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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO" *by* P. F. DeCarlo et al.

**Anonymous Referee #1**

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This paper presents a case study analysis of organic aerosol data from an AMS on the C-130 during the MILAGRO intensive. Two flights are analysed, chosen on the basis of one being heavily influenced by biomass burning and one being largely free of it. PMF analysis of the high resolution data is performed and compared with other in situ data sources and regional model outputs in an effort to apportion the different OA components and explore atmospheric processing. The authors employ what they call 'postprocessing' to reconcile the data from the two flights and quantify the influence of biomass burning on OA components normally ascribed to urban sources. This



is an interesting technique, however given than only two flights were used in this paper, it is also intrinsically impossible to validate here (see below) and so some of the conclusions risk being overstated.

This paper is generally well written, topical and well within the scope of the MILAGRO special issue and the scientific scope of Atmospheric Chemistry and Physics. I recommend that this be published after the authors consider the following (hopefully minor) comments:

General Comments

The authors give very little discussion of the altitude of measurement. This is important, because if any data collected in the free troposphere or any residual layers were included in the analysis, these would add an additional transition beyond the plume/background interaction model that forms the basis of this analysis (fig 1) and artificially inflate correlation statistics. I would strongly recommend filtering these data from the analysis and if they have already been removed, the authors should state as such.

The 'postprocessing' technique is interesting, but given that there are only two flights being analysed, there is no way that the validity of this technique can be verified within this work. Unless additional flights can be included in the analysis (which I'm guessing they can't), the authors need to be more upfront about this intrinsic limitation in the discussion. I would also recommend reporting the non-postprocessed values in the conclusions section in addition to those given.

In a more specific case, the analysis performed in 3.5.4 seems a little tenuous. As described, there are many potential confounding issues that are not adequately discounted (the two flights having 'roughly similar' SO4 concentrations is not a particularly compelling argument). In addition, other factors not discussed may include the effect of seed ambiguity in the PMF solutions when comparing the two flights, the effect of aqueous processing or the influence of regional transport from outside of the Mexico

City basin. Given the large amount of scatter between LV-OOA and SO4 within the individual flights (as shown on figure 4), this rough calculation of the addition of mass from these processes seems to be a bit of a stretch. Without better justification, I'd be wary about carrying these numbers forward to the conclusions section.

Given that Crounse et al. informed the postprocessing (e.g. apportioning CO to biomass burning), the comparison in section 3.6 seems a little self-fulfilling. In order for the comparison to be fair, influences of this work should be excluded from the data processing presented here. As a general point, the comparison is not performed very critically in the text; currently, phrases like 'agrees well' are used without any quantitative measure or context as to what would constitute a good agreement.

The analysis of seed ambiguity is very interesting and the authors deserve credit for giving a concise account of this as an appendix rather than attempting to bury it in the supplementary material. However, one could suggest that the strong dependence on the seed could be indicative of inappropriate convergence criteria and tweaking this (in conjunction with the error model) might be the more appropriate way of dealing with the issue. The authors' thoughts on this matter would be informative. Also, the ambiguity should also be carried forward and reflected in the numbers shown in the discussion and conclusions sections in some way (see other comments).

As regards presentation, I would recommend fewer subheadings be used; many sections are only comprised of single paragraphs, which is frowned on in many journals.

Specific Comments

Given that sources and processes of organic aerosol is a common theme of many MILAGRO papers, I recommend the running title is changed to something more specific to this work.

The fact that this paper presents data from only two flights should be stated in the abstract; currently, it gives the impression that an entire campaign's worth of data was



used, whereas in actuality this work is more akin to a case study. Also, as explained below, the '>90% anthropogenic' statistic reported is currently unsupportable and should be removed.

P2449: 'Traditional' and 'recent' are not really sufficient descriptions of the different models; they use fundamentally different approaches (suited to different scientific applications) and as such, they can't be compared on the basis of when they were developed. Better descriptions are warranted.

P2460: The discussion in section 2.5 is mostly redundant and a little self-contradictory in places; if the large correlation is mainly due to plume transactions, then the sources are still effectively 'collocated', only on the scale appropriate to the measurement (i.e. regional rather than local). Generally speaking, much of the caution that must be exercised when interpreting simple correlations is nothing more than sound scientific practice and really doesn't really need covering in as much detail. Specific cases where correlations could be misleading could be dealt with in the discussion.

P2463: While using the 'LV-OOA' and 'SV-OOA' terminology gives consistency with recently published works, it should be noted that the AMS alone does not measure the volatility of the particulates. As such, the basis for aligning the factors observed here with previous volatility works should be stated.

P2463: How much of the difference in HOA/CO ratios could be due to the ambiguities in the PMF analysis reported in the appendix?

P2465: A strong correlation with CO is not adequate to reach the conclusion that the SV-OOA is mainly anthropogenic in origin; biogenic precursors could easily be emitted within the same geographic area as the anthropogenic equivalents. Also, anthropogenic NOx could also be stimulating the formation of biogenic SOA, which would lead to an apparent enhancement within the polluted plume.

P2465: I would recommend caution when explaining how atmospheric processes 'convert' between the two OOA types; it should be stressed that the two factors represent end points in a continuum of organic composition. The way it currently reads, a reader would be forgiven for thinking that they are two discrete chemical components.

P2475, L5: Many previous studies have shown the contributions of m/z 60 and levoglucosan to vary according to fire type and airmass history, so this conclusion is not new. As a minimum, it would only be fair to cite previous works in the literature. What would be better is if the authors could somehow put numbers to this phenomenon, applicable to this case.

Technical Comments

P2448, L24: It should be mentioned that a more fundamental difference between CMB-OMM and AMS multivariate analysis is that the latter uses data from all of the organic mass, rather than a subset.

P2450, L8: Given the wide variation of OH concentrations in the atmosphere, the concentration relevant for the 'approximately 1 month' statistic should be given.

P2450, L24: The open, non-agricultural, biomass burning events during MILAGRO were as much grassland fires as forest fires.

P2452, L12: Stylistically, the end of the introduction reads more like a conclusions section. Suggest a reword to focus on the hypotheses being tested rather than the findings.

P2452, L19: A citation for the gas phase instrumentation should be specified.

P2453, L6: The ODRPACK95 package used in Igor Pro should probably be referenced.

P2458: Technically, PMF is an 'algorithm', not a 'model'. Also, it is PMF2 which is being referred to, as a trilinear version (PMF3) exists. Finally, the key constraint is that the solutions are non-negative as opposed to positive (i.e. zeros are allowed).

P2459, L16 (and elsewhere): '1 atm' should be specified in SI units.



P2460, L3 (and elsewhere): 'LST' should be defined (relative to UTC), as it is not a normal time zone code.

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2445, 2010.