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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of CCN activity of Arctic aerosol and Canadian biomass burning during summer 2008" *by* T. L. Lathem et al.

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Responses to Reviewer #1

We thank the reviewer for the helpful and detailed comments which have served to improve the manuscript. Italicized responses to the issues raised are shown below.

General comments

1) The authors find best agreement in CCN closure studies if they assume an external mixture of organics and inorganics. This assumption does not have any physical basis, in particular in aged air masses, and thus one obtains 'the right answer for the wrong reasons'. While they discuss some other reasons in the text, in the abstract only the preference of an external mixture is mentioned. I suggest a more careful statement of

the preference of an external mixture in choosing mixing state and rather stating it as a coincidental agreement for such aged air masses.

We have changed the language in the abstract to be consistent with the rest of the text, by stating that the available observations could not determine the true degree of external mixing and therefore may not always be representative of the environments sampled.

2) Does your finding that 'fresh' and 'aged' biomass burning air masses have the same kappa and the discussion in the text of some previous lab and chamber studies suggest that in the atmosphere there is basically no 'fresh' biomass burning plume since everything is already aged after such short time scales (distances) that they are rarely detected? – Could you put an approximately number on such time scales/distances? Such number would be of high value as input to models where possibly only one (a few) grid box(es) might need to consider fresh biomass burning.

The data (in this study and other cited ARCTAS studies, p. 24707) does indicate that the aging of biomass burning plumes is rapid, on the timescale of 1-2 hours. The distance (and number of model grid boxes) from point of origin that this corresponds to depends on transport; we hence feel that citing the aging timescale is most general and informative.

p. 24680, l. 3: add the percentage value of the bias in CCN number that is caused by a bias in kappa of 50%.

Good point! The bias in CCN is up to 25%, which has been added to the text.

p. 24683, l. 21: Why is N(CCN) more sensitive to composition in the Arctic than in other areas?

This is a good question and is discussed by Moore et al. (2012) in detail. Briefly, the maximum supersaturation in Arctic stratus/stratocumulus (∼0.1%) is around the point of maximum sensitivity in the CCN spectrum (i.e., dCCN/ds). This does not seem to

occur in other locations.

p. 24684, l. 10ff: Do all these studies use kappa(org) as the only 'fitting variable' to achieve ideal closure? How about other parameters (mixing state, size-resolved composition) that could also contribute to the goodness of closure? I suggest putting the text somewhat more careful here along the lines that it is known that using kappa(org) does not to represent the true aerosol composition distribution but is simply used as a fitting parameter that does not reflect the true ambient aerosol properties.

Within a given closure scenario (pre-determined mixing state and composition assumption), kappa(org) is the only fitting parameter that is varied, and the text has been clarified to state that kappa(org) is a fitting parameter. One can choose to perform multiple additional closure scenarios (variations of mixing state and composition) to determine the impact of those assumptions on predictability and identify which combination of assumptions provides the best closure.

p. 24684, l. 29: How does the epsilon(org) defined here differ from the epsilon(WSOM) as defined e.g. in the abstract?

They are the same; epsilon(org) has been changed to epsilon(WSOM)

p. 24685, l. 27: Add some references for global climate models that use such detailed composition information (aerosol composition, organic fraction etc.).

The following references have been added: Mann et al., (2010), Bauer et al., (2010), Meskhidze et al., (2011), Zhang et al., (2012).

p. 24687, l. 4 ff: Was the AMS not on board of the DC-8?

Yes, the AMS was on board the DC-8. Please refer to section 2.3 for details.

p. 24687, l. 27: Can you quantify 'not important' here? – Bias of x%?

Thank you for pointing this out. The bias from calibrations was determined to be less than 8%.

p. 24690, l. 26 and following line: Remove 'cloud' since 'cloud drop' formation implies competition for water vapor under ambient conditions which is not the case in the CCNC.

Done.

p. 24692, l. 4: The CCN number concentration as determined in the CCNC gives the maximum number of particles that might be activated in 'cloudy air parcels' where time and water vapor limitations exist. Reword your sentence accordingly.

This statement is a general statement referring to Köhler theory itself, not specifically to the CCNC

p. 24693, l. 5: Kohler theory describes sc for an individual particle, thus mixing state is not considered at all in Eq. (1)-(3). Do you want to say that 'for internally mixed aerosol populations kappa is the same for all particles' (or similar)? – Reword accordingly.

This is a good point; the text has been clarified as suggested.

p. 24695, l. 12: what was the minimum NCCN ($<$ 100 cm-3)? I assume it was not zero.

The lowest CCN concentration levels were constrained within 50-100 cm^{-3}, to provide adequate counting statistics, while also minimizing supersaturation depletion to provide an unbiased supersaturation calibration. This concentration range has been added to the text.

p. 24696, l. 16: add 'aerosol' after 'biomass burning'

Done.

p. 24697, Sect 4.1: Refer here to Fig. 2

Done.

p. 24698, l. 9/10: Be more specific. 'Significant impacts' on what?

The text has been clarified to suggest significant impacts on the aerosol direct/indirect

radiative forcing in the Arctic environment.

p. 24698, l. 20: why do heterogeneous processes decrease the activated fraction? Such processes usually lead to larger particles by adding more hygroscopic (e.g. sulfate) mass.

Not all aging processes may lead to more hygroscopic particles. Formation of oligomers, breaking of surface active films with oxidation, etc. have been shown to lead to decreases in particle hygroscopicity.

p. 24699, l. 6: 'Reduction' is ambiguous here (might be read as opposite to oxidation).

Better 'decrease'.

Done.

Sect. 4.1.3. Are you saying that these air masses include a significant fraction of boreal/background aerosol? Is your main point that industrial aerosol comprises the mode at $\sim$ 52 nm that dominates the particle NUMBER (p. 24700, l. 19) whereas most of the MASS is from background aerosol (_142 nm)?

Yes, we are suggesting the number is dominated by industrial particles, but the majority of the mass (as detected by the AMS) is dominated by the background aerosol.

p. 24701, l. 9: Do you mean the 'bulk composition of the total aerosol population' by 'aerosol composition' here?

Yes; wording has been changed as suggested.

p. 24701, l. 15 ff: The large variability in kappa is not surprising given the large variability in the inorganic fraction. How large is the variability in the inferred kappa(org) between different studies? Can this be related to different air masses, seasons, etc.?

The wide range of kappa(org) in the Arctic is discussed in the introductory text (p. 24684, l. 11-20) with previous studies suggesting a kappa(org) varies from 0.02 to 0.11.

More data is certainly needed to relate changes in kappa(org) to differing seasons, locations, etc., specifically data with complementary information on the composition of the aerosol and degree of oxygenation, as these are variables which also lead to variations in kappa(org).

p. 24704, l. 3: It might be helpful to repeat here the conversion factor OM/OC and some discussion that assuming different ratios does not impact the results significantly even though it is likely that OM/OC increases with ageing.

Having already been discussed in earlier sections, we do not feel a repeat of the information is necessary here. A reference however to the earlier material will be made here to remind the reviewer of the relevant discussion.

p. 24704, l. 27: The assumption of kappa(org) = 0 is clearly not appropriate. However, I expect that assuming kappa(org) = 0.1 or 0.2 still gives reasonable closure results. Thus, how 'correctly' does kappa have to be determined? You might want to soften the expression 'correctly determining' here.

The text has been clarified to suggest that the WSOM fraction is important for improving predictions of the CCN activity of Arctic aerosol, rather than 'correctly determining'.

p. 24707, l. 14: What is meant by 'net aging'?

We have removed "net" and changed to "aerosol"

p. 24707, l. 16ff: An increase in O/C with constant kappa might have several reasons: (i) Organic material becomes successively oxidized to volatile material (e.g. CO2, HCHO etc) and the fraction of highly oxygenated aerosol mass remains roughly constant which might be reflected by a constant organic mass or even decrease. (ii) The oxidation pathways in the organic aerosol fraction result in compounds that are not very hygroscopic (e.g. organic nitrates, sulfates that should differ from their inorganic counterparts in the AMS spectrum). Do you have any evidence for either of these possibilities?

The reviewer raises an interesting point. While the composition may be changing in a way that O/C alone doesn't capture, additional (simultaneous) aerosol composition measurements are required to discern this (and not available for the dataset here). For example, organosulfates cannot be distinguished from inorganic sulfate in the AMS (Farmer et al., PNAS, 2010); organonitrates, while producing different spectral patterns in the AMS than inorganic nitrate, seem to hydrolyze in the aerosol resulting in typically low concentrations (Browne et al., Atmos. Chem. Phys. Discuss., 13, 201-254). We will make note of the above in the revised manuscript.

p. 24709, l. 20: Better: 'Neglecting the hygroscopicity of the water-soluble organics: : :' (As it is written now, it might imply that you also neglect their mass/volume in the aerosol sizes.)

Good point. Done.

Table 2: Use same order of 'aces' as in text (switch (1) and (2))

Done.

Figure 3: Increase the font for numbers in the boxes.

Done.

Figure 6: Why are the results for other two cases (internally mixed, in/soluble organics) not shown here?

For clarity, the CCN closure plot of Fig. 6 is used to show the results for the best closure scenario studied. For an overview of all cases, we refer the reader to Table 2.

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