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 (N_{drop}) gets clipped, when cloud water content (qc) falls below a minimum threshold (1 e^{-15} g m^{-3}). Within these clipping routines it is necessary to fill the cloud condensation nuclei (CCN) budget again with the amount of 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, N_{drop} was set to a minimum value (2 cm^{-3}) for qc > 1 e^{-9} g m^{-3}. This routine induced an artificial source of CCN above cloud top. Given the stable stratification in the inversion, the CCN released above cloud top were only re-entrained at very slow timescales into the cloud. Additionally, we had to adjust the weights calculation that determines the redistribution of CCN after evaporative processes. Through a miscalculation in this routine, CCN were lost within the boundary layer, which impacted our total N_{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 <u>times</u> 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).