Atmos. Chem. Phys. Discuss., 9, C3620–C3625, 2009
www.atmos-chem-phys-discuss.net/9/C3620/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Origin of aerosol particles in the mid latitude and subtropical upper troposphere and lowermost stratosphere from cluster analysis of CARIBIC data" *by* M. Köppe et al.

Anonymous Referee #2

Received and published: 6 August 2009

Overall this paper fall into scope of ACP and represents a valuable contribution to our understanding of aerosol distribution in UT/LS region, where airborne measurements represents key source of reliable in-situ observations. Data presented are based on measurements carried out between Europe and SE Asia with the CARIBIC project instrumented airfreight container on board of civil airplane. Analysis is centered on use of multivariate statistics – cluster analysis in attempt to identify typical air masses and corresponding nucleation and total aerosol concentrations. After addressing several issues of moderate importance and comments it deserves publication in ACP. First

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Discussion Paper

more general issues are raised followed by more detail comments.

Whole paper is more of data presentation nature with little discussion. Key part is cluster analysis and typical air mass types derived using this multivariate statistical method. Unfortunately, there is no uncertainty level mentioned for cluster division and various cluster size. This part should be added and discussed.

Authors claim that this data represent the first statistically sound submicrometer particle number concentration data set in the UT/LMS over Eurasian continent and analyses the origin of these particles. Summer data set, which covers two summers (2006 and 2007) represents 15 609 data points (approx. 43 hours of measurements) unequally distributed over the summer period. Winter period measurements correspond to approximately 70 hours for winters 2006/7 and 2007/8, also unequally distributed with breaks up to 3 months. Size of the data set is not exceeding significantly several other airborne campaigns performed over SE Asia (ACE-Asia or INDOEX for example) and Europe (ICAART-ITOP, LACE or INCA for example).

Overall, this paper is more of a case dependent study than one would expect from statistically robust seasonal dataset. On my opinion, this study is first step towards such data set and analysis, but it is still a long way towards statistically sound seasonal data set and this claim should be removed. The measurements from previous CARIBIC flights on board of B767 between Germany and Maldives/Sri Lanka is larger than presented one (Hermann et al, JGR, Vol 108, D3, doi:10.1029/2001JD001077, 2003). Join analysis of both can be next step towards statistically sound data set significantly exceeding previous studies. Both datasets are obtained over Eurasia and within the same latitude range. Both also cover with similar proportions mid-latitudes, subtropic region and tropics.

Flight routes and flight altitudes for specific flights are often planned to use advantage of jet-stream on outbound flights from Europe and in opposite avoid the jet stream for inbound flights to Europe. Thus, fuel efficient flight planning preferentially follows

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specific regions of atmosphere and synoptic scale features, which eventually result in oversampling of specific conditions with respect to the others. Discussion on this subject is missing.

Two years period is covered with 32 flights (16 return flights between Frankfurt and Manila). Summer period is based on 7 return flights. Winter period is based on 9 return flights. Some of the clusters and analysis discussed are likely dominated by single flight or pair of flights (inbound-outbound within two days).

Authors should discuss the effect of measurements during specific time of the day (local time). With fixed timetable, the measurements were performed over certain regions during specific narrow period of time. This can significantly affect sampling of air masses influenced by deep convection and short lifetime tracers like O3, ultrafine aerosol N4-N12, NOy, cloud occurrence (HC cluster occurrence and its separation from BL cluster).

Absence of cluster indicating aircraft emissions is surprising. In CARIBIC study by Hermann et al. (Tellus 60B, 2008) on route between Germany and Caribbean it was estimated to represent 5 – 10 % of measurements for N12 > 3000 cm-3 STP and 5 – 20% of measurements for N4-12 > 1000 cm-3 STP. The air traffic density on Europe – SE Asia is likely not by order of magnitude lower than in North Atlantic corridor and over Europe and SE Asia is comparable. Can authors explain and comment on why this cluster was not identified?

It is hard to believe that high altitude clouds were not encountered during the whole winter season in sufficient amount to be clustered. Can you please comment on this?

Final dataset includes only 26 % of data collected during summer period and 38 % for winter period. Authors claim that this huge dataset reduction does not affect general results of multivariate analysis (lines 123-125). With some reservations I can agree that number of clusters is still the same, but frequency occurrence and distribution of clusters very likely will be influenced by missing data. What is the main source of

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missing data? In-flight calibrations can be evenly distributed, but it is hard to believe that instrumental malfunctions have the same pattern. Authors should clearly prove their claim and show level of uncertainty for each cluster size during both seasons as well as discuss on what statistical significance level are data affected.

Perfectly normally distributed data are of course ideal, but any of applied techniques do not require strictly normally distributed data. Can authors explain why they claim requirement for strict normal distribution?

Large part of the paper is devoted to description of statistical methods. However, information about the data treatment prior application of cluster analysis is often missing. How did you defined outliers? How large fraction of data was considered as outliers?

Similar results from the CARIBIC projects were previously reported by Hermann (2003, 2008). I do like more the analysis and format of these papers and I wonder why the same approach was not used also here.

Detail comments:

Line 78-79: with respect to frequency of flights and flight routes, the CARIBIC project provides wide regional coverage, but it is far from a nearly global data set.

Line 117: how exactly were measurements with longer time resolution than 10 sec interpolated?

Lines 146 and 161: Based on Table 2, there are 16 return flights FRA-MNL-FRA and one single flight FRA-MNL. Together it is 33 long-distance flights, not 36.

Lines 151-152: what synoptic weather situations were considered? From data selection pattern there is not a sign of flights excluded or included based on synoptic situation and division is done arbitrary between summer and winter half years.

Lines 152 – 153: Authors assessment of too small number of flights with respect to specific meteorological situations (synoptic?) is a contradiction to their claim in summary

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of statistically sound data set. On what scale does mean “finer division”?

Lines 387-400: What is a fundamental difference between BL air masses and high clouds cluster (besides presence of more dense clouds)? If the high clouds cluster is linked to deep convection as authors claim, it means that it is also air mass, which was rather recently in BL. Table 3 indicates that Lat/long is nearly the same for both. Both clusters show presents of clouds, similar O3 and CO. Thus, it will make more sense from point of view of interpretation and discussion to present HC cluster as a sub-cluster for BL air mass cluster.

Line 423: If the pressure altitude was used, than static pressure and altitude is the same as the later is derived from former parameter

Line 467: If the main season for agricultural fires over Ukraine and S Russia is during July/August. What is a difference between BL cluster between years 2006 and 2007 as the majority of summer flights for 2006 was conducted during this period, but not in 2007? Can this uneven sampling lead to oversampling of specific feature and thus lead to overestimating of its importance? Similar analogy can be applied to all HC and BL clusters for both summer and winter. Moreover, main burning season in Eastern Russia and Ukraine is during late spring/early summer. Why this is not reflected in data analysis? Part starting with line 477 and similar part for boreal winter analysis: Observed aerosol concentrations (upper and lower quartiles) in this study are compared with several other studies. It is not clear, however, if also upper and lower quartiles are reported from other studies or it is full range of measurements. In case that full range of measurements is reported, information value from such comparison is very low and relevant upper and lower quartiles or median values should be compared. One can fit almost any measurements (unless there is a major instrumental malfunction) into minimum-maximum range of values from field campaign. Can you please clarify? It will be nice to include comparison also with measurements obtained during INDOEX, ABC and ACE-Asia projects, for example.

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Line 516-517: Figures 5 and 8 show that certain air routes were used more often in summer and others more often in winter.

Line 563: it will be worth to analyze deeper frequency occurrence of FT-1 and FT 2 clusters. Is it because of different synoptic situations during the late autumn and late winter over mid-latitudes or maybe changes in monsoon circulation over SE Asia?

Fig. 4 and 7: The figures can be clearer if instead of cloud of points projections of each point on 3D planes are drawn

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 13523, 2009.