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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source identification and airborne chemical characterisation of aerosol pollution from long-range transport over Greenland during POLARCAT summer campaign 2008" *by* J. Schmale et al.

Anonymous Referee #1

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This manuscript presents selected results from 8 flights by the Safire ATR-42 during the summer phase of POLARCAT-France (June-July, 2008). All flights were conducted from Kangerlussaq in south western Greenland and targeted forecasted pollution plumes from a wide range of source regions that were predicted to be transported to and over the Greenland ice sheet. As suggested by the title, this manuscript primarily seeks to use aerosol composition (measured by AMS) and size distributions (measured by SMPS) to assess dominant sources for each plume that was encountered, thereby helping to validate the chemical forecasts.

Given the stated focus on aerosol characteristics, I am concerned by the criteria used to identify and define the start and stop times of plume encounters. Perhaps it is more accurate to say that the 5 case studies presented make me fear that the application of these criteria resulted in questionable plume selection and may have made analysis less informative than it might have been. Succinctly, criteria 1 and 2 are based on observed aerosol characteristics, hence would appear to be objective means to identify aerosol plumes, while criteria 3 focuses on CO and O3 which can increase with aerosol, but can also increase with constant or even decreasing aerosol. It might still be objective and informative to identify "pollution" plumes using these 3 criteria and examine whether aerosol size distributions and composition markedly differ in plumes with abundant aerosol plus either of the gas tracers compared to more pure aerosol plumes, or how aerosol characteristics may change in a CO or O3 plume even where aerosol abundance remains constant or even decreases. However, the authors added a 4th essential criteria, i.e. that the FLEXPART model could identify a clear link between a source region and any possible plume encountered by the aircraft, before any enhancement in CO, O3, aerosol sulfate, organic aerosol, or aerosol volume would be accepted as a plume. Examination of Figures 3 and 6 suggests that this last criteria resulted in plumes being identified on the basis of enhancements in O3 or CO more or less regardless of variations in aerosol properties. Specifically, plume I starts with a marked increase in both gases and stops when they drop back down, plume II is marked by a pulse of higher O3, plume III does contain notable enhancements in OC and SO4 but start and stop times appear to be defined by drop in CO, IV is based on O3 increase, V reflects simultaneous CO increase and O3 decrease, as does V(ref). Granted, in several, perhaps most, of these example plumes there were also increase in SO4, OC or volume, but in all cases the aerosol properties varied substantially within the defined plume encounter. More importantly, there are many instances of similar, or even more pronounced enhancements in aerosol mass or volume on both of these flights that do not meet the definition of plume, presumably because FLEXPART was ambiguous about where the airmass came from.

These two figures provide significant indications that FLEXPART is not yet a perfect tool even within the O3 and CO plumes that were identified. Out of 6 test cases presented, the predicted excess CO was reasonably close to observations twice (II and V), way too high twice (III and IV), and way too low twice (I and V(ref)). Therefore, in addition to suggesting that the authors reconsider the analyses presented in Section 4 using just the first 3 criteria to identify plumes, I feel that Sections 5.2.1-3 also need to be reconsidered since plume types defined for these analyses relied entirely on FLEXPART (see last two sentences in 5.2). My opinion, based on Figs 3 and 6, is that FLEXPART identified O3 and CO plumes that partially overlapped with aerosol plumes causing artificial mixing of aerosol plume/background, or multiple aerosol plumes when averaged over the defined plume intervals.

Specific comments/suggestions keyed to page/line #

7597/3 "rather" should be "more often"

7597/27 "rather" should be "primarily" or "preferentially"

7600/24 more detail needs to be given regarding which inlet on the DC-8 was copied, there were nearly 20. Suspect the model was the UH (or Clarke group) inlet, but the cited reference describes 3 different inlets that were tested during the DICE experiment.

7601/1-13 I am surprised that AMS sensitivity was so low, and wonder if the reduced mass spectra approach is valid (but am not an AMS expert so will assume it is ok). However, the explanation of how the raw spectra were filtered needs to be more clear.

7606/13-16 It might be very interesting to compare ATR-42 observations during phase 2 to those obtained on the DC-8 ARCTAS flight on 9 July when a similar (maybe the same) plume was encountered north of Thule

7607/12 constraints

7610/1-2 greater than 12

7612/10-11 enhanced at 145

7612/22-23 reflects a strong FF contribution rather than BB

7615/20-22 in the discussion of Fig 10 I do not understand how mass increases in upper trop while volume appears to decrease slightly. As noted several other places, the SMPS sees refractory particles that AMS does not, so that could explain the opposite case, but makes this discrepancy even larger. This needs to be explained, else the reader will suspect serious problems with one or both of the aerosol instruments.

7617/28-29,7618/1-2 My impression is that growth factors reflect ability of aerosol to take up water, which does not seem to explain why there is more SO4 in BB plumes over Greenland

7618/10-12 Not at all clear to me how one 2 minute observation of anything above the ice sheet can be consistent with interpretation of several hundred years of an ice core record.

7618/16-20 Explain the estimate that 89% of aerosol is SO4 more clearly and specifically. Also, reconcile this with statements elsewhere (including abstract) that OC accounts for 71% of mass in background and more in most plumes.

bottom7619/top 7620 Also possible that the binning based on FLEXPART smoothed out any differences that were in the different plumes. Detailed work on snow and ice on the icesheet clearly shows compositional differences in different layers (from different storms or dry deposition events)

7620/24 I would hesitate to say something "is always greater" based on just 2 cases (see Fig 12)

7621/7622 I do not find section 5.3 very convincing or useful.

7622 and Fig 14 Fit in the plot is not so good that I am convinced 9 days is better than any other choice in the 7-11 day range. Could say something about what the different

fits extrapolate back to at the source (day 0) and compare to measurements over that region.

7623/7624 The conclusion that OC in well aged aerosol is the same regardless of source is a strong statement. Given possible concerns about the AMS data processing applied to mitigate poor sensitivity, and the potential that FLEXPART combined plumes, I am not convinced that this is true.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 7593, 2011.