

Interactive comment on “Contributions from transport, solid fuel burning and cooking to primary organic aerosols in two UK cities” by J. D. Allan et al.

Anonymous Referee #1

Received and published: 18 September 2009

Allan et al. present a factor analysis on AMS organic data measured in two UK cities with an emphasis on primary organic aerosol.

As one major result, the authors highlight the importance of food cooking (specifically from oil frying) as a main source to ambient OA in a plausible way.

The manuscript is well-written and well-structured, and contains an objective and consistent source apportionment study.

This paper is topical and of great interest to ACP readers.

I have no major objections, but a few comments (line by line) that need to be addressed.

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————— Specific comments —————

P19105, L17: "...greatest fraction..." – revise

P19105, L20: "...travel..." - revise

P19105, L26-L27: "At distances removed..." – revise

P19106, L22: "applied to other sources of atmospheric aerosol data" – revise

P19106, L27: In this study, OA was separated into HOA, charbroiling, wood burning, and food cooking OA.

P19106, L27: With the MCA, more than two components can be resolved (Zhang et al., 2007)... but it is certainly not restricted to three components. Revise.

P19106, L29: Also mention that OOA, type 2, and OOA, type 1 could be associated with different volatilities (see e.g. Lanz et al., 2007).

P19107, L4-5: "chemically younger" – revise

P19107, L25: "These are much larger cities compared to Zurich" – why is this important here?

P19108, L14: "lightly-used" – revise

P19109, L15: I do not understand why a newer instrument would measure more organics (but less chloride, as an example). Please explain. Are these intercepts significantly different from 0? Please indicate the uncertainty associated with these numbers.

P19109, L10: "the instrument" - mention again the type of instrument (C-ToF-AMS)

P19109, L22: I suggest estimating the collection efficiency, CE, via total AMS aerosol + black carbon, BC (derived from the mentioned MAAP instrument), versus PM1.

P19111, L8: maybe "as provided by P. Paatero (University of Helsinki)"

P19111, L10: Why is PMF applied to high-resolution AMS data is still in its "relative

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infancy" (compared to PMF applied to unit-mass resolved data)? Please explain.

P19111, L17-18: 300 cumulative steps seem rather small given the large datasets.

P19111, L29: I do not understand this cut: are there no low signal-to-noise fragments at $m/z < 200$? In contrast, I expect some relevant PAHs to be found at $m/z > 200$.

P19112, L4 sqq.: I agree that m/z 18 is depending on (scaled to) m/z 44, but there are more such interdependencies in AMS mass spectral data. One could also argue that a strong weighting of m/z 's 18 and 44 is justified, as they make up for such a large fraction of the organic mass.

P19112, L12: In contrast, such plumes (e.g. wood burning plumes) may introduce the variability necessary to separate distinct sources (e.g. wood burning) by PMF. . . but I agree that singular emission events might also cause some positive artefacts for those sources: in such cases, I suggest relaxing the non-negativity constraints for the factor time series.

P19112, L24: maybe "non-refractory"?

P19112, L25: "...organics dominate with contributions from sulphate. . ." – revise

P19112, L26: "behaviour"?

P19113, L12: "where" or "when"?

P19113, L27: I doubt if the AMS represents the appropriate instrument to detect organic sulphur. After vaporization and ionization, it may be measured as OM + SO₄²⁻

P19114, L7: "oxidized" or "oxygenated"?

P19114, L14: the "slight dip around midday" in the OOA diurnal cycle is plausible. In the morning hours, aged OOA is down-mixed into the increasingly high boundary layer and the photochemical aging is not prominent before the after-noon hours.

P19114, L15: Please expand on this question: was OOA, type 2, not present in these

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campaigns or was it always condensed in the aerosol-phase and has therefore no distinct temporal variability (i.e. it can not be separated by PMF)? Is the OOA-factor derived here a mixture between type 1 OOA and type 2 OOA (2:1, 3:1)?

P19116, L19: "consistent with combustion HOA" – revise

P19117, L9 and elsewhere: replace "4 factor solution" by "4-factor solution"

P19117, L19: "...giving alternative solutions that while typically not the most optimal. . ." this sentence will not get your ideas across to the reader. Furthermore, a Paatero-citation would be more appropriate at this instance.

P19117, L22: What is meant by "mass spectral space" and "time series space"?

P19118, L1: The high Q/Q.exp-values are rather surprising, also in the view of similar studies (Lanz et al., 2007, Ulbrich et al., 2009) that reported Q/Q.exp values close to 1. I wonder if A.) this difference is due to the instrumentation (ToF-AMS vs. Q-AMS with relatively low S/N) and B.) if this ratio can be reduced by specifying a model error > 0% in PMF2 v4.2.

P19118, L8: As a matter of taste, I would put these mass spectra into the supplementary section.

P19118, L11: "...it was clear that some of the mass that would otherwise have been assigned to SFOA was being assigned to the other factors." – this sentence is not clear.

P19118, L : I agree that rotational ambiguity and other program settings (including the number of factors) determine the worst-case uncertainty of the PMF-AMS analysis. However, I disagree to call this uncertainty "confidence interval". This term should not be used in that context.

P19119, L16: "other local combustion sources" – such as . . .? It should also be mentioned that no perfect agreement between PM from combustion and gaseous pollutants from combustion can be expected (different formation and removal processes).

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P19119, L15: "...also...also..." - revise

P19119, L16-17: "...using...using..." - revise

P19119, Equation 1: What is meant by $f(\text{HOA}, \text{SFOA})$? Primary combustion OA ($=\text{HOA}+\text{SFOA}$)? The equation reads as if you modelled HOA and SFOA from HOA and SFOA. ... I am not sure if I got the authors' idea, but maybe one should include the terms CO and NO_x in this equation?

P19119, L21: "gas phase concentration" maybe you mean mixing ratios of gaseous pollutants

P19119, L26 : "...improve the NO_x values..." – usually, models are not used to improve measurement values.

P19120, L8: "...dwarfed..." – revise

P19120, L24: why was there not enough "OOA variation" to separate OOA1 and OOA2?

P19120, L1: "...OOA, which assuming it is secondary in nature..." please note that Weimer et al., 2008 characterized primary wood combustion OA by an OOA-like mass spectrum (depending in burning conditions...).

P19122, L11: Please use a more descriptive title (4.2 Combustion factors) (disambiguation: "emission factors")

P19122, L17: I am not sure if a measurement campaign can be called "experiment"

P19123, L12-13: given the high m/z 44-to- m/z 57 ratio in the OOA by Weimer et al. (2008) it is rather likely that this primary OOA from wood burning (smouldering phase) would be classified as OOA by PMF and usually (but wrongly) interpreted as SOA.

P19123, L22: "raw AMS spectra" – do you mean: average organic MS?

P19126, L6-7: "...using a quadrupole-based AMS, the PMF analysis failed to identify

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the COA component..." why? Do you have explanations for this behaviour?

P19130: It could be added to this discussion that "charbroiling aerosol" comprises a broad mixture of all sorts of aerosols from char, oil, accelerant, meat, vegetable (wood), fat etc. burning, and that these latter contributions have variable importance and are difficult to separate. In any case, I think it is not very surprising that "charbroiling aerosols" as characterized by Mohr et al. (2008) had no (a minor) impact on ambient OA in UK cities, in wintertime.

P19132, L19: "...not significant enough..." what are the "significance"-criteria? Either it's significant or not.

P19132, L25-27: What are the standard deviations of these emission ratios?

P19133, L21: "a priori" - revise

P19144: Please indicate uncertainties associated with intercepts and slopes

P19148: Figure 1: the term "Volume concentration" (y-axis) does not apply to AMS data shown in figure.

P19149: Figure 2: time-axes should be labelled "date" rather than "date and time" (this also applies to some other figures). The text in the caption could be more descriptive.

P19152: Figure 4: I assume that the correlations OOA vs. AMS-sulphate etc. are not very sensitive to rotations. Is this correct?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 19103, 2009.

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