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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 #2

We thank the reviewer for their very detailed comments and contributions that have lead to an improved manuscript. Italicized responses are shown below.

1a. Lines 5-9 of p24681 – I agree, but it is important to note that there is no evidence for transport of Asian pollution reaching the surface in the high Arctic. It could happen in the future, but to date it has not been a significant factor at the surface (see Gong et al., 2010).

The pollution does not have to reach the surface to have an impact on the Arctic region. Refer to the discussion in the introduction with regards to the radiative effects of

aerosols on Arctic clouds (changes to cloud emissivity, radiative forcing, etc.).

1b. Lines 18-19 of p24681 - Some of the first observations of small particles emanating from polynias and open leads were made in the Canadian Arctic (see Leaitch et al.,1983; 1994).

This is now mentioned.

1c. Lines 9-12 of p24682 – The references in Moore et al. (2011) are missing the Canadian Arctic CCN measurements of Leaitch et al. (1983).

Thank you for pointing this out. References are now included.

1d. Lines 20-26, p24683 – I understand "CCN prediction" to mean computing the CCN spectrum based on a known or predicted number and chemical size distributions; I don't see it defined here. Also, in most deterministic approaches to CDNC prediction, CCN are not explicitly predicted, so I suggest explaining more clearly why the uncertainty in CCN prediction is important.

We will include a clarification for the term "CCN prediction". We will also more clearly explain that GCM parameterizations often require a computation of CCN spectrum, hence errors in the latter introduce a prediction uncertainty in CDNC and the indirect effect.

1e. Lines 17-18, p24685 – One of those few studies is Suda et al. (2012; see below). It should be referenced here.

Done.

1f. Lines 2-4, p24686 – Perhaps you are the first to demonstrate this for the DMT counter, but it has been known for at least 40 years that vapour depletion occurs in CCN counters for such high number concentrations. Your observation (and later on your correction) is useful, but please be clear that this is not something that people making CCN measurements in the past have ignored.

A fraction of researchers may be aware of vapor depletion effects on CCN measurements, but this issue is very rarely mentioned in the published literature (and virtually ignored during the last decade). The issue has not, to our knowledge, been systematically explored for its effect on data quality. Therefore, the statements are appropriate and no change is required.

2a. It seems a little surprising that the UHSAS did not operate well (above?) 2130m altitude, but the SMPS did. Did the customized SMPS operate well at all altitudes; e.g. were there no arcing problems?

The NASA LARGE SMPS is a custom built instrument specifically designed for aircraft measurements with parameters (ramp time and maximum voltage) carefully selected to avoid arcing at high altitudes and additional flow controls to maintain adequate sample flow. The UHSAS is a relatively new instrument that is not specifically designed for operation at high altitudes. The lower pressures led to internal flow control problems that affected data quality.

2b. P24689 – The WSOC is submicron (presumably, there was a 1 um impactor or cyclone in front of it), but the AMS is not (e.g. AS&T publications: Jayne et al., 2000; Rupakheti et al., 2005; Liu et al., 2007). Therefore, the WSOM fraction of the OM measured by the AMS will be an upper limit. This needs to be discussed, and carried through the analysis.

Indeed, a 1 um impactor was placed upstream of the WSOC measurement, while the AMS was subject to analysis of a larger size fraction. We agree that this is an upper limit. As reported in Section 2.3, the sensitivity of the CCN closure errors to the WSOM fraction was small (a 15% increase in WSOM fraction would increase reported CCN closure errors by an average of 4% and increase kappa by an average of 0.02).

2c. How was the AMS collection efficiency (oven bounce issue) treated, or was it? This is another potentially significant uncertainty.

These uncertainties are all included within the reported uncertainties in the AMS mass concentrations, as described in Section 2.3, which are dominated by uncertainty in the particle collection efficiency due to particle bounce (Huffman et al., 2005). This has been clarified in the text.

2d. Is the measurement of WSOC, as determined from the PILS, the same as WSOC derived from soaking particle-laden filters in water and analyzing?

The two methods are comparable, and this is characterized in Sullivan et al. (2004), where integrated 24-hr filter sampling and extraction methods were compared with online 24-hr PILS-WSOC averages , and agreement was found to be within 35 +/- 20%.

2e. You discuss the ability to derive O:C from the AMS, but you did not use it adjust the WSOC to WSOM. Instead, you use a constant 1.6 and then going through a relatively elaborate discussion to suggest the uncertainty is small. Why?

Determining an OM/OC ratio from O:C (Aitken et al., 2008) is a correlation based primarily on chamber measurements, not ambient data, therefore its application to ambient data is subject to uncertainty. For this, we chose a value representative of moderately aged organics (1.6) and tested the sensitivity of our results to this chosen value as discussed in Sect. 2.3.

2f. Line 21, p24690 – How “precise”?

0.05% (absolute) as mentioned in Sect. 2.5

3a. Line 11, p24693 – How “low”?

Less than 4% by volume (added to text).

3b. P24695 – I agree that your correction for supersaturation depletion is useful, but you need to discuss that such a correction may not be able to account for potential differences in the hygroscopicity of the particles at much higher concentrations.

The total surface area for condensation of water vapor onto the CCN in the CFSTGC is primarily responsible for the depletion effect. There is a weak dependency of the depletion term on aerosol hygroscopicity, because the correlation is expressed in terms of CCN (not aerosol) concentration. Given this, we feel that a discussion on the subject is not necessary.

3c. P24696, lines 21-24 – Is this true for the aged as well as the fresh plumes you characterized?

Yes

3d. Lines 2-3, p24697 – A CO concentration of 170 ppbv is much higher than is typical of "background" or clean air masses. How high was the CO (i.e. add it to Table 1), and how can you justify calling this clean or background?

These filters were chosen to be consistent with previously published studies. CO for the Arctic background was 92 +/- 5 ppbv. CO for the boreal forest background was 100 +/-3 ppbv. CO values rarely exceeded 100 ppvb. The airmass is background in so much as it is not influenced by biomass burning.

4a. Lines 13-25, p24698 – ": : :fresh biomass burning sampled in Canada has median number densities of 7832 cm-3." Although, I am sure you did not mean it this way, the phrasing makes it sound like it applies to all fresh BB in Canada. Of course, not only will number concentrations differ between burns, but it also depends on where in the "fresh" plume you sample as to what the number concentration might be. To me, this entire paragraph has little substance as these numbers (concentrations and sizes) depend on a large number of factors. If your sampling was for many cases there might be some value, but for a few cases separated by large distances and potential altitudes they are not appropriate as general characterizations.

These points are well taken. We will further emphasize that these are median values reported from our sample data (not for Canada as a whole), and represent the best

achievable results from the available high-quality flight data. The use of the numbers in the text (and the variability between plumes expressed by the width of the Figure 3 box plots) is to illustrate the magnitude of change in number concentrations when influenced by biomass burning and to contrast with the other sources in the airmasses sampled.

4b. Line 26, p24698 – You refer to OC and to WSOM. How was the OC derived? The C vs M reference is a little confusing to me in other parts of the paper. It would be useful to ensure consistency of use throughout.

OC is organic carbon derived from the AMS. OC is converted to OM via the OM/OC ratio as defined in Sect. 2.3. Each is used as appropriate in the text and it would be incorrect to define them differently.

4c. Lines 25-27, p24699 - One example of NPF in the Arctic summer is given by Chang et al. (JGR, 2011).

Done.

4d. Pierce et al. (ACP, 2012) shows that the organic kappa for a recent and pure forest biogenic aerosol may be as low as 0.06.

Reference noted

4e. Lines 5-18, p24703 – If the composition measurements are appropriate to larger sizes, then why not compare with CCN at lower supersaturations?

The automatic stepping of supersaturation limited its dynamic range availability when sampling a small scale plume. Thus, we had to compare at the available supersaturations, which were at the higher end (0.42% and 0.57%, as shown in Table 2).

4f. Section 4.3 –You say (p24701, lines 8-10) that the industrial case is an example of external mixing, and of course we expect the more aged aerosols to be more internally mixed (as you say on p24705, lines 19-20), yet when you do your closure you the

lowest uncertainty for the externally mixed assumption except for the industrial case. You suggest that we are to represent all these aerosols as external mixtures, except for the one that is most likely to be the strongest external mixture. This is a bit of a paradox, and it really deserves a little more attention and clarity in the discussion.

Industrial pollution is an external mixture, as stated, but because of size varying composition, the closure errors are still large. The large overprediction errors are due to this size-dependent composition, where a different kappa for each mode is required, and this is described in detail in the text (p24706, lines 11-25).

4g. Section 4.4, p24707 – So, according to figure 7, the aerosol becomes much more oxidized but kappa does not change. Does that not imply that the fundamental assumption about oxidized organics being more hygroscopic is incorrect? It seems like BB is one of the best situations to observe something like that, since the aerosol is so dominated by OM. If it doesn't work, then is there not a fundamental flaw in the assumption?

Yes, and this is one of the interesting findings of our study. There may be other processes involved such that the net effect of aging is that the hygroscopicity is not increased significantly. These observations should motivate additional research into this area.

4h. It is curious that there is no discussion or indication of how altitude plays a role in the observations and results.

Most of the data was constrained to within the boundary layer, due to instrument operating issues at high altitude.

5a. Line 9, p24708 – replace "captured" with sampled. Captured implies a representative sampling, which clearly this is not. Source is not clear with the Arctic background. Also, what about shipping emissions? Are they not a main source?

'Captured' has been replaced with 'sampled' and 'main source' replaced with 'some of

the primary sources'. Shipping emissions will be an increasing source (as mentioned in the introduction text), but were not quantified in this study.

b. Lines 20-25 – OM being such a significant component of the fine aerosol in the Arctic is an important factor contributing to this statement.

OK

6a. Figure 2 – The tiny plots of the size distributions with a log scale for the ordinate are uninformative. You discuss the CCN activity of Aitken- and accumulation-mode particles, not coarse particles such sea salt or dust; a linear ordinate is more appropriate for comparing with such CCN activity. Increasing the size of these plots, in combination with using a linear ordinate scale, will make these plots useful to others who might be interested (such as me).

Due to size constraints, the plot size cannot be increased further and the log-scale is a consistent way of presenting size distribution data with multiple modes and high variations in concentration. Important variables for model input (such as median size and standard deviation) are also tabulated in Table 1 for quick reference.

6b. Figure 7 – What about all of the other data points? Why are only the BB cases shown?

We chose to focus our analysis only on BB cases, since the other air masses would be expected to have already reached their limit in aging and therefore not exhibit large variations in O:C or kappa.

References:

Sullivan, A. P., Weber, R. J., Clements, A. L., Turner, J. R., Bae, M. S., and Schauer, J. J.: A method for on-line measurement of water-souble organic carbon in ambient aerosol particles: Results from an urban site, Geophys. Res. Lett., L13105, doi:10.1029/2004GL019681, 2004.

Huffman, J.A., Jayne, J. T., Drewnick, F., Aiken, A. C., Onasch, T., Worsnop, D. R., and Jimenez, J. L.: Design, Modeling, Optimization, and Experimental Tests of a Particle Beam Width Probe for the Aerodyne Aerosol Mass Spectrometer, Aerosol Science and Technology, 39(12): 1143-1163, doi: 10.1080/02786820500423782, 2005.

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