

Interactive
Comment

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.

J. Schmale et al.

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General response

Thank you for your review and comments that have helped to improve this paper. Based on the review comments we have performed a number of changes to the manuscript. In the following we present a detailed response to each point raised.

Reviewer's comment:

The paper presents AMS measurements made on the ATR-42 research aircraft over
C4653

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Greenland as part of POLARCAT. FLEXPART products are heavily used in the analysis. There is much useful information in the paper but it is difficult to read as the details hide many of the results. The paper could be publishable after the concerns described below are addressed.

Most significantly, readability of the paper should be improved by highlighting major findings and using data examples to support them (rather than vice versa)

Authors' response 1:

In the revised version we will try to improve the readability as suggested by the referee. We now focus on the variability in the trends of gas phase mixing ratios and aerosol concentration as a result of transport pathways and meteorology. The subsection starts with 4.2.2 "Aerosol and trace gas enhancement from Canadian BB", a case (Plume I) where all gaseous and particulate tracers are elevated. From there we move on to 4.2.3 "Aerosol enhancement from North American BB and FF pollution with minimal trace gas signature" where Plume III is discussed when almost no enhancement in the CO tracer was found, but aerosol is clearly elevated. Subsequently the contrary, the wash-out case, Plume V and Vref, is discussed in 4.2.4 "Strong CO enhancement and aerosol wash-out in WCB". To show that not all WCB events result in efficient aerosol scavenging Plume II is elaborated in 4.2.5 "Significant aerosol and trace gas enhancement despite transport associated to WCB". Finally, Plume IV, is discussed in the light of the stratospheric intrusion in 4.2.6 "Stratospheric air mass intrusion into upper tropospheric Asian pollution plume" and the associated change in trace gas signatures compared to Plume V which was sampled in the same large scale pollution episode.

Reviewer's comment:

and discussing and quantifying the uncertainties of the FLEXPART products used since they make up a large part of the analysis.

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Authors' response 2:

It is difficult to quantify the uncertainties of the FLEXPART products for this campaign. Quantifying uncertainties of source-receptor relationships needs independent data allowing to establish such source-receptor relationships. Ideally, one should know the source strength of a substance exactly and have accurate measurements of this substance downwind. This is the purpose of tracer experiments and FLEXPART has been extensively evaluated using data from such tracer experiments (Stohl et al., 1998; Forster et al., 2007). In the absence of a unique tracer, carbon monoxide enhancements are often used as an indication of how well a model can reproduce them along a flight track. However, in addition to the sources of CO being highly uncertain (especially biomass burning sources), the CO enhancements encountered during the campaign were all relatively small, which makes the comparison between measured and modeled CO enhancements highly dependent on the assumed background CO concentrations, which may have varied throughout the campaign and even within individual plumes. Other substances that could serve as tracers for model evaluation were not measured during the campaign. Therefore, a thorough evaluation of FLEXPART using the campaign data is not possible and we have to refer to previous papers (especially, Stohl et al., 1998; Forster et al., 2007) for model evaluation.

Reviewer's comment:

In addition, I have questions about the data quality given the lack of a reasonable comparison between AMS total mass and SMPS volume (based on Figure 10). A formal comparison of these parameters with an uncertainty analysis should be provided.

Authors' response 3:

Please refer to response 17.

p. 7595, line 18: change to "low-volatility, highly oxygenated aerosol"

Ok.

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p. 7595, lines 19 – 20: Instead of merely stating that the volume size distributions were “rather similar”, please describe the characteristics, i.e., modal diameter and width.

Authors’ response 4:

The abstract has been changed to:

“The volume size distribution of out-of-plume aerosol was markedly smaller with a diameter of 24 nm in the dominant mode and a width of 0.16. Nearly pure BB particles from North America exhibited an Aitken mode of 66 nm and a width of 0.53 and an accumulation mode diameter of 387 nm (width 0.80), while size distributions grew larger for longer travelled particulate matter from East Asia and Siberia (411 nm, width 0.50).”

Reviewer’s comment:

p. 7595: Why is only the aerosol lifetime for North American emissions given in the abstract? Also, it is not clear what this term is without more of an introduction. Lifetime since emission? Lifetime in the Arctic?

Authors’ response 5:

Only the aerosol lifetime of North American emissions could be derived, no statement about East Asian and Siberian particles can be made. The sentence in the abstract has been rephrased to:

“An aerosol lifetime, including all processes from emission to detection, in the range between 7 and 11 days was derived for North American emissions.” *Reviewer 1 prefers to explicitly name the range instead of saying 9 ± 2 days.*

Reviewer’s comment:

p. 7601, line 6: LOD should be defined (i.e., what are the values based on) when it is first introduced.

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Authors' response 6:

The sentence has been changed to:

“This adds noise to the organic signal and increases the limit of detection (LOD). The LOD depends on the instrument's background signal (I_b) and is calculated from three times the standard deviation of I_b 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.”

Reviewer's comment:

p. 7601, line 6: Should this be “Therefore, only a selection of m/z corresponding to the most abundant ions observed in all flights was chosen. . .”?

Authors' response 7:

Basically yes. However, it is important to mention that certain m/z have contributions from different chemical species. m/z 44, for example, has a contribution from the organic mass spectrum but also a signal from air. So, only the contributions to the organic spectrum of each m/z were considered. The explanation of how the ions were selected follows below, so we prefer not to mention yet that the method resulted in the selection of the most abundant ions. The sentences have been rephrased to:

“Therefore, only a selection of m/z , i.e. their contribution 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

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the most abundant ions observed that exhibited a clear signature in all flights."

Reviewer's comment:

p. 7601, line 14: Was the number of increased points above LOD compared to the total organic mass signal included in the analysis in any way?

Authors' response 8:

We are not quite sure where this question leads to. The organic mass derived by means of the jump mass mode for which the number of points above LOD in the selection of m/z is crucial was used throughout the entire analysis of this paper. However, there is no weighting of plumes during a flight where more points for above LOD than during other flights. Does this answer the question?

Reviewer's comment:

p. 7601, line 19: Please quantify "a high representation of mass of the total organic mass."

Authors' response 9:

The statement has been specified to:

"3. the highest representation of mass of the total organic mass after fulfillment of points 1 and 2." The actual values are displayed in Table 2 as referenced in the paragraph below the bullet points.

Reviewer's comment:

p. 7602: Based on the aerodynamic lens used in the AMS, what size range of particles of that actually made it into the instrument?

Authors' response 10:

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The limiting factor for the transmitted particle size range is the pressure controlled inlet (PCI). The transmission range for this PCI is between 80 and 1000 nm dva as stated in line 16 on page 7600. As it might not be completely clear that the given range is the total transmission range the sentences has been changed to:

“...the overall efficiency over the complete transmission range of 80 to 1000 nm (dva) is 54 percent.”

p. 7603, line 7: change to “...because the vacuum pressure decreases over time.”

Ok.

p. 7603, line 8: omit “still”.

Ok.

Reviewer's comment:

p. 7605: The treatment of “aerosol-like BC tracers” is confusing. If it is truly an aerosol-like BC tracer, why is it treated like sulfate? It should be termed a “hygroscopic aerosol tracer”.

Authors' response 11:

The reviewer makes a good suggestion. However, the model calculations are not made for any particular emission source but use generic “aerosol-like” properties comparable to sulfate to derive an emission sensitivity. This emission sensitivity is calculated backward in time from the measurement point and does not consider any specific emission source (or a specific “tracer”). Only later, as part of the post-processing, the emission sensitivity is folded with particular emission fields such as for sulfur, BC or CO. Thus, the only difference between our various “aerosol-like” tracers is the emission source distribution. While this may seem oversimplified to an aerosol modeler (and “aerosol-like” carbon monoxide certainly also cannot be compared directly to any mea-

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sure substance), the resulting aerosol lifetimes are reasonable also for BC and well reflect expected differences in different regions of the world (Cassiani, paper in preparation). The authors use the FLEXPART common terminology for the aerosol-like BC tracer which is also indicated as such in the online reference given on p. 7604, line 17. As a compromise line 19 on p. 7605 has been changed to “while the hygroscopic aerosol-like tracer represents the lower limit.”

Reviewer’s comment:

p. 7605, lines 20 – 22: I am confused by the statement that “even though CO and BC do not necessarily have the same sources, it seem suitable to compare these tracers within the uncertainties of the source-receptor analysis.” Are the two species being used to discriminate between sources or combined to represent single sources?

Authors’ response 12:

Thank you for mentioning this. This sentence is a relic from a former version of the manuscript. For the ACPD paper no such analysis was applied any more. The sentence was removed from the manuscript.

p.7607, line 12: Should be “constraints.”

Ok.

Reviewer’s comment:

p. 7608, lines 18 – 19 and Figure 3: On p. 7605 it is stated that the aerosol-like tracer’s wet deposition is overestimated. How, then, can its use to derive an aerosol wash-out ratio be justified?

Authors’ response 13:

The wash-out ratio needs to be considered as a qualitative value rather than as a quantitative measure. P. 7605, line 20 states that the ratio of the aerosol-like and passive tracer indicate the “potential” wash-out not the actual. Simply by the logic of how the

tracers function the concept of this parameter becomes clear: The passive tracer is not subject to wet deposition and is therefore the upper limit of modeled tracer concentrations and the aerosol-like tracer is washed-out more heavily than real particles would be and is therefore a lower limit estimation. During the POLARCAT campaign flights, all plumes were subject to wet deposition along the transport pathways so that the ratio of the FLEXPART tracers is always very low and little variability is seen. In other campaign, however, the tracer ratio covers a much larger range of values. Here, the low values are used to illustrate qualitatively that strong scavenging occurred during the long-range transport.

Reviewer's comment:

p. 7611, lines 10 - 15: Without a BC measurement and with a highly uncertain "aerosol-like" modeled tracer of BC, nothing can be said about BC concentrations in particular plumes.

Authors' response 14:

This is correct. This passages intended to give an explanation why the SMPS signal is stronger during the encounter of plume I and weaker for plume III. Plume I is clearly associated with BB while the late episode of plume III is dominated by FF. Assuming that BB would emit more BC than FF the variance in the signal can be explained. This argument is not based on FLEXPART results. However, we understand that this might be inferred since this explanation is given directly after the passage discussing FLEXPART tracers. Therefore, lines 8 – 13 have been deleted.

Reviewer's comment:

p. 7612, lines 2 – 4: It is stated that pollutions plumes were sampled (Event III). SO₄ and organics are elevated but measured CO is not. Any explanation?

Authors' response 15:

P. 7612, lines 2-4 say the following: "Pollution plumes sampled between these two

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low-pressure systems were in a region of stretching and filamentation leading to the formation of fine-scale. . .” Do you mean p. 7610, lines 24 ff where event III is discussed? Fig. 1 at the end of the document shows the 1 Hz CO data (black) and the flight altitude (green). Smoothed CO data (red) generated by the boxcar method averaging over 30 points is added for better illustration of the trends in the CO signature. The red box indicates the plume episode. An increase in the CO mixing ratio from below 100 nmol mol⁻¹ to almost 110 nmol mol⁻¹ can be observed during the pollution episode. Unfortunately, the instrument conducted a self-calibration during the plume encounter. Due to this and due to the fact that the CO mixing ratio does not decrease after the plume encounter but increases dramatically, most likely because the aircraft’s own emissions were sampled during landing, it is difficult to derive a delta value and to state whether there is a true enhancement in CO caused by the pollution plume. Therefore, in the manuscript the formulation “Gas-phase parameters do not show a signature which can be identified as pollution” was chosen.

Reviewer’s comment:

Sections 4.1. and 4.2. I found Section 4.1 difficult to read as the main points (which are summarized on p. 7614, lines 17 – 22) are lost in all of the detail that is given. I suggest the section be re-written so that the main points are delivered and the data are summarized in order to illustrate the main points, not vice versa.

Authors’ response 16:

We will rewrite section 4.1 for the revised version, please see response 1.

Reviewer’s comment:

Fig 10: Has any attempt at closure between the AMS mass and the SMPS volume been performed? I realize that the AMS only detects non-refractory mass but SO₄ and organics should make up the majority of the total aerosol mass in lieu of any major dust events. Yet, AMS total mass and SMPS volume as displayed in this figure appear to

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not be in agreement. Why are the two peaks in total AMS mass below 3.5 km not seen in the SMPS data? Why is there more structure at the high altitudes in the AMS data than the SMPS data?

Authors' response 17:

In the meantime we discovered an error in the conversion of the vertical profile of the SMPS volume concentration from ambient conditions to standard temperature and pressure. Figure 10 (see Fig. 2 at the end of the document) 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:

p. 7615, line 23: what non-plume aerosol is being referred to here? The entire experiment? The altitudes of low total aerosol mass concentration? The altitudes of low CO mixing ratio?

Authors' response 18:

The entire campaign's out-of-plume aerosol is meant. The sentence has been clarified: "In general, the entire campaign's non-plume aerosol is composed of. . . This encompasses all altitudes, however signals from take-off and landing were not considered."

Reviewer's comment:

Figure 11 and in general: The paper relies heavily on FLEXPART products. Uncertainties associated with these products (e.g., age spectrum and average plume age) should be discussed as quantified to the extent possible. Is there a significant difference between 10 and 15 days in plume age given the uncertainties in the transport model?

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In addition to our response 2, we would like to emphasize that calculations have been made with two different types of meteorological input data sets, to characterize uncertainties associated with the input meteorological data. All plume identifications have been cross-checked also with GFS data and are consistent with each other. We are not quite sure what the reviewer means by "significant differences". Do you refer to chemical, microphysical or other differences? As far as the age of plume is concerned, in the former subsections 4.1 and 4.2 an age range and in graph 11 the average plume age \pm one standard deviation for each category is given. Since most detected plumes originate from a large source area, e.g. East Asia and Yakutsk, Siberia, or Canada and Great Lakes area, it is not possible to determine the age of the pollution event as a single number of days. Air masses joint and mixed along the transport pathway all contributing to the signal detected above Greenland. Therefore, ranges are given accounting for contributions from all source areas.

Reviewer's comment:

p. 7620, lines 1 – 4: Isn't it possible to have similar O:C ratios for different organic compounds? Why is the similarity in O:C ratio assumed to mean that the composition of the organic aerosol is the same regardless of origin?

Authors' response 20:

It is correct that similar O:C ratios can represent different organic compounds. Having had this argument in mind the formulation "chemical characteristics" (p. 7620, line 1) was chosen instead of chemical composition. The following sentence has now been added for clarification:

"However, this does not imply that the chemical composition is the same across all plume categories as different organic compounds can exhibit the same O:C ratio."

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Reviewer's comment:

p. 7622: What is "chemical quality?"

Authors' response 21:

The term "same chemical quality" was replaced by "similar chemical and hygroscopic properties" which includes the chemical composition but also hygroscopicity.

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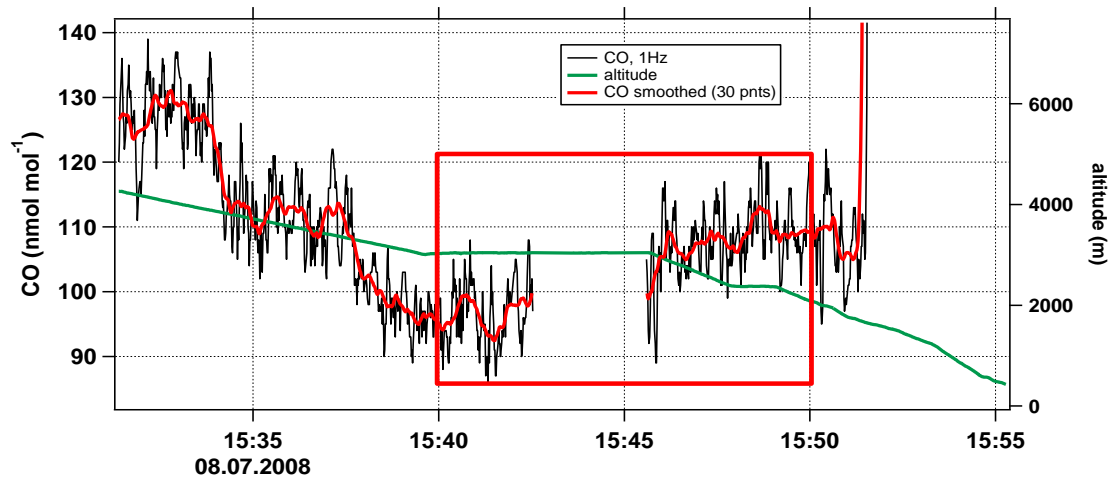


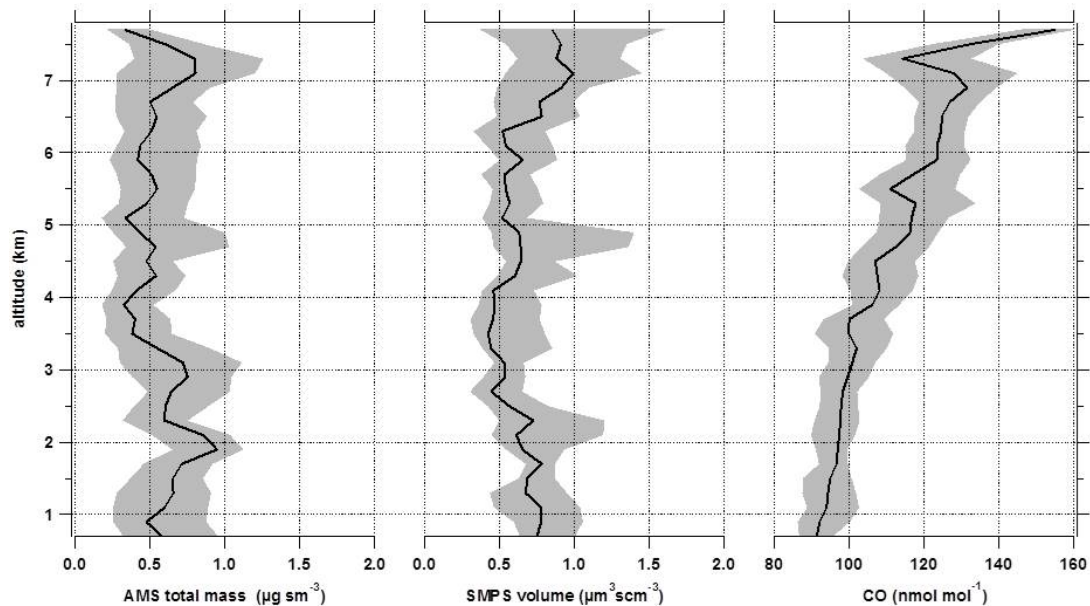
Fig. 1.

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