# Interactive comment on “Single-particle characterization of ice-nucleating particles and ice particle residuals sampled by three different techniques” by A. Worringen et al.

Anonymous Referee #3

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Review of “Single-particle characterization of ice-nucleating particles and ice particle residuals sampled by three different techniques” by Worringen et al.

This manuscript presents the characterization of individual ice nucleating and ice residual particles using SEM/EDX analysis. The ice nucleating particle and ice particle residuals were collected by three different instruments. This study provides very useful information/insights on the comparison of sampling instruments/techniques for ice nuclei. The subject of the manuscript is suitable for publication in ACP. I recommend the authors to consider addressing the following comments/issues for the revision.

Major comments:

1. the manuscript didn’t provide sufficient description on the FINCH+IN-PCVI experi-ments. First, was there size selected inlet applied (i.e, were the ice crystals removed before FINCH)? or it take all the particles including ice crystals and evaporate the ice before their inter the FINCH for ice nucleation activation? What were the experimental temperature and RH conditions for FINCH ice nucleation measurements? These are crucial information for the comparison with other two techniques. If the ice crystals were removed before FINCH, then it is comparing the non-activated particles with ice crystal residuals (activated particles) even if they were sampled the same air mass spontaneously.

FINCH+IN-PCVI is operated at the total inlet, which samples aerosol and all cloud elements without a size selection. No ice crystals are removed, all hydrometeors are evaporated. We have added an according statement.

Experimental conditions of FINCH are shown in Fig. 1 and Table S1 in the supplement now.

2. It is not very clear what samples are compared. In Figure 1, were only the samples form the marked periods (A-F) compared? The only overlap of the sampling for all three instruments was the F period around Feb. 20, which also across about 24 hours. How many particles/samples were analyzed during this F period? It would be very useful to provide air mass backward trajectories (HYSPLIT data) in the supplement materials to validate that the same particle source during the sampling times which were compared. The sampling time for the background aerosol particles was only less than 30 min, additional evidences (e.g., HYSPLIT data) are needed to validate this comparison with background aerosol composition. Same for the comparison with LA-MS data set. It is also not clear when LA-MS data were collected, the sampling time of LA-MS data should also be marked in the Figure 1.

Periods B to F have no longer a meaning so they are removed from the graph. In the summarizing graphs, all samples are compared, and period A is used as an example, as for this one, comparatively many particles were available.

Trajectories for Period A are given in the Supplement now.

Discussion of background samples was removed from the paper as requested by other reviewers.

Fig. 1 shows now also the sampling times of LA-MS. SEM and LA-MS are unavoidably non-parallel due to the sample flow limit.

3. Section 4.2, a major part of the discussion on the comparison of ice nucleation ability for the different particle classes is not appropriate, since the manuscript didn't provide sufficient supporting information of this type of comparison. First, Strictly speaking, when comparing the nucleation ability, data on freezing temperature/RH, activation fraction, or nucleation rate are needed for each particle types.
We agree and are aware of that problem. Due to the low particle numbers, however, we are not able to differentiate all the different thermodynamic/kinetic conditions during the campaign.

Second, the enrichment is not quantitatively determined and background aerosol information is not sufficient (this study only has background aerosol data on a very short period of time).It is recommended to rewrite this section or leave it out.
We removed the section 3.4 (total aerosol) and modified section 4.2 accordingly.

4. Section 4.3, the discussion on the comparison of these three techniques can be extended in more details.
We think that the present data set is not suited for a comparison of details of the different sampling techniques. In particular, we do not have enough parallel measurements for such a comparison. However, the INUIT consortium will carry out a closure experiment in March 2015 at a large aerosol chamber (AIDA, Karlsruhe) in which we hope to get a data set consisting of strictly parallel measurements.

It would be also important for the community if the manuscript can discuss briefly the limitations and advantages of each technique and provide some recommendations to improve the sampling or measurements in the future.
We have added a set of recommendations regarding the techniques and their development. Lacking a reference method, it is at this point not possible to rate each technique against another.

Specific comments:

1. P23030, l21-24, these two statements are oversimplified and it is not true. This should be reworded. For example, sea salt and sulfate can be efficient ice nuclei, e.g. Gregory P. Schill and Tolbert [2014] G. P. Schill and Tolbert [2013], Abbatt et al. [2006].
We have rephrased this section.

2. P23031, l3-4, there are a few studies that investigated the effect of mixing state of particles on ice nucleation and characterizations of ice nucleating particles from differ-ent field campaigns using SEM/EDX and other X-ray analysis technique (Hiranuma et al. [2013], [Knopf et al., 2010; Knopf et al., 2014]; Wang et al. [2012]).
We have rephrased the sentences and added references.

3. P23031, l25, the sampling for three techniques was not in parallel.
Corrected.

4. P23032, l23-25, Please provide a more detailed description on collecting particles onto different substrates. Are both two substrates used in all three different sampling instruments? How chemical composition was quantified for the particles these two substrates (the X-ray background of these two substrate is different)?
Both substrates were used with all techniques. This is stated explicitly now.
Chemical quantification in SEM/EDX always has to regard the contribution from the substrate. For that reason, usually non-interfering substrates are chosen (B and thin-film C, in our case). This is stated now. More details are later discussed in section 2.2.1.

5. P23033, l16 and l19-21, Do you mean “INUIT-JFJ 2013” or “INUIT-JFJ 2014”? Please provide more information regarding the ice nucleation experiments. It is not sufficient by just saying “supersaturation and freezing temperature were varied during the campaign.”
Section was rephrased.
We give now details on the working conditions in the text, in Fig. 1 and as supplement.

6. P23035, Section 2.3, the manuscript didn’t provide sufficient descriptions/criteria on the particle classification for both SEM/EDX and LA-MS techniques. What is the “Droplet”, any chemical information on these particles? It could be very useful to show representative SEM images and X-ray spectra for each particle class (can be in the supplemental materials).

We have extended Table 2.

For the LA-MS classification, the paper of Schmidt et al. (2014) provides the details. Our table of classification was modified.

Information on droplet composition is given already in the original manuscript (page 23034, lines 4-9) – they mainly consist of sulfates.

We show in the manuscript specific artifacts of this investigation. Information on the Pb-rich particles can be found in more detail in Ebert et al. (2011). Other aerosol types are already described throughout the literature (Ebert et al., 2004; Kandler et al., 2007; Vester et al., 2007; Adachi and Buseck, 2008).

7. P23037, L10-16, since there are abrupt increases in particle concentrations which may come from local source, why it is only a “minor local influence”?

The samples were actually collected before and after that distinct spike in particle concentration. As this is not obvious from the graph, we have added a statement to the text.

8. Combining discussion in P23041, L18-19; p23045, L2-13; and P23050, L23-25, It is stated that lead-bearing particles in the whole INUIT campaign is 1% for FINCH+IN-PCVI and from Figure 6,no lead-bearing particle was detected from ISI sampling. If assuming that FINCH+IN-PCVI captured all the lead-bearing particles that nucleated ice (or nucleated ice on all the lead-bearing particles), does that mean 90% of lead-bearing particle (9% out of 10%) determined by Ice-CVI was artifacts?

This conclusion could only be drawn, if no variation would occur during the campaign, and if no statistical uncertainty would exist (which is high here due to the low numbers).

We have added a new discussion on lead as artifact to section (now) 3.1.1. We conclude that the homogeneous lead-rich particles (<10 % of all lead-bearing) are probably artifacts, but the others are probably not.

## References used in this reply

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