Atmos. Chem. Phys. Discuss., 15, C9453–C9458, 2015
www.atmos-chem-phys-discuss.net/15/C9453/2015/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

Received and published: 18 November 2015

The manuscript uses size distribution measurements from two Arctic sites together with a state-of-the-science global aerosol model to investigate the processes that control seasonal cycle of the Arctic aerosol properties. While the topic of the study is interesting and the observational and modelling methodology used of very good quality, the analysis and interpretation of the results are strongly biased and at times simply wrong. The main problem is that the results are in many places misrepresented in order to play up one of the simulations (NEWSCAV+COAG), while frequently neglecting the aspects where the other simulations perform well. I agree that NEWSCAV+COAG performs quite well, but it is not even close to being as superior compared to the other runs as the authors make it sound.

Regarding my recommendation, I'm torn between rejection and major revisions. While

the sloppy and biased analysis of the results shows poor scientific conduct, the actual science behind the partly false conclusions is in fact quite good. The study can become publishable without new simulations, but I want to stress that this requires a careful reanalysis of the data and completely rewriting the results section so that it truthfully reflects the data. Simply rewording the specific sentences that are highlighted below will not be enough!!! I'm simply giving examples of the most evident biased statements, but the discussion as a whole needs to become much more balanced before publication.

Specific comments:

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

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?

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.

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* overestimate particle number below 30 nm (actually 40 nm for NEWSCAV) at Alert.

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

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.

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)."

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.

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

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.

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.

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

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. 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?

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.

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.

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

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.

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 process 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.

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

20. P29094, L16-: "The 3-fold wintertime overprediction —" Which simulation does this refer to?

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

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.

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?

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.

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.

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.

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

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 > 10^5 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).

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.

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

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

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 29079, 2015.