We thank the referee for their helpful, constructive comments, which enabled us to reconsider our interpretations, discuss some details more clearly, and eliminate relevant problems.

The red sections comprise the comments proposed by Reviewer 1; our responses are outlined in black; *and the modified portions of the manuscript are shown in italics.*

– Specific comments –
Line 113: The molar concentration of typical seawater is at the upper end of the experimental range of concentrations used here, and the authors do an excellent job discussing relevant concentration ranges in Line 583 ff. I believe it would aid the reader to have this concentration information presented earlier in the work, but this may be a matter of personal preference.

We have added to this section the information about the natural occurrence of salt in the snow and ice, as shown below:

The concentrations are proxies for natural salt occurrence in coastal snow (5 mM) (Beine et al., 2012;Douglas et al., 2012), up to the concentration in sea water (0.5 M) (Massom et al., 2001;Thomas, 2017).

Lines 120-137: Did the authors have any suspicions or intentions on how each freezing method would cause the solutes to be organized in the sample? If so, it would be helpful to include that information here, even if (actually, especially if) those expectations were later found to be untrue.

Thank you for the comment; our preference is to leave the descriptive part without personal opinions. The relevant expectations are outlined in the Discussion section to a certain extent.

As mentioned above, the ice crystal size difference between the samples frozen under high and low supercooling was expected. However, we were rather surprised that the ice crystals in the non-seeded supercooled droplet appeared smaller than those in the ice spheres prepared by spraying a solution into the LN.

Lines 138-141: While it makes sense to protect the surface from sublimating, how can the experimental procedure ensure the sublimation reveals only the original ice surface? Is it possible the ice surface is also sublimated to a certain degree, as subsequent experiments suggest? If so, what impact would this have?

The responses are provided in a related part of the paper, as shown below:

The layer exhibited a structure very different from that the frozen samples (Figure S18); thus, the desublimed ice was always readily distinguishable from the original ice sample. The sublimation of the condensed layer was monitored at the start of the ESEM imaging; as soon as the layer had sublimed, the imaged surface of the frozen sample was affected by the sublimation process to only a very small extent. We can infer this fact because we had also followed further sublimation of the ice samples, as will be reported in the near future. By further extension, the effect of ice sublimation could be more pronounced in temperature cycling and ice dynamics experiments.

Line 218: It is interesting that the surface became flooded with brine over the course of the procedure. How long did this take? Do you have a hypothesis what physical process would explain this?

The event most likely resulted from the slow, gradual sublimation of the ice. The flooding typically took about 5 minutes in the given samples.

during the procedure, the surface became completely brine-flooded due to the slow, gradual ice sublimation.

Line 322-323: It may be useful to discuss the uncertainties this introduces into the analysis.

As this comment was proposed also by Reviewer 2 we are giving same answer.

The uncertainty of the surface coverage calculation associated with the manual threshold selection can be indirectly deduced from the variance of the surface coverage in the sequence of the images recorded at the same temperature in Figure 4. Five images were recorded at the temperature -23.4 °C at the beginning of the experiment; their estimated surface coverages equaled 22.5, 21.1, 22.9, 20.4, and 25.8 %. Similarly, the values of 36.0, 35.7, and 34.4 were estimated for the surface coverage of the sample at -25.1 °C. Thus, we assume the surface coverage uncertainty due to the manual threshold selection is in units of percent. The systematic error due to a difference in the signal intensity would be larger, and therefore only the surface coverages based on the samples with similar signal intensities can be directly compared.

We modified and extended the Ch. “2.3 Estimating the brine surface coverage” accordingly:

The uncertainty of the manual threshold selection procedure is indirectly deducible from the variance of the surface coverage in the sequence of the images recorded at the same temperature soon after one another; the relevant value was in units of percent.

Line 315 ff: The idea of surface coverage is an interesting one. I think I understand the limitations of the technique, and I agree it is unfortunate the depth of the brine layer cannot be determined. However, is it worth including even some rough calculations of the total CsCl mass present in the sample, and determining if the freezing method assumptions are supportable? For example, you could calculate the surface area of the brine layer and assume it is present to a depth of 1,500 nm (the ESEM interrogation depth); does that volume account for all the CsCl, or just a tiny fraction of it?

The details of the performed calculations have been added to the Supplementary information section and theirs results into the text:

we can nevertheless estimate the relative amount of the salt on the surface of the sample, presuming that the thickness of the surface brine is similar to channels’ width. Therefore, we estimated the relative amount of the salt on the surface of the frozen samples prepared from 0.05 M CsCl via the freezing methods I-III, exploiting the assumption that the thickness of the surface layer equals 1.5 µm. As a result, the relative amounts of the salt on the surface were 9, 4.5, and 26% for the non-seeded droplets, seeded droplets, and ice spheres, respectively. The calculations are provided in the SI.

As the authors (and some of the cited literature) suggest, assumptions about where freezing methods place solutes can be very speculative; any opportunity to constrain this information would be welcome.

We consider the current observation a suitable contribution to the general discussion on these issues; further experimental methods are being designed to facilitate better understanding of the problem.

Line 329: I'm not convinced the seeded sample would grow from the exposed surface downwards. Why wouldn't crystallization be favored at the basal surface, where presumably maximum heat transfer was happening and the droplet temperature was slightly lower?

The temperature of the sample was only 2 °C below the freezing point of pure water (and slightly less in an aqueous solution), and it remained constant within the sample. Spontaneous crystalization did not occur; this process started after the sample had been seeded with an ice crystal positioned onto the sample's surface. Horizontal movement of the ice-solution interface could be observed, proceeding from the point of touch between the sample and the ice crystal towards the other side of the sample. Although the vertical movement of the interface could not be monitored, we assume that the crystallization had started at the surface, where the crystallization nucleus was in contact with the droplet. The temperature gradient was supposed to be minimal, as the solution had been thermostated before seeding at -2°C. The thermal camera measurement suggested thermal stabilization within 3 minutes at -18 °C, and it will probably be less for -2°C. Furthermore, a dedicated study proposes that crystalization on the surface is 1e10 more likely than in the bulk (Shaw et al., 2005).

We modified the text accordingly:

Conversely, the seeded sample probably started to crystallize from the surface, close to the edge of the sample as the first place in touch with the seeding crystals. Even though small vertical temperature gradients were to be expected through the solution, we considered crystallization from the surface more likely, in accordance with a related study which had suggested that crystallization on the surface was 1×10^{10} *more probable than the corresponding process in the bulk (Shaw et al., 2005).*

Line 341-343: Was the droplet used here the same droplet presented in Figure 2b? If it is or if it isn't, I would clarify this point in the text. If it is a different droplet, what was the surface brine coverage for the droplet in 2b? This comparison might give the reader some idea of the variability in the system. Section 3.3.2 discusses surface coverage for the 0.05 M sample; while I understand it represents additional work, it may be worth at least tabulating surface coverage for all 9 samples presented in Figure 2 as a Supplemental figure.

The applied droplet was the same as that presented in Figure 2b; the observed spot, however, was different. The surface coverage variabilities are summarized in Table S3, where the coverage for 5 independent, identically prepared samples is presented in relation to each freezing method. We prefer avoiding a direct comparison of the surface coverage values for various concentrations, as this may become misleading due to the arguments presented in Section 2.3.

Line 356-359: This, to me, is an interesting and important finding. As a suggestion, would it make sense to include the relative brine volume versus temperature (based on the phase diagram) as a second line on Figure 4, or add an additional scale on the right-hand side indicating relative brine volume? This would allow a more direct comparison between brine surface area and brine volume.

We are grateful for good idea. Figure 4 and the text has been completed with the relevant information.

The strong relationship between the brine volume and surface coverage is indirectly demonstrated in Figure 4; the coverage follows the same trend as the relative amount of brine in the frozen sample (w_{brine}) *calculated from the phase diagram using the formula*

$$w_{brine} = \frac{m_{aq}}{m_{fr} - m_{aq}},$$

where, m_{aq} *is the molality of the aqueous solution of salt (that can be approximated by the concentration of the salt in case of low concentrations) and* m_{fr} *is the molality of the brine in the frozen sample (it is dependent on temperature and can be inferred from the liquidus curve of the phase diagram). Thus, we are of the opinion that the change of the brine volume is the main cause of the surface coverage variation.*

Line 381-383: I'm a bit confused by the assertion that the brine volume has increased dramatically. You have previously stated the experimental method cannot evaluate brine volume, only surface area coverage. What data supports the idea that the brine volume has increased dramatically? Is it possible the brine volume is unchanged, and the brine has simply spread laterally? I would suggest addressing this uncertainty in the paper.

We cannot evaluate the brine volume directly; such a procedure is possible only in the surface coverage, and therefore the corresponding formulation in the text may have been rather vague. Nevertheless, we suppose the brine volume and the surface coverage are related parameters, as spreading of the brine on the surface of the ice is governed mainly by the surface tension.

The following text has been incorporated in the manuscript:

Even though it is not possible to evaluate the volume from the microscopic images, we documented well that the brine surface coverage had risen four times during the warming, becoming much larger than the increase implied by the phase diagram. Presumably, the brine volume can be inferred indirectly from the surface coverage if spreading of the brine on the ice surface is governed mainly by the surface tension. We interpret the change in the brine volume as the central cause of the observed surface coverage variations, and we also suppose that the observed 4-fold rise in the coverage indicates an even larger volume alteration because the brine thickness will increase too. The strong relationship between the brine volume and surface coverage is indirectly demonstrated in Figure 4;

Line 432ff: While reading the first paragraph of this section, the question that came to mind was, "Would ice sublimation also explain the apparent motion of grain boundaries, as the surface ice erodes?" a possibility I'm glad to see discussed in lines 486-490. I would suggest considering rewording some of the introductory statements of this section to acknowledge the motion is apparent, and may be an experimental artifact, not "true" motion.

Thank you! We appreciate the suggestions. The appearance of the ice boundaries' movement is discussed from the beginning of the paragraph now.

With the time progressing, the grain boundary on the right-hand side performed an apparent gradual movement through the hump, while the hump did not change.

Do you have a sense how quickly sublimation occurs off the ice surface? The fact that the surface coverage of CsCl stays the same suggests sublimation is not happening on a timescale relevant for the apparent boundary movement, but an explicit reference to the amount of time required for sublimation (perhaps in section 3.4) would be welcome. It may be possible to combine the apparent lateral motion of the grain boundaries with a known sublimation rate to estimate the angle of the grain boundaries relative to the ice surface. If all the calculated angles are shallow, which would be geometrically unlikely, the finding would support the idea that the grain boundaries are actually moving and not just appearing to move.

We materialized the answer in the text of the manuscript:

We do not know the complete time required for our samples to sublimate, because the process was not aimed at in our experiments; contrariwise, we intended to prevent sublimation of the samples. We usually imaged the samples for about 30 minutes, and they never sublimated during this period. The sample height at the start of the imaging corresponded to approximately 1 mm (based on the working distance of the ESEM). The upper limit of the (vertical) sublimation rate can thus be calculated if full sublimation of the sample is assumed to materialize within 30 minutes; the (vertical) sublimation rate would then be 33 µm/min. The brine channel (Figure 6) moved by 19 µm/min. If the apparent movement of the brine channel is caused solely by the sublimation, the angle between the channel and the horizontal line will be 60°.

However, the uncertainty of the sublimation rate discourages us from discussing the further details in the manuscript.

Line 441: If it's easy to do, I would suggest adding text to the movie (Figure S1) including a timescale, or a caption stating the overall elapsed time in the movie. Otherwise the timescale is indirectly available in the caption for Figure 6.

The video has been completed with the elapsed time.

Line 512: This line seems to suggest the method of freezing a droplet using a seed crystal should exclude impurities to the surface of the droplet during freezing. However, Line 329 suggests the opposite, that the droplet would crystallize from the surface downward. I suggest reconciling these two statements (or clarifying them if I have misunderstood either), or discussing it as an uncertainty.

Thank you for identifying the conflicting statements. We attempted recording the process on camera; however, this did not deepen our knowledge in this respect thus far. We are of the opinion that, as discussed above, the freezing proceeds from the surface laterally and also vertically downwards. The fact that we did not observe that many air bubbles may have been caused by freezing slow enough to allow the bubbles to escape, either to the ambient air or towards the bottom of the sample.

Conversely, the seeding method allows slow ice crystalization, leading to major expulsion of the air either out of the solution or downward, towards the bottom; we are nevertheless unsure of the details of such scenarios.

Figures 744-749: This is a useful finding. It is worth noting that in at least one other related work, the same investigator suggested crushing an ice sample would preferentially expose impurities because the ice would cleave "along defect sites such as veins and pockets" (Kahan et al., 2010). If the research presented here can be used to address this issue as well, and it appears to me it could, I would suggest the text be modified accordingly.

Thank you for identifying the connection. We have changed the text accordingly.

In the present paper, we propose evidence for structural differences between the surface (Figures 2-7) and the interior of the investigated ice samples (Figure 9). In the ice broken via mechanical impact, we cannot bring any evidence for the supposed cleavage "along defected sites such as veins and pockets"(Kahan et al., 2010). From Fig. 9 we can infer that the sample disintegrated perpendicularly to the lamellae. Certainly, after cutting the ice, some amount of impurities will surface; however, our *current observations indicate that most of the compounds still remain inside the ice interior. No flowing out of the brine was observed at −25 °C, even though the sample had slowly sublimated.*

– Technical corrections –

Line 266: Figure 21a should read Figure 2a. Line 373: Suggest that "expected" is a better word choice here than "expectable". Line 672: I think "special" should be replaced by "spatial". Line 729: I think that "expected" is a better word choice here than "expectable". Line 744: "Figure 2-7" should be "Figures 2-7".
Thank you for careful reading – good suggestions.

Line 769: I believe the correct temperature here should be -16 °C, not 16 °C.

We think that 16-degrees supercooling refers to -16 °C.

– References –

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