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Atmospheric Chemistry and Physics Discussions

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

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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 comments
This manuscript provides a statistical summary of European aerosol microphysical observations taken from two aircraft over a period of one month. The data are segregated into two time periods, one of anticyclonic circulation and stagnation, and a second of active frontal passages. The data are geographically separated into four regions and into vertical bins. They are interpreted with the assistance of a trajectory

and emissions model.
The paper is reasonably well written and is easy to understand. The topic is of interest to ACP readers, and the dataset described is unique. The manuscript consists primarily of a recitation of statistical results, with relatively little interpretation and certainly no surprising conclusions. That said, there is some value to the summary that is presented here, as it can provide a 1-month dataset against which models that incorporate detailed aerosol microphysics and chemistry can be compared. Thus I recommend it be published in ACP following revision as described below.

Specific comments

1) The manuscript indicates that "nucleation events" could be observed (e.g., p.20400 line 2). You are actually observed the end product of new particle formation, not the nucleation of stable molecular clusters.

The reviewer is right in stating that we actually observed the end product of new particle formation. We edited the following sentences in the manuscript:

p. 20385, line 8:
Ultra-fine particles as indicators for nucleation events were observed. . .
p. 20400, line 2:
. . . and UCN particles could be observed in the cloud outflow at high altitudes indicating preceding new particle formation.
p. 20406, line 5:
. . . where nucleation events appeared.
p. 20406, line 7:
. . . and only few nucleation events were indicated at high altitudes.
p. 20409, line 3:
Nucleation events were indicated inside the continental boundary. . .

2) p. 20401, lines 1-5. This discussion of sampling issues should be moved to Section 2.2, where the measurement details are discussed.

The respective lines were moved to Section 2.2.

3) p. 20401, line 12, define RH(overbar) as the arithmetic mean RH.

We added the following to p. 20401, line 12:

. . . (RH(overbar)=55 % between 0–3 km, with RH(overbar) being the arithmetic mean of the relative humidity within the given altitude range) . . .

4) p. 20401, line 20. This section discusses decoupled, stable layers of enhanced aerosol concentrations above the boundary layer. However, the only discussion of how the BL is identified has been with respect to the ECMWF gridded data. How was the BL height determined from the aircraft data? Or did you use the ECMWF BL height determined whether samples were in the FT or BL? If so, these decoupled layers might actually be within the BL.

To clarify that the classification of the observations into several layers was based on the airborne observations themselves we added the following to p. 20401, line 18:

Each horizontal sequence was analysed within the respective vertical profile of aerosol properties and meteorological conditions which were observed with the aircraft and classified into one of the following vertical layers: . . .

5) p. 20401, lines 25-27. It is not clear if the lognormal parameters were fitted to each individual size distribution and then averaged, or whether the size distribu-

tions were averaged and then the lognormal parameters fitted. These can produce drastically different results (particularly for standard deviation), with the former method being more correct.

We have edited p. 20401, lines 25-27 to clarify the calculation of the fitted parameters:

The size distributions obtained by PCASP-100X and FSSP-300 measurements were first averaged for each horizontal flight leg. A log-normal number size distribution in the accumulation and coarse mode size range was fitted to each of the averaged size distributions. The fitted size distributions were classified into the respective vertical layer. Figure 6 shows the median and quartiles of each group of fitted size distributions.

6) p. 20402, lines 19-21. I don't fully understand this sentence. Are you suggesting a growth of particle sizes between time periods (a) and (b) in the free troposphere? Surely transport would result in advection of a completely different airmass between these 2, two-week periods–not a temporal evolution of aerosol characteristics.

To make this part more understandable we change the sentence on p. 20402, lines 19-21 to:

Larger particles were more present within the LFT and UFT during period (a) while smaller particles occurred during period (b). This indicates that the meteorological conditions for particle growth were more favourable during period (a).

7) p. 20403, lines 9-19. It is very hard to discern much from the grey-shaded points shown on Figure 7. Would you please bin the data into the regions shown on

Fig. 4 and then produce a box plot showing the changes in age class as a function of longitude? This would more clearly show the spatial variation in age class, as well as the variability.

A box plot showing the changes in age class as a function of longitude would remove the information we obtain from the latitudinal transport from Finland via the Baltic Sea towards the north-east of Germany. Therefore we decided to retain the map. However we changed the grey scale in the plot into a colour scale to highlight the geographical differences in the age classes.

8) What is a "classified sequence", (e.g., line 17 and elsewhere). Is this one of the horizontal stacked legs that comprised the vertical profiles? Please explain your nomenclature.

The expression "sequence" was replaced by "(horizontal) flight leg" throughout the manuscript to maintain a consistent nomenclature.

9) Table 4 (discussed on p. 20404) is hard to follow–there is such a range of literature values that it's difficult to see what is consistent between the current observations and the literature. Is there a clear way to show this graphically (e.g., a plot showing each parameter and the range of measured and literature values)?

The authors agree that the table is not easy to follow. However, a condensed graphical solution did not improve the presentation of the data (see Figure S1). One would have to split the plot into several sub-plots for each component and maybe altitude range to give a proper overview of the comparison between observed and literature data. However, this would increase the already high number of figures within the manuscript. The authors therefore decided to keep the table as a source of additional information.

10) p. 20405 lines 10-11. Surely airmass exchange during frontal passages also explains much of the changes between periods (a) and (b). You seem to focus on vertical transport and removal processes here.

We changed the following paragraph to add the possibility of air mass exchange to the discussion. However, wet removal and vertical mixing play an important role especially within the boundary layer above a region rich in aerosol sources. Here, the uptake of particles and aerosol precursors into the boundary layer is very effective as could be observed during the advection of rather clean air masses from the Baltic Sea across Northern Germany. p. 20405 line 10:

The changing meteorological conditions between period (a) and period (b) resulted in a change of the particle load due to an exchange of air masses during frontal passages. However, the change in atmospheric stability caused a change in vertical mixing of pollutants linked to the surface, too. High UCN number concentrations indicating nucleation events were most frequently observed inside the boundary layer during the dry period (a). In contrast, wet scavenging and convection during period (b) reduced the entrapment of possible aerosol precursors close to the surface and within the boundary layer and inhibited their accumulation. In contrast, destabilisation and convection increased the ratio of volatile matter within the lower free troposphere above the boundary layer.

11) p. 20406 lines 12-16. Are aircraft particles non-volatile? I thought chemi-ionassisted nucleation and condensational growth resulted in many volatile particles (e.g., Schroeder, Kaercher, Schumann).

Both, non-volatile (soot) and volatile particles can be observed in aircraft plumes with volatile components dominating the number concentration (Schröder et al., 1998;

Petzold et al., 1999). The paragraph focuses on ultra-fine condensation particles above 8 km altitude which consist of secondary volatile aerosol matter and states that non-volatile particles featured a minimum in number concentrations above 8 km altitude. Lines p. 20406, lines 8-16 say (we added the two aforementioned citations and edited line 14 by deleting "However, . . . "):

Hardly any nucleation events occurred at low altitudes whereas an increased number concentration of UCN particles could be observed above 8 km. Updrafts during frontal passages and a destabilisation of the atmosphere due to the dissolving high pressure system lead to a transport of ground based emissions and aerosol precursor gases into the upper free troposphere. In addition, aircraft emissions within the highly frequented air space serve as possible particle sources in the upper troposphere (Schröder et al., 1998; Petzold et al., 1999; Singh et al., 2002; Voigt et al., 2010). Secondary aerosol showed an increase of number concentrations at high altitudes, whereas primary non-volatile particles featured a minimum above 8km.

12) p. 20407, lines 22-26. I don't know what you mean by the "static nature" of sulfate. Time scales for gas-phase SO2 oxidation are probably of order of a few days at this latitude and season, so you could expect to see substantial secondary sulfate formation over the aging time scale studied here.

Here we emphasize the relatively more dynamic evolution of organic aerosol particles compared to sulphate aerosol particles as stated by Jimenez et al. (2009). We focus here on the particle phase. We changed p. 20407, lines 25-26:

Organic aerosol undergoes a more dynamic chemical evolution with time compared to the relatively static nature of sulphate aerosol particles. Most organic aerosol evolves due to oxidation to oxygenated organic aerosol (Jimenez et al., 2009).

13) p. 20408, lines 11-16. I had a hard time understanding the point of these sentences until re-reading them. You are saying that synoptic-scale descending motion limited convection so that there was little exchange between the free troposphere and the polluted boundary layer. Please clarify.

The sentences should explain that due to the definition of cloud free conditions which was used for the trajectory analysis only flight legs at low altitudes could be used for the presented analysis method. To make it more understandable we changed the sentences of p. 20408, lines 11-16 to:

Only trajectories where the relative humidity did not exceed 80 % within the last 96 h were selected to avoid cloud processes. Those relatively dry air masses occurred dominantly during the high pressure situation when air masses tended to descend on a synoptic scale rather than to rise. As a consequence, it was mostly flight legs below 3 km altitude that could be connected to ground based emission sources and consequently were selected for the present trajectory analysis method. Thus, almost no cases of uplifted emissions could be included into the analysis.

14) In the abstract, introduction, and conclusion, you state that understanding spatial variability in aerosol microphysical properties is "essential" to improve understanding of aerosol climatic effects. However, you don't really say why this is the case - and this presumption is the motivation for the entire manuscript. Are there studies you can cite that look at the effect of regional-scale aerosol variability of this magnitude on radiative forcing? If so, please discuss and cite.

We added the following to the introduction at p. 20386, line 22:

Considering the direct and indirect effects of aerosol particles on the earth climate a fundamental knowledge of the particle properties and their temporal and

spatial variability in the atmosphere is required. In-situ observations showed that regional variations of aerosol microphysical properties lead to variations in the vertical column aerosol optical depth due to both, direct and indirect aerosol effects (Clarke and Kapustin, 2010). Different aerosol types feature different radiative forcing above various surface types. While primarily light scattering aerosols have a larger impact above surfaces with low albedo such as urban areas, light absorbing aerosols effect the radiative forcing above surfaces with high albedo, e.g. snow-covered mountain regions (Iorga, 2007). The aerosol direct radiative forcing during cloud free conditions above the continent can lead to a radiative cooling of the surface temperature and radiative heating within the planetary boundary layer resulting in a stabilisation of the lower troposphere. A positive feedback arises as the stable lower troposphere increases the possibility of further accumulation of aerosol particles (Pere et al., 2011). Further feedbacks induced by aerosol radiative forcing may also have larger regional effects which show sensitivity to the type and location of the forcing. E.g. the change in short-lived-species like aerosols at northern mid-latitudes contributed to rapid warming of the Arctic climate (Shindell and Faluvegi, 2009). Further studies showed that aerosols, also comprising anthropogenic emissions of aerosols, can drive North Atlantic climate variability (Booth et al., 2012).

15) Please add the dates of the project to the Abstract.

The first sentence of the Abstract includes the date of the project:

In-situ measurements of aerosol microphysical properties were performed in May 2008 during the EUCAARI-LONGREX campaign.

Technical corrections

1) Please define the "Benelux" (Belgium, Netherlands, Luxembourg) States. This is

not commonly used outside of the EU.

The term is now defined in the text.

2) p. 20389, line 25. The instrument is a condensation particle size analyser. However, Stein et al. is a conference abstract and is not a citable, peer-reviewed paper. Please remove.

The citation was changed to Fiebig et al. (2005) and Feldpausch et al. (2006).

3) p. 20389, line 19 and elsewhere (e.g., p.20394, lines 21-27, etc.). Please do not capitalize "South of England" (and, elsewhere, South Germany, etc.). Unless these are formal geographical place-names the geographic word should not be capitalized.

Capitalized geographic words like “South of England” were set to lower-case letters while geographic names like “South Germany” remained capitalized.

4) p. 20390, line 4, change "low volatile" to "low-volatility"

Changed.

5) p. 20392 line 7, change "weather" to "whether"

Changed. Thank you for spotting that.

6) p. 20395 lines 16, etc. Do not capitalize "mean", "median", or "quartile".

Changed.

7) p. 20397, line 3 and elsewhere. "Data" is a plural noun. "the data were. . ." is correct.

You are right. Changed.

8). p. 20397, lines 13-16. This information belongs in the figure caption, not here.

The lines were removed from the text as they are already present in the figure caption.

9) p. 20398, lines 16-17. How did the vertical mixing occur? Deep convection? Frontal uplift?

We added the following sentence to p. 20398:

The vertical mixing during period (b) was enabled by less vertical stability of the troposphere and frontal uplift.

10) p. 20399, line 11. Change to "Only a few events. . . ."

Changed.

11) p. 20400, line 6. Reference Fig. 5 here.

Reference added.

12) p. 20404, line 2. Change to "deviations for all OTHER age classes vary

from . . . ."

Changed.

13) p. 20404, line 15. Should this be 40

Thank you for spotting this. The standard deviation is 40 %.

14) p. 20404, line 21. Change "conform with" to "are consistent with". "Conform" means "is constrained to agree" in this context.

Changed.

15) p. 20409, line 16. Change "effects" to "interactions".

Changed.

16) Figure 1. I don't understand the vertical plot. Is this showing the median altitude and the 1st and 3rd quartiles of the altitude range?

Yes, the caption says:

The box plot in the right panel shows the vertical extension of the measurement flights (median and 1st and 3rd quartiles).

17) Figure 3. Change "floating columns" to "bars".

Changed.

18) Table 3. Change "sinlge" to "single"

Changed.

19) Fig. 8. The axis labels are inconsistent. Fig. 8c is labeled "Rv(vol/pm2.5)" while the others are labeled with the ratios (no "R").

Changed.

20) Fig. A2. Could you show this as a scatterplot instead? It would give a better sense of bias and variability.

The figure was changed into a scatterplot.

21) I have not checked the references for errors. The one citation I did look at (Stein et al.) had an error - the page numbers are S381-S382, and the non-peer-reviewed abstract should not be cited. Please check all references thoroughly.

Stein et al. is removed. References are checked.

References:

Booth, B. B. B., Dunstone, N. J., Halloran, P. R., Andrews, T. and Bellouin, N.: Aerosols implicated as a prime driver of twentieth-century North Atlantic climate variability, Nature, Nature Publishing Group, a division of Macmillan Publishers Limited. All Rights Reserved., 484, 228-232, 2012

Clarke, A. and Kapustin, V.: Hemispheric Aerosol Vertical Profiles: Anthropogenic Impacts on Optical Depth and Cloud Nuclei, Science, 329, 1488-1492,

doi:10.1126/science.1188838, 2010

Feldpausch, P., Fiebig, M., Fritzsche, L. and Petzold, A.: Measurement of ultrafine aerosol size distributions by a combination of diffusion screen separators and condensation particle counters, Journal of Aerosol Science, 37, 577 - 597, doi:10.1016/j.jaerosci.2005.04.009, 2006

Fiebig, M., Stein, C., Schröder, F., Feldpausch, P. and Petzold, A.: Inversion of data containing information on the aerosol particle size distribution using multiple instruments, Journal of Aerosol Science, 36, 1353 - 1372, doi:10.1016/j.jaerosci.2005.01.004, 2005

Iorga, G., Hitzenberger, R., Kasper-Giebl, A. and Puxbaum, H.: Direct radiative effect modeled for regional aerosols in central Europe including the effect of relative humidity, J. Geophys. Res., AGU, 112, D01204, 2007

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., E., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U. and Worsnop, D. R.: Evolution of Organic Aerosols in the Atmosphere, Science, 326, 1525-1529, doi:10.1126/science.1180353, 2009

Péré, J. C., Mallet, M., Pont, V. and Bessagnet, B.: Impact of aerosol direct radiative forcing on the radiative budget, surface heat fluxes, and atmospheric dynamics during the heat wave of summer 2003 over western Europe: A modeling study, J. Geophys. Res., AGU, 116, D23119, 2011 Petzold, A., Döpelheuer, A., Brock, C. A. and Schröder, F.: In situ observations and model calculations of black carbon emission by aircraft at

cruise altitude, J. Geophys. Res., AGU, 104, 22171-22181, 1999
Schröder, F. P., Kärcher, B., Petzold, A., Baumann, R., Busen, R., Hoell, C. and Schumann, U.: Ultrafine aerosol particles in aircraft plumes: In situ observations, Geophys. Res. Lett., AGU, 25, 2789-2792, 1998
Shindell, D. and Faluvegi, G.: Climate response to regional radiative forcing during the twentieth century, Nature Geosci, Nature Publishing Group, 2, 294-300, 2009
Singh, H. B., Anderson, B. E., Avery, M. A., Viezee, W., Chen, Y., Tabazadeh, A., Hamill, P., Pueschel, R., Fuelberg, H. E. and Hannan, J. R.: Global distribution and sources of volatile and nonvolatile aerosol in the remote troposphere, J. Geophys. Res., AGU, 107, 4121, 2002
Voigt, C., Schumann, U., Jurkat, T., Schäuble, D., Schlager, H., Petzold, A., Gayet, J.-F., Krämer, M., Schneider, J., Borrmann, S., Schmale, J., Jessberger, P., Hamburger, T., Lichtenstern, M., Scheibe, M., Gourbeyre, C., Meyer, J., Kübbeler, M., Frey, W., Kalesse, H., Butler, T., Lawrence, M. G., Holzäpfel, F., Arnold, F., Wendisch, M., Döpelheuer, A., Gottschaldt, K., Baumann, R., Zöger, M., Sölch, I., Rautenhaus, M. and Dörnbrack, A.: In-situ observations of young contrails – overview and selected results from the CONCERT campaign, Atmospheric Chemistry and Physics, 10, 9039-9056, doi:10.5194/acp-10-9039-2010, 2010

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C9600/2012/acpd-12-C9600-2012-supplement.pdf

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