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Comment

Interactive comment on “Airborne observations of aerosol microphysical properties and particle ageing processes in the troposphere above Europe” by T. Hamburger et al.

T. Hamburger et al.

thomas.hamburger@itm.su.se

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We thank the reviewer for her/his valuable comments on the manuscript. The reviewer's comments are in regular type and our responses are outlined in italic type.
General

The paper describes vertical and horizontal measurements of aerosol number size distributions and aerosol volatility in central and northern Europe in a very wide area ranging from the Atlantic Ocean west of Ireland to the Baltic states in the east and from northern Italy in the south to southern Finland in the north and vertically from the

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lower boundary layer up to stratosphere. The volatility measurements tell about aging processes and the authors have combined this information with estimates of aerosol source regions and emission strengths. All this information is very valuable, especially for evaluating various models. The paper is definitely suitable and worth publishing in ACP. It is mainly well written the amount of graphs and tables is almost ok but I did find some issues that I did I wish you could answer and/or modify.

Detailed comments

p. 20387, L 3 – 6, “Airborne measurements mostly cover areas on a regional scale (Ansmann et al., 2002; Minikin et al., 2003) or focus on certain altitude levels (CrumeYrolle et al., 2010; Voigt et al., 2010). “ You should also refer to the CARIBIC project and tell what there has been done.

We added the following sentences to p. 20387, l 3-6 to consider the CARIBIC project:

Airborne observation campaigns mostly cover areas on a regional scale (Ansmann et al., 2002; Minikin et al., 2003) or focus on certain altitude levels (CrumeYrolle et al., 2010; Voigt et al., 2010). Long-term airborne observations mostly of the upper troposphere/lower stratosphere have been carried out within the CARIBIC project on a monthly base using civil aircraft on transcontinental flights (Brenninkmeijer et al., 1999; Brenninkmeijer et al., 2007).

Section 2.1. I did not find an information on how many flights were there altogether. What time of the day were the flights? Time of the day affects a lot of things so that information should also be carried to the discussion of the results.

Detailed information on flights and flight times can be found in McMeeking et al.

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(2010) or Hamburger et al. (2011). To give some information on the observation flights in the present manuscript we added the following lines to p. 20388, line 20 and added Table 1:

A total of 15 DLR Falcon 20 flights and 16 BAe-146 flights were performed within the EUCAARI-LONGREX campaign (Table 1). Thereby 17 flights were performed during the morning hours and at noon time and 14 flights during the afternoon hours (McMeeking et al., 2010; Hamburger et al., 2011). Thus, the retrieved data span the whole daytime.

P. 20389, L 9 – 11. “The regions for the vertical profiles were limited by ATC like the air routes. Thus, vertical profiles frequently had to be performed during take-off and landing and in Temporary Reserved Areas (TRA)”. Do you have any idea whether the profiles would be significantly different outside the TRA?

This is an interesting point. However, no evident effect concerning whether the observations took place during take-off and landing or within TRAs was observed. Due to the dense population in the region of interest no difference between inside or outside a TRA should occur. In addition, TRAs themselves are restricted areas in the troposphere above the ground and not necessarily related to e.g. restricted military areas on the ground. I.e., “common” aerosol sources can be found also on the ground of TRAs.

Section 2.2. Measurements

I did not find any discussion on the uncertainties of the instruments. For instance, you had several size distribution measuring instruments with overlapping size ranges. In my experience, they never agree within 1%. How about in your measurements? I would suggest adding at least one scatter plot where the sums of the number

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concentrations in the overlapping ranges are shown. And how about the different aircraft: you had similar instruments at least for some parameters – how did these agree? Then about the CPSA: what were the CPCs? I did not get the paper referenced (Stein et al, 2001), it is a conference proceedings, more difficult to get access. Did the CPCs actually all the time show concentrations in the right order, i.e., $N_4 \geq N_{10} \geq N_{14}$? I have also done similar stuff with TSI CPCs and there are often cases when the order is not that and that means something is wrong. Write some discussion on this.

Considering the observations on different aircraft, PCASP data from two inter-comparison flights, a dual aircraft intercomparison with the instruments mounted on both aircraft separately and a single aircraft intercomparison with the instruments mounted on the BAe-146. The total concentrations measured by the two instruments matched well during both flights (Figure S1), the aerosol size spectra showed reasonable agreement (Figure S2). The detailed analysis of the aircraft comparison is an interesting and important topic. However, this technical question exceeds the scope of the present manuscript.

We added the following note to the methods section, p. 20390, line 14:

Both PCASP-100X were compared during two intercomparison flights. The comparisons showed very good agreement in the total number concentrations and good agreement in the aerosol size spectra.

To clarify the use of the CPSA and CPC we edited p. 20389, lines 24-28 and p. 20390, lines 1-2:

Particle number concentrations were measured aboard the DLR Falcon 20 using a custom build condensation particle size analyser (CPSA) (Fiebig et al., 2005; Feldpausch et al., 2006) and a condensation particle counter (CPC, TSI model 3760). The CPSA consists of a battery of four condensation particle counters which can be

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set to different lower cut-off diameters. Two channels of the CPSA at lower cut-off diameters of 4 and 10 nm were used for the present analysis. The TSI CPC was set to a lower cut-off diameter at 14 nm. Number concentrations of ultra-fine condensation nuclei were defined as the number concentrations of particles with diameters between 4 and 10 nm. The TSI CPC was used in combination with a thermodenuder to measure the number concentration of non-volatile particles.

All instruments undergo a calibration before and after the aircraft field campaign and routine checks during the campaign to assure the quality of the observations or detect possible malfunction. In addition, the instruments are monitored online during the measurement flights. Routine checks of the CPCs on ground and in the air with the thermodenuder switched off show the right response ($N_4 \geq N_{10} \geq N_{14}$). Comparisons of the optical probes showed also good agreement within the overlapping size ranges (Figure S3). We added the following information to the manuscript, p. 20390, line 27:

The optical particle spectrometer probes were calibrated using monodisperse aerosol particles of various non-absorbing substances with known refractive index. The used calibration aerosol consisted of Di-Ethyl-Hexyl-Sebacat (DEHS), polystyrene latex (PSL), ammonium sulphate and sodium chloride.

Section 2.3

P 20392 – 20393 You explain about calculating the normalized emission factor EN. I find this discussion much too qualitative. For example, how do the different compounds affect the value of the EN? There should be a formula so that also other people can calculate the same thing. Has the formula been presented in some other paper? If not, write it here. You could add another appendix for that.

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The normalized emission factor E_N was introduced to define regions rich (or poor) in ground based emissions. Instead of using a rather coarse classification of different regions (e.g. in Lammel et al, 2003) the authors decided to use a finer resolution and an objective method based on the EDGARv4.1 database. We do not seek to model aerosol processes or compare aerosol model results with our observations within this manuscript (of course, we encourage using the presented observations for model comparisons within further studies!). We use the transport analysis to classify the observations with respect to the history of the air masses above regions with different emission intensities. Therefore we think that the pure qualitative approach is sufficient.

We added the following formulas to clarify the calculation of E_N , p. 20393, line 6:

The normalized emission factor E_N at the respective grid cell i can be derived as follows::

$$E_G = \{x \mid x \in \{E_{S,0}, \dots, E_{S,n}\} \wedge x > 0\}, \quad (1)$$

$$E_{P99} = P99(E_G), \quad (2)$$

$$E_{N,i} = E_{S,i}/E_{P99}. \quad (3)$$

$E_{S,i}$ represents the sum of the source types at grid cell i . E_G is the global set of all $E_{S,i}$ not zero and E_{P99} the 99th percentile of E_G . Finally, E_N at the grid cell i is derived by normalising $E_{S,i}$ to E_{P99} .

P 20393 L12 – 14 “The observed air masses are classified by the time between the measurement and the time of the strongest emission events occurring along the

air mass transport pathway within the last 96 h before the measurement.” OK, this is a simple approach but it disturbs me a bit. Let us say along the air mass transport route there are four ENs, 0.9, 0.8, 0.8. and 0.8. Then you calculate the aging from the 0.9 only. The three 0.8 values most probably have a larger impact on the observations. I have a suggestion that would require a bit work but not much: why don't you calculate the weighted average transport time from them different source regions with $\text{ave}(t) = (\text{sum}(\text{dt}(i) * \text{EN}(i)) / \text{sum}(\text{EN}(i)))$ or something similar? I don't require this, I leave it up to you to consider whether this is a good or a bad idea. Anyway, if you do this, I would expect that the ranges in the results of the aging analyses might get smaller.

Thank you for this interesting comment. The reviewer is right in stating that along an air mass transport pathway multiple sources with different emission intensities appear. However, the final analysis was binned into 12 hourly time bins. If a region features several consecutive high E_N , e.g. 0.9, 0.8, 0.8. and 0.8, the analysis would classify the air mass into the same time bin at a trajectory output of 30 min.

From our experience with the measurements the last strong emission source covered most of the signal from previous emission sources which we could detect with the instruments presented in this analysis. Previous trajectory analysis using a time weighted average of E_N along the transport pathway was used to consider this effect. However, to avoid artificial bias of the selected weighting function and to remain with the simplicity of the presented trajectory analysis we decided to continue with the analysis method described in the manuscript. As we could show even this simplistic analysis method results in valuable information on the average age of the observed aerosol particles.

Section 3.

A general point: I would like to know, how large a fraction of the total aerosol column lies within boundary layer or other altitude ranges in the various regions. That

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data would be useful for instance in evaluating AOD measurements. Your data would show that but I don't find that information anywhere explicitly. Consider whether this would be something to present.

This a very interesting point. To address your suggestion we added the following paragraph to Section 5 (Discussion), p. 20408, line 26:

The dry and stable meteorological conditions in the first half of May 2008 not only allowed an increase of aerosol lifetime but also an increase of the particle load within the planetary boundary layer. The vertical profiles in Section 3.4 illustrate the distinct negative gradient in number concentrations between the boundary layer and the lower free troposphere. Thus, a large fraction of the column aerosol optical depth (AOD) in the troposphere might be related to the aerosol emissions and the subsequent particle growth within the boundary layer. Considering the accumulation mode particles as the optically active fraction in the submicron diameter range for visible sunlight according to the Mie scattering theory, we can use the vertical profiles of accumulation mode particles as an indicator for the relative contribution of the boundary layer and the free troposphere to the fine-mode (i.e. submicron) AOD. The fraction of accumulation mode particles within the boundary layer relative to the total column reached 81-89% during the anticyclonic conditions with lowest values above the Baltic Sea and highest values above the Benelux States. The observed boundary layer height was between 2-2.5km above ground level. The fraction of accumulation mode particles decreased during period (b) to 41-80% with a decreasing boundary layer height (1.5-2km a.g.l.). Number concentrations of coarse mode particles were close to the detection limit within the free troposphere during cloud free conditions. Hence, estimates of a relative fraction of coarse mode particles between boundary layer and free troposphere cannot be done to extend the procedure to the total AOD. However, even though the coarse mode particles in the boundary layer contributed to a large fraction to the total aerosol volume (50%), they contributed to a rather small

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fraction of 8% to the total aerosol surface. The total aerosol surface, being a main parameter of the bulk aerosol optical properties (Brock et al., 1993), was dominated by the accumulation mode.

Another thing I don't find in the results is comparison with ground based observations at the same time at some representative station. You could very easily get that data from the EBAS (<http://ebas.nilu.no/>) data base. Then you could add a few symbols to your plots at ground level (ave, median, a couple of percentiles), for instance in Fig 5, no new figures.

Thank you for mentioning this. So far, we did not mention the comparison of aircraft data with ground-based observations which was done in Hamburger et al. (2011). As the comparison in Hamburger et al. (2011) is quite extensive we will only mention the comparison in the introduction of this manuscript p. 20387, line 21:

In addition, Hamburger et al. (2011) compared the airborne observations to the EUSAAR ground sites at Hyytiälä, Vavihill, Mace Head, Hohenpeißenberg, Melpitz, and Cabauw.

P 20407 L19 – 20 “Coagulation of particles caused an almost entirely internal mixing state... Coagulation plays a minor role here, growth by condensation is the dominant effect.

We changed p. 20407, lines 17-20 to:

The available measuring methods presented in this work enable the analysis of aerosol ageing processes based on coagulation of particles and condensation of precursor gases on pre-existing particles. Both processes caused an almost entirely internal mixing state...

Table 2 could be Table 1. In table 1 you could give also more information on the flights: dates or date ranges, regions etc.

We added Table 1.

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/12/C9615/2012/acpd-12-C9615-2012-supplement.pdf>

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