

## Response to Referee no. 1:

Dear Referee no. 1,

we would like to thank you for your very accurate and constructive revision of our manuscript. We appreciate the time and effort that you and referee no. 2 have dedicated to providing your valuable feedback on our manuscript. We are able to include most of your corrections and suggestions and are sure that they will improve our manuscript. The changes will be highlighted in the revised manuscript.

Please find below our detailed responses to your individual comments.

*(Reviewer comment; Author's reply)*

*1) Title : I find the title misleading as it suggests that the observed effect is caused specifically by biotite. However, the authors seem to argue that it is not specifically the biotite that causes the observed effect but initially (?) reduced porosity in the biotite-bearing layers. If they are right, then a biotite-free but denser salt layer in the sample would show the same layer-scale mass transport phenomenon reported here and described by Merino et al. I would recommend a title more along the lines of "CT imaging demonstrates interlayer mass transport in layered halite-biotite aggregates undergoing dissolution-precipitation creep".*

We agree with the reviewer's suggestion. The original title suggests indeed that biotite has a specific impact upon the transport length scales of DPC. In the manuscript we argue that the observed effect is rather a consequence of textural heterogeneity and porosity fluctuations between biotite-bearing layers and pure NaCl layers. However, as we discuss in comment 4, the differentiation between the phyllosilicates involved as well as the rate controlling process seems to be important when discussing phenomena related to DPC. Therefore, the revised manuscripts will be titled "Biotite supports long-range diffusive transport in dissolution-precipitation creep in halite through small porosity fluctuations"

*2) Abstract, lines 5-6 read: "We used time-resolved (4D) microtomographic data to capture the dynamic evolution of the transport properties in layered NaCl-NaCl/biotite samples". This is not true. No attempt was made to calculate transport properties (or measure them). Only porosity evolution was studied. Best correct to porosity rather than "transport properties" – throughout the ms.*

Thank you for pointing this out. The reviewer is correct as transport properties are by definition: conductivity, diffusivity, and viscosity. In our study, we did not measure those. Accordingly, throughout the revised manuscripts, the term "transport properties" will be replaced by "porosity" or "hydraulic properties" where a more general view is emphasized.

*3) Abstract, lines 12-14 reads: "We propose that, in our experiments, the diffusive transport processes invoked in classical theoretical models of DPC are superseded by chemo-mechanical feedbacks that arise on longer length scales." This cannot be said if in the main text it is claimed that the sample scale compaction behaviour is consistent with compaction experiments on pure NaCl. The effect of interlayer transport in the present experiments is argued not to influence overall compaction strain,*

*so it does not dominate over pressure solution as a deformation mechanism, it merely contributes and dominates porosity reduction in the biotite-bearing layers.*

We agree with the reviewer and will replace “superseded” with “complemented” or an equivalent synonym in the revised manuscript to address the contributing character of the process.

*4) Introduction, lines 29-30 read: “Phyllosilicates have been recognised to have a reinforcing effect on the dissolution process ..”. Yes, a but others have notes that pressure solution (compaction) can be inhibited or unaffected by phyllosilicates, e.g. Niemeijer & Spiers (2002). The enhancement effect comes mainly from observations on natural rocks where advective mass removal along phyllo-rich layers cannot be eliminated as playing a role.*

We agree that this is a crucial question that needs to be addressed. However, we think that a detailed discussion is beyond the scope of this manuscript and may require another study or publication that focuses on the effect of different types of phyllosilicates upon the three types of DPC systems (dissolution-, diffusion- or precipitation-controlled). A general statement like “all phyllosilicates have the same effect upon all types of DPC systems” is probably too broad and needs to be tested in a series of scientific experiments.

As pointed out by the reviewer we agree that observations from natural samples always include an effect of advective flow and mass transport. However, experimental studies by e.g. Renard et al. (2001) show as well that the presence of clay enhances the DPC rate in a closed NaCl system. As DPC in NaCl is diffusion-controlled (see Spiers et al. 1990.) it makes sense that a phyllosilicate that can incorporate an additional layer of water into its structure (like clay minerals can do) provides a good diffusion pathway, hence, accelerates the DPC rate.

In contrast to the diffusion-controlled NaCl system, Niemeijer & Spiers (2002) investigated muscovite + quartz which is under the experimental conditions of 500°C identified as a dissolution-controlled system. Here, the authors argue themselves that Al<sup>3+</sup> is expected to decrease the solubility of quartz hence, impede DPC. It would be interesting to see if a low Al mica like biotite has the same effect upon DPC in quartz.

*5) Intro, lines 30-35: What does the present study actually add to the paper by Macente et al (2018)? Would be wise to make this clear somewhere, e.g. in lines 46-48. Just seems like a technical refinement at present.*

While we appreciate the reviewer’s feedback, we respectfully disagree that this study is just a technical refinement of Macente et al. (2018). We think this study makes a valuable contribution to the field because it uses technical advances in form of an evolved experimental setup as well as advanced analysis techniques and codes. Analyses that distinguish out study from Macente et al. (2018) only became available recently and allow for example to track the NaCl redistribution. In addition to that, while our study focuses on the qualitative analysis of transport length scales during DPC, Macente et al. (2018) emphasize the impact of phyllosilicates upon the evolving porosity.

Scientific progress sometimes happens by groundbreaking discoveries but most of the time carefully planned small steps prepare the ground for innovation. We acknowledge that our study may fall into the latter category however, even testing the fundamental observations

by Macente et al. (2018) and confirming the results should be regarded as a successful step towards understanding the process of DPC. We believe that a problem exists in geosciences which is that experiments are often run once and never again. Hence, most discoveries lack replication and confirmation by subsequent studies either through technical difficulty, expense, or because of data handling limitations.

6) Intro lines 48-49 read: *“Our aim was to determine length scales of diffusive transport in a dynamically evolving porosity during DPC”. What about trying to explain them??*

We will rephrase the sentence in the revised manuscript and will add an explanatory part.

7) Section 2, line 53. *Peach and Spiers 1996 is a study of the percolation threshold in dilating salt, not a study of deformation mechanisms. A far more relevant reference here and in line 57, would be the study of pressure solution in compaction by Spiers et al 1990, which specifically addresses the creep law for pressure solution in NaCl in 1D compaction and deviatoric creep – and emphasizes the analogue aspect.*

Thank you, for pointing this out. We have changed the reference accordingly as we appreciate the relevance of Spiers et al. 1990 in this context.

8) Section 2, lines 58-59 read: *“It is further a material used in geological nuclear waste repositories (Powers et al.,1978), and its deformation behaviour is well-characterised”. Salt is not a material used in radioactive waste repositories – it has been and still is widely considered as a HOST ROCK for repositories. A more recent ref than Powers should be added and refs should be added to underpin “well characterised”. Urai , Schleder, Spiers and Kukla 2008 would be suitable here.*

We agree with the reviewer that salt is considered as a host rock for nuclear waste repositories rather than it is a material used in these repositories. The sentence will be rephrased to "It is further considered as a host rock for geological nuclear waste repositories (citations) and its deformation behaviour is well-characterised (citations)". Further, relevant references will be added.

9) Section 2.3 Experimental setup, lines 95-96 reads: *“The experiments were run inside a thermally insulated box where the temperature was logged and found to be stable within  $\pm 1.7$  °C over the course of the experiments.” This is quite a large range in T for such a soluble material as NaCl (which would certainly cause sample-wide dissolution-precipitation effects) and raises questions regarding temperature GRADIENTS in the sample and their possible effect on convection and advective transport. Was temperature measured at different points along the length of the sample and if so what was the T profile or gradient? Could this have driven advective transport in the samples? Some calculation is needed to answer this. Of further interest here is the possible effect of differential heating of the sample during CT-imaging, as a result of x-ray attenuation – e.g. differential heating of biotite-bearing versus pure NaCl layers. Can effects such as this be eliminated?*

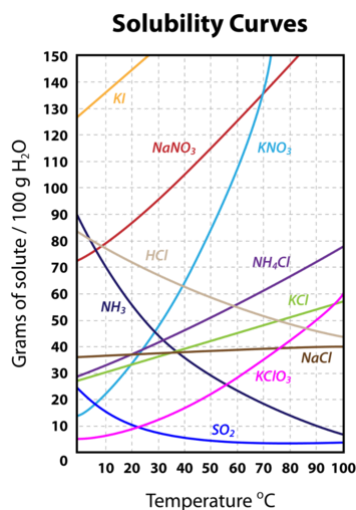
Thank you for pointing this out. Although we agree that this is an important consideration, it is not appropriate for inclusion in this manuscript because the solubility of NaCl is relatively unaffected by the temperature in the range of  $\pm 1.7$  °C at room temperature (see figure

below). The solution of NaCl is only a very low endothermic reaction, therefore temperature does not affect the solubility as it would e.g. KCl.

In addition to that, we consider the peek cell surrounding the samples as an additional thermal insulation layer due to its low thermal conductivity ( $0,25 \text{ W m}^{-1} \text{ K}^{-1}$ ). Hence, temperature changes inside the sample cell are expected to be below  $1 \text{ }^\circ\text{C}$  and therefore negligible for changes in the solubility of NaCl. Unfortunately, we were not able to measure a temperature gradient along the length of the sample but following up from the argumentation above, a measured gradient would have been too low to drive convection or advective transport.

Differential heating of layers with different composition is a function of the dose and the specific heat capacity of the minerals involved. In the case of NaCl and biotite the specific heat capacity is  $c=0.88 \text{ kJ/kg}\cdot\text{K}$  and  $c=1.035\text{-}2.064 \cdot 10^4 \text{ kJ/kg}\cdot\text{K}$ , respectively. From a theoretical point of view this may cause differential heating by x-ray radiation hence, a temperature gradient within the sample between layers that contain biotite and those that do not.

However, even at high doses at synchrotron facility the increase of temperature of geological samples is in the range of single digits. We would expect to see an even lower effect in lab based x-ray CT scanners. The x-ray source used for the scans was operated with a target power loading of 2.8 W. Compared to e.g. the Diamond Light Source (I12) which at 53 keV and 300 mA ring current would be about 16 kW, our source is a dim heat source. In addition to that, the biotite-bearing layers contain 20 wt% of biotite, are therefore dominated by NaCl which itself will exchange heat with neighbouring biotite-grains. The temperature gradient that can be achieved between the mixed and the pure NaCl will be limited and negligible.



(From: CK-12 Foundation – Christopher Auyeung)

10) A further point related to the above is the issue of radiation damage and its effect on NaCl solubility. Recent measurements that I have witnessed in a similar scanner show heating of NaCl by a few degrees accompanied by significant radiation damage of the salt – it turns yellow or purple at high doses. So my question to the present authors is: did the samples change colour after CT scanning? Did they check? And, if the colour did change, can they eliminate the possibility of damage gradients influencing dissol-precip transfer between layers of different composition hence different damage in the NaCl? Note that from a theoretical point of view, if the deposited energy due to radiation damage of NaCl is  $E$ , the increase in solubility for small  $E$  is  $100.E/RT \%$ . Could this effect, or the heating due to attenuation, be significant?

Thank you for pointing this out. We are aware that radiation damage may occur in the samples. The described change in colour was observed in NaCl recovered from the sample cells. Source of this changes in colour, from white to yellow or even purple, is the formation of F- and H- centres in the anion lattice of the crystal. As the bulk sample experienced a similar dose, we do not expect a lateral damage gradient to emerge and drive mass transfer from higher damaged domains (increased solubility of NaCl) to lower damaged domains. We would also like to point out, that the experiments conducted by Renard et al. (2003) at a synchrotron source do not report any significant effect of radiation damage on the solution rate. Compared to these data, our samples were irradiated at much lower doses and only for a short amount of time (1hr).

We will acknowledge the problem of irradiation damage with an addition to our revised manuscript.

*11) Also under Section 2.3, it is mentioned in line 98 that the applied effective stress on the compaction experiments was 6.64 to 10.5 MPa. That means that local stresses at NaCl and NaCl-biotite grain contacts would have been much higher – in the range 12 to 50 MPa. These stresses are well inside the regime where salt deforms plastically at room T, leading to a coupling between work-hardening plasticity on the grain scale and dissolution-precipitation transfer, as opposed to classical pressure solution seen in compacting NaCl at stresses below 3 -4 MPa (see Urai et al 2008 above; also Spiers & Brzesowsky. *Densification behaviour of wet granular salt: Theory versus experiment. Seventh Symposium on salt 1, 83-92, 1993*). The likelihood that this plasticity-coupled mechanism played a role in the present experiments, rather than classical pressure solution, should be pointed out, especially as it is a mechanism where pore volume diffusion plays a role as opposed to the grain boundary diffusion process that controls “normal” pressure solution.*

We appreciate the reviewer’s feedback and will acknowledge this limitation in our revised manuscript. Although, we agree that locally plastic deformation may have occurred at small contacts where stress was concentrated, the effect upon the bulk deformation was negligible. One reason why DVC works in our experiments is that the centres of the NaCl grains did not deform, hence deformation must have happened at the grain boundaries and not within the grain.

*12) Section 3 Results, Figure 3. The apparently straight portion of the compaction curves shown in this plot is referred to by the authors as steady state creep, whereas the inset in the Fig clearly shows that the strain rate is continuously decreasing within the resolution of the data. Moreover, the authors actually say that the compaction curves show asymptotic behaviour (e.g. line 271), which in itself means that steady state is not achieved. In addition, it is quite impossible to reach a steady state compaction rate in a compaction experiment of any kind, as porosity is continuously decreasing and therefore so must the strain rate – regardless of deformation mechanism. In this study, apparent steady state seen in the compaction curves is an artifact of the few, rather scattered strain-time data (clearly understood from the inset in Fig 3). Perhaps use of the term “apparent steady state” would be acceptable, but the term steady state creep should be removed throughout and all related points corrected accordingly.*

As suggested by the reviewer we have corrected in the revised manuscript the term “steady state” to “apparent steady state”.

13) Figure 7. Lines 223-224: "Figure 7 shows the vertical displacement rate of the biotite-bearing layer and the bulk sample for different increments of progressing deformation". And in Lines 225-226 "At the beginning of the experiment the rate of both bulk samples was elevated compared to the biotite-bearing layers." OK for the displacement rates, but any meaningful comparison requires normalization with respect to the thickness of the NaCl and NaCl-biotite layers considered, i.e. the average strain rates in each zone should be plotted versus compaction stage (time proxy). This is crucial because of later discussion around the issue of enhanced compaction (lower contact stresses) causing interlayer mass transfer.

We chose to plot the vertical displacement rate instead of the average bulk strain rate as the latter would give a similar result to the DVC analysis. While the average bulk strain rate of a layer would describe the magnitude of strain, DVC gives strain resolved on the grain scale which we believe is a better measurement to analyse strain localisation and enhanced compaction.

14) Section 3.2 Strain analysis. The usage of the terms volumetric strain (isotropic), deviatoric strain and compaction strain becomes a bit confused from here on, I feel. In 1-D, compaction strain is equal to volumetric strain, but not equal to the isotropic strain component of the strain tensor of course. However, the isotropic vol strain does seem to be referred to as compaction at some points in the ms. Somewhere early in the ms, these terms need to be strictly defined and differentiated from each other, and then used consistently. It is also important to note that deviatoric strain cannot occur during 1D compaction independently of the isotropic component of volume reduction, because the pressure solution process (even when accompanied by plasticity) is serially coupled to intergranular sliding – you cannot have one without the other (in pure NaCl or in NaCl-biotite mixtures). In isotropic compaction under 3D loading with  $S_1=S_2=S_3$  you can get compaction with little or no intergranular sliding.

Thank you for pointing this out. We agree that these terms need to be defined in the manuscript. Accordingly, we will add those changes to the methods section. We further emphasize the differentiation between macro- and micro-strain. While the vertical shortening and compaction of the bulk sample refers to the macro-strain, isotropic (volumetric) and deviatoric strain refer to strains resolved on the grain scale. Here, the volumetric strain is the first invariant and isotropic strain component of the strain tensor. The deviatoric strain is equal to the second invariant of the strain tensor and describes deformation at constant volume.

We would like to point at that the term „compaction strain“ was not used in our manuscript.

15) The above point comes into play in Figs 8-11, where isotropic volumetric strain (called volumetric strain) is used as an indicator of compaction, whereas macroscopically measured compaction is 1-D compaction. I would strongly advise the authors to present a complete picture in Figs 8-11 by adding contour plots of vertical compaction strain, in addition to the isotropic vol and deviatoric strains. This would make what is going on clearer with a complete set of all information.

As suggested by the reviewer we will adjust figures 8-11 by adding a vertical shortening component.



*16) Section 3.4 NaCl redistribution, Fig 13 and text referring to it (e.g. lines 254-255). Here, changes in NaCl content within the samples are specified per horizontal slice through the sample. That should be made clearer in the text as it reads as though the mass of the samples is not constant. That also raises the question as to whether the mass of NaCl in the samples is indeed constant. Do the changes in NaCl mass/vol fraction seen in individual samples add up to the original NaCl solid mass? This needs to be clarified.*

We agree with the reviewer and will rephrase the text to clarify the case. An increase in NaCl could indeed be an addition of NaCl from an outside source. This is not the case in our samples. Instead, the total NaCl content of the samples remains constant throughout the experiment.

We will add a measure of the total NaCl in the samples and give mass balance calculations to prove that no additional NaCl enters the sample and the pure redistribution of NaCl was observed.

*17) Section 4 Discussion, lines 269-270 read: "The general compaction behaviour we observed was consistent with previous studies on NaCl compaction". Well, yes, the data do show increasing compaction with time. But that is no basis to claim consistency with previous work. First, no other compaction data on salt show the apparent steady state portion claimed by the authors, so they are not qualitatively consistent. Second, a comparison with the isostatic compaction tests of Schutjens & Spiers is not expected to be consistent because of the different boundary conditions imposed. Third, no evidence is presented that the present amounts and rates of compaction are consistent with previous 1D compaction tests on samples of controlled grain size, such as those reported by Spiers et al (1990 – low applied stresses) or Brzesowsky and Spiers (1993 – stresses similar to the present). To claim any consistency or detect any interesting differences, a quantitative comparison should be made by adding a few curves from previous 1D compaction studies on salt of the same grain size – or calculating compaction curves for the present conditions from the compaction data or laws given by previous authors.*

We agree that claiming "consistency" of our results with previous work requires a proof which we do not establish in the original manuscript. In the revised manuscript we will therefore exchange the term "consistency" with "in accordance with".

A direct comparison of our results with previous studies is difficult as grain sizes and deformation conditions vary between the individual studies. According to the rate law by Spiers et al. (2004) the strain rate in diffusion-controlled systems is affected by the grain size, the applied effective load, the temperature and the porosity. Further, the duration of the here presented experiments exceeds the duration of most published data.

Hence, compaction curves could be calculated based on the rate law by Spiers et al. but previously published data should not be plotted for direct comparison.

We will follow the suggestion by the reviewer and add a calculated compaction curve to our data plots in the revised manuscript.

*18) Lines 272-276. The authors claim a change in deformation mode beyond 200 hours here. But they also argue that their data are continuous and show a continuous asymptotic decrease in strain rate. The continuous nature of their strain rate data is also apparent from Fig 3 (see point 12 above). It does not seem justified then to claim a change in deformation mode here, so the point should be removed or weakened.*

We agree with the reviewer, the two stages of the process are rather related to a transition from a loosely packed aggregate where mechanical compaction significantly contributes to the strain rate, to an interlocked aggregate dominated by chemical compaction. In this case mechanical and chemical compaction are active simultaneously throughout the experiment but dominate at different times depending on the state of the aggregate. We will add a clarification to the revised manuscript.

*19) Lines 305-306 read: “the upper NaCl layer did develop a pronounced gradient towards the interface with the biotite-bearing layer though, which could be evidence for a diffusive salt redistribution”. Yes agreed. But it could also be evidence of advective redistribution if there were even small internal T-gradients. Can this possibility be eliminated? If not that should be stated.*

Thank you for pointing this out. As discussed for comment 9 we think that internal temperature gradients are negligible.

*20) Lines 309-319: Here it is proposed, quite reasonably, that Merino’s model of diffusion from more porous to denser layers may occur because of a higher solute concentration (supersaturation) in the more porous material than the denser material. This is consistent with pressure solution theory and fine. However, appealing to the high supersaturations discussed by Desarnaud et al (2014) or Zimmerman et al (2015) is misplaced here as these are concerned with pre-nucleation supersaturations. There is no evidence for a nucleation stage in the present experiments as it is quite clear from the grain scale images, and from previous compaction work on NaCl, that precipitation occurs mainly by OVERGROWTH on the pre-existing grain (pore) walls. If fine grains are nucleated in the pores in the present experiments, that would be new and should be described. Only then should the above nucleation argument can be kept.*

We appreciate the reviewer’s feedback, but we respectfully disagree with the suggestion to remove the argument.

The reviewer’s statement that NaCl precipitation occurs by overgrowth as a simplistic process, ignores the fact that crystal growth mechanisms are varied and largely subject to supersaturation. In fact, the resolution of the CT data does not enable us to make any statement about growth mechanism at the NaCl surface. We chose these references as they confirm that it is possible to achieve a significant supersaturation of NaCl in solution without homogenous nucleation (which we do not observe). This is a prerequisite for long distance transport.

We agree that long distance transport is consistent with DPC theory (e.g. Gunderson et al 2002), but our argument, as presented, indicates that such behaviour is consistent with the specific material studied as well as theory in general.

*21) Lines 320-322. Here the authors argue that the Merino model may apply because the biotite-bearing layers compacted more than the pure NaCl layers in the early stages of the experiments, so had lower porosity, lower contact stresses and hence a lower supersaturation on NaCl in the pores – giving a driving force for diffusion of dissolved NaCl from the pure to the mixed layers. For the reader, however, this seems to be a strange statement after so much emphasis has been placed on the lack of evidence for any strain enhancement in the biotite-bearing layers (at many points, but also again in lines 336-337). The argument seems inconsistent. Can the authors please clarify this picture – it is*



*most confusing in the present form???? Was strain only uniform in the late stages but not initially? If so, please make this clearer.*

We will clarify this in the revised manuscript. The inconsistency and confusion results from an error in the wording. Instead of a higher compaction, the packing density of the biotite-bearing layers was greater than that of the pure NaCl domains. Hence, the porosity was lower. We suggest adding the initial porosity measurements in the results section to demonstrate that the lower porosity existed right from the beginning and is not an artifact of localized compaction in the biotite-bearing layer.

*22) In relation to the above point, I also wonder if the authors should mention the possibility that the preferential “cementation” of the biotite rich layers that they see could reflect an INSTABILITY caused by the Merino effect progressively reducing grain contact stresses and supersaturation in the biotite layers faster than in the NaCl layers.*

We will add this to the revised manuscript. We assume the reviewer means an instability that is the reinforcing counter part to the stress increase in high porosity domains in the Merino feedback mechanism.

*23) Line 331. The authors suggest here that electrochemical effects at the NaCl-biotite interface may enhance dissolution at those sites, following the references cited. However, as far as I recall those refs deal with the effects of micas at mica-quartz interfaces. I do not think one can then assume that the same enhancement effects will occur at a mica-salt (ionic solid) interface. Line 331 should read “.....which MAY accelerate dissolution of NaCl.”.*

Line 331 will be updated as suggested. It is correct that the cited literature deals with mica-quartz interfaces. However, the authors already argue themselves that their results may be applicable to dissimilar interfaces in general.

*24) Lines 336-334: This explanation of what goes on inside a biotite-bearing layer is reasonable. However, is it not a remarkable coincidence that “the additional NaCl contributes to a load-bearing framework whose compaction rate is in sync with the bulk sample’s”??? Would it not be better (i.e. more accurate) to replace “is in sync with” by “roughly matches” ?? Otherwise there would have to be some strong coupling which is hard to argue.*

As suggested by the reviewer we have replaced “is in sync with” by “approximates”.

*25) Lines 374-386. The issue of volumetric strain versus compaction strain versus isotropic strain raises its confusing head here again, further underpinning the need for better definition of these terms at an early stage in the paper, followed by consistent use in a way that distinguishes between physical compaction and the math properties of the isotropic part of the strain tensor – see point 14 above. MORE INTERESTING though is the issue of what was observed in the glass bead layers in the biotite-bearing samples. Presumably there was no actual compaction of these layers, beyond some rearrangement effects or possible bead breakage or chipping. This should be clarified in the Results. There, it should also be made clear whether there was any precipitation of salt in the bead layers. If there was at both sample ends, this would support the Merino model, as there would be no*

*stress-induced supersaturation in the brine in the pores between beads. If there was precipitation between beads at one end of the sample but not the other, this would suggest a role of convection and advective transport, or double diffusive convection. If there was no precipitation at all between the beads, this could be explained by the nucleation barrier at these sites – thus supporting neither the Merino model nor an advective transport model.*

As suggested by the reviewer we will add a paragraph about the glass beads in the result section. In the SBS sample which kept a connected porosity throughout the experiment no nucleation of NaCl was observed in either of the two glass beads layers indicating a nucleation barrier. In the second biotite-bearing sample (SB) we observed indeed the nucleation of an NaCl crystal after the porosity breakdown in the biotite-bearing layer and the loss of connection to the pure NaCl layer. We will explore the origin of this nucleation and possible shift of the Merino effect.

*26) The issue of the glass beads does raise the question of why the authors did not do an additional control compaction experiment with a layer of denser NaCl instead of a layer containing biotite? This would more rigorously test whether the Merino model may apply, i.e. whether diffusive transport is caused by porosity hence supersaturation differences, as opposed to some special effect of biotite. This would be a worthwhile addition to the paper, if time and money allow - as would an experiment substituting calcium fluoride cleavage flakes for biotite flakes. That would be useful because the diffusive properties of NaCl-CaF<sub>2</sub> interfaces have been directly measured during active pressure solution of the NaCl by De Meer et al (2002 EPSL 200).*

Thank you for this suggestion. It would have been interesting to explore this aspect as discussed by Schwichtenberg (2021). However, the time frame to run an experiment comparable to the presented experiments would exceed a feasible revision time. Each experiment ideally runs for ~ 2000 hours (~ 3 months) in which the in-house CT scanner has limited availability (e.g. we are restricted to our transmission source which precludes large sample scanning). In addition to that, we need to run time consuming image analyses which add a few months to the total time required to get meaningful results.

We agree that there remain interesting questions challenges and which should ideally be addressed in future experiments!

#### **TECHNICAL ISSUES (language, typographics etc)**

Overall the paper is well written and in good English. Nonetheless a few small improvements can be made as follows:

1. i) an asterisk \* is not a mathematical symbol. Proper multiplication and scalar, vector or tensor product symbols should be used.
2. ii) Figures 8-11 would benefit from an explicit indication of which sample is being displayed.

The technical issues will be resolved in the revised manuscript as suggested by the reviewer.