

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 2; our responses are outlined in black; and the modified portions of the manuscript are shown in italics.

Anonymous Referee #2

Main points

1. I'm a little confused about the detector sensitivity and variation in acquisition settings. It peaks for the authors that they mention this point so explicitly. I agree with them, that this -while being nasty- does not impact their conclusions. Looking at Figure 3, this might imply that the samples with a high signal intensity, might even have a higher surface coverage of brine, because weaker spots from smaller puddles might be overseen. I might suspect that sample g and f show the same fine structure as i their dark areas – but I would not think that this adds significantly to the brine covered area. However, in line 319 to 323 it is not clear to me how the “normalised brine surface coverage” is defined and why this is free from this artefact. I would suggest to move the discussion from the experimental section here, use Figure 3 more explicitly to discuss the potential impact on your conclusions, and maybe even include estimates of this systematic error in the uncertainties mentioned in lines 315-320.

The formulation “the samples with a high signal intensity might even have a higher surface coverage of brine because weaker spots from smaller puddles might be overseen “ is true.

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.

2. I suggest to add concentration and temperature information of the samples during measurement and ice production to the phase diagram (Figure 1).

As the displayed phase diagram ranges from 0 to 18 mol kg⁻¹, the two lower salt concentrations (0.005 M and 0.05 M) would be on the very left of the phase diagram, thus becoming hardly recognizable.

Throughout the text, I would encourage to be more precise in using the words brine volume, concentration, and salt amount. I'm not so convinced that you can discuss brine volume, as you are not sensitive to the thickness. Your observable is "only" the amount of salt, is it not?

Thank you for the comment.

What we observed was the surface coverage with the salt. However, we suppose that the brine volume and surface coverage are related parameters, as spreading of the brine on the surface of the ice is governed mainly by the surface tension. The relationship between the brine coverage and volume can be inferred from Figure 4. We attempted to use the indicated words with greater care throughout the text.

Would you expect the shock frozen samples (in N₂) to crystallize once temperatures drop below the eutectic and could you comment on the question whether or not the salt deposits would liquify again upon warming to the temperature of investigation taken that this process might be slow.

We know from the differential scanning calorimetry (related to the NaCl solution, paper accepted for publication in J. Chemical Physics: "Vitrification and Increase of Basicity in between Ice Ih Crystals in Rapidly Frozen Dilute NaCl Aqueous Solutions") that the brine in the shock-frozen samples crystallizes at some temperature below T_{eu} , depending on the cooling rate. In our ESEM micrograms, crystallized CsCl was not observed in the frozen spheres due to poor thermal conductivity between the cooling stage and the spheres; however, we examined CsCl crystallization in the freezing method IV, where the thermal conductivity was sufficient. The deposited salt liquefies upon warming; Figure S19 was added to demonstrate the process.

3. Protecting the sample with a condensed ice layer is a excellent idea. However, could you give more details how you ensured that you "revealed the original surface" for the measurements and not removed too much ice (including some layers of the original surface). (lines 138)

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.

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.

Minor points

lines 12: I'm not a native speaker: What means "threading"? I find the first sentence of the abstract a slightly to abrupt and suggest to start more general.

The word "threading" was adopted from (Cheng et al., 2010) in the hope of its being an acceptable option. In the context of the article, *threading* means *winding around*: (<https://www.dictionary.com/browse/threading?s=t>)

lines 34: reword: ice and snow are also part of the environment.

Certainly: The problem has been eliminated.

Lines 41: Add reference to Grannas 2007 and/or Bartels-Rausch 2014

The reference has been added.

Lines 43: Rather “into the ice lattice at high concentration”. Domine’s work clearly shows that ice can hold impurities as solid solution and for some species at sufficient high concentration to impact partitioning to the atmosphere.

The relevant text section has been expanded accordingly.

Excepting low concentrations of HF, NH₄⁺, HCl, HNO₃ and formaldehyde (Perrier et al., 2002; Thibert and Domine, 1998, 1997), impurities are usually not incorporated into the ice lattice (Krausková et al., 2016; Hobbs, 2010; Wilson and Haymet, 2008).

Lines 87: I found the discussion in this paragraph too detailed and slightly off topic and suggest to remove it. This paper does not add to the question of interior vs. surface distribution of impurities. It is only sensitive to the surface of the samples.

The interior of the frozen samples was, in fact, approached by using the method 4, where the samples frozen in the test tubes had been broken and examined. Another applicable technique of reaching inside the sample was sublimation.

Lines 168: Is it correct that you never observed a supercooled solution? The brine always started crystalizing at the eutectic temperature? That would be an interesting observation.

Thank you for the comment. Regrettably, we cannot discuss your point properly, as we did not measure the temperature of the sample in a direct manner; the temperature of the cooling stage was adjusted. The temperature difference between the sample and the cooling stage is characterized in the Methods section. Moreover, the cooling is markedly slower in the sample than in the Peltier stage. We nevertheless know that supercooling is common in the DSC of NaCl solutions.

Lines 171: What is an BSE detector?

The abbreviation refers to a detector of back-scattered electrons.

Figure 2: Could you give an estimate on the smallest features that you would detect with the specific detector settings? (lines 213: no brine puddles larger than.... Were detected...).

We imaged the samples with the resolution of 0.5 μm. However, the size of the structures is not the decisive factor determining the visibility of the structures. The detector settings are influenced by the salt concentration and surface roughness. Further, if the crystalline salt is present, the visibility of other structures becomes greatly reduced because the salt signal is intensive too, and the detector sensitivity is thus reduced.

In figure 4 the surface coverage seems to increase continioiusly, while T shows steps between -24 to -18°C. Could you comment on this?

The temperature in Figure 4 is that set on the Peltier stage. When the temperature of the stage has been changed, it takes some time for the temperature of the sample to settle; thus, the changes on the surface coverage are continuous, not stepwise.

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