Response to Referee # 1

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), NEWS- CAV gives a better match in summer.

AC: 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: 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 (Eq. 7 in text), although the performance in terms of the mean fractional error (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: 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: 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 accumula- tion 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: 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 overprediction —" True that it cannot account for all, but it clearly could account for a lot (if not the majority) of it.

AC: This statement is removed in the revised text. We acknowledge in the revised text that new-particle formation 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: 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: 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: 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: 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: 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: 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: 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 (SO_2) 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: 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: 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: 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: 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: 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: 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 overprediction —" Which simulation does this refer to?

AC: 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: 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: 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: 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: 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: 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: 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 between 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 discussing Figs. 9 and 10.

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

Response to Referee 2

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

RC: 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: 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 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: 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: 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: Page 29091, line 18. What did the authors mean by "aerosol formation"? new particle formation? And by "reducing the condensation sink"? "condensation sink" on accumu- lation mode aerosols?

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

AC: 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: 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?

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

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

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

RC: Page 29087, line 8. Is there any justification for 1x10-3 s-1?

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

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

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

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

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

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

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

AC: This sentence is removed in the revised analysis.

RC: 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: This sentence is removed in the revised text.

RC: 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: 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 NH_3 simulation near Alert is evaluated in Wentworth et al. (2016) relative to recent shipboard measurements. Unfortunately there are no NH_3 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: 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 NEWSCAV simulation and the NONUC simulation is nearly as large as (or even larger in the Fall season) than that between the NEWSCAV simulation and the NEWSCAV+COAG sim- ulation. This suggests that the nucleation may be as important as the in-cloud coagula- tion, 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 NEWSCAV+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: 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: Figure 6. Please clarify the gray shaded region.

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

RC: Figure 7. Can the authors explain why all simulations over-predict aerosol numbers for JFM?

AC: 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: 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: 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: 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: 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 springtime 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 transported 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.

Summary of relevant changes:

All relevant manuscript changes are documented in the author responses above and are highlighted in red in the following revised manuscript.

Processes controlling the annual cycle of Arctic aerosol number and size distributions

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Abstract

Measurements at high-Arctic sites (Alert, Nunavut and Mt. Zeppelin, Svalbard) during the years 2011 to 2013 show a strong and similar, annual cycle in aerosol number and size distributions. Each year at both sites, the number of aerosols with diameters larger than 20 nm exhibits a minimum in October, and two maxima, one in spring associated with a dominant accumulation mode (particles 100 nm to 500 nm in diameter), and a second in summer associated with a dominant Aitken mode (particles 20 nm to 100 nm in diameter). Seasonal-mean aerosol effective diameter from measurements ranges from about 180 nm in summer to 260 nm in winter. This study interprets these annual cycles with the GEOS-Chem-TOMAS global aerosol microphysics model. Important roles are documented for several processes (new-particle formation, coagulation scavenging in

clouds, scavenging by precipitation, and transport) in controlling the annual cycle in Arctic aerosol number and size.

Our simulations suggest that coagulation scavenging of interstitial aerosols in clouds by aerosols that have activated to form cloud droplets strongly limits the total number of particles with diameters less than 200 nm throughout the year. We find that the minimum in total particle number in October can be explained by diminishing new-particle formation within the Arctic, limited transport of pollution from lower latitudes, and efficient wet removal. Our simulations indicate that the summertime-dominant Aitken mode is associated with efficient wet removal of accumulation-mode aerosols, which limits the condensation sink for condensable vapours. This in turn promotes new-particle formation and growth. The dominant accumulation mode during spring is associated with build up of transported pollution from outside the Arctic coupled with less-efficient wet removal processes at colder temperatures. We recommend further attention to the key processes of new-particle formation, interstitial coagulation, and wet removal and their delicate interactions and balance in size-resolved aerosol simulations of the Arctic to reduce uncertainties in estimates of aerosol radiative effects on the Arctic climate.

1. Introduction

The climate impact of aerosols strongly depends on aerosol number and size distributions (Haywood and Boucher, 2000; Lohmann and Feichter, 2005). These aerosol properties, in addition to chemical composition, contribute to aerosol effects on the Earth's climate. Aerosols influence the global climate directly through scattering and absorption of radiation (Charlson et al., 1992), and indirectly by modifying cloud properties

(Twomey, 1974; Albrecht, 1989). 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.

Observations at Arctic sites show a strong and similar annual cycle in aerosol number and size distributions (e.g. Ström et al., 2003; Leaitch et al., 2013; Tunved et al., 2013). In the high Arctic, at Mt. Zeppelin, Svalbard, and Alert, Nunavut, Canada, the observed annual cycle in aerosol number exhibits two maxima: one in March-April associated with dominance of accumulation-mode particles and one in July associated with smaller, Aitken-mode particles. The inter-seasonal transition from accumulation-mode-dominated springtime distributions to Aitken-mode-dominated summertime distributions has been observed not only at surface sites, but also in the free troposphere (Engvall et al., 2008). This inter-seasonal transition from spring to summer has been extensively studied; evidence suggests control by changes in aerosol wet removal efficiency, new-particle formation, and transport patterns (e.g. Korhonen et al., 2008, Garrett et al., 2010, Sharma et al., 2013). More-efficient wet removal in the mid latitudes and within the Arctic in late spring and summer inhibits transport of aged accumulation-mode aerosols into the Arctic. 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).

Korhonen et al. (2008) conducted a pioneering global aerosol model study to interpret the processes controlling the spring–to-summer transition in Arctic aerosol number and size observed from Svalbard and the shipboard campaigns of Heintzenberg et al. (2006). The focus of that study was limited to spring-summer and the transition between these seasons. In our study, we extend the temporal scope to consider the entire annual cycle and use observations from both Svalbard and Nunavut, about 1000 kilometers apart. Over recent years, numerous studies have focused on the spring-summer transitions in aerosol mass abundance using observations and models to examine the role of transport and scavenging (Garrett et al., 2010; Garrett et al., 2011; Browse et al., 2012; DiPierro et al., 2013; Sharma et al., 2013, Stohl et al., 2013). However, there has been considerably less focus on Arctic aerosol number and size distributions. To our knowledge, ours is the first global modeling study to consider the complete annual cycle in Arctic aerosol number and size.

In this study, we examine aerosol number and size distributions over recent years (2011-2013) at the Canadian high-Arctic measurement site at Alert, Nunavut (82.5 °N) and the European site at Mt. Zeppelin, Svalbard (79 °N). We use the GEOS-Chem global chemical transport model (Bey et al., 2001; www.geos-chem.org) with the size-resolved aerosol microphysics package TOMAS (D'Andrea et al., 2013; Pierce et al., 2013;

Trivitayanurak et al., 2008) to examine the relative importance of various aerosol processes (NPF, emissions, removal, and microphysical processes such as condensation and coagulation) in controlling the annual cycle of aerosol number and size distribution in the Arctic.

While the importance of wet removal is well known (Korhonen et al., 2008; Garrett et al., 2010; Browse et al., 2012), relatively less attention has been given to coagulation of interstitial particles in clouds, which is another sink process for aerosol number. We implemented a mechanism in GEOS-Chem-TOMAS that represents coagulation between aerosols that have activated to form cloud droplets and interstitial aerosols (defined as particles within clouds, but outside of cloud droplets). This mechanism accounts for the ~100-fold increase in size (due to water uptake) for particles that are cloud condensation nuclei (CCN) and have activated to form cloud droplets. This size change increases the coagulation rate of smaller Aitken-mode aerosols with these larger activated aerosols. Pierce et al. (2015) showed that the inclusion of this mechanism to GEOS-Chem-TOMAS brings aerosol size distributions to closer agreement with observations, globally. Cesana et al. (2012) analyzed CALIOP retrievals using the cloud-phase detection algorithm and found that low-level liquid clouds are ubiquitous in all seasons in the Arctic. Thus, this in-cloud coagulation process is particularly relevant for the Arctic.

The following section describes the 2011-2013 high-Arctic measurements and gives an overview of the GEOS-Chem-TOMAS simulations conducted for this study. Section 3 examines the monthly and seasonal mean in-situ observations of aerosol number and size from scanning mobility particle sizer (SMPS) at Alert, and differential mobility particle

sizer (DMPS) at Mt. Zeppelin. The GEOS-Chem-TOMAS model is used to interpret the annual cycle of these measurements. We subsequently present the process rates that control the aerosol annual cycles in our simulations.

2. Method

2.1. Measurements at Alert

Measurements of particle size distributions at Alert have been ongoing since March 2011 with the exception of a few technical interruptions. Sampling of the ambient aerosol size distribution at Alert was conducted as described by Leaitch et al. (2013). Briefly, the particles are sampled through stainless steel tubing with a mean residence time for a particle from outside to its measurement point of approximately 3 s. At the point of sampling, the aerosol is at a temperature (T) of approximately 293K and the relative humidity (RH) is <50%. The total number concentration of particles larger than 10 nm in diameter at Alert is measured with a TSI 3772 Condensation Particle Counter (CPC) operating at a flow rate of 1 litre min^{-1} (lpm). The size distributions for particles from 20 nm to 500 nm in diameter are measured with a TSI 3034 Scanning Mobility Particle System (SMPS), operating at a flow rate of 1 lpm and verified for sizing on site using mono-disperse particles of polystyrene latex and of ammonium sulphate generated with a Brechtel Manufacturing Incorporated (BMI) Scanning Electrical Mobility Spectrometer (SEMS) and for number concentrations through comparison with the TSI 3772 CPC. The Alert SMPS data are accurate to within 15%, in terms of number concentration and sizing. The TSI 3772 CPC was initially compared with a TSI 3775 CPC temporarily operating at the site and measuring the number of particles with sizes larger than 4 nm.

The differences between the TSI 3772 and 3775 CPC were found to be <10% when there was no evidence of particles smaller than 10 nm. The TSI 3772 CPC also compares to within 10% with the SMPS when particle sizes are large enough for all particles to be counted by both instruments (e.g. during periods of Arctic Haze).

2.2. Measurements at Mt. Zeppelin

The Department of Environmental Science and Analytical Chemistry, Section for Atmospheric research (ACES), Stockholm University (SU), has monitored the sub-micron aerosol number size distribution at Mt. Zeppelin since 2000 with a differential mobility particle sizer (DMPS). Today, this more-than-15-year continuous dataset constitutes one of the longest unbroken aerosol number size distribution observation series in the Arctic.

During the 15 years of operation, the DMPS system has undergone a number of modernizations. Initially a single differential mobility analyzer (DMA) system was used covering a size range between roughly 20-600 nm. A major overhaul was performed during late 2010, and since then the setup has remained unchanged, covering a size range of 5-800 nm. Thus, the data used in our study (2011-2013) come from the same instrument configuration.

This DMPS-system utilizes a custom-built twin DMA setup comprising one Vienna-type medium DMA coupled to a TSI CPC 3772 covering sizes between 25-800 nm and a Vienna-type long DMA coupled with at TSI CPC 3772 effectively covering sizes

between 5-60 nm. The size distributions from the two systems are harmonized on a common size grid and then merged. Both systems use a closed-loop setup. The inlet hat is a whole air inlet according to EUSAAR standard. At current setup, the inlet operates with a flow rate of about 100 lpm and consists of several stainless steel tubes. The 25 mm diameter DMPS sampling tube is in total 4.5 m long. Inside the station, the flow is split into progressively smaller tubing until reaching 1 lpm at the DMPS. Laminar flow condition applies throughout the sampling line. On the outside, the inlet temperature is kept above 273K using active heating. Inside the station the temperature increases gradually to room temperature (maximum temperature of 298K, but typically around 293K). RH and T are internally monitored and measurements are at dry conditions with RH<30%. The system is regularly checked with latex spheres and flow controls. The data are manually screened and crosschecked with other available observations.

2.3. GEOS-Chem-TOMAS model description

In this study, we use the GEOS-Chem-TOMAS model, which couples the GEOS-Chem global chemical transport model (www.geos-chem.org, Bey et al., 2001) with the TwO-Moment Aerosol Sectional (TOMAS) microphysics scheme (Adams and Seinfeld, 2002; Lee and Adams, 2012). All simulations use GEOS-Chem version 9.02 at 4°x5° resolution globally, corresponding to 440 km x 95 km at 80 °N. The model has 47 layers extending from the surface to 0.01 hPa. Simulations at Mt. Zeppelin are sampled at the station altitude of 500 m. Assimilated meteorology is from the National Aeronautics and Space Administration (NASA) Global Modelling and Assimilation Office (GMAO) Goddard Earth Observing System version 5 (GEOS-5). All simulations use meteorology and emissions for the year 2011 following 3 months spin-up at the end of 2010. GEOS-

Chem includes simulation of more than 50 gas-phase species including oxidants such as OH and aerosol-precursor gases such as SO_2 and NH_3. Emissions in GEOS-Chem-TOMAS are described in Stevens and Pierce (2014). In addition, we implement seabird-colony NH_3 emissions from Riddick et al. (2012) with modifications for additional colonies in the Arctic region based on the on-line Circumpolar Seabird Data Portal (Seabird Information Network, 2015) as described and evaluated in Wentworth et al. (2016) and Croft et al. (submitted). Our simulations include secondary organic aerosol (SOA), both biogenic (~19 Tg yr^{-1}) and enhanced anthropogenic non-volatile (100 Tg yr^{-1}) spatially correlated with anthropogenic CO emissions (D'Andrea et al., 2013).

The TOMAS microphysics scheme tracks the number and mass of particles within each of 15 dry size sections. The first 13 size sections are logarithmically spaced, including aerosol dry diameters from approximately 3 nm to 1 μm, while 2 additional size sections represent aerosol dry diameters from 1 μm to 10 μm (Lee and Adams, 2012). Simulated aerosol species are sulphate, sea-spray, hydrophilic organics, hydrophobic organics, internally mixed black carbon, externally mixed black carbon, dust and water. Aerosol hygroscopic growth is a function of grid box mean relative humidity (RH) capped at 99%. Simulated aerosols are treated as dry (RH=0%) for comparison with the measurements presented in this study

For these simulations, NPF is treated according to the state-of-the-science ternary H_2SO_4-NH_3-H_2O nucleation scheme described by Baranizadeh et al. (2016). The formation rate of particles at circa 1.2 nm in mass diameter is determined from a full kinetics simulation

by Atmospheric Cluster Dynamics Code (ACDC; Olenius et al., 2013) using particle evaporation rates based on quantum chemistry. The scheme is implemented as a comprehensive look-up table of simulated formation rates as a function of sulphuric acid and ammonia vapour concentrations, relative humidity, temperature, and condensation sink for condensable vapours (existing aerosol surface area). Growth and loss of particles with diameters smaller than 3 nm are approximated with the Kerminen et al. (2004) scheme (evaluated in TOMAS in Lee et al. (2013a)). In our simulations, we do not include NPF by organic vapours such as those arising from the oceans (O'Dowd and de Leeuw, 2007; Fu et al., 2013). Currently, no single nucleation scheme includes contributions from organics, sulphuric acid, bases, and water. As well, Giamarelou et al. (2016) found that nucleation-mode particles in the Arctic are predominantly ammonium sulphates.

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.

In our simulations, aerosols are removed from the atmosphere by precipitation both in and below clouds (Liu et al, 2001), and also by dry deposition using a resistance in-series approach (Wesley, 1989) assuming an aerosol dry deposition velocity of 0.03 cm s^{-1} over snow and ice. Wet deposition is an important sink process for aerosols larger than about 50-100 nm in diameter. The in-cloud wet scavenging parameterization in the standard GEOS-Chem-TOMAS module uses the same equations for the removal efficiency and the precipitation fraction as in the bulk-aerosol GEOS-Chem module described in Liu et al. (2001) with updates implemented by Wang et al., (2011) to account for wet removal in mixed-phase and ice clouds. The aerosol in-cloud wet removal in GEOS-Chem-TOMAS is limited to the aerosol size range that is assumed activated into cloud hydrometeors.

2.4 Simulations and revisions to model parameterizations

Table 1 summarizes the four simulations conducted with the GEOS-Chem-TOMAS model. These simulations include 1) a standard, 2) updates to wet removal, 3) updates that add the process of interstitial coagulation of aerosols in clouds, and 4) a sensitivity test with no NPF. The first (simulation STD) uses the standard GEOS-Chem-TOMAS model as described above.

Simulation NEWSCAV introduces developments to the wet removal parameterization to allow for variable in-cloud water content, to implement a temperature-dependent aerosol activation fraction, and to more closely relate in-cloud aerosol scavenging to cloud fraction. The standard GEOS-Chem-TOMAS wet removal efficiency β for large-scale

clouds is based on a parameterization originally developed by Giorgi and Chameides (1986):

$$\beta = k_{min} + Q/L \qquad \text{Eq. 1}$$

where Q is the grid- box mean precipitation production rate [g cm^{-3} s^{-1}] from the GEOS-5 meteorological fields, L is the in-cloud liquid and ice water content [g cm^{-3}] of the precipitating clouds (an assumed constant) and *kmin* is a constant, 1x10^{-4} s $^{-1}$. The k_{min} term represents autoconversion processes that produce precipitation. The Q/L term represents accretion processes. The standard GEOS-Chem model uses a globally fixed value for L of 1x10^{-6} g cm^{-3}. While this value performs well for wet scavenging in a global sense (Liu et al., 2001; Wang et al., 2011), the value does not well represent observations in certain regions. Measurements by Shupe et al. (2001) and Leaitch et al. (2016) show an Arctic spring-summer-mean cloud liquid water content that is an order of magnitude lower (1x10^{-7} g cm^{-3}). During the spring and summer, more efficient aerosol removal in liquid clouds plays a key role in the control of aerosol distributions (Garrett et al., 2010). An overestimation of the liquid water content of Arctic clouds (by using a globally fixed value for L) in our simulation would yield under-vigorous wet removal efficiency, particularly for cases of low intensity precipitation (low Q). To address this issue, we replace the fixed value with the cloud liquid and ice water contents from the GEOS-5 assimilated meteorology fields and calculate the efficiency as the ratio of the grid-mean precipitation production rate and the grid-mean liquid and ice water contents. We impose a maximum efficiency (1x10^{-3} s^{-1}) to prevent over-vigorous

removal. This value is consistent with the upper limit for these process rates given in Gettelman et al. (2013).

In addition, we implement a temperature-dependent representation of the aerosol activated fraction (Verheggen et al., 2007) to account for the fraction of aerosol susceptible to wet removal in mixed-phase clouds. In mixed-phase clouds, only a fraction of the aerosols are contained in the cloud hydrometeors and susceptible to removal when cloud water and ice converts to precipitation. As clouds glaciate, cloud droplets evaporate and release aerosols from the condensed phase because ice crystals grow at the expense of cloud droplets due to differences in the saturation vapour pressure over liquid water and ice. The Verheggen et al. (2007) parameterization for activated fraction accounts for this effect, such that only a fraction of the total in-cloud aerosol is susceptible to wet removal as precipitation forms in mixed phase clouds. However, in strongly riming-dominated regimes, this may lead to an under-estimate of the removal.

We also develop the representation of the precipitation fraction. In the standard GEOS-Chem model, the fraction of the grid box that is precipitating, F, is

$$F = Q/\beta L \qquad \text{Eq. 2}$$

Replacing β with Eq. 1 and simplifying yields

$$F = 1/(1+(k_{min}L/Q)) \qquad \text{Eq. 3}$$

where $k_{min}L$ has a fixed value of $1x10^{-10}$ g cm^{-3} s^{-1} in the standard model version. Thus, the precipitation fraction increases with precipitation production rate. We replace this parameterization by treating the precipitation fraction for aerosol scavenging in clouds as the cloud fraction from the GEOS-5 meteorological fields in the model layers where precipitation is produced. These wet scavenging developments were also implemented in a GEOS-Chem v9-03-01 simulation of ^{137}Cs (also using GEOS5 met fields) and evaluated against ^{137}Cs measurements taken for several weeks following the March 2011 Fukushima Dai-Ichi nuclear power plant accident. Implementation of these scavenging revisions yielded improved agreement with the radionuclide measurements (median ratio of measured to modeled surface layer concentrations changed from 5.53 to 0.52) and reduced e-folding times from 21.8 days to 13.2 days, which is close to the measurement value of 14.3 days (Kristiansen et al., 2015). These wet removal revisions also slightly reduced the mean bias relative to measurements of the number of aerosols larger than 40 nm (N40), 80 nm (N80) and 150 nm (N150) for the same global set of 21 geographically diverse sites as described in D'Andrea et al. (2013) (not shown).

Simulation NEWSCAV+COAG includes additional developments to the interstitial aerosol coagulation mechanism in clouds for the TOMAS microphysics scheme as explored in Pierce et al. (2015). This revised coagulation parameterization accounts for the order 100-fold increase in the wet size of aerosols that activate to form cloud droplets. This simulation assumes for the purposes of coagulation only that 1) aerosols that activate to form cloud droplets must have a dry diameter larger than 80 nm, 2) super-cooled clouds persist to temperatures as low as 238K and 3) all cloud droplets are 10 μm in diameter. While these are crude assumptions, they are within reasonable bounds and

allow examination of the potential of interstitial coagulation to control aerosol size distributions. The grid-box mean coagulation kernel between two size bins is calculated as

$$J_{i,j} = (1-f_{cloudy})K_{clear;i,j,}N_iN_j + f_{cloudy}K_{cloudy;i,j}N_iN_j \qquad \text{Eq. 4}$$

where $J_{i,j}$ is the coagulation rate between particles in bins i and j, f_{cloudy} is the fraction of the grid box that is cloudy, K_{clear} is the coagulation kernel between bins i and j in the clear portion of the gridbox, K_{cloudy} is the coagulation kernel between bins i and j in the cloudy portion of the gridbox, N_i is the number concentration of particles in bin i, and N_j is the number concentration of particles in bin j. While the activated particle is treated as having a diameter of 10 μm, the unactivated collision-partner aerosol is treated as having a diameter following hygroscopic growth under grid-box mean relative humidity. If the in-cloud relative humidity is considerably greater than the grid mean, then the coagulation kernel could be overestimated. These developments to the interstitial aerosol coagulation parameterization in clouds are applied and evaluated in Pierce et al. (2015) and yielded improved agreement with in-situ aerosol size distributions at 21 geographically diverse sites in the Northern Hemisphere.

Simulation NONUC turns off new particle formation (NPF) globally to examine the contribution of NPF to aerosol number in the Arctic. This simulation is otherwise identical to simulation NEWSCAV.

3. Observations and GEOS-Chem-TOMAS simulations of annual cycles in Arctic aerosol number and size

3.1 Observed annual cycle in Arctic aerosol number and size

Figure 1 shows the 2011-2013 monthly median aerosol number distributions from the SMPS at Alert and DMPS at Mt. Zeppelin. At both sites, the accumulation mode (defined here as particles with diameters from 0.1 μm to 0.5 μm due to instrument range, although typically extending to 1 μm) gradually builds during winter to a maximum in March and April. Afterward, the accumulation mode decreases while the Aitken mode (defined here as particles of 0.02 μm to 0.1 μm in diameter due to instrument range, although typically extending to 0.01 μm) increases in number to a maximum in July-August. October is characterized by the lowest number concentrations in both modes until the accumulation mode starts to build again in November. As a result, the total aerosol number at both locations has a shallow maximum in both spring and summer. 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 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). The complete annual cycle is remarkably similar at both sites and similar to that observed at Mt. Zeppelin over an earlier ten-year period from 2000 to 2010 (Fig. 7 in Tunved et al., 2013). The similarity in these number distributions across the one thousand km that separates Alert and Mt. Zeppelin suggests an annual cycle that spans the high Arctic. In the following sections we use the GEOS-Chem-TOMAS model to interpret the processes that control these cycles.

Figure 2 shows the monthly median aerosol effective diameter calculated from the 2011-2013 measurements with SMPS at Alert, and DMPS at Mt. Zeppelin. The effective diameter is the ratio of the second and third moments of the aerosol number distribution, and is useful in determining the optical properties of an aerosol distribution, and for comparing between distributions. The effective diameter is defined as

$$D_{eff} = \int_{Dmin}^{Dmax} D^3 N(D) dD \;\; / \;\; \int_{Dmin}^{Dmax} D^2 N(D) dD \qquad \text{Eq. 5}$$

where D is the aerosol diameter and $N(D)$ is the aerosol number distribution. The integral here is taken over the instrument size range from D_{min} = 20 nm to D_{max} = 500 nm. Despite the geographic distance of these two sites, the annual cycle of the aerosol effective diameter is remarkably similar. At both sites, the aerosol effective diameter shows a strong annual cycle with a minimum during the summer months of about 180 nm and a maximum in the winter of about 260 nm. The effective diameter at Mt. Zeppelin exceeds Alert by about 10-20% throughout the year. In the next sections, we interpret these observed annual cycles in number and size using the GEOS-Chem-TOMAS model.

3.2. Interpreting processes controlling the annual cycle of aerosol number and size with GEOS-Chem-TOMAS

Figures 3 and 4 show the seasonal-median number distributions from measurements at Alert and Mt. Zeppelin, respectively, for winter (DJF), spring (MAM), summer (JJA), and fall (SON), and also for our four simulations. The measurement distributions exhibit

the key features of Arctic aerosol size distributions, a dominant Aitken mode in summer, a dominant accumulation mode with suppressed Aitken mode in non-summer seasons, and minimum number in fall. To assist in interpreting Figs. 3 and 4, we calculate the fractional bias between the observed and simulated total number of aerosols over two size ranges available from the measurement data: 1) Aitken particles 20-100 nm in diameter and 2) accumulation particles 100-500 nm in diameter. We apply a size limit of 20-500 nm to the Mt. Zeppelin measurement data and to our simulations to be consistent with the available data from Alert. We define fractional bias (*FB*) as

$$FB = (C_m - C_0)/C_0 \qquad \text{Eq. 6}$$

where C_m is the model value and C_0 is the observed value. These seasonal fractional bias values are presented in Tables 2 and 3. Among all four simulations, simulation STD never has the fractional bias closest to zero for the size ranges considered in Tables 2 and 3.

The strong control of wet removal on Arctic aerosol number and size distributions throughout the annual cycle is highlighted by comparison of simulations STD and NEWSCAV in Figs. 3 and 4 and in Tables 2 and 3. For both Alert and Mt. Zeppelin, the standard GEOS-Chem-TOMAS model (simulation STD) overestimates the observed number of 100-500 nm diameter particles in all seasons as quantified by the positive fractional bias values in Tables 2 and 3. At both Alert and Mt. Zeppelin, this bias is reduced in spring and summer for simulation NEWSCAV relative to STD. The bias reduction is greatest in summer when aerosol wet removal by precipitation is more efficient within the Arctic boundary layer, and strongly limits the accumulation-mode

number at the surface sites. The efficiency of wet removal is parameterized to increase with temperature (from 238 K to 273 K) in our simulations. In seasons other than summer, wet removal in the Arctic boundary layer is less efficient. However, wet removal outside the Arctic boundary layer continues to influence the number of accumulation-mode particles transported to the measurement sites. Over a limited size range (200-500 nm diameter particles) and in all seasons at both sites, NEWSCAV is a closer match to measurements than STD, but the difference between STD and NEWSCAV is very small at Alert in winter and spring.

Wet removal also has feedbacks that particularly influence Aitken-mode and 100-200 nm diameter particle numbers indirectly through changes in NPF and subsequent particle growth to these sizes. Figures 3 and 4 show that at both sites and in all seasons, more vigorous wet removal in simulation NEWSCAV relative to STD yields more numerous Aitken-mode particles (although the springtime difference is very small), and in fall and winter, also more numerous 100-200 nm particles. A reduction in surface area of 200-500 nm aerosols by more vigorous wet removal (simulation NEWSCAV relative to STD) promotes NPF and particle growth. Other than in summer, this NPF occurs primarily outside the Arctic boundary layer and growth occurs during transport to the measurement sites. As a result of the increase in number of 20-200 nm particles in simulation NEWSCAV relative to STD, the accumulation-mode bias is greater for NEWSCAV in fall and winter at both sites and the Aitken mode bias is greater for NEWSCAV in fall, winter and spring at both sites (Tables 2 and 3).

The balance of these processes of NPF, growth, and wet removal is a challenge for Arctic simulations of number and size. In all seasons at both sites (except for summer at Mt. Zeppelin), NEWSCAV strongly over estimates the number of 20-40 nm diameter particles. Nonetheless, among the four simulations NEWSCAV has the closest-to-zero bias for the 20-100 nm and 100-500 nm diameter particles at Mt. Zeppelin in summer. As well, at Alert, the summertime Aitken-mode bias for simulation NEWSCAV is second smallest (after NEWSCAV+COAG), but the shape of the distribution shown in Fig. 3 is not a perfect match with the observations for either simulation as there are sizes that are strongly over- and under-predicted within the 20-100 nm diameter range.

Figures 3 and 4 demonstrate the importance of in-cloud coagulation (NEWSCAV+COAG relative to NEWSCAV) in reducing the number of 20-200 nm diameter particles in all seasons but to varying degrees. In spring and summer at both sites, this additional coagulation for simulation NEWSCAV+COAG reduces the number of 40-100 nm diameter particles excessively relative to measurements. As a result, simulation NEWSCAV is a somewhat better match to measurements in this 40-100 nm diameter range in spring and summer at both sites. However among the four simulations, NEWSCAV+COAG has the smallest fractional bias for the Aitken mode in winter and summer at Alert, and in winter and spring at Mt. Zeppelin, as well as the smallest springtime accumulation-mode bias at both sites (and smallest summertime accumulation-mode bias at Alert).

Simulation NONUC was designed as a means to assess the relative contribution of NPF processes to the Arctic aerosol size distributions. In our simulations, NPF contributes

most strongly to the number of particles smaller than 200 nm. These contributions occur in all seasons as shown by the differences between NEWSVAC and NONUC in Figs. 3 and 4. In the summertime, NPF occurs within the Arctic boundary layer both in our simulations and in observations (Chang et al, 2011; Leaitch et al., 2013; Allan et al., 2015; Croft et al., submitted). At this time of year, the Arctic region has greater production of oxidants such as OH and has greater dimethyl sulphide (DMS) emissions from the oceanic biological activity, such that oxidation of DMS by OH produces sulphur dioxide (SO_2) and ultimately, sulphuric acid, which contributes to particle formation processes in the boundary layer (e.g. Leaitch et al., 2013). In seasons other than summer, transport of particles arising from NPF outside the Arctic or NPF above the Arctic boundary layer contribute more to the number of particles with diameters smaller than 200 nm. The NONUC simulation coincidentally has lowest bias for the accumulation-mode number in fall and winter at both sites, and for the Aitken mode in fall at both sites, as well as in spring at Alert. Shutting off the NPF process (a source term) in the model appears to compensate for errors in the key sink terms for aerosol number, such as wet removal and coagulation, and related feedbacks. In reality, NPF makes a significant contribution to the number concentration in the Arctic (e.g. Chang et al., 2011; Leaitch et al., 2013). The Arctic is a challenging region that tests the performance of the entire set of model mechanisms. Nevertheless, our results, presented in Figs. 3 and 4, highlight NPF and particle growth, wet removal, and coagulation as key processes for controlling Arctic aerosol size distributions throughout the annual cycle.

Figures 5 and 6 show the annual cycle of the monthly median total number of particles with diameters between 20-500 nm (N20), 80-500 nm (N80), and 200-500 nm (N200) from simulations and from measurements at Alert and Mt. Zeppelin. To assist with interpreting Figs. 5 and 6, Tables 4 and 5 contain the mean fractional bias (MFB) and mean fractional error (MFE) following Boylan and Russell (2006).

$$MFB = \frac{1}{N}\sum_{i=1}^{i=N}\frac{(C_m(i) - C_o(i))}{(C_m(i) + C_o(i))/2}$$

Eq. 7

$$MFE = \frac{1}{N}\sum_{i=1}^{i=N}\frac{|C_m(i) - C_o(i)|}{(C_m(i) + C_o(i))/2}$$

Eq. (8)

where $C_m(i)$ is the ith monthly model value, $C_o(i)$ is the ith monthly measurement value and N is the total number of months in a year.

Figures 5 and 6 demonstrate the key features of the annual cycle of integrated Arctic aerosol number distributions. Measurements from both Alert and Mt. Zeppelin show a shallow maximum in the N20 in both spring and summer. The measurement N80 and N200 have a maximum in March-April at both sites. The minimum for the N20, N80 and N200 from measurements occurs near September-October at both sites. All four simulations capture the general trend of N80 and N200 being higher in spring than in fall, but there are some notable mismatches discussed below.

Similar to our findings in examining the seasonal-mean size distributions (Figs 3 and 4), Figs. 5 and 6 show that the N200 is highly sensitive to the wet removal parameterization. Simulation STD over-predicts the N200 at both Alert and Zeppelin as evidenced by the greatest magnitude of the N200 MFB and MFE among the four simulations at both sites for simulation STD. Wet removal revisions for simulation NEWSCAV reduce the N200 MFB and MFE towards zero, whereas implementation of the new coagulation mechanism has a lesser effect on these N200 biases. NONUC has the closest-to-zero MFB for N200 among the four simulations at both Alert and Zeppelin and also the lowest MFE at Alert. However, the MFE for the N200 is similar between NONUC and NEWSCAV+COAG at both sites. As noted earlier, suppressing particle formation in NONUC likely compensates for errors in sink processes.

The N20 and N80 are sensitive to the wet removal and coagulation schemes. Tables 4 and 5 show that interstitial coagulation (NEWSCAV+COAG relative to NEWSCAV) reduces the MFB and MFE, for N20 and N80 at both Alert and Mt. Zeppelin. However, changes to the wet-removal parameterization increase the N20 and N80 MFB and MFE at both sites for simulation NEWSCAV relative to STD, except for the N80 MFB at Mt. Zeppelin. As discussed in reference to Figs. 3 and 4, NPF increases when the wet removal is more vigorous, and these new particles grow to increase the number of Aitken-mode aerosols in the simulations (i.e. the condensation sink for condensable vapours on to existing aerosols is lower, which favours NPF and growth and reduces losses of new particles by coagulation). At Mt. Zeppelin for the N20 and N80, NONUC has the smallest MFB but NEWSCAV+COAG best represents the annual cycle (smallest

MFE) among the four simulations. At Alert, for the N20 and N80, NEWSCAV+COAG best represents the annual cycle (smallest MFE and MFB).

Figures 5 and 6 also show the annual cycle of aerosol effective diameter at both Alert and Mt. Zeppelin for our simulations and from measurements. The simulation NEWSCAV+COAG has the closest agreement (smallest MFE) with the annual cycle of effective diameter from the measurements for both sites. At Alert, the aerosol effective diameter has the smallest bias for both NEWSCAV and NEWSCAV+COAG, whereas, at Mt. Zeppelin, STD has the smallest bias for the effective diameter due to cancellation of errors between months of over- and under-prediction. The simulations over-predict the aerosol effective diameter in July and August, except for NEWSCAV at Mt. Zeppelin. The over-prediction of summertime effective diameter is pronounced for the simulation NONUC that removes NPF, illustrating the importance of NPF in yielding the summertime minimum effective diameter. The effective diameter in winter at Mt. Zeppelin is strongly underestimated in all simulations, reflecting too many small (Aitken mode) particles, even for simulation NONUC.

The similarity in the annual cycle of effective diameter from measurements at both Alert and Zeppelin suggests a cycle that occurs throughout the Arctic. Figure 7 shows the seasonal-mean pan-Arctic geographic distribution of the surface layer effective diameter for the NEWSCAV+COAG simulation. Throughout the Arctic, the simulated effective diameter declines to a minimum in summer. In Fig. 7, we superimpose the effective diameter from observations at Alert and Mt. Zeppelin. The simulated effective diameter

at the altitude of Mt Zeppelin (500 m) is smaller than the surface value shown here (by 35 nm in summer, 20 nm in fall and 5 nm in winter and spring).

3.3. Process rates controlling the annual cycle in Arctic aerosol number and size

Figure 8 shows the monthly- and regional-mean process rates that control aerosol number in four size ranges for the entire troposphere north of the Arctic Circle (66 °N) for simulation NEWSCAV+COAG. Source processes for aerosol number are positive and sink processes are negative.

The number of aerosols smaller than 10 nm in diameter (nucleation-mode size) is primarily controlled by NPF (particle formation, also termed nucleation), coagulation, and transport. There are two maxima in the particle formation rate shown in Fig. 8 (top-left panel), one in early spring (March) and one in summer (July). In spring, simulated NPF occurs mainly in the free troposphere, whereas in summer, NPF occurs also in the boundary layer. In the summertime Arctic boundary layer, NPF is enhanced by the low aerosol surface area due to efficient wet removal of accumulation-mode aerosols by episodic rain and summer enhancements in sulphuric acid production rates (from oxidation of DMS). 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. This simulated springtime maximum in NPF occurs because the precursors for sulphuric acid (DMS, SO_2) are

transported from open ocean areas and pollution sources at lower latitudes. Then NPF proceeds in locations where the condensation sink for sulphuric acid on existing aerosols is low such as following wet scavenging episodes.

The top-left panel of Fig. 8 shows that transport reaches a maximum during winter, while NPF reaches a minimum such that the two are comparable sources for the entire Arctic troposphere. 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 (SO_2) transported into the Arctic in winter. Our simulated Arctic wintertime sulphuric acid mixing ratio 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.

Figure 9 shows aerosol number transport rates at different altitudes by decomposing the rates from Fig. 8 into four altitude bands. Nucleation-mode particles are mostly transported in the mid to upper troposphere (at altitudes between 4 and 10 km) where the coagulation sink is sufficiently low that nucleation-mode particles can persist. 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. Transport rates for nucleation size-particles are greatest from January to March.

Figure 8 (top-right panel) indicates that several processes control the simulated Aitken-mode number in the Arctic troposphere. Northward transport is the dominant source process for the Arctic Aitken mode during all months of the year. This transport of simulated Aitken-mode aerosols occurs throughout the troposphere as shown in Fig. 9. Figure 8 shows that during the Arctic spring (March-April), when the total aerosol mass is greatest, condensational growth of existing aerosols makes a relatively greater contribution to the total source rates for Aitken-mode particles. This net enhancement in condensational growth includes condensational loss of Aitken-mode particles to accumulation-mode sizes such that the nucleation mode is a larger source of Aitken-mode particles than apparent in the figure. Simulated primary particle emissions within the Arctic have a relatively constant source rate for the Aitken mode throughout the year, quite similar in magnitude to the maximum condensational growth rate in March-April. Coagulation is the dominant sink for the Aitken mode with dry deposition accounting for the majority of the remaining sink. Simulated removal of the Aitken- mode number by wet deposition is a weaker sink than dry deposition because the smaller Aitken-mode aerosols have inefficient removal by activation scavenging (the process of aerosols acting as the seed for cloud-droplet and ice-crystal formation and subsequent removal during precipitation). 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., 2016) and we likely under-estimate this removal in our simulations. Figure 8 does show an increase in wet removal as a sink for the Aitken mode in summer as this process

becomes more efficient at warmer temperatures and aerosols larger than about 60 nm are removed by activation scavenging in our simulations.

For the accumulation-mode particle number simulation, Fig. 8 (bottom-left panel) indicates that the dominant sources are northward transport and condensational growth, which also includes production of sulphate by in-cloud oxidation. These two simulated source terms are roughly equal in magnitude in the Arctic throughout April to October. Northward transport of accumulation-mode aerosols persists in the simulation in all seasons, with a minimum in winter and an increase in March-April. Figure 9 shows that transport of accumulation size aerosol at altitudes between 1.5 km and 4 km reaches a maximum in April, which would contribute to the well-known Arctic haze phenomena. Figure 9 also shows that the majority of simulated accumulation-mode number transport is below 1.5 km. This low-level transport is persistent though diminished throughout the summer; suggesting that the summertime cleanliness of the Arctic near-surface atmosphere relies heavily on the increased efficiency of the removal processes in the lower troposphere during the summer months. Indeed, Fig. 8 shows that wet removal is the dominant accumulation aerosol number sink process in all seasons, but increases in magnitude and relative importance with respect to dry deposition in the summer, accounting for about 90% of the total summertime sink rate. In winter, the relative simulated importance of dry deposition for accumulation aerosol number increases, although remains below 25% of the total sink rate.

Since wet removal has large effects on the accumulation aerosol number associated with Arctic springtime pollution, we further examined its annual cycle. Figure 10 shows the

monthly- and regional-mean accumulation-mode number lifetime with respect to wet removal for layers of the lower troposphere. Longer lifetimes from December to March contribute to the build up of the Arctic haze layer, particularly as this is combined with transport of pollution into the Arctic during wintertime. The spring to summer transition period is characterized by a rapid increase in the efficiency of wet scavenging that contributes to removal of the Arctic haze in April-May. Figure 10 shows about a 5-fold decrease in wet removal lifetime in the Arctic 1.5-4 km layer from February to April. Simulated wet removal lifetimes in the Arctic boundary layer below 1.5 km reach a minimum in October, such that when combined with diminishing new particle formation as the sunsets and limited transport yields the simulated total aerosol number minimum in the fall season, similar to that observed at Alert and Mt. Zeppelin. To put the Arctic region in context, Fig. 10 also shows the lifetimes with respect to wet removal for the region north of 50°N, indicating that wet removal processes are generally more efficient for a region with greater southward extent and at lower altitudes.

Figure 8 shows that the simulated coarse mode is controlled primarily by emissions, transport and wet deposition. In early spring (March-April), northward transport of coarse-mode aerosols (dust and sea-salt emissions) is not quite matched by the removal processes. The resultant residual (black line on Fig. 8) gives the net rate of either aerosol build-up or loss for the regional monthly mean number. In early spring, there is a net build-up of coarse-mode aerosol in the Arctic region. However as spring progresses, there is a net loss such that the net residual integrates to zero over the annual cycle. Wet removal is the primary loss process in all seasons in this simulation. 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.

In this section we examined process rates over the entire troposphere north of 66 °N. To put these Arctic process rates in context with other regions, Fig. 11 shows the same set of processes for the same four aerosol size ranges over the entire troposphere north of 50 °N. Several differences are apparent. For the nucleation, Aitken and accumulation sizes, transport is of negligible importance relative to other source processes, unlike for the Arctic region. Coagulation remains the main sink for the number of nucleation- and Aitken-mode aerosols as shown in Figs. 8 and 11, but the relative importance of wet removal of the Aitken mode in summer has diminished. Wet removal rates for the accumulation-mode aerosol number reach a maximum in May in the Arctic whereas the maximum is in July for the entire region north of 50°N. For the Aitken and accumulation modes, condensational growth is the dominant source north of 50 °N, unlike for the Arctic region only (Fig. 8) where transport was of similar or greater importance. The coarse mode north of 50 °N shows a peak in the transport source in April, similar to Fig. 8, associated with transport of dust from lower latitudes in spring. Coarse-particle number wet removal also shows an April maximum in both Figs. 8 and 11. The global mean simulated number process rates (not shown) show a relative importance of processes similar to that in Fig.11, except in the global mean, primary emissions are the only non-negligible source of coarse aerosol number throughout the year. Wet deposition remains the dominant sink of accumulation and coarse-mode number, followed by dry deposition at the global scale, as in the Arctic.

4. Conclusions

In this study, we examined the annual cycle of aerosol number and size distributions in the Arctic from measurements made during 2011-2013 by scanning mobility particle sizer (SMPS) at Alert and by differential mobility particle sizer (DMPS) at Mt. Zeppelin. There was a strong and similar annual cycle in measurements of aerosol number and size at both sites despite their geographic separation of 1000 km. The annual cycle in the total number of aerosols larger than 20 nm had two maxima. The maximum in spring was dominated by accumulation-mode aerosols (particles 100 nm to 500 nm in diameter) and in summer was dominated by Aitken-mode aerosols (particles 20 nm to 100 nm in diameter). At both sites, total aerosol number reached a minimum in October. The annual cycle of aerosol effective diameter derived from measurements had an inter-seasonal range between 180 nm and 260 nm, with a minimum in the summer. These annual cycles were similar to those presented by Tunved et al. (2013) based on earlier data at Mt. Zeppelin between the years 2000 and 2010.

We interpreted these annual cycles in Arctic aerosol number and size with the GEOS-Chem-TOMAS aerosol microphysics model. Our simulations indicated a strong sensitivity of the annual cycle of Arctic aerosol number and size to several key processes; new-particle formation (NPF), interstitial coagulation scavenging in clouds, wet removal through precipitation, and transport.

Our GEOS-Chem-TOMAS simulations demonstrated that wet removal had a strong control on Arctic aerosol number distributions throughout the annual cycle, similar to the findings of earlier studies focused on spring-summer (Korhonen et al, 2008) and Arctic aerosol mass abundance (e.g. Garrett et al., 2010; Browse et al., 2012; Sharma et al., 2013). In our study, wet removal updates were developed for the GEOS-Chem-TOMAS model that together increased the efficiency of wet removal. We replaced the global-constant cloud liquid water content with the values from GEOS-5 assimilated meteorology fields, updated the gridbox precipitation fraction, and implemented the Verheggen et al. (2007) temperature-dependent aerosol activation fraction to account for the fraction of aerosol assumed to be susceptible to wet removal in mixed-phase clouds. In our updated-removal simulation, efficient wet removal in the Arctic summertime boundary layer strongly limited the accumulation-mode number despite an ongoing source through transport and condensational growth. The wet removal updates reduced model-measurement bias (relative to the standard model) for the number of aerosols larger than 200 nm in all seasons at both Alert and Mt. Zeppelin (although the changes in winter and spring at Alert were relatively small).

More vigorous wet removal promoted NPF and growth in our simulations and contributed to a summertime dominant Aitken mode since a reduction in the surface area of accumulation size aerosols (the condensation sink for sulphuric acid) influences the likelihood that sulphuric acid will participate in NPF as opposed to condensing on existing aerosols. Indeed, the more vigorous wet removal scheme increased the simulated Aitken-mode number in all seasons at Alert and Mt. Zeppelin (although the springtime

Aitken mode was relatively less sensitive to the changes made in our study). Outside of summer, NPF and growth occurred mostly outside the Arctic boundary layer. A sensitivity study with no NPF globally indicated that NPF strongly controls the number of particles with diameters smaller than 200 nm in all seasons in the Arctic, while particularly important in yielding the summertime Aitken-mode dominance.

From February to April, the simulated accumulation-mode wet removal efficiency at altitudes of the springtime Arctic haze layer (between 1.5 and 4 km) increased by 5-fold, contributing to our simulation of the spring-summer transition from Aitken- to accumulated-mode dominated Arctic size distributions (e.g. Engvall et al., 2008; Korhonen et al., 2008). In the boundary layer, simulated wet removal efficiency reached a maximum (lowest accumulation-mode aerosol number lifetime) in October. The observed total aerosol number minimum in October was reproduced in our simulations due to efficient wet removal combined with diminished boundary layer NPF due to lower sulphuric acid concentrations and limited transport.

We also found an important role for coagulation of interstitial aerosols in clouds with aerosols of larger size that have activated to form cloud droplets. There has been relatively less attention given to the importance of this process in controlling Arctic size distributions despite the Arctic being a region with widespread cloud cover in all seasons. Implementation of an interstitial coagulation mechanism in clouds in our simulations reduced the number of aerosols with diameters smaller than 200 nm in all seasons at both Alert and Mt. Zeppelin. In some seasons this reduction in the Aitken-mode number worsened model-measurement agreement, highlighting the delicate balance between the

processes of coagulation, NPF, growth and wet removal in control of the Arctic size distributions that is challenging to simulate. Our simulations tended to under predict the number of larger Aitken-mode aerosols (40-100 nm in diameter) in summer and this is the subject of ongoing investigation related to aerosol sources and growth.

The high sensitivity of aerosol number to interstitial coagulation in clouds suggests that size-resolved models should include this process. However, many present-day global models neglect this process, including previous versions of GEOS-Chem-TOMAS (D'Andrea et al., 2013; Pierce et al., 2013; Trivitayanurak et al., 2008), GISS-TOMAS (Adams and Seinfeld, 2002; Pierce and Adams, 2009), GLOMAP (Spracklen et al., 2005a,b; 2008, Mann et al., 2012), GLOMAP-Mode (Mann et al., 2010; 2012, Lee et al., 2013b), GEOS-Chem-APM (Yu and Luo, 2009; Yu, 2011) and IMPACT (Herzog et al., 2004; Wang and Penner, 2009). To our knowledge, only a few models such as MIRAGE and ECHAM-HAM (Herzog et al., 2004; Ghan et al., 2006; Hoose et al., 2008) represent this process.

Our results highlight the importance of aerosol processes (as well as their delicate balance and interactions) that continue to be poorly understood; 1) new-particle formation (NPF) and growth, 2) in-cloud interstitial coagulation, and 3) wet removal as playing a key role in the control of the annual cycle of aerosol number and size in the Arctic. The relative importance of the processes that control aerosol number could change in a future warming Arctic climate and also as emissions within the Arctic change.

Acknowledgements:

The authors acknowledge the financial support provided for NETCARE through the Climate Change and Atmospheric Research Program at NSERC Canada. Thanks to Sangeeta Sharma, Desiree Toom, Andrew Platt and the Alert operators for supporting the Alert observations. We are also grateful to Ilona Riipinen, Jan Julin and Tinya Olenius for helpful discussions and for providing the Atmospheric Cluster Dynamics Code (ACDC), applied in our GEOS-Chem-TOMAS simulations.

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Figure Captions:

Figure 1: Measured monthly median number distributions from the scanning mobility particle sizer (SMPS) at Alert for 2011-2013 and the differential mobility particle sizer (DMPS) at Mt. Zeppelin for 2011-2013 for particle sizes between 20 nm and 500 nm. Error bars show the 20-80th percentile of the measurements.

Figure 2: Measurement monthly median aerosol effective diameter from SMPS and DMPS at the two high-Arctic sites, Alert (2011-2013) and Mt. Zeppelin (2011-2013), respectively, for particle sizes between 20 nm and 500 nm. Error bars show the 20th and 80th percentiles.

Figure 3: Seasonal median number distributions from SMPS measurements at Alert (2011-2013) and for the GEOS-Chem-TOMAS dry size distribution simulations (described in Table 1). The measurement 20-80th percentile is in grey shading. Simulations are shown in color as indicated by legend.

Figure 4: Seasonal median number distributions from DMPS measurements at Mt. Zeppelin (2011-2013) and for the GEOS-Chem-TOMAS dry size distribution simulations

(described in Table 1). The measurement 20-80th percentile is in grey shading. Simulations are shown in color as indicated by legend.

Figure 5: Monthly median number concentration for aerosols with diameters of 20-500 nm (N20), 80-500 nm (N80), and 200-500 nm (N200), and effective diameter from the 2011-2013 Alert SMPS measurements and for the four GEOS-Chem-TOMAS dry size distribution simulations described in Table 1. The measurement 20-80th percentile is in grey shading. Simulations are shown in color as indicated by legend.

Figure 6: Monthly median number concentration for aerosols with diameters of 20-500 nm (N20), 80-500 nm (N80), and 200-500 nm (N200), and effective diameter from the 2011-2013 Mt. Zeppelin DMPS measurements and for the four GEOS-Chem-TOMAS dry size distribution simulations described in Table 1. The measurement 20-80th percentile is in grey shading. Simulations are shown in color as indicated by legend.

Figure 7: Geographic distribution of the simulated pan-Arctic surface-layer seasonal-mean dry effective diameter [nm] for the NEWSCAV+COAG simulation. The coloured stars indicate the effective diameter from measurements at Alert (SMPS) and Mt. Zeppelin (DMPS).

Figure 8: Monthly and Arctic mean aerosol number process rates for the entire Arctic troposphere (north of 66°N) for simulation NEWSCAV+COAG. Processes considered for each of four size ranges are condensation, coagulation, particle formation, primary

emissions, wet and dry deposition, transport across 66 °N and net regional buildup or loss rates. The aerosol size ranges are nucleation (D_p<10 nm), Aitken (10<D_p<100 nm), accumulation (100<D_p<1000 nm), and coarse (D_p>1000 nm).

Figure 9: Monthly and Arctic mean aerosol number tendency due to transport within each of four vertical layers between 1) 0-1.5 km, 2) 1.5-4 km, 3) 4-10 km, and 4) above 10 km for the simulation NEWSCAV+COAG for the entire troposphere north of 66°N. Summation of the 4 layers for any given month and size range yields the transport tendency shown in Fig.8. Positive values indicate a net northward transport into the Arctic.

Figure 10: Regional and monthly mean aerosol number lifetime with respect to wet deposition for accumulation-mode aerosol number (100<D_p<1000 nm) nd in the altitude bands of 0-1.5 km, and 1.5-4 km for the GEOS-Chem simulation NEWSCAV+COAG.

Figure 11: Monthly- and regional-mean aerosol number process rates for the entire troposphere north of 50°N for simulation NEWSCAV+COAG. Processes considered for each of four size ranges are nucleation, emissions, coagulation, condensation, wet and dry deposition, transport across 66°N and net regional accumulation or loss rates. The aerosol size ranges are nucleation (D_p<10 nm), Aitken (10<D_p<100 nm), accumulation (100<D_p<1000 nm), and coarse (D_p>1000 nm).

Tables:

Table 1: Summary of the simulations conducted for this study.

Simulation Name	Revised Wet Removal	With Interstitial Coagulation	With New-Particle Formation
STD	no	no	yes
NEWSCAV	yes	no	yes
NEWSCAV+COAG	yes	yes	yes
NONUC	yes	no	no

Table 2: Model-measurement fractional bias (Eq. 6) for total number of aerosols with diameters of 20-100 nm and 100-500 nm at Alert (in reference to Fig. 3). Bias values closest to zero for each season are highlighted in red.

Bias	STD	NEWSCAV	NEWSCAV+COAG	NONUC
20-100 nm				
Winter	1.95	3.45	**0.18**	1.47
Spring	0.83	1.12	-0.46	**-0.36**
Summer	-0.58	0.56	**0.23**	-0.92
Fall	0.15	3.53	0.52	**0.07**
100-500nm				
Winter	0.66	0.87	0.40	**0.34**
Spring	0.38	0.30	**-0.01**	-0.40
Summer	0.98	0.21	**0.05**	-0.43
Fall	0.40	1.34	0.78	**0.01**

Table 3: Model-measurement fractional bias (Eq. 6) for total number of aerosols with diameters of 20-100 nm and 100-500 nm at Mt. Zeppelin (in reference to Fig. 4). Bias values closest to zero for each season are highlighted in red.

Bias	STD	NEWSCAV	NEWSCAV+COAG	NONUC
20-100 nm				
Winter	6.73	12.87	**3.17**	5.43
Spring	0.68	1.01	**-0.40**	-0.43
Summer	-0.65	**-0.21**	-0.54	-0.90
Fall	0.34	4.59	1.14	**0.10**
100-500nm				
Winter	3.24	3.42	2.18	**2.09**
Spring	0.96	0.49	**0.19**	-0.22
Summer	0.60	**0.02**	-0.15	-0.61
Fall	1.50	1.63	0.99	**0.12**

Table 4: Model-measurement mean fractional bias and mean fractional error (Eqs. 7 and 8) for N20, N80, N200 and effective diameter at Alert (in reference to Fig. 5). Bias and error values closest to zero for each season are highlighted in red.

	STD	NEWSCAV	NEWSCAV+COAG	NONUC
MFB				
N20	0.22	0.57	**0.06**	-0.53
N80	0.24	0.36	**0.05**	-0.43
N200	0.74	0.24	0.27	**0.17**
Eff. Diam.	0.17	**-0.05**	**0.05**	0.21
MFE				
N20	0.45	0.57	**0.23**	0.80
N80	0.32	0.37	**0.23**	0.60
N200	0.74	0.30	0.30	**0.29**
Eff. Diam.	0.20	0.10	**0.08**	0.22

Table 5: Model-measurement mean fractional bias and mean fractional error (Eqs. 7 and 8) for N20, N80, N200 and effective diameter at Mt. Zeppelin (in reference to Fig. 6). Bias and error values closest to zero for each season are highlighted in red.

	STD	NEWSCAV	NEWSCAV+COAG	NONUC
MFB				
N20	0.46	0.66	0.21	**-0.18**
N80	0.65	0.57	0.31	**-0.11**
N200	0.86	0.20	0.22	**0.12**
Eff. Diam.	**0.04**	-0.17	-0.07	0.06
MFE				
N20	0.76	0.86	**0.75**	1.03
N80	**0.68**	0.77	**0.68**	0.88
N200	0.86	0.66	**0.54**	0.56
Eff. Diam.	0.12	0.17	**0.10**	0.17

Figures:

Figure 1: Measured monthly median number distributions from the scanning mobility particle sizer (SMPS) at Alert for 2011-2013 and the differential mobility particle sizer (DMPS) at Mt. Zeppelin for 2011-2013 for particle sizes between 20 nm and 500 nm. Error bars show the 20-80th percentile of the measurements.

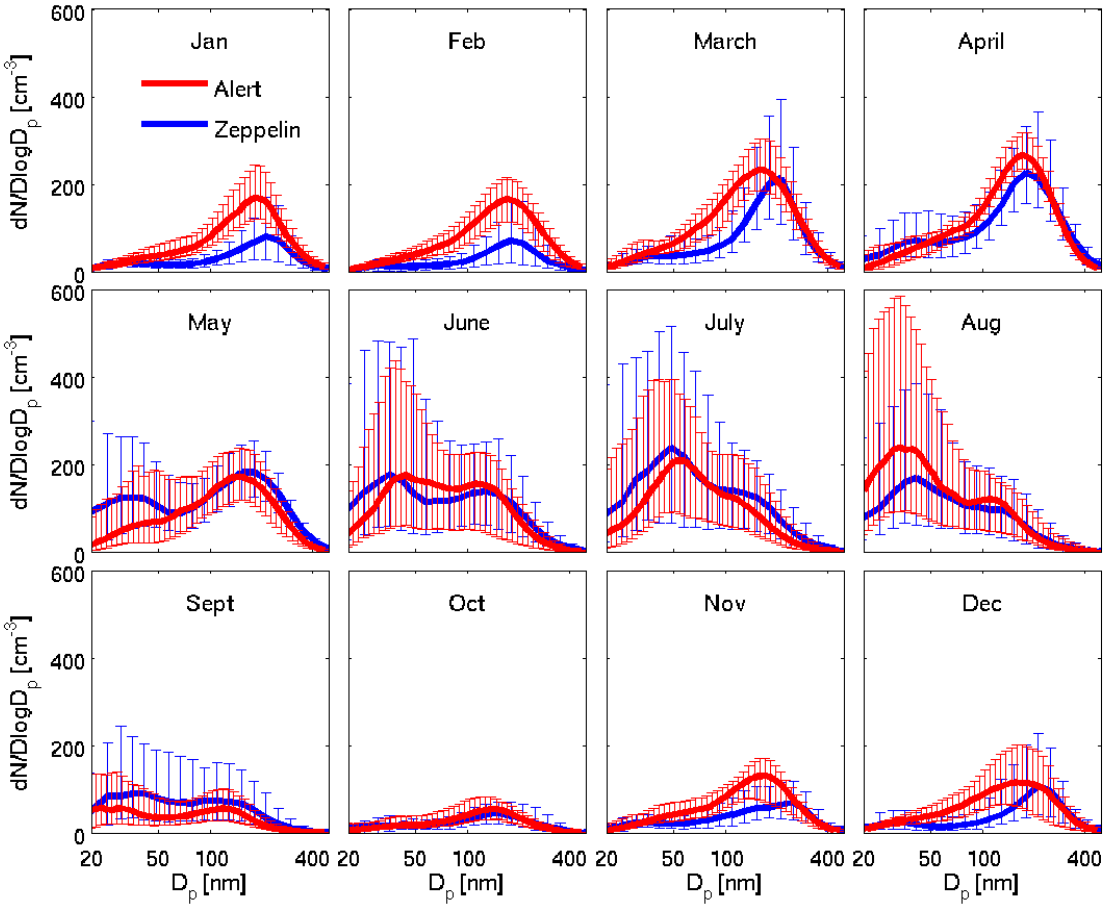


Figure 2: Measurement monthly median aerosol effective diameter from SMPS and DMPS at the two high-Arctic sites, Alert (2011-2013) and Mt. Zeppelin (2011-2013), respectively, for particle sizes between 20 nm and 500 nm. Error bars show the 20th and 80th percentiles.

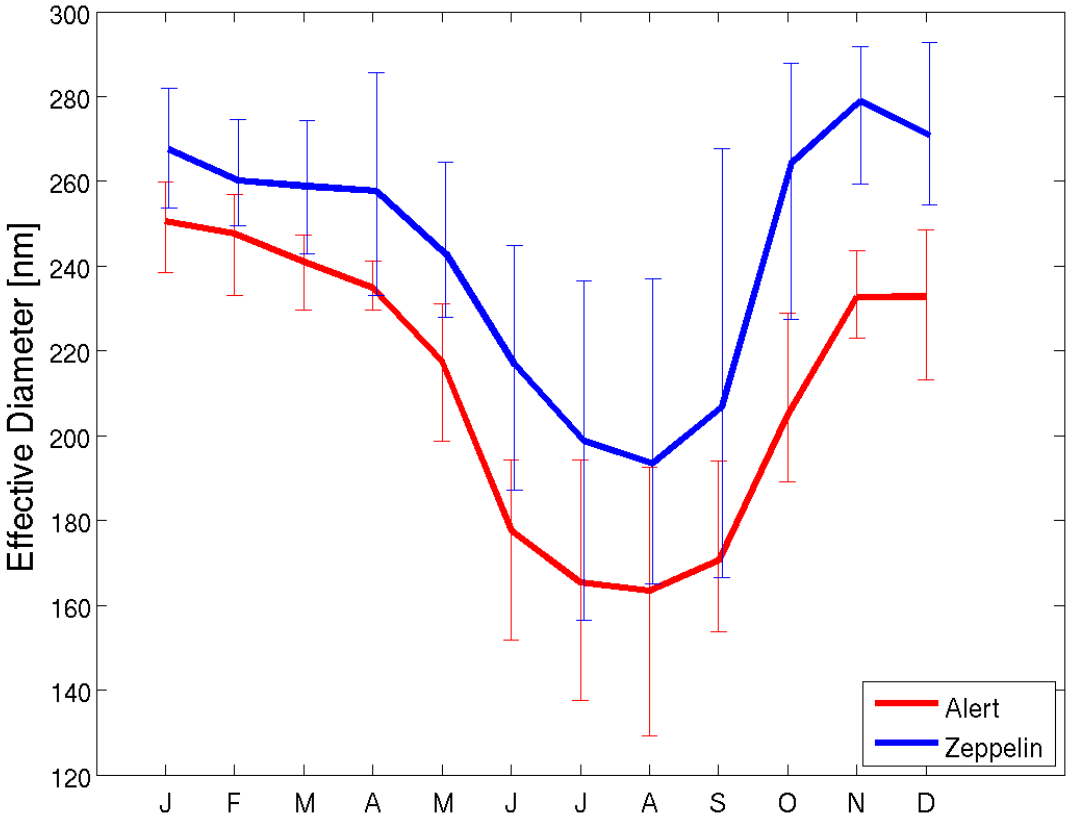
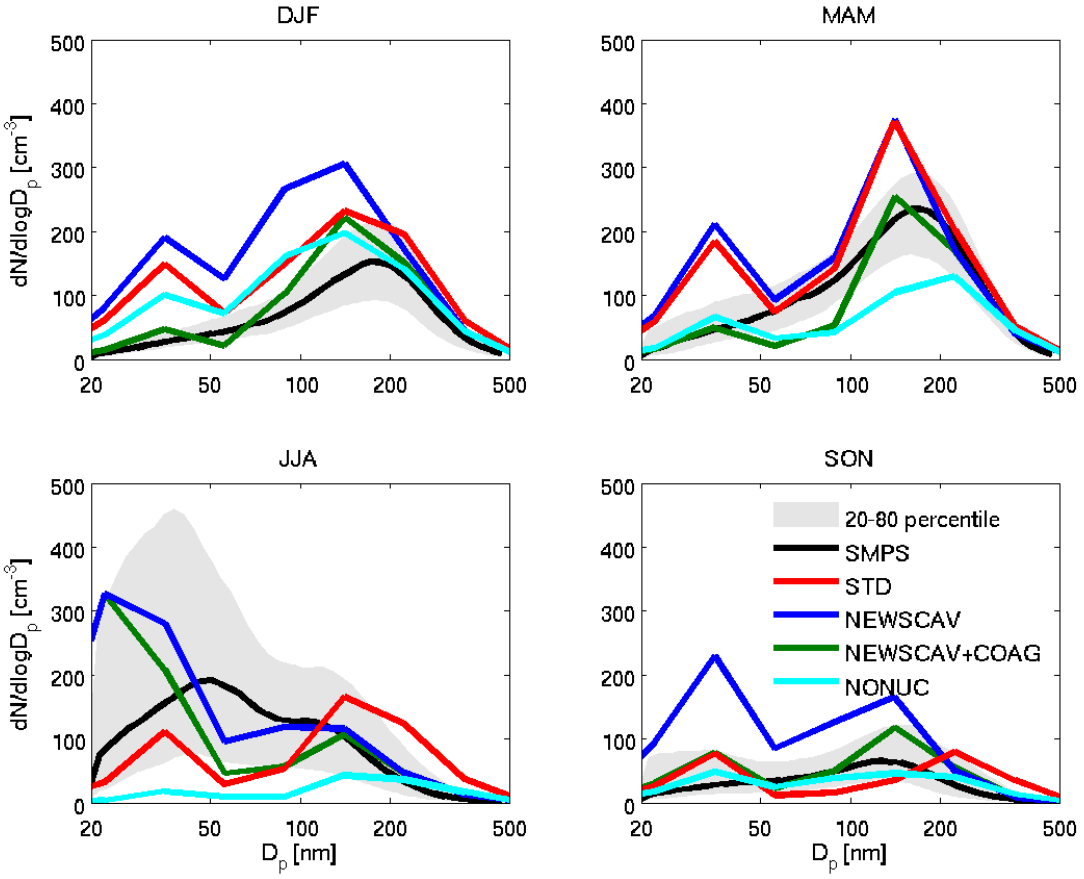


Figure 3: Seasonal median number distributions from SMPS measurements at Alert (2011-2013) and for the GEOS-Chem-TOMAS dry size distribution simulations (described in Table 1). The measurement 20-80th percentile is in grey shading. Simulations are shown in color as indicated by legend.



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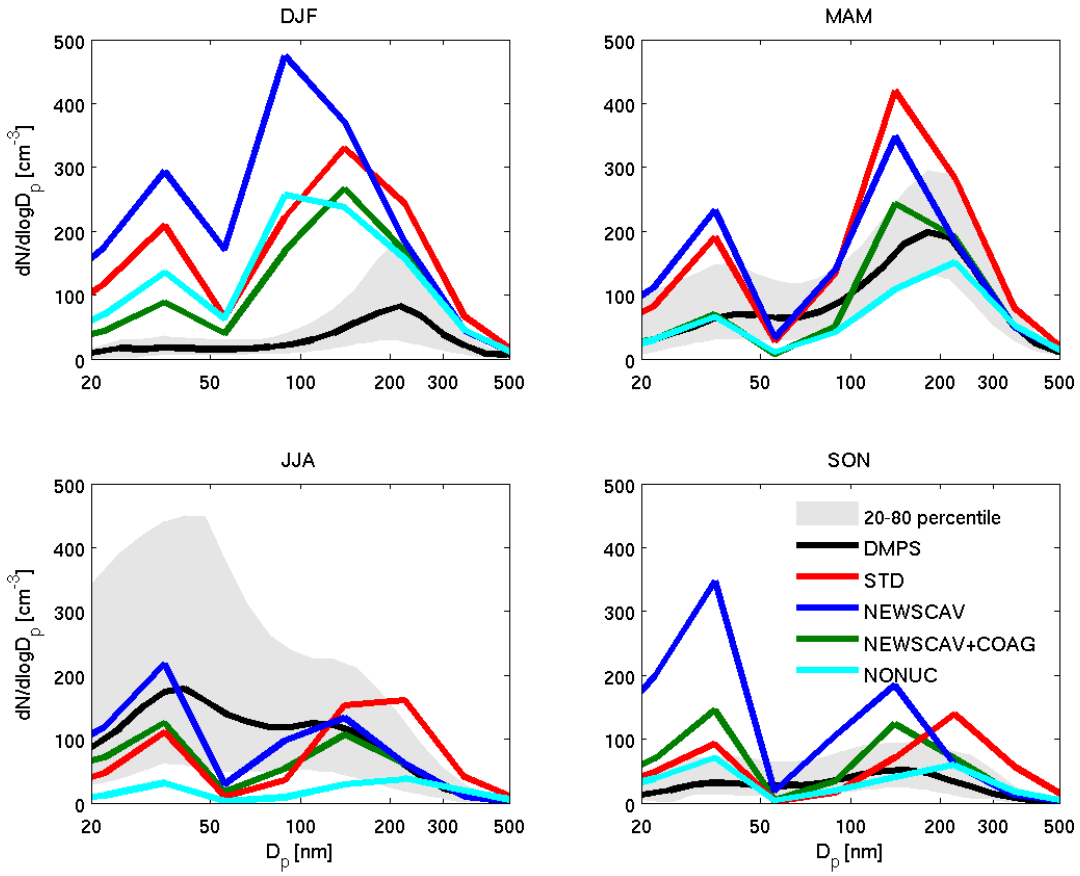

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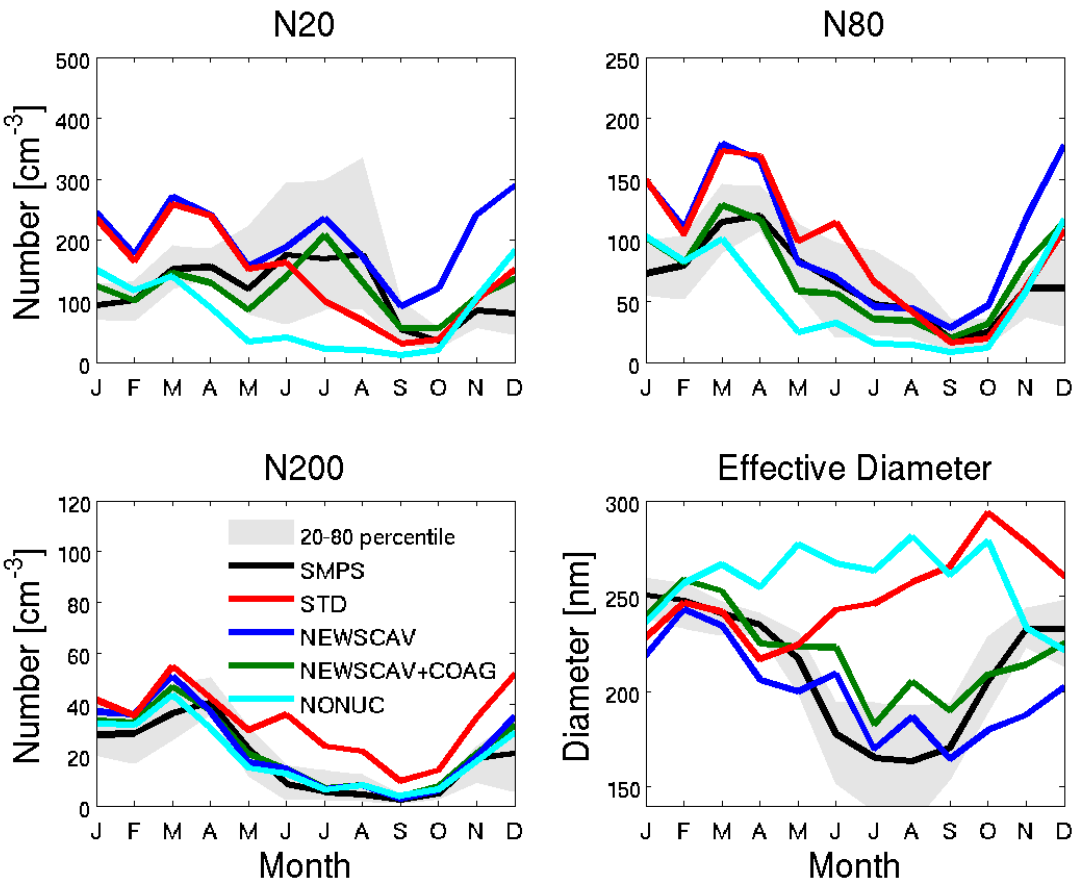

Figure 6: Monthly median number concentration for aerosols with diameters of 20-500 nm (N20), 80-500 nm (N80), and 200-500 nm (N200), and effective diameter from the 2011-2013 Mt. Zeppelin DMPS measurements and for the four GEOS-Chem-TOMAS dry size distribution simulations described in Table 1. The measurement 20-80th percentile is in grey shading. Simulations are shown in color as indicated by legend.

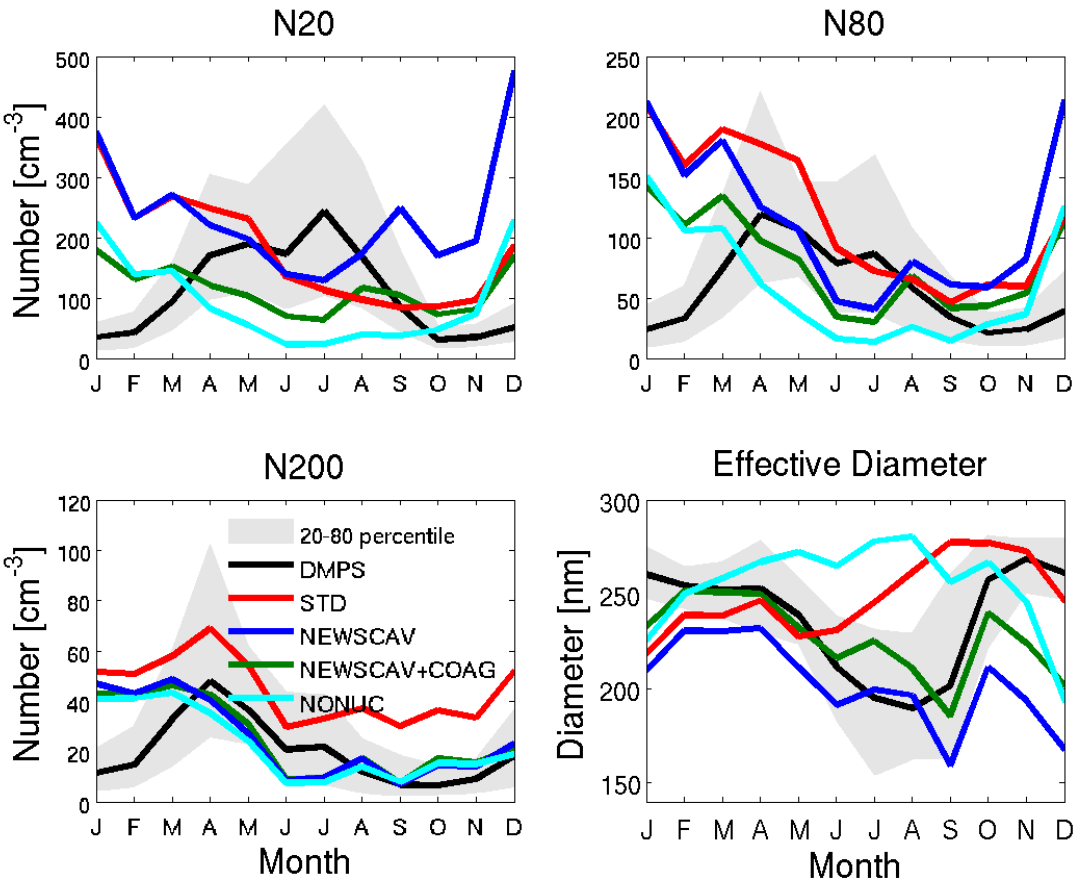

Figure 7: Geographic distribution of the simulated pan-Arctic surface-layer seasonal-mean dry effective diameter [nm] for the NEWSCAV+COAG simulation. The colored stars indicate the effective diameter from measurements at Alert (SMPS) and Mt. Zeppelin (DMPS).

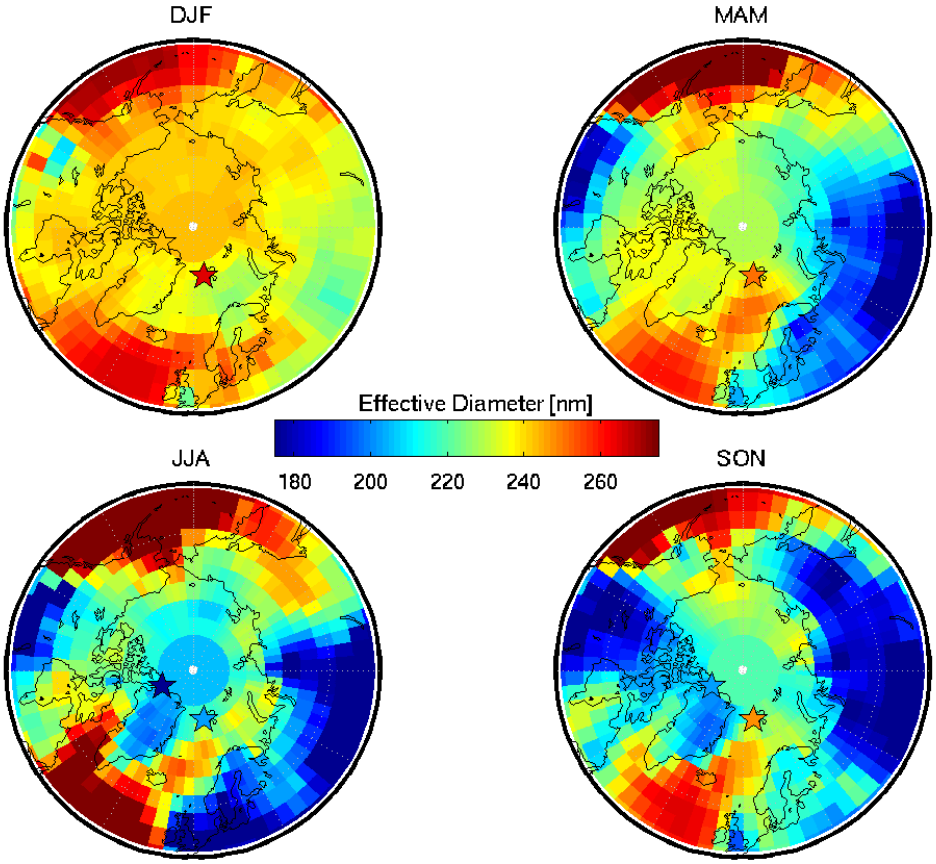

Figure 8: Monthly and Arctic mean aerosol number process rates for the entire Arctic troposphere (north of 66°N) for simulation NEWSCAV+COAG. Processes considered for each of four size ranges are condensation, coagulation, particle formation, primary emissions, wet and dry deposition, transport across 66 °N and net regional buildup or loss rates. The aerosol size ranges are nucleation (D_p<10 nm), Aitken (10<D_p<100 nm), accumulation (100<D_p<1000 nm), and coarse (D_p>1000 nm).

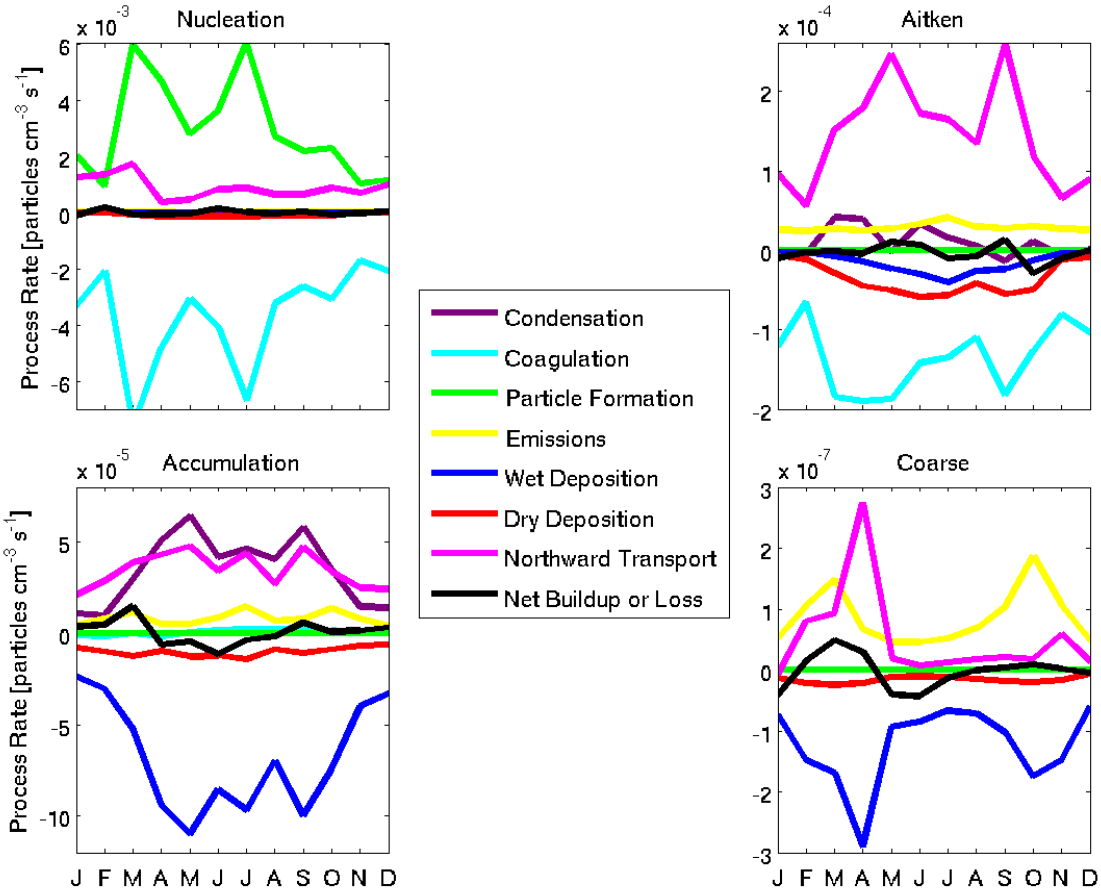

Figure 9: Monthly and Arctic mean aerosol number tendency due to transport within each of four vertical layers between 1) 0-1.5 km, 2) 1.5-4 km, 3) 4-10 km, and 4) above 10 km for the simulation NEWSCAV+COAG for the entire troposphere north of 66°N. Summation of the 4 layers for any given month and size range yields the transport tendency shown in Fig.8. Positive values indicate a net northward transport into the Arctic.

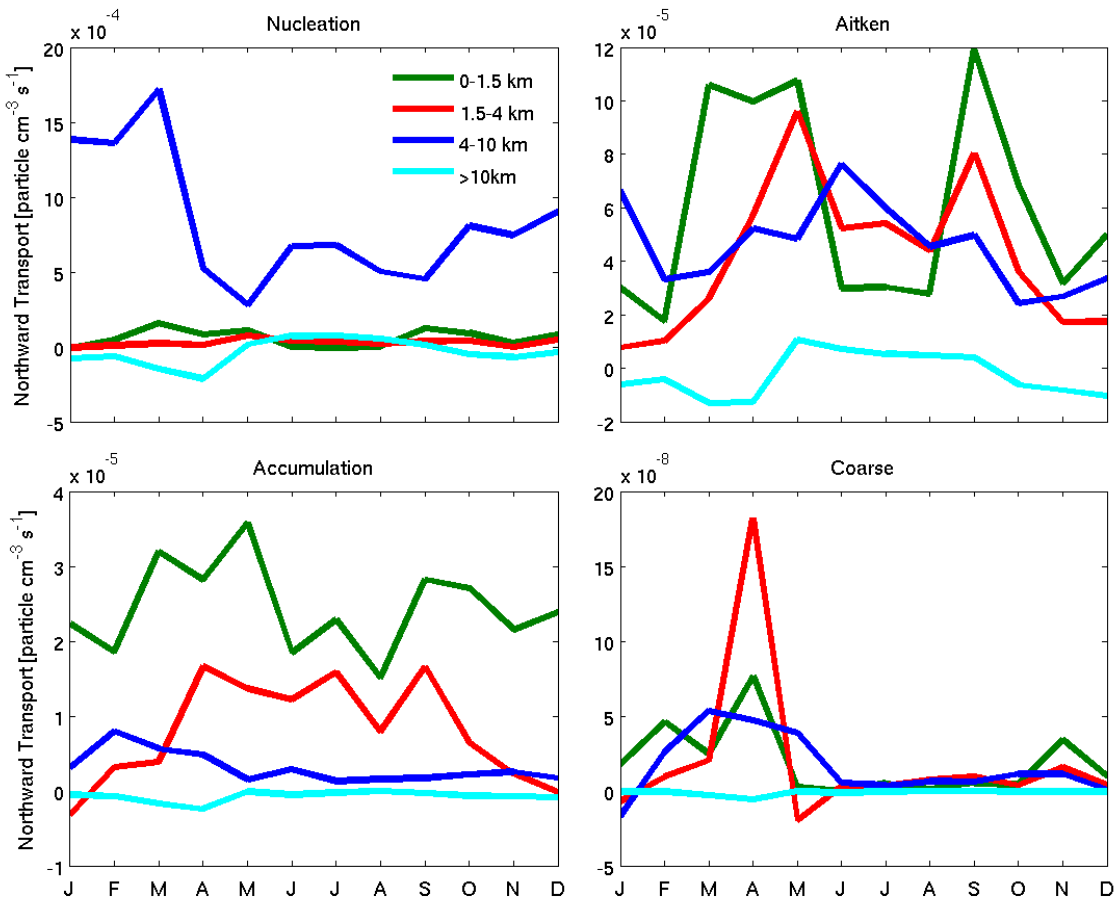

Figure 10: Regional and monthly mean aerosol number lifetime with respect to wet deposition for accumulation-mode aerosol number ($100<D_p<1000$ nm) in the altitude bands of 0-1.5 km, and 1.5-4 km for the GEOS-Chem simulation NEWSCAV+COAG.

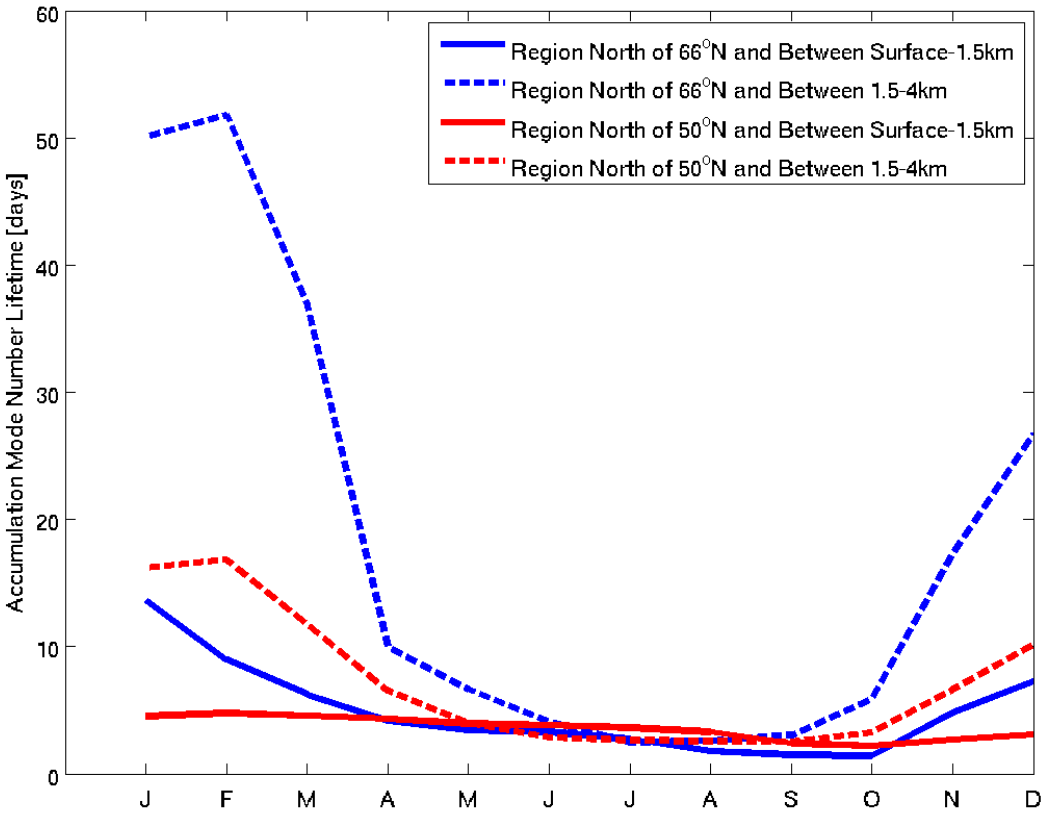

Figure 11: Monthly and Arctic mean aerosol number process rates for the entire troposphere north of 50°N for simulation NEWSCAV+COAG. Processes considered for each of four size ranges are nucleation, emissions, coagulation, condensation, wet and dry deposition, transport across 66°N and net regional accumulation or loss rates. The aerosol size ranges are nucleation (D_p<10 nm), Aitken (10<D_p<100 nm), accumulation (100<D_p<1000 nm), and coarse (D_p>1000 nm).