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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.

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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 #2 Received and published: 13 May 2011

Referee Comment:

General scientific comments: The manuscript presents interesting results on Green-

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landic aerosols influenced by anthropogenically polluted air masses. Generally, the manuscript is written in good English language and only a few comments with concern to the language have to be addressed. The figures are easy to understand based on their structure and figure captions are also clear. Nevertheless, figures cause some problems as the colour codes are sometimes difficult to distinguish and figures should appear larger in the final version. In addition, I will greatly encourage the authors to take advantage of the DMPS and AMS data by showing a mass comparison if possible. Also, it is recommended to use the nephelometer data for further analysis in dependence on air mass origin.

Summing up, the manuscript is worth for publication in ACP, but I would like the authors to address the general scientific comments listed above and the detailed scientific comments listed below.

General response: We thank referee n°2 for his constructive comments on our manuscript. We have revised the manuscript attempting to take into account all the comments raised. In general, the figure quality and thus readability has been improved. This concerns particularly figures 1, 2, 3, 4, 7, and 8, where we now used bold border lines, changed colour codes to make figures clearer, and zoomed on necessary information. Furthermore the data quality has been evaluated more carefully. Datasets of AMS, SMPS, and nephelometer measurements are coherent when compared to each other. A paragraph is added in the manuscript to describe the data quality evaluation.

Detailed scientific comments:

Abstract

Please clarify a few expressions that you use as "aerosols size distribution". Is it number, mass, etc. distribution? Or "aerosol volatility fraction". Is it volume, mass etc. fraction? You have to be exact when discussing these parameters. This is valid for the whole manuscript.

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Authors response: The entire manuscript has been checked to clarify when talking about number, mass, or volume, volatile, and non-volatile distributions, however we've tried not being repetitive. Page 11773, line 21: This is not a full sentence. Please clarify! Authors response: The sentence has been changed to: This is a valuable finding with respect to potential climate impacts of black carbon in the Arctic.

Introduction

Page 11774, line 10-11: The sentence has a bad wording.

Authors response: The sentence has been changed to: The principal objective within the French POLARCAT program has been to increase our knowledge about long range transport of short-lived pollutants (particulates and gases) to the Arctic.

Page 11775, line 18: ... aims at characterizing ...

Authors response: Expression changed to:...aims at characterizing...

Page 11775, line 18: I personally would reformulate the expression "pollution aerosol particles".

Authors response: The sentence has been changed to: This study aims at characterizing aerosol particles transported from North-American pollution sources to Greenland during the Arctic summer and their consequences for the Arctic climate.

2. ATR-42 aircraft measurements

2.1 Physical aerosol properties

As you conclude from number measurements to volume and mass measurements, you should describe your assumptions about shape of particles used in this model.

Authors response: We assumed spherical particles. This is now mentioned in the text.

In addition, how was the overlap between SMPS and OPC done, they are based on different diameter definitions? Any calculation of full size distributions needs some

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theory behind it. I can also not see any reference on the combination of these two instruments.

Authors response: On the one hand, the SMPS system consisted of a TSI model 3010 condensation particle counter, a DMA (Differential Mobility Analyzer) as described by Villani et al. (2007) and a krypton aerosol neutralizer (Kr-85). SMPS electrical mobility distributions (in number) have been converted to number size distributions. The SMPS system and inversion algorithm have been evaluated within the frame of EUSAAR. On the other hand, the PCASP is regularly calibrated at DMT with latex spheres (refractive index 1.59). PCASP and SMPS instruments measure dry aerosol particles sizes, since both instruments are working with dry sheath air. When merging SMPS and PCASP data we consider the mobility shape factor to be one. In general SMPS and PCASP number size spectra are matching pretty well within the common size range of 0.11-0.467 μm . We performed Mie calculations to study the response of PCASP size spectra as a function of the complex refractive index. This allows us to recalculate the entire PCASP size spectra for whatever complex refractive index. With respect to the measurement performed on the ATR-42 during the POLARCAT summer campaign, however, we consider the chemical information (no information about sea salt, mineral particles, chemical composition of supermicron particles...) to be insufficient to deduce a non-ambiguous size dependent complex refractive index. This means that correcting the PCASP data for another complex refractive index would have been somewhat a guess. Thus, we decided to keep the size spectra as given for the calibration refractive index of spherical latex spheres for this study. This means we assume the particles to be latex type particles. To merge SMPS and PCASP data we decided to linearly weight the SMPS spectrum from 1 to 0 and the PCASP data from 0 to 1, respectively, within the common size range. An explanation has been added in the manuscript.

2.2 Chemical composition and trace gases

Comments on the use of the AMS are a little beyond my level in this area and are hopefully addressed by other reviewers.

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2.3 Aerosol optical properties

A small discussion on the choice of the specific absorption coefficient would be reasonable as there are different numbers which can be found in the literature.

Authors response: Most of the specific absorption coefficients found in the literature have been deduced for the aethalometer. For the PSAP we used in this study the specific absorption coefficient of 11.6 that has been found by Sharma et al (2002) for Arctic BC aerosol measured during summer at Alert station. Sharma also gave other values for the PSAP wavelength corresponding specific absorption coefficient for Arctic winter BC aerosol particles and also near source regions (Toronto). Finally, BC data can be easily recalculated for other specific absorption coefficients, since the relationship between absorption coefficient, specific absorption, and BC is linear.

Can you deduce from the chemistry, which absorbing material was present?

Authors response: No, thus we have no justification what specific absorption to use. Consequently, the specific absorption used here remains a little arbitrary, the used value of 11.6 is clearly mentioned.

3. Classification of sampled air masses

Please give an idea about the length of these time windows, which will serve for your further calculations.

Authors response: The start and end time of these time windows has been calculated and added in Table 2.

4. Results and discussion

4.1 Aerosol chemical composition

Page 11781, line 8-12: I do not understand this. Why summing up the masses by knowing you will overestimate the total mass. It would be more clear, showing the non-refractory part as an extra column.

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Authors response: In practice, non-refractory material includes species such as ammonium sulfate and bisulfate, ammonium chloride, ammonium nitrate, and volatile organic compounds but excludes black carbon, crustal materials, and sea salt/sodium chloride. Within the discussion of data quality imposed by referee n°1 the aerosol chemical composition chapter has been rewritten. In a first step the AMS mass (organics, sulfate) and SMPS+PCASP mass data (refractory, non-refractory) are presented separately, in a second step we then merge both data sets. Thus, in merged mass contributions we are comparing total mass from volatile aerosol size distributions (spectra at ambient temperature minus those at 280°C) with AMS mass (material volatilized/ionized below 600°C). The corresponding refractory mass (beyond 280°C) is added accordingly. We cannot avoid the fact that the AMS accounts for the material between 280 and 600°C, whereas the mass deduced from volatile aerosol size spectra does not.

In addition, I would like to have this paragraph providing more information on the size fractions you measured.

Authors response: The inlet was designed for particles smaller 800nm in size.

The SMPS was designed for particles smaller 500nm in size. What about the AMS. Was that set to size-selective mode? Please clarify!

Authors response: The AMS PCI (pressure controlled inlet) is designed to keep the pressure in front of the AMS inlet constant to guarantee stable inlet transmission efficiency and stable conditions for particle sizing. The PCI design for the ATR-42 aircraft has been described and characterised in Schmale et al. (2010). The transmission efficiency is 100 % for particles in the range between 200 and 400 nm d_{va}, then the efficiency is decreasing. A suggested approximation (Schmale et al. (2011)) is to take into account an overall transmission efficiency between 80 and 1000 nm of 54 %. D_{va} denotes the vacuum aerodynamic diameter which is equal to the product of the mobility diameter (d_{mob}) with the "Jayne shape factor" and the particle density divided by unit density (DeCarlo, 2004). A second systematic uncertainty is caused by the collection

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efficiency (CE). CE accounts for losses within the standard AMS inlet and lens system, the non-focusing of particularly shaped particles, and the bounce-off from the heater which occurs for certain types of particles. For this study, CE is assumed to be 0.5 due to the lack of comparable aerosol chemical composition measurements. AMS data have been corrected for both, collection efficiency CE and PCI efficiency. The sections 3 and 4.1 have been rewritten according to the requests of reviewer 1.

4.2 CO concentrations

Page 11782, line 25-28: I would like to see some averaged numbers on this finding giving evidence that CO levels can distinguish between air mass origins.

Authors response: CO enhancements are presented in Table 2. We only state that encountered air masses of different origins revealed different CO enhancement. Of course we (do not and) cannot conclude from CO enhancement levels on air mass origins.

Page 11782, line 4: ... and the related "source strength" ...

Authors response: Spelling corrected.

Figure 6a and 6b: Take different colours here.

Authors response: Figures have been deleted due to comments of reviewer 1.

4.3 Aerosol size distributions

Page 11782, line 21: If you say raw aerosol size distributions, what do you mean? In order to get real number values, you have to apply an inversion scheme. Please clarify!

Authors response: Raw number size distributions just mean non fitted number size distributions after inversion (of electrical mobility spectra).

As discussed in the abstract section, please always specify the parameter in question. A size distribution can be a number, area, volume, mass, etc. size distribution!

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We specified the parameter in question more frequently, nonetheless avoiding to be repetitive, where not necessary. With regard to your combination of a SMPS and a PCASP, please again comment on how you adjusted for different diameters! See the authors' answer above.

Page 11783, line1: ... "because of its mounting position outside the plane" ... What do you mean, does it say that other instruments inside were not measuring the truth?

Authors response: The CAI inlet designed for the ATR-42 allows for sampling total submicron and with an upper 50% sampling efficiency for particle sizes at $d = 5 \mu\text{m}$ (McNaughton et al., 2007; Gomes et al., in preparation). For this study the transfer function (the size dependent sampling efficiency) of the CAI inlet for aerosol particles is a minor issue since the AMS is not analysing supermicron particles. The uncertainties are negligible in the submicron size range, where size spectra are compared to AMS data. Thus, for the size distributions (to be compared with AMS) we decided to use preferentially SMPS+PCASP since the PCASP size bins are better resolved as compared to the GRIMM OPC. This is now better clarified in the text.

Table 1: You have a few values, which look like being set by the fitting routines, please comment on this!

Authors response: The fitting of number size distribution has been entirely checked. When not necessary we abandoned the fourth mode, thus, just using three modes. However, parameter which still stuck to the boundary are always very low and thus, implicating no major changes in the number or volume distributions.

Figure 7: These figures should be larger and a more clear colour code is needed to distinguish between the different time windows.

Authors response: Figures have been improved.

Page 11784, line 3-5: Is this statement reasonable for an air mass that has been transported for about ten days? The statement is rather weak and funded on the findings

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(number, diameter) concerning the Aitken mode particles.

Authors response: Given the case that FLEXPART yielded reasonable results, the transported particles obviously stayed small in size, thus there should neither have been large amounts of coating to lead to significant particle growth, nor coagulation should have reduced tremendously the Aitken mode number. Additional pollution sources during the transport over the Atlantic are not likely to explain the rather small particles. The sentence has been changed accordingly.

4.4 Aerosol volatility

Figure 10: Give a legend for the colour code of this figure!

Authors response: A legend has been added

It would be great to see if there is any link between volatile volume fractions and aging. Have you looked into that?

Authors response: An entire paragraph has been added in the "results and discussion" section with an extended discussion of aerosol size distributions and masses of refractory and non-refractory aerosol as been recommended by referee n°1. Unfortunately, there is no clear correlation between air mass ages and volatile volume fraction. It might be due to the fact that after so long transportation times almost all the available gases were already condensed.

4.5 Aerosol absorption properties

Table 2: It would be nice to see that comparison also for the other time windows for a general comparison.

Authors response: We changed fourth to fifth column in figure caption of Table 2. It is not in the scope of this study to compare NA air masses to air masses from other non-pollution and pollution origins like Asia, etc...

Conclusions

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Page 11787, line 22-24: What do you mean when saying "the accumulation mode is less important". It is less important for what? If you talk about scattering efficiency over a white surface (snow, ice) then say it or do you mean it is less pronounced?

Authors response: The sentence has been changed to: The accumulation mode is less pronounced as compared to Fiebig et al. (2003) and Petzold et al. (2007) and is mainly composed of sulphate and more volatile organics. We'd like to point out that only very few data on submicron aerosol size distributions have been published for long-range transport plumes above Greenland. Unfortunately, delta CO are available for Petzold et al. but not for Fiebig et al.. At least we have to keep in mind that delta CO values for the Petzold size spectra on average have been larger by a factor of almost 5 (varying between 3-10) compared to this study. This of course reduces effective differences in figure 8 by the above factors. Therefore we added figure 8b, to compare size distributions of Petzold et al and of this study, both normalised with their respective delta CO.

Page 11787, line 24-26: Wording! The Aitken mode is not derived from SMPS ...
Authors response: The sentence has been changed to: Absorbing aerosols, i.e., black carbon, are found to be predominantly contained in the pronounced Aitken mode which is identified as well in SMPS as in NVSMPS size distributions, yielding ambient and refractory aerosol size distributions, respectively.

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

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