Review of "Sublimation of frozen CsCl solutions in ESEM: determining the number and size of salt particles relevant to sea-salt aerosols" submitted to ACP by Vetráková et al.

Reviewer #2

General comments:

Vetrákova et al. present a thorough study of sublimation from CsCl/water in an environmental SEM (ESEM). The research is well motivated, carried out with expertise, and features a good discussion. It is of good value for the scientific community, including atmospheric science. It is also well written, although slightly too much targetted to specialists.

The paper is rather large, and features a plethora of images and topics, and the results are described in great detail. It could profit either from substantial cutting, putting a narrower focus, or - in the other extreme - a more complete view. This would consist the "revision", while scientifically almost everything is very sound (see below).

In case of a more complete view, valuable information would be the phase diagrams NaCl/ice/water and CsCl/ice/water, discussions on crystal morphology of all species (would that include NaCl.2H2O?), a discussion of the regular patterns (e.g. fig. 7c), comparison to simple optical microscopy studies, (semi-)quantitative evaluations, etc.

Thank you very much for your comments. In the following text, we reply to your comments and answer the questions. We are currently working on implementing the suggested changes into the manuscript.

Specific comments:

It is not always clear how much the paper builds on results from Závacká et al. (2022), and what is new (except of course the cation, and the advantages of using a heavy ion).

The Letter by Závacká et al. (2022) shows sublimation residua of sea salts for a range of concentrations and sublimation temperatures (−16, −30, −40 °C). We observed low temperature was needed for formation of small salt particles, however, due to the complexity of the sea salt, difficulty in defining eutectic temperature, and its poor visibility on the ice surface, the relation between formation of small particles and eutectic solidification was not straightforward.

All the samples in Závacká et al. (2022) were prepared only by one freezing method (spontaneous freezing). In the present manuscript we wish to detail three ways of sample freezing, compare the results (size and number of particles) in terms of freezing method (rate and directionality of freezing, …).

The observation of crystallization of CsCl below and above the T_{eu} allowed us to present hypotheses about the mechanisms how the particles are formed.

The discussion and interpretation of figure 2 is, compared to all others, very short. Specifically, the droplet-like features on figures 2a and 2b require an explanation. Rather few readers are experts in ESEM, let alone ESEM of multiphase systems!

Thank you for your comment, the features are newly described in more detail.

The brine (section 3.3) must be supercooled, and indeed the droplet-like features suggest the liquid state. But is there any independent proof of the liquid nature?

We infer the liquid state of the droplet-like features from their visual appearance, and from their transformation to salt crystals during the observation. We did not use any further detection methods to proof their liquid state.

Technical corrections:

The surface pretreatments are not provided. They are very important for the Peltier surface and for the silicon wafer (for which the reader requires additionally how it was fixed and thermally coupled to the Peltier).

We provided the details of the silicon surface of the Peltier stage. Following text was added to the chapter 2.1.

Surface of our cooling stage is made from very pure, commercially available silicon wafer that is usually used for the production of semiconductor components. The wafer had no additional surface treatment. It was glued to the Peltier cell with a highly thermally conductive adhesive, compatible with the low temperature and reduced gas pressure environment of the microscope.

Line 137: 650 Pa should not be called "ambient" pressure, which is ca. 101000 Pa.

The “ambient pressure” refers to the pressure around the sample inside the specimen chamber of the ESEM. The term is used in the meaning of “surrounding”. It shall not express the atmospheric pressure. We can use the term “chamber pressure” instead.

Line 186: The temperature sensor and its setup are not described.

Following text was added to chapter 2.1:

Due to confined space and electrical interference between the temperature sensor and the detector, it is not able to directly measure the temperature on the surface of the sample during the experiment inside the ESEM chamber. Therefore, the sample temperature is inferred from the temperature of the Peltier cooler. The actual sample temperature was validated outside the ESEM at atmospheric conditions using Pt1000 temperature sensor (P1K0.161.6W.A.010, Innovative sensor technology IST AG, Switzerland) frozen inside the sample. The bias between the temperature of the Peltier cooler and the actual sample temperature was no more that 2 °C.