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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Anthropogenic and forest fire pollution aerosol transported to the Arctic: observations from the POLARCAT-France spring campaign" *by* B. Quennehen et al.

**B. Quennehen et al.**

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Anonymous Referee #2 Received and published: 29 March 2012

This study presents airborne observations of aerosol properties in different air masses encountered over northern Scandinavia. European, Asian, Anthropogenic, and natural aerosol plumes are contrasted. The authors use model tools to try to explain observed differences in the aerosol characteristic.
There is yet a shortage of information about aerosol properties aloft and how these properties evolve during transport. As such there is always room for an observational study such as this. The strong section of this study is the relation between the model



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Interactive Discussion

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estimated “age” of the air and the observed properties. This has a great potential for exploring new, but also perhaps older data. Linking in-situ observations with numerical models like this will be very useful. Weak points are speculations used to try to explain the data. Revisiting the same plume is not a Lagrangian study, and the authors actually contradict themselves in the interpretation of the data.

*General answer: We thank anonymous referee #2 for her/his interesting and valuable comments. The manuscript has been modified following her/his and referee #1 and #3 recommendations. For each point, a specific answer is given.*

Abstract contains a listing of observations, but no scientific punch line. No statement of what implications data might have or how the observations might be useful.
*Authors' answer: Modification of the abstract was also recommended by referee #3. The abstract has been shortened and modified as suggested by referees #2 and #3.*

Introduction P4543, L25- It is not clear if the authors describe the phenomenon Arctic Haze (that only occurs a certain time of the year) or sources of pollutants to the Arctic in general. In the latter case non-land emissions is of interest as well.

*Authors' answer: It is Arctic Haze that is describe on page P4543, line L25. The sentence has been rewritten as follows: “Arctic Haze originates...”*

P4544, L5 True, but a broad distribution can make be effective in scavenging the smallest particles.

*Authors' answer: This statement has been added to the manuscript.*



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P4544, L15 Dilution would rather be mixing. The air that is used to “dilute” also has a characteristic aerosol.

*Authors’ answer: The sentence has been modified accordingly.*

Instrumentation P4546, L5- Which inlet was used, the backward facing inlet (estimated upper cut-off?), or the CVI as aerosol inlet? Are data from both inlets used, if so how might the results be affected by the different inlets (SMPS, OPC)?

*Authors’ answer: The CVI inlet was used without counterflow as an isokinetic aerosol inlet for the clear sky measurements (which are the subject of this study), while the CVI inlet with counterflow was used in cloudy conditions. The backward facing inlet has not been operated within this campaign.*

P4546, L15- How was the size distribution treated in the overlap region (with respect to size), or more precisely was the SMPS data dependent on the OPC size distribution for the inversion?

*Authors’ answer: In the overlap region, we only consider the SMPS measurements, since we are very confident in the SMPS and because of their finer resolution. In addition, concentration comparisons have been made in the overlap region showing good agreement between SMPS and OPC concentrations. The following sentences have been added to the manuscript: “In the overlap region, only SMPS measurements were considered. Comparison between OPC and SMPS concentrations in the overlap region show good agreement.”*



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P4547, L19 . . . running average over 180 (units?). . .

*Authors' answer: The unit (s) has been added.*

Some PSAP instruments are very sensitive to rather fast variations in the relative humidity that is sometimes experiences in aircraft measurements (in and out of clouds or vertical stratification etc). Was this observed, or any special precautions taken with respect to this issue? Bond et al., 1999 is given as reference, does that also mean that the PSAP data was corrected for light scattering aerosols?

*Authors' answer: RH measurements were performed upstream (flow splitter) the PSAP. At least during the 3 considered flights, the measured RH never reached values higher than 20%. This is mainly due to the temperature difference between sampled air outside the cabin (0° C and below) and inside cabin temperature of approximately 20° C.*
*As indicated in the manuscript we didn't correct the PSAP data for light scattering (no direct measurement of light scattering). Since the spot size has not been measured explicitly for the specific PSAP, we have used the spot size value of 19.23 mm2, suggested in Müller et al (2009). We consider this value an average value for PSAP instruments. The flow rate has been measured (critical orifice) and calibrated. Since the correction of the absorption coefficient for the scattering is in the order of a few percent, the conclusion on Fig8b remains unchanged*

P4548, L8 What is really meant by "function" here? I suspect that ON/OFF was decided on while making the measurements given current ambient conditions, or was this a post flight selection routine?



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*Authors' answer: Referee #2 is right, the decision of ON/OFF was made during flight by an operator on-board the aircraft using real-time measurements available inside the cabin. The number of impactor samples taken per flight depended for example on how many stacked levels in different altitudes were eventually performed in identified polluted air masses. Thus the duration of sampling was mostly limited by the time allocated for each level flight. Especially, the duration of cloud residual sampling was mainly a question of how long the aircraft could stay continuously in a cloud patch. As a consequence, the number of samples collected per flight, as well as the duration for each sampling fluctuated remarkably depending on the ambient conditions (e.g. free troposphere vs. MBL, clean vs. polluted, cloud free vs. cloudy).*

P4548, L21 How was this random procedure performed?

*Authors' answer: It certainly doesn't mean that the analyst subjectively or preferentially selected and analyzed only a certain type of particles. What is meant here by random procedure is the fact that several field of views (each containing a number of particles) were picked up randomly over the sampling substrate and every single particle found within each field was analyzed. This procedure was repeated until the total number of analyzed particles per sample summed up to > 30 particles.*

P4550, L15 The "typical polar background" distribution, how did you arrive to this? In what way is this typical, for this time of year, at this altitude, or this latitude?

*Authors' answer: This distribution corresponds to a sampling period with local origins (as determined by the FLEXPART model) at the same flight altitude as the encountered pollution plumes mean altitude (∼4000m). It is a background distribution valid for the measurement position (lat, lon) at this altitude, and at the time of the year in the Arctic, when the measurement campaign took place.*



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P4550, L22 “This evolution is explained. . .” Is not supported in anyway at this part of the document and should be removed. With diurnal variability, a semi-Lagranian approach such as this would at least have to compare distribution at equivalent times the different days.

*Authors' answer: The sentence mentioned has been removed.*

P4551, L8- In what way is Aitken and Accumulation mode defined in this study? Simple estimates based on the data given do not support the statement that coagulation is a key player for the reduction of accumulation mode particles. The 11th April have fewer but larger particles, which show a tendency of nearly 20 % more volume presumably acquired over the last 2 days. The reduction in number density is about 400 particles per cc. Perhaps 25% of that reduction can be explained by coagulation (assuming a coagulation constant of 1e-9 1/s). In Figure 3 around 0930, there is a feature which I would interpret as cloud processed air. If the authors disagree (based on the statement that no clouds occurred during the transport), I suggest adding some comment in the paper with respect to this feature in the data.

*Authors' answer: In case 1 study (European anthropogenic plume), Aitken and accumulation modes are defined for diameter ranges of 20-90 nm and 90-500 nm, respectively. These ranges have been used to constrain the parameterization (fitting log normal modes) of the modes. The modal diameter ranges of case study 1 and 2 (Eurasian polluted plumes) have been added in the manuscript in Table 2.*
*The cloud processing of the air mass has been investigated using the ECMWF cloud cover (vertically resolved) and precipitation products. These ECMWF analysis show that, while no precipitation occurred during the air mass transport, particles possibly may have been processed in clouds on April 10 and April 11. A figure has been added to the manuscript (Figure 9 in the revised manuscript, see below Figure 1) to support this statement. The respective paragraph in section 3.1 of the manuscript has been*



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*rewritten, also adding figure 6). In particular, the sentence that no cloud formation and precipitation scavenging took place (p 4551, line 12-13), has been deleted since we may rule out precipitation scavenging, but not the possibility of aerosol transformation (processing) in non-precipitating clouds (based on ECMWF products).*
*With respect to the particular feature in figure 3 around 9h30 we have no explanation. First guess would be that we hit a small cloud, thus creating error counts due to cloud droplet break-up in an aerosol inlet. Looking at FSSP cloud spectrometer and 2D-C ice particle imager there is absolutely no cloud! The peak in the SMPS spectrum happened at the moment when the aircraft started to climb. It's also impossible that the aircraft had hit its own exhaust! Furthermore the aerosol Lidar data (see Figure 2 in this document) do not suggest cloud presence. For us it could be electronic noise that created the spike on one single SMPS size scan, when scanning between 350-450 nm. We neither wanted to delete the scan, nor filter/edit the data for this spike. By the way, the spike can be seen also in figure 4 of the manuscript. this does not impact the study since we are working with the fitted functions.*

P4551, L20- Did you consider that particles heated, may end up just below the 20 nm detection limit?

*Authors' answer: Yes, we are aware of the fact that heated particles may end up below 20 nm. In particular, we cannot document the evolution of Aitken mode particles when heating these particles to 280°C. A large fraction of these particles drops below 20 nm in diameter and thus, is not seen by the VSMPS. Thus, we only study the behaviour of accumulation mode particles passing through the thermodenuder. The resulting modal mean diameters of ambient temperature accumulation mode particles are 43.4 and 58.9 nm. The calculation of the ratio Fconc,ac is using the log-normal parameterisations of the "accumulation mode particles" before and after thermodesorption. In conclusion we think that our study of accumulation mode*



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*particles is not biased by particle diameters falling below 20 nm after thermodesorption.*

P4552, L13- The conclusion that the plume was sampled at different conditions and that dilution is an important process for the interpretation of these data, feels as a contradiction to several other statements in this manuscript.

*Authors' answer: The statement here is that the pollution plume is not homogeneous. The Lagrangian matches support that in general we sampled the same plume on April 9th, 10th, and 11th. However, depending of the spatial collocation of the flight track with the plume (center or border of the plume), we may have sampled the plume edges, etc. . ., The manuscript has been modified to make this statement more visible.*

P4552, L20- The speculative statement about gravitational settling is one of several in this manuscript that is ill founded. The terminal velocity of an accumulation mode particle (assuming BC is mainly associated with this mode) is on the order 1 micrometer per second.

*Authors' answer: The sentence of the mentioned speculative statement has been removed.*

P4553, L15- I'm a little confused with the numbers in the text and the references to figure 8 and table 2 (and Table 1?). But more interesting is that I find the coagulation calculation results surprising. First a large drop in simulated accumulation mode number density, which is not captured in the observations. Then the opposite occurs for the following day. As stated by the authors on the previous page, sampling is done in the plume with different characteristics (not Lagrangian) so this type of exercise have limited use. My impression from the simulated coagulation is that the mass is not conserved and that some dilution must be assumed.



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*Authors' answer: The text has been made clearer. The coagulation calculations were done with mass conservation, however subject to the restriction that for larger diameters than the last bin diameter the mass is indeed lost. The cloud processing mentioned in a previous comment and now mentioned in the manuscript could partly explain that the drop in accumulation mode simulated by the model was not observed, particularly not in the plume sampled on April 10. This would favor the idea of possible cloud processing of the air mass sampled during April 10. Moreover, at least the second plume sample for April 10 indicates that the plume was sampled nearer to the center of the plume than was the case for the other samples, which may be concluded from figure 8a showing higher ïĄĎCO values for the second plume sample of April 10. This may explain to some extent the significant measured accumulation mode compared to that simulated (simulation starting from the April 09 measured distribution) for this day.*

P4556, L17 "These assumptions. . ." I don't follow this sentence.

*Authors' answer: We now denote air masses (i), (ii), and (iii) as (RF) for Russian Fires, (AF) for Asian Fires and (AA) for Asian Anthropogenic. The sentence has been reworded as follows: "These assumptions are based on Adam de Villers et al. (2010) who discussed these two air masses (AF) and (AA). Both air masses contained a mix of biomass burning and anthropogenic contributions. In their study, Adam de Villiers et al. (2010) found evidence on the different intensity of the anthropogenic contribution in these two air masses.".*

P4557, L1 The hypothesis listed needs more explanation. The bottom line is to suggest reasons for why there are more Aitken mode particles in the European Plume



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vs the Asian plume.
1) This implies that there were a lot of Aitken mode particles in the Asian plume, but they grew to become accumulation mode particles in 5 days. Asian particles are larger, but they are also fewer. Would a case with a strong surplus of condensable vapors not also make more particles?
2) More gives less? It needs rather exotic size distribution to arrive at less by starting with more, through coagulation.
3) Plausible, as a large condensational sink would suppress secondary nucleation.
4) What is meant with large scale here? Time, space, number of particles?

*Authors' answer: We agree with referee #2, and therefore entirely abandoned hypothesis 1 and 2. The final part of section 4.1 has been entirely modified as suggested by referees #1 and #2 to better match the above suggestions.*

P4557, L17 "Concentrated" As you are to discuss chemistry, this word may be ambiguous in this context.

*Authors' answer: The term "particle" has been added in order to make clearer the context.*

P4558, L1-L25 Perhaps this section can be shortened to highlight some particular aspects, but refer to tables for all the numbers.

*Authors' answer: Section 4.2 has been modified to better match with the end of section 4.1. In addition, the section has been shortened and made clearer.*



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Conclusions This section must be split in one summary and one conclusions section with perhaps a couple of bullet points. I'm not convinced about the interpretation with respect to coagulation vs condensation.

*Authors' answer: The final section (former section 6 of conclusions) has been rewritten and splitted into 2 separate sections (summary+discussion & conclusions).*

Figure 2 The small numbers in the figures are hard to see. I suggest replace with fewer but larger or only keep the ones with arrows.

*Authors' answer: We only keep the numbers with arrows in the revised version of the manuscript.*

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 4541, 2012.





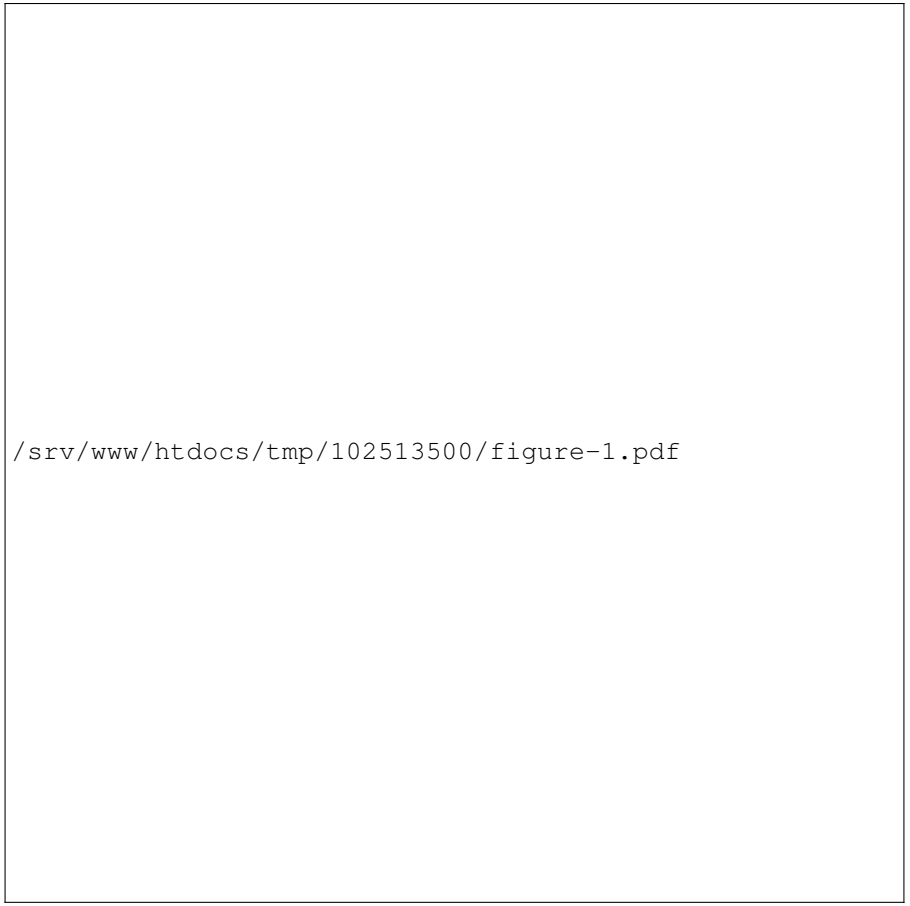


**Fig. 1.** Figure 9 (in revised manuscript): ECMWF could cover profile along the HYSPLIT back-trajectories starting from the anthropogenic pollution plume measurements on 10 and 11 April 2008



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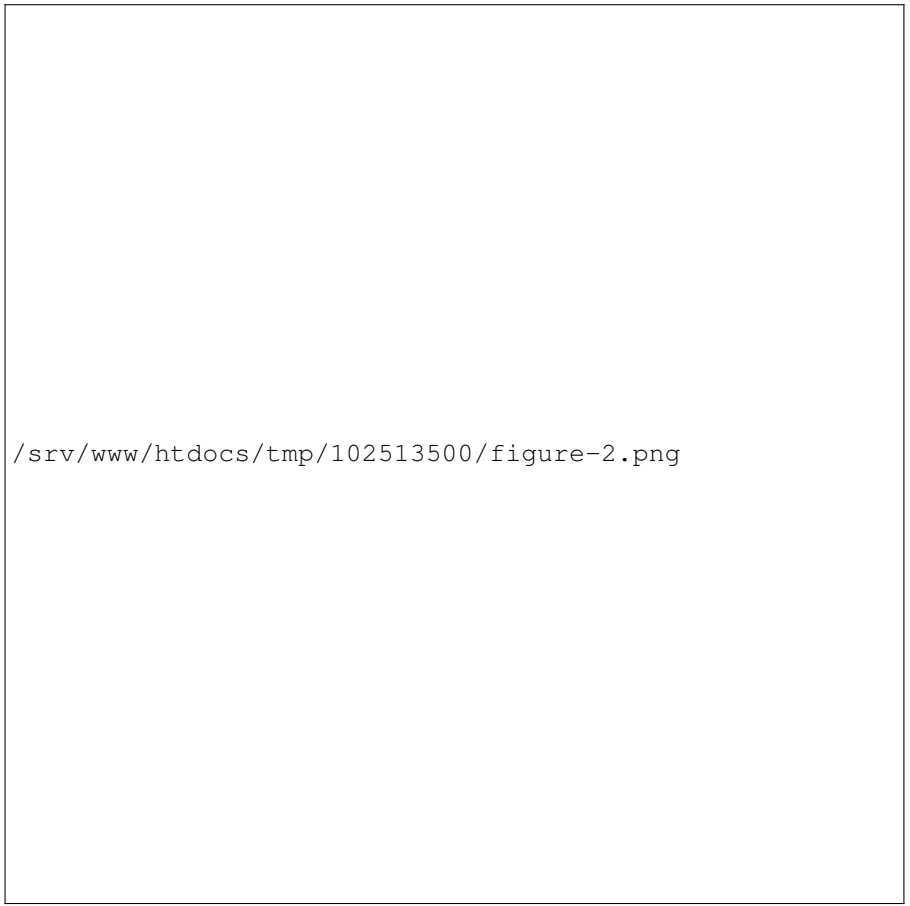


**Fig. 2.** Lidar on aircraft: plot between 09:08 and 09:45 for April 09, 2008.



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