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Interactive comment on "Airborne measurements of the spatial distribution of aerosol chemical composition across Europe and evolution of the organic fraction" by W. T. Morgan et al.

Anonymous Referee #1

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This papers presents very interesting data and analysis from a series of aircraft campaigns. The paper provides ideas and information which add significantly to our knowledge of the formation of OM in the atmosphere over Europe, and which can help validate or improve models in future.

The paper has some weaknesses, however, which should be addressed before if can be recommended for publication.

1. The paper says that it presents evidence to support the framework suggested by Jimenez et al., 2009. This is fair to a certain extent, but really the paper supports the general idea that VOC is oxidised more the further downwind one looks. The paper

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misses the opportunity to evaluate the time-scales of the conversion from HOA to SV-OOA to LV-OOA that would really underpin the Jimenez framework. I think that such an evaluation is possible from the data, and would make the paper much more useful to the community.

2. The paper makes no attempt to put the AMS results in the context of what is already known in Europe. In particular, the authors find good correlation of OOA with nitrates and sulphates, which at first sight suggests an anthropogenic source. However, these results needs to be compared and contrasted to conclusions from other studies, notably the 14C studies of Gelencser et al. (JGR, 2007), or Szidat al. (e.g. JGR, 2006, ACP, 2009), Yttri et al.(ACP, 2009), or the PMF studies (e.g. Saarikoski, ACP 2008). These all suggest BSOA as the source of most OM.

3. The reason for the correlation with sulphates is also interesting to discuss. The spatial distribution of S in the UK for example is very different to that of NOx and VOC (or BVOC). Don't the high time-resolution data allow any conclusions to be drawn concerning whether S is indeed involved in SOA formation or not?)(The recent review of Hallquist et al., ACP 2009 suggests many possible pathways to OM formation. Do the current data not give any hints as to sources?)

4. There is no discussion or presentation of vertical profiles of OM or OM:PM ratios. Did these change much with height? Over 2-3 km there is a substantial temperature change, which could be expected to affect condensation of SOA - is there any sign of such effects?

5. The paper is rather long and heavy to read. This is partly as the authors have erred on the side of providing more information than less (that's okay I think), but some things could be improved to help the reader. Ideas follow.

6. Figs. 2-4 could be usefully condensed into one map, with pies to show the OM, SO4 and NO3 splits. This would give a much faster visual impression of the data. I would also like to see a Table with these numbers (in the Supplementary would do), since

figuring out the masses and ratios by eye, and in comparison to the flight paths of Fig. 1 is difficult.

7. Tables would also help the discussion in section 3 (p27223–27224). Here I can read that OM is significant in both background and polluted conditions, but I can't see where this statement comes from.

8. p27229. Lines 25 on. Here the authors use OOA-2, OOA-2 etc. This notation reminds of that used previously by many authors (e.g. Ulbrich et al. 2009). Make it clear the extent to which the notation is consistent across papers, or simply a result of PMF?

9. p27230, 4.2.1 etc. Data are discussed without any supporting figs or tables for the reader to refer to. Addition of Tables would be best.

10. p27231, similar point here. On lines 7-8 the "slope" is compared against literature, without either this study's slope or the literature values being given. Give both.

11. p27231, emission ratios cont. 2 papers is a very limited resource when citing emission rates. What about official emission rates of OM, NOx, CO, etc.?

Figures:

12. Fig. 1. I found the mix of periods and labels confusing. Each period consists of many days. How do the individual flights shown relate to the full period - did the aircraft follow exactly the same flight path for every day of each period?

13. Fig. 4 The caption presumably means Nitrate total mass, not AMS total mass.

14. Fig. 5a. One usually refers to vertical and horizontal bars or lines, not "sticks and bars". Sticks could be in either direction anyway, so this new notation is only confusing.

15. Fig. 5b. Why does this figure again use just 2 literature values, and why different from that used above (now Allan, not Lanz). Would you expect these emission values to apply to NW Europe anyway?

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16. Fig. 6a. The flight names are so long as labels that it isn't clear which correlation coefficient applies, and the plot is sometimes so messy that the labels can't be rad anyway. Use e.g. a cross to indicate where the r2 values are.

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