nature portfolio

Peer Review File

Micro-scale dissolution seams mobilise carbon in deep-sea limestones

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

1st Feb 21

Dear Dr. Schrank,

Your manuscript titled "Cryptic micro-dissolution mobilises carbon in Earth's largest carbon reservoir" has now been seen by 2 reviewers, whose comments are appended below. In the light of their advice I regret to inform you that we cannot publish your manuscript in Communications Earth & Environment.

You will see that the reviewers raise substantive concerns. We are particularly concerned that the main argument of the manuscript is rather speculative and is not strongly supported by the presented data. Taking these points together with our editorial considerations, we are unable to conclude that a straightforward revision of your manuscript would be likely to allow you to strengthen your case sufficiently. Unfortunately, these reservations are sufficiently important to preclude publication of this study in Communications Earth & Environment.

I am sorry that we cannot be more positive on this occasion and thank you for the opportunity to consider your work.

Best regards,

Mojtaba Fakhraee, PhD
Communications Earth & Environment
orcid.org/0000-0002-2461-6374

P.S. You might want to consider Scientific Reports as a potential venue for the publication of these results. Scientific Reports publishes primary research from all areas of the natural and clinical sciences that is judged to be scientifically valid and technically sound, whatever the considered significance. (see below for more information.)

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funding, and advice and support from Springer Nature.

Reviewers' comments:

Reviewer #2 (Remarks to the Author):

Review about "Cryptic micro-dissolution mobilises carbon in Earth's largest carbon reservoir" by Schrank, C. E., Jones, M. W. M., Kewish, C. M., van Riessen, G. A., Elphick, K. E., Sloss, C. R., Nothdurft, L. D., Webb, G. E., Paterson, D. J., and Regenauer-Lieb, K.
This manuscript presents the microstructural analysis of microdissolution features in pelagitic limestones, analyses the budget of carbon released during the process. It then uses the results to discuss on the fate of this carbon, and how much of this carbon is indeed lithified and buried, versus how much can be released in the ocean.
Deep-sea limestones represents one of the largest carbon sinks in the Earth, and the rate of burial, and efficiency of the process in terms of carbon budget is a very important issue to understand the dynamics of atmospheric and oceanic carbon, which are issues interesting a wide range of researchers in Earth and environmental science.
The study of the amount of carbon mobilized is based on the analysis of stylolites and microdissolution features, and on the enrichment in non-soluble species, such as Fe, with respect to the surrounding rock matrix. It allows the estimate of dissolved $CaCO3$ during the formation of the solution seams. The authors use a set of powerful Xray fluorescence microscopy and ptychography techniques to obtain microstructural analysis and precise chemical elemental maps of their samples with a resolution around 100 nm to 1 micrometer. This allows, among other, to analyse microdissolution features, which are commonly disregarded due to their small size.
The analysis, carried out on a significant set of rock thin sections, is sound and clearly presented. The results are clear, well presented, and the discussion and consequences for the main question are interesting.
I hence recommend this manuscript to be published in Communications Earth and Environment, in a form close to this one.
I have a few minor questions that the authors could consider, which hopefully can help solving connected issues and extend the consequences of the study:
Some of the questions arising during the discussion are the following: the amount of carbon released during the dissolution is well established.
1. The techniques used by the authors should allow to see how much the different dissolution features identified contribute respectively to this mobilization: in particular, how much do the microsolution features contribute with respect to the large stylolites? Since structural geologist often do not consider the smallest features, the percentage to which they contribute with respect to ,the usually analysed large solution seams would be interesting to know.
2. The authors discuss the fact that large discontinuities can contribute to fluid mobility in subduction zones, and avoid reprecipitation of carbon in the surrounding porosity, while driving the carbon in species in solution up to the Sea. Can the authors evaluate the distance travelled by the carbon, based on their experimental techniques and/or on modelling, for the samples they study? What would be the contribution of joints in the transport of dissolved carbons, in addition to the dykes mentioned? When stylolites develop, the stress perturbation often leads to the formation of joints (see e.g.:
Ben-Itzhak, L. L., Aharonov, E., Karcz, Z., Kaduri, M., & Toussaint, R. (2014). Sedimentary stylolite

networks and connectivity in limestone: large-scale field observations and implications for structure evolution. Journal of Structural Geology, 63, 106-123.

Or

Aharonov, E., & Karcz, Z. (2019). How stylolite tips crack rocks. Journal of Structural Geology, 118, 299-307.
)

Can these structures contribute to the transport of the dissolved carbon, and to which extent?

Reviewer #3 (Remarks to the Author):

While this manuscript contains some interesting results, it is both far too specialized for a wider audience and contains numerous assumptions that aren't well supported, making it very speculative. The authors assume that all subjected carbonates are similar to the massive, shallow water carbonates they are investigating, when the literature describes a var wider (and still poorly understood) range of depositional forms for this mineral. The depositional environment will surely dominate the possible pathways for release of carbon via their process, from no release to full release. However a full breakdown of this matter would be beyond the scope of a paper in this Journal. The sediments that they are examining are hardly 'deep-sea', these are continental margin sediments undoubtedly (with both extensive sandstone and massive carbonate layers). Thus the whole area of basaltic-associated carbonates is not addressed nor mentioned. There is also a great deal of specialist jargon in the manuscript, making it a difficult read for non-geologists.

Re: COMMSEN-20-0508-T

Dear editorial team, dear reviewers,

We thank you and the two reviewers R2 and R3 for the comments on our manuscript. In the following, we address the reviewers' remarks in detail.

Thank you very much for your kind consideration,

With the best wishes from Australia

Christoph Schrank (on behalf of the author team)

Replies to Reviewer R2

R2: The techniques used by the authors should allow to see how much the different dissolution features identified contribute respectively to this mobilization: in particular, how much do the micro-solution features contribute with respect to the large stylolites? Since structural geologist often do not consider the smallest features, the percentage to which they contribute with respect to the usually analysed large solution seams would be interesting to know.

Answer: Thank you for this interesting suggestion. These large stylolites, shown in Fig. 1 of the article, are of the "simple wave-like type", using the recent classification of Koehn et al.[1]. However, the same authors demonstrate that this type of stylolite is "not useful for compaction estimates". Unfortunately, the rocks do not have suitable macroscopic markers for compaction estimates, and a comprehensive, systematic geochemical mass-balance study at the scale of the entire formation is a tremendous task well beyond the scope of the present paper. As a result, we only offer a very conservative estimate for the minimum amount of compaction accommodated by these features in our results section, based solely on wave amplitudes: ~ 20%. We realise that this estimate is possibly too low, considering that we occasionally observe very thick macro-stylolites such as the one shown in Fig. 1b – which may indicate complete loss of a dissolving limestone bed. Nevertheless, without much further fieldwork – sadly impossible at the moment – we cannot provide a better estimate. However, we also now discuss a related study that looked at large stylolites in the stratigraphic onshore equivalent of the studied limestones in the Canterbury basin. This study estimates a volume loss between 7 and 12% accommodated by large stylolites, very similar to that accommodated by our micro-seams.

R2: The authors discuss the fact that large discontinuities can contribute to fluid mobility in subduction zones, and avoid reprecipitation of carbon in the surrounding porosity, while driving the carbon in species in solution up to the Sea. Can the authors evaluate the distance travelled by the carbon, based on their experimental techniques and/or on modelling, for the samples they study?

Answer: To our knowledge, the most successful chemical tracers for quantifying mass fluxes in rocks and sediments are isotopes[2]. The XFM method cannot measure isotopes, and we did not attempt to conduct a bulk-rock or in-situ isotope study with laser-ablation mass spectrometry. The reasons: the bulk rock method is not suitable because we cannot selectively sample material from within and outside micro-dissolution seams. In-situ laser ablation has spot sizes on the order of 20 µm, which is two to four times larger than the width of a micro-seam and thus also not suitable.

We agree that a modelling study would be very interesting but note that it is beyond the scope of the current paper. A modelling study would be very challenging because one would need to employ a coupled model of fluid flow, chemical reactions, and mechanics (and perhaps heat flux) to capture the essential physics underpinning micro-seam formation and growth. These coupled models constitute the forefront of modelling research in solid-earth science[3,4] and therefore require substantial research effort. We hope that our unique microchemical dataset will inspire modelling studies down the track. Our research team currently explores the notion of cross-diffusion waves[5,6] to explain localisation structures such as the examined micro-seams. However, this is work in progress and will require standalone publications.

R2: What would be the contribution of joints in the transport of dissolved carbons, in addition to the dykes mentioned? When stylolites develop, the stress perturbation often leads to the formation of joints […]. Can these structures contribute to the transport of the dissolved carbon, and to which extent?

Answer: Thank you for this excellent suggestion. We now treat this suggestion in detail in our discussion (augmented by ca. 40%) and include the suggested references. In a nutshell: there is no observational evidence that cogenetic veins formed during micro-seam activity, and thus we rule out that veins acted as major vertical fluid pathways together with micro-seams. However, if thin, transient joints formed together with the micro-seams cannot be assessed. We did not observe any but note that their preservation potential would be very low.

Replies to Reviewer 3

R3: While this manuscript contains some interesting results, it […] contains numerous assumptions that aren't well supported, making it very speculative.

Answer: We provided strong supporting evidence for our conclusions and even add new data to the revised paper from additional competitively acquired beamtime at the Australian Synchrotron. Furthermore, R2 in their well-reasoned and referenced review clearly disagrees with R3: "*The analysis, carried out on a significant set of rock thin sections, is sound and clearly presented. The results are clear, well presented, and the discussion and consequences for the main question are interesting. I hence recommend this manuscript to be published in Communications Earth and Environment, in a form close to this one.*"

R3: The authors assume that all subjected carbonates are similar to the massive, shallow water carbonates they are investigating, when the literature describes a var wider (and still poorly understood) range of depositional forms for this mineral.

Answer: This assertion is incorrect. First, we do not claim that "all subjected carbonates are similar to the massive, shallow water carbonates [we] are investigating". Second, we do not claim that the samples we study are shallow-water limestones. There is consensus in the existing literature that the studied limestone is a deep-sea limestone[7,8]. This interpretation is consistent with our own field observations and the tectonic context of the studied basin[9]. Please, compare our detailed response below. Third, although it is fair to say that deeper marine carbonate depositional environments are less well understood than shallower environments, the overall field constitutes a mature research field[10] that provides abundant context for our new findings.

R3: "The depositional environment will surely dominate the possible pathways for release of carbon via their process, from no release to full release. However a full breakdown of this matter would be beyond the scope of a paper in this Journal.

Answer: The main point of our work is to report on the surprising discovery of the micro-seams and to estimate how much carbon they can release. We achieve this for the first time because of our world-first use of advanced synchrotron techniques, capable of quantifying elemental composition on the micro-scale on centimetre-scale thin sections. We then proceed to use existing, well cited modelling and field evidence to estimate the depth of formation for the micro-seams before we discuss, again conservatively and carefully, where the dissolved carbon could go. In fact, we state already in the abstract that the carbon could be locked in completely as cement or, the other endmember, leave the sediment completely when advective drivers operate. Therefore, the reviewer reiterates our own points, which does not constitute a controversy. Please, also note that we augmented the discussion carefully by about 40% to elucidate this point better.

We agree that further research is needed. Spurring such work is one of the main aims of the paper: to stimulate further work on micro-seams. Thanks to our paper, we now know that they exist and play an important role in mobilising carbon. As noted by the reviewer and in our discussion, the fate of that carbon will vary depending on individual settings, and each will require additional work. That work will be made possible by the present paper where we also provide a unique methodology that can help to answer all the exciting new questions our work raises.

R3: The sediments that they are examining are hardly 'deep-sea', these are continental margin sediments undoubtedly (with both extensive sandstone and massive carbonate layers). Thus the whole area of basaltic-associated carbonates is not addressed nor mentioned.

Answer: As stated above and detailed in the Appendix below, these rocks are classified universally as deep-sea limestones[7-9]. The reviewer does not provide any arguments or reference to literature that refutes this classification. It is unclear why or how "basaltic-associated carbonates" are relevant, as the point of our work is that significant carbon may be mobilised during carbonate deposition, regardless of the substrate upon which it is deposited. The substrate is not a factor in the processes we have elucidated. These shallow-subsurface and syndepositional-compaction processes are expected to be widespread across a broad range of deeper carbonate sediments because there is no evidence to suggest that the studied rock package is, in any way, special.

R3: There is also a great deal of specialist jargon in the manuscript, making it a difficult read for non-geologists.

Answer: This is a subjective, unsubstantiated statement. Our team consists of physicists, structural geologists, geophysicists, and sedimentologists, all of whom contributed to writing the draft. Therefore, we believe that the current choice of language would be accessible to a diverse readership. Nevertheless, if specific examples of unclear passages were pointed out to us, we would be happy to consider alternative approaches to writing. Language can be fixed easily.

Appendix: Were the studied limestones truly deposited in the deep sea?

The most comprehensive and recent sedimentological analyses of the studied Paleocene limestone series by Frank Chanier and co-workers[7,8,11] in the 1990's designate these rocks as pelagic (i.e., "deep sea") in origin. This classification is consistent with older work from the 1960's. For example, Eade wrote that "sedimentation during Upper Cretaceous and Tertiary times […] took place almost entirely in deep water" in the research area[12]. This interpretation is consistent with the tectonic context that clearly shows that the studied limestones are overlain by turbidites "deposited on a deep sea plain"[13]. The most recent literature on the sedimentation and tectonics of the research area in the 2000's also designates the limestone series as "marls and pelagic limestones"[9,14].

Our own field studies did not provide a compelling reason to disagree with > 50 years of literature consensus on the depositional environment of these rocks. The intercalated siliciclastic beds in the limestone series represent episodic deep-sea turbidite deposits, as indicated by the typical Bouma-type sedimentary structures, which was already recognised as early as 1957 (see Waterhouse and Bradley[15], or Browne[16]), and the trace-fossil assemblage (Fig. I). The trace-fossil assemblage consists of the Nereites ichnofacies, which is common in bathyal to abyssal depositional environments characterised by oxygenated deep-marine environments with slow rates of continuous sediment supply from suspension. These conditions are commonly interrupted by periodic turbidite deposits sourced from the continental shelf. Between turbidite events, traces represent complex and delicate feeding and welling structures below the sediment/water interface (graphoglyptids)[17-19]. Traces identified in the Pahaoa area include: *Chondrites* (Ch), *Cosmorhaphe* (Cr), *Helminthorhaphe* (He), *Lorenzinia* (Lr), *Nereites* (Ne), *Paleodictyon* (Pd), *Scolicia* (Sc), *Spirophycus* (Sh), *Spirophyton* (Sp), *Spirorhaphe* (Sr), *Thalassinoides* (Th), and *Zoophycos* (Z). The high diversity of nerites trace fossils in conjunction with the sedimentary evidence demonstrate that the intercalated siliciclastic deposits are turbidites in a deep-water environment (Fig. I). Moreover, smectite is the most common clay mineral within the studied limestones and the covering mudstone sequence[7,8]. This clay mineral is characteristic of distal, deep-marine depositional environments[20].

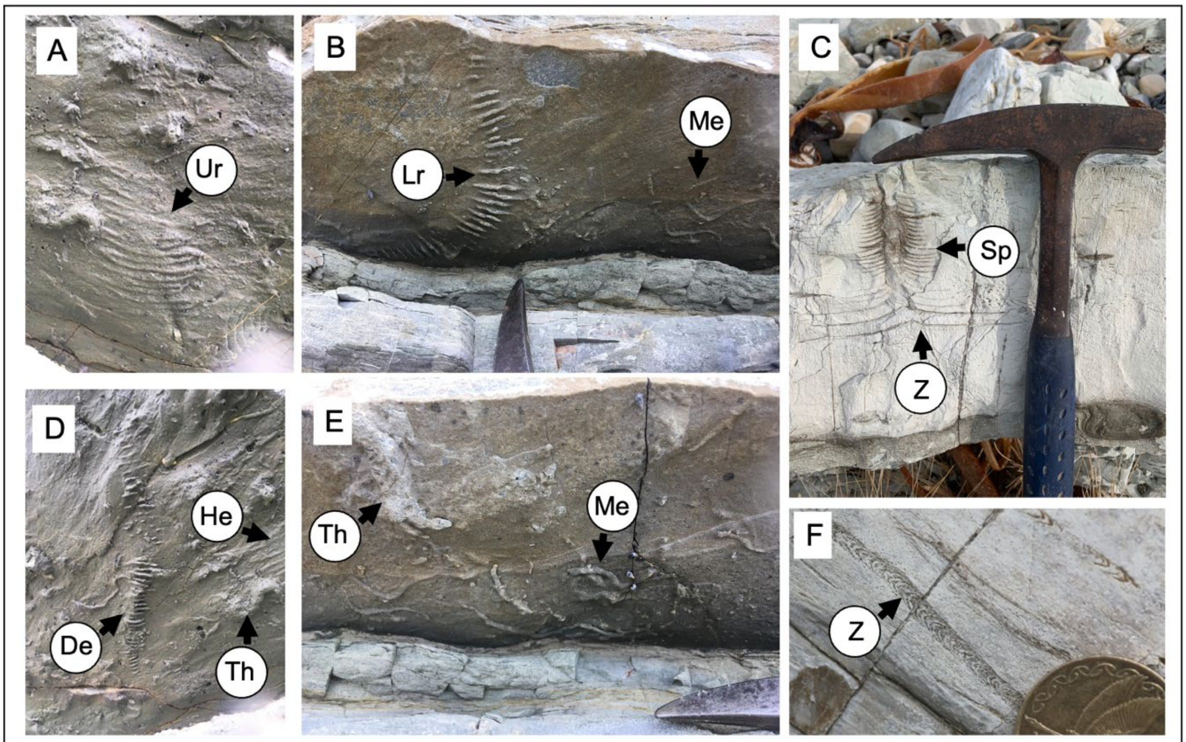

Figure I. Common ichnofossils preserved in the siliciclastic and carbonate intervals representing a Nereities ichnofacies. A: Urohelminthoida (Ur) : Desmograpton (De), Helminthopsis (He), Thalassinoides (Th); B: Lorenzinia (Lr), Megagrapton (Me), Urohelminthoida (Ur); C: Zoophycos (z); Spirophyton (Sp), D: Desmograpton (De), Helminthopsis (He), *Thalassinoides* (*Th*) : E: Megagrapton (Me), Thalassinoides (Th); F: Zoophycos (Z)

Finally, we emphasise that the most important physical property of the studied rocks is that they largely consist of (now lithified) carbonate mud. Our study clearly demonstrates (see above) that this carbonate mud matrix, the micrite, is affected by micro-dissolution during pre-burial diagenesis. We have no indication that our rocks are in some way "special" in terms of their physical dissolution behaviour and can thus confidently predict that other marine sediments largely made of carbonate mud – namely other deep-sea carbonate deposits – should experience diagenetic micro-dissolution of the mud. One of the most important aspects of our study is that we employ, to our knowledge, the only suitable microanalytical method (high-resolution synchrotron XFM) that can make these tiny dissolution seams visible – and also permits us to quantify how much calcite was dissolved and thus determine how much carbon was lost.

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29th Jul 21

Dear Dr Schrank,

Please allow me to apologise for the delay in sending a decision on your manuscript titled "Cryptic micro-dissolution mobilises carbon in Earth's largest carbon reservoir". It has now been seen by three reviewers - the original Reviewer #2 and two new Reviewers - whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License) provided you acknowledge the limitations associated with the small number of samples from a single location and caveat your conclusions accordingly. Please also consider using the term 'sink' instead of 'reservoir' when referring to the 'largest carbon reservoir'.

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,

Mojtaba Fakhraee, PhD
Editorial Board Member
Communications Earth & Environment
orcid.org/0000-0002-2461-6374

Joe Aslin
Associate Editor
Communications Earth & Environment

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

Second Review about “Cryptic micro-dissolution mobilises carbon in Earth’s largest carbon reservoir” by Schrank, C. E., Jones, M. W. M., Kewish, C. M., van Riessen, G. A., Elphick, K. E., Sloss, C. R., Nothdurft, L. D., Webb, G. E., Paterson, D. J., and Regenauer-Lieb, K.

In this manuscript, the authors analyse microdissolution features in pelagitic limestones, and analyse the budget of carbon released during the process. This allows to discuss the fate of this carbon in a common type of sedimentary rock during lithification and burying.
Deep-sea limestones are one of the largest carbon sinks in the Earth, and the rate of burial, and efficiency of the process in terms of carbon budget is a very important issue to understand the dynamics of atmospheric and oceanic carbon, which are issues interesting a wide range of researchers in Earth and environmental science.
The manuscript is clear and well-written, with issues of interest to many scientists in the field and beyond. The study is sound and convincing. The methods are original, based on a set of powerful Xray fluorescence microscopy and ptychography techniques to obtain microstructural analysis and

precise chemical elemental maps of their samples with a resolution around 100 nm to 1 micrometer, which allows to analyse microdissolution features often disregarded due to their small size. This allows to assess the enrichment in non-soluble species, such as Fe, with respect to the surrounding rock matrix. It allows the estimate of dissolved $CaCO_3$ during the formation of the solution seams. and offer new pathways to deal with such issues and perform studies on other locations or other types of rocks.
The answer of the authors and modifications address all points raised in my first review. The manuscript is of very high quality and reports both new discoveries of general interest, and a fruitful new methodology. I support the publication of the manuscript in the current form.

Reviewer #4 (Remarks to the Author):

I Keep the a neutral opinion for this paper. The analysis and results look OK, the only thing I concern about is the number of samples; such research always need very large number sample sets to exclude the heterogeneity. I do not believe the number of samples in this research is enough.

Reviewer #5 (Remarks to the Author):

COMMSENV-20-0508A-Z Review

The paper “Cryptic micro-dissolution mobilises carbon in Earth’s largest carbon reservoir” provides micro-scale XFM and nano-scale X-ray ptychography analysis of dissolution structures observed in samples of the deep ocean Kaiwhata limestone obtained in field work . By analyzing the samples with micro- and nano-scale techniques, the authors were able to observe micro-scale dissolution seams that are invisible in hand specimen. Mass balance indicates that there is an excess of carbon, which is either trapped in calcite cement or released in the ocean. This observation has implications on the carbon budget.

The authors have made substantial effort to provide answers to all concerns of reviewer #3 and therefore I think that the paper can be recommended for publication.

In contrast to previous reviewer #3, I do not find too much specialist jargon in the manuscript (and I am from a different discipline of geoscience). I find the balance between communication to a broad audience and specificity in stating research findings is well done.

Comments:

R2: The authors discuss the fact that large discontinuities can contribute to fluid mobility in subduction zones, and avoid reprecipitation of carbon in the surrounding porosity, while driving the carbon in species in solution up to the Sea. Can the authors evaluate the distance travelled by the carbon, based on their experimental techniques and/or on modelling, for the samples they study?
Answer: To our knowledge, the most successful chemical tracers for quantifying mass fluxes in rocks and sediments are isotopes[2]. The XFM method cannot measure isotopes, and we did not attempt to

conduct a bulk-rock or in-situ isotope study with laser-ablation mass spectrometry. The reasons: the bulk rock method is not suitable because we cannot selectively sample material from within and outside micro-dissolution seams. In-situ laser ablation has spot sizes on the order of 20 µm, which is two to four times larger than the width of a micro-seam and thus also not suitable.

For future work, it might be worth to look into atom probe tomography and if it can be used to analyze the isotopic composition inside/outside the dissolution seams.