

***Interactive comment on “Source apportionment of submicron organic aerosols at an urban site by linear unmixing of aerosol mass spectra” by V. A. Lanz et al.***

**Anonymous Referee #2**

Received and published: 12 January 2007

The manuscript is very interesting and offers a new methodology for utilising AMS data to probe sources of atmospheric aerosol particles. It certainly should be published and ACP is an ideal journal for the work. It will be a valuable contribution to the literature. Both referees have offered some very useful comments and I have no wish to cover similar material. I will therefore restrict my comments to those not made by the other referees, or where I feel it is worth commenting on the other referees statements. The more important comments are listed, before some minor typographical and other language errors.

General Comments: 1) Referee 3 also raises this question in point 5. The way the

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charbroiling MS is retrieved isn't too clear. As I understand it the signal was isolated (how?) and the background removed (what background was chosen?). There appears to be a significant degree of temporal similarity between charbroiling and wood burning (figure 6). If the charbroiling MS is retrieved from the dataset as is implied isn't there potentially a contamination problem in the source spectrum? The authors should make it very clear how this spectrum is derived, why they believe it is free from contamination and how they justify comparing it with the dataset used to source it in the first place..

2) Page 11695 line 4-5, and elsewhere <The original first, OOA-like factor is split into highly aged background aerosol (high similarity to aged rural aerosol:  $R^2=0.97$  and to fulvic acid:  $R^2=0.93$ ) and one that mostly resembles aerosol from isoprene oxidation in the presence of  $\text{NO}_x$  ( $R^2=0.82$ ).> I do not see how you can use a rural ambient spectrum as a diagnostic when you are separating out the secondary aerosol. You argue that one component is typical of rural air and the second one due to isoprene oxidation. It is recognized that rural aerosol are less well modeled by the Zhang approach than urban aerosol, how do you know that rural aerosol, when analysed using PMF approaches do not also split the oxygenated fraction into two discrete source spectra as you have found here. I would think this quite likely in fact. It seems to me therefore to be unwise to compare with rural spectra as a "source" function. A discussion on this would be helpful to the reader

3) I agree with Prof Paatero that the authors should strive to convey details of the technique as well as the science (and ensure that the details are correct) at the expense of a little more length. This is important the as I suspect that this paper may well become something of a blueprint for analyses of this kind by a, now large, AMS community, as the Zhang papers have done for PCA

4) I agree with the comments by Prof. Paatero that factor 6 is rather overplayed given the statistics and the lack of supporting data, this should be downplayed a little more

Minor Corrections Page 11684 lines 5-10 This is rather clumsy, it isn't obvious on first

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reading that the authors are referring to the Zhang method all the way through this discussion. I suggest a reword.

Page 11684 lines 13-14 <Ea signal mainly from di- and poly-carboxylic acids functional groups, CO+)> should read <acid>

Page 11685 line 22 <are capable to reproduce emission events of> suggest rewording to <are capable of reproducing emission events of>

Page 11687 line 5-8 The authors use a CE of unity, rather than 0.5, presumably because the area is impacted by significant NH<sub>4</sub>NO<sub>3</sub> rather than a sulphate dominated inorganic fraction though this is not shown. This is validated using a total mass closure with a PM<sub>10</sub> measurement. That implies virtually all the PM<sub>10</sub> is PM<sub>1</sub> and considerably less than half of the mass is in the coarse mode. Is this true? It does not seem a reliable way of validating the choice of CE. Though it does not matter for this paper it may make a difference for later analyses.

Page 11688 line 16-17 <at t samples in time samples,> correct

Page 11690 line 17-18 It is best to stress that fulvic acid is not a <source> nor is it a <tracer> it is a model compound that describes the chemical functionality of aged, oxygenated aerosol.

Page 11693 line 6 <The source profiles and its activities> should read <Eand their activities>

Page 11694 line 4: It is worth citing Zhang et al again here; what you show is the consistency between the two approaches for the 2 component model but this may be missed without citation.

Page 11695 line 12 <is losing most of its> should be <losing>

Page 11696 line 1-4 The comments here on factor six seem somewhat overblown. Surely the comment is that there is some indication from a fragment that resembles an

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oleic acid type signature that this source may arise from food cooking. Further work to obtain a source fingerprint for food cooking is necessary.

Page 11697 line 22-23 Are the differences between modeled and measured  $m/z$  random or systematic? Our experience measuring bush fires suggests that  $m/z$  60 is preferentially removed over time, is this what you are observing? Is the remaining source  $m/z$  preserved whilst  $m/z$  60 decreases? Is levoglucosan the dominant source of both  $m/z$  60 and 73?

Page 11698 line 20-21 <Periods of nice weather> The text really ought to define the meteorological condition rather better than this. I assume you mean anticyclonic, stable, clear sky periods?

Page 11699 line 6, I assume that the fireworks are the explanation for the morning of August 1, perhaps this should be stated again here?

Page 11699 line 4 and figure 5. Do you have an explanation for the reduced error for both the 2 and 3 component model outside the periods of photochemical activity compared with the periods when photochemistry is active?

Page 11699 line 17-18 Yes but as I recall whilst the oligomerized fraction appear to be involatile in the chamber studies of Kalberer et al, AMS mass spectra taken under similar conditions (Alfarra et al ACPD) do not show similarities to fulvic acid, at least under the high concentrations in that study. This should be noted

Page 11702 line 15-16 I suspect it is the fires that are causing the wood smoke not the fireworks, maybe the argument should be put the other way around.

Page 11703 I am less than sure of this discussion. The first paragraph does not convince me that this may be food. The wind direction analysis is interesting but doesn't definitively show a source. The last section should appear above as it defines what you mean by fine weather a little better.

Page 11723 Figure 7 Do you do not mean that the notched median band for the 5th

hour of the day is LARGER than that for the 6th hour of the day?

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