Editor Initial Decision: Reconsider after minor revisions (Editor review) (25 Aug 2015) by Dr. Ryan Sullivan

Our responses are in red.

Comments to the Author:
Thank you for submitting a further revised version of your manuscript. This has largely addressed and clarified the questions raised in the last round of reviews.

The causes of the discrepancies and disagreements found using different ice nucleation methods is an important topic that will likely continue to see active discussion for many years to come. This paper contributes to this discussion, by focusing on the possible effects of particle coagulation and sedimentation on the ice nucleation properties derived from cold plate droplet freezing methods where particle suspensions in water are studied. This is in contrast to the "dry" methods where particles are dispersed in air as an aerosol before they are nucleated into droplets and subject to ice nucleation conditions.

No comment required

This manuscript is now almost acceptable for publication in ACP. I do feel the clarity of the manuscript could be significantly improved however in some places, and I ask that the authors consider the following changes and suggestions as they revise their manuscript for final publication in ACP. There are also some errors and typos I noticed that should be corrected.

No comment required here

It strikes me that it is not made clear that the MICC experiments were focused on condensation/immersion mode freezing. This should be made very clear early on, so that the applicability of the comparison to the cold plate and other immersion freezing experiments is very clear. You might also briefly mention of any depositional ice nucleation below water saturation is ever observed, and if so do you simply ignore this data and just focus on the total/maximum ice crystal concentration produced in the chamber after the expansion and cooling?

We have added a sentence at the end of the intro to say we are focussed on condensation / immersion freezing. We always observed drops in our experiment before significant ice nucleation.

Similarly, I found it hard to fully understand how the MICC expansion chamber probe data was analyzed to determine the ice crystal concentrations. It seems that when possible the 3V-CPI measurements of particle shape are used to distinguish ice from droplets. If that is correct please make this clear.
We have made this clear in the methods section.

In some cases the ice crystals were too small/round to distinguish from their shape, and so instead ice is determined by particle size from the CDP. If that is also accurate please make this clear, and also state what particle size was used as the split between cloud droplet and ice crystal, and a justification for this cut size. In the few experiments where size and not shape was used to measure the ice crystal concentration, you could draw a line on the CDP figure indicating this cut size.

Yes, that is correct. The cut size for the warm experiments is 20 microns because the ice crystals grow rapidly. For the cold experiments it is 13 microns from the CDP, taken after the droplets have evaporated due to the Bergeron-Findeison process – because the ice crystals were smaller in these experiments; however, this doesn't necessarily mean the ice crystals are 13 microns in size – it is just their optical size. The justification is that the drop distribution from the CDP forms a distinctive `mode' and the ice crystals are clearly separated from this mode. See new Figure 4. We've said this in the observations section. While not perfect it is a conservative estimate of the ice concentrations (yet it is still higher than the cold stage derived results) so our conclusions still hold.

Also, how is the aerosol particle concentration (used to derive the aerosol surface area) measured, using only the UHSAS data?

Yes. This is said in the methods now.

The size range of the WELAS probe should also be mentioned on page 5 for completeness. How is the WELAS data used?
Done. See above for how WELAS was used.

I'm not sure that all the plots of the different expansion experiments are necessary or add much to the main paper. It is probably hard for a non-expert to really glean much from these plots, and they take up a lot of space in the paper. It is good to document them, and the discussion of each where different features of each expansion are discussed. I think most of these could be moved to Supporting Information instead, as there are already 17 figures in the paper, which is quite a lot. A few exemplary expansion figures could remain in the main paper. You could describe the general features and the data analysis more generally, instead of discussing each expansion separately. Any important unique features of particular expansions could still be discussed in the main paper, or the supplemental.

We've separated these plots into supplementary information now and just discussed the background experiments and a warm and cold ice nucleation experiment.

The expansion experiment figures (Figs. 2-9) could be annotated to indicate the specific features (cloud droplet formation/evaporation, ice formation) mentioned in the text. Color coded vertical lines could be used to indicate these main events, for example.

We have done this for the main figures in the paper – figures 4 and 5. The other figures showing chamber time-series have been moved to the supplemental material and figures 2 and 3 (now in the supplementary material) have been annotated too.

The typical range of particle concentrations used in the cold plate experiments should be mentioned, such as on the top of page 4. Right now these numbers are missing. Then it will be more clear why you probed the range of particle concentrations you used in your experiments and models, such as on page 14, line 12.

done

This is a rather long and confusing sentence in the Abstract: "Results from applying a coagulation model to the size distribution of mineral particles present in the suspensions, as used in the cold-stage-derived parameterisations, were used to investigate the idea that the mineral particles coagulate in suspension, which either removes the particles from the drops by sedimentation, whilst in the bulk suspension, or reduces the total particle surface area available for ice nucleation to take place."

Shortened and made clearer.

Section 3 – At the beginning, should reiterate that condensation/immersion mode freezing was

studied in these experiments.
Done.

Paul DeMott recently published an empirical parameterization for mineral dust INP, based on laboratory and field studies. As this is highly relevant to this work you should discuss the salient points from that paper that relate closely to your work.

DeMott, P. J.; Prenni, A. J.; McMeeking, G. R.; Sullivan, R. C.; Petters, M. D.; Tobo, Y.; Niemand, M.; Möhler, O.; Snider, J. R.; Wang, Z.; Kreidenweis, S. M. Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles. Atmospheric Chemistry and Physics 2015, 15, 393–409, doi:10.5194/acp-15-393-2015.

Thank you for pointing me to this paper. Indeed it does corroborate the previous statement by Hiranuma et al that CFDCs may underestimate in the supersaturated regime when operated at 105% Rhw. It also goes on to state that more are activated at higher RH and this agrees better with AIDA data. Therefore this addresses further the concerns of Dr Murray (in a previous comment) and adds to the discussion. We have cited it in the discussion.

Page 4, line 17: Do you mean "> 5 nm"?

Yes, sorry for the confusion.

The fact that you do multiple expansions on the same mineral aerosol is important, as the most efficient ice nucleants are removed in the first expansion, thus changing the derived n_s values. While you do discuss this on page 10, it seems very odd to me that you averaged the n_s values from the successive expansions together. This does not seem like an accurate reflection of the aerosol's actual ice nucleation properties. Why not create an additional figure similar to Fig. 10 where you plot the n_s values for the different expansions as a different colour, for the different aerosol types. This would be a very interesting plot as it would show just how much each expansion changes the remaining aerosol's ice nucleation properties. Just averaging the different expansions together seems inappropriate as it is not the same aerosol in each experiment – the nest ice nucleants have sedimented out.

Sorry, they are not averages of all expansions. The n_s values are averages for one experimental run. Each experimental run is a data point. This is now said more clearly in the method section.

Section 4.2.1: Cite some key references to the ACPIM model.
Done.

Page 12, line 1: "as observed". Do you mean depositional ice nucleation was or was not observed here?

Not observed within error of measurements. We've changed the manuscript to make this clear.

Page 12, line 8: In discussing these parameterizations it would help if the parameters that they are based on was briefly summarized. Are they all based on particle surface area, as is used here?

Yes, we've now said this explicitly.

Section 4.2.2: Switched from past to present tense. Should stick with past tense.

OK, done.

Page 14, line 21: Regarding the initial mineral dust size, I am quite sure these is a large distribution of particle sizes, extending into the supermicron range for these mineral powders. That has certainly been my experience doing experiments with many types of mineral dust powders. So while I see the point that the mean (?) particle size increased with time, I don't think one can assume there were no initial supermicron particles.

Good point, we are not assuming there are no initial supermicron particles, but that there were very few sub micron particles left over; hence, they must have aggregated. We've clarified this in the manuscript.

Page 14, line 26: Are these the Dynamic Light Scattering experiments? DLS is often referred to but never stated what these experiments are. Is that what the Malvern Zetasizer Nano ZS measures? Much more details are required here, specially as most of the atmospheric science community is not familiar with DLS as it is used for colloids not aerosols.

We've included an explanation of this now..

Why is only one particle size listed for the DLS measurements in Table 2? Is this the mean or median value? Surely a distribution of particle sizes is measured. Would be great to include the variance of the measured size distribution, as that can indicate if the size distribution narrowed or widened with time in the water. That can provide further information about the physical processes causes the change in particle size.

It is the mean. We have also provided the standard deviations now. Unfortunately we feel it is difficult to offer too much of an explanation into the observed standard deviation but they are now in the table for completeness.

Page 16, line 30: should state that an /increase/ in wt % results in an /decrease/ in fractional surface area.
done

As the particles in droplets on a cold plate are subjected to a temperature gradient, are thermophoretic effects possibly important in causing particle aggregation/coagulation? Should discuss if you can or can't rule out the role of thermophoresis.

Thermophoresis is potentially important, yes. The exact dimensions of the instrument and the cooling rate would be required to model this. We have mentioned this and given a back of the envelope calculation in 4.2.2.

Page 18, line 18: Should spell out what "it" is.
Done (suppression of coagulation due to double charge layer)

Page 20, line 11: Typo, "it" instead of "in".
done

Page 20, first bullet point: Should reiterate the differences in the Feldspar versus other mineral data that lead to this conclusion. If I recall the Feldspar data agreed even when high particle concentrations were used.

Done. This is much clearer now.

Yes the Feldspar agreed for high particles concentrations, but only in pico litre drops at low temepratures. Here we are suggesting the generation of pico litre drops (drops of 10 microns in size) breaks up aggregates.

Appendix: I found several typos, please proof read carefully.

Page 20, line 2: solved, not solve

done

Line 5: "When by the"

multiplied, this is inserted.

Line 7: delete "the"

we disagree. "the" is needed here.

Page 23, line 16: "(source)" – missing reference?
Yes, reference added.

Page 24, line 20: "like changed"

changed (to charge)!

Table 1: Are the two values listed in [ ] for the two lognormal modes of the size distribution? Please specify in the table caption.

Yes, this has been done now.

Table 2: See comment above about why only one (average?) value for the particle size from DLS measurements is given.
This has been addressed for completeness we included the variances.

Figure 1 caption: Typo, "monitor"

Changed to "monitored."

Figure 2, and similar figs 3-9: I found it hard to know which y-axis to use for the lower panel. The red text of the right y-axis suggests that only the 3V-CPI data is on the right y-axis, but the legend says the blue traces are also on the right y-axis. Perhaps you can see the confusion with the colour coding here. Perhaps blue could be used for the left y-axis and the droplet concentration only. And/or you can add left/right arrows beside the traces or legend entries indicating which axis to follow for each.

Have done this.

See earlier comment about moving most of the chamber expansion plots to supporting information.

done

Figures 13 & 14 should be combined into one 2 panel figure.

done

figure 15: Specify in caption if this model included Brownian only or Brownian and gravitational forces.

This included only Brownian forces. Done.

Figure 16 should have a more explicit caption, hard to understand what data is used here. What system is the Atkinson data for? What does “shifted to Niemand line for natural dust” mean?

Sorry the `Niemand line' statement was wrong and referred to something else. We've changed the legend and updated the caption to explain exactly what was done.

Ryan Sullivan