We thank Dr. Soenke Maus for an extensive and detailed review, which, in our opinion, has enabled us to substantially improve the manuscript.

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.*

I find the manuscript interesting, well written and to some degree worth publishing, as there is limited information available about the distribution of solute in ice formed in the environment, especially with the noted spatial resolution of 5 _m. However regarding the relevance for ice in the environment (section 4.) and the straightforward implications for naturally occurring salty ices proposed by the authors, I strongly disagree.

We agree that the "*straightforwardness*" of the presented results is directed more towards laboratory-prepared samples than natural ices. As we work exclusively with the former, we may not have sufficiently appreciated the specific requirements of the latter in the original manuscript. After realizing this, we modified the text accordingly.

While the paper puts many observations in the light of possible processes and phenomena described in the literature, the discussion is very vague. Some quantitative results are given but not compared to theories.

We definitely agree in that the article misses the approach of a theoretician and would like to incorporate such an aspect in the future; currently, however, we focus on experimental microscopy, which, previously, was not covered in the literature on a comprehensive basis.

It is neither clearly presented which of the observed ice-brine pattern are created by the method, and which relate to the freezing process.

The text has been revised to better separate and highlight these facts.

I. While the authors apply different freezing methods, it is not clear to which degree these resemble natural freezing conditions. For (I), freezing of a filtered (0.45 _m) solution, in a silicon sample holder rather high supercoolings were likely to exist followed by unidirectional freezing. This may resemble nucleation in the atmosphere, yet filtering, silicon holder and unidirectionality introduce considerable differences. Method (III) and (IV), freezing within liquid nitrogene, is easy to perform but may lead to rather artificial freezing conditions. Hence only (II) seeding a slightly supercooled droplet with ice crystals resembles natural conditions.

We appreciate the expert analysis of the relevance towards natural freezing conditions. As a matter of fact, our experiments are indeed centered especially on laboratory-frozen samples, and the step towards conditions closer to environmental freezing is yet to be taken.

In general, the freezing of solutions is a morphological stability problem for which the pattern depends on temperature and solute gradients (that may be derived based on the degree of supercooling) and the solidification velocity. None of these boundary conditions is documented or monitored.

We have made an attempt to determine the freezing velocity, when technically possible.

II. The technique only yields information about the distribution and pattern of ice and solute at the sample surface. For these brine films the thickness remains undetermined. The brine film coverage on the surface may not only depend on its (unknown) thickness, but also on processes in the interior of the samples. E.g. noting that brine will be expelled from the interior, the surface to volume ratio or specific surface area of samples can be expected to be a highly relevant parameter. However, sample size has not been varied during the experiments. Another important property that affects expulsion of brine is the bulk density of an ice-brine aggregate. Some results obtained here (e.g., line 339-346) should be discussed in terms of the non-linear behaviour of the expansion coefficient of an ice brine-mixture that may take positive or negative values depending on solute concentration and temperature (e.g. Timco and Frederking, 1996). Overall, the discussion of brine coverage is incomplete and speculative - see specific notes below.

III. For all samples the author create (line 138) a thin layer of ice from condensed moisture on the samples surface, to protect the surface from sublimation. It should be tested how this thin layer of ice might affect the surface brine pattern:

A deeper description of the desublimed layer has been added, together with the relevant images (Figure S18).

could the layer be porous and imply suction of brine into it?

The condensed layer of ice is porous; we do not assume the layer could alter the underlaying ice surface in a notable manner. The sample is exposed to the condensed layer only briefly, for the period of a few minutes.

Does the creation of the layer change the surface temperature?

The formation of the condensed layer was inevitable in the open-chamber freezing. The layer was formed gradually during cooling the frozen sample; thus, no abrupt temperature change was observed. During lowering the sample temperature, the surface of the sample was likely warmer than the bottom due to imperfect heating; the condensed layer could also participate in slower cooling of the surface. The experiments, however, were conducted after the sample temperature had been thermally equilibrated and the condensed layer had sublimated.

May it lead to dissolution of ice in surface brine? How is the time determined at which the thin layer has sublimated? The behaviour of this layer needs to be described better, possibly through time-series of images showing its sublimation.

The images showing the sublimation of the condensed layer were incorporated in the SI (Figure S18). In our experiments, the time of the presence of the desublimated ice layer was kept short, and we thus do not expect the brine to wick in substantially. Other details on the layer sublimation are given below.

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We have modified the text to distinguish the effects of the freezing from those caused by the sample sublimation.

We admit that the low pressure sample sublimation is only an approximation to the natural conditions, progressing much faster than would have been relevant to sea ice surface or frozen aerosols. The approximation may be relevant to a certain extent if the surface reconstruction occurs at a low rate; such an assumption nevertheless remains to be validated.

1. Introduction
L29 straightforward implications –> I would hardly call the results/conclusions as straightforward

The portion of the text has been modified to read as follows:

The results have straightforward and indirect implications for artificially prepared and naturally occurring salty ices, respectively.

L103–104 and the location of sea water brine on sea ice can be inferred –> sea ice is a rather different system, involving natural convection, unidirectional growth, a freeboard, the presence of snow, etc....

The sentence has been modified to read as follows:

 thus, the presence of brine on the surface of the frozen samples can be clearly monitored, with indirect implications towards the location of sea water brine on sea ice.

L138–141 –> This process of thin layer application and sublimation needs to be better

The formation of an ice layer on the surface of the frozen sample cannot be excluded unless the sample has been frozen in an inert atmosphere. At the start of the imaging, we identified a condensed ice layer on the surface of the sample. The surface's structure is very different from that of the actual samples, as demonstrated in Figure S18. We imaged the samples from the very beginning until the ice sublimated completely. The data were evaluated by using the images recorded soon after the experiment had begun; thus, the samples' surfaces are not at all or only very slightly affected by the sublimation.

A discussion of these details has been incorporated in the manuscript, as follows:

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.

2. Methods
L146– (2.2.) –> Here I would expect information about image resolution (mentioned later as 0.5 µm in the results section.

A sentence concerning the resolution has been added to complete the methods:

The images were typically recorded with the magnification of 500 (image resolution of ∼ 0.5 µm), although a resolution up to ten times higher is feasible.

L163–168 We estimate that the surface of a frozen sample is up to 2 °C warmer compared to that of the sample holder...–> The crystallization temperature you observe is -25 °C which, allowing for some uncertainty, is within the eutectic temperature range you mention from other sources for aqueous CsCl solutions. There is thus no need to assume a warmer surface. Why is it assumed/likely then?

Our suspicion of a warmer surface was based on the observed CsCl crystalization. Based on the referee's comment, however, we employed a thermal camera to study the temperature behavior more closely. The details are now specified within the manuscript:

The thermal camera measurements in atmospheric conditions (without the reduced pressure environment of the ESEM sample chamber) showed that the sample was about 2 °C warmer than the cooling stage. Further warming of the sample during the ESEM experiments was to be expected, especially due to the effects of the electron beam and the relatively warm gas purged through the specimen chamber; conversely, the surface cooling embodied an expectable process too, considering the ice sublimation and water evaporation from the brine. These factors could contribute to the salt crystallization on the sample surface, even when the temperature of the holder does not change; still,

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the overall temperature difference between the surface of the frozen samples and the cooling stage will likely not exceed 2 °C.

In this context, we propose the following explanation, that is also detailed in Chapter 2.3:

The pixels of the grayscale image recorded with the ESEM exhibit brightness in the range from 0 (black) to 255 (white). The Mountain ® Software enables us to select pixels with the brightness value above the chosen threshold; these pixels are highlighted in the software. By overlaying the highlighted mask and the white areas in the original image, the best-fitting threshold is chosen to represent the brine and to exclude the ice (Figure S1).

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, Figure 4. Five images were recorded at the temperature -23.4 °C at the beginning of the experiment; their estimated surface coverages equalled 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 that the surface coverage uncertainty due to the manual threshold selection ranged within units of percent.

3. Results and discussion

We performed additional open-chamber freezing experiments, utilizing a high-speed camera to determine the freezing rate in the methods I and II. The movement of the freezing front was recorded for the 0.05 M CsCl solution. In the freezing method I, the freezing rate of 150 mm/s was established; in the technique II, the freezing rate of 0.2 mm/s was roughly estimated, as the freezing interface was very poorly visible on the camera in this freezing process. Regrettably, the camera could not be used to monitor the freezing front in the methods III and IV (freezing in liquid nitrogen).

As regards the structure of the text, we would like to separate the observation of the surfaces from that of the samples' interiors, thus facilitating separation of the samples frozen with the method IV.

A table comparing all the freezing methods has been incorporated in the text. (Table 1)

We agree: The text concerned has been deleted.

L274–277 Therefore, based on analysing the crystal sizes, we can infer that the spontaneous freezing of the non-seeded droplet supercooled to about -16 °C occurred at the highest freezing rates experimentally attempted in this.–> Based on the facets in Figure 9, I would rather suggest that freezing in LN2 with method IV was faster than in method I. Next, the freezing rate of droplets sprayed onto LN2 may have been be largest overall, with the difference that this interface has not become morphologically unstable. Such a planar growth can be expected based on morphological stability theory for high growth rates (e.g., Sekerka, 1973; Coriell et al., 1994).

We thank the reviewer for referring to the literature and appreciate his valuable comment. The text has been modified accordingly.

An alternative reason for comparatively large ice crystals forming during the nebulization of a solution into LN may consist in a different freezing mechanism: at freezing rates from 210 to 2×10^{-6} mm/s, morphological instability of the ice surface is predicted, whereas outside this range linear progression of the ice front is expected (Wettlaufer, 1992;Maus, 2019).

The method IV allows the estimation of freezing rate to be less than 0.1 mm/s.

Using the theoretically predicted instability for the 0.035 % NaCl solution, (Wettlaufer, 1992) we can conclude, for the observed 10-μm spacing, that the freezing rate can range from 0.1 to 10^{-5} mm/s. Thus, the freezing of the 16 °C supercooled sample can proceed 1,000 times (or more) faster than that of the LN-immersed capsules.

L309– (table 2) –> I would skip one decimal, if the image resolution is roughly 0.5 μm. It would be interesting to see statistics for higher temperature - this is discussed in detail for the brine surface coverage. Also, and estimate of salt content based on the surface grooves should be presented, to illustrate if pattern in the interior need to be different to reflect the nominal salt content.

One decimal place was omitted in the Table. The grain boundary groove widths are difficult to measure at higher temperatures, as a large portion of the ice surface is covered with brine pools. The surface salt content has been estimated, and the calculations are provided in the SI.

L356-359 –>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 coverage implied by the phase diagram.–> During cooling the coverage changed from 80 to 40%, consistent with the phase diagram. This indicates that thickness changes are sometimes involved and sometimes not. And should be discussed.

We do not have any reliable method to determine the thickness of the brine layer; however, we suppose the area of the layer is, to some extent, related to the brine volume (see below).

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L379-381 –>The results of this temperature cycling experiment indicate that, as the ice from the surface was required to melt during the heating in order to double the volume of the brine, a formerly inaccessible portion of the brine (restrained due to being trapped below the surface layer of the ice) surfaced.–> The results can also be interpreted in terms of a thinning of the brine layer.

It cannot be excluded that the brine layer became thinner and more widespread. However, we suppose that the ratios of the volume and thickness of the layer are related, as spreading of the brine on the surface is governed mainly by the surface tension. In Figure 4, the surface coverage is compared to the amount of brine in the sample, calculated from the phase diagram; it clearly shows that the two parameters are related.

The following text has been added:
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.

L395-397 –>It cannot be excluded that the sample surface was several degrees warmer than the holder due to the effects of the electron beam; purging relatively warm gas through the specimen chamber.–> Could you please present some numerical estimates?

As already cited above the text has been rephrased as follows:
The thermal camera measurements in atmospheric conditions (without the reduced pressure environment of the ESEM sample chamber) showed that the sample was about 2 °C warmer than the cooling stage. Further warming of the sample during the ESEM experiments was to be expected, especially due to the effects of the electron beam and the relatively warm gas purged through the specimen chamber; conversely, the surface cooling embodied an expectable process too, considering the ice sublimation and water evaporation from the brine. These factors could contribute to the salt crystallization on the sample surface, even when the temperature of the holder does not change; still, the overall temperature difference between the surface of the frozen samples and the cooling stage will likely not exceed 2 °C.

L405-409 –>Thus, the temperature increase with the subsequent spread of the brine on the surface may lead to the surface darkening, resulting in higher solar radiation absorption and further increase of the temperature.–> The effect of a brine film on sea ice albedo has to my knowledge not been discussed yet - rather the number and size of inclusions in the bulk ice are relevant.

The number and sizes of inclusions are certainly factors of considerable importance.(Light et al., 2003) However, the optical properties are undoubtedly influenced also by the phase state of the salt/brine, as measured and discussed in (Light et al., 2016;Carns et al., 2016) The specific references have been added together with an explanatory sentence:

Conversely, the brine crystallizing on the ice surface would result in larger reflection of the radiation, as the crystals would scatter the light substantially more effectively compared to the liquid brine (Carns et al., 2016;Light et al., 2016). These effects, together with the number and sizes of inclusions, ice grains, and location and characteristics of the absorbing particles, influence the overall absorption properties of icy bodies (Warren, 2019).

L432–434 (and whole 3.5.) –>The dynamics of the frozen samples' surfaces could be observed even at -23 °C. The CsCl brine-filled grain boundaries were not static: their positions changed swiftly in time. Interestingly, the positions of the humps on the ice surface did not change accordingly. –> The two explanations (i) migration of grain boundaries in a temperature gradient and (ii) exposure of inclined grain boundaries during surface sublimation are not so expected and their discussion is lengthy. Could they be validated by some data and estimates (possible temperature gradients, sublimation rates, internal sample observations, etc...)?.

We incorporated the answer for both explanation into the text:
(i)

The sample was prepared from a hemispherical droplet by cooling from the bottom. Thus, the height was most likely not constant across the sample and would be the most prominent in the center, allowing the temperature gradient to rise across the surface. A thermal camera was used to estimate the temperature gradients throughout the sample during cooling outside the ESEM chamber; it was not experimentally feasible to insert the camera into the chamber. As the temperature of the sample was dropping from 0 to −18 °C, the temperature at the center of the surface was approximately 2.7 °C higher than at the periphery. This difference had been caused presumably by the greater thickness of the sample at the central section. However, after the sample was cooled down and its temperature became approximately constant (which typically took about 3 minutes), the gradient was reduced, albeit in an opposite manner: The center was about 0.2-0.3 °C colder than the periphery, producing a small temperature gradient throughout the sample even after the cooling.

The sample presented in Figure 6 was sufficiently cooled down, as enough time had elapsed since the last temperature adjustment; therefore, we assume there might be a temperature gradient of only several tenths of °C across the sample.

(ii)

The sublimation rate largely depends on both the amount of water vapor purged into the chamber and the temperature. 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°.

L526-528 –> 10 µm indicates rather fast freezing - see note on freezing rates above.

We very much appreciate the help that Dr. Soenke Maus provided through interpreting our observations. We have added the following text:

Using the theoretically predicted instability for the 0.035 % NaCl solution, (Wettlaufer, 1992) we can conclude, for the observed 10-μm spacing, that the freezing rate can range from 0.1 to 10^{-5} mm/s. Thus, the freezing of the 16 °C supercooled sample can proceed 1,000 times (or more) faster than that of the LN-immersed capsules.

4. Relevance to previous observations

L578–591 –> It is corrrect that the range 0.005 to 0.5M solute concentration is found in nature. However, the classification into relevant salinity regimes and processes in nature sounds a bit artificial: sea ice can have 0.05 M solute content (brackish ice, Baltic Sea ice), and for salt concentration near roads I would expect a large range depending on environmental conditions. Surface snow on sea ice may be much more saline than 0.005 M, and a range of 0.01 to 0.1 M is more representative.

We appreciate the expert comment and advice by Dr. Soenke Maus; we have changed the text accordingly:

The tested concentrations ranged over two orders of magnitude. The values within 500 - 50 mM define the concentration of NaCl in seawater, and therefore they are also descriptive of fresh sea ice in the given context (Massom et al., 2001;Thomas, 2017). NaCl concentrations reaching up to 160 mM were detected immediately next to a highway treated against road icing; 50 mM of a salt solution can thus be considered a concentration potentially found farther from roads or also in their close vicinity when the salt was already partly flushed away (Notz and Worster, 2009;Labadia and Buttle, 1996). The values discovered in surface snows in Arctic coastal regions or on the surface of frost flowers span between 10-100 mM but may also be as low as 5 mM.(Beine et al., 2012;Douglas et al., 2012;Maus, 2019).

L592-595 –>The principal finding presented within our study is embodied in the very strong sensitivity of the ice-brine morphology and brine distribution to the freezing method (Figure 2, 3). Even for identical solution concentrations, the method and direction of freezing strongly modify the appearance of the ice surface morphology.–> These principal findings are not new.

Yes, we agree on that point, as relevant items of literature and also theoretical models have been composed and released. With our experiments, we primarily intended to understand the locations of impurities in laboratory-made ices.

L601-610 –> It is not clear that natural convection played a role for solute redistribution within the different freezing experiments, as freezing was either fast or upwards. The comparison to sea ice formation and desalination is thus not useful here. While directionality is important, it is not due to natural convection.

The comparison to natural convection indeed is rather remote from the central subject; our original intention was only to summon the convection from that field. The following text has been complemented in relation to this information:

Our freezing methods differ from that related to natural sea water in the rates and directionality. By extension, we would like to imply that the role of material convection in relation to the drop size should be considered for the modeling of the freezing process.

L662–664 –>Apparently, the amount of the brine on the ice surface was not sufficient
to fill the groove around all the ice grains due to low brine concentration and/or the freezing method concentrating the brine towards the interior of the ice matrix–> As mentioned, directionality, local brine expulsion and bulk ice-brine density changes are relevant here and this statement needs to be validated.

We agree with the comment and have attempted to discuss the topic accordingly. However, we are unsure of further validation besides the presented micrograms.

L678 –>frozen aqueous solution (without added salt)–> Do you mean frozen (almost pure) water?.

Yes, we meant pure water. Thank you!

L710–712 –>The present study suggests that the remaining solute is likely to be found in not only the puddles of the highly concentrated solution but also, the veins or grain boundaries grooves threading the crystals.–> This is not a new observation. To produce something new you should at least give some estimates of the solute contained in the veins/grain boundaries, based on your observations.

Estimates of the relative amount of the solute present on the surface layer have been added to the text. The calculations are outlined in the SI.

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.

5. Conclusion
L759 –>amount of brine–> You have not determined the amount, just the surface coverage.

We agree: "surface coverage" is a more accurate expression.

L766–767–>The presented micrographs clarified the possible porosity and pore microstructure
of sea ice.–> The relevance of the freezing conditions/sample sizes for sea ice has not been demonstrated. Regarding microstructure of sea ice there are textbooks that 'clarify' this much better (e.g., Weeks, 2010; Shokr and Sinha, 2015).

We agree: Our sample preparation methods are not directly related to sea ice formation. The sentence has been modified as follows:

The presented micrographs clarified the possible porosity and pore microstructure of salty ices.
L768–770–>From the ice crystal sizes we infer the actual 769 freezing rates–> In fact no freezing rates are inferred. Also, freezing rates are very likely largest for the LN2 freezing (III and IV) - see discussion above.

If our interpretation of the lamellar spacing in the samples prepared via the method IV is correct, the freezing rate could be comparable to or slower than those in the method II. Nevertheless, in the Conclusion we abandoned the inference of the freezing rate from the grain sizes, as it does not seem very reliable after we have studied Dr. Soenke Maus' comments and explanations.

Instead we state:

The rheology of ice is strongly related to the freezing rates; in our experiments, these were directly measured or inferred from the spacing of the lamellae in the ice samples' interior, with the latter of these approaches being applied in cases of morphological instability.