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.

General comments:

1. The paper does not suitably cite previous work on this case study from the ACCACIA campaign. The work is novel in its prognostic representation of both CCN and INP; however, similar studies have been already conducted which compare cloud microphysics over sea ice and over ocean. Please ensure all previous literature is cited more appropriately. For example, the differences between the boundary layer structure over sea ice and over ocean has been discussed as an observational study by Young et al., 2016a, and large-eddy simulations of this case study have been presented by Young et al., 2017. Observational conclusions should not be repeated here as conclusions of this study unless these earlier works are cited appropriately.
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?
3. Readability and clarity could be improved – for example, it is often not clear whether the model simulation results or measurements are being discussed.
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.
5. The INP perturbation experiments are lacking analysis and discussion. Please add to this section of the study or remove it.
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.

Specific comments:

<u>Abstract:</u>
Page 1:
Line 4: was ACCACIA conducted in the central Arctic? I would’ve taken this to be >80N?

Line 5: define COSMO

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.

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

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

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

Line 20: Can you say more here about how the aerosols are transported out of the boundary layer?

<u>1. Introduction:</u>
Page 2:
Lines 13-15: Please rephrase – sentence meaning unclear

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

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

Line 23: Please define ECMWF.

Line 29: please rephrase

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.

<u>2. Model description and setup</u>
Page 3:
Line 27: Dropsondes were discussed in detail in Young et al., 2016a – please cite here for reference.

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

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?

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

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

2^{nd} 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?

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 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.

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.

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)?

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?

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

Line 28: Please provide references for this statement.

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

3. Evaluation of background state
Page 5:
1^{st} 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.

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?

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.

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

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

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?

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)?

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.

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

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?
Lines 10-11: Why? Opportunity to discuss.
Line 14: "*most perturbed simulation*" – please quote run label for clarity

Page 11:
Line 4: "*increase in the ice phase*" – please be more specific, do you mean number concentration? Mass concentration?
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.

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)?

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.

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

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?

5.3 Invariance of results across temperature regimes

Page 14:
Lines 6-10: Please improve clarity
Line 12: Again, is this the parametrization used? This is not the same as in Solomon et al., 2015.

5.4 Consistent response independent of perturbation injection period

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.
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.
Line 13: "*possible pathway for cloud-aerosol interactions*" – what is meant by this statement? Not clear/vague.
Line 15: Define τ
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.
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.

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.
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?

7. Conclusions:

Page 19:
Line 2: References for "*… the subject of a number of recent studies*"
Lines 8-10: Opportunity to link with Young et al., 2018 ACCACIA study (cumuli tower development – inter-model agreement)
Lines 8-14: Make stronger links to previous ACCACIA studies and make novelty of results more distinct from previously published conclusions.
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.
Lines 25-26: How? Please provide details.

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.

Technical corrections:

Page 1, line 19: typo → "**properties**"
Page 2, line 2: "high **model** uncertainties"
Page 2, lines 12: "**mid-latitude**"
Page 4, line 23: "to be **at** slightly smaller **sizes** than"
Page 4, line 25: "successively" → "**in stages**"
Page 5, line 8: "according to" → "**agreeing with**"
Page 5, line 15: "In a dry run,…" – new paragraph?
Page 7, line 7: "allow the **cloud** droplets as well as the **ice** crystals"
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.
Page 7, lines 15-16: "The more numerous ice crystals" → "**Higher concentration of** ice crystals"
Page 8, Figure 4 caption: typo → "**interquartile**"
Page 9, line 11: "upon seeding" → "**after seeing**"
Page 10, line 8: "The initial… on the cloud regime" – remove, vague and not required.
Page 10, line 9: "cloud droplet growth **is** limited"
Page 19, line 19: should this be a new paragraph?
References: Some references are incomplete or incorrect.

Figures and Tables:

Table 1: Please list columns as "Background CCN/INP".
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
- 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
- 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?

Please increase legend size on all figures.

Figure 1:

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

Figure 2:

- How do these profiles compare with observations?
- Would it be clearer to have: ice control (black), ice perturb (grey), ocean control (red), ocean perturb (orange)?

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.
- In caption, define LWMR

Figure 5:

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

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

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?

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)?

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).

List of referenced works:

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, P. Natl. Acad. Sci. USA, 74, 2293–2314, https://doi.org/10.1073/pnas.0910818107, 2010.

DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Möhler, O., Snider, J. R., Wang, Z., and Kreidenweis, S. M.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, Atmos. Chem. Phys., 15, 393–409, doi:10.5194/acp-15-393-2015, 2015.

Solomon, A., Feingold, G., and Shupe, M. D.: The role of ice nuclei recycling in the maintenance of cloud ice in Arctic mixed-phase stratocumulus, Atmos. Chem. Phys., 15, 10631–10643, https://doi.org/10.5194/acp-15-10631-2015, 2015.

Solomon, A., de Boer, G., Creamean, J. M., McComiskey, A., Shupe, M. D., Maahn, M., and Cox, C.: The relative impact of cloud condensation nuclei and ice nucleating particle concentrations on phase-partitioning in Arctic Mixed-Phase Stratocumulus Clouds, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-714, in review, 2018.

Young, G., Jones, H. M., Choularton, T. W., Crosier, J., Bower, K. N., Gallagher, M. W., Davies, R. S., Renfrew, I. A., Elvidge, A. D., Darbyshire, E., Marenco, F., Brown, P. R. A., Ricketts, H. M. A., Connolly, P. J., Lloyd, G., Williams, P. I., Allan, J. D., Taylor, J. W., Liu, D., and Flynn, M. J.: Observed microphysical changes in Arctic mixed-phase clouds when transitioning from sea ice to open ocean, Atmos. Chem. Phys., 16, 13945–13967, https://doi.org/10.5194/acp-16-13945-2016, 2016a.

Young, G., Jones, H. M., Darbyshire, E., Baustian, K. J., McQuaid, J. B., Bower, K. N., Connolly, P. J., Gallagher, M. W., and Choularton, T. W.: Size-segregated compositional analysis of aerosol particles collected in the European Arctic during the ACCACIA campaign, Atmos. Chem. Phys., 16, 4063–4079, doi:10.5194/acp-16-4063-2016, 2016b.

Young, G., Connolly, P. J., Jones, H. M., and Choularton, T. W.: Microphysical sensitivity of coupled springtime Arctic stratocumulus to modelled primary ice over the ice pack, marginal ice, and ocean, Atmos. Chem. Phys., 17, 4209–4227, https://doi.org/10.5194/acp-17-4209-2017, 2017.

Young, G., Connolly, P. J., Dearden, C., and Choularton, T. W.: Relating large-scale subsidence to convection development in Arctic mixed-phase marine stratocumulus, Atmos. Chem. Phys., 18, 1475-1494, https://doi.org/10.5194/acp-18-1475-2018, 2018.