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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Competition for water vapour results in suppression of ice formation in mixed phase clouds" *by* Emma L. Simpson et al.

Anonymous Referee #2

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This manuscript presents laboratory experiments and parcel model simulations of ice nucleation at temperatures mixed phase clouds are observed. The experiments simulate expansion cooling in a chamber, in which ice formation on K-feldspar particles (INP) was measured at low and high ammonium sulfate (CCN) concentrations, respectively. A parcel model is then used to simulate the chamber experiments.

The parcel model simulates droplet activation for insoluble as well as soluble aerosols, and simulates ice nucleation based on ice active sites per unit surface area. The authors used four different criteria for initiation of heterogeneous freezing. Two of the criteria (threshold mass of condensed water, threshold water activity) that were derived by fitting the chamber results are non-traditional.

The parcel model is then used to simulate ice formation for a range of vertical veloci-

ties and INP size distributions. Model results are compared between low and high CCN simulations, which suggests that competition for water vapour may lead to suppression of ice formation in mixed phase clouds. This conclusion is not supported by the chamber experiments (Line 14, Page 9). However, the experiments and model simulations are still interesting and are appropriate for publication in ACP. For these reasons a major revision is recommended.

Major Comments:

1. There is a lack of discussion on possible reasons why a suppression of ice formation by high CCN was not observed in the chamber experiments or what assumptions made in the parcel model may be inaccurate that led to the suppression.

2. If "a minimum depth" of aqueous shell is required for ice nucleation (page 6, line 28), then the critical water mass required for freezing should be proportional to the K-feldspar particle surface area, not the volume as shown in Eq. (7).

3. A threshold water activity may not be a good criterion, as indicated by the narrow range (0.9997 to 0.99994) of fitting results. Furthermore, ice nucleation has been observed to occur at lower water activities, although decreasing rapidly as the activity decreases (e.g., Knopf and Alpert, 2013).

4. The two new criteria (threshold water mass and threshold water activity) were derived from experiments with INPs of a specific size distribution. Would the same values be estimated when the INP size distribution is changed or the soluble mass fraction is changed in the chamber experiments? The estimated criteria are used in Section 5 for various size distributions and soluble mass fractions.

Minor Comments:

1. Parcel model simulations were made for a range of INP size distributions and vertical velocities, using the four different freezing criteria (shown in Figs. 5, 6, 7, S6-S15). The results demonstrate the suppression of ice nucleation when CCN is numerous and

droplet growth effectively competes for water vapor. The interpretation is reasonable for Regime 1 (small size and low vertical velocity). But Lines 5-8 on Page 15 may need some clarification:

"In the second regime, Regime 2, towards the top right hand corners of panels c and d in Fig. 5, INPs have a significant soluble fraction that allows them to act as giant CCN at large diameters, towards 2 um. This results in a suppresion of ice formation because the INPs are in competition with themselves as well as the CCN for available water vapour."

Is it possible that the suppression results from earlier ice nucleation (i.e., at higher T) with larger INPs and subsequent vapour deposition to ice enhances the effect of CCN?

Similar clarification is suggested for Lines 7-11 on Page 16:

Is "kinetic limitations to growth of large particles" meaning more rapid growth of ice particles?

"This is because at warmer temperatures..." the WBF process is also more effective at lower updrafts?

2. Line 28 on Page 2. "do not take competition for water vapor into account as it has already been corrected for" seems to be saying the opposite.

3. Move the text on Lines 1-15 on Page 14 (description of FHH, the hygroscopicity parameter, and soluble fraction) to between Lines 27 and 28 on Page 4, as a new paragraph. Start a new paragraph at "Freezing rates of INPs ..." (Line 28).

4. What is the soluble mass fraction and hygroscopicity parameter for K-feldspar particles used in the chamber experiments and in the parcel model? Give the values near Line 23 on Page 4 or in Table 2 on Page 12.

5. Figure 2(b) on Page 10. The peak concentrations slightly beyond 250 seconds appear to be spurious.

6. Table 2 on Page 12. One modal size distribution is given for K-feldspar particles in Table 2. But a two-modal size distribution is shown in Figure S4. Need to explain why the fine mode is neglected.

7. Lines 10-11 on Page 13. "INPs with four different soluble fractions" could also be given earlier in model description (see minor comment 3).

8. Lines 29-30 on Page 19. "the INPs act as giant CCN" is not as effective as ice formation and subsequent vapor-to-ice deposition for INPs in competition with themselves.

9. Lines 4-5 on Page 20. This statement is confusing.

10. Figure S1. Why are the curves for frozen fraction not monotonic as a function of INP diameter (in units of nm)?

11. Figure S16. Is the unit for Dp micron rather than nm? Also the caption is not clear: what are the chamber conditions (e.g., soluble mass fraction?), what is the difference between (a) and (b)?

12. Typos. "approached" on Line 9, Page 4. "location long" on Line 5, Page 8. "where" on Line 8, Page 13. "reach" on Line 3, Page 20. "its" on Line 6, Page 20.

References:

Knopf, D.A., and Alpert, P.A.: A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplet, Farad. Discuss., 165, 513-534, 2013.

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