

The Cryosphere Discuss., referee comment RC2
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Comment on tc-2021-376

Anonymous Referee #2

Referee comment on "Sublimation of frozen CsCl solutions in ESEM: determining the number and size of salt particles relevant to sea-salt aerosols" by Ľubica Vetráková et al., The Cryosphere Discuss., <https://doi.org/10.5194/tc-2021-376-RC2>, 2022

This is a very interesting laboratory study investigating the impact of freezing method, salt concentration and sublimation temperature on the formation of salt aggregates after water ice removal via sublimation. The main finding is that the eutectic point represents a threshold below which salt aggregates are formed small enough to potentially form sea salt aerosol (SSA).

GENERAL COMMENTS

My main concern is regarding how the experiments presented do apply to the natural environment. A more critical assessment is required so the reader is aware that salty icy interfaces in the polar regions and associated processes may actually behave quite differently. I suggest to expand the discussion (section 4.6) and critical assessment of the experiment relevance, in particular on two points:

A) salt origin and transport - the experiment uses as a surrogate for the natural system a well mixed salt solution droplet, which is cooled down, then sublimated. However, the transport of salt to snow (which then gets airborne to release SSA after sublimation) occurs primarily via upward migration driven by capillary action from the sea ice surface and atmospheric deposition of SSA (e.g. from open ocean and frost flowers), all well described in Dominé et al. (2004). Thus, the salt solution or particles are delivered to the surface of already frozen water, i.e. ice crystals. Therefore, the formed salt aggregates may be quite different in the natural environment than what is seen here, in agreement with that "structures of these residua therefore depend on the sizes of the ice crystals and the location of the salt in the frozen sample prior to sublimation" (L335).

B) freezing method - the first method, labeled as "spontaneous freezing" insinuates homogeneous freezing, which it is not. Ice nucleating particles introduced with the CsCl must be present in solution to initiate freezing at -11degC, way above the -37degC homogeneous freeze point. INPs could be anywhere in the droplet and initiate localised formation of an ice crystal, therefore I question the concept of a slowly upward moving freezing front (Fig1a), has this been observed? Have UHP water blanks been run to check for contamination and background? This needs clarification.

Furthermore, the title promises also some information on the size of salt particles/aggregates formed. However, only number densities are reported whereas particle size information is only semi-quantitative along the images. Has any attempt been made to extract from the image analysis also the fraction of observed residual particles falling into the coarse aerosol size range (0.5-30 µm)? This would importantly add to a more quantitative evaluation of the experiment.

Overall the paper is very long, in part due to the very descriptive discussion, and occasional repetitions of text from Section 3 in Section 4. I'd therefore recommend to combine Section 3 & 4 into a section "Results and discussion" & shorten the text; e.g. by remove all text/ references to chemical reactivity, pH, QY ... as these are rather speculative and were not part of this study; refer more often to Figure 15, which is a good summary.

In summary, I recommend some major revisions. More detailed comments are listed below.

SPECIFIC COMMENTS

L28 in general from the aerosol or ice (snow) phase containing bromide. The relative contribution of aerosol and snowpack source in the high latitudes is under debate

L32 source strength and their relative contribution

L41 better: ... not supported by ...

L52 spell out ESEM

L68 dry air?

L73 I suggest to move description and reason of choosing CsCl as a sea salt proxy to the method section

L222-240 I suggest moving this paragraph of describing choice/ advantages and limitations of CsCl as a sea salt proxy to the method section. You need this in order to understand the results.

L230 similar eutectic point between NaCl and CsCl supports this; however would differences in salt crystal structure itself impact the size, shape and number of salt structures formed after sublimation? CsCl due to the similarity in respective ion sizes forms a cubic lattice with eightfold coordination, whereas NaCl with the cation smaller than the anion forms octahedral structures with sixfold coordination. Please comment and expand the discussion accordingly.

L231 tasks?

L236-37 why mention if you don't share the details (composition, figure in appendix)?

Either remove or add the required detail.

L274-77 Please clarify the terms 'stick-slip' mechanism and 'pined and mobile contact lines evaporation' and what they are supposed to mean in this context.

L278 this is a far too strong statement; it is true for the experimental conditions, however it remains far from clear if this is the case in the natural environment. Distinguish more critically between the this experiment and nature. Also what aerosol size range is being referred to?

L332 spontaneous freezing - typically homogeneous freezing without the addition of nucleators occurs at -37degC, however here freezing occurs at -12C so "warm" INPs are present (introduced with the CsCl); see general comments above.

L399-404 I don't think this is correct, please clarify. After lead opening in winter/spring new sea ice forms quickly and frost flowers grow on top of it into very cold air; the coldest air temperatures during MOSAiC were observed in March at around -42C (Shupe et al., 2022). I expect surface snow and filigran ice structures on the surface such as frost flowers to be in equilibrium with air temperature above.

L469-78 While potential implications of freezing on pH, reactivity etc are interesting I suggest to remove these (and references) as this is not relevant to the experiment of this study, pH was not measured ...

L500 clarify or define "aerosol-forming potential"; can you give an average particle size? what was the fraction of observed residual particles falling into the coarse aerosol category (0.5-30 μm)?

Supplement

L8 it should really be here and throughout the text particle number density (number of particles per area or volume), or surface/ volume number density; the term particle density, in my view, refers to a composition dependent density, as in mass per volume, of the particle itself.

L14 surface density is ambiguous (see above), better surface number density or particle number density per area ...

REFERENCES

Dominé et al., The origin of sea salt in snow on Arctic sea ice and in coastal regions, Atmos. Chem. Phys., <https://www.atmos-chem-phys.net/4/2259/2004/>, 2004.

Shupe et al., Overview of the MOSAiC expedition - Atmosphere , Elementa, <https://doi.org/10.1525/elementa.2021.00060>, 2022.