seems relevant, among many others.
5. Lines 79-81: no explanation is provided for the range of conditions examined (choice of minerals, salinity).
6. Experiments: are the diffraction patterns sensitive only to the degree of particle alignment, or do they or do they also depend on solid fraction?
7. Experiments: how were the mineral samples prepared/purified? For example, the "illite" sample contains 75% illite (line 222) with many impurities. Were these removed?

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Version 1:

Decision Letter:

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Dear Dr Kuehn,

Your revised manuscript titled "Clay alignment by sedimentation: new insights from 4D-synchrotron experiments" has now been seen by the original reviewer #1, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Carolina Ortiz Guerrero, Ph.D.
Associate Editor
Communications Earth & Environment

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed all of the comments and suggestions provided in the previous review. The manuscript has been significantly improved as a result. Given the thorough revisions and improvements, I believe the manuscript is now ready for publication. I have no additional comments or suggestions.

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Response to reviewer comments

Reviewer Evaluations

Reviewer #1:

The manuscript reports on a new approach to measuring clay-mineral alignment in subaqueous environments. In addition to quantifying the degree of alignment, the authors consider the time factor, cumulative overburden, and the mineral composition. The manuscript presents interesting and, to the best of my knowledge, new findings with interesting conclusions regarding the extent of overburden's effect on clay sedimentation alignment. The introduction is well-written, and the results are presented in an informative fashion. I would suggest addressing and clarifying the following:

- While the experiment's sedimentation time is clearly dependent on the available synchrotron beam time, the method for determining this time itself is unclear. The mode of the particle size distribution, despite neglecting the shape factor, falls within the smaller range for clay minerals. This suggests a longer sedimentation time, as predicted by Stokes' Law (for instance).

Response:

The settling time was experimentally determined at Halle University prior to the synchrotron experiments in order to adjust for the injection method and the density of the injected slurry. We added this explanation to the methods section. Back-of-the-envelope calculation of settling times for 5 μm particles and 30 cm column height also gave us ~3 hours settling time.

- There could be more details provided on the quantification of CFO from the X-ray diffraction data analysis.

Response:

We added more details on the extraction of CPO data to the methods section and hope that the information is now sufficient.

- Figure 3d suggests a nearly uniform distribution for illite, implying minimal contribution or even potential interference with the overall CPO? For clarity, this should be further addressed.

Response:

We further clarified the role of illite for sedimentation and CPO development.

- The manuscript does not discuss the impact of clay-mineral type (e.g., kaolinite and illite) on platelet bond energy and surface charge. For instance, kaolinite platelets interact primarily via F-F hydrogen bonds, while illite relies on potassium coordination. These differences should be considered in the context of particle alignment. Additionally, increased ionic strength is known to compress the electrical double layer, potentially promoting better alignment. However, the authors' observations seem to contradict this expectation, making it an interesting point for discussion.

Response:

This is a very helpful comment! According to the reviewer's suggestion, we added this point to the discussion.

- There are some experimental details that could be provided for a more comprehensive description of the experiment such as: the ionic strength of the seawater, the chemical composition, solutions pH (which could also affect the edge charge), etc.

Response:

We added the chemical composition and pH of the sea water, mixed from Tropic Marin Classic sea salt and distilled water to the methods section.

Reviewer #2 (Remarks to the Author):

1. Overall, this is an interesting short communication. The main weakness is that the authors do not really review the current state of knowledge at the beginning of the manuscript, and therefore they cannot point out exactly how their work advances the current state of knowledge. The experiments are promising but also seem rather preliminary. For example, the work would be more fundamental and generalizable if carried out using pure clay samples and systematically selected salinities or counterion types. The writing is good, but the manuscript would benefit from more work aimed at sharpening the narrative and, in particular, at identifying and highlighting the novel aspects of the work.

Response:

Yes, we agree, our experiments are new and exploratory and we have to explain the state of knowledge in further detail. Therefore, we added a section tackling the electrochemical interaction of clay particles suspended in a fluid and highlighting that our experimental results indicate the incomplete knowledge about electrochemical interactions and alignment processes within the sediment.

As such experiments have not been done before and as we investigated two standard water types,

average seawater and de-ionized water, which are fundamental and instructional for further research we think that the study is worth publishing on its own and based on the current data set. Due to synchrotron beam time availability and application procedure we cannot repeat the experiments at the suggested variability of conditions within a reasonable time frame. However, we worked on sharpening the narrative and highlighting the novel aspects as suggested by the reviewer. The current set of experiments allows for a comparison of NaCl-rich and deficient water conditions, representing a systematic difference. On this base, the study opens up a new research avenue on clay-in-water sedimentation experiments at variable salinities and conditions.

2. Lines 60-61, 65-66: I'm not sure I understand these sentences.

Response:

We rewrote these sentences for clarification.

3. Figure 1: what is the basis of this figure? Is this well-established knowledge? Is it a summary of the literature (and if so, based on what references)? Is it a new framework proposed in this work? The classifications of different structures seems ambiguous and qualitative (e.g., what distinguishes “aggregate” from “non-aligned particles”)?

Response:

According to the reviewer's comment, we improved Figure 1 at several points: we included tactoids in order to be conform with other studies and now point out the difference between “aggregate” and “non-aligned particles”. We also added the studies which inspired our conceptual illustration.

4. The synthesis of previous knowledge of the microstructural properties of kaolinite is very minimal. This synthesis is based on eight references (8-15), several of which focus on clay minerals other than kaolinite, with very different aggregation properties. I would recommend a more thorough review of the existing state of knowledge on clay (and particularly kaolinite and illite) microstructure and consolidation. This would help the authors highlight the new insight that is generated by their results. For example, work by Eric Ferrage, Carlos Santamarina, Etela Tombacz, seems relevant, among many others.

Response:

Yes, we agree. We extended the section on the microstructural properties of kaolinite and illite within the scope of this manuscript. This also enforces us to highlight that the outcome of our experiments must be highly influenced by gravity and overburden.

5. Lines 79-81: no explanation is provided for the range of conditions examined (choice of minerals, salinity).

Response:

Such an explanation is now added to the paragraph.

6. Experiments: are the diffraction patterns sensitive only to the degree of particle alignment, or do they or do they also depend on solid fraction?

Response:

The intensities above background certainly depend on the amount of solid relative to water, wall thickness of the sedimentation tube and the amount of solid traversed by the beam. However, pole figures are derived after background subtraction and are given as normalized probability density functions (with a mean of 1). Hence, density variations in the pole figure and the percentage of diffracting crystallites are independent on sample volume.

7. Experiments: how were the mineral samples prepared/purified? For example, the “illite” sample contains 75% illite (line 222) with many impurities. Were these removed?

Response:

Impurities were not removed. We used the samples in the described composition, and added a sentence to the methods sections for clarification. At these impure conditions, the material is perhaps closer to a real sediment composition, but for further systematic investigations it would be interesting to study pure illite samples and vary the content and composition of impurities.

Rebuttal letter

Reviewer #1

The authors have addressed all of the comments and suggestions provided in the previous review. The manuscript has been significantly improved as a result. Given the thorough revisions and improvements, I believe the manuscript is now ready for publication. I have no additional comments or suggestions.

Authors

Thank you very much. for your review.