Atmos. Chem. Phys. Discuss., 12, C9466–C9471, 2012
www.atmos-chem-phys-discuss.net/12/C9466/2012/

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 #2

Received and published: 16 November 2012

This paper describes aerosol measurements conducted from the NASA DC-8 during ARCTAS in the summer of 2008 over northern Canada. The analysis focuses on the CCN activity of the aerosol, including characterizing the kappa values and CCN closure. The data are separated among five different aerosol types: fresh BB; aged BB; Arctic "background"; biogenic aerosol from boreal forest; and industrial emissions. The observations are useful and the paper is well written; although the discussion becomes more complicated near the end. I have number of mostly minor comments.

1. The introduction is a very nice discussion. A few comments and suggestions:

a. 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).

b. 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).

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

d. 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.

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

f. 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.

2. Section 2 – It is clear that the observations and analysis are very carefully done. Some comments:

a. 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?

b. 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.

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

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

e. 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?

f. Line 21, p24690 – How “precise”?

3. Section 3 –

a. Line 11, p24693 – How “low”?

b. 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.

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

d. 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?

4. Section 4 –

a. 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.

b. 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.

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

d. 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.

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

f. 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.

g. 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?

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

5. Conclusions –

a. 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?

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.

6. Figures -

a. 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).

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

Some of references mentioned above:

1) Gong, S. L., T. L. Zhao, S. Sharma, D. Toom-Sauntry, D. Lavoué, X. B. Zhang, W. R. Leaitch, and L. A. Barrie, Identification of trends and interannual variability of sulfate and black carbon in the Canadian High Arctic: 1981–2007, J. Geophys. Res., 115, D07305, doi:10.1029/2009JD012943, 2010.

2) Leaitch, W.R., Hoff, R.M., Melnichuk, S., and Hogan, W., 1984: Some chemical and physical properties of the Arctic winter aerosol in northeastern Canada. J. Climate Appl. Meteorol., 23, 916-928.

3) Leaitch, W.R., L.A. Barrie, J.W. Bottenheim, S.-M. Li, P. Shepson, and Y. Yokouchi, 1994: Airborne observations related ozone depletion at polar sunrise. J. Geophys. Res., 99, 25499-25517.

4) Suda, S. R., M.D. Petters, A. Matsunaga, R.C. Sullivan, P.J. Ziemann and S.M. Kreidenweis (2012), Hygroscopicity frequency distributions of secondary organic aerosols, J. Geophys. Res., 117, D04207, doi: 10.1029/2011JD016823.

5) Chang, R.Y.-W., S.J. Sjostedt, J.R. Pierce, T.N. Papakyriakou, M.G. Scarratt, S. Michaud, M. Levasseur, W.R. Leaitch, J.P.D. Abbatt, Relating Atmospheric and Oceanic DMS Levels to Particle Nucleation Events During the Canadian Arctic Summer. J.Geophys. Res., 116, doi:10.1029/2011JD015926, 2011.

6) Pierce, J. R. et al., Nucleation and condensational growth to CCN sizes during a sustained pristine biogenic SOA event in a forested mountain valley, Atmos. Chem. Phys., 12, 3147-3163, 2012.

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