Zürich, May 2019

**Letter to the Editor**

Dear Holger Tost,

We are very thankful to the two reviewers for their effort and the detailed analysis of our submitted paper. In response to some of the criticisms raised by the 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 ($\mathrm{N_{drop}}$) gets clipped, when cloud water content (qc) falls below a minimum threshold ($1\ \mathrm{e}^{-15}\ \mathrm{g\ m^{-3}}$). Within these clipping routines it is necessary to fill the cloud condensation nuclei (CCN) budget again with the amount of $\mathrm{N_{drop}}$ 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, $\mathrm{N_{drop}}$was set to a minimum value ($2\ \mathrm{cm^{-3}}$) for qc > $1\ e^{-9}\ \mathrm{g\ m^{-3}}$. 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 $\mathrm{N_{drop}}$ 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.

After we performed these fixes we reran our simulations and obtained different results as compared to the previous experiments. Overall the new simulations maintained a realistic CCN profile throughout the simulations. However, our background CCN got depleted by precipitation too efficiently, such that we adjusted our initial conditions to fixed CCN in the control simulations, but keeping the perturbations prognostic (similar to Possner et al., 2017).

Concerning the impact of aerosol perturbations we find that over the ocean our conclusion of a buffered aerosol response remains still valid, however the buffering is not due to aerosol transport, but to a shift of the response from the liquid to the ice phase as well as a delayed precipitation increase. In contrast, over sea ice CCN perturbations now significantly impact the MPC throughout our simulated period. 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 the reviewer's comments remain valid, such that we included many their suggestions in the revised manuscript. We addressed their comments in the separate files attached.

We would like to thank you again for your patience and thank the reviewers for helping us detect these errors in our simulations.

Best regards,

On behalf of all coauthors, Gesa Eirund

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 ($\mathrm{N_{drop}}$) gets clipped, when cloud water content (qc) falls below a minimum threshold ($1\ \mathrm{e}^{-15}\ \mathrm{g\ m^{-3}}$). Within these clipping routines it is necessary to fill the cloud condensation nuclei (CCN) budget again with the amount of $\mathrm{N_{drop}}$ 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, $\mathrm{N_{drop}}$ was set to a minimum value ($2\ \mathrm{cm^{-3}}$) for qc > $1\ e^{-9}$ $\mathrm{g\ m^{-3}}$. 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 $\mathrm{N_{drop}}$ 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 studieshave 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 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 $cm^{-3}$ 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 $N_{drop}$. To provide a better comparison to the observed cloud properties from Young et al., 2016, we kept the background CCN concentrations fixed at 100 $cm^{-3}$, which then leads to a more reasonable $N_{drop}$ (as compared to observations). Additionally, it matches the fixed $N_{drop}$ 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).

**2nd 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 $\mathrm{N_{drop}}$ in Young et al., 2017 and the $\mathrm{N_{drop}}$ from the observations as our new reference for choosing our initial CCN concentration (100 $\mathrm{cm^{-3}}$). 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 $\mathrm{L^{-1}}$ 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 $\mathrm{L^{-1}}$ distributed as prescribed by D10 over the 250.5 – 258 K temperature range. We note that this is higher than 0.5-1.5 $\mathrm{L^{-1}}$ 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. ~16.6 $L^{-1}$) – 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:**

**1st 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 $L^{-1}$ in Table 2?

As you suggested, we use the label *observations* to describe the observed characteristics form 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 $cm^{-3}$ 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 $N_{drop}$ 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 $\mathrm{cm}^{-3}$ 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 "$N_{ice}$ *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:**

**1st 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 $N_{drop}$ >> $N_{ice}$ 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 phase 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 τ

Throughout the manuscript this has been changed to "cloud optical depth" to avoid confusion.

**4th parapraph:** 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 clod 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 is 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*$^{-3}$.", 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 qc and qi in the caption. We used the qc threshold for the cloud liquid properties and the qi 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 Qc (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 qc. 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 $\mathrm{N_{drop}}$ 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 $\mathrm{N_{drop}}$ 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 $\mathrm{N_{ice}}$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.

**Response to Reviewer #2:**

**Review of "Relaxation times of Arctic mixed phase clouds to short-term aerosol perturbations under different surface forcing" by Gesa K. Eirund, Anna Possner and Ulrike Lohmann**

This manuscript deals with the impact of short-term aerosol perturbations on Arctic mixed-phase clouds (MPCs). Based on an observed case from the ACCACIA campaign, the authors use large eddy simulation to investigate how the structure and microphysical properties of the modelled MPC change with changing cloud condensation nuclei (CCN) and ice nucleating particle (INP) concentrations, changing surface fluxes (characteristic for a surface covered with ice or open water) and air temperature. The manuscript is in general well written and contains interesting results.

However, there are a few points that I think should be clarified before the manuscript is accepted for publication.

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 ($\mathrm{N_{drop}}$) gets clipped, when cloud water content (qc) falls below a minimum threshold (1 $\mathrm{e^{-15}}$ g $\mathrm{m^{-3}}$). Within these clipping routines it is necessary to fill the cloud condensation nuclei (CCN) budget again with the amount of $\mathrm{N_{drop}}$ 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, $\mathrm{N_{drop}}$ was set to a minimum value (2 $\mathrm{cm^{-3}}$) for qc > 1 $e^{-9}$ g $\mathrm{m^{-3}}$. 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 $\mathrm{N_{drop}}$ 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.

**Major comments:**

1. Introduction: It is not clear to me *why* a short-term aerosol emission perturbation should generate a *long-term* cloud response. Since (accumulation mode) aerosols are efficiently scavenged by precipitation, I don't see a clear reason why the response would last longer than a few hours if you have precipitating clouds? I was actually quite surprised that you did indeed see a quite pronounced response up to almost a day after the perturbation. I think that a motivation would be good to include in the introduction.

Note that we changed the focus of the revised study away from adjustment timescales and the MPC response times to aerosol perturbations, but moved towards the differences in aerosol

response between the open ocean and sea ice regime as well as microphysical pathways in clouds as a response to aerosol perturbations.
Nevertheless, we did expect that with an aerosol perturbation of 1000 $cm^{-3}$, which is 10 times more CCN than the observed value, we would simulate a strong response (i.e. eventually including complete suppression of rain), especially considering the fact that the simulated precipitation (especially over sea ice) was very low (0.59 mm $d^{-1}$ over sea ice and 1.12 mm $d^{-1}$ over the open ocean). Therefore, aerosol concentrations in the boundary layer are depleted over very long time-scales in these simulations and concentrations remain high after seeding throughout the simulated period.
Also, the surface fluxes and higher updrafts over the open ocean provide a source of moisture and high relative humidity, which favors new droplet activation as response to an aerosol perturbation. Hence, we were not surprised to see a strong sensitivity (especially initially) to aerosol perturbations.
According to the shifted focus of our revised manuscript however, we removed the timescale aspect from the introduction completely.

2. Model description and setup: The model description and simulation setup is incomplete. I would suggest adding information on:
a. Which hydrometeors are considered? Is it only one category for liquid and one for ice or do you have separate categories for cloud droplets, rain drops, pristine ice, graupel/hail, snow,….? And what is really $q_c$ and $q_i$?
b. Are any secondary ice formation processes represented in the model?
c. At which altitude is the model top?
d. At which latitude is the case simulated?
e. What is the time step and how long is the integration time?
f. What is the assumed habit of the ice crystals?
g. Is the sounding performed over ice or open water?
Related to the last comment, I'm not convinced that it's appropriate to use the same sounding to initialize the "open water" and "ice surface" simulation. I would assume that a sounding over ice looks quite different to a sounding over water?
We added the following information to the model description (section 2):

a. Hydrometeors: 5 hydrometeor types (cloud droplets, raindrops, cloud ice, snow, graupel) represented as gamma distributions with prescribed shape parameters and prognosed bulk mass and number concentrations
b. In our simulations, only the HP-mechanism is included, other secondary ice processes are omitted. However, due to the very cold cloud temperatures (-15 to -20 °C), secondary ice processes do not play a strong role (Hallett and Mossop, 1974).
c. 23 km model top
d. A box of 20 x 20 $km^2$ around the position of dropsonde 5 release (75°N, 24,5°E)
e. Model time step 2 s with model output every 6 minutes, integration time 20h
f. Snowflakes and ice crystals are assumed to be dendrites
g. We used only the dropsonde over open ocean. Initially we tried to set up the sea ice case with the dropsonde over sea ice, however, the dropsonde profile over sea ice was too dry to simulate a cloud for our setup.
   We agree that over sea ice the boundary layer profile might highly differ from the boundary layer over open ocean (Young et al., 2016). But by having the same initial conditions for the open ocean and the sea ice case we can narrow any differences in cloud dynamics and microphysics down to surface fluxes, which become

increasingly important over freshly melted sea ice or polynyas (Gultepe et al., 2003). This is now more clearly stated in the manuscript.

3. Evaluation of background state:
a. The definition of the cloud extent is confusing (but it may become clearer if point 2a above is clarified). Does the limit of $q_c$ applied to define the cloud boundary include rain? In Section 3, you say that "Our model successfully simulates a liquid topped MPC with ice sedimenting out of the liquid layer in both control simulations, according to observations". But this structure is not really clear from Figure 4. Looking at figure 4, I seems like you have a cloud between approximately 600 and 1500 m (in the ocean case) and that the rest is falling precipitation?
The cloud illustrated in Figure 12 is also quite different from the clouds plotted in Figure 4, the simulated cloud over the ice surface is not substantially thinner than the cloud over ocean (at least not according to the values in Table 2).

Our cloud extent was determined only based on cloud water content (qc), i.e. no rain. However, we agree that the report of mean values here is confusing. As the cloud over the open ocean features convective structures, the calculation of the cloud extent based on the highest/lowest level with qc>0.01 g $m^{-3}$ leads to a sampling of the updraft towers, which distorts the cloud extent of the main stratus layer.
Instead we now calculated the cloud extent of the stratiform cloud layer, which is defined as the layer where 80% of the domain grid points have qc>0.01 g $m^{-3}$. We included the stratiform cloud base and top as horizontal, dashed lines in Figure 4. Thus, everything below and above this line represents convective structures. Note that the black line and shading in Figure 4 is only cloud water mixing ratio (excluding rain mixing ratio), which we also stated more clearly in the figure caption of Figure 4. In our revised manuscript we removed the column with cloud extent values in Table 2 to avoid confusion, but refer in the Text to Figure 4.

Thanks very much for pointing out that deficit in Figure 12, we adjusted the figure (now Figure 11 in the revised manuscript) to better fit our findings from sections 3 and 4.

b. Droplet number concentration: It is not clear to me how you could possibly get a droplet number concentration of 63 or 110 $cm^{-3}$ (as observed over ocean and ice, respectively) in your simulations if you have a CCN concentration that is only 49 $cm^{-3}$. On the other hand, the CCN concentration plotted in Figure 2b is approximately 100 $cm^{-3}$ at ~2000 m. Why is the concentration so high at this altitude? This seems to be much higher than the cloud top, so I don't think it could be transported CCN?

That is indeed true. This very strong aerosol accumulation at the cloud top in our previous Fig. 2b resulted from two errors in our aerosol scheme (as described in the first paragraph). As we changed our setup in the revised manuscript, we initialized our new simulations with a fixed CCN background concentration of 100 $cm^{-3}$. This equals the fixed $N_{drop}$ concentration in LES simulations for the same case from Young et al., 2017 and ensures a more accurate representation of $N_{drop}$ as has been observed during the campaign in Young et al., 2016. Also, we kept the background CCN fixed over time (as in Possner et al., 2017) to prevent a loss of background aerosols by precipitation and freezing. With this new setup we are able to reproduce the observed $N_{drop}$ better than in our previous setup with 48±15 $cm^{-3}$ as compared to 63±30 $cm^{-3}$ in the observations (see Table 2).

4. Robustness to perturbations in microphysics: Figure 6 shows that the LWP increase with increasing CCN concentration is more sustained over ocean than over ice. The authors also discuss this result on page 11-12, but I cannot really find an explanation/hypothesis to why this is the case? And, as mentioned above in comment 1, I was actually surprised to see that the CCN perturbation response was sustained for such a long time after the initial perturbation – in particular for the open ocean case.

Over open ocean updrafts are stronger and the vertical moisture flux is increased as compared to the cloud regime over sea ice. This allows for fast additional cloud droplet activation and formation, especially in the updraft cores (see Figure S2). We included more discussion on these differences between the cloud regime over the open ocean and sea ice in section 5.1 (page 13, lines 16ff) and in the discussion (page 17, first paragraph).
Resulting from the fast increase of $N_{drop}$ over the ocean and the initial precipitation suppression (Fig. S1a), the response of the liquid phase in the cloud over the ocean can be maintained for approximately 16 h, until the response shifts from the liquid to the ice phase through increased ice crystal growth by deposition (Fig. 6c and Fig. 7c).
Over sea ice, total surface precipitation is very low (as mentioned earlier; Fig. S1b) and the ice water path (IWP) is substantially lower as compared to over the ocean (Fig. 7d), such that CCN can perturb the cloud even beyond 20 h.

5. Discussion:
a. The simulated transport of CCN out of the cloud layer is interesting, but also a bit puzzling to me. In another Arctic mixed-phase cloud study, Igel et al. (2017) found that entrainment/mixing *from* the free troposphere could actually be an efficient *source* of aerosols/CCN to the mixed-phase cloud layer. I think this would be worth mentioning/discussing. I'm not sure why you (and Solomon et al., 2018) get a different response, but one possibility could be that the cloud simulated by Igel et al. extended into the inversion, and that the inversion layer and lower free troposphere was actually moister than the below-cloud layer. Another possibility could be that Igel et al., impose a vertical gradient in their aerosol concentrations (based on observations). In general, I think it would be interesting to see how efficient the CCN recycling is in your study.

This aspect has been removed from our revised manuscript, as the aerosol accumulation at cloud top was due to bugs in our aerosol scheme.
As a side note, prior to fixing the model, we did some tests with a varied inversion strength which had little to no effect on that strong aerosol sink at cloud top. However, we plan to investigate the impact of boundary layer stability on aerosol-cloud interactions and cloud dynamics in a future study.

b. Another thing (related to the above point) that would be interesting to know is how well COSMO-LES simulates the entrainment processes at the cloud top. Do you have any observations of TKE dissipation from ACCACIA that you could compare with?

This point also becomes redundant in our revised manuscript, as we don't discuss aerosol transport any more.

c. In general, the authors could extend and contrast their results to other studies on aerosol effects on mixed-phase clouds. The increase in ice water content and subsequent decrease in liquid water content with increasing INP has been described by e.g. Avramov and Harrington (2010), Ovchinnikov et al. (2014), Young et al. (2017), Stevens et al. (2018). The increase in liquid water path with increasing CCN has been discussed in Stevens et al. (2018)

Thanks for these additional references. We extended the comparison of our results to previous ACCACIA work and further studies on aerosol effects on Arctic MPCs.
More specifically, we added a comparison to LES results from Young et al., 2017 for the same case in section 3 as well as in the discussion.
Additionally, we expanded our discussion on INP perturbations and included references additional in section 5.2 and the discussion (e.g., Morrison et al., 2008, Ovchinnikov et al., 2014, Possner et al., 2017, Young et al., 2017, Solomon et al., 2018, Stevens et al., 2018, Young et al., 2018).

**Minor comments:**
Abstract:
1. Although it's mentioned in the title, I think it should be clarified also in the abstract that you are looking at instantaneous aerosol perturbations.
We removed the term "short-term" from the title, as we have moved the focus of the manuscript away from timescales. But we added a statement in the abstract, that the aerosol perturbations are instantaneous (page 1, line 3).
2. The sentence starting with "Motivated by ongoing sea ice retreat…" does not read very well. It's not clear what you contrast with what and that it is model simulations you are referring to.
We changed this sentence to "*Motivated by ongoing sea ice retreat, we performed all sensitivity simulations over open ocean and sea ice to investigate the effect of changing surface conditions.*" (page 1, lines 7-8).

Introduction:
3. Page 2, lines 4-6: Deep convective clouds are also mixed-phase.
Thank you, that is indeed true, we added that (page 2, lines 6-7).
4. Page 2, line 10: "… potentially causing a warming effect…" – why potentially? Isn't the LW (surface) effect always warming?
True, we deleted "potentially".
5. Page 2, line 15: "… eventually accelerating…" – why accelerating? Isn't it also possible that we could have a negative feedback from clouds?
That's true, especially during summer a higher cloud fraction might lead to a net cooling. We changed it to "*accelerating or slowing*" (page 2, line 16)
6. Page 2, lines 25-29: This sentence does not read very well.
We changed this sentence 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 compared to over sea ice.*" (page 2, lines 28-31).
7. Page 2, line 33: Why would sea salt and dimethyl emissions dominate ship emissions? Do you mean that these (sea salt and DMS) emissions generally are larger than ship emissions?
According to Gilgen et al., 2018 the impact on CCN and hence on $N_{drop}$ of natural emissions from the ocean (such as sea salt and DMS) together with a changed future meteorology

exceeds the impact that predicted ship emissions have on the cloud properties. Even with 10-fold ship emissions there was no significant impact on cloud properties.
However, for clarification we rewrote this section to "*An increased availability of cloud condensation nuclei (CCN) resulting from both, sea salt and dimethyl sulfide emissions from the ocean and predicted ship emissions may lead to increased cloud formation and a net surface cooling during summer, as projected by global climate and earth system models (Gilgen et al., 2018, Stephenson et al., 2018).*" (page 3, lines 6-8).

Model description and setup:
8. Page 3, line 30: "km" should be "$km^2$".
We adjusted this to 20 km x 20 km.

Evaluation of background state:
9. Page 5, line 3: I would specify that "both control simulations" refer to the control simulations over open water and ice, respectively (and thereby define *ocean_control* and *ice_control*).
Specified (page 6, lines 5-6).
10. Page 5, line 9: "… lower in our model simulations". Lower than what? I assume you mean compared to observations?
Lower as compared to *observations*, we included that specification in the text (page 6, line 16).
11. Figure 1: Perhaps refer to table 2 for the simulations?
We included a reference to Table 1 (simulation overview) in the figure caption.
12. Table 2: "Cloud extend" should be "cloud extent".
This has been removed from the table (see reply to comment 3a).

Surface flux impact on cloud dynamics:
13. Figure 3: What time step is plotted?
After 3 h, we included that in the figure caption.
14. Page 6, line 5: "Cloud extend" should be "cloud extent".
This has been removed from the table (see reply to comment 3a).

Invariance of results across temperature regimes:
15. Page 13, line 10: I would suggest adding that the RH is kept constant, just as a clarification.
Added (page 17, line 3).

Consistent response independent of perturbation injection period:
16. Page 15, line 9: I would suggest adding "substantial" in between "no" and "change".
This section has been removed in the revised manuscript.

Discussion:
17. Page 16, line 15: What is tau?
We now consistently use the wording "cloud optical depth".
18. Page 17, line 4: Define WRF?
This sentence has been deleted in the revised manuscript (as we do not compare the aerosol accumulation to results by Solomon et al., 2018 any more).
19. Page 19, line 15: I suggest changing "resembling" to "providing" or something similar.
We changed to "providing" (page 22, line 16).

# Response of Arctic mixed-phase clouds to aerosol perturbations under different surface forcings

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**Abstract.** The formation and persistence of low lying mixed-phase clouds (MPCs) in the Arctic depends on a multitude of processes, such as surface conditions, the environmental state, air mass advection and the ambient aerosol concentration.
In this study, we focus on the relative importance of different instantaneous aerosol perturbations (cloud condensation nuclei and ice nucleating particles; CCN and INPs, respectively) on MPC properties in the European Arctic. To address this topic, we performed high resolution large eddy simulations (LES) using the Consortium for Small-scale Modeling (COSMO) model and designed a case study for the Aerosol-Cloud Coupling and Climate Interactions in the Arctic (ACCACIA) campaign in March 2013. Motivated by ongoing sea ice retreat, we performed all sensitivity studies over open ocean and sea ice to investigate the effect of changing surface conditions. We find that surface conditions highly impact cloud dynamics, consistent with the ACCACIA observations: over sea ice, a rather homogeneous, optically thin, mixed-phase stratus cloud forms. In contrast, the MPC over the open ocean has a stratocumulus-like cloud structure. With cumuli feeding moisture into the stratus layer, the cloud over the open ocean features a higher liquid (LWP) and ice water path (IWP) and has a lifted cloud base and cloud top compared to the cloud over sea ice.
Furthermore, we analyzed the aerosol impact on the sea ice and open ocean cloud regime. Perturbation aerosol concentrations relevant for CCN activation were increased to a range between 100 to 1000 $\mathrm{cm}^{-3}$ and INP perturbations were once doubled as compared to the background concentration and once increased by a factor of 3 (at every grid point and at all levels). The perturbations are prognostic to allow for fully interactive aerosol-cloud interactions. Perturbations in the INP concentration increase IWP and decrease LWP consistently in both regimes. The cloud microphysical response to potential CCN perturbations occurs faster in the stratocumulus regime over the ocean, where the increased moisture flux favors rapid cloud droplet formation and growth, leading to an increase in LWP following the aerosol injection. In addition, IWP increases through new ice crystal formation by increased immersion freezing, cloud top rise, as well as subsequent growth by deposition. Over sea ice, the maximum response in LWP and IWP is delayed and weakened compared to the response over the open ocean surface.
Additionally, we find the response to aerosol perturbations being highly dependent on the cloud regime. Over the open ocean, LWP perturbations are efficiently buffered after 18 h simulation time. Increased ice and precipitation formation relax the liquid cloud properties back to their unperturbed range. On the contrary, over sea ice the cloud evolution remains significantly perturbed with CCN perturbations ranging from 200 to 1000 CCN $\mathrm{cm}^{-3}$.

## 1 Introduction

Clouds play a crucial role in the hydrological cycle and the radiative balance of the Earth-atmosphere system. However, clouds still comprise high uncertainties and their behavior under climate change scenarios is not yet well-understood. Hence, the magnitude of the cloud radiative forcing in the upcoming years remains unclear (IPCC, 2013). Mixed-phase clouds (MPCs) contain both phases, i.e. ice and water, and are important for the radiative balance (Lohmann, 2002) and climate sensitivity (Tan et al., 2016). MPCs occur either in regions of deep convection, where the cloud top reaches temperatures low enough for ice formation (Rosenfeld and Woodley, 2000), in mountainous terrain (Lloyd et al., 2015a; Farrington et al., 2016; Lohmann et al., 2016), or in cold regions of the planet, i.e. in high latitudes (Morrison et al., 2011). In the Arctic, MPCs occur approximately 40% of the time (Shupe et al., 2006) and are often observed as persistent low clouds (Shupe et al., 2011). Their radiative forcing at the surface is still ambiguous and determined in part by the distinct seasonal cycle at high latitudes. In summer, the reflection of incoming radiation dominates, while during the rest of the year absorption and emission of longwave (LW) radiation prevails, causing a warming at the surface (Curry et al., 1996). In recent decades the Arctic has been warming at a faster rate than the rest of the globe (Serreze and Barry, 2011). As changes in the Arctic can impact mid-latitude weather conditions, the climate state of the Arctic is important not only regionally but also hemisphere-wide (Cohen et al., 2014; Ye et al., 2018). Due to their strong radiative impact, MPCs can alter the Arctic climate system (e.g. Bennartz et al., 2013; Van Tricht et al., 2016), potentially accelerating or slowing the current high latitude warming.

Arctic MPC fraction and phase partitioning are governed by a multitude of processes operating in conjunction across a wide range of spatial scales; such as the large-scale dynamical forcing, surface processes, as well as the ambient aerosol concentration. The large-scale dynamical forcing determines air mass and hence water vapor advection, which is found to be crucial for the persistence of Arctic MPCs (Morrison et al., 2011; Sedlar et al., 2012; Loewe et al., 2017). With ongoing sea ice loss and the possibility of an ice-free Arctic by mid century (Overland and Wang, 2013), the impact of surface conditions on Arctic MPCs has gained increasing attention in the past decade (e.g. Schweiger et al., 2008; Palm et al., 2010; Vavrus et al., 2010; Liu et al., 2012; Sotiropoulou et al., 2016; Young et al., 2016). A more exposed open ocean surface has potential implications for cloud dynamics (Schweiger et al., 2008; Sotiropoulou et al., 2016; Young et al., 2016, 2018). Schweiger et al. (2008) using the 40-yr European Centre for Medium-Range Weather Forecasts Re-Analysis (ERA-40) product demonstrated that sea ice loss increased boundary layer height and led to more midlevel clouds. In addition, Sotiropoulou et al. (2016) found increased stratocumulus or cumulus cloud formation over the ocean in contrast to thin stratus clouds over sea ice in observations from the Arctic Clouds in Summer Experiment (ACSE) campaign. 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 compared to over sea ice. In LES simulations for the same case, Young et al. (2017) could reproduce these observations and Young et al. (2018) simulated cumuli tower development over a warming open ocean surface, in agreement with previous results of more convective cloud systems over a destabilized surface.

In addition to increased surface heat fluxes, aerosol emissions may increase in the Arctic (Struthers et al., 2011; Browse et al., 2014; Gilgen et al., 2018; Stephenson et al., 2018) which could impact cloud microphysics. Since the Arctic is a pristine environment and aerosol concentration are generally lower than in the lower and mid-latitudes (Moore et al., 2013; Schmale et al., 2018), any aerosol perturbations could significantly impact MPC formation and persistence. With decreasing sea ice, trans-Arctic shipping is also projected to increase, exerting local aerosol perturbations (Hobbs et al., 2000; Khon et al., 2010; Peters et al., 2011). An increased availability of cloud condensation nuclei (CCN) resulting from both, sea salt and dimethyl sulfide emissions from the ocean and predicted ship emissions may lead to increased cloud formation and a net surface cooling during summer, as projected by global climate and earth system models (Gilgen et al., 2018; Stephenson et al., 2018). Locally, aerosols released in ship tracks alone can change cloud liquid and ice water content (LWC and IWC, respectively) as found in studies of Christensen et al. (2014) and Possner et al. (2017). Equivalently, a reduction in the ambient CCN and hence cloud droplet number concentration ($N_{drop}$) could induce cloud dissipation (Mauritsen et al., 2011; Loewe et al., 2017; Stevens et al., 2018). However, disentangling the competing effects of environmental conditions and aerosol disturbances appears challenging (Jackson et al., 2012). In the past, Stevens and Feingold (2009) argued for a buffered aerosol response in certain cloud regimes. For mid-latitude convective clouds, Miltenberger et al. (2018) showed that cloud fraction is not impacted by aerosol perturbations, but that aerosols may affect the organization of cloud pockets with fewer, but larger cloud cells under levels of increased pollution. In simulations of trade wind shallow cumuli by Seifert et al. (2015) an initial aerosol response is seen, with an increased number of cumulus structures and decreased precipitation. Yet the system efficiently returns to an organized cloud structure in a quasi-stationary state after some hours, which is insensitive to the background aerosol concentration. Turbulent mixing and de- and entrainment of aerosols out of polluted regions could potentially also impact the aerosol concentration and the long-term aerosol response, as has been simulated by Berner et al. (2015) in large eddy simulations (LES) of ship tracks in the Monterey Bay. On the other hand, Igel et al. (2017) found that entrainment of aerosols from the free troposphere into the boundary layer represents an important source of aerosol particles for Arctic MPCs as the authors showed in observations and LES simulations of the Arctic Summer Cloud Ocean Study (ASCOS) field campaign.

In this study, we investigate how the response to increased aerosol concentrations may differ for different cloud regimes of Arctic MPCs. For this purpose we perform high-resolution idealized LES to resolve the multitude of boundary layer processes that impact the cloud state. We contrast our results for different surface conditions (open ocean surface versus sea ice) and apply different perturbations across a $\pm 2$ K temperature range. To validate our simulations we use observations obtained during the recent ACCACIA campaign (Lloyd et al., 2015b; Young et al., 2016) in the European Arctic.

## 2 Model description and setup

LES are performed with the Consortium for Small-scale Modeling (COSMO) model in its configuration for idealized LES experiments (Schättler et al., 2000). COSMO-LES has been proven to simulate MPCs in the Arctic with reasonable accuracy (Possner et al., 2017). Here, we simulate a single-layer stratocumulus case during the ACCACIA campaign on March $23^{rd}$, 2013. All simulations are initialized with the dropsonde profile number 5 released during the campaign (Young et al., 2016).

The obtained profiles are smoothed to exclude small-scale variability from the measurements as model input. In addition, the water vapor mixing ratio ($q_v$) was increased by 20% to account for the dry bias in dropsonde data (Ralph et al., 2005; Young et al., 2016). Note that in contrast to Young et al. (2017) we initialize the open ocean as well as the sea ice simulations with the same atmospheric profile, to narrow down dynamic changes in the cloud-topped boundary layer to changed surface conditions alone (i.e. turbulent surface fluxes) and exclude any impact from varying large-scale conditions or boundary layer stability.

The domain covers a 19.2 $\mathrm{km}$ x 19.2 $\mathrm{km}$ large area centered around the location of the release of dropsonde number 5 (75°N, 24.5°E). The horizontal resolution is 120 $\mathrm{m}$, the vertical resolution is variable and specified with 20 to 25 $\mathrm{m}$ within the entire boundary layer and coarser resolution above cloud top up to the model top at 23 $\mathrm{km}$. The temporal resolution is 2 $\mathrm{s}$ and the model has been run for 20 h, including a 1.5 h spin-up period. Radiation is treated interactively according to the Ritter and Geleyn (1992) radiation scheme and includes a diurnal cycle. The cloud microphysical tendencies are parameterized following the Seifert and Beheng (2006) two-moment scheme. The scheme considers five hydrometeor types (cloud droplets, rain drops, cloud ice, snow and graupel) represented as gamma distributions with prescribed shape parameters and prognosed bulk mass and number concentrations. As in Possner et al. (2017) we use a prognostic treatment of ice nucleating particles (INPs) while we keep the background CCN fixed, with cloud droplet activation calculated according to Koehler theory (Nenes and Seinfeld, 2003). Throughout the simulations the CCN are assumed to be pure ammoniumbisulfate particles. Prognostic INPs are implemented as in Solomon et al. (2015). The scheme parameterizes immersion freezing following the DeMott et al. (2015) temperature dependence and captures the depletion and replenishment of INPs. Following the COSMO setup for the model intercomparison performed by Stevens et al. (2018), ice crystals and snow flakes are assumed to be dendrites. As secondary ice processes are observationally poorly constrained, only the HP-mechanism (Hallett and Mossop, 1974) is included in our model, which is inefficient at cold temperatures (-15 to -20°C).

We initialize the simulations with one background mode of potential CCN (0.2 µm mean diameter and 1.5 standard deviation), represented by a lognormal size distribution. For direct comparison to obervations and the Young et al. (2017) model study, the CCN concentrations were chosen to match the observed $N_{drop}$ over the ocean (Young et al., 2016) and the fixed $N_{drop}$ in Young et al. (2017) and were set to 100 $\mathrm{cm}^{-3}$ . INPs were initialized with a concentration of 3.3 $\mathrm{L}^{-1}$, which is at the high end of predicted ice crystal number concentrations ($N_{ice}$) by different parameterizations in Young et al. (2016) (assuming one INP per ice crystal). Due to the interactive INPs in our simulations, we used a relatively high initial INP concentration to prevent an underestimation of $N_{ice}$. For simplicity we assumed a constant aerosol profile with height. As for the background thermodynamic conditions, we kept the background aeorsol concentrations the same in the open ocean and sea ice case.

We performed control simulations over sea ice and open ocean and evaluated these against available observations. For the sea ice case, the COSMO sea ice model (Mironov et al., 2012) was switched on. To exclude influences from variable turbulent fluxes, the sensible and latent heat flux were set to 25 and 23 $\mathrm{W\,m^{-2}}$ over ocean and to 1 and 0.8 $\mathrm{W\,m^{-2}}$ over sea ice. These prescribed fluxes are at the lower end of the observed range (Young et al., 2016). However, larger fluxes were found to increase the strength and size of the convective cells in sensitivity simulations not shown here. Therefore we would need larger domain sizes to simulate cases with larger surface fluxes. This was not possible due to the high computational demand of each simulation. Surface roughness length was assumed to be higher over the ocean with 0.0002 $\mathrm{m}$ in contrast to 0.0001 $\mathrm{m}$ over sea ice. Divergence was prescribed as zero at the surface and was relaxed linearly to $4*10^{-6}\ \mathrm{s}^{-1}$ at the inversion height and kept constant above. To compensate for the subsidence heating, we included negative horizontal advective temperature tendencies, while all other tendencies were set to zero to prevent any influence of boundary layer moistening or drying by large-scale advection.

### 2.1 Setup perturbation experiments

In order to study the effects of aerosol perturbations, an additional mode of potential CCN or INPs was released at every grid point at every height after 1.5 h of simulation time, i.e. following the initial surface precipitation peak. At this time step, the full aerosol perturbation was released. The perturbation mode was assumed to have the same chemical composition but to be at a slightly smaller size than the background mode (0.19 $\mu$m). The smaller size ensures the perturbation mode to activate later than the background mode according to its implementation in the aerosol scheme. Both aerosol perturbations are prognostic, meaning that aerosols are advected throughout the domain, are depleted by cloud droplet or ice crystal formation and precipitation, and are released back into the atmosphere through evaporation or sublimation.

Perturbation aerosol concentrations relevant for CCN activation were increased by 100, 200, 500, and 1000 $\mathrm{cm}^{-3}$. For INP perturbations we once perturbed with the background concentration ($3.3\ \mathrm{L}^{-1}$ for a temperature range of 250.5-258 $\mathrm{K}$) and once increased the initial INP concentration by a factor of 3 ($10\ \mathrm{L}^{-1}$). A summary of all performed simulations can be found in Table 1.

Given the pronounced sensitivity of high latitude cloud processes to atmospheric temperature (e.g. Devasthale and Thomas, 2012), we test the robustness of our results across a $\pm 2$ K temperature change of the background state. In these experiments the entire initial temperature profile was shifted towards colder or warmer temperatures at constant relative humidity.

| Name | CCN perturb ($\mathrm{cm}^{-3}$) | INP perturb ($\mathrm{L}^{-1}$) | T perturb (K) |
|---|---|---|---|
| *ocean_/ice_control* | - | - | - |
| *ocean_/ice_100CCN* | 100 | - | - |
| *ocean_/ice_200CCN* | 200 | - | - |
| *ocean_/ice_500CCN* | 500 | - | - |
| *ocean_/ice_1000CCN* | 1000 | - | - |
| *ocean_/ice_3INP* | - | 3 | - |
| *ocean_/ice_10INP* | - | 10 | - |
| *ocean_/ice_control+2K* | - | - | +2 |
| *ocean_/ice_control-2K* | - | - | -2 |
| *ocean_/ice_1000CCN+2K* | 1000 | - | +2 |
| *ocean_/ice_1000CCN-2K* | 1000 | - | -2 |
| *ocean_/ice_10INP+2K* | - | 10 | +2 |
| *ocean_/ice_10INP-2K* | - | 10 | -2 |

**Table 1.** Summary of all experiments performed. In all simulation the fixed background CCN concentration is 100 $\mathrm{cm}^{-3}$ and the prognostic INP concentration is set to 3.3 INP $\mathrm{L}^{-1}$. All settings listed here were run over open ocean and sea ice surface.

## 3 Evaluation of background state

The local atmospheric conditions over open ocean as observed during the ACCACIA campaign (hereafter named *observations*) are characterized by a single temperature inversion at 1.3 $\mathrm{km}$, capping a single-layer MPC between approximately 0.3 and 1.2 $\mathrm{km}$ (Young et al., 2016). Our simulated case similarly features a strong inversion ($\Delta\theta$ =6 $\mathrm{K}$) at a height of 1.4 $\mathrm{km}$, capping a single cloud layer below (Fig. 1). The boundary layer in both *control* simulations (named *ocean_control* and *ice_control* over open ocean and sea ice, respectively) is stably stratified, as seen in the positive gradient in the ice-liquid potential temperature ($\theta_{il}$) and the negative gradient in the total water content ($q_t$) in Fig. 1. Over the ocean surface an unstable surface layer forms due to the non-zero surface fluxes. The remainder of the boundary layer is stably stratified, which prevents the formation of a well-mixed boundary layer. As a result of stronger surface fluxes, the boundary layer retains more water vapor over the ocean as compared to sea ice (Fig. 1a,b).

Our model successfully simulates a liquid-topped MPC with ice sedimenting out of the liquid layer in both *control* simulations, in agreement with *observations*. The observed cloud properties obtained from Young et al. (2016), our simulated values of the unperturbed simulations, as well as the LES results from Young et al. (2017) are summarized for comparison in Table 2. From Young et al. (2017) we only included the simulation using the ice parameterization that was fitted to the *observations* (termed ACC), which best reproduced the observed case (Young et al., 2017).
The simulated mean $N_{ice}$ of 0.27 $\mathrm{L}^{-1}$ in *ocean_control* (Table 2) is slightly lower compared to *observations*, but within the observed range. $N_{drop}$ agrees well in our model simulations as compared to *observations*, but the maximum $N_{drop}$ in Figure

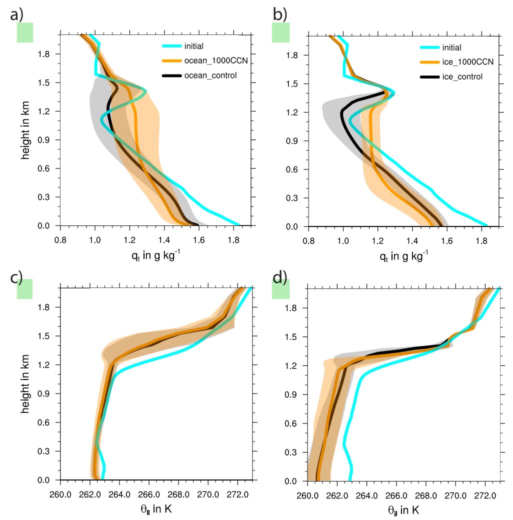


**Figure 1.** Time and domain averaged (±1 standard deviation) a) total water content $q_t$ ($q_t = q_c + q_v + q_i$) in the *ocean_control* simulations as well as most perturbed *ocean_1000CCN* simulation, b) total water content $q_t$ in the *ice_control* and *ice_1000CCN* simulation, c) ice-liquid potential temperature ($\theta_{il}$) in the *ocean_control* and *ocean_1000CCN* simulation and d) $\theta_{il}$ in the *ice_control* and *ice_1000CCN* simulation (for an overview of the simulations refer to Table 1). The blue lines represent the modeled initial values (i.e. timestep zero).

2a is simulated at a higher altitude (1.4 km instead of 1.0 km) due to the upward shift of the simulated stratiform cloud deck. The cloud droplet radius ($R_{drop}$) is smaller than observed, due to an underestimation of the liquid water mixing ratio (LWMR) in the *ocean_control* simulation by a factor of 2. This underestimation of the liquid phase is a general issue in high-resolution simulations of mixed-phase clouds. In particular, the potential impact of the autoconversion rate on cloud evolution in a similar context has recently been discussed in Stevens et al. (2018).

The *ice_control* simulation can only be compared to *observations* in qualitative terms, as the initialization relies on the open ocean dropsonde profile (see section 2). As in the *observations*, the LWMR is smaller over sea ice than over the ocean. Our simulated $N_{ice}$ is also significantly lower over sea ice than over ocean. In contrast to *observations*, $R_{drop}$ is only 0.7 µm smaller in *ice_control* than in *ocean_control*, instead of 5 µm in the *observations*. Additionally, $N_{drop}$ is smaller instead of larger in *ice_control* (Table 2 and Fig. 2b). We relate these differences in cloud properties between our simulated and the observed MPC to the difference in the observed and simulated initial conditions.

## 4 Surface flux impact on cloud dynamics

The simulated effect of surface fluxes is illustrated in Fig. 3, showing a snapshot of the updraft velocities and LWP over ocean and sea ice after 3 h of simulation time. The different surface conditions lead to two different cloud regimes: over ocean, where surface fluxes are increased, the updrafts are higher, leading to cumulus towers detraining into the stratus deck and to a domain

**Table 2.** Averaged (±1 standard deviation) cloud properties derived from the ACCACIA in-situ *observations* (Young et al., 2016, 2017), the Young et al. (2017) LES simulations, and the *ocean_control* and *ice_control* simulations (as temporal means over 2-20 h). As in the observations, all modeled quantities represent in-cloud values (cloud liquid content $q_c$>0.01 $\mathrm{g\,m^{-3}}$ and cloud ice content $q_i$>0.001 $\mathrm{g\,m^{-3}}$).

| | LWMR ($\mathrm{g\,kg^{-1}}$) | $N_{drop}$ ($\mathrm{cm^{-3}}$) | $N_{ice}$ ($\mathrm{L^{-1}}$) | $R_{drop}$ (µm) | $R_{ice}$ (µm) |
|---|---|---|---|---|---|
| *observations* ocean | 0.24±0.13 | 63±30 | 0.55±0.95 | 10 | - |
| *ocean_control* | 0.11±0.08 | 48±15 | 0.27±0.20 | 6.5±1.7 | 15.5±2.0 |
| Young et al. (2017) LES | 0.06 | 100 | 0.34 | 10 | 30 |
| *observations* sea ice | 0.05±0.04 | 110±36 | 0.47±0.86 | 5 | - |
| *ice_control* | 0.06±0.05 | 40±18 | 0.08±0.05 | 5.8±1.8 | 18.0±2.9 |

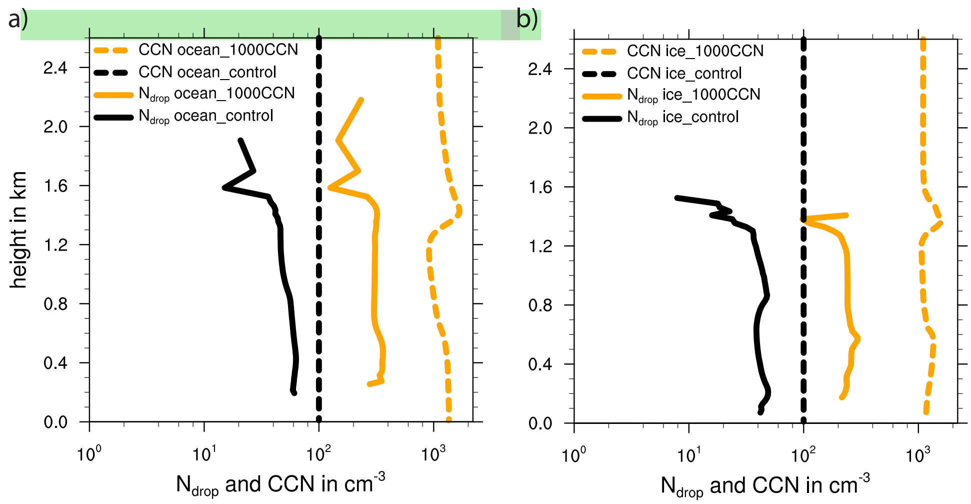


**Figure 2.** Average (2-20 h) $N_{drop}$ (solid lines) and the sum of all CCN tracers, i.e. background and perturbation mode, (dashed lines) in the a) *ocean_control* simulations as well as most perturbed *1000CCN* simulation and b) *ice_control* simulations as well as most perturbed *1000CCN* simulations.

wide shallow stratocumulus cloud structure. Within the shallow cumuli the LWP increases up to 300 $\mathrm{g\,m^{-2}}$, 4 times higher than in the surrounding stratus layer. In contrast, over sea ice the updrafts are low and a spatially homogeneous stratus forms. The LWP of the stratus cloud remains below 80 $\mathrm{g\,m^{-2}}$.

These dynamic differences feed back onto the vertical cloud structure (Fig. 4). Supported by the stronger updrafts over the open ocean, the cloud base and top of the stratiform cloud deck are lifted by 200 m and 100 m, respectively, as compared to the cloud over sea ice (dashed lines in Fig. 4). These high updrafts over the open ocean sustain an increased rate of cloud droplet activation. Despite this increased rate of cloud droplet activation, the mean effective radius remains unchanged between the two cloud regimes due to the increased rate of condensate forming in the updraft. The higher updrafts also facilitate rain

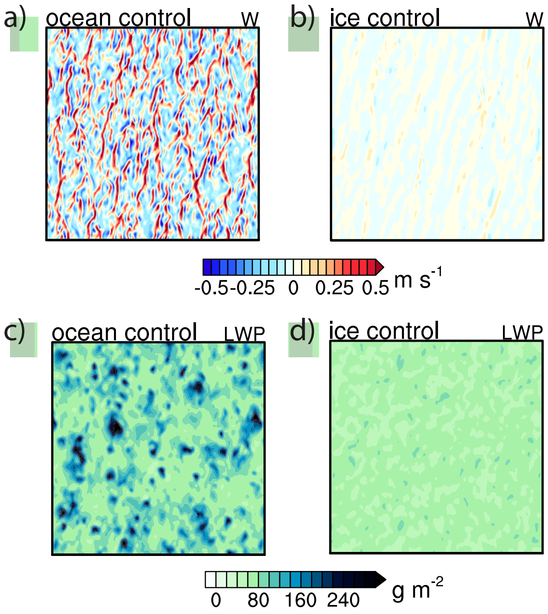


**Figure 3.** Snapshot at 3 h simulation time of a,b) sub-cloud updraft speed at 100 m and c,d) LWP for the *ocean_control* (left) and the *ice_control* case (right).

formation over the open ocean, where droplets can grow at a faster rate than in the surrounding stratus cloud. As a result, total precipitation is increased over the open ocean (on average 1.12 $\mathrm{mm\,d^{-1}}$ as opposed to 0.59 $\mathrm{mm\,d^{-1}}$ above sea ice, Fig. S1a,b). Over sea ice, relatively low updraft speeds prohibit a strong upward moisture flux into the cloud layer due to the large thermodynamic stratification in the sub-cloud layer. This results in a drier boundary layer at cloud height and an optically thinner cloud (Table 3).

In addition to $N_{drop}$, $N_{ice}$ is also increased in *ocean_control* as compared to *ice_control*. As suggested by Garrett and Zhao (2006), the higher liquid water content in the air column increases the cloud longwave (LW) emissivity. Thus, the higher LWP over the open ocean increases LW cloud top cooling, which initiates immersion freezing at cloud top (Fig. 5a,b). Through cooling in the updrafts and more available moisture, ice crystals can grow more efficiently by vapor deposition over the ocean (Fig. 6a,b). Overall, these processes lead to a higher IWP over open ocean than over sea ice. Note that over the ocean sedimenting ice in the form of snow contributes to 20% of total rain and snow at the surface, while over sea ice this is reduced to 2%.

These differences in cloud structure and properties (i.e. changes in cloud base and top, liquid and ice content, and precipitation efficiency) between the two cloud regimes agree with *observations* and previous LES results (Young et al., 2017).

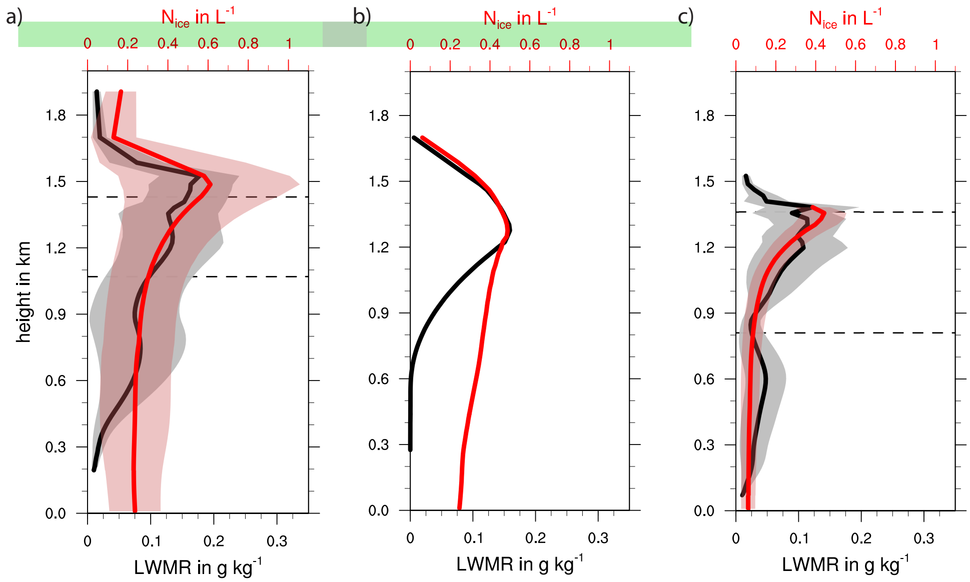


**Figure 4.** Domain and time averaged (2-20 h) ±standard deviation of $N_{ice}$ (red) and cloud liquid water mixing ratio (black) in the a) *ocean_control*, b) Young et al. (2017) LES simulations and c) *ice_control* simulation. Only in-cloud values are plotted. The horizontal dashed lines represent the modeled cloud base and cloud top, where 80% of the domain grid points are cloud-covered ($q_c$>0.01 $\mathrm{g\,m^{-3}}$).

Due to the distinctly different cloud dynamics in both regimes, also the effect of the aerosol perturbations on the clouds differs. In the following we present results from several sensitivity simulations, where we investigated the cloud response to CCN and INP perturbations across different temperature ranges for the two cloud regimes.

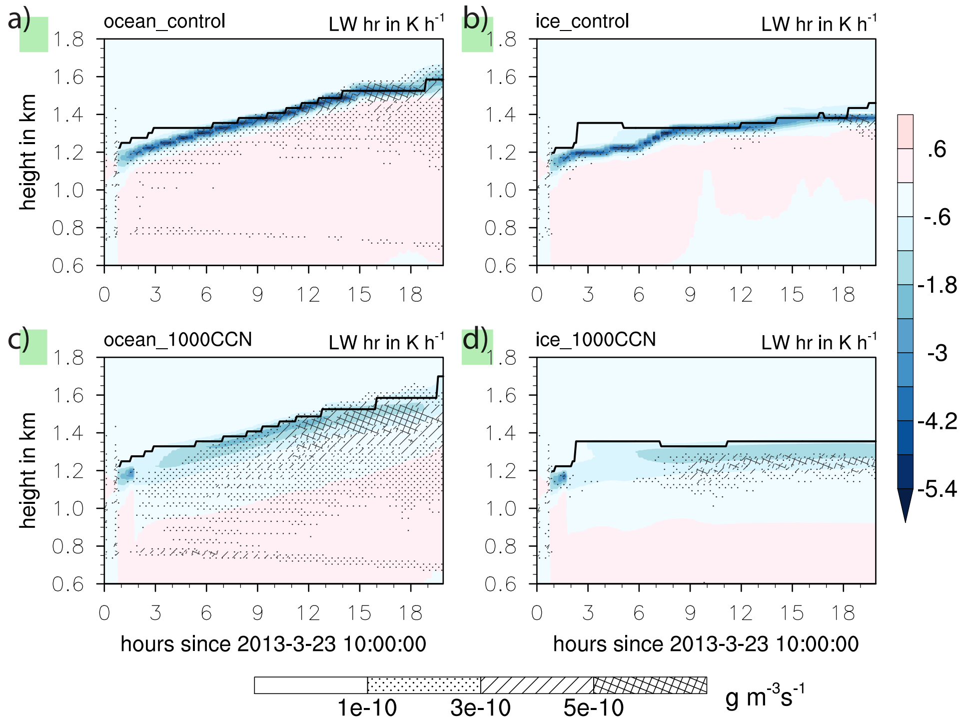


**Figure 5.** Domain averaged LW heating rate (color), immersion freezing rate (hatching) and cloud top of the uppermost cloud layer, where 80% of the domain grid points are cloud-covered ($q_c$>0.01 $\mathrm{g\,m^{-3}}$) are shown for the a) *ocean_control* and b) *ice_control* simulations and c,d) the respective *1000CCN* simulations. Only the range where immersion freezing occurs (T <258 K) is shown.

## 5 Robustness to perturbations in microphysics

### 5.1 Response to CCN perturbations

We performed simulations with potential CCN perturbations ranging from 100 to 1000 CCN $\mathrm{cm^{-3}}$. These number concentrations are higher than what would locally be expected from sea ice loss (Browse et al., 2014), but within the range of CCN concentrations measured in ship exhaust plumes (Hobbs et al., 2000) or Arctic haze conditions in spring (Rogers et al., 2001). The perturbations were applied (as described in section 2) following the strong precipitation event 1.5 h after initialization. Over the ocean, the cloud responds almost immediately to CCN perturbations with an increase in LWP (Fig. 7a). A doubling of the initial CCN concentration (100 CCN $\mathrm{cm^{-3}}$) is sufficient to increase mean LWP by 13 $\mathrm{g\,m^{-2}}$ within the first hour after seeding. This equals a 13% change in LWP between *ocean_control* and *ocean_100CCN* and is comparable to the observed LWP change in ship tracks by Christensen et al. (2014).

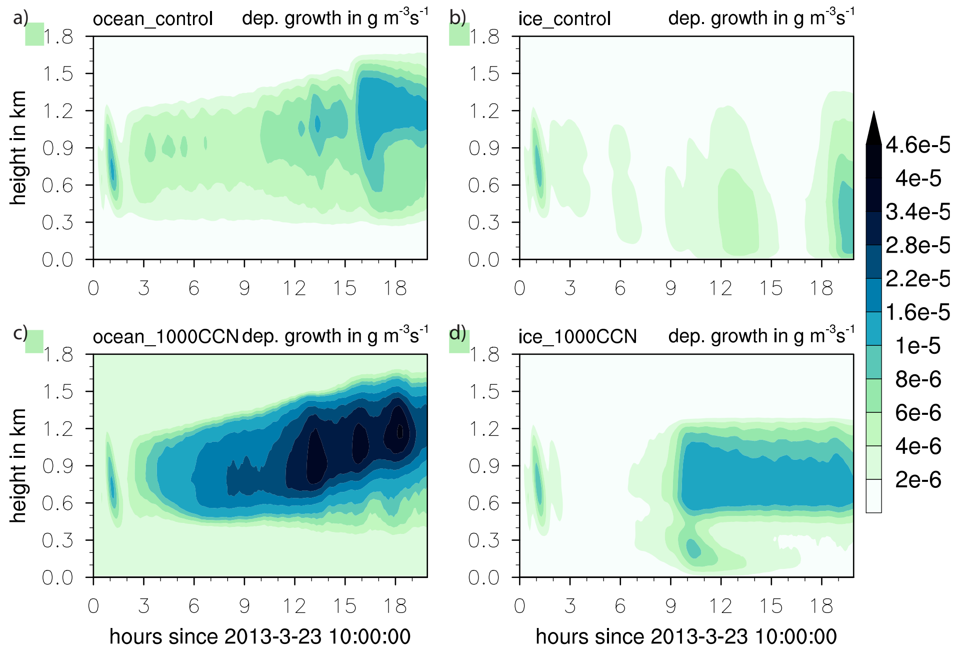


**Figure 6.** Domain averaged depositional growth rate for the a) *ocean_control* and b) *ice_control* simulations and c,d) the respective *1000CCN* simulations. Only the range where immersion freezing occurs (T <258 K) is shown. Note the non-linear colorbar.

Elevated CCN concentrations in combination with strong updrafts allow fast additional droplet formation, which immediately increases the in-cloud vertical mean $N_{drop}$ from 49 to 201 $\mathrm{cm}^{-3}$ directly after seeding and decreases $R_{drop}$ from 6 to 4 µm in the *ocean_1000CCN* simulation (Fig. S2). This decrease in radius is expected according to the Twomey effect (Twomey, 1974) and argues for an immediate 20% increase in liquid water content. Consequently, with increasing CCN perturbation, LWP successively increases, however, a further increase in perturbation strength from 500 $\mathrm{cm}^{-3}$ to 1000 $\mathrm{cm}^{-3}$ does not induce an additional increase in LWP. As the total water content is similar for *ocean_500CCN* and *ocean_1000CCN* (Fig. S3), the boundary layer seems to be saturated for a CCN perturbation of 500 $\mathrm{cm}^{-3}$. All available precipitation has been suppressed and further growth of the mixed-layer is inhibited for CCN perturbations >500 $\mathrm{cm}^{-3}$. Additionally, in these two most perturbed simulations, the cloud top rises and the cloud deepens through overshooting cumulus towers that detrain moisture into the free troposphere and pre-condition the layers above cloud top for further cloud growth (Fig. 5a,c and S4). The cloud top rise in simulations perturbed by CCN could be a result of latent heat release during cloud droplet formation which feeds back onto the updraft velocities. For CCN perturbations below 200 $\mathrm{cm}^{-3}$, this additional latent heating might not be enough to sustain further cloud growth and the cloud top does not rise in *ocean_100CCN* and *ocean_200CCN* as compared to *ocean_control*.

Apart from changes in LWP, also $N_{ice}$ and IWP are affected by CCN perturbations (Fig. 7c and Fig. S5a). Firstly, the cloud deepening in *ocean_500CCN* and *ocean_1000CCN* (Fig. S4) results in an increase in $N_{ice}$ in the respective simulations, as at higher altitudes new INPs can be entrained and become available for immersion freezing. Immersion freezing is also more efficient throughout the cloud, as the higher LWP radiatively cools the cloud layer over a larger area as compared to *ocean_control* (Fig. 5c), which additionally increases $N_{ice}$ in the perturbed simulations. Secondly, growth by vapor deposition is significantly increased in the perturbed simulations (Fig. 6c). The radiative cooling in addition to slightly colder temperatures at cloud top through the cloud deepening create favorable conditions for ice crystal growth through the Wegener-Bergeron-Findeisen (WBF) process (Wegener, 1911; Bergeron, 1935; Findeisen, 1938). This cooling of the cloud-driven mixed layer together with higher $N_{ice}$ favor more efficient depositional growth in all CCN sensitivity simulations. Besides, the sub-cloud and cloudy layer become increasingly well-mixed and moistened with respect to *ocean_control* in all sensitivity simulations (Fig. 1 and Fig. S3), such that the boundary layer remains supersaturated with respect to water and the liquid as well as the ice phase can grow simultaneously. In *ocean_500CCN* and *ocean_1000CCN* the stronger cloud top rise and cloud layer cooling sustain an immediate increase in the depositional growth rate, which increases IWP in these simulations as compared to *ocean_100CCN* and *ocean_200CCN* throughout the simulated time period. The importance of depositional growth in simulations perturbed by CCN agrees with recent results from Solomon et al. (2018).

The response to CCN perturbations strongly depends on the cloud regime. Due to the lower updrafts and the decreased vertical moisture transport over sea ice, the increase in $N_{drop}$ after the CCN injection is lower than over the ocean (Fig. S2). Limited by moisture transport into the cloud layer over sea ice, the increase in LWP is weaker than over the open ocean (Fig. 7b). Additionally, the spatial variability of LWP is reduced over sea ice due to the more stratiform cloud deck. Hence, a perturbation of 100 CCN $\mathrm{cm}^{-3}$ significantly increases the LWP up to 7 h after seeding.

Over sea ice, IWP and $N_{ice}$ reach a maximum shortly after the maximum increase in LWP (Fig. 7d and S5b). As over the ocean, LW cooling over a larger vertical range in the CCN perturbation simulations triggers immersion freezing in the upper 300 $\mathrm{m}$ of the cloud (Fig. 5d). Similar to the open ocean case this radiative cooling and higher $N_{ice}$ in the perturbed simulations favor increased depositional growth. However, the depositional growth rate in *ice_1000CCN* is only one third of the growth rate simulated in *ocean_1000CCN* (Fig. 6d).

As evident from Fig. 7a, over the open ocean the elevated LWP decreases after reaching its maximum and returns to the LWP range of *ocean_control*. Independent of the strength of the CCN perturbation, LWP in all simulations relaxes back to the unperturbed state over the open ocean. On the contrary, over sea ice any CCN perturbation >200 $\mathrm{cm}^{-3}$ significantly perturbs LWP and IWP beyond 20 h simulation time. We relate this different aerosol response of the stratocumulus cloud over the ocean and stratus cloud over sea ice mainly to differences in cloud dynamics. Over the open ocean, the cloud response to CCN perturbations is shifted from the liquid to the ice phase, where the strong and rapid increase in ice mass reduces the liquid-phase response. Due to the increase in cloud ice and snow, increased surface precipitation after 12 h simulation time in the perturbed simulations additionally adds to the attenuated CCN response over the open ocean (Fig S5).

Fig. 8 visualizes the spatio-temporal evolution of LWP within the domain over the open ocean. In the first hours after the initiation of the perturbation the LWP throughout the domain and within the updraft towers is increased (Fig. 8b). However,

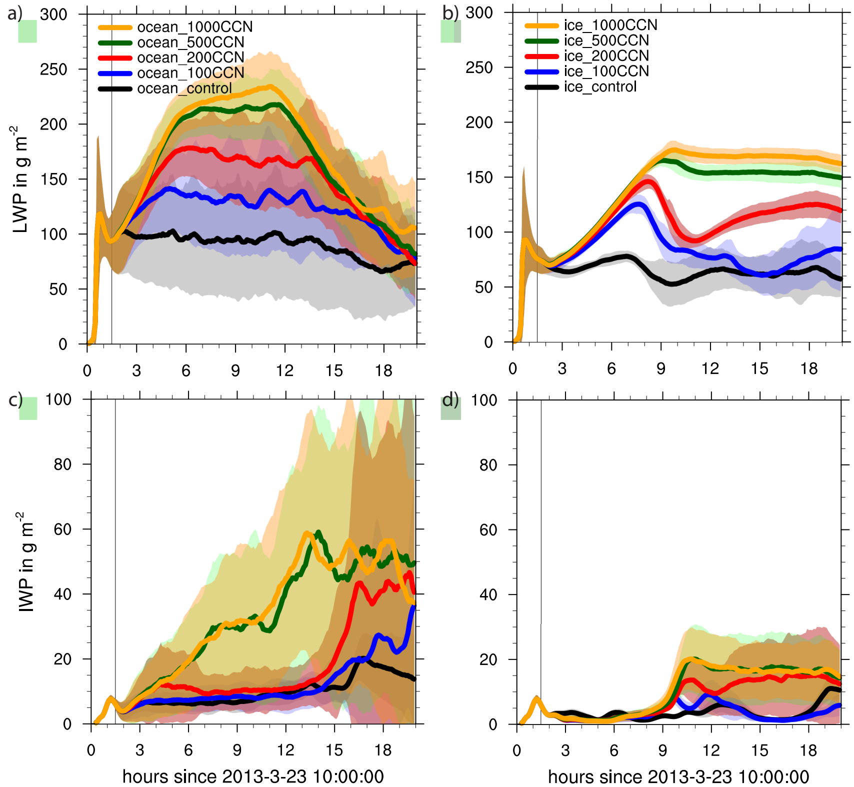


**Figure 7.** Domain averaged a,b) LWP and c,d) IWP over the open ocean (left) and sea ice (right) in the *control* and all CCN sensitivity simulations. The solid lines depict the means, the shadings the standard deviations. The vertical black lines indicate the CCN perturbation injections.

towards the end of the simulation, the cloud organizes back to structures similar to those observed in the control simulation (Fig. 8a). This behavior is qualitatively similar to what has previously been observed in numerical aerosol-perturbed simulations of warm-phase shallow cumuli (Jiang et al., 2006; Seifert et al., 2015). There, evaporative processes caused the limited sensitivity of the cloud field to aerosol perturbations. In our study, the main mechanism controlling the liquid-phase response of the stratocumulus cloud is the increased ice and precipitation formation.

### 5.2 Response to INP perturbations

Similar to the CCN perturbation simulations, we applied two INP perturbations of 3 and 10 INP $L^{-1}$ after 1.5 h simulation time. INP concentrations of over 10 $L^{-1}$ are not uncommon in Arctic spring conditions, representing Arctic haze (Rogers et al., 2001). In both dynamic regimes, IWP increases and LWP decreases with more available INPs (Fig. 9). As a result, the amount of precipitating ice and snow is increased in the perturbed simulations, while the amount of rain is decreased (not shown), similarly to the simulations perturbed by CCN. Total surface precipitation is increased within 2 h following the INP injections

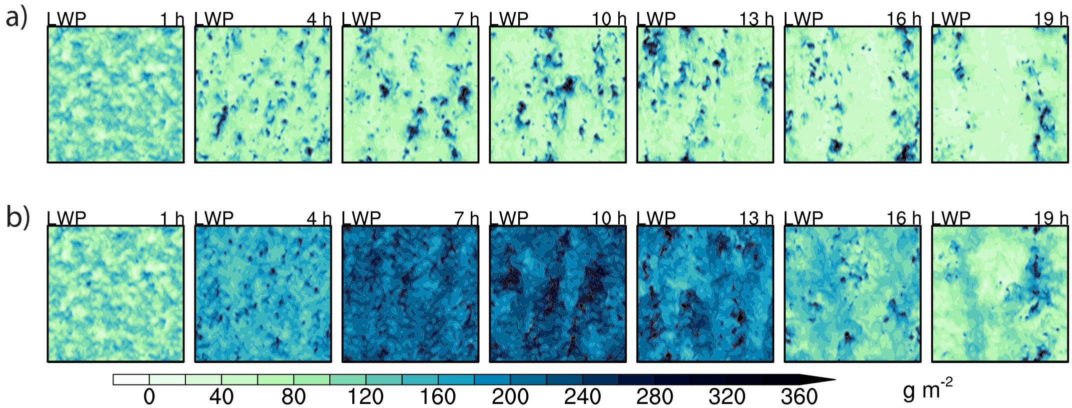


**Figure 8.** LWP for a) *ocean_control* and the b) open ocean *1000CCN* simulation.

to 1.93 $\mathrm{mm\,d^{-1}}$ over the open ocean and 2.20 $\mathrm{mm\,d^{-1}}$ over sea ice in the *10INP* simulations, but thereafter not significantly impacted (Fig S1c,d).

The relative impact of INP perturbations is considerably larger than compared to CCN perturbations. A perturbation of 3 INP $\mathrm{L^{-1}}$(i.e. double the background concentration) doubles the peak IWP over the ocean from 5 to 10 $\mathrm{g\,m^{-2}}$, and decreases LWP by 12% from 100 to 88 $\mathrm{g\,m^{-2}}$ (Fig. 9a,c) one hour after INP injection. An equivalent change of CCN in *ocean_100CCN* increases LWP by merely 13% and does not (yet) increase IWP (section 5.1). Over sea ice, IWP increases initially by almost 400% from 3 to 12 $\mathrm{g\,m^{-2}}$ and LWP decreases also by 12% from 66 to 58 $\mathrm{g\,m^{-2}}$ for a perturbation of 3 INP $\mathrm{L^{-1}}$ as compared to *ice_control* (Fig. 9d).

Considering the full simulation period, the mean IWP increase through INP perturbations remains below the response of the ice phase to CCN perturbations of 500 $\mathrm{cm^{-3}}$ or higher (Fig. 7c,d and Table 3). Investigating this increase in the ice phase in clouds perturbed INPs, we conclude that in the *3INP* and *10INP* simulations ice crystal growth on the expense of liquid water through the WBF process (as seen in the increase in IWP accompanied by a LWP decrease) as well as changes in $N_{ice}$ (Table 3) through immersion freezing on INPs dominate the total IWP increase. The higher $N_{ice}$ follows the Twomey effect in the sense that $R_{ice}$ is smaller (Table 3), but IWP is still increased (Kärcher and Lohmann, 2003). This is insufficient to exceed the IWP increase in clouds perturbed by CCN, where growth by deposition in the colder and destabilized cloud layer dominates any changes in $N_{ice}$.

Also, even though the relative impact of INP perturbations is large, in neither regime does a perturbation of 10 INP $\mathrm{L^{-1}}$ glaciate the cloud. This finding is consistent with other studies investigating cloud glaciation under INP perturbations (e.g. Morrison et al., 2008; Solomon et al., 2018), but in contrast to Young et al. (2017), who simulate cloud glaciation using different (but more simplified) ice nucleation parameterizations for the same case. Considering $N_{drop}$»$N_{ice}$ throughout the simulation, a complete glaciation of the cloud seems surprising with an INP perturbation of only 10 $L^{-1}$.

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. However, the more dynamic cloud structures over the open ocean are able to maintain an elevated IWP by 300% throughout the simulation.

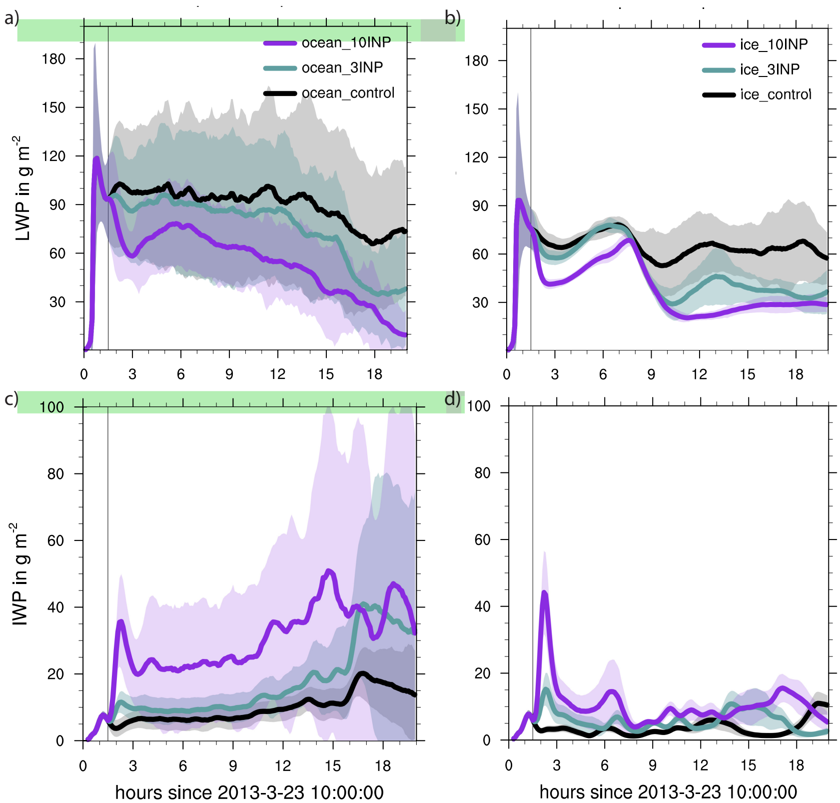


**Figure 9.** Domain averaged a,b) LWP and c,d) IWP over the open ocean (left) and sea ice (right) in *control* and all INP sensitivity simulations. The solid lines depict the means, the shadings the standard deviations. The vertical black lines indicate the CCN perturbation injections.

### 5.3 Sensitivity to different temperature regimes

To address the robustness of our conclusions to different temperature ranges, we performed the *control*, the *1000CCN*, and the *10INP* simulations over sea ice and open ocean in 2 K warmer and colder conditions. The relative humidity was kept constant.

The environmental conditions mainly determine the partitioning of moisture between the liquid and the ice phase (Fig. 10). Focusing on the open ocean case first, the response to CCN perturbations is intensified in the cloud liquid phase under warmer conditions, as LWP increases compared to *ocean_1000CCN* and IWP decreases. This is of course related to the fact that at warmer temperatures less INPs nucleate, which decreases $N_{ice}$ (Fig. 10c and Table S1). In contrast, at colder temperatures more INPs nucleate, IWP increases earlier on as in *ocean_1000CCN* and LWP is significantly reduced (Fig. 10a,c and Table S1). However, even under warmer conditions LWP in *ocean_1000CCN+2K* relaxes to its unperturbed state and returns to the range of *ocean_control* at the end of our simulated time period (Fig. 10a). Hence, our conclusion concerning the buffered aerosol response in the liquid phase over the open ocean remains valid for warmer environmental conditions.

Over sea ice the aerosol response of LWP is also sensitive to the environmental conditions. Under warmer conditions, the cloud shows a similar behavior to the open ocean case. LWP in the *ice_1000CCN+2K* shows a similar increase to the *ocean_1000CCN* case and relaxes to the unperturbed conditions after 18 h. The temporal evolution of the LWP (Fig. S6) indicates small convective cells between 4-16 h in the *ice_1000CCN+2K* simulation in contrast to *ice_1000CCN*. As ice processes play a minor role in the *ice_1000CCN+2K* simulation, a strong precipitation event around 13-14 h likely causes the LWP to relax back to the unperturbed state (Fig. S1f).

For INP perturbations, the temperature change initiates increased freezing and a higher IWP for the colder simulations and vice versa for the warmer simulations (Fig. S7). Determined by the nature of the DeMott et al. (2015) immersion freezing parameterization that is based on observations, more (less) INP nucleate at colder (warmer) temperatures.

## 6 Discussion

To summarize the cloud micro- and macrophysical responses to both, INP and CCN perturbations, we calculated the mean cloud properties in Table 3. Additionally, a schematic of our findings is shown in Fig. 11. The first panels in each row conclude our results from section 4, indicating the existence of two different cloud regimes, a stratocumulus regime over open ocean and a homogeneous stratus regime over sea ice. These distinct regimes mainly result from differences in updraft speed, leading to different efficiencies in vertical moisture transport, subsequent cloud droplet growth, precipitation and ice formation. Our results agree with previous findings obtained from satellites and measurement campaigns as well as the ACCACIA observations and modeling results. As has been observed by Young et al. (2016) and simulated by Young et al. (2017), we also simulate a MPC over the ocean with a higher cloud top, larger droplets, increased LWP and IWP and increased precipitation rates. The development of cumuli over the ocean as a response to increased surface fluxes additionally supports findings by Young et al. (2018).

As in Schweiger et al. (2008) and in agreement with previous ACCACIA studies our results indicate a higher cloud base over the open ocean and geometrically thicker clouds than over sea ice (supporting findings by Palm et al., 2010). Similarly to

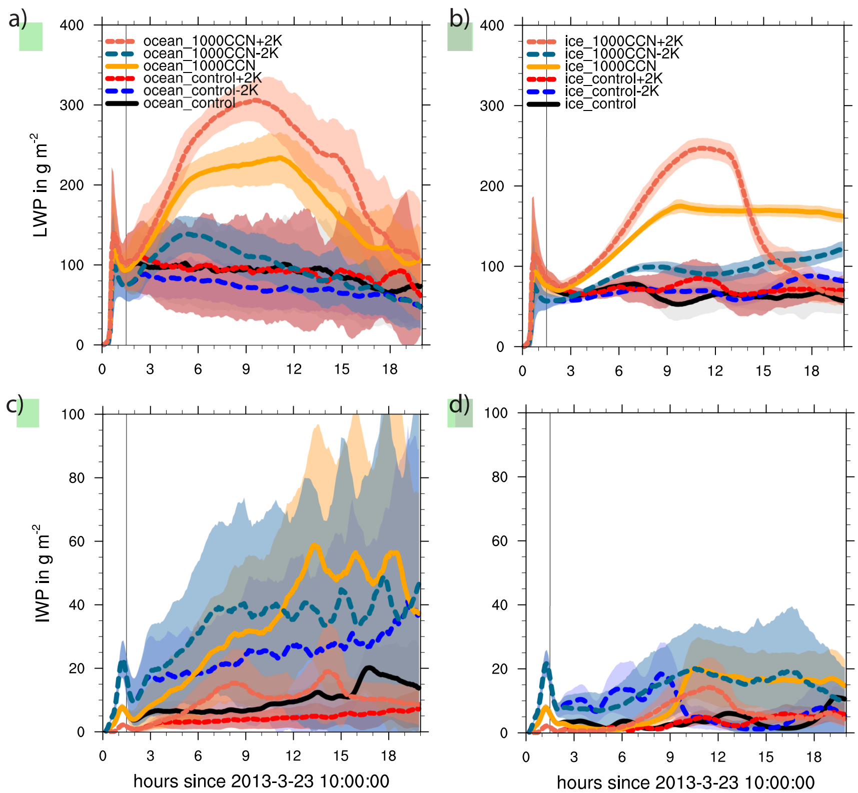


**Figure 10.** Domain averaged a,b) LWP and c,d) IWP over the open ocean (left) and sea ice (right) in *control* and the respective *1000CCN* simulations in their regular state and 2 K warmer and colder conditions. The solid lines depict the means, the shadings the standard deviations. The vertical black lines indicate the CCN perturbation injections.

Sotiropoulou et al. (2016) we also note structural differences over both surfaces with a stratocumulus cloud regime over the ocean versus a stratus cloud over sea ice. However, while Sotiropoulou et al. (2016) relate changes in cloud properties mainly to changes in atmospheric stability over the open ocean and sea ice, our case studies are initialized with the same atmospheric stability profile, hence we suggest that the differences in surface fluxes may play a stronger role than previously suggested. In terms of radiative effects, we find the cloud base height to be the dominating factor determining the net surface LW radiative balance for clouds sufficiently optically thick in the LW spectrum (note that net surface LW radiation is defined to be positive downwards, i.e. absorption by the surface throughout our study). As the cloud over sea ice has a lower cloud base, the cloud re-emits LW radiation at warmer temperatures, which reduces the net surface LW cooling (Table 3).

In a next step, we applied aerosol perturbations to the two contrasting cloud regimes. 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, which is a novel aspect as compared to previous ACCACIA modeling studies (Young et al., 2017, 2018).

Both studies Young et al. (2017) and Young et al. (2018) used a prescribed $N_{drop}$ concentration and parameterized $N_{ice}$ concentrations (not considering interactive INPs) in their model setup, which have been adjusted in sensitivity simulations by Young et al. (2018). In their study, the authors found smaller droplets in a simulation with increased $N_{drop}$, but found little effect on LWP or IWP. In contrast, we see a strong initial sensitivity of Arctic MPCs to CCN perturbations. Over ocean and sea ice, the LWP is already significantly increased with a perturbation of 200 and 100 CCN $\mathrm{cm}^{-3}$, respectively. With increasing CCN perturbations, $N_{drop}$ ($R_{drop}$) increases (decreases), accompanied by an increase in LWP (in agreement with Morrison et al., 2008; Possner et al., 2017; Solomon et al., 2018; Stevens et al., 2018). As a result of the larger LWP, LW cooling increases in the perturbed simulations throughout the cloud and the cloud deepens, such that more ice crystals nucleate through increased immersion freezing. Additionally, ice crystals grow by enhanced deposition rates in the perturbed simulations. This increased IWP in simulations solely perturbed by CCN was noted before by Possner et al. (2017) as well as Solomon et al. (2018). As a result of higher IWP and LWP, the cloud becomes optically thicker and reduces the LW emission from Earth surface (Table 3), which warms the surface, as also modeled in Stevens et al. (2018). Changes in the radiative properties are overall only moderate between the *control* and *1000CCN* simulations, ranging from 6-13% over sea ice and ocean, respectively. Most likely the change in cloud structure between the two regimes determines the smaller response in net surface LW radiation to CCN perturbations over sea ice than over the open ocean. The temporary transition from a stratocumulus to a stratus cloud over the ocean for a perturbation of 1000 CCN $\mathrm{cm}^{-3}$ (Fig. 8) increases the cloud re-emittance throughout the domain. On the contrary, the additional thickness of the stratus cloud over sea ice has a smaller effect, as the cloud structure is not significantly changed. Interestingly, the change in cloud base as simulated between *ocean_control* and *ice_control* has a stronger radiative effect on the Earth's surface (4.3 $\mathrm{W\,m^{-2}}$) than CCN perturbations of 1000 $\mathrm{cm}^{-3}$ (3.4 $\mathrm{W\,m^{-2}}$ over the ocean and 1.3 $\mathrm{W\,m^{-2}}$ over sea ice).

There is a strong regime-dependence of the MPC response to CCN perturbations, which is novel in the context of aerosol-cloud interactions. Over sea ice, the cloud evolution remains significantly changed throughout the simulation period for any CCN perturbation >200 CCN $\mathrm{cm}^{-3}$. Over the open ocean, ice formation and growth as well as an increase in precipitation buffer the LWP response and lead to a relaxation of the liquid phase to its unperturbed state after 18 h simulation time. Additional observations such as the ACCACIA campaign, but in polluted environments, could help to constrain such regime-dependent aerosol-cloud interactions. Also further model studies including prognostic aerosols could expand our findings to a wider range of meteorological conditions (which we touched upon with our temperature change sensitivity tests).

The initial relative impact of increasing INP concentrations is larger as compared to CCN concentrations. With more potential INPs, more particles are available for ice crystal formation by immersion freezing, which increases $N_{ice}$ and IWP. The increase in IWP is accompanied by a decrease in LWP through the removal of liquid water by deposition via the WBF process. This is consistent with previous studies investigating the effect of increasing INP or $N_{ice}$ on Arctic MPCs (Morrison et al., 2008; Ovchinnikov et al., 2014; Stevens et al., 2018; Young et al., 2018). The lower LWP in the simulations perturbed by INPs leads to an optically thinner cloud in the *10INP* simulations which increases net LW cooling at the Earth's surface (Table 3). This is an opposing effect to CCN perturbations, which generally have a moderate warming effect on the underlying surface. Interestingly, the IWP increase for a perturbation of 10 INP $\mathrm{L}^{-1}$ is smaller than the IWP increase in the *1000CCN* simulation

**Table 3.** Averaged cloud properties $\pm 1$ standard deviation throughout the simulated time period following the aerosol injection (hour 2-20) for the unperturbed and perturbed simulations.

| | *ocean_control* | *ocean_1000CCN* | *ocean_10INP* | *ice_control* | *ice_1000CCN* | *ice_10INP* |
|---|---|---|---|---|---|---|
| Cloud mean $N_{drop}$($\mathrm{cm}^{-3}$) | 48.1±15.4 | 306.6±68.5 | 52.0± 15.4 | 40.1±18.4 | 230.5±53.6 | 39.1±15.4 |
| Cloud mean $R_{drop}$(µm) | 6.5±1.7 | 4.1±1.0 | 6.1±1.6 | 5.8±1.8 | 4.4±1.0 | 6.4±2.0 |
| LWP ($\mathrm{g\,m^{-2}}$) | 89.8±50.9 | 176.5±57.3 | 52.8±31.4 | 64.9± 17.7 | 147.6± 34.2 | 37.0±14.8 |
| Cloud mean $N_{ice}$($\mathrm{L}^{-1}$) | 0.27±0.20 | 0.44±0.34 | 0.84±0.75 | 0.08±0.05 | 0.17±0.10 | 0.27±0.19 |
| Cloud mean $R_{ice}$ (µm) | 15.5±2.0 | 17.5±3.2 | 14.4±2.6 | 18.0±2.9 | 18.4±3.4 | 15.9±2.6 |
| IWP ($\mathrm{g\,m^{-2}}$) | 10.1± 7.1 | 35.2± 34.8 | 31.6±34.7 | 3.6± 2.8 | 11.0±9.9 | 10.8±8.0 |
| Cloud optical depth | 9.5±4.9 | 28.9±8.8 | 5.8±3.3 | 7.6±2.2 | 23.0±5.1 | 4.0±1.7 |
| Net surface LW ($\mathrm{W\,m^{-2}}$) | -25.1±3.9 | -21.7±4.0 | -28.6±11.5 | -20.8±4.5 | -19.5±4.4 | -26.4±6.5 |

over the open ocean (Table 3). We relate this difference to more efficient ice crystal growth by deposition in *1000CCN* than in *10INP*, supported by higher deposition rates (not shown) in experiments perturbed by CCN (Table 3). These results also generally agree with findings from Possner et al. (2017). The authors found that in an INP-limited regime, perturbations in INP had a significant impact on the cloud state and CCN perturbations had a stronger impact on LWP. However, once a sufficiently high INP concentration was reached, further perturbations in INP had no additional impact on the cloud state, and the macrophysical response of the cloud to CCN perturbations was heavily buffered by ice-phase processes. Similarly, we do see an effect of INP perturbations on LWP, IWP, $N_{ice}$ and $R_{ice}$, but the longer-term cloud response is more affected by CCN perturbations.

The stratus cloud over sea ice initially shows a stronger response to INP perturbations than the stratocumulus cloud over open ocean. This different sensitivity to INP changes between surfaces is consistent with findings from Morrison et al. (2008). Similarly, Jiang et al. (2000) found Arctic stratus over sea ice to be specifically vulnerable to INP perturbations. Note that with time, the IWP increase (LWP decrease) is more pronounced over the ocean, which can be related to stronger updrafts and cooling as well as the continuous cloud deepening over open ocean.

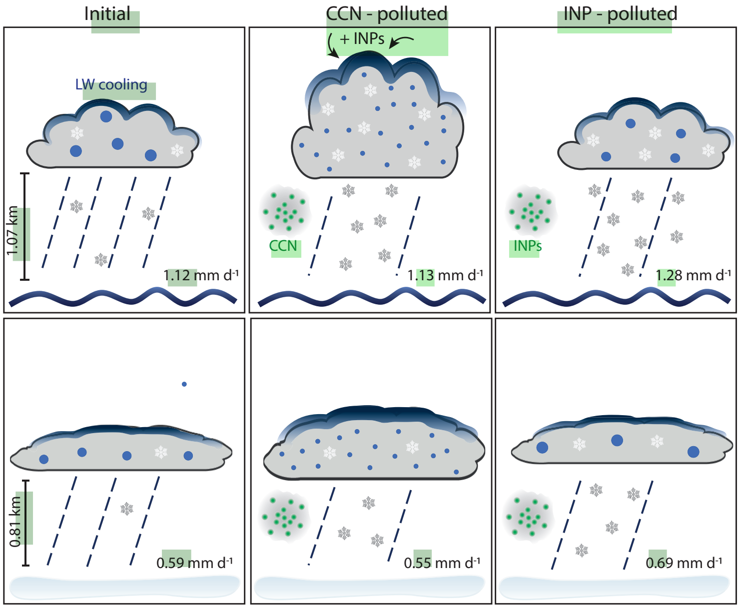


**Figure 11.** Conceptual overview of the cloud response to increased CCN and INP concentrations. The first row illustrates the open ocean stratocumulus regime, the lower row the stratus over sea ice.

## 7 Conclusions

The analysis of MPCs within a changing Arctic environment has been the subject of a number of recent studies (Browse et al., 2014; Christensen et al., 2014; Young et al., 2016; Possner et al., 2017; Gilgen et al., 2018). Here, we addressed the cloud properties of MPCs in two differing regimes (i.e. sea ice and open ocean) in a series of high resolution LES. The robustness of the response to an aerosol perturbation was evaluated by applying our perturbation scenarios in warmer/colder environmental conditions.

Our key findings are summarized as follows:

1. The surface properties have a significant impact on MPC properties. Our simulations support previous results obtained for the ACCACIA campaign (Young et al., 2016, 2017, 2018): over the open ocean, strong turbulent surface fluxes increase the updraft velocities, which in turn favor the development of cumuli towers feeding moisture into the stratus layer. This increased vertical 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. Over sea ice, surface fluxes and in turn updraft velocities are low, which confines the cloud to a homogeneous stratus cloud. As the boundary layer is generally less moist, cloud droplet and ice crystal formation and growth are limited as compared to the cloud over open ocean.

2. Aerosol perturbations providing potential CCN significantly impact the cloud LWP and IWP immediately after the perturbation injection. The MPC over the ocean responds with an increase in $N_{drop}$ and LWP. Through increased LW cooling throughout the cloud, new ice crystal formation by immersion freezing and subsequent growth by vapor deposition, IWP increases. Over sea ice, CCN activation is less efficient and the maximum response is delayed and weakened.

3. The relative initial response of the cloud to INP perturbations is larger than to CCN perturbations. The response is relatively straightforward and agrees with previous results. INP perturbations immediately increase the IWP and decrease the LWP in both cloud regimes. In our simulations, none of the applied INP perturbations (3 and 10 $\mathrm{L}^{-1}$) is sufficient to cause complete cloud glaciation.

4. The cloud response to aerosol perturbations is highly regime-dependent. Over the open ocean, LWP perturbations are efficiently buffered after 18 h simulation time. Increased ice and precipitation formation relax the liquid cloud properties back to their unperturbed range. Over sea ice the cloud evolution remains significantly perturbed with CCN perturbations ranging from 200 to 1000 CCN $\mathrm{cm}^{-3}$. For INP perturbations, an intense ice formation and precipitation peak is triggered with no further subsequent change in cloud properties. Over the open ocean, LWP and IWP remain perturbed throughout the simulation for an INP perturbation of 10 $\mathrm{L}^{-1}$.

Extrapolating our findings to a future ice-free Arctic, increased ship traffic, and higher levels of pollution at the high latitudes, we find that changed surface conditions are likely to highly affect MPC dynamics, properties and hence the radiative budget of the surface. The effect of pollution will be most effective in stratiform clouds over sea ice, where INP perturbations on the order of 10 $\mathrm{L}^{-1}$ lead to a strong cloud thinning and thus the relatively largest contribution to the radiative balance on the order of a 6 $\mathrm{W\,m^{-2}}$ cooling at the surface. On the contrary, CCN perturbations have a moderate warming effect, which is increased over the open ocean. Considering that ship exhaust plumes may consist of both, CCN and INPs (Hobbs et al., 2000; Thomson et al., 2018), the combined aerosol effect on Arctic MPCs is unlikely to offset Arctic warming, as also found by Christensen et al. (2014) and Possner et al. (2017).

Nevertheless, we note that our study has come caveats. We used the open ocean initial dropsonde profile to initialize both our cases (open ocean and sea ice), which is in contrast to Young et al. (2017). Over vast sea ice covered surfaces the boundary layer profile might highly differ from the boundary layer over open ocean (Young et al., 2016) and thus the clouds may evolve differently. However, we wanted to narrow possible differences over open ocean and sea ice case down to surface fluxes, which become important over freshly melted sea ice or polynyas (Gultepe et al., 2003). In addition, due to runtime limitations it was not possible to simulate these high-resolution simulations for a longer time period. Thus, we unfortunately cannot draw any conclusions concerning cloud persistence beyond 20 h.

*Author contributions.* GKE conducted the simulations, analyzed the results, and was the main author of the paper. AP and UL contributed to the design of the study and the analysis of the results. All authors contributed to the writing of the study.

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