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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Physical and chemical properties of pollution aerosol particles transported from North America to Greenland as measured during the POLARCAT summer campaign" *by* B. Quennehen et al.

Anonymous Referee #1

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This manuscript provides an interesting analysis of a unique airborne dataset collected near Greenland in summer 2008. This dataset is one of the few collected in summertime in this region of the troposphere, and includes measurements from state-of-the-art sensors such as the AMS. The data are of high quality and the topic, aerosol characteristics relevant to Arctic climate forcing, are important and should be published. However, the manuscript has several significant issues that must be addressed by major revision before publication in Atmos. Chem. Phys. There are four primary issues:

1) There is no evaluation of data quality, and the surprising results of a soot-dominated

Aitken mode are dependent upon this quality,

2) The analysis relies upon the absolute accuracy of the long-distance FLEXPART simulations, rather than upon values measured aboard the aircraft,

3) There is over interpretation of highly scattered data of limited or no statistical significance, and

4) The logic behind the conclusion that the Aitken mode is dominated by soot particles is unclear, and perhaps flawed.

Each of these primary issues is addressed in turn below, followed by less significant issues.

1) Evaluation of data quality. There are at least three sets of measurements that must be compared and discussed to demonstrate that the data are of sufficient quality to allow for quantitative analysis: size distributions, chemical composition, and light scattering. Size distributions can be easily integrated over the diameter range of efficient transmittance of the AMS inlet/aerodynamic lens. Assuming a density (or calculating one based upon the AMS composition + PSAP estimates of BC contribution), one can quantitatively compare the calculated and measured aerosol mass using a scatterplot and two-sided (orthogonal distance) linear regression analysis. Agreement within the stated uncertainties would permit further quantitative analysis. Similarly, using Mie theory one can calculate the light scattering of the aerosol and compare it to the nephelometer values. Further, nephelometer-based scattering and AMS concentration should be highly correlated. If an independent CPC were available on the aircraft, one could also integrate the size distribution and compare it to the concentration reported by the CPC. Differences in sample RH (especially for the PCASP) should be considered in all of the above calculations. Finally, the heated size distribution and non-heated size distribution should be compared for any cases that may exist when the thermodenuder was not on.

Figure 3 suggests that there are serious discrepancies between the aerosol mass calculated from size distributions and the AMS mass. These discrepancies need to be examined more quantitatively and evaluated before proceeding with any further analysis. Because the size distributions (Figs. 7, 8) are so different from earlier measurements in plumes transported long distances from forest fire sources, it is imperative that confidence in these measurements be established at the outset.

2) Over-reliance upon FLEXPART simulations. According to p. 11780, lines 10-15, periods of flight influenced by Canadian boreal fires (BF), Alaskan boreal fires (AK), and anthropogenic pollution from North American sources (AN) were identified solely by qualitative reference to plots of the column-integrated potential emission sensitivity (PES) from FLEXPART. The FLEXPART transport model is an exceptional tool, and one that is capable of providing substantial insight into emission sources and transport pathways. However, it is only as good as the meteorological fields driving it and the parameterizations for convective transport and boundary layer height within its code. In no case should it be used as the only source of information regarding airmass origins following transport of many days–it is simply not consistently accurate enough for such use. In particular for aircraft observations, minor altitude errors can translate into complete mismatches between in situ data and the FLEXPART transport paths. For example, in Fig. 5 there are periods where FLEXPART identifies that the aircraft should be in emissions from Alaskan fires, yet the CO mixing ratios are near 100 ppbv–the hemispheric background CO values. Yet observations from these time periods are analyzed as if the aircraft is in the midst of an Alaskan forest fire plume–which it clearly cannot be! Similar, if less extreme, issues are found for the identification of the BF airmasses in the other flights. It is likely that a plot of FLEXPART-reconstructed CO at the location of the aircraft would show little similarity to the actual measured values. Airmass properties must be evaluated first on the basis of measured values, and then the sources evaluated in part by using the FLEXPART results.

On p. 11782, lines 5-11, the measured number concentration enhancements are ra-

tioed with FLEXPART-calculated CO emissions to examine the change in particle concentrations with transport time. This analysis presumes that the FLEXPART simulations precisely track the transport of CO directly to the aircraft, including quantitatively determining the dilution of the plume over transport times of 6-13 days. It is unlikely that FLEXPART has this level of skill. It would make much more sense to used measured enhancements of CO over background to calculate this change in aerosol concentration over time. After all, if one can calculate a delta-N from the measurements, why not also calculate a delta-CO from the measurements, and use that internally consistent ratio?

I suggest that the authors select time periods of high CO mixing ratios as the foundation of their analysis, and use the FLEXPART simulations to help identify source regions of the layers. They need to reconstruct in-flight concentrations of CO from the FLEXPART simulations to demonstrate that the model can be used ascribe the source regions to specific features in the measurements. Analyses need to be based on measured values whenever possible, with interpretation only supported by–not dependent upon–the simulations of transport over such long distances.

3) Over interpretation of highly scattered data. In Section 4.2, the ratio of measured delta-N to modeled CO_emi is calculated (see point 2 above). These data are then plotted in Fig. 6, and exponential fits are made to calculate the decay of the dilution-corrected number concentration as a function of time, and its increase as a function of CO. It is difficult to believe that there is any statistical significance to these scattered data–particularly the second panel. The legends on the panels indicates "R2" values of 0.42 and 0.23, but R2 values are not applicable to nonlinear regression. Is this a reduced Chi-squared value, perhaps? Are the uncertainties in the values used as inverse weights to the fits, as would be appropriate? In any event, these plots should be redone using values normalized by measured delta-CO, rather than modeled CO_emi, as noted above.

4) Assessment of soot to Aitken mode. In Sections 4.4 and 4.5, an argument is made

that the small amount of absorbing material found, presumably soot, is attributable to ∼20 nm particles in the Aitken mode. This is deduced because ". . . large fractions of the SMPS measured Aitken mode are detected in the NVSMPS distribution of refractory particles below the detection limit of 20 nm". This sentence is unclear. How can large fractions be detected if they are smaller than the detection limit? This analysis would be more supportable if number size distributions of the SMPS and the NVSMPS for example conditions were shown, and the particles could be quantitatively mapped from the refractory Aitken mode to the non-refractory Aitken mode. The presence of the soot mass at sizes <20 nm would be quite surprising, and counter to extensive evidence collected in broad regions of the atmosphere in a range of conditions (see Schwarz, et al., Global-scale black carbon profiles observed in the remote atmosphere and compared to models, Geophys. Res. Lett., 37, L18812, doi:10.1029/2010GL044372, 2010.) The claim in Section 4.5 that "the ratio. . . between V_psap and V_aitken gives information about the fraction of BC particles within the Aitken mode" is true only if we accept the premise that the BC is within the Aitken mode–it does not provide independent evidence that this is the case.

Minor issues.

A) There are several more minor issues that should be corrected in a substantially revised manuscript. Several of the figures are very difficult to read. In particular, in Fig. 1 it is hard to see the blue and yellow portions of the flight, indicating Alaskan forest fire and anthropogenic influence, respectively.

B) In Fig. 2 it is difficult to see the continental outlines and so gather a geographical reference for the transport pattern. In this figure the authors should keep in mind (and point out to the reader) that the PES is plotted on a logarithmic scale, and that many of the features to which the eye may be drawn might be very minor contributors to the budget of the measured airmass. This is why reconstructed, source-apportioned values of CO along the aircraft flight track are so valuable, and more quantitative than these maps (see Fig. 1 of Warneke et al., Biomass burning in Siberia and Kazakhstan

as an important source for haze over the Alaskan Arctic in April 2008, Geophys. Res. Lett., 36, L02813, doi:10.129/2008GL036194, 2009).

C) In Fig. 3, it would be helpful to remove the cloudy portions from the data (since they are discarded anyway), and to provide a plot of aircraft flight altitude on the right-hand axis.

D) The use of yellow for the "AN" category begins in Fig. 3 and continues throughout the figures and tables. The yellow is very hard to see in many of these elements. In Fig. 3D the big plume of organic material at 14:30 and 16:15 is not identified as a haze layer of interest; the same with the 18:30-19:30 time period in Fig. 3C. Also, the AMS and size distribution curves appear to be poorly correlated in these two panels, drawing the reader's concern.

E) In Figure 4 the y-scale changes from panel to panel; it would be helpful to keep these fixed as in Fig. 3.

F) In Figure 7 it is very hard to distinguish the solid line from the curves with symbols. I would also suggest using a linear y-scale rather than logarithmic. If particles of some size range do not show up on a linear plot, then they are not important contributors to the number distribution. If they contribute to mass and you wish to highlight this, then show a volume distribution as well.

G) In Figure 8, the curves between the literature values and the current study are very different, drawing the reader's concern. Perhaps they could be normalized by delta-CO (if available in the literature values) to reduce dilution effects? Also, the reanalysis I suggest using data selected by in situ measurements rather than FLEXPART transport patterns might reduce the differences.

H) Figure 9 is very hard to read because of small symbols and lines.

I) Figure 10 needs a legend or caption to explain the color code.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 11771, 2011.