Atmos. Chem. Phys. Discuss., 12, C2867–C2874, 2012
www.atmos-chem-phys-discuss.net/12/C2867/2012/

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.

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Received and published: 23 May 2012

Anonymous Referee #1 Received and published: 12 March 2012

This manuscript presents analysis long-range transported fire smoke emission transport to the arctic based mainly on air craft measurements. The paper focuses on two case studies. The text is quite well written and most parts of it can be considered scientifically sound. The paper also fulfills the originality requirement. In my opinion, the paper is worth to get published in ACP after the authors have addressed the following, mostly minor issues.

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

Scientific issues:
The last paragraph of section 2.2 and interpretation of figures 5 and 7: It remains a bit unclear how the "age" of air masses has been determined. Especially, how the zero point of air has been obtained for different air transport situation. I suppose one needs somehow to define the dominant source area of measured air masses? How this is handled when multiple sources affect the measured air? More explanation is needed here.

Authors' answer: As mentioned on page 4549 line 6-10, "the FLEXPART model was repeatedly initialised along the flight trajectories for aircraft position changes of more than 0.20° in horizontal dimension and/or 150 m in vertical dimension. For every single run, 40000 particles were released from a small volume around the aircraft position and then were tracked during twenty days (backward in time).". When considering a single run, the position of each particle within the 40000 at each time step backward in time (up to 20 days) is known. Thus, when a particle reaches the ground (or the lower vertical layer of the model for biomass burning emission areas), it is considered that it reached its emission point. The time between such emissions and the measurement periods is corresponding to the FLEXPART age of the particle. When the 40000 particles are considered, an "agespectrum" is derived. The integration of this "agespectrum" leads to the estimation of the FLEXPART age as described in section 2.2 and as used in Figs 5 and 7.
The continental source apportionment is also derived from the FLEXPART backward simulations. It expressed the main continental origins and the main type (anthropogenic or biomass burning) of air masses. For the first case study (the one that use

the FLEXPART age), the combination of the two products discussed above and the footprint potential emission sensitivity indicate that Central Europe is the main source region of the studied air mass. In the case of multiple sources, the FLEXPART age must be considered carefully.
The manuscript has been modified using the above explanation.

Page 4551, lines 3-5: It is extremely dangerous to extrapolate the particle size backward to the emission point as done here. Different processes can affect particle growth rates, and in very different ways, during different stages of atmospheric transportation.

Authors' answer: The sentence has been removed and replaced by the followings: "During the measurement period (9 to 11 April), the mean modal mean diameter of the Aitken mode is increasing exponentially. Because of the different processes affecting particle growth rates in very different ways, during different stages of atmospheric transportation, the exponential parameterization presented here however, becomes more and more uncertain when extrapolating the modal mean diameter backward close to the emission region.".

Page 4554, lines 23-24. The authors should state more clearly what they mean by the Aitken mode being affected more than the accumulation mode by condensation. Since they refer to eq. 4, I suppose they mean the particle diameter growth rate, which indeed is expected to be somewhat larger for the Aitken mode. Many people might be more interested in secondary aerosol mass formation by condensation, and this is usually dominated by mass flux into the accumulation mode (or coarse mode if substantial amounts of sea salt or dust are present).

Authors' answer: Referee #1 is right, the particle diameter growth rate is considered

here and this has been clarified in the manuscript.

The last statement of section 3.4: I do not buy this explanation, as modeling the coagulation process is expected to have relatively low uncertainties. I would rather seek for a process other than coagulation or condensation explaining the observed shift in the accumulation mode. How about cloud processing which is know to add material into this mode effectively in the atmosphere?

Authors' answer: Using the ECMWF cloud cover product, it appears that HYSPLIT back-trajectories related to the anthropogenic plume indeed experienced cloud processing (without precipitation) within the 2 days preceding our measurements. The manuscript has been modified to take into account the possible contribution of cloud processing to the evolution of the aerosol size distribution. (see Figure 1 below, the figure has been added and is discussed in the revised manuscript).

The authors should make some interpretations of the relatively large volatile fraction of about 80 per cent. How this value compares with observation made by others and what it reveals about the aging of measured particles?

Authors' answer: the aerosol volatile fraction at 280° C of such anthropogenic air mass transported to the Arctic free troposphere, to the authors' knowledge, has never been studied. Recently, Häkkinen et al. (ACPD, 2012) presented results from long-term non-volatile aerosol fractions at the ground based station of Hyytiälä. Their results support our findings, since Häkkinen et al. found that during spring season, the main source of aerosol was anthropogenic with a non-volatile aerosol fraction of 0.17. In an urban background study, Birmili et al. (ACP,2010) found a non-volatile aerosol fraction of 0.31.

Clearly, the small non-volatile fraction observed indicate that particles are mostly composed of volatile condensable material, with potentially significant amounts of secondary particular material. A comparison of the observed volatile fraction with previously observed values and a short discussion have been added to the manuscript.

The discussion in section 4.2 is a bit difficult to follow and should therefore be improved. Basically, the mean size of particle modes any measurement point results from the combination of two things: 1) the means size of particles at the emissions, 2) the processing of particle during atmospheric transportation. The first of these depends on the source type and in case of fire emission also with the burning conditions. The second one depends on the initial particle number concentration and size distribution (coagulation), the amount of aerosol precursors (condensation, cloud processing, nucleation) and the time available for these processes (transport distance). The analysis should somehow be arranged along these points. Now it is very difficult to see whether and how section 4.2 supports the hypothesis 1 (page 4557, lines 2-3) as claimed in lines 9-11 on page 4557. Why cannot the authors test the hypothesis 2 with their coagulation model similar to what was done in section 3.4? One could easily give different numbers of Aitken mode particles at the initial condition and simulate the time evolution of the system by coagulation.

Authors' answer: Since referee #2 suggested that hypotheses 1 and 2 might be unrealistic since (i) a more significant condensation process would produce more accumulation mode particle in an Asian air mass than in European ones (which is not the case) and (ii) only non-realistic particle size distributions would justify hypothesis 2. Therefore, hypothesis 2 has not been tested with the coagulation model. Overall, the end of section 4.1 has been entirely reworked in order to fulfill the recommendations of referee #2, and thus referee #1. Furthermore, section 4.2 has been shortened and made clearer as requested by referees #1 and #2.

Technical issues: The instruments measuring the aerosol size distribution (or size fractions) rely on at least 3 different particle diameters. This should be brought up and explained in section 2.1.1 and perhaps elsewhere in the text.

Authors' answer: 3 different particle diameters are introduced: geometric, electrical mobility and optical diameters related to electron microscopy, SMPS and OPC measurements, respectively. However, if the measurement techniques are different, the diameter types can be translated into each other, of course under certain assumptions (refractive index, sphericity, DMA transfer function, etc...). 2 sentences have been added to the text to present the different particle diameters.

Page 4552, line 10: Figs. 7, 7 and 7? Should be either Fig. 7 or Figs. 7a, 7b and 7c.
Authors' answer: References to Figs, 7a, 7b and 7c have been corrected.

There is something strange in the way the paper by Adam de Villers et al (2010) is either referred to or located in the reference list. Should it read "de Villers et al."?

Authors' answer: The last name of the author is Adam de Villiers. Therefore, the reference is now in correct alphabetic order.

References:
Birmili, W., Heinke, K., Pitz, M., Matschullat, J., Wiedensohler, A., Cyrys, J., Wichmann, H.-E., and Peters, A.: Particle number size distributions in urban air before and after volatilisation, Atmos. Chem. Phys., 10, 4643-4660, doi:10.5194/acp-10-4643-2010, 2010.

Häkkinen, S. A. K., Äijälä, M., Lehtipalo, K., Junninen, H., Backman, J., Virkkula, A., Nieminen, T., Vestenius, M., Hakola, H., Ehn, M., Worsnop, D. R., Kulmala, M., Petäjä, T., and Riipinen, I.: Long-term volatility measurements of submicron atmospheric aerosol in Hyytiälä, Finland, Atmos. Chem. Phys. Discuss., 12, 11201-11244, doi:10.5194/acpd-12-11201-2012, 2012.

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

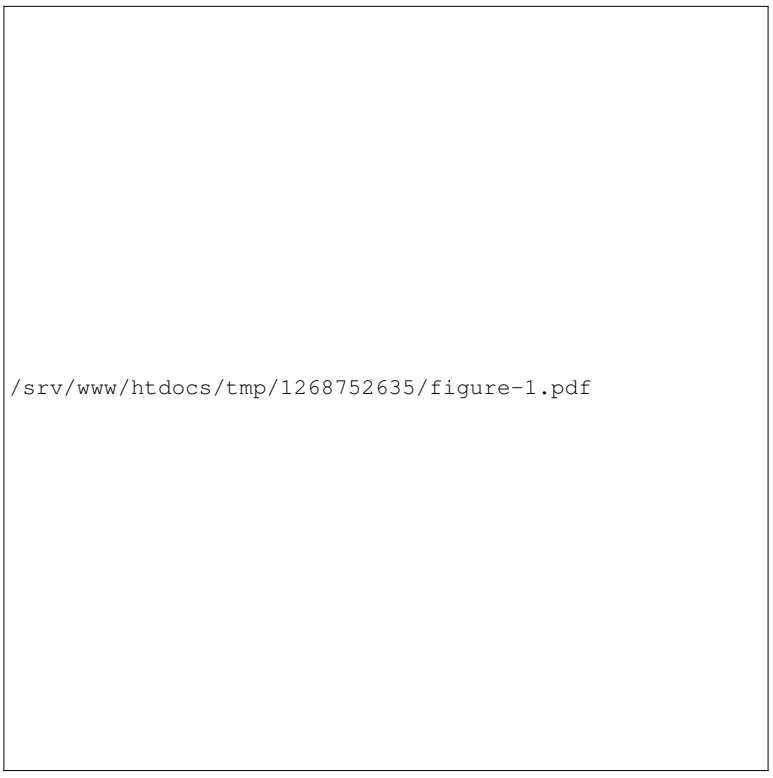

Fig. 1. Figure 9(in revised manuscript)- ECMWF could cover profile along the HYSPLIT back-trajectories starting from the anthropogenic pollution plume measurements on 10 and 11 April 2008