Response III to Reviewers

6 December 2022

We would like to thank the editors of the ACP for the assessment of our manuscript and for in principle allowing us to revise the manuscript. On 18 November 2022 we sent a direct letter to the handling editor. While we have not received a response by the day of the deadline for our revised submission (6 December 2022), we are proceeding with this Author's response.

Response to the 3rd review by anonymous reviewer 1

This is my last review of this manuscript: I will decline to review this paper again. I strongly feel – as I said in my previous review, included below – that the process worked poorly for this submission. I was not able to find responses to my comments from the previous cycle. I requested major revision and the authors included only cosmetic adjustments to the previous version (modifying a few figures), basically ignoring almost all of my specific comments. My recommendation is to reject this paper and start the discussion phase again so the reviewers can see how the authors responded to their comments. I appreciate that the Editor may disagree with my recommendation and accept the manuscript. Either way is fine with me. I have to add that I am disappointed with the process and will seriously consider any further review invitations from ACP. I am uploading this comment together with me previous review (from June 2022) below for the Editor convenience.

We are sorry to hear that the reviewer feels that way. However we have to point out that we had fully addressed reviewer's comments on the first 8 pages of the Author's response submitted on 31 July 2022

(acp-2021-888-author_response-version2.pdf).

The changes in the manuscript were substantial, and included modifying figures as well as thoroughly reworking whole paragraphs of our manuscript (particularly in part 5). The extend of the changes could be clearly seen in Author's track-changes file (acp-2021-888-ATC2.pdf, submitted on 31 July 2022), particularly on pages 13, 23, 24, 25, and 27.

Unfortunately, we have not received any new specific comments from the reviewer 1. For clarity we are enclosing our previous response to this reviewer on the following pages.

Previous Response to anonymous reviewer 1

Review of a manuscript "Aerosol impacts on the entrainment efficiency of Arctic mixed phase convection in a simulated air mass over open water", previously titled "Aerosol-cloud-turbulence interactions in well- coupled Arctic boundary layers over open water" by Chylik et al.

Overall recommendation: publish after substantial (major) revisions

This is a revised manuscript that I reviewed last fall. The paper has been significantly revised and has a new title. With all the changes, the paper should be treated as a new submission, requiring a careful reading from scratch. Overall, I feel the discussion of the revised manuscript should start once again (and be published with comments and responses available online), and not be treated as just a check if the revisions addressed the reviewers' concerns. So either the Editor should reject the original submission and request a new discussion phase, or the authors should withdraw the paper and resubmit the new version for the discussion. In a nutshell, I feel something went wrong, especially considering the length of time between the original submission (November 2021) and submission of the revision (May 2022).

We thank the reviewer for the careful assessment of our revised manuscript. We were surprised to read the assessment that "something went wrong", and about the advice to treat the revised paper as a new submission. In our opinion this criticism is unjustified, for various reasons.

Firstly, we followed the revision instructions from the editorial office to the letter, including the invitation to submit a revised version (and not a new submission). We also obeyed the deadline for submission, so the length of time can not be considered overly lengthy. Accordingly, we feel that the comments on the followed procedure should not be part of the response to us authors, but should be aimed at the editorial office instead.

Secondly, while the modifications in the revised manuscript are indeed substantial, this is not unusual for a major revision. In my experience in submitting and reviewing papers I have seen much more drastic changes under the label 'major revision', including changes in the title. And the first (and largest) part of the paper describing the case, the LES experiments and their evaluation against observations, has seen relatively little change. Accordingly, in our opinion (which is apparently shared by the editor) the verdict "reject" would be overly harsh.

As for the science, the new emphasis provides a better focus of the paper. However, I see significant problems with the technical aspects of the manuscript as presented in my major points and specific line- by-line comments. I have to add that the authors ignored some of my specific comments as a few of those below address the same issues as in the previous review. We tried hard to follow up on the original recommendation to improve the technical aspects of the paper. We think this has substantially improved the manuscript, in particular some of the figures. We apologize if we omitted some of these recommendations in our first revision; we hope that in the second revision we have adequately addressed these points.

General (major) comments

1. This is the same as in my previous review: Manuscript title should have "modeling" word in it. Perhaps start with "Modeling of aerosol impacts...".

The title now has the word "simulated" in it. "Simulating" is a subset of "modeling", which can include many other types of numerical integrations (such as global circulation modeling). So we did follow up on this comment. We think "simulated" is more appropriate in our title, because i) it is more precise and accurate and thus delineates better from other atmospheric modeling studies, and ii) we hereby follow many other previous LES studies that have the word "simulation" (and not modeling) in the title.

2. I find the analysis of the assumed initial CCN concentration interesting and worth pursuing. However, I feel the explanation of the simulated dramatic impact is mostly missing. For instance, even without the feedback into dynamics, increasing droplet concentration has to lead to the increase the reflected solar radiation as in the classical Twomey effect. This is not even mentioned in the manuscript. Of course there is a significant dynamical feedback as shown, for instance, by Figs. 11 and 12. The dramatic increase of the LWP feeds back on the Twomey effect, correct? Is that all what happens? I am not sure. For instance, with the low CCN (10 per cc), is there any impact on the precipitation initiation and fallout? In ice-free clouds, low CCN typically implies more drizzle and rain, and thus reduced LWP. How this is modified by the presence of ice? Is the ice initiation in the parameterized microphysics affected by mean droplet size, presumable larger in the low CCN simulation? In a nutshell, I find the new emphasis of the paper interesting, but the analysis of the results leaves much to be desired. This is why I think the paper should go to the open discussion phase again so the authors can openly reply to the reviewers' comments.

We fully agree that increasing droplet concentration leads to the increase in reflected solar radiation, and the dramatic increase of the LWP is likely to feed back on this effect. As suggested, we have added the estimation of the Twomey effect. We have incorporated the this effect directly after the paragraph on 2nd cloud-aerosol indirect effect, where it fit thematically. Our comparison shows that there is a good agreement for the pristine case, however there are significant differences for the control case and the continental case.

Regarding the impact of CCN concentrations the ice processes, we have added further explanation of the effect of the cloud droplet size, as well as figure showing the tendencies in the mass of ice hydrometeors

3. Fig. 1 in the manuscript is as in the previous version. I commented on it in my previous review and the authors tried to respond. My suggestion

is to show a small fraction of the figure, focusing on the area targeted in the LES study. The rest of the figure, difficult to see what it shows, is irrelevant. For instance, where are land boundaries? Another possibility is to add a second panel with the enlarged area targeted by the modeling study to better document cloud systems there.

In the new version we follow both suggestions: the main panel now focuses on the target area, while the smaller secondary panel (a) show the general location of the mission with the land boundaries clearly marked. We agree that this configuration of the figure is more efficient in describing the flights and the simulated domain.

4. This is following point 6 from my previous review. I still claim that the technical quality of the presentation needs to be improved. Some figures require adjustments to make them legible when included in the printed version, some figures can be improved, some show quantities that are not clearly defined, some captions remain unclear. See specific examples below.

Thanks for this critical but constructive comment. We have again looked at all figures, and accommodated all suggested improvements as provided below. We hope that by doing so the figures now have the scientific quality required for publication.

Specific minor comments

1. Caption of Fig. 3 is incorrect (see panels b and c, and their description). Please correct.

The caption has been corrected to agree with the panels.

2. Fig. 4. The caption says "histogram" but the vertical axis says "pdf". So which one is it? Pdf is a histogram with each bin value divided by the bin width. Also, I feel the caption should say that this is the histogram of time during which a given concentration was observed. I feel more details of those observations would be needed. For instance, what was the time resolution of those observations (1sec? 10 sec?).

The caption of the figure was indeed misleading. We have corrected it now to "pdf". We have also added some of the main properties of the observations and for further information we refer the data publications.

3. L 144: "We speculate...": Do model results show that aerosol processing is responsible for local fluctuations? If so (as suggested by Fig. 12c), "speculate" is a poor word.

Replaced with the word "hypothesize"

4. Section 3.2, microphysics. It is not clear if all ice variables (cloud ice, snow, graupel) include prediction of both mass and number, that is, as in a complete double moment scheme. It would be appropriate to mention this explicitly.

Adding explanatory sentence: This is a full 2-moment implementation, the mass concentration and number concentration of each of five hydrometeors are thus prognostic variables.

5. Section 3.3 misses information about the longwave radiation. This was my point 6 from the previous review.

This was an unfortunate oversight on our side. The composite large-scale state above the model ceiling is indeed applied also for the downward longwave radiative flux. We have modified the text accordingly. For further information on long wave and short wave radiation, readers are referred in the section 3.1 to Pincus & Stevens (2009).

6. Around 1. 245 and related to my previous point 8. I am not sure why "Newtonian" is important to say wrt to the relaxation. It would be more important to explicitly state that the relaxation dumps small-scale perturbations, in contrast to the approach when the mean is relaxed to the prescribed profiles. BTW, such relaxation also serves as gravity wave absorber, correct? So why an extra absorber is added in the top 1 km of the domain? Maybe because of a much shorter time scale is used there.

We explicitly state that the continuous nudging applied is Newtonian relaxation as opposed and not some more complicated spectral nudging. The continuous nudging here is towards the prescribed profiles with the purpose of preventing excessive model drift in this height range, and reducing perturbations in the temporal domain. At each vertical level, the difference between the prescribed profile and the current horizontal mean is added to all gridpoints (Neggers et al., 2012), independent of how far from the horizontal mean they are. Therefore, it neither acts as gravity wave absorber nor removes perturbations small-scale spatial perturbations.

The sponge layer instead nudge values at each gridpoint towards the horizontal mean. The strength of this nudging is increasing with altitude. At the top of domain it reaches the timescale of ≈ 6 minutes (Heus et al., 2010).

For clarity we have added references to sources that describe continuous nudging and sponge layer.

7. L. 258: Considering statistics shown in Fig. 4 from observations, I would like to see similar statistics from model simulations. In particular, what is the range of CCN spatial fluctuations simulated by the model?

The full 3D fields of CCN concentration were unfortunately not amongst the stored outputs of the simulations. That said, such comparison would be statistically inconsistent: the LES follows the trajectory of the air mass, while

the aircraft sampled on a race track which can be also affected by mesoscale variability.

8. L. 262: "... using a maximum initial cloud droplet number concentration...". I do not understand what this statement refers to. Please explain.

Replaced with brief description and explanation:

The initial cloud droplet number concentration is supersaturated areas is set accordingly: the initial mean droplet size must be higher than the threshold $\bar{x}_{\min} = 4.2 \cdot 10^{-12}$ g used by Seifert & Beheng (2006), and at most 1/2 of the initial CCN number concentration is activated. The motivation for this initial concentration is straightforward: the simulation can starts with neither too large nor too small droplets, and spins-up without encountering lack of CCN.

9. Fig. 5. The most appealing feature of the figure is the sharp maximum of cloud water near the cloud top (also evident in Fig. 8). This is in contrast to warm stratiform clouds that typically feature a uniformly increasing cloud water with height (e.g., Fig. 4 in Stevens et al. 2005 cited by the authors). Is that because of the presence of ice? If one adds ice and water in Fig. 8a, would the total nonprecipitating water follow close-to-linear profile? Also: the variable show is the mixing ratio, not "mass concentration".

We understand the maximum near the cloud top seems to be sharp, however it can be explained by the effect of ice microphysics: the ice precipitation is removing cloud water from the area below the maxima. This can be clearly seen in the Figure 8, which shows the precipitation in the panel b and the suspended cloud liquid water and ice water in the panel b. We can clearly see a match between the gradient of cloud water mass around 800–1300 m and the mass of ice hydrometeors. We can also see relative change in the mases of snow to graupel, indicationg the riming of cloud droplets (showing the riming tendencies and other hydrometeor tendencies was considered beyond the scope of this paper).

With regard to cited literature The Figure. 4 in Stevens et al. (2005), we also have to consider the spatial scales. Firstly, the distance between the cloud top and the cloud bottom there is there only about 250 m. Both the gradient below and the gradient above are sharper than in our simulations (as seen in Figure 5 and ore clearly in Figure 8).

We have corrected the caption of figure 5 from "mixing ratio" to "specific mass".

10. L. 322: Why does the secondary ice production allow formation of large and heavy ice hydrometeors?

The secondary ice production in supercooled liquid clouds leads to high number of rimed ice hydrometeors (and partially rimed ice hydrometeors) as well as to spread in sizes of ice hydrometeors, and thus also their terminal velocities. Due to differences in the terminal velocities of ice particles they are more likely to further aggregate. In the text we added reference to Sullivan et al. (2017) and Georgakaki et al.(2022) that both deal with this phenomenon. 11. Figs. 9 and 10. The figures show box-whiskers data for the observations, and only mean for the model. Can box-whiskers plots be done for the model as well? I appreciate difficulties in comparing a 1D flights with 2D model data, but I feel comparing statistics would make the analysis better. L. 344 states that comparing particle size distributions is impossible, but comparing spatial variability of mass mixing ratios (and maybe concentrations) would make sense.

We had also consider this comparison, but we have realised that it is better not to. The main reason is that the whiskers would then be statistically inconsistent: the sample size of the LES data is much larger compared to the observations. This is already mentioned in the text. To avoid raising the wrong expectations, we choose here to only show the 1st statistical moment, the mean.

12. L. 359: Something is missing in this sentence.

We have added missing preposition. The sentence is now "by the strong variability in w in the close vicinity of convective cells."

13. Figs. 11 and 12 shows the dramatic impact of assumed initial CCN concentration on cloud macro- and microphysics. However, the analysis leaves me wondering where this impact comes from. BTW, liquid cloud fraction is not defined. For instance, is it calculated locally and then averaged horizontally, or is it based on the horizontally-averaged water and ice mixing ratios? L. 385 suggests the latter per "area fraction". Please make it clear.

The *liquid cloud fraction* is caculated locally. It is defined as the fraction of the gridpoints at each vertical layer where the cloud liquid water content is above the threshold (set to 0.01 gm^{-2} for cloud liquid water).

We have added the description to the caption of the figure.

14. L. 394. How important is the fact that the entrained air is warmer when the boundary layer grows deeper in the 1000 per cc simulation? This aspect is not mentioned in the discussion and I feel it should.

It is indeed an imortant point. We have added the following sentence:"The strengthening can be explained as a consequence of a deep mixed layer growing into a weakly stable overlying layer."

15. Fig. 13 and L. 399: I think "evolutions" would be better than "time series".

adjusted as proposed

16. How is a1 shown in Fig. 13 defined?

The following definition of the model output has been added to the manuscript:

The liquid cloud cover a_l is in the model defined as the fraction of the domain where the vertically integrated cloud liquid water exceeds the minimum threshold of 0.01 g m⁻².

17. Figs 14 and 16. Please plot bars side-by-side, not one behind the other. Initially I thought there was some logic in having some bars wider than others, only to notice that they were simply partially hidden. How is dF rad in Fig. 14 exactly defined? How are LW and SW in Fig 16 exactly defined? For instance, are those across the entire boundary layer, or just near the inversion? The latter drives entrainment, correct?

We have adjusted the bars in order so they are not overlaying each ther anymore.

The $dF_{\rm rad}$ was a typo in the notation. The correct is $\Delta F_{\rm rad}$, the net long wave radiative flux divergence across the liquid cloud layer. We have corrected the notation.

The values of radiative components in the figure 17a (previously 16a) explicitly refer to radiative surface fluxes (as stated in the caption), horizontally averaged over the whole surface of the domain (this is now added to the caption).

18. L. 405: "This suggests that ice formations...". Yes, I agree, but it would be nice to see how. This is where the analysis needs to be expanded.

A few explanatory sentences were added before this statement, as well as Fig. 15 showing the tendencies in ice water budget.

19. L. 421: "Fig. 14f confirms that entrainment rate increases...". I feel additional analysis should explain what physical mechanism(s) are involved.

The phenomenon is briefly described five lines earlier, the entrainment rate is further investigated in the following paragraphs, and the consequences further discussed in (6.3).

20. L. 440. One may wonder if the classical Twomey effect is sufficient to explain 200 W m -2 difference. I doubt it. The dynamical feedback is likely the key. See major point 2.

The Twomey effect would suggest even larger difference than 200 W m⁻² due to increase in both the number of cloud droplets, as well as due to dramatic increase in Liquid water path.

This result is now desribed earlier in the manuscript, and here we are adding just a brief reference to the figure.

21. Since the SST is prescribed, I find the Eq. (3) and its discussion irrelevant. My suggestion is to focus on the dynamical and microphysical effects steaming from the factor 10 CCN concentration change. We do not agree. Equation (3) does not define the SST, but the Surface Energy Budget (SEB) that consists of energy fluxes, which is something entirely different. While the SST is indeed fixed the CCN change does significantly affect the SEB, in ways not yet reported in previous studies. Because the SEB plays a crucial role in Arctic climate change, in particular sea ice melt in response to cloud cover, we feel this insight is relevant and should be reported.

22. In view of the above comments, I did not read sections 6 and 7. I expect these will change significantly after all above comments are addressed.