

Interactive comment on “Processes controlling the seasonal cycle of Arctic aerosol number and size distributions” by B. Croft et al.

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AC: The authors thank anonymous referee #2 for the helpful suggestions and questions, which have led to strong improvements in our manuscript. We indicate how we have addressed each item in the responses below.

RC: 1. Overall, the manuscript is very useful and well-written. I found no major scientific error, but I wish the authors would present the model/model comparisons a bit more carefully. I recommend this work for publication after the following comments are addressed.

AC: 1. We agree with the referee that the manuscript would be improved by more careful model-model comparisons. To assist with making these comparisons, we quantified

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the bias and error (defined in the new Eqs. 6-8) between the measurements and simulations for Figs. 3-6 and presented these results in the new Tables 2-5. Please note that we also removed Fig. 2 as being redundant with Fig. 1, and as a result the original Figs. 4-7 are now Figs. 3-6. Please note that in response to the comments of referee #1, large sections of the discussion related to data analysis (Section 3) have been rewritten to provide more balanced model-measurement and model-model comparisons. Revised text is indicated in red in the updated manuscript. We think these revisions have improved the manuscript considerably since the manuscript does address several figures that contain considerable information and it is necessary to interpret this information carefully.

Please also note that the appendix is removed in the revised manuscript as we considered that Fig. A1 was redundant with Fig. 1. We also have moved Fig. A3 into the main text and removed A2 and 4 as being unnecessary, based on the focus of our discussion. Please also note a correction on the new Figs. 3 and 5 since we had erroneously truncated the Alert size distributions at 10 nm as opposed to 20 nm in the original Figs. 4 and 6. This correction is most evident in the summertime simulation. As well, following the comments of referee 1, Fig. 10 has been revised to have a simpler, more focused presentation.

RC: 2. Page 29081, line 7. The authors jump from the global aerosol to Arctic aerosol suddenly. The transition seems abrupt. Also, the motivation for studying Arctic aerosol seems a little weak to me. The authors should add a little bit discussion more on why particularly on Arctic aerosol. For example, the climate in Arctic is more sensitive to aerosol perturbations than other regions due to the complex positive feedback system there such as snow albedo feedback. This would make the transition more smooth and the motivation stronger.

AC: 2. Thank you for pointing out this abrupt transition in the introduction. We have revised the first paragraph of the introduction to provide clearer motivation for the reasons for studying Arctic aerosols. We now state that ‘Aerosols play an important role in the

Arctic climate, and changing aerosol concentrations are believed to have contributed to the rapid Arctic warming observed over the past few decades (Shindell and Faluvegi, 2009). However, in the Arctic there are complex aerosol feedbacks and strong seasonal aerosol cycles that make study of aerosol-climate interactions particularly challenging in this remote region (Browse et al., 2012; 2015). To address a portion of this challenging puzzle, this study focuses on understanding the processes that control the Arctic aerosol number and size distributions over the entire annual cycle.'

RC: 3. Page 29091, line 18. What did the authors mean by “aerosol formation”? new particle formation? And by “reducing the condensation sink”? “condensation sink” on accumulation mode aerosols?

AC: 3. In the revised text, we consistently use the terminology ‘new-particle formation (NPF)’ as this terminology is more widely understood to refer to the process of stabilized clusters of gas molecules forming new particles. The revised text states ‘These summertime conditions favour new-particle formation (hereafter referred to as NPF) from precursor vapours within the Arctic boundary layer due to the low condensation sink for particle-precursor vapours on to existing aerosol surface area, and the low coagulation sink for newly formed, growing particles (Leaitch et al., 2013; Heintzenberg et al., 2015).’.

RC: 4. Page 29083, line 11. What is the difference between TSI 3776 CPC and TSI 3772 CPC?

AC: 4. We revised the text in Section 2.1 to explain that this difference relates to the aerosol size ranges measured by these instruments. The lower size limits are 4 nm and 10 nm for the TSI CPC 3775 and 3772. Please note that we had erroneously referred to a TSI CPC 3776 and this is now corrected to 3775 in the revised text.

RC: 5. Page 29084, line 7. “the same instrument configuration”? the same as what? Can the authors also clarify which year’s data they used at Mt. Zeppelin site and Alert site?

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AC: 5. We revised the sentence to indicate the instrument configuration was the same over the measurement period considered in our study. The revised text reads ‘Thus, the data used in our study (2011-2013) come from the same instrument configuration.’. The revised text now states that we use measurement data from 2011-2013 for both Mt. Zeppelin and Alert. We added this information both in the abstract and in the last two paragraphs of the introduction.

RC: 6. Page 29084, line 10. What purpose is the Ni-63 neutralizer used for?

AC: 6. The neutralizer is used to apply a Boltzmann charge distribution to the particles before entering the differential mobility analyzer. We removed this sentence since we did not include a similar discussion for Alert.

RC: 7. Page 29084, line 21. “4 degrees by 5 degrees resolution”. Do the authors have any sense how this coarse grid resolution would affect the model results?

AC: 7. Since this study was conducted in the Arctic region, the grid boxes at this resolution are smaller than they would be in regions towards the tropics at this resolution. This increases our confidence in using the 4x5 resolution for these Arctic simulations. We added the following comment in Section 2.3 describing the model ‘All simulations use GEOS-Chem version 9.02 at 4°x5° resolution globally, corresponding to 440 km x 95 km at 80 °N.’. In any global model study, resolution plays a role in the model results and thus we agree that it is important to document the resolution used in the study.

RC: 8. Page 29085, first paragraph. Can the authors briefly clarify how they treated the con- densation growth and coagulation of particles in the model? I believe it would help readers to understand results. Did they consider the effect of nitrate or/and non-volatile SOA on condensation growth? on which size sections? Did they treat coagulation among all size sections? Or just between size sections that are next to each other?

AC: 8. We agree that adding this information would be helpful to readers in understanding the results. We added the following paragraph near the end of Section 2.3.’

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Growth of simulated particles occurs by condensation of sulphuric acid and organic vapours, which we assume to be non-volatile. These vapours condense proportional to the Fuchs-corrected aerosol surface area distribution (Donahue et al., 2011, Pierce et al., 2011, Riipinen et al., 2011). Condensational growth is not a sink for aerosol number but does transfer aerosol number between size bins while increasing aerosol mass. Coagulation is an important sink for aerosol number (particularly for aerosols with diameters smaller than 100 nm), and moves aerosol mass to larger sizes. Our simulations use the Brownian coagulation scheme of Fuchs (1964), and consider coagulation between all particle sizes.’

RC: 9. Page 29085, line 25, Liu et al. (2001) is not appropriate for dry deposition, though it suits well for wet deposition.

AC: 9. We added a reference to Wesley (1989) for the dry deposition scheme. Thank you for noting this omission.

RC: 10. Page 29087, line 8. Is there any justification for $1 \times 10^{-3} \text{ s}^{-1}$?

AC: 10. This threshold is consistent with the maximum process rates indicated in Gettleman et al. (2013). The revised text states ‘This value is consistent with the upper limit for these process rates given in Gettelman et al. (2013).’.

RC: 11. Page 29088, line 12. Pierce et al. (2014) is not seen in the reference list.

AC: 11. We corrected this reference to D’Andrea et al. (2013). Thank you for noting the need for this correction.

RC: 12. Page 29091, line 8-9. This is not consistent with what the authors stated on page 29090, line 9.

AC: 12. The sentence at page 29091, line 8-9 is removed in the revised text.

RC: 13. Page 29091, line 20. Why does wet scavenging have less control on accumulation mode number in the non-summer seasons than the summer season? Because

of less precipitations?

AC: 13. This sentence is removed in the revised discussion. The revised text now indicates that wet removal has a role in controlling the accumulation mode in all seasons. However we discuss in more detail about how the efficiency of wet removal is greater in the Arctic boundary layer in the summer. In our simulations, we parameterize this process with a dependence on temperature. In non-summer seasons wet removal does occur within the Arctic but is less efficient at lower temperatures, and as well wet removal outside the Arctic does influence how much accumulation mode aerosol reaches the Arctic. We discuss this in detail in the revised Section 3.2.

RC: 14. Page 29091, line 25. “Reduces the condensation sink”. the sink of sulfuric acids?

AC: 14. This sentence is removed following the revisions. However we are careful in the revised text to explicitly state ‘the condensation sink for sulfuric acid’ where applicable.

RC: 15. Page 29092, line 26. “Not enough material to contribute to new-particle growth”. Did the authors consider the condensation of SOA on it?

AC: 15. In the revised model description (Section 2.3) we now state that we allow particle growth by SOA condensation, however this source may not be well represented in the Arctic. The revised text states that ‘Growth of simulated particles occurs by condensation of sulphuric acid and organic vapours, which we assume to be non-volatile’

RC: 16. Page 29093, line 7-9. Do these volatile organic compounds come from ocean as well? Is that possible that the deposition of Aitken mode aerosol is underestimated at Alert site?

AC: 16. These VOCs can come from the ocean and these sources for the Arctic are likely not well represented in the model. This is an important and complex problem for control of aerosol number and will be examined in future studies In the revised text we

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acknowledge that there are uncertainties related to deposition of the Aitken mode. The related text reads, 'Recent studies indicate that aerosols as small as 50 nm - 60 nm can activate in the clean Arctic summertime conditions (Leaitch et al., 2013; Leaitch et al., 2015) and we likely under-estimate this removal in our simulations.', although as the revised text indicates 'aerosols larger than about 60 nm are removed by activation scavenging in our simulations.'

RC: 17. Page 29093, line 11. "at both sites and for all seasons". I would say except for the summer season at Alert site.

AC: 17. This sentence is removed in the revised analysis.

RC: 18. Page 29093, line 12. This sentence is a digression. Also, the authors still discuss figures 4 and 5 in the following paragraph. I suggest removing this sentence.

AC: 18. This sentence is removed in the revised text.

RC: 19. Page 29093, line 19-21. Do the authors imply here that the overestimation of Aitken mode aerosol numbers can be explained by the errors in nucleation scheme (NPF)? This is in contrary to the statement in previous paragraph. In addition, is there any way to evaluate NH₃ in the model at Alert site? Did the authors think about the possibility that nucleation is actually associated with organic compounds while the model neglected this?

AC: 19. This sentence is removed in the revised text. However we do discuss that error in the NPF scheme can play a role in this overestimation (simulation of NPF is challenging for global model). This is likely not the entire reason for Aitken mode over prediction since when we shut off all NPF in the model, we still found over estimate of the Aitken mode in winter. Thus, coagulation also has an important role as discussed in detail in the text and there is a delicate balance between the processes of wet removal and NPF.

The NH₃ simulation near Alert is evaluated in Wentworth et al. (2016) relative to recent

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shipboard measurements. Unfortunately there are no NH₃ measurements at Alert. The model does not include NPF by organic compounds. As now noted in the revised text, at present-day we have no good way of combining NPF by both organics and sulphuric acid-ammonia-water ternary schemes in a single mechanism. Recent work by Giamarelou et al. (2015) suggests that nucleation-mode particles in the Arctic are predominantly ammoniated sulfates and thus we prefer to continue with the ternary scheme. We added this information to our model description for clarification about our choice of NPF scheme.

RC: 20. Page 29093, line 25-27. This is a misleading statement. The authors imply that aerosol nucleation is not important for non-Summer Aitken mode aerosol. This is not supported by Figures 4 and 5. The difference of Aitken aerosol numbers between the NEWS-CAV simulation and the NONUC simulation is nearly as large as (or even larger in the Fall season) than that between the NEWS-CAV simulation and the NEWS-CAV+COAG simulation. This suggests that the nucleation may be as important as the in-cloud coagulation, at least a non-negligible process, for non-Summer Aitken mode aerosol. Also, the 20-50 nm aerosol number concentration predicted by the NONUC simulation is even closer to measurements than the NEWS-CAV+COAG simulation at both sites on Fall (SON). This paper do show the importance of the coagulation in clouds, but it also shows the importance of nucleation.

AC: 20. We agree that NPF processes are important for control of the Aitken mode in all seasons and the above statement is removed from the revised text in this section. The revised text indicates that both NPF and coagulation have important controls on the non-summer Aitken number.

RC: 21. Figure 6. Please clarify the gray shaded region.

AC: 21. We added a sentence to the caption to indicate that this gray area bounds the 20-80th percentile.

RC: 22. Figure 7. Can the authors explain why all simulations over-predict aerosol

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numbers for JFM?

AC: 22. The simulations at Mt. Zeppelin do strongly over predict aerosol number in winter (as shown in Fig. 4 and Table 3). This is the subject on ongoing investigation as the revised text notes, the delicate balance between emissions, new particle formation, growth and wet removal is challenging to simulate in the Arctic.

RC: 23. Page 29094, line 13. This sentence is not accurate. The STD simulation captures measured N₂₀ on JJA at Alert site better than the NEWSCAV+COAG simulation.

AC: 23. This sentence is removed in the revised analysis. As well, we found an error in the plotting of this N₂₀ figure for Alert. We had plotted N₁₀ as opposed to N₂₀. The figure is now corrected. In the revised figure, the performance of NEWSCAV+COAG is better in JJA at Alert and is best among the four simulations in terms of the N₂₀ mean fractional bias and mean fraction error across the entire annual cycle as shown in the new Table 4. However, we caution that these metrics across the entire annual cycle miss certain details, such as the close agreement with measurements for simulation STD in June at Alert and areas of over prediction and under prediction can cancel in the bias metrics. The revised discussion is more balanced and does point out when each of the simulation perform well. As well, Table 4 does indicate that the N₂₀ MFE and MFB for STD at Alert are second to NEWSCAV+COAG in being closest to zero (due to this reasonably good performance of STD in early summer).

RC: 24. Page 29095, line 24. Did the authors imply that most of precursors for the nucleation in early spring are transported from the outside Arctic? Because the authors stated next line that in summer there are greatest local precursor emissions.

AC: 24. The second paragraph of Section 3.3 has been revised to give a more detailed explanation about the summertime NPF occurring in the boundary layer and the spring-time NPF occurring in the free troposphere in our simulations. In our simulations, NPF proceeds when the condensation sink for sulphuric acid is low but still some sulphuric acid is being produced. In spring, those precursors of sulphuric acid are likely trans-

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ported in the free troposphere from lower latitudes over regions open water or pollution sources, and NPF occurs when the condensation sink is low. In summer there is more ice-free ocean within the Arctic that can emit dimethyl sulfide (a precursor for sulphuric acid) directly into the Arctic boundary layer and form sulphuric acid there. Then since the boundary layer is cleaner in summer, NPF proceeds here in our simulations. This discussion can be found in the revised second paragraph of Section 3.3.

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