Review of **"A numerical study of aerosol influence on mixed-phase stratiform clouds through modulation of the liquid phase"**, by de Boer et al., submitted to *Atmospheric Physics and Chemistry*

General comments

This paper describes simulations of aerosol impacts on Arctic mixed-phase clouds through modulation of droplet immersion freezing rates. These clouds have important climate impacts yet are poorly understood and represented in weather and climate models. There have been a number of papers in recent years studying Arctic mixed-phase clouds, and in particular processes explaining their maintenance and dissipation. The current paper adds to this body of literature, and presents some interesting results. The authors find much larger sensitivity to insoluble mass type than other parameters such as soluble mass fraction and CCN concentration. Overall, the paper is well-written, although aspects of the presentation could be improved as detailed below. I also have several comments regarding the methodology and analysis, including a general comment about placing this study in light of observational constraints and uncertainties. Furthermore, I found the discussion in section 5 tying the modeling analysis together with the picture of Arctic clouds as a complex system rather distracting and incomplete (see specific comment #6 below). Overall, my recommendation is major revisions before the paper can be accepted for publication in *ACP*.

Specific major comments

1. As noted above, a major question is how the ranges of aerosol/freezing parameters tested here are constrained by observations, in light of large observational uncertainties in freezing characteristics of various particles. For example, the authors find much larger sensitivity to insoluble mass type than soluble mass fraction and aerosol concentration, but how well constrained are the ranges of these parameters tested? How do the differences in freezing rate parameter across these ranges (Y in Eqs. 2-3) compare with observational uncertainty in the freezing rates derived from the work of Diehl and Wurzler (2004)? While I recognize that observational (laboratory) constraints on these parameters are limited, some discussion of this issue is needed.
2. While presumably the authors utilized a 2D model for computational efficiency (though this was not stated in the paper), there can be significant differences in 2D versus 3D representations of the cloud dynamics and turbulence (e.g., Bretherton et al. 1999, QJRMS). While I wouldn't expect the authors to redo these simulations in 3D, I would suggest the authors mention this point, and explicitly state their rationale for using a 2D cloud model.
3. One potential issue in terms of the model setup and experimental design is that the cloud dynamics/turbulence were not allowed to spin up (likely taking roughly 1-1.5 hours) prior to allowing ice formation. Hence, ice was allowed to occur in a system that was in a state of significant imbalance between liquid water, cloud top radiative cooling, and turbulence. This could have potential implications on interactions between ice microphysics, the occurrence of liquid, and buoyant production of turbulence that I would like to see the authors address. For

example, the system may tend to glaciate too early by introducing ice before the dynamics able to support liquid water growth have fully spun up.

4. Some of the microphysical budget analysis and discussion is not clear (including Figs. 7, 9, 12). In the figures and discussion, there often doesn't appear to be a balance in the process rates, which does not seem possible given that the simulations reach fairly steady conditions after several hours. For example, liquid water condensation rate in Fig. 7 is overall positive for SOO and NOICE, yet the LWP is fairly steady in time. This suggests some kind of sink process balancing the overall net (i.e., vertically-integrated) positive condensation/evaporation rates. Is this from collision-coalescence and subsequent sedimentation of rain? A bit more discussion here would be illuminating. Another example is the strongly negative total vapor tendency near cloud top in most of the simulations in Fig. 7. I don't understand how this can be the "total vapor tendency", because that would result in significant drift over time in the vapor field at these levels if it were this the case. Is the "total vapor tendency" only the microphysics tendency (i.e., excluding resolved and sub-grid transport and large-scale advection)? In general, the authors need to better clarify what is actually shown in these budget figures, and ensure these results are consistent with a balance (or lack thereof) in timeseries of quantities like liquid water and water vapor mixing ratios.
5. I don't follow the analysis and discussion of the soluble mass fraction sensitivities on p. 22075-22076. The authors provide an explanation for these results through changes in critical activation radius with changes in soluble mass fraction, leading to larger droplet sizes. While the critical radius does increase with an increase of soluble mass fraction according to Kohler theory, the critical supersaturations decrease (in some cases rather substantially). This would have an opposing effect, as more droplets will activate with higher soluble mass fraction due to lower critical supersaturation, leading to smaller droplet sizes. I am surprised that the critical radius effect is more important than the critical supersaturation effect. Is it possible that the effect is exaggerated because relatively few aerosol bins are used in the model? Did the authors investigate sensitivity to bin number/resolution? In general the authors seem to emphasize the importance of differences in overall condensation arising from changes in droplet size, (e.g., in their diagram in Fig. 14, and on lines 21-22 on p. 22078), which I am skeptical of, especially for stratocumulus clouds with relatively weak dynamics. This is because supersaturations in these clouds are quite small, meaning that the clouds are not far from equilibrium saturation regardless of droplet size (except perhaps right near cloud base, where most droplet activation should occur). Without "clean" sensitivity tests in which condensation/evaporation are the only processes allowed to operate, it seems difficult to associate changes in condensation rate with changes in droplet size, since there are numerous other processes that are also impacted by droplet size which can in turn impact condensation rate. Hence, overall, I would suggest the authors try and better clarify the effects of changes in droplet size and concentration. One suggestion for addressing this issue would be to show plots droplet concentrations and size (e.g., droplet size spectra) – I found it surprising that this was not done in the manuscript given its focus on the impacts of ice formation via liquid droplet properties.
6. As stated above, the discussion on system complexity including Fig. 14 seems rather out of place in this paper. I completely agree about the importance of considering system complexity in the

context of multiple interacting processes, but Fig. 14 and the discussion on p. 22078-22079 are confusing and do not really seem to contribute to an improved understanding of this. Moreover, this discussion seems rather out of place since this issue was not described in terms of motivation of the work, or discussed in the introduction. Thus, it has the feel of being an add-on at the end. There are also issues of generality here; the authors show various interactions with arrows denoting the sign of the interaction in Fig. 14, but might these differ for different cases (e.g., different temperature, cloud thickness, etc.)? Finally, many interactions hypothesized in Fig. 14 are not well supported by the results presented here. For example, the authors suggest the importance of changes in aerosol properties leading to changes in droplet size, which in turn impacts condensation rate and the positive feedback between liquid water, radiative cooling, and production of turbulence (e.g., lines 21-22 on p. 22078). However, as discussed in comment #5 above, without "clean" sensitivity tests it can be very difficult to isolate specific mechanisms driving the response to aerosols. While bulk condensation rate may change with droplet size, this doesn't necessarily mean that changes in droplet size drive the changes in condensation rate since there are many other interacting processes also affected by droplet size.

7. As alluded to in comment #5, I am somewhat concerned about the bin resolution. The authors used only 40 bins for liquid, 20 for ice, and only 10 for aerosols. Did they investigate sensitivity of results to bin resolution? This seems especially pertinent given the emphasis of this study on the effects of droplet activation and growth on mixed-phase clouds; these processes in particular can be impacted by numerical effects from coarse bin resolution.
8. I would suggest using a, b, c, d, etc. to label all multipanel figures, instead of top, center, bottom, etc. I found it very hard to follow which plots different parts of the figure captions were describing.

Additional comments

1. P. 22061, lines 1-2. I don't believe that Kay et al. 2008 specifically discuss their results in terms of mixed-phase clouds, contrary to what is implied here.
2. P. 22062, line 23. I don't think Jiang et al. 2000 explicitly simulated the IN budget and allowed depletion of IN, in contrast to Harrington and Olsson (2001). This should be clarified.
3. P. 22064, line 7. It is stated here that AMPS evolves size spectra, but then later it is stated that it uses mass-based bins. Is it size or mass? I realize for liquid drops these are equivalent, but this isn't necessarily the case for ice if particle density is not assumed constant, and regardless this should be worded consistent to avoid confusion.
4. P. 22064-22065. Some aspects of the aerosol model are unclear. Is the aerosol size distribution assumed to be fixed in time? Or does aerosol processing occur through cycles of condensation, collision-coalescence, and evaporation below cloud base? Is aerosol lost at the surface via precipitation?
5. P. 22065. I am confused by the term "potentially activated mass bins". Are these bins that experience at least partial activation? I'm assuming that the model allows activation of some fraction of aerosols within a bin, which is important given the low bin resolution (only 10 bins). This should be clarified.

6. P. 22066. The symbol for deltaT changes between eqn. (1) and eq. 3 as well as later in the text on p. 22067. A consistent symbol should be used.
7. P. 22068, line 17 and elsewhere in the text. There are several places where the authors use "CCN", but strictly speaking these should be condensation nuclei (CN) concentrations (for example, the aerosol size distribution fit in Eq. 4 is based on CN measurements). In general, CCN concentration depends on the critical supersaturation assumed (or measured), so CCN concentrations should include the relevant supersaturation.
8. P. 22069, lines 20. The dynamical timestep is 2 sec, but what is the microphysics time step?
9. P. 22070, lines 11-12. It is stated that the NOICE simulation has a LWP "several orders of magnitude" larger than the observations, but this seems an exaggeration. It appears the overestimate is closer to one order of magnitude in Fig. 3.
10. P. 22070, line 25. The authors state that the freezing of haze particles via the condensation mode may be important at low levels in the cloud. However, the definition of cloud base in mixed-phase clouds that precipitate ice is not clear. Do the authors mean freezing of haze may be important below the base of the mixed-phase layer containing supercooled liquid?
11. P. 22071, lines 12-13. I don't follow what is meant by "wave like pattern". I would suggest not using "wave like" here, perhaps "oscillating" would be better.
12. P. 22072, line 17. "…is reduced in the SOO simulation…". Reduced relative to what? The NOICE simulation?
13. P. 22074, lines 22-25. This sentence could be confusing to readers and could be reworded. I think the authors mean that with few hydrometeors in the MON and ILL simulations, there is little loss of vapor due to surface precipitation, so there is a net (vertically-integrated) positive tendency of water vapor in the column; in other words, water vapor increases over time in these runs.
14. P. 22076, line 1. See comment #11 above.
15. P. 22077, line 14. How would droplet collision-coalescence increase LWP? This alone doesn't impact bulk mass of liquid. Furthermore, collision-coalescence combined with sedimentation of larger coalesced hydrometeors should reduce, not increase the bulk amount of liquid water.
16. P. 22088, Figure 1. First, the y-axis is missing a label. Second, I think what is plotted here are contributions to Y in Eqs. 2-3, not $B_{h,i}$ as implied in the caption.
17. P. 22092. What are the "droplet volume distributions"? This is never clearly defined in the paper.