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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.

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Final response to anonymous Referee 1

General Response:

Thank you for your review. Based on the points addressed in the review we performed several changes in the manuscript. In the following we give a point by point answer to each comment.

Reviewer's comment:

"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."

Authors' response 1:

The title states that sources of aerosol pollution plumes are identified in this study and that the focus is laid on the chemical characterization of submicron aerosol from long-range transport. The title does not state that we primarily use aerosol chemical signatures for the identification of emission sources. Source regions are located by means of the FLEXPART model and back trajectory analyses and chemical composition and size distributions of aerosol belonging to the same category of source type are then related to it. We do not state that the purpose of this paper is to validate chemical forecasts, this has neither been attempted nor discussed in this work.

Reviewer's comment:

"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."

Authors' response 2:

The purpose of this work was to identify ***pollution*** *plumes, as opposed to "aerosol" plumes, that reached the Greenlandic troposphere via long-range transport and to characterize particle chemical properties of plumes that could clearly be attributed to a source region. Two aspects need to be considered in this respect:*

(1) A pollution plume according to our understanding will have a signature in both gas and particulate phase. Considering the process from emission over transport to detection several factors contribute to this understanding of pollution plumes. An emission source will emit gaseous tracers and likely primary particulate matter. With time some gaseous tracers, aerosol precursors, will undergo chemical change and partition into the particle phase (e.g. oxidation of SO2 to sulfuric acid which has a low vapor pressure and therefore is mainly found in the particle phase) and semi-volatile compounds might condense onto particles (e.g. secondary organic aerosols (SOA)). Therefore, for combustion sources as considered in this paper, be it anthropogenic FF combustion or forest fires, it is expected that both gaseous and particulate tracers are present. During transport concentrations of both types of tracers might change. Dilution will happen and decrease mixing ratios. Photochemical production of ozone and photochemical destruction of CO might occur. At the same time precipitation events can diminish the load of particulate matter and new formation of particles is also possible. Considering the long transport pathways from North America, East Asia and Siberia to Greenland

all these factors will have had an effect on the detected plumes. As shown in the case study of plume V, nearly complete wash-out of aerosol occurred, but CO is elevated and thus a pollution episode was crossed. Therefore, it makes sense to consider all available gas phase parameters and the signals from the AMS and SMPS. The logic of our approach was to identify all pollution episodes independently of aerosol wash-out and then to characterize the chemical properties of aerosol for all events. Since the chosen time resolution of the AMS measurements was 2 minutes, and the requirement set to have at least 3 data points, the start and endpoint of a pollution plume is defined by this factor in the case of short events.

(2) The purpose of this work was to establish a source-receptor relationship and then analyze the chemical properties and size distribution of submicron aerosol for each source-receptor category observed. It is thus the nature of this approach that requires as a precondition that a source-receptor relationship can be attributed to the pollutions plumes which were considered in the analysis. We are aware of the fact that an episode where the FLEXPART model could not establish a clear source-receptor relationship is automatically excluded from being a pollution event. Such cases, however, were not considered in this work for above stated reason.

Reviewer's comment:

"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."

Authors' response 3:

Please refer to the definition of pollution plumes as mentioned in response 2. In Fig. 3 panels b and e the enhancement of aerosol starting and stopping at the same time is visible. Plume III is clearly defined by the AMS signal (Fig. 3 panel b). The SMPS signal might be weaker in this case because of lower amounts of BC which is expected to be higher in e.g. Plume I where the source is BB while sources are mixed for Plume III. Plume IV is also characterized by an extended period of particulate sulfate elevation. The observation that "in all cases the aerosol properties varied substantially within the defined plume encounter" is correct. Referring to the explanations given in response 2 a number of factors will have influenced the characteristics of the individual pollution plumes. This includes plume filamentation, stronger dilution near the edges of the plume and less dilution within the center. It also has to be kept in mind that the aircraft might have crossed plumes near the edges and in the center or even slightly out of the plume during the identified episode. Consequently, it cannot be expected that an episode reflects a homogeneous concentration of submicron aerosol, especially when source regions and types are mixed.

Reviewer's comment:

"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."

Authors' response 4:

The authors are not sure which episodes the reviewer refers to. Table 3 lists all considered plumes for all flights. During the flight on 8 July (Fig. 3) 10 plumes and on 13 July (Fig. 6) 9 plumes were identified. As the reviewer states above, the plumes discussed in section 4 are "example plumes" or case studies selected from a total of 48 plumes presenting the variety and diversity of pollution events encountered during

the campaign.

Reviewer's comment:

"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))."

Authors' response 5:

The reviewer is absolutely right by stating "that FLEXPART is not yet a perfect tool". There are clearly limitations with respect to determining in-situ trace gas and trace aerosol species. With respect to trace gases it has to be kept in mind that local background concentrations are not available and thus only excess concentrations are reported. Therefore, variable background conditions which have an influence on the overall trace gas mixing ratio cannot be accounted for. This weighs heavier in cases such as reported here where the enhancement is very low due to the long transport times and associated dilution of plumes. Additionally, it has to be kept in mind that the aircraft will not always sample the plume in its center but frequently only on the side or on its lower edge, and that the resolution of the model is not sufficient to cover such small scale effects. For aerosol tracers, the wash-out is overestimated, as explained in the paper on p. 7605 lines 13 ff, and therefore it can only be used as the lowest estimate. However, we would like to point out that for establishing the source-receptor relationship the meteorological input data is most important separately from of the accuracy of the modeling of CO or other tracers' concentrations. Additionally GFS input data were considered (see p. 7604, lines 12 ff) and only source region matches between ECMWF and GFS data were used. Based on this we are convinced that FLEXPART is an adequate tool for the determination of pollution plume origin.

Reviewer's comment:

"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)."

Authors' response 6:

Based on the arguments given in responses 2 and 5 we do not see the necessity to reconsider either section 4 or 5.2.1-3. The purpose of this work requires establishing source-receptor relationships. Plumes which could not be attributed to a source and emission type category (3 plumes and 2 marine influenced air masses) are not declared as non-existent but are rather not considered within the context of this study. It is, unfortunately, impossible to determine the source region of a sampled plume only on the basis of the measurement data. Therefore, transport modeling is needed for our study and we think FLEXPART is a good choice for this task. Other transport models would likely have similar problems.

Reviewer's comment:

"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."

Authors' response 7:

We are not sure what is meant by "FLEXPART identified O3 and CO plumes that partially overlapped with aerosol plumes causing artificial mixing. . .". In our opinion gas phase and particles cannot be decoupled. The concept suggested by the reviewer, that CO or O3 plumes overlap with independent and distinct aerosol plumes does not seem realistic. The main source emission types during the POLARCAT campaign were fossil fuel combustion and biomass burning that both emit CO, primary particles and

aerosol precursor gases. It may be conceivable that aerosol particles have settled from a higher altitude plume and coincidentally mixed with a CO plume below where aerosol wash-out occurred beforehand. However, such a case is very unlikely and would most likely not have been recorded by the AMS since only particles in accumulation mode are considered which have a very low settling velocity. Therefore, we don't think that artificial mixing of plume and background aerosol happened beyond the uncertainties related to the given time resolution. Inhomogeneities in particle concentrations within a plume have been explained in response 3. Additionally, the process of selecting plumes involved first consideration of in-situ data with a minimum length of 3 AMS 2 minutes data points. Subsequently, FLEXPART sensitivities were checked for this period. If source regions changed during that minimum interval the plume was attributed to a mixed category (e.g. Sib BB / Asia FF). Any further breakdown of plume episodes can simply not be represented by the in-situ aerosol data. Thus the potential averaging of short and distinct pollution intervals within a 6 minutes interval has to be accepted as experimental bias. In case the source region changed within an in-situ data identified plume with a longer period than 5 AMS data points the plume was split based on FLEXPART data. We don't see how a different approach of plume identification could have been realized. Only considering aerosol data is on the one hand difficult because the data are often close to the detection limit and highly variable which makes it difficult to separate individual plumes occurring shortly after each other. On the other hand, wash-out event would have been completely neglected which would result in a loss of information. Thus in-situ trace gas and aerosol data were combined. The fact mentioned before response 2 that aerosol concentrations may increase, decrease or remain at the same level with enhanced or decreased CO or O3 is correct. However, all plumes considered in this study are long-range transport plumes where wet deposition played a crucial role. Therefore, the lifetime of CO is estimated to be longer than aerosol lifetime and thus only the combination of both parameters can lead to a comprehensive plume identification and description.

Reviewer's comment:

"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."

Authors' response 8:

The jump mass mode has been shown to be applicable to Q-AMS measurements (Crosier et al., 2007). We can't think of a reason why this method should not be applicable in this context. Moreover, Drewnick et al. (2009) showed that restricting the number of selected m/z to those with highest signal-to-noise ratio results in an improvement of the limit of detection. The reason for the low sensitivity of the AMS is explained on p. 7603, lines 5-10. The background concentrations in the instrument's vacuum chamber were high due to the small amount of time available for pumping. Due to this both the closed and open signals are large so that for the determination of the actual difference two large numbers need to be subtracted from each other which results in a high limit of detection. The passage has been adapted to (also based on comments by reviewer 2):

"This adds noise to the organic signal and increases the limit of detection (LOD). The LOD depends on the instrument's background signal (Ib) and is calculated from three times the standard deviation of Ib times sqrt(2) to account for the noise in the background and measurement signal which are subtracted from each other for the determination of the aerosol mass concentration. Therefore, only a selection of m/z, i.e. their contributions to the organic mass spectrum, was chosen to represent the total organic mass similar to the jump mass mode (JMS) used for quadrupole AMS data analysis as described by Crosier et al. (2007) and as suggested by Drewnick et al. (2009). The considered mass to charge ratios were derived from an organic mass spectrum obtained during sampling a pollution plume, thus accounting for mass to charge ratios present in the background and pollution events. The final selection criterion was a combination of three factors:"

. . .

"This method resulted in the selection of five m/z (15, 29, 43, 44, and 59) corresponding to the most abundant ions observed that exhibited a clear signature in all flights. "

Reviewer's comment:

"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"

Authors' response 9:

This is certainly true. According to the available list of Lagrangian matches between the DC-8 and ATR-42 there is unfortunately no match for the mentioned 9 July flight. As the 2nd reviewer notes, much information and many details are already given in this paper so that another paragraph on comparing DC-8 and ATR-42 results would be beyond the scope of this work.

Reviewer's comment:

"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."

Authors' response 10:

Thank you for pointing this out. We discovered an error in the calculation of the vertical profile of the SMPS volume concentration. Figure 10 (see figure-1) shows now the corrected data. Now both profiles agree much better. On average, the SMPS detects more particulate mass by a factor of two (assuming a density of 1.7 g cm-3). Although the SMPS detects black carbon and other refractory material that the AMS

is not sensitive to, a factor of two appears to be rather large. Additional reasons might be underestimation of the correction factors for the AMS inlet system. However, this remains speculative.

Reviewer's comment:

"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"

Authors' response 11:

The term "growth factor" was utilized in the style of the cited reference. To avoid confusion we rephrased and extended the passage to:

"It has been observed that in ageing BB plumes the relative increase in mass of particulate sulphate is higher than of organic carbon (Formenti et al., 2003; Yokelson et al., 2009) which can explain the relatively high percentage of sulphate in the detected BB plumes over Greenland. Enhanced particulate sulphate fractions in aged forest fire plumes have been reported before (Hudson et al., 2004)."

In fact, the high particulate sulphate content is an indicator for aged plumes since it is a secondary aerosol tracer. Since SOA is formed faster than particulate sulphate it is conceivable that during the initial uplift of the plumes which was often associated with precipitation SOA has been scavenged and thus diminished while particulate sulphate was formed afterwards.

Reviewer's comment:

"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."

Authors' response 12:

The phrasing might be a bit misleading. No comparison of a mass concentration from a single pollution event sampled during the POLARCAT campaign was meant to be compared with sub-seasonal resolved ice core data. We intended to say that this type of plume may give a contemporary example of what has been deduced from ice core data before. The comparison has been eliminated nevertheless in this passage as reference in the sense as described above has been made in the introduction of the paper.

Reviewer's comment:

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

Authors' response 13:

Plume category V (episodes dominated by Asian FF and little influence from Siberian BB) exhibit a fraction of 0.37 particulate sulphate. We were curious how high the contribution of particulate sulphate would have been if a plume of pure Asian FF origin had been observed (which was not the case but could occur outside of the fire season). Thus, the contribution of Siberian BB was derived by means of the FLEXPART passive CO fire tracer which indicated between 54 and 85 percent BB contribution for the respective plumes. Hence, it was assumed that in the BB contribution 22 percent particulate sulphate were contained as learned from plume category I ("pure" BB). Based on this the theoretical composition of a pure Asian FF plume was calculated resulting in a sulphate fraction of roughly 0.89. Since this is not an observation but a theoretical consideration it cannot be reconciled with the reported observational results that pollution plumes advected primarily organic matter. The passage has been rewritten for clarification to:

"During the POLARCAT campaign no "pure" Asian FF plume was observed as East Asian outflow was always mixed with Siberian forest fire influenced air masses. How-

ever, it is conceivable that outside of the fire season air masses exclusively characterised by East Asian anthropogenic activity are transported towards Greenland. Therefore, the theoretical composition of a “pure” Asian FF plume was calculated. The FLEXPART CO passive fire tracer indicates BB contributions between 54 and 85 percent for the respective episodes. From the aerosol composition of category I (“pure” BB) we know that the BB contributions contain roughly 22 percent particulate sulphate. Based on this a percentage of 88.7 $\pm$ 34.5 percent of particulate sulphate is estimated for a “pure” Asian FF plume.”

Reviewer’s comment:

"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)"

Authors’ response 14:

Based on the explanation given in response 7 it is clear that binning based on FLEXPART happened only in special cases where aerosol and gas phase tracers indicated a plume over a period of time during which the source region changed. Hence, the authors do not think that the similarity in the O:C ratio in all plume categories is an artifact due to the selection method of plumes. Also, the variance within each plume class encompasses a range of values which shows that differences were present. However, these differences cannot be resolved any further. It also has to be kept in mind that the O:C ratio is approximated by means of a linear regression based on literature as clearly stated in the paper. Nevertheless, it has been stated in several publications that aged organic matter homogenizes (e.g. Jimenez et al., 2009 and Andreae et al., 2009). It is difficult to give an answer to reviewer’s general statement that compositional differences were found on snow and ice on the ice sheet. Details on source regions, source types, age, transport patterns, and especially analytical methods etc. would

be needed to evaluate if a comparison is possible. What we can state is that similar O:C ratios and compositional differences do not exclude each other. For example, the following organic functional groups all have one oxygen atom: aldehydes, alcohols, and ketones. In case they all have the same R- they would exhibit the same O:C ratio. Likewise oxalates or dicarboxylic acids have both 4 oxygen atoms.

Reviewer’s comment:

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

Authors’ response 15:

Very few data on submicron aerosol size distributions have been published for long-range transport plumes above Greenland (Brock et al., 1989 and 1990). In the authors’ opinion this bears the responsibility to publish the available data. As comparison with data from Petzold et al. (2007) of similar measurements of emissions from comparable source regions and of comparable age over Europe shows, data cannot simply be adopted because there are significant differences. This Greenland specific information can be valuable for modeling studies and considerations especially with respect to aerosol-cloud interactions where the size of the particles plays a crucial role. However, we understand that the presentation of the results can be optimized. For better comparability of the size distributions all number fits are now displayed in one graph as well as all volume fits are shown in a second graph (new Fig. 13, see figure-2). The distributions had to be recalculated due to the mentioned error in conversion from ambient to STP conditions (see response 10). Nevertheless, the main observations and trends remained.

Reviewer’s comment:

"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."

Authors' response 16:

We agree with the reviewer that any tau-value in the range from 7 to 11 days can reflect the aerosol lifetime. We changed the notation from "9 $\pm$ 2 days" to "7 to 11 days". Fig. 4 b gives an example of the source region for the plumes considered in this calculation. Since this is a very large region including a variety of emission source types, it will prove difficult to find measurements in this source region that can be related to the measurements obtained during POLARCAT. Measurements made in the source region area will most likely not reflect the variety of emissions that contributed to the signal above Greenland.

Reviewer's comment:

"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."

Authors' response 17:

The conclusion states that the O:C ratio in the well aged aerosol is similar regardless of the source region. The O:C ratio must not be considered to be equivalent to particulate organic carbon or organic matter as different organic chemical compounds can have the same O:C ratio (see also response 17). Additionally, it has been reported in several publications (e.g. Jimenez et al., 2009, Ng et al., 2010) that well aged aerosol tends to adopt high but uniform O:C ratios. With respect to the concerns related to the AMS data analysis and the involvement of FLEXPART results, please refer to responses 11 and 12. Additionally, the O:C ratio is based on the ratio of the organic contribution to m/z 44 and the total organic mass. The organic m/z 44 signal had a strong and clear signal and is trustworthy. The total organic mass has been determined as described

in the manuscript including statistical parameters and is still a standard method for quadrupole MS versions of the AMS. In our point of view the presented results are apt to allow the stated conclusion.

Specific comments/suggestions keyed to page/line 7597/3 "rather" should be "more often"

Ok, the revised manuscript has been adapted accordingly

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

Ok, the revised manuscript has been adapted accordingly

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.

Thank you for pointing this out, the revised manuscript specifies that the UH inlet was used.

7607/12 constraints

Ok, the revised manuscript has been adapted accordingly

7610/1-2 greater than 12

Ok, the revised manuscript has been adapted accordingly

7612/10-11 enhanced at 145

Ok, the revised manuscript has been adapted accordingly

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

Ok, the revised manuscript has been adapted accordingly

7620/24 I would hesitate to say something "is always greater" based on just 2 cases

(see Fig 12)

Ok, "always" has been omitted

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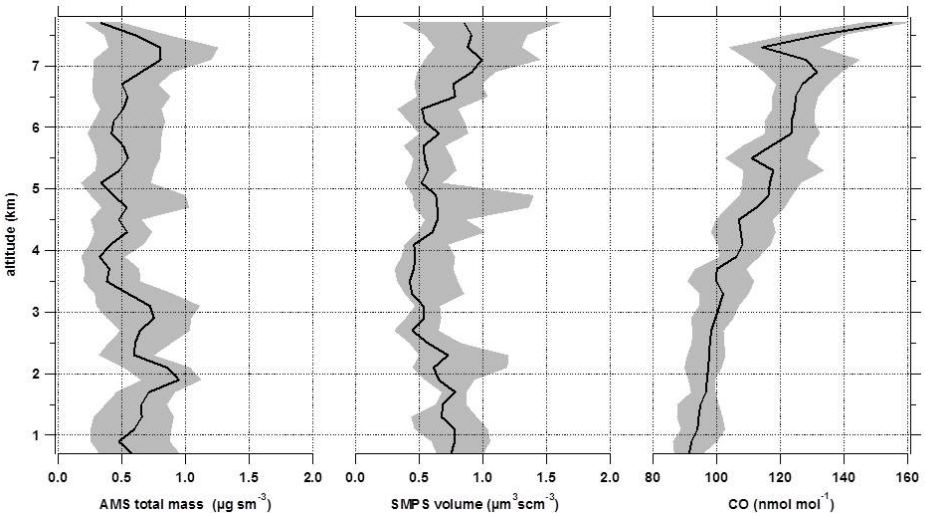

Fig. 1.

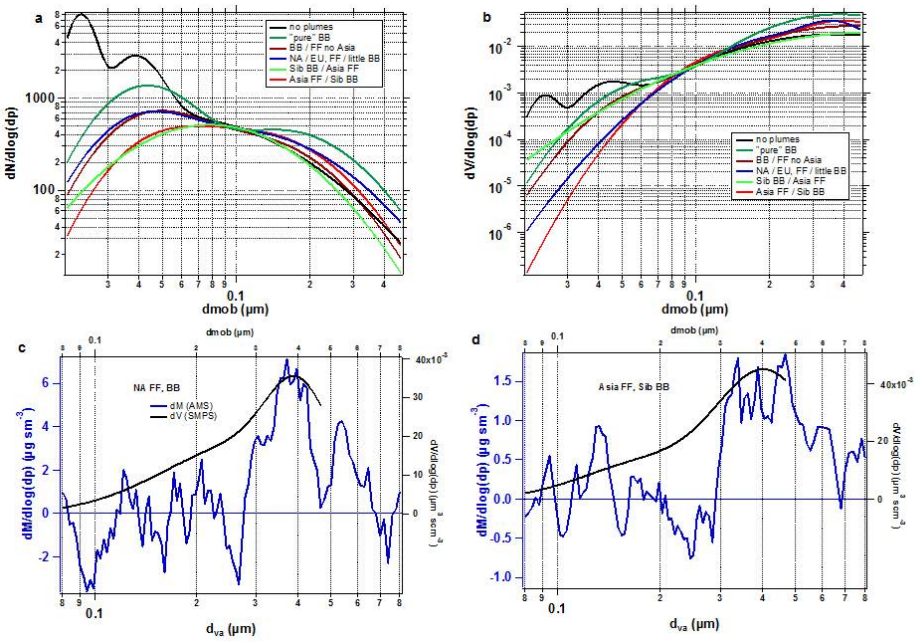
a
dN/dlog(dp)
dmob (µm)
no plumes
"pure" BB
BB / FF no Asia
NA / EU, FF / little BB
Sib BB / Asia FF
Asia FF / Sib BB
b
dV/dlog(dp)
dmob (µm)
c
NA FF, BB
dM (AMS)
dV (SMPS)
dM/dlog(dp) (µg sm-3)
dV/dlog(dp) (µm3 scm-3)
dva (µm)
d
Asia FF, Sib BB
dM/dlog(dp) (µg sm-3)
dV/dlog(dp) (µm3 scm-3)
dva (µm)

Fig. 2.