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Atmospheric
Chemistry
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Discussions

Interactive comment on “Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data” *by* C. Mohr et al.

Anonymous Referee #2

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General comments:

This manuscript presents and interprets results from PMF (positive matrix factorization) analysis on HR-AMS (high resolution time-of-flight aerosol mass spectrometer) data taken during the DAURE campaign in Barcelona. The authors identified five factors which have also been identified in previous studies. Cooking organic aerosol (COA) comprised on average 17% of total OA. The authors note that due to the mass spectral similarity of COA and HOA (hydrocarbon-like OA), especially at unit mass resolution (UMR), COA may be more prevalent than previous studies, esp. those with UMR data only, might suggest. The authors suggest a method to estimate COA when high resolution data are not available.

Overall, the manuscript is well written and reports interesting results which are relevant to the community and within the scope of ACP. I recommend publication of the manuscript in ACP after my comments below have been addressed. My specific comments below include two major comments, followed by smaller comments.

Specific comments

Major comment 1: Choice of PMF solution

I have several comments and concerns about the choice of the particular PMF solution.

a) Currently, the choice of PMF solution is discussed exclusively in the supplemental information. Some of this discussion should be moved to (or summarized in) the main text since it is quite important for the information, interpretations and conclusions presented in the main text.

b) (Page 13, and Fig S20) SEED = 64 is not shown in the figure. I will assume that the authors meant SEED = 46, which is highlighted (circled) in Fig. S20. The authors chose SEED = 46 since this solution yielded the best correlations with ancillary data. Figure S20 shows that the SEED = 46 solution has by far the highest Q/Qexpected, i.e. the highest error (worst fit) compared to other SEED solutions. I am aware that choosing the best PMF solution for MS data involves more than mathematical diagnostics. But, it is nonetheless striking that the best solution based on ancillary data is by far the worst solution based on mathematical diagnostics. This may suggest that one or more of the following is not appropriate (at least for this dataset): 1. the mathematical model or error estimation (see below), 2. choosing factors based on correlations with ancillary data, and/or 3. the choice of ancillary data. This should be discussed in the revised manuscript. The authors generally follow procedures and methods accepted and used by the AMS community, but the community is also still debating the meaning of PMF factors and the best way to select them. Thus, it is important to discuss these issues rather than glance over them.

c) Q/Qexpected is about 12.5 for the HR PMF solution. This suggests an overestimate of the uncertainty in the HR signal (Qexpected) and should be discussed in the revised manuscript. Incorrect estimation of Qexpected could in fact (partially) explain the discrepancy between mathematical diagnostics and those based on ancillary data discussed in b) above.

Major comment 2: Fossil vs. non-fossil carbon (p. 27398, starting line 17)

This is an important finding in the paper, but as currently written it can be misleading and/or confusing for two main reasons:

a) It is not clear how the authors go from estimates of OM:OC displayed in Fig 2 to estimates of fossil vs. non-fossil (lines 20-22) and suggest that they explain this in more detail.

b) I also suggest to be more cautious with identifying factors as “POA” as it is not clear to what extent COA or BBOA (or maybe even HOA) are truly primary (have not undergone chemical reactions after emission).

Other specific comments

- Introduction (1st paragraph): I suggest that the authors point out that “organic matter” is composed of many chemical species, one of the ways in which it is much more complex than the other major PM1 components.

- p. 27392, line 6: “. . . organosulfates and organonitrates also contribute to their concentrations. . .”. I suggest stating that they may also contribute since the authors cannot confirm the presence of organonitrates and –sulfates in this study.

- p. 27392, line 23 (and elsewhere in the manuscript): It would be appropriate to point out that the PM1 composition shown and analyzed is the composition of dry PM1 since

particles are dried before detection. Water is probably a major constituent in PM1 which is not discussed in the manuscript.

- p. 27393, line 4: It would be appropriate to include correlations of the filter data and AMS data for SO4, NH4, OA and nitrate.

- p. 27395, line 11: It is not clear what the authors consider a “high-enough R2” to confirm correlation. They seem to be content with R2 = 0.22 here but not with R2 = 0.17 on p. 27396 line 13.

- p. 27395, line 26 and following discussion: The authors’ use of “traffic emissions” here and later is confusing since they earlier discussed ship traffic as one potential source, but the “traffic emissions” discussed here do not include ship traffic. I suggest revising to avoid this confusion (e.g. light-duty vehicle emissions? road traffic emissions?)

- p. 27403 line 21 (and following): suggest changing “is linear” to “is assumed to be linear”, or explain why explain why it must/should be linear.

- p. 27405, line 27405: suggest changing “first-generation secondary OA” to “early-generation secondary OA”. While the data suggest that this OA is fresher, they do not show how many generations of chemistry the OA has undergone.

- p. 27405 lines 12-13 (last sentence of section): The authors note that it will be of great interest to validate the coefficients they find with other datasets. Considering that different AMSs can yield (somewhat) different mass spectral signatures when measuring the same aerosol, COA extracted from other datasets would not necessarily be expected to yield the same coefficients. “validate” therefore does not seem to be the appropriate term. I suggest to discuss instead the usefulness of e.g. comparing coefficients from different datasets.
The potential difference in mass spectra from different instruments and, therefore, the potential difference in coefficients is an important caveat which the authors should address in the revised manuscript.

Technical corrections

- p. 27385 (and elsewhere): change O/C to O:C to be consistent with convention and with the rest of the manuscript.

- p. 27386, line 29: suggest rewriting “keeps being added” since it is colloquial

- p. 27388, line 11: suggest changing “specially” to “especially”

- p. 27391, line 17: the Wiedensohler et al., 2011 reference is not in the reference list. The authors should add this reference and check the reference list for completeness and accuracy.

- p. 27395, lines 13-15: separating “rather” and “than” by two lines makes this sentence difficult to read/understand. I suggest revising it.

- p. 27406, lines 7-8: suggest to italicize m/z

- p. 27406, line 26: suggest changing “emissions site” to “emission sites”

- Supplemental information page 6, line 54: “relied” should probably be “related”

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 27383, 2011.