

Response to Reviewer #1:

The study titled “**Relaxation times of Arctic mixed-phase clouds to short-term aerosol perturbations under different surface forcings**” by Eirund et al. illustrates how mixed-phase clouds, modelled using large-eddy simulations, respond microphysically to short bursts of high aerosol number concentrations, such as those which would be experienced in the vicinity of shipping emissions. By simulating two cloud scenarios – one over sea ice, the other over ocean – the authors show how the surface conditions, moderated by chosen sensible and latent heat fluxes, can affect how the clouds respond to the influx of high aerosol particle number concentrations.

The study builds upon previous work using measurements from the Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaign, and tests these observations with a more complex model representation of aerosol-cloud interactions than has been done before; therefore, the results are an important addition to the scientific literature. However, before publication, I have a few concerns which I feel should be addressed. The study has some potential implications for the stability of Arctic clouds in the face of increased shipping emissions across the region; however, these are not suitably discussed at present.

Furthermore, the authors come close to repeating some conclusions from Young et al., 2016a and 2017, and should distinguish the novelty of their results more so from these published works.

Thank you very much for the detailed review. We incorporated your suggestions within the revised manuscript, which substantially improve the quality of the manuscript. In response to some of the criticisms raised by both reviewers, we discovered two errors in our previously submitted model simulations that affected our results substantially. These two errors were found within the aerosol- and two-moment scheme, which induced an accumulation of aerosol above the cloud within the inversion layer. In more detail, the COSMO model has several clipping routines, where the cloud droplet number ( $N_{\text{drop}}$ ) gets clipped, when cloud water content ( $qc$ ) falls below a minimum threshold ( $1 \text{ e}^{-15} \text{ g m}^{-3}$ ). Within these clipping routines it is necessary to fill the cloud condensation nuclei (CCN) budget again with the amount of  $N_{\text{drop}}$  that have been removed in order to conserve number. We generally included this adjustment in the code, but we were missing to include the CCN adjustment in one routine. Here,  $N_{\text{drop}}$  was set to a minimum value ( $2 \text{ cm}^{-3}$ ) for  $qc > 1 \text{ e}^{-9} \text{ g m}^{-3}$ . This routine induced an artificial source of CCN above cloud top. Given the stable stratification in the inversion, the CCN released above cloud top were only re-entrained at very slow timescales into the cloud. Additionally, we had to adjust the weights calculation that determines the redistribution of CCN after evaporative processes. Through a miscalculation in this routine, CCN were lost within the boundary layer, which impacted our total  $N_{\text{drop}}$  throughout the simulation. As these two errors had compensating effects on the total CCN budget, we discovered them only once we had the first one fixed.

In the substantially revised manuscript we now focus more on the ocean/sea ice difference and the microphysical pathways of the cloud response to aerosol perturbations rather than the timescales of cloud adjustment. However, many of your comments remain valid, such that we included many your suggestions in the revised manuscript. Some sections of the previous manuscript were deleted/rewritten, which we clearly stated in the specific answers to your comments. Please find below our responses, which we marked in red.

#### **General comments:**

1. The paper does not suitably cite previous work on this case study from the ACCACIA campaign. The work is novel in its prognostic representation of both CCN and INP; however, similar studies have been already conducted which compare cloud microphysics over sea ice and over ocean. Please ensure all previous literature is cited more appropriately. For example, the differences between the boundary layer structure over sea ice and over ocean has been discussed as an observational study by Young et al., 2016a, and large-eddy simulations of this case study have been presented by Young

et al., 2017. Observational conclusions should not be repeated here as conclusions of this study unless these earlier works are cited appropriately.

We thank the reviewer for pointing out that we were not sufficiently clear in distinguishing the novelty of this work from previous works. For a thorough comparison to work performed by Young et al., 2017, we included their LES simulation profiles (as shown in Figure 7 from their study) into our Figure 4 and Table 2.

Moreover, we emphasized more strongly in the text, which results were already obtained from observations and which ones are new.

2. The same boundary layer properties are used to compare how clouds form over the ocean or sea ice from the same state. Whilst this is an interesting perspective, why did the authors not use a boundary layer profile measured over the sea ice for these simulations? Do the authors expect the resulting cloud to compare well with observations when the initial profiles used are not the same as that measured? The boundary layer over sea ice is different to that over the ocean (as presented by Young et al., 2016a; 2017); therefore, can the authors comment on why they used the oceanic profile for the sea ice simulations?

Concerning the boundary layer profile over sea ice and ocean we included a more detailed explanation in the text.

Initially, we did use the sea ice dropsonde profile to initialize the modeled sea ice case, however, the profile was too dry to simulate a mixed-phase cloud for the given atmospheric state. Hence, we used the profile over the open ocean, which also provides a better comparison to the modeled open ocean case and reduces differences between the two cases solely down to differences in surface fluxes. We agree with the reviewer that a quantitative direct comparison of the sea ice case with the observations obtained over sea ice (as in Young et al., 2016) is not completely valid, since the initial state differs. However, we argue that a qualitative response (i.e. lower liquid and ice water content over sea ice than over the open ocean, higher cloud top and cloud base over the open ocean) is still valid and is consistent with observations from Young et al., 2016.

3. Readability and clarity could be improved – for example, it is often not clear whether the model simulation results or measurements are being discussed.

We improved clarity and readability and introduced a naming convention to distinguish more clearly between the observations and simulations (see Table 1).

4. The Discussion section could be significantly improved – it currently focuses on validating findings against previous studies; however, there is an opportunity to compare with previous ACCACIA studies which is currently not being capitalised upon. Specifically, there is an opportunity to conduct comparisons with the LES findings of Young et al., 2017 – your results are similar, and therefore there is scope to make some preliminary statements about the ability of two different models to reproduce these observations.

We included the comparison with results from Young et al., 2017 in the results section (see comment 1) and in the discussion.

5. The INP perturbation experiments are lacking analysis and discussion. Please add to this section of the study or remove it.

Thank you for pointing this out. We agree that the section discussing INP perturbations lacked more in-depth interpretation. We added more analysis to section 5.2 and additional discussion on the INP perturbation experiments in section 6. In particular, we included a summary of the mean cloud

properties for the simulations perturbed by INP in Table 3, as well the temperature sensitivity simulations for a perturbation of  $10 \text{ INP L}^{-1}$  in the appendix and a panel demonstrating the effect of INP perturbations in our final schematic summary (Fig. 11).

6. The authors have the opportunity to make some preliminary statements about the stability and microphysical response of Arctic MPCs in the face of pollution transport/shipping emissions (as suggested in the Introduction); however, little discussion of this is included. Please comment on the potential real-world implications of this modelling study.

First, we added references for our chosen CCN and INP perturbations. Our chosen numbers are within the range of CCN observed in ship exhaust plumes (Hobbs et al., 2000) and Arctic haze (Rogers et al., 2001). Additionally, we added more discussion on a comparison to MPC satellite observations in ship tracks from Christensen et al., 2014 in section 5.1.

To make assumption about the future impact of sea ice loss and pollution in the Arctic (as stated in the Introduction), we included a statement at the end of our conclusions (page 22, line 30ff).

### Specific comments:

#### Abstract:

#### Page 1:

**Line 4:** was ACCACIA conducted in the central Arctic? I would've taken this to be >80N?

Changed to "European Arctic".

**Line 5:** define COSMO

Defined.

**Lines 6–11:** these findings read very similarly to those presented by Young et al., 2016a; 2017. Are they conclusions from your modelling work? It currently reads like conclusions from the measurements used to initialise the model – measurements that have already been published. Additionally, LES studies of this case study have already been published. Please distinguish your conclusions more so from these studies to highlight its novelty.

We added that our obtained results agree with observations, to point out that these findings are rather a reproduction than a new result (page 1, lines 8-9).

**Line 12:** "two dynamically different regimes" – this is quite vague, can you expand on this?

We changed this line to "sea ice and open ocean cloud regime" (page 1, line 13).

**Line 16:** "the maximum response" – response in what?

No longer applicable. Entire abstract was rephrased.

**Line 18:** Could you specify more about your aerosol perturbations here? For example, their duration/altitude?

We added this information on page 1, line 13ff:

*"Perturbation aerosol concentrations relevant for CCN activation were increased to a range between 100 to 1000  $\text{cm}^{-3}$  and INP perturbations were once doubled 15 as compared to the background concentration and once increased by a factor of 3 (at every grid point and at all levels)."*

**Line 20:** Can you say more here about how the aerosols are transported out of the boundary layer? Previously the aerosols had been accumulating in the inversion layer above cloud top, as explained in the first paragraph. However, this was due to a bug within the simulation, which has been resolved for the revised manuscript.

#### 1. Introduction:

#### Page 2:

**Lines 13-15:** Please rephrase – sentence meaning unclear

Sentence changed to "Due to their strong radiative impact, MPCs can alter the Arctic climate system (e.g. Bennartz et al., 2013; VanTricht et al., 2016), potentially accelerating or slowing the current high latitude warming" (page 2, lines 15-16).

**Line 16:** "amount" is vague – "fraction"?

Changed to "fraction" (page 2, line 17).

**Line 23:** Please provide references for the sentence ending “*potential implications for cloud dynamics*”.

We added references from the follow up section on cloud dynamics (Schweiger et al., 2008; Sotiropoulou et al., 2016; Young et al., 2016; Young et al., 2018).

**Line 23:** Please define ECMWF.

Defined.

**Line 29:** please rephrase

Rephrased to “*These observed changes in cloud height were also observed during the Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaign (Young et al., 2016). Besides, the authors reported fewer and larger cloud droplets as well as increased precipitation rates over the open ocean as compared to over sea ice.*” (page 2, lines 28-31).

**Line 34:** Can the Arctic still be called pristine? It is cleaner than the mid-latitudes and is very clean in the summer, but the Arctic haze strongly increases aerosol mass concentrations in the Arctic atmosphere during the spring, where the data used to initialise these model simulations was collected. Please consider rephrasing this statement.

That is true, but even in spring the Arctic is still more pristine than other regions on Earth, especially the mid latitudes (Moore et al., 2013, Schmale et al., 2018). We added this clarification in the text (page 3, line 3).

## 2. Model description and setup

**Page 3:**

**Line 27:** Dropsondes were discussed in detail in Young et al., 2016a – please cite here for reference.

Included.

**Lines 29-31:** Spatial domain size and resolution are specified, please include similar information regarding run length and temporal resolution.

Included (page 4, lines 8-9).

**Page 4:**

**1st paragraph:** Solomon et al., 2018 (ACP) use the DeMott et al, 2010 (PNAS) parametrization, not the DeMott et al, 2015 (ACP) parametrization. They use a prognostic derivation of the temperature-dependent fit from Fig. 2 of DeMott et al., 2010, removing the dependence on aerosol number concentration from the standard version of the parametrization. Please can the authors clarify which ice nucleating particle parametrization they used, and note whether they included the aerosol number concentration dependence or used the version (described in Solomon et al., 2015, ACP) which is dependent only on temperature? DeMott et al., 2015 (ACP) is designed for mineral dusts; therefore, if this relationship was used, can the authors comment on the validity of doing so in the Arctic where there is some dust (Young et al., 2016b, ACP) externally mixed with other aerosol species?

We clarified in the text that for our study (as for Possner et al., 2017), the implementation is the same as in Solomon et al., 2015, but the curve to be discretized into the 15 temperature dependent INP bins follows the new DeMott parameterization (page 4, lines 15-17).

**Lines 7-8:** Are these median diameters that are quoted? Please clarify.

No, they are mean diameters. We included this in the text.

**Line 9:** “*low altitudes*” – please specify. Did the authors exclude in-cloud measurements with the PCASP (standard practice)? If so, how?

No, we did not use PCASP measurements within the revised manuscript.

With our fixed aerosol scheme we noted a fast depletion of CCN by precipitation in our *control* simulations, which led to an underestimation of the observed  $N_{\text{drop}}$ . To provide a better comparison to the observed cloud properties from Young et al., 2016, we kept the background CCN concentrations fixed at  $100 \text{ cm}^{-3}$ , which then leads to a more reasonable  $N_{\text{drop}}$  (as compared to observations). Additionally, it matches the fixed  $N_{\text{drop}}$  in the Young et al., 2017 LES simulations for

the same case, which we included for comparison (see General comment 1). Hence, we rewrote this section of the model description (page 4, lines 22ff).

**2<sup>nd</sup> paragraph:**

Please say more about how the aerosol observations are used as input to the model. The PCASP only measures >0.13 micron, do you have any other aerosol measurements for the smaller size mode? When fitting these two modes to the observations, what geometric standard deviation was used for each? Was a lognormal distribution assumed? Did you fit to data collected over the entire ocean segment of the ACCACIA flight in question, and did you use different inputs for the sea ice simulations?

As mentioned above, we used the constant  $N_{\text{drop}}$  in Young et al., 2017 and the  $N_{\text{drop}}$  from the observations as our new reference for choosing our initial CCN concentration ( $100 \text{ cm}^{-3}$ ). Hence, we don't mention PCASP measurements any more.

Generally, our CCN modes are represented as lognormal size distributions. We added the standard deviation in the explanations (page 4, lines 21-22). For simplification, the aerosol input profile is constant with height and we assumed the same aerosol background profile for the open ocean and the sea ice case. Similar to the thermodynamic background conditions, we wanted to keep everything except the surface conditions the same in both control simulations, to reduce differences to those in surface fluxes.

**Lines 10-11:** How did you arrive at this INP concentration? In Young et al., 2016a, the parametrizations listed are temperature dependent and were evaluated at the coldest temperature measured over the sea ice or ocean. Is the same technique used here?  $3.3 \text{ L}^{-1}$  is higher than presented in Young et al., 2016a for PCASP data over both the sea ice and ocean (their Table 3), and seems particularly so given the sensitivity to ice particle number concentration presented by Young et al., 2017. Please provide more information about where this number concentration has come from.

We agree that the cloud properties are highly dependent on the INPs (as indicated by the INP sensitivity experiments and as shown in Young et al., 2017). Given the tremendous uncertainties with regard to secondary ice formation, the INP concentration was constrained to provide realistic INP and IWP within these simulations. Under these constraints we arrived a total INP concentration of  $3.3 \text{ L}^{-1}$  distributed as prescribed by D10 over the 250.5 – 258 K temperature range. We note that this is higher than  $0.5\text{-}1.5 \text{ L}^{-1}$  during ACCACIA and ISDAC, but we wanted to prevent a large underestimation of the ice phase in our simulations. With this initialization we are able to reproduce LES results from Young et al., 2017 using the ACC-ice parameterization (Table 2 and Fig. 4).

**Line 14-15:** How did you arrive at these values? They are within the range measured (Young et al., 2016a), yet they are very specific choices which should be justified.

We did some sensitivity analysis towards higher surface fluxes but we noted that with stronger surface fluxes, updraft speeds increased such to create stronger convective cells. For increased convection and stronger cloud organization the domain size should be increased. As the response to higher fluxes was linear and to save computational power without increasing the domain size even more we kept the fluxes at the lower end of the observations. We included this explanation in the text (page 4, lines 32-35).

## 2.1 Model perturbation experiments

**Line 22:** are all model analyses taken after 1.5h? Is this taken to be the spin-up period of the model? From Fig. 6, it looks like the spin up may be until 2h? Can the authors discuss whether other diagnostics (such as TKE or W) were used to define the spin up? Additionally, how was the perturbation time chosen? Was this taken to be immediately after spin up (inferred from Fig. 6)? Spin-up was identified after the major precipitation event was over in the beginning of the simulation. We added the spin-up length to our model setup description on page 4, line 9. As we wanted to capture the full aerosol response and our simulation time was limited due to computational cost, we could not delay the aerosol injection further.

The perturbation injection was chosen to be directly after the initialization. For clarification we rephrased this in the text (page 5, lines 6-7).

**Lines 22-23:** Why is the pollution mode smaller? I agree it should be, but more could be done to justify this choice. Why was 0.19 micron specifically chosen? Again, is this the median diameter of the mode?

The pollution mode is motivated by anthropogenic particle compositions such as sulfates, nitrates, black carbon and organic carbon. These particles are on average smaller than natural aerosol sources such as sea salt and dust. With the pollution aerosol mode we wanted to include both, pollution from ship exhausts as well as transported aerosol from the mid-latitudes representing Arctic haze during spring (Law and Stohl, 2007). With a size of 0.19 micron we are still within the range of ship emissions (Hobbs et al., 2000) and include similar particles as the background mode.

As wanted to ensure the perturbation mode CCN to activate after the background CCN, its size needed to be slightly smaller due to its implementation in the code (as now included in the text, page 5, lines 9-10).

**Line 26:** This reads like you have both doubled the background and increased by a factor of 3 (i.e.  $\sim 16.6 \text{ L}^{-1}$ ) – please clarify

This has been clarified in the text (page 5, line 15).

**Line 28:** Please provide references for this statement.

We added a reference (Devasthale and Thomas, 2012).

**Line 21:** How long is a single time step? (see previous comment on temporal resolution)

One single model time step is 2 s, this has been added in the model description (page 4, line 9).

### 3. Evaluation of background state

Page 5:

**1<sup>st</sup> paragraph:** Here the authors seem to jump between the simulations and the initial conditions and discuss these interchangeably. Please describe the initial conditions (dropsonde measurements) first, then the controls to avoid confusion.

We changed the paragraph by first describing the observations as reported in Young et al., 2016 and then describing the simulated profiles shown in Figure 1.

**Line 9:** *ocean\_control* is used interchangeably to refer to the observations and the control model simulations.

This label should refer to model results only. Please use a different label (e.g. *observations*) to describe the dropsonde measurements. Also, it lists the mean  $N_{ice}$  is  $0.17 \text{ L}^{-1}$  in Table 2?

As you suggested, we use the label *observations* to describe the observed characteristics from Young et al., 2016 to avoid confusion.

Thanks for pointing that out, that was a previous typo in the text. As our simulations changed due to the new model setup, we adjusted all values in Table 2 and the text accordingly.

**Lines 11-13:** Did you try changing the background concentration of CCN to improve the cloud microphysical agreement with observations?  $3.39/3.99 \text{ cm}^{-3}$  is significantly less than observed – are these comparisons with the observations robust? There are significant differences in cloud base/top height and cloud properties, the study would therefore benefit from some discussion on why this is the case and how these differences may affect the real-world implications.

This underestimation was largely due to a bug within our simulations, which induced an unrealistic vertical distribution of aerosol and low CCN concentrations within the cloud and sub-cloud layer. This has now been fixed. To further avoid a slow decline in CCN throughout the simulated period due to losses by surface precipitation, the background CCN were now held constant. This section is updated accordingly.

**Line 14:** Why do those aerosols in the inversion layer not activate? Is it sub-saturated? Is there too much competition for water vapour?

Yes, the air above cloud was sub-saturated and the entrainment into the cloud occurred on very slow timescales. However, this no longer applies to the bug-fixed simulations as the strong aerosol accumulation above cloud top is resolved.

**Line 15:** Please provide a comment on the realism of this finding (poor re-entrainment of aerosols due to low turbulence).

Also this is resolved with the new setup.

**Lines 15-17:** Please clarify. Are you talking about the dry run where you have  $N_{\text{drop}}$  formation? How is this possible if the boundary layer is kept below water saturation?

Resolved with new setup/statement deleted.

**Page 6:**

**Line 1:** Do you show this anywhere? (Mixing of CCN from above)? Or is it inferred from Fig. 2? Perhaps some  $w'$  tendencies could be shown to illustrate upwards/downwards motion (if these diagnostics are available)?

Resolved with new setup/statement deleted.

#### 4. Surface flux impact on cloud dynamics

**Page 7:**

**Lines 1-2:** Young et al., 2018 discuss similar simulated effects from oceanic surface fluxes using these observations for initialisation – please consider cite/comparing here.

As we compare similarities/differences to other studies in the discussion, we added a comparison to Young et al., 2018 in the discussion (page 17, lines 29-31). Thank you for the additional reference.

These results agree nicely with our open ocean case.

#### 5.1 Response to CCN perturbations

**Page 9:**

**Line 6:** “*is sufficient to significantly perturb*” – please elaborate. By how much? Do smaller perturbations not change the cloud physics as much? There is an opportunity to discuss real-world consequences here.

For the following Figures 7,9 and 10 (showing the LWP and IWP) we changed to showing the mean plus/minus one standard deviation instead of the median. Also, with the new simulations our figures have changed substantially.

Regarding your comment, we now state in the text that a perturbation of  $100 \text{ cm}^{-3}$  CCN is sufficient to perturb the mean LWP by 13% within the first hour upon seeding. To include some reference to observations, we compared that to estimates of Christensen et al., 2014, who found an equivalent change of LWP in ship tracks (page 11, lines 9-10).

**Line 15:** For reference to Fig. 6C to be valid, need to mention IWP here too.

As we rewrote this paragraph the reference to IWP has been moved. We mention IWP and the respective reference to Fig. 7c now on page 11, line 7.

**Page 10:**

**Line 7:** Young et al., 2018 showed that these detraining layers of moisture can be reduced by implementing strong large-scale subsidence. Cloud top increases with time in your ocean simulations due to the heat and moisture fluxes from below – have you looked at the effect of increasing your imposed subsidence to reduce cloud deepening?

No, unfortunately we did not perform any sensitivity studies concerning subsidence rates. However, we plan to investigate the influence of large-scale variability (inversion strength which might impact aerosol detrainment, subsidence, boundary layer stability etc.) in a separate study.

**Lines 10-11:** Why? Opportunity to discuss.

The section comparing the response to CCN perturbations over the open ocean and sea ice has been rewritten/moved to page 13, line 16ff. But the general statement that the response over sea ice lasts longer remains still valid, which is discussed now on page 13, lines 26ff.

**Line 14:** “*most perturbed simulation*” – please quote run label for clarity

Generally, we use the simulation names as stated in Table 1 now for reference to the individual simulations.

**Page 11:**

**Line 4:** “*increase in the ice phase*” – please be more specific, do you mean number concentration? Mass concentration?

In the rewritten paragraph we now refer to “ $N_{\text{ice}}$  and IWP” on page 13, line 1.

**Lines 5-6:** Even though the largest perturbation simulation LWP relaxes back to similar trends as the control simulation, it’s magnitude is still approximately 2-3× that of the control. Given the low LWPs

simulated, this small difference could have an effect on the radiative properties of the clouds. Please discuss.

This has changed now with the new simulations. These statements in the text have been deleted.

**Page 12:**

**Line 1:** Figure 2b does not show transport out of the boundary layer, it just shows non-zero number concentrations. To prove transport, could you show some tendencies (perhaps some relationship with  $w'$ )? Or perhaps a time series of aerosol particle number concentration profiles (like Figures S2-S4)?

This has changed now with the new simulations. These statements in the text have been deleted.

## 5.2 Response to INP perturbations

**Page 13:**

**1<sup>st</sup> paragraph:** Could the authors show how the INP perturbations affect  $N_{ice}$ , in addition to LWP/IWP? Also, there is little analysis on this section's findings in comparison to the CCN perturbations, why is this? As the manuscript stands, this section reads like an afterthought.

We agree that the discussion of the INP perturbations was insufficient in our manuscript and added more analysis/discussion. We added the cloud properties as obtained from the 10INP simulations in our Table 3 (which includes  $N_{ice}$ ).

**Lines 6-7:** Is this illustrated anywhere? If not, please include a figure (like Figure 7) in the supplementary as evidence.

This has been rewritten to *"the stratus cloud over sea ice is initially very susceptible to INP perturbations, which induce an initial peak in IWP and surface precipitation before the cloud returns to the unperturbed state"* (page 16, lines 3-4). For reference, we added a figure showing surface precipitation in the appendix, Fig. S1c,d.

**Side note for Discussion:** The authors show that the cloud does not glaciate (in agreement with other studies). These findings are in contrast to Young et al., 2017's ACCACIA LES results, who use a more simplified representation of cloud microphysics and aerosol-cloud interactions. Could this mixed-phase persistence be because the ice number concentrations are much lower than observed, and modelled in that case? There is an opportunity to compare with their findings, which the authors do not capitalise on. Also, there is a lack of analysis/discussion on the INP perturbation experiments – does the extra ice created by the INP injection precipitate out of the cloud as snow? If so, how does the INP injection affect precipitation rates?

Thank you for this additional discussion point. We included a comparison to Young et al., 2017. As is our new simulations  $N_{ice}$  is close to what has been modeled by Young et al., 2017, we don't think that  $N_{ice}$  is responsible for the lack of glaciation in our simulated case. As  $N_{drop} \gg N_{ice}$  in our simulations (see our Table 3) and also in simulations from Young et al., 2017, we generally think a complete glaciation of the cloud in Young et al., 2017 is very surprising.

We added this as well as a more in-depth comparison to the impact of CCN perturbations in section 5.2 as well as in the discussion.

## 5.3 Invariance of results across temperature regimes

**Page 14:**

**Lines 6-10:** Please improve clarity

The wording in this paragraph has been changed to improve clarity.

**Line 12:** Again, is this the parametrization used? This is not the same as in Solomon et al., 2015.

We clarified this already in the model description section (see comment above, only the implementation is the same as in Solomon et al., 2015, but the newer DeMott parameterization (DeMott 2015) is used).

## 5.4 Consistent response independent of perturbation injection period

Note: this section has been completely removed, as the cloud response to CCN perturbations has been changed with the fixes in the aerosol scheme.

**Page 15:**

**Lines 2-4:** This should be made clearer at the start – to me, it was not clear until now what the aerosol perturbations represented in model terms.

**Side note:** This section seems to be “in response” to some discussion, perhaps it should be relocated to a sub-section of Section 6?

## **6. Discussion:**

### **Page 16:**

**Line 5:** Reference to Young et al., 2016a – this study uses a range of aircraft measurements to show this, and model results here should be presented as successfully reproducing these conditions rather than new conclusions.

*Yes, we included a reference and comparison to Young et al., 2016.*

**Line 11:** As previous, did you try using a sea ice dropsonde profile? Like that presented in Young et al., 2017 for this case? These conclusions are very similar to the observational conclusions of Young et al., 2016a and modelling conclusions of Young et al., 2017. Please reference these studies here – there is a great opportunity to show how these results compare with the previous studies, especially since a more complex microphysical modelling representation is used here. There is novelty in these results; however, the distinction between conclusions from this study and those from previous ACCACIA work is not clear.

*As explained previously we tried but finally did not use the sea ice profile.*

*We also added a comparison to Young et al., 2017, as we also included their data into our results section (Table 2 and Figure 4).*

**Line 13:** “possible pathway for cloud-aerosol interactions” – what is meant by this statement? Not clear/vague.

*We changed this phrase to “As our model setup allows for a prognostic treatment of aerosol-cloud interactions, we are able to quantify the cloud response to spatio-temporally resolved aerosol perturbations [...]” (page 18, lines 10-12).*

**Line 15:** Define  $\tau$

*Throughout the manuscript this has been changed to “cloud optical depth” to avoid confusion.*

**4<sup>th</sup> paragraph:** The authors refer to “polluted”/“unpolluted” – are you referring to the CCN perturbation experiments only? There is no reference to the INP perturbation experiments here, and there is a lack of analysis/discussion on these simulations.

*The focus on polluted/unpolluted periods has been removed in the revised manuscript (see our first comment on page 1 of the response). The quoted values in Table 3 and the text are now averages over the full time period. Additionally, we included the 10INP simulations.*

**Lines 32-33:** It has been previously stated that the perturbation experiments relaxed back to their initial state but the authors have here clarified that there is some difference in magnitude (as per my previous point). This is confirmed in the values quoted in Table 3 between the controls and “post-polluted” rows. Please ensure analysis and discussion is consistent throughout the manuscript.

*This has been removed, as we deleted the division into a polluted and post-polluted period.*

### **Page 17:**

**Line 2:** Do you show aerosol particle transport out of the boundary layer? It is possible I have missed it, but non-zero values above don’t necessarily show that aerosols are being transported vertically. Perhaps some microphysical tendencies (if you have the relevant diagnostics) could show the upward transport of aerosols?

Or a time evolution of aerosol number concentration (like Figures S2-S4)? As it stands, this statement does not seem to be supported by any figure in the manuscript or supporting information.

*With the new set of simulations and the fixes to the scheme this has been deleted/become redundant.*

**Lines 5-7:** There are more caveats to this study than listed here. For example, the fact that a sea ice boundary layer profile was not used for the sea ice simulations is a significant caveat that requires discussion. Why was this not used?

*We do not consider the use of the ocean profile for both cases (sea ice and open ocean) a caveat of this study, as we now (as already explained previously) can clearly attribute differences between the two cases to differences in surface fluxes. In the case of freshly formed sea ice, reduced surface fluxes may impact the overlying clouds. Similarly, surface fluxes increase over polynyas which is also likely to impact clouds (Gultepe et al., 2003).*

We agree that the results might not be valid for a large sea ice covered domain, as there the initial conditions might be different. But also here, season, local conditions, location etc. play a large role as well. We added the point of sea ice observations/model comparison due to the different initial profiles, but as mentioned we don't consider this to be a shortcoming of this study.

Note that this section has been moved to the end of the study, following the conclusions.

## 7. Conclusions:

### Page 19:

**Line 2:** References for "... the subject of a number of recent studies"

We added some references (page 22, lines 2-3).

**Lines 8-10:** Opportunity to link with Young et al., 2018 ACCACIA study (cumuli tower development – intermodal agreement)

We added this link to the discussion and added a reference to Young et al., 2016, 2017, 2018 (page 17, first paragraph of section 6) as well as to the conclusions (page 22, line 10).

**Lines 8-14:** Make stronger links to previous ACCACIA studies and make novelty of results more distinct from previously published conclusions.

See our comment above, we added "*Our simulations support previous results obtained for the ACCACIA campaign (Young et al., 2016, 2017, 2018)*" to this section (page 22, line 9-10).

**Line 18:** "Over sea ice, cloud droplet growth is less efficient..." – why? There has been little discussion of why microphysical processes occur differently over sea ice and ocean.

This statement has been changed to "*This increased moisture flux leads to an increase in the cloud LWP and IWP, larger cloud droplets and ice crystals and a higher cloud base and cloud top*" (page 22, lines 12-13). As mentioned, we relate the (slightly) larger cloud droplets to an increased vertical moisture flux over the ocean through cumuli towers feeding moisture into the stratus layer. Is now also stated in point 1 in the conclusions.

**Lines 25-26:** How? Please provide details.

We removed this statement from the conclusions, as it is not one of the main points of this study but rather a further sensitivity experiment.

**Line 28:** "*possible pathway for aerosol-cloud interactions*" – this statement has been used before and the meaning is not clear. Do the authors just mean that aerosol plumes may affect cloud structure in the Arctic? Please clarify.

This statement has been deleted from the revised manuscript.

## Technical corrections:

**Page 1, line 19:** typo → "**properties**"

Thank you for pointing this out, but this has now been deleted in the revised manuscript.

**Page 2, line 2:** "high **model** uncertainties"

Here, we refer to general uncertainties in cloud physics. Hence, we left the current wording.

**Page 2, lines 12:** "**mid-latitude**"

Changed.

**Page 4, line 23:** "to be **at** slightly smaller **sizes** than"

Changed.

**Page 4, line 25:** "successively" → "**in stages**"

This has been reworded to "*Perturbation aerosol concentrations relevant for CCN activation were increased by 100, 200, 500 and 1000 cm<sup>-3</sup>.*", so explicitly state the concentrations of the perturbations (page 5, line 13)

**Page 5, line 8:** "according to" → "**agreeing with**"

Changed to "*in agreement with*" (page 6, line 12).

**Page 5, line 15:** "In a dry run,..." – new paragraph?

This has been deleted in the revised manuscript.

**Page 7, line 7:** "allow the **cloud** droplets as well as the **ice** crystals"

This has been slightly rewritten in the revised manuscript, but we changed "droplets" to "cloud droplets" (page 8, lines 6-7).

**Page 7, line 15:** “LW cooling is increased up to...” → “**LW cooling increases up to...**” – the former reads like you are modifying the LW cooling, not a simulated effect.

This section has also been rewritten in the revised manuscript, but thanks for pointing this fact out. This has been changed to “[...] *the higher liquid water content in the air column increases the cloud longwave (LW) emissivity*” (page 9, line 8).

**Page 7, lines 15-16:** “The more numerous ice crystals” → “**Higher concentration of ice crystals**”

This has been deleted in the revised manuscript.

**Page 8, Figure 4 caption:** typo → “**interquartile**”

This has been removed as we changed to mean/standard deviation.

**Page 9, line 11:** “upon seeding” → “**after seeing**”

With “*upon seeding*” we refer to the time past the aerosol injection. For clarity, we changed it to “*after seeding*” (note that this paragraph has also been rewritten, but the relevant wording can be found on page 11, line 8 and page 12, line 2).

**Page 10, line 8:** “The initial... on the cloud regime” – remove, vague and not required.

We use this sentence as a transition from the impact of CCN perturbations on the open ocean and the sea ice case. Hence, we decided to keep it, but we reworded to “*The response to CCN perturbations strongly depends on the cloud regime*” (page 13, line 16).

**Page 10, line 9:** “cloud droplet growth is limited”

This has been removed in the revised manuscript.

**Page 19, line 19:** should this be a new paragraph?

We added a new pullet point for the INP conclusions.

**References:** Some references are incomplete or incorrect.

We went through the references and corrected everything that was incorrect.

#### **Figures and Tables:**

**Table 1:** Please list columns as “Background CCN/INP”.

We deleted this column, as it is the same number for all simulations.

**Table 2:** Total values taken over how long? The entire run? Excluding spin up? Please clarify.

- Caption: “*Note that the airplane did not sample the lower and upper levels*” – of what? The model domain? Please clarify/rephrase

We removed this restriction in the analysis and accordingly also in the text. As our maximum liquid water mixing ratio in Figure 4 is at a slightly higher altitude (at approx. 1.55 km height) than in the observations, we didn’t want to exclude this maximum from the analysis by restricting the averaging to heights <1.5 km.

- in-cloud criteria: both the liquid and ice mass thresholds? Or just one or the other? Please clarify, and define  $q_c$  and  $q_i$

We defined  $q_c$  and  $q_i$  in the caption. We used the  $q_c$  threshold for the cloud liquid properties and the  $q_i$  threshold for the cloud ice properties. This explanation is added in the caption.

- Can you comment on the very low cloud base height with comparison to the observations over the ocean? Or the cloud top height which is almost double the altitude of that observed over the sea ice?

We removed the column with cloud extent values, but refer in the text to Figure 4. Our cloud extent was determined only based on  $Q_c$  (no rain, only cloud water). However, we agree that the report of mean values here is confusing. Instead, we calculated the cloud base and top for the stratiform cloud layer, i.e. the layer where 80% of the domain grid points are cloud-covered. The reason our previously calculated cloud base was so low, was because we sampled the updraft cores that are spatially limited, but regions of high  $q_c$ . Now, we added horizontal lines in Figure 4 to indicate the cloud extent of the stratiform cloud.

**Please increase legend size on all figures.**

Changed.

**Figure 1:**

- please choose different colours – hard to read
- improve readability – perhaps split into 4 panels? Over ocean/sea ice?

We split the figures into 4 panels to improve readability. Thus we could also decrease the number of lines and colors.

**Figure 2:**

- How do these profiles compare with observations?

We added a comparison of the simulated  $N_{\text{drop}}$  with observations over the ocean in the text.

- Would it be clearer to have: ice control (black), ice perturb (grey), ocean control (red), ocean perturb (orange)?

To improve readability, we changes the figures to a) ocean case and b) sea ice case and plotted  $N_{\text{drop}}$  and CCN in the same figure, such that these two quantities can be directly compared.

**Figure 3:**

- There is no increase in ice number with decreasing altitude like in the observations (Young et al., 2016a ACP), please comment on this. Similarly, for the LWMR – these trends are in contrast to those observed, please comment on why.

Over the ocean, we reproduce model results of Young et al., 2017 very well. The increase in  $N_{\text{ice}}$  in the observations in Young et al., 2016 are most likely related to a shattering event and hence not physical.

- In caption, define LWMR

Done.

**Figure 5:**

- Why are the panels not shown to 0 m? Or at least to cloud base?

Because we wanted to restrict ourselves to the temperature range ( $T < 258$  K) where freezing occurs (at cloud base temperatures are too warm for immersion freezing as parameterized in our model).

We added this explanation in the figure caption.

**Figure 6:**

- it may be beneficial to show Fig. S4 as additional sub-panels of Fig. 6 to show how the cloud structure evolves with time in the different scenarios

We stayed with only showing Fig. 6 (now Fig. 7), as otherwise the whole figure would be too crowded with more than 4 subpanels. We included a figure in the appendix showing the cloud top for the different sensitivity simulations. The Hovmoeller plots shown in the previous appendix are replaced, as we find our new figure showing the cloud top being of equal importance.

**Figure 7:**

- Just a side note, this figure does not print well (not clear which line is which). Consider changing colours used, or splitting into sea ice/ocean sub-panels?

We removed this figure and included Figure 6, which shows the depositional growth in a Hovmoeller plot. We note that depositional growth is generally most important to contribute to an IWP increase, hence we switched these figures.

**Figure 10:**

- This figure is particularly crowded and individual traces are hard to distinguish. Perhaps separate into further sub-panels? (e.g. ocean+1000CCN, ocean control, ice+1000CCN, ice control)?

We kept this figure, but changed the color of the 1000CCN simulation to orange, to keep it consistent with Figure 7 and increase readability.

**Figure 12:**

- Is precipitation always as rain? Again, do you refer solely to the CCN perturbation experiments for “polluted” case analysis. Please define what you mean by “polluted”, and include some analysis on the INP perturbation experiments (or remove).

Precipitation contains rain and snow, thus we added snow in the schematic. As mentioned earlier, we removed a distinction between “polluted” and “unpolluted” and now included a schematic showing the impact of INP perturbations.