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Interactive comment on "Investigating the discrepancy between wet-suspension and dry-dispersion derived ice nucleation efficiency of mineral particles" *by* C. Emersic et al.

Anonymous Referee #1

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Review of "Investigating the discrepancy between wet-suspension and dry-dispersion derived ice nucleation efficiency of mineral particles" by Emersic et al.

Using a cloud chamber, the authors investigated the ice nucleation efficiency of three types of mineral particles (kaolinite, NX-illite, and K-feldspar) generated by dry-dispersion. The results are then compared to ice nucleation efficiencies of the same minerals determined using wet-suspensions. The authors argue that at warmer temperatures the ice nucleation efficiencies associated with dry-dispersion are consistently higher than ice nucleation efficiencies associated with wet-suspensions. The authors then go on to argue that the differences may be due to coagulation of particles in the

wet-suspensions.

This topic is well suited for ACP and the subject is an important one since both the dry-dispersion and wet suspension techniques are commonly used to measure ice nucleation efficiencies of atmospherically relevant particles. I appreciate the authors' novel and systematic studies to try and determine the contribution of coagulation to the wet-suspension experiments. Listed below are concerns and/or suggested revisions. Once the authors have adequately addressed these comments, I recommend the paper for publishing in ACP.

Major comments:

1) Hiranuma et al. ACPD, 2014, stated "Though the number of immersed particles can vary from droplet to droplet and the random placement of particles in the drop may be of an effect on the ns values, the ns spectra from suspension measurements are in general in reasonable agreement even over a wide range of wt% of illite NX samples. Thus, the influence of the random placement of particles in the drop and agglomeration on the ns estimation for suspension measurements seems small." This statement seems contradictory to the coagulation calculations in the current manuscript and conclusions reached in the current document. Please discuss.

2) Were microliter samples with high concentrations of minerals used in the wet suspension experiments reported by Hiranuma et al. at temperatures of -28 C to -34 C? If microliter samples and high concentrations of minerals were used at these temperatures in the experiments reported by Hiranuma et al., then coagulation seems like an unlikely explanation for the difference between the dry dispersion and wet suspension experiments. Please discuss.

3) It was not clear how the authors determined the ice particle concentrations reported in Table 1. For K-Feldspar at -21C the authors indicated with a footnote that the CDP was used. For clarity, please indicate that CDP >18 microns was used (assuming this is correct). Also, does this mean the ice numbers from the 3V-CPI were used in all

other cases or were 3V-CPI > 35 used in some of the other cases?

4) In the text it was not clear how the authors determined the number of ice particles in the experiments and in some cases the decision sounded subjective. In some cases it sounded like they relied on ice particle concentrations from the 3V-CPI, but in other cases it sounded like they didn't rely on this result because the ice particles were somewhat rounded due to a lack of vapor growth. What criteria did they use to decide when to use and when not to use the ice measurements from the 3V-CPI. In addition in some cases it sounded like they used the data from the 3V-CPI > 35 microns to determine ice particle concentrations while in other cases it sounded like they did not since the 3V-CPI often over-sizes out of focus images of droplets (Connolly et al., 2007). What criteria did they use to decide when to use and when not to use the results from the 3V-CPI > 35 to determine ice particle concentrations? From my reading of this document it sounds like the results from the ice 3V-CPI should be used as a lower limit to the ice particle concentrations and the 3V-CPI > 35 microns should be used as an upper limit to the ice particle concentrations. Is this a valid statement?

5) What is the uncertainty in the ice crystal concentrations determined for use in Equation 2? In table 1 the authors report an uncertainty from Poisson counting statistics, but what is the uncertainty from under counting with the 3V-CPI due to a lack of vapor growth and rounded ice particles and what is the uncertainty from over-sizing out of focus images of droplets with the 3V-CPI > 35 microns?

6) In Figures 8 and 9 the authors should include the uncertainty in their results from the uncertainty in determining the ice crystal concentrations in their experiments (i.e. uncertainty from under counting with the 3V-CPI due to a lack of vapor growth and uncertainty from over-sizing out of focus images of droplets with the 3V-CPI > 35 microns). Also, in Figure 9, have the authors included uncertainties in the parameterizations from Murray et al. 2011 (assuming uncertainties were given in the manuscript by Murray et al. 2011).

7) Page 892, line 13-15, "However, the droplets lasted for a brief period (less than < 40 s)." Here the authors are referring to Figure 3, but in Figure 3 the black solid line suggests that the liquid droplets persist for at least 300 seconds. Please explain and give some explanation on how to interpret the black solid line in the bottom panels of figures 2-7.

8) Related to the comment above, on Page 893 the authors indicate that in Figure 5 the kaolinite particles nucleated ice in the absence of cloud droplets. I am not sure how the authors reach this conclusion since the presence of cloud droplets are indicated by the black solid line in the third panel of Figure 5.

9) In the dynamic light scattering experiments, why not do the experiments as a function of time to determine coagulation rates. This seems more relevant since coagulation rates would be more directly comparable with the coagulation calculations?

10) It would be helpful to list the point of zero charge for the different surfaces of kaolinite.

11) Section 5.1.1 The discussion on colloidal forces in suspensions is useful and does suggest that it may be reasonable to neglect repulsive forces in the coagulation calculations. However, this section does not provide conclusive evidence that repulsive forces can be neglected. I also came to the same conclusion from Table 2. It would be more convincing if the authors did a time dependent study using dynamic light scattering to show that the force of repulsion can be neglected.

Minor comments:

1) Page 890, line 9-10, are there any particles > 5 nm in the filtered air.

2) Figure 11 and 12. For the red solid lines there is a sudden drop at the earliest times, and then a straight line. Why is this line not an exponential curve? Please comment.

3) In Figure 2, consider changing "droplets" in the annotation to "droplets (CDP)" to be consistent with the other annotations.

4) Figure 2. Please indicate that the dashed blue line corresponds to the right axis.

5) It may be helpful to include a short description on each of the instruments used to measure droplet concentrations and ice crystal concentrations (i.e. PALAS WELAS 2000, CDP, SPEC 3V-CPI) since they are crucial to the interpretation of the data.

6) The figure caption indicates CDP > 20 microns, but the text refers to CDP > 18 microns. These two numbers should be the same to avoid confusion.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 887, 2015.