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Atmospheric Chemistry and Physics
Discussions

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 #1 for the detailed and constructive review of our manuscript. We agree with the referee that the manuscript would be improved by major revisions to the data analysis sections. The manuscript has now undergone a major revision, which includes a rewriting of the majority of the results section in order to give a more balanced presentation of the results. We think that the major revision in response to these referee comments has strongly improved the manuscript presentation. We thank the referee for carefully itemizing each concern and below we respond to each item with an explanation about the changes made to address each point.

RC: The study can become publishable without new simulations, but I want to stress that this requires a careful re-analysis of the data and completely rewriting the results section so that it truthfully reflects the data.

AC: The analysis section of the manuscript (Section 3) has been rewritten. Revised text is highlighted in red. The focus of this rewriting was to provide a more balanced and complete presentation related to the model-model and model-measurement comparisons. To assist with interpretation of the results in a more quantitative framework, we conducted calculations of the bias and error (Eqs. 6-8) between model and measurements. These results are presented in the new Tables 2-5 and are used in the revised discussion of Figs. 3-6. Please note that we have removed the original Fig. 2 as we agreed with the referee that this figure was redundant to the information presented in Fig.1. As a result, the old Figs. 4 and 5 are now Figs. 3 and 4. As well we removed Appendix Figure A1 as being redundant with Fig. 1. As well, please note that Figs. 3 and 5 include a correction that is particularly evident in summer. We had erroneously truncated the size distributions at 10 nm as opposed to 20 nm for the original Alert figures (original Figs 4 and 6). This error is corrected in the revised Figs. 3 and 5.

RC: 1. Fig 4 and 5.: The following statement is simply not true: "Of the four simulations, NEWSCAV+COAG provides the closest agreement with the measurements at both sites and for all seasons". For example, NONUC gives a better match in autumn for both sites. At Zeppelin (and for large part of the size distribution also at Alert), NEWSCAV gives a better match in summer.

AC: 1. Following our reanalysis, we have removed this statement. We calculated the model-measurement fractional bias (Eq. 6 in text) for each of the four simulations over two particle-diameter ranges shown in Figs. 3 and 4 (20-100 nm and 100-500 nm). The new Tables 2 and 3 give these bias values. We use red/bold highlights in each table to indicate the simulation with the fractional bias value closest to zero. These tables indicate (as the referee noted) that NEWSCAV does perform better than NEWSCAV+COAG in summer at Zeppelin for both size ranges. As well NONUC is best in autumn for both size ranges and at both sites. These points are discussed in the revised text. We also added discussion to indicate that NONUC may be right but for the wrong reasons. Shutting off all new-particle formation may compensate for errors in the wet removal or coagulation sink terms.

RC: 2. Fig. 6 and 7: “Among our four simulations, the simulation NEWSCAV+COAG yields the closest agreement with the integrated number measurements (N20, N80, N200) in all seasons at both sites.” I’m extremely confused by this statement, as it is so obviously untrue. Are we not looking at the same figures?

AC: 2. Following the data reanalysis, we have removed this statement. As the new Tables 4 and 5 indicate, NONUC does perform best among the four simulations at Zeppelin for the N20, N80 and N200 in terms of the mean fractional bias (MFB, Eq. 7 in text), although the performance in terms of the mean fractional error (MFE, Eq. 8 in text) is best for NEWSCAV+COAG at Zeppelin. This is included in our revised discussion. We also include discussion about the MFB and MFE at Alert being closest to zero for N20 and N80 for NEWSCAV+COAG, but NONUC performing better for N200 MFB and MFE at Alert.

RC: 3. Fig 4 and 5: The following statement is not true for all seasons: “Among our four simulations, the NEWSCAV+COAG simulation gives the closest representation of the number of non-summer Aitken and accumulation mode aerosols relative to the in-situ measurements at both Alert and Mt. Zeppelin.” For example, during autumn (SON), both figures indicates better match for both modes with NONUC. At Zeppelin, also STD seems to capture the Aitken mode number better. At Alert in DJF, NONUC may also perform better for accumulation mode (difficult to say exactly without access to numerical data). These facts must be mentioned.

AC: 3. The above statement is removed following the rewriting of the results section. As indicated in our reply to RC: 1 above, we added a discussion about the best performance of NONUC in autumn at both sites and this is also shown in Tables 2 and 3.

We did find for the accumulation mode that NONUC performed best in winter at Alert and also Zeppelin and this is noted in red in Tables 2 and 3 and part of the revised discussion.

RC: 4. The following statement is misleading: “Figures 4 and 5 show that in summer, the simulations NEWSCAV and NEWSCAV+COAG capture the dominant Aitken mode.” For Zeppelin, STD captures this features in practice just as well. Further down page 29092, one should stress that both NEWSCAV and NEWSCAV+COAG *strongly* over- estimate particle number below 30 nm (actually 40 nm for NEWSCAV) at Alert.

AC: 4. The above statement is removed in the revised text. The text now discusses that the particle number is strongly overestimated at Alert for sizes smaller than 40 nm (and underestimated from 40-100 nm). We also note the need for care in interpreting the fractional bias values over this range where errors of over and under prediction will cancel over a given size range. For Zeppelin in summertime, we found that NEWSCAV gave the best match to the Aitken mode based on the bias values and this is included in the revised discussion and shown by Tables 2-3.

RC: 5. Fig 4 and 5: It is true that NEWSCAV improves the match with measured accumulation mode number (> 100 nm) most in the summer. However, the fact that it improves the match with the observed number of particles larger than 200 nm also in some other seasons is very significant for correctly simulating the aerosol direct effect, and thus deserves a mention.

AC: 5. We agree that this improved match with measurements of particles larger than 200 nm for NEWSCAV in other non-summer seasons should be mentioned and we added this discussion. This highlights the control of wet removal on the accumulation mode throughout the annual cycle as we now emphasize in our revised discussion.

RC: 6. Fig 4 and 5: “Thus, errors in the new-particle formation processes cannot account for the non-summer Aitken mode over prediction –“ True that it cannot account for all, but it clearly could account for a lot (if not the majority) of it.

AC: 6. This statement is removed in the revised text. We acknowledge in the revised text that new-particle formation (NPF) and growth can play a role in the Aitken mode over prediction. As part of our related discussion, we state in Section 3.2 that 'The balance of these processes of NPF, growth, and wet removal is a challenge for Arctic simulations of number and size. Among the four simulations and in all seasons at both sites (except for summer at Mt. Zeppelin), NEWSCAV strongly overestimates the number of 20-40 nm diameter particles.'.

RC: 7. Fig 6 and 7: This statement is not true for Zeppelin: "The summertime minimum in N200 is over-predicted by about a factor of two for simulation STD. Wet removal revisions for simulation NEWSCAV yield a factor of two reduction to give very close (within 20 %) agreement with the measurements)."

AC: 7. This statement is not included in the revised discussion.

RC: 8. Fig 6 and 7: "The simulation NEWSCAV+COAG has the closest agreement with the seasonal cycle in the measurements." At Alert, NEWSCAV also performs similarly well (in summer even better), which should be acknowledged.

AC: 8. In regard to the old Figs 6 and 7 (now Figs. 5 and 6), the Tables 4-5 and revised text now acknowledges when NONUC and NEWSCAV performs better than NEWSCAV+COAG. Please note that the original version of this figure for Alert, we had erroneously plotted the N10 as opposed to the N20 for Alert. This error is corrected in the revised figure.

RC: 9. Fig 6 and 7: "STD also over-predicts the summertime effective diameter by about a factor of two" Not true for Zeppelin.

AC: 9. We have revised the text to read the text to read 'The simulations over-predict the aerosol effective diameter in July and August, except for NEWSCAV at Mt. Zeppelin.' As well, Tables 4 and 5 quantify the mean fractional bias and error for the simulation relative to measurements over the annual cycle.

RC: 10. It should be stated more clearly what new knowledge this study contributes to our understanding of the Arctic aerosol cycles. For example, the importance of transport and accumulation of pollution in the spring months as well as of the summertime removal processes has been well known for a long time. On the other hand, interstitial coagulation has previously reached much less attention.

AC: 10. We agree with this suggestion that the presentation would be helped by a greater emphasis on the new knowledge that the study contributes. We have made changes throughout the text in response to this comment. As there has been much attention on the spring-summer period, we point out in the introduction that our study is unique in considering number and size distribution over the entire annual cycle. "To our knowledge, ours is the first global modeling study to consider the complete annual cycle in Arctic aerosol number and size. "We also now use the word 'annual' as opposed to 'seasonal' in the title and throughout the text to emphasize the focus on the complete annual cycle. Further to this, we place a greater emphasis on the importance of the coagulation mechanism by giving this greater focus in the abstract and introduction starting with the comment 'While the importance of wet removal is been known, there has been relatively less attention given to coagulation of interstitial particles in clouds, which is an important sink process for the number of particles smaller than about 200 nm.' and also emphasize the development in Section 3.2.

RC: 11. Intro: P29081, L2: How does the climate impact of aerosols strongly depend on the mass distribution (in addition to number and size distribution)? L13-17: Tunved was hardly the first one proposing this.

AC: 11. The word 'mass' has been removed as redundant and the text reads as 'The climate impact of aerosols strongly depends on aerosol number and size distributions.' We did not mean to suggest that Tunved et al. were the first to propose these controls on the number and size distribution. We added the following sentence, 'This inter-seasonal transition from spring to summer has been extensively studied; evidence suggests control by changes in aerosol wet removal efficiency and transport patterns (e.g. Korhonen et al., 2008, Garrett et al., 2010, Sharma et al., 2013).' Thus we indicate work dating back to 2008 related to proposed controls on the spring-summer transition.

RC: 12. P29082, L 25: "through stainless steel" – missing word (inlet)? P 29083-4: The description of Alert site instrumentation is much more detailed than that of Zeppelin site -> harmonize

AC: 12. Thank you for noticing this error – we added the word 'tubing' here. We have also added further details about the instrumentation at Zeppelin in Section 2.2 in order to match better with the level of detail in the Alert description.

RC: 13.Section 2.3: Which model levels are used in comparison? Zeppelin is located on a mountain on an island and thus shouldn't be compared to model surface layer results.

AC: 13. We use the model level at about 500 m for comparisons shown. This is noted in the methods (Section 2.3) 'Simulations at Mt. Zeppelin are sampled at the station altitude of 500 m.'.

RC: P.29085: The validity of the nucleation mechanism is impossible to evaluate at this stage, since the manuscript detailing it is "in preparation" and not accessible to the reviewers. What seems odd is that this mechanism produces significant nucleation in Arctic winter months, i.e. when there is extremely little solar radiation need to produce sulfuric acid. Where is the sulfuric acid coming from in the model? What are the modelled winter-time sulfuric acid levels in the Arctic and how do they compare with observations/other models?

AC: We have updated the citation for the nucleation mechanism as the related study is now published in GMDD. In our simulations, the nucleation (new-particle formation) that occurs in the Arctic winter occurs in the middle/upper troposphere. We added the following discussion related to Fig. 8. 'Simulated NPF occurs in the dark Arctic wintertime since the oxidant OH is produced through reaction of ozone and volatile organic

compounds, although the OH mixing ratios are three-fold less than in summer. As a result, sulphuric acid (a particle precursor vapour) can be produced though oxidation by OH of DMS and sulphur dioxide (SO2) transported into the Arctic in winter. Our simulated Arctic wintertime sulphuric acid is about 0.01 ppt near the tropopause and diminishes towards the Earth's surface. Measurements by Möhler and Arnold (1992) indicate wintertime sulphuric acid levels in Northern Scandinavia of about 0.1 ppt near the tropopause decreasing to 0.01 ppt near the Earth's surface, implying the true nucleation rate could be even higher.'

RC: 14. Section 2.4: Eqs. 2 and 3: It is unclear how one arrives at Eq 3. There is no beta in Eq 2 to be replaced with Eq. 1.

AC: 14. Thank you for pointing out this omission. The equation has been corrected and beta now appears in the denominator.

RC: 15. I suggest removing Fig. 2 since it adds very little (if any) additional information to Fig. 1. The discussion on total number concentration can be kept.

AC: 15. We agree with this suggestion and removed Fig. 2 as the information was redundant with Fig. 1, and we kept a brief comment about the total number concentration.

RC: 16. P29091, L 8-9: Isn't the summertime variability more likely to be associated with nucleation event and non-event days?

AC: 16. The text to discuss Fig. 1 has been revised to read 'In Fig. 1, the magnitude between the 20th to 80th percentiles for particles smaller than 100 nm is greatest during the months of June to August when new-particle formation (NPF) processes in the Arctic boundary layer are expected to make strong and episodic contributions to the aerosol number (e.g. Korhonen et al., 2008; Leaitch et al., 2013).'.

RC: 17. P29093, L4-5: "Although the over prediction of the number of 20-30 nm at Alert is reduced." This is not a full sentence and it is unclear what it refers to.

AC: 17. This sentence has been removed in the revised discussion.

RC: 18. P29093, L23: “This unphysical simulation. . .” NONUC is ‘unphysical’ in the sense that it does not include one microphysical process – but given that including this pro- cess doesn’t seem to capture all the physical processes either (match to observations isn’t super good anyway), I would not call this one simulation more unphysical than the others.

AC: 18. We have removed this terminology. We added discussion about how NONUC can be right for the wrong reasons due to cancelling errors in the sink terms of wet removal and coagulation with a removal of the process of new particle formation. We added this discussion to the text regarding Figs. 3 and 4.

RC: 19. P29094, L1-2: What is “more than 75%” based on?

AC: 19. The revised discussion does not include this statement and we now quantify the differences between simulations using the bias metrics presented in the new Tables 2-5 and defined in Eqs. 6-8.

RC: 20. P29094, L16-: “The 3-fold wintertime over prediction –“ Which simulation does this refer to?

AC: 20. This statement does not appear in the revised text. We now use the mean fractional bias and mean fractional error as metrics for comparing the simulated N20, N80 and N200 with measurements as presented in Tables 4-5.

RC: 21. P29095, L24: precursors of what?; L26-27: maxima -> maximum (or ‘maxima which ARE’)

AC: 21. The sentence at P29095 does not appear in the revised text. We corrected to word maxima to maximum in the following revised sentence ‘The simulated early-spring NPF rate maximum for nucleation-size particles is associated with NPF in the middle and upper troposphere, and as a result is not evident in the measurements at Alert and Mt. Zeppelin.’.

RC: 22. Fig. 9: Why isn’t condensation seen as a loss process for nucleation mode (it is a source process for the Aitken model)? What is the logic of giving the *inverse* of accumulation or loss (black line)? I found it very confusing.

AC: 22. Condensation is a loss process for the nucleation mode but we find that coagulation is dominant such that condensation does not show up on the linear scale. As well, we re-plotted this figure (now Fig. 8) such that the sign is flipped for the net build-up or loss.

RC: 23. Fig. 9: “Primary particle emissions within the Arctic account for about 10–20% of the source rate throughout the year in our simulation” Of the Aitken mode source rate? How can it be 10-20% throughout the year with such a constant emission rate and such a highly varying transport rate? " – dry deposition accounting for about 20–25% of remaining sink.” Since dry and wet deposition seem to be the only two factors affecting the *remaining sink* (i.e. if coagulation not taken into the account), doesn’t the figure imply that dry deposition is responsible for more than 50% of the remaining sink?

AC: 23. The sentence regarding primary particle emissions has been revised to read ‘For the Aitken mode, simulated primary particle emissions within the Arctic have a relatively constant source rate throughout the year, quite similar in magnitude to the maximum condensational growth rate for the Aitken mode in March-April.’. We revised the sentence regarding dry deposition to read ‘Coagulation is the dominant sink for the Aitken mode with dry deposition accounting for the majority of the remaining sink.’.

RC: 24. What causes the minimum in the simulated size distributions around 60 nm (Hoppel minimum), if not cloud processing of activated particles? Here activation size to cloud droplets is 80 nm.

AC: 24. In our simulations, the larger of the Aitken mode particles (about 60-100 nm) do activate to form cloud droplets and are removed as precipitation forms. To avoid confusion we now state in the methods that the assumption about 80 nm is only for the purpose of the interstitial coagulation parameterization.

RC: 25. Fig. 10: From which latitudes are the nucleation mode particles transported (4-10 km altitude) - i.e. from how far they travel without growing or coagulating? Where does the spring time peak transported dust come from? It is stated that "Figure 10 shows that the early spring-time transport occurs mainly at altitudes above 4 km, a time when the polar dome still extends relatively far southward." This is not true for the coarse mode that is the topic of this paragraph. Perhaps the authors are talking about the other modes here, but since it is in no way indicated, it is impossible to know.

AC: 25. In regard to the possible latitudes of origin of the nucleation mode, we expect that there are episodes such as after scavenging when the troposphere may be very clean and particularly towards the upper troposphere such clean conditions can occur such that the lifetime of nucleation mode particles could be quite long (about one week). Thus these very small particles could be transported over considerable distances. We added this following comment about the potential for this longer lifetime with respect to coagulation, 'At these altitudes and particularly when the atmosphere just been cleaned by a precipitation event, if the Aitken and accumulation mode concentrations are low (5-10 cm-3), then nucleation-mode particles can have a lifetime of about one week with respect to loss by coagulation.'. In regard to the question about springtime transport, we have revised this sentence to explicitly refer to the coarse mode, which was the topic of the paragraph. The sentence now reads 'Figure 9 shows that the early springtime transport of the coarse mode occurs mainly at altitudes between 1.5 and 4 km, a time when the polar dome still extends relatively far southward.'

RC: 26. I find Figures A2-A4 quite redundant and suggest leaving them + the one paragraph discussing them out. If the authors insist on keeping these figures, take them out of the appendix and justify their significance better.

AC: 26. We agree that some of this presentation regarding aerosol processes in other latitude ranges could be removed. For example, there are quite a few similarities be-

tween the 78N and 66N figures, and also similarities between the 50N and global figures. We decided to remove the 78N and global figures and retain the 50N figure, putting it into the main text with a discussion at the end of Section 3.3 that better justifies the significance of the figure in putting the 66N figure in context. As a result of these changes, the manuscript no longer has an appendix section.

RC: 27. P29098, L16-17: there is no clear mention of latitudinal dependencies when dis- cussing Figs. 9 and 10.

AC: 27. This sentence is removed in the revised discussion.

RC: 28. P29098 L20-21: "may be considered as the inverse of the wet removal efficiency" Don't you mean "are approximated here as"? What is the logic for showing the wet removal lifetime for all these altitudes? At 10 km, the lifetime seems to be > 10ˆ5 days → clearly this is not the dominant process here. To evaluate the conclusions, it would be important to know the corresponding lifetimes also for other processes (all altitude ranges)

AC: 28. This sentence is removed in the revised discussion We agree that showing such an extensive set of lifetimes at many altitudes was excessive and distracting from our main point. We removed this figure and replaced the figure with the simplified Fig. 10, which better illustrates our points that 1) there is a change in accumulation aerosol number lifetime during the annual cycle and that the timing of the sharp decrease in lifetime coincides with the time when the Arctic haze layer diminishes and 2) there is a minimum in the Arctic boundary layer lifetimes in October, coincident with the total particle number minimum.

RC: 29. P29098, L22-24: "This simulated aerosol lifetime with respect to wet removal has a summertime minimum in the Arctic for aerosols in the Aitken, accumulation and coarse size ranges throughout the troposphere". Do you refer to north of 66 deg here? If so, the green line (closest to ground) has a minimum in the autumn, not summer.

AC: 29. These sentences are removed. The revised figure (Fig. 10) only includes two layers below 4 km and two regions (north of 50N and north of 66N) and we now state in ‘In our simulation wet removal lifetimes in the Arctic boundary layer below 1.5 km reach a minimum in October ‘.

RC: 30. P29099, L4-6: Not true for coarse mode.

AC: 30. This sentence is removed in the revised discussion.

RC: 31. Note: I have not reviewed the conclusions section, since I expect it to change significantly once the authors redo their analysis.

AC: 31. The conclusion has been extensively revised to reflect our data reanalysis. The changes are highlighted in red in the revised manuscript.

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