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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.

Anonymous Referee #1

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The study by Lathem et al. presents a data analysis of CCN data in the Arctic during the ARCTAS experiment. The authors perform CCN closure studies using different assumptions (solubility of organics, mixing state and find reasonable agreement for the different air masses explored here (biomass burning, boreal forest, Arctic background, industrial). This study represents one in many studies that address the role of chemical composition and mixing state in CCN properties for different environments. Since the Arctic is particular prone to climate change and also receives very different air masses, understanding the role of different emissions there in impacting CCN properties is particularly challenging and important.

The manuscript describes a solid data analysis and is well and clearly written. I only have several minor comments that might help to clarify some conclusions and state-

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ments that should be considered prior to publication in Atmospheric Chemistry and Physics.

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.

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.

Minor comments

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

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

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)

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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.

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

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

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

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

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.

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.

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.

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

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

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

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

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

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

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 ($\sim$142 nm)? p. 24701, l. 9: Do you mean the ‘bulk composition of the total aerosol population’ by ‘aerosol composition’ here?

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?

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.

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.

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

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?

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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.)

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

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

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

Technical comments

p. 24686, l. 14: . . .are provided

p. 24695, l. 11: remove 'concentration of CCN' (has been defined earlier)

p. 24696, l. 17/18: . . .characterized . . .confirmed

p. 24698, l. 3: remove 'plots of'

p. 24697, l. 8: . . . as few as. . .

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24677, 2012.