Response to reviewer comments for ACP-2012-655

We would like to thank the three reviewers for providing helpful comments on this manuscript. We have taken time to go through these comments and have responded to them individually below. Please note that the original referee comments are provided in *italics*, while the author responses are below them in plain font. We would like to apologize to the reviewers for the mislabeled figure captions. These have been updated as outlined below. In general, we believe that changes made to the original manuscript as outlined in our responses greatly improve the quality of this paper.

Anonymous Referee #1

General Comments

This manuscript describes numerical simulations aimed at an improved understanding of aerosol effects on mixed-phase clouds, in particular through impacts of the liquid phase. Specifically, these authors characterized the impact of insoluble particle type, soluble mass fraction, freezing point depression, and aerosol number concentration. Aerosol insoluble mass type (i.e. freezing efficiency) was found to be the most important factor controlling cloud lifetime. Overall, the paper explores several important processes and yields interesting results. My biggest complaint about the manuscript is in regards to the figures, which I think are at times unclear (and mislabeled).

Thanks to this reviewer for these helpful comments – again, we apologize for the mislabeled and unclear figure captions. We have worked to improve these and have attempted to address your additional concerns as outlined below.

Specific Comments

p. 22061, line 24. CCN activity of a variety of dust particles has been demonstrated in the lab. A relevant reference here would be: Koehler et al., GRL, 2009, DOI: 10.1029/2009GL037348.

We thank the reviewers for bringing this study to our attention and this reference has been added at the location mentioned above.

Section 2.2. The authors show that insoluble aerosol type is an important factor in their study. Yet the authors only use freezing efficiencies from one study 8 years ago (Diehl and Wurzler, 2004), which ignores a lot of laboratory work that has been completed since that time. The authors note that these values really just represent a range of freezing efficiencies (p. 22071 lines 22-25), and not strictly these components, but I think it would be worthwhile to direct the reader to some of these studies to demonstrate the wide range of values measured for the broad range of particle types *in the atmosphere (e.g. Broadley et al., 2012, ACP, doi:10.5194/acp-12-287-2012; Zimmer- man et al., 2008, JGR, doi:10.1029/2008JD010655; Murray et al., 2012, Chem Soc Rev, DOI:10.1039/c2cs35200a and references therein, to name just a few).*

This is indeed an important point. We have added a statement about the existence of other studies along with some of the above-mentioned references in section 3. In general, we believe that the values used from Diehl and Wurzler represent an appropriate range for this specific case, mainly because this range results in everything from a very rapidly glaciated cloud (with high freezing efficiencies) to a thick, long-lived cloud (using low freezing efficiencies). While there are certainly other particle types and freezing efficiencies existing, it seems as though we are covering a good range here for this specific cloud.

Section 2.2 and Figure 1. I think the authors need to make some effort to make this figure clear and consistent with Section 2.2. First, all red lines and all pink lines appear to be identical; these should be differentiated. Second, why 'ln' in Eq. 3 and 'log' in Figure 1 for Bh,i and Vd? Variables should also be identical in the equation and figure (e.g. Bh,i in both). What is 'T10' in the figure?

Figure 1 has been modified, and in our opinion is now clearly labeled and consistent with the text/Equation 3. T10 was an example of the impact of the last term in Equation 3 (ln(deltaT)), but has been removed from the latest figure.

Section 4.2, 2nd paragraph and Figure 4. Immediately following a paragraph in which the authors state that freezing point depression is not an important process for this study, except in the case of haze droplets (which the model does not consider for freezing), the manuscript goes on to show and discuss results related to freezing point depression. Is this figure necessary?

We have reworded this section and integrated the main points of the second paragraph into the first paragraph. We believe that Figure 4 (when compared with Figures 10 and 13) still provides nice visual evidence of the limited impact of freezing point depression when compared to other aerosol influences, and have therefore left it as part of the manuscript.

Discussion. Given that the authors show results from a mixed-phase cloud case from SHEBA (Figs 3, 6, 8, and 11), it would be worthwhile to add some discussion (in the Discussion) relating their modeling results to the measurements and giving some recommendations as to how to this study has improved our understanding of this particular case.

The intent of this current effort was not necessarily to better understand a particular case. The SHEBA case was used out of convenience, as it had already been run as part of a model intercomparison study (Morrison et al., 2011,. *J. Adv. Model. Earth Syst.)*. For various reasons, we do not expect the simulated clouds to replicate what was observed – an example of this is the lack of a liquid water spin-up period in the simulations. Ice is allowed to form from the initial timestep and act upon a liquid profile that is matched to observations, which would result in the departure from the observed steady state even in a perfect model. The observational estimates were included simply for reference, to give an example of a reasonable liquid to ice ratio for the large scale conditions used in the current study. We have added discussion along these lines in Section 5.

Figure 3. It is interesting that there was no ice from the observations until >3 hours. Does this correspond to a change in temperature? What does this say about your ice parameterization?

This is actually the result of instrument downtime. It was snowing during this time period as well. This has been stated in the manuscript in section 4.1.

Technical Comments

p. 22061, line 27. Deposition nucleation occurs on SOME IN.
The word "some" was added here.

p. 22067, line 13. 'complementary'.
Fixed – thanks!

p. 22070, lines 6-7. The 'Bergeron-Findeissen effect' was earlier referred to as 'Wegener-Bergeron-Findeissen mechanism.' Be consistent throughout.
We've edited the text to state Wegener-Bergeron-Findeisen throughout (and fixed the spelling for Findeisen).

p. 22070, line 9. I believe 'simulations' should be 'observations'.
Yes, it should be and has been replaced. Thanks.

p. 22070, lines 11-12. Is it really several orders of magnitude? It looks like <2 orders of magnitude.
Correct – this has been reworded to state that the simulated LWP is roughly one order of magnitude larger than observed.

p. 22078, line 2. Do you mean 'droplet number concentration'?
This was supposed to say "droplet size and droplet concentration on riming rates", and has been fixed.

p. 22080, line 7. 'reached'.
Fixed – thanks.

p. 22080, line 7. Why the '±'?
+/- has been replaced with approximately.

p. 22080, line 21. Add 'in'.

Fixed – thanks!

Figure 3 caption. 'NOICE, black and KAO, green'
We have modified the document to accurately represent the line colors.

Figure 5 and Figure 5 caption. The text notes that only the snapshot at 10 min includes information about the ILL or MON simulations, but I see no indication of these in the bottom 2 panels of the figure, even at 10 min. Also, the caption states that for the top panel that the 'NOICE line and markers are not included,' and yet they appear to be included, while I see no lines for ILL and MON. And why is there a particle freezing efficiency in the top of Figure 5 for NOICE?

Thank you for bringing this to our attention – the lack of ILL and MON results at 10 minutes (and beyond) was the result of changes to our routines that calculated cloud-top location. The NOICE simulation was indeed included in this version of the figure. There is a particle freezing efficiency because in terms of aerosol properties, the NOICE simulation is essentially the same as the KAO simulation. The only difference between the two is that the actual freezing parameterization is turned off, and the values shown in the previous figure represent what the freezing parameterization would have been doing if it were active. We have now updated this figure to remove the NOICE simulation and adjusted our definition of cloud (qc>1e-8 g/kg) to include the MON and ILL simulations as well.

Figure 6 caption. There is not a 'time series for ice number density (bottom left)' in the manuscript, as the caption suggests.

This caption has been updated to match the current figure.

Figure 7. Don't change color scheme for this figure; i.e. don't change MON to purple.

This apparent change in color resulted from a figure conversion from RGB to CMYK. All of the plots were created with the same color schemes for individual simulations. This has been corrected in the current version (all figures now RGB).

Figure 8 and p. 22075, lines 17-18. The caption does not accurately describe the figure (ice # concentration is missing).

This caption has been updated to match the current figure.

Figure 11 and p. 22077, lines 6-9. Again, the caption does not accurately describe the figure.

This caption has been updated to match the current figure.

Figure 14. One of the positive feedbacks is blue. What do the black symbols represent?

The positive feedback loop was in red – the negative feedback loop was in blue. While there is a + correlation within that negative feedback loop, the loop itself is still negative (the loop being that supersaturation with respect to ice results in ice depositional growth, which subsequently reduces ice supersaturation). Black lines represent pathways that are not included in one of the example feedback loops. In the end, we decided to remove this figure all together, as reviewers believed it to be distracting to the rest of the work.

Anonymous Referee #2

General Comments

The paper explores how variations in CCN properties can affect the persistence of mixed-phase clouds through alteration of the intensity of ice nucleation via immersion mode. The study is based exclusively on the numerical simulations from a 2D cloud-resolving model and the sensitivity of simulated cloud is tested with respect to the soluble mass fraction of the CCN as well as the composition of the insoluble component. The latter affects the ice nucleating properties of aerosol immersed in cloud droplets. The ice nucleating efficiencies of various insoluble components, such as soot and various dust types, are taken from an independent study. The paper follows the footsteps of dozens of studies from the last few years that used model sensitivity studies to gain insight into various processes in mixed-phase clouds. For the approach to yield useful results, however, this study needs to do a better job in justifying the range of explored parameters and analyzing the simulations, as indicated in comments below.

We thank the reviewer for their insight and suggestions. We've adapted the manuscript as outlined below.

Specific Comments

1. If I understand the simulation setup correctly, in each model run a soluble mass fraction and insoluble mass type are assumed to be the same for CCN of all sizes. Noting that nearly all of the CCN are of sub-micron sizes, with about half of them having radius smaller than 0.1 micron, and knowing that dust particles tend to fall into the coarse-mode aerosol, is it realistic to assume that every aerosol particle contains a dust particle? Furthermore, the majority of "laboratory-derived parameters" used here for dust were obtained for particle sizes of hundreds of microns and extrapolating them to particles 1000 times smaller calls for providing some justification. I wonder if it will be more realistic to assign dust fractions only to coarse mode CCN (called "large" mode in this paper) and what effect that change would have on the results?

This criticism is justified and we agree (and state in the manuscript) that the assumption that all aerosol particles have the same composition is not realistic. We agree that it may be more realistic to only include dust particles in the coarse mode of the aerosol size distribution, but unfortunately, the aerosol scheme is not currently configured to accommodate multiple aerosol species. We agree that it is fair to ask what impact a revised aerosol scheme would have on the results from this study, but do not currently have the tools necessary to answer that question.
Having said this, the droplet freezing parameterization used favors freezing of the largest droplets. It is likely that droplets activated on larger coarse mode aerosol particles will fall under this category, while droplets activated on smaller aerosol particles will struggle to find sufficient supersaturation to grow to sizes where freezing results. Therefore, while the potential influence of insoluble material on liquid nucleation and growth is not accounted for, the droplet freezing parameterization limits the ability for these small droplets to freeze.

2. I like the attempt to analyze the sensitivity of the immersion freezing parameterization (Eq 1) to various parameters, but I find much of the discussion in section 2.2 to be confusing. It would be more instructive to illustrate how the freezing rate changes (in relative terms) when uncertain (e.g., Bhi) or variable (e.g., Vd, Ta) parameters are altered within a probable range. For example, the freezing point depression contribution for aerosol with a dry size of 6 microns is practically irrelevant here since there are virtually no such large particles in the presented simulations. For the vast majority of considered CCNs, the freezing point depression for droplets larger than 1-2 micron would be very close to zero. On the other hand, varying Bhi by five orders of magnitude or so would be comparable to changing the droplet temperature (deltaTa) by about 10 degrees. This brings up a question: Is a series of 2D model simulations needed to demonstrate, for example, that the insoluble aerosol type affects the cloud structure more that accounting for freezing point depression? The study currently considers 17 different simulations, many of which are not discussed in any details. Dropping a few simulations from consideration would also can help to make the discussion of others more structured and systematic.

We have modified section 2.2 to (hopefully) clarify it. We have also updated Figure 1 to clean it up and make the labels more clear. As to whether the 2D simulations are necessary – I believe that Figure 1 demonstrates that the insoluble mass type is most influential in terms of affecting the freezing rate. However, establishing how these influences play out dynamically and radiatively ultimately requires more complex simulations. The 17 cases included here were designed to cover a wide range of conditions, and with the exception of the BIGNFPD simulation, we believe all are presented with enough detail to justify their existence. We have removed BIGNFPD from table 2.

3. The presentation and the analysis of the results can be improved. Surprisingly for a study on aerosol effects on clouds, the paper does not present any results on simulated droplet or ice number concentrations. (A caption to figures 6,8, and 11 list ice number concentration but the corresponding panels are not found in these figures.) Providing *this information would help to put the widely varying properties of the simulated clouds in perspective. For example, does any combination of IN and CCN properties result in a more realistic simulation than others? Do some aerosol properties lead to completely unreasonable cloud microphysics? Figures, in general, need some work. Many green and red lines are hard to distinguish, a number of 2D plots are too small, and captions often do not correspond to the content shown.*

While concentration was not explicitly presented in the figures, there was discussion on the impact of droplet and ice concentrations in the text (e.g. p. 22074, line 21; p. 22075, line 16; p. 22077, line 7), in addition to text that discusses the impact of droplet size on processes such as precipitation and riming rates. Admittedly, this discussion is limited in scope. As outlined in the text, for these simulations IN (as defined by the model) do not impact these simulations at all since ice nucleation processes that would utilize IN (deposition, condensation and contact freezing) are not active. Therefore, only CN are relevant to the current work. In that senst, yes, there are simulations that are not realistic when compared to observations for this time period, such as those that lead to rapid glaciation of the liquid layer. This is not the result of CN/CCN concentration, however, but rather demonstrates the strong modulating impact of the freezing efficiency.

With respect to the figures, we have updated some of them. It would have been helpful for the reviewer to state precisely which green and red lines are hard to distinguish and which 2D plots are too small. We believe that the current figures are legible, but if there are specific examples we'd be very happy to revisit specific elements. We do agree that some of the captions were outdated and we have updated them accordingly.

4. Is an overcrowded flowchart in fig 14 really needed to make a point that the system is complex and that many interacting processes operate in a mixed-phase cloud? There are many other processes that could be included and more arrows could be drawn as well. What is not clear is how this study contributes to the refinement of this conceptual model. The authors are upfront that they do not quantify the strength of the indicated feedbacks. So what is the purpose for this diagram? Does this study identify any new processes or feedbacks not previously reported? It is not clear that it does, but if so, the paper should make a stronger case on what these processes are, how they change our understanding of the mixed-phase cloud system, and when they would be most important to account for. Basically, the paper should tell readers why they should care about these effects.

All of the reviewers brought up that this figure was distracting to the rest of the work and that discussion of this figure was incomplete. Therefore, we have decided to remove it from the manuscript. Additionally, we have updated the discussion section with an aim of including limited cloud system discussion as an example of how aerosol-induced changes to microphysics can have far-reaching impacts on the cloud system as a whole.

Technical comments

p.22060, ln5: remove "the"
This has been done.

p.22060, ln13-16: Sentence is not clear.
This sentence has been reworded to say "Alteration of the aerosol properties in simulations with identical initial and boundary conditions results in large variability in simulated cloud thickness and lifetime, ranging from rapid and complete glaciation of liquid to the production of long-lived, thick stratiform mixed-phase cloud".

p.22061, ln13: Findeisen
Corrected – thanks!

p.22064, ln15-16: Be more specific on how you set your size grid, or just say that you use 40 bins. Saying that you split them in two groups of 20 does not add much.
The reason we split into two groups was that the first group of 20 (<5 microns) was considered to be haze, for which droplet freezing does not occur, while the second group of 20 were considered to be cloud/rain droplets for which freezing does occur. Having said this, we have eliminated the splitting into two groups but have preserved a statement about the bin sizes not representing a linear spectrum.

p.22068, ln7: "from 10 to 35"
We have modified this as suggested.

p.22070, ln9: relative to what simulation?
This should have said "relative to observations" and has been corrected.

Table 1, figures: use consistent notation in text, table, and figures. E.g., it is "A" in table, "a" in text (p22066); there is a mixture of dT and deltaT in text and figures, etc
We have gone through the document to make notations used consistent between figures, tables and text.

Anonymous Referee #3

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.*

We would like to thank the reviewer for their suggestions and insight. Hopefully the modifications we have made in response to these comments and those of the other two reviewers help to improve the manuscript.

<u>Specfic Comments</u>

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.

Unfortunately neither the study of Diehl and Wurzler, nor the studies cited within this work, provide an in-depth analysis of uncertainty associated with their freezing efficiency parameters. This means that our results are only meaningful if the range of freezing efficiencies tested is significantly larger than the uncertainty associated with the freezing efficiency of any one material. Having said this, out of the aerosol properties tested (soluble mass fraction, insoluble mass freezing efficiency and aerosol number concentration), only the insoluble mass freezing efficiency is perhaps largely uncertain. The range of values chosen for freezing efficiency is rather broad (4-5 orders of magnitude), and while the values themselves may be uncertain, the influence on the cloud is less so. Therefore, while we cannot definitively state that kaolinite does one thing while soot does another, we can state that based on the current parameterization, the freezing efficiency of a particle generally plays the largest role in determining how active droplet freezing may be in a given environment, unless the freezing efficiencies are uncertain enough to cast significant doubt on the range of values tested. Section 5 text has been modified in order to include a statement regarding the possible uncertainty in this result.

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.*

The reviewer is correct in that this was done solely for computational efficiency. The microphysics scheme used would not be practical for multiple 3D simulations of this sort. The 2D simulations took roughly 3-4 times longer than the period simulated (e.g. 36-48 hours to complete a 12 hour simulation). Completing these in three dimensions would have taken too long (particularly given the need to rerun the entire set of simulations several times!). We have explicitly stated in the text that this was our reasoning for running in 2D, and have included the Bretherton et al. reference as an example of a work that demonstrates the potential hazards of doing so.

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.

We completely agree that this is an issue. While we are not defending this practice, the simulations were completed in this manner in order to match the experimental set up of the recent SHEBA-based model intercomparison study (Morrison et al., 2011). We do include mention of this as a potential problem in the discussion in Section 5, and state that we do not expect the simulation results to replicate observations in part because of this shortcoming. With some digging through old code (the latest version of these simulations were completed about 2.5 years ago), I suppose that it is possible to re-run a limited set of these simulations turning off all ice processes for the first 2 hours or so to demonstrate the impact of liquid-water spin up. This would take some time, but could likely be done if the reviewer and editor specifically require it for publication of this work.

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.*

These are valid questions and demonstrate to us that our explanation of these figures is not clear. First, these terms are not supposed to balance, as they do not include all relevant processes (e.g. sedimentation of liquid). In response to the comment about liquid water condensation tendencies, the reviewer is correct in their assessment that precipitation is aiding in keeping the simulations at steady state. Both the SOO and NOICE simulations have some liquid precipitation (which explains the positive condensation rate below the cloud layer) and the SOO simulation also features removal of liquid water through droplet freezing. In the end, we decided that the tendency figures were not necessarily adding much to the analysis, so we decided to limit them to only the droplet freezing and ice depositional growth tendencies, which are discussed in the most detail within the manuscript. The text has been updated accordingly.

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.*

This is a very good point, and the droplet size distributions demonstrate that both effects appear to be contributing to evolution of the liquid droplet spectrum. This new figure (Figure 9) demonstrates that while there is an increase in the amount of small droplets with increasing soluble mass fraction due to the decrease in critical supersaturation, there is also an increase in the size of the largest droplets and the number of larger droplets due to an increase in critical radius. Therefore, the net impact of increased soluble mass fraction appears to be a broadening of the droplet spectrum, which ultimately still results in more droplet freezing. This information has been added to the manuscript in Section 4.4.

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.

We have removed Figure 14 from this work and reworded some of our discussion in Section 5. In general, the text edits were meant to include some of the system complexity discussion in the framework of impacts of aerosol properties on the cloud (and how they likely extend beyond the first order effects through feedback loops).

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.

When we first began setting up these simulations (nearly 5 years ago!), we did perform some sensitivity studies to evaluate the impact of bin resolution. I cannot remember exactly what range of values was tested, but the current resolution was selected as being the most efficient without providing wildly different results from higher 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.

This change has been made as suggested.

<u>Technical Comments</u>

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.

The reviewer is correct – the reduced cloud cover that is discussed in Kay et al. is not specifically stated to include mixed-phase clouds. It was inappropriately phrased in this way in the manuscript based on the author's own knowledge about the occurrence of cloud types in the portion of the Arctic discussed in that paper, and based on the relative radiative influence of different Arctic cloud types. Nevertheless, Kay et al. do not specifically state that mixed-phase (or even liquid-containing) would be largely responsible for many of discussed changes to the radiative budget. Therefore, we have re-written this sentence to state: "Stratiform mixed-phase clouds have been shown to commonly occur and impact the surface energy budget at high latitudes (e.g de Boer et al., 2009; Shupe et al., 2006}, and cloud-induced changes to the surface energy budget have been hypothesized to contribute in modulation of sea-ice extent (Kay et al., 2008).

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.

This is correct. We have clarified this in the text by adding "(in the case of Harrington and Olsson, 2001)" to the statement about IN removal via precipitation.

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.

The reviewer is correct and this should be mass, not size. This has been changed in the manuscript.

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?*

To clarify, the aerosol budget works as follows: In regions with cloud present, CN are removed via CCN activation. As long as a cloud is present (Nd>1 cm-3 or Dd>1 micron), CN continue to be adjusted in this manner. Once there is no longer a cloud present, CN are reset to the initially assigned distribution. The CN spectrum in the cloud is allowed to evolve via its number concentration and mean mass. The standard deviation is held constant. Precipitation scavenging of aerosol is not considered in these simulations. This method follows that of Phillips et al., 2007 (JAS). We have updated the text to clarify how aerosol are handled.

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.

Yes, a bin can experience partial activation, as calculated by the ratio of supersaturation to the amount of water necessary to result in complete activation of a bin. We have reworded this section and (hopefully) clarified it. The word "potentially" was used because at the time of calculation of water necessary for activation of the whole bin, the fraction of aerosol to be activated is unknown (and we did not want to imply that the whole bin would be activated).

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.

We have updated the text so that deltat in equation 1 matches that in equation 3 and beyond. Additionally, deltaTd in equation 3 has been changed to deltaTf to match equation 1.

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.

The reviewer is correct and CCN has been changed to CN where necessary throughout the text.

8. P. 22069, lines 20. The dynamical timestep is 2 sec, but what is the microphysics time step?

The microphysics timestep is 0.5 seconds. This information has been added to the text.

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.

The reviewer is correct and this has been modified to say "an order of magnitude".

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?
This is exactly what is meant and the text has been updated to clarify that this is the case.

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.
This is a good suggestion, and we have changed wave-like to oscillating.

12. P. 22072, line 17. "...is reduced in the SOO simulation...". Reduced relative to what? The NOICE simulation?
Yes, relative to the NOICE simulation -- this has been clarified in the text.

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.
This is mostly correct, though we had not explicitly made the link to removal of water by precipitation (since this is water vapor tendency, and not total water tendency). We have changed the sentence to say: "Because there are very few hydrometeors present in the MON and ILL simulations, the large scale advective tendency along with evaporation of any liquid droplets results in a net positive vapor tendency throughout the column in those simulations, resulting in an increase of water vapor with time", which we believe clarifies that we mean water vapor is increasing with time due to very limited condensation and reduced depositional growth of ice crystals in the simulations with reduced hydrometeor amounts.

14. P. 22076, line 1. See comment #11 above.
This statement has been updated to change "wave" to oscillation.

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.
We agree that this should have been explained better. The mechanism resulting in increased LWP involves the production of larger (faster falling) ice crystals through the production of a small number of (relatively) larger droplets via coalescence. Because this results in a small number of large crystals (instead of a larger number of smaller crystals), fewer water vapor is removed via the Wegener-Bergeron-Findeisen process (resulting in lower water evaporation rates) and the sink mechanism that is ice precipitation removes a smaller amount of total water from the cloud level. When combined these elements result in larger LWP. We have

updated the manuscript to (hopefully) clarify this sequence of events.

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.*

This is correct and we have modified the figure and caption to reflect that this is the case.

17. P. 22092. What are the "droplet volume distributions"? This is never clearly defined in the paper.

These are distributions of droplet volume at a given time in the simulation (in the form of box/whisker plots). The caption has been reworded to clarify.