

Responses to

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 #1

Received and published: 1 December 2011

This paper covers the PMF analysis of HR-TOF-AMS data taken during the DUARE campaign in Barcelona, Spain. The factors identified are consistent with similar recent studies in urban environments and comprise of low- and semi-volatile oxygenated, biomass burning, hydrocarbon-like and cooking organic aerosols. Attempts are also made to extend the ‘poor person’s PMF’ (Aiken et al., 2009) to COA. Overall, this paper is very well written, clearly and concisely presented and relevant to the science.

I would recommend that the paper be published in ACP after the following comments have been considered:

The authors thank Referee #1 for his or her thorough review of the paper. We will address first the main critical points and then reply on the specific comments individually further below.

General comments:

The discussion of the choice of solution set seems to be somewhat marginalised in the supplementary material. Given how much the later conclusions depend on the choice of the number of factors and p_{peak} value, it would be informative if the authors could include a précis of this process in the main text.

The following text was incorporated into the main manuscript (see also replies to comments by referee #2): “For a discussion of the number of factors chosen and the criteria used to select the best UMR solution, we again refer the reader to the SI, section 3.3. HR solution criteria are briefly outlined here, figures can be found in the SI, section 3.4. The chosen 6-factor solution for the HR dataset is presented in Fig. S15. The two SV-OOA factors were recombined into a single SV-OOA factor using the sum of the time series and the concentration-weighted average of the spectra. The 5-factor solution (Fig. S16) was discarded due to the high similarity of two factors (spectra and time series). The 7-factor solution (Fig. S17) features three LV-OOA spectra: PMF seems to assign an individual LV-OOA factor to three different sections in the time series, which can be due to small variations in instrument tuning with time. As the variation in the solution space $p = 6$ as a function of f_{peak} , a user-specified rotational parameter, is negligible (Figs. S18 – 19), the most central solution ($f_{\text{peak}} = 0$) was chosen and different SEED values (random initial values) were explored (Figs. S20 – 21). SEED = 46 was chosen as the best solution due to correlations with reference spectra, even though it exhibits the highest Q/Q_{expected} value (12.75 compared to ~12.55 for SEED \neq 46) and is thus not the mathematically optimal solution (see SI 3.3). A boxplot of the scaled residuals (boxes are +/- 25% of points) per m/z is shown in Fig. S22, time series of the residuals and Q/Q_{expected} are shown in Fig. S23. $Q/Q_{\text{expected}} \gg 1$ (12.75) indicates an underestimation of the number of factors or of the errors in the input data, possibly due to fitting errors in the HR data which are currently not included in the total error estimation. In addition to $p > 6$ (which was explored and shown not to give a more plausible solution, see above), the introduction of a “model error term” might also reduce Q/Q_{expected} , however, as discussed in Ulbrich et al. (2009), this is usually not applied in AMS data sets.

Q/Q_{expected} values $\gg 1$ for HR-ToF-AMS data have been shown recently by other authors, e. g. Allan et al. (2010) published a Q/Q_{expected} value of 10.5.“

While the method for estimating the cooking fraction is potentially of much use (especially when interpreting ACSM data), experience has taught the AMS community that the ‘poor person’s PMF’ is not particularly robust between instruments and campaigns. It should be stressed in the text that it is possible (and indeed likely) that the derived coefficients presented will vary between datasets.

It is possible and indeed likely that the coefficients will vary between datasets. We feel that this is also stated clearly enough in the last sentence of section 3.3.2 (“The application of this approach to other datasets will be of great interest to validate the coefficients found here.”), and this is also the reason why we are interested in comparisons with other datasets. In addition, as pointed out by referee #2, different AMSs can yield somewhat different mass spectral signatures. To consider this important point, the last sentence of this section was changed as following: “The application of this approach to other datasets and the comparison of coefficient values will be of great interest, as discrepancies can also be influenced by some variation of mass spectral patterns between different instruments.”

Throughout the manuscript, the authors use the R^2 statistic to compare the consistency between mass spectra. However, due to the nature of the mass spectral data, it is probably more appropriate to use an uncentred Pearson’s r (normalised dot product) instead. Note that R^2 is still appropriate for comparing time series.

The uncentered R values were added in brackets to the R^2 for the spectra comparisons.

Specific comments:

Page 27387: The final paragraph of the introduction should be restructured slightly. As it is currently worded, it would imply that the technical development of the COA estimation method is the principal focus of the paper, which I do not believe to be the case.

The final paragraph and especially its first sentence, “This paper presents a new method for identifying and estimating cooking contributions to organic aerosol.”, were indeed meant to refer to both the method for identifying COA in urban datasets (Fig. 6) AND the simple approach to estimate COA based on marker mass fragment signals (Fig. 8). As this was maybe not fully clear, the beginning of the paragraph was changed as following: “This paper provides a characterization of the chemical composition and organic aerosol sources of PM_{10} in Barcelona during winter. In addition, a new method for identifying COA in ambient air and estimating cooking contributions to organic aerosol is presented.”

Page 27393: The authors refer to two factors as ‘LV-OOA’ and ‘SV-OOA’. While these are the commonly preferred terms, a caveat should be added that the AMS alone does not measure volatility. Rather, the factors identified can be related to low- and semivolatile organics that have been measured elsewhere.

It is correct that the AMS does not measure volatility, and that LV- and SV-OOA refer to two subtypes of OA which can be quantified with an AMS but whose physical properties cannot be measured with just the AMS. This should be clear from the description in the methodology section. It is stated later on in the manuscript that the O:C ratio and volatility are inversely related, and that LV-OOA is usually correlated with the low-volatile SO_4 , while SV-OOA is correlated with the more volatile NO_3 . In addition, we state on p. 27395, lines 12 – 17: “[...] the diurnal patterns of both SV-OOA and NO_3 (not shown) indicate that the temporal evolution of both components is rather driven by similar processes such as primary emissions, rapid chemical formation, and the diurnal cycles of land/sea breeze and boundary layer height than gas-to-particle partitioning based on volatility characteristics. SV-OOA can

thus be described as of more local origin, less processed than LV-OOA". To fully clarify this point, the following sentence was added to the description of the LV- and SV-OOA factor: "We use the terms "LV-OOA" and "SV-OOA" as introduced by Jimenez et al. (2009) although we did not explicitly measure the volatility of the compounds within this study."

Page 27396: The method for calculating b_{880nm} traffic (and corresponding biomass burning product) should be briefly described.

To briefly describe the model, the following sentence was introduced: "The model uses the Lambert-Beer law to obtain equations which relate the absorption coefficients (b_{abs}) measured at two different wavelengths with the Ångström exponents for conditions of pure traffic and pure biomass burning."

Page 27397: Rather than remove spikes, why not average the AMS data down to the same time grid as the Aethalometer data? Better still, why not select the data corresponding to the times within the Aethalometer scanning cycle that correspond to the 880nm measurement?

The AMS data were averaged to the same time grid as the Aethalometer data for the sake of the comparison, however, the initial high time resolution of the AMS might still lead to very short-termed peaks in the AMS signal which might not be caught by the Aethalometer. To adjust the times very precisely to when the Aethalometer is measuring absorption at 880 nm lies outside the scope of this paper.

Supplement, line 93: The reference spectra used in comparisons should be stated.

The following reference was added: Ng et al. (2011)

Figure S18: I would recommend that the authors try values lower than -1 for f_{peak} and try to identify the point at which the solutions begin to change.

PMF was run on the HR data for $f_{peak} = -1$ until $f_{peak} = -5$. For $f_{peak} < -1$, $Q/Q_{expected}$ starts to increase slightly. The dependence on f_{peak} of the mass spectral pattern and the mass loading is also very weak for $f_{peak} < -1$. The following sentence was added to the figure caption of Fig. S18: "For $f_{peak} < -1$, $Q/Q_{expected}$ starts to increase again (not shown)."

Technical comments

Page 27390, line 6: The 1.107 and 1.108 are two different models of GRIMM dust monitor. The authors should check which instrument was used here.

The model used here was 1107. The corresponding text in brackets was changed to "Grimm Labortechnik GmbH & Co. KG; model 1.107".

Page 27390, line 15: The authors should give a typical relative humidity of the sample line if available.

The relative humidity of the sample line was unfortunately not logged.

Page 27390, line 23: References and/or web addresses should be given for the SQUIRREL and PIKA software.

The following reference was added:

Sueper, D.: ToF-AMS High Resolution Analysis Software – Pika, online available at: <http://cires.colorado.edu/jimenez-group/ToFAMSResources/ToFSoftware/PikaInfo/>, 2008.

Page 27392, line 21: Rather than 'a' and 'b', the authors should specify 'intercept' and 'slope'.

“a” and “b” were replaced by “intercept” and “slope”, respectively.

Page 27395, line 3: The C_nH_{2n-1} series does not necessarily come from alkenes and cycloalkanes as stated. Alkanes also contribute to these peaks.

The sentence was rewritten as following: “The substantial signal at the ion series C_nH_{2n+1}⁺ (m/z 29, 43, 57, 71,...) and C_nH_{2n-1}⁺ (m/z 41, 55, 69,...) from saturated alkanes, alkenes, and cycloalkanes, [...]”.

Page 27396, line 12: The sentences “The time series babs(880 nm)traffic and the HOA time series show a similar trend. However, the R² value of 0.17 is too low to confirm correlation.” Seem somewhat contradictory.

We agree that this text seems contradictory. However, a possible explanation is already given in the original text of the manuscript (p. 27396, lines 13 – 19): “This can be due to a lower time resolution of the aethalometer compared to the AMS or peaks in the organics time series assigned to HOA without or with lower concurrent BC emissions (i.e. possible variations in HOA/BC emission ratios of various vehicles or other fossil sources in the area). Removing 18 data points from spikes in the HOA time series (see Fig. S24) increases the R² value to 0.52.”

Figure 6: Correct 'Allan et al. 2009' to 'Allan et al. 2010'.

The reference in Fig. 6 was changed to “Allan et al., 2010”.

Figure S2: The parameters used to estimate PM₁ from the GRIMM should be given.

GRIMM PM₁ was gravimetrically corrected using the following formula:
PM₁[corrected]=(PM₁[uncorrected] - (-0.0024))/0.8236

This information was added to the description of the Grimm PM₁ data in the manuscript.

Responses to

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

Received and published: 5 December 2011

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.

We thank Referee #2 for his or her thorough review of the paper. We will first address the specific comments individually and then reply on the technical comments further below.

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.

The corresponding section on HR solution criteria (3.4 in the SI) was moved to the main manuscript and introduced as following: “For a discussion of the number of factors chosen and the criteria used to select the best UMR solution, we again refer the reader to the SI, section 3.3. HR solution criteria are briefly outlined here, figures can be found in the SI, section 3.4. The chosen 6-factor solution for the HR dataset is presented in Fig. S15. The two SV-OOA factors were recombined into a single SV-OOA factor using the sum of the time series and the concentration-weighted average of the spectra. The 5-factor solution (Fig. S16) was discarded due to the high similarity of two factors (spectra and time series). The 7-factor solution (Fig. S17) features three LV-OOA spectra: PMF seems to assign an individual LV-OOA factor to three different sections in the time series, which can be due to small variations in instrument tuning with time. As the variation in the solution space $p = 6$ as a function of f_{peak} , a user-specified rotational parameter, is negligible (Figs. S18 – 19), the most central solution ($f_{peak} = 0$) was chosen and different *SEED* values (random initial values) were explored (Figs. S20 – 21). *SEED* = 46 was chosen as the best solution due to correlations with reference spectra, even though it exhibits the highest $Q/Q_{expected}$ value (12.75 compared to ~12.55 for *SEED* ≠ 46) and is thus not the mathematically optimal solution (see SI 3.3). A boxplot of the scaled residuals (boxes are +/- 25% of points) per m/z is shown in Fig. S22, time series of the residuals and $Q/Q_{expected}$ are shown in Fig. S23.

$Q/Q_{\text{expected}} \gg 1$ (12.75) indicates an underestimation of the number of factors or of the errors in the input data, possibly due to fitting errors in the HR data which are currently not included in the total error estimation. In addition to $p > 6$ (which was explored and shown not to give a more plausible solution, see above), the introduction of a “model error term” might also reduce Q/Q_{expected} , however, as discussed in Ulbrich et al. (2009), this is usually not applied in AMS data sets. Q/Q_{expected} values $\gg 1$ for HR-ToF-AMS data have been shown recently by other authors, e. g. Allan et al. (2010) published a Q/Q_{expected} value of 10.5.“

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/Q_{expected} , 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.

We meant indeed $SEED = 46$, and corrected that in the text which is now to be found in the main manuscript (see comment above). This solution has the highest Q/Q_{expected} (12.75), however it is not that different from the solutions with $SEED$ values other than 46 (~12.55). The comparison to ancillary data is a very important quality measure for the PMF solution chosen and gives more information on atmospherically relevant processes than the mathematical criteria for the best fit. The choice of ancillary data for the present PMF solution is appropriate and comparable to other studies (e. g. Lanz et al., 2007; Aiken et al., 2009). To not just glance over this issue, the corresponding sentence within the PMF solution discussion was modified as following (see above): “ $SEED = 46$ was chosen as the best solution due to correlations with reference spectra, even though it exhibits the highest Q/Q_{expected} value and is thus not the mathematically optimal solution”. For the comment on the mathematical model or error estimation, see below.

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

$Q/Q_{\text{expected}} \gg 1$ indicates an underestimation of errors in the input data, possibly due to fitting errors in the HR data which are currently not included in the total error estimation. A number of factors > 6 was explored and shown not to give a feasible solution (see reply to comment above). The introduction of a “model error term” might also reduce Q/Q_{expected} , however, as discussed in Ulbrich et al. (2009), this is usually not applied in AMS data sets. Q/Q_{expected} values $\gg 1$ for HR-ToF-AMS data have been shown recently by other authors, e. g. Allan et al. (Allan et al., 2010) published a Q/Q_{expected} value of 10.5. This discussion was implemented in the text on the PMF solution criteria in the main manuscript, see reply to a).

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.

The corresponding sentence was changed as following: “Considering the OM:OC ratios displayed in Fig. 2 of the three primary-dominated components (BBOA, HOA, and COA), and assuming that all the carbon in HOA is fossil and all the carbon in BBOA and COA is non-fossil, we can estimate that 59% of the carbon in POA in Barcelona is non-fossil while 41% is fossil.”

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).

With photochemical aging, the spectra of organic aerosol sources measured by AMS become progressively similar and lose their source fingerprint (Andreae, 2009; Capes et al., 2008): They resemble OOA, related to SOA (Zhang et al., 2005). The different spectra of HOA, COA, and BBOA are clearly different from OOA and indicate that they have an origin that is not related to SOA formation, thus of primary emissions. In addition, the similarity of the COA and BBOA factor found here with measured spectra of primary cooking and biomass burning emissions (e. g. Mohr et al., 2009; Weimer et al., 2008) confirms their identification as POA, even if they may have undergone a small degree of aging after being emitted.

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.

The corresponding section in the 1st paragraph of the introduction was changed as following: “However, knowledge about sources, fate and mutual interaction of gas-phase and aerosol organics, of which it is estimated that there are between 10 000 and 100 000 different compounds is still limited (Goldstein and Galbally, 2007). Therefore, the atmospheric evolution of organic aerosol (OA) is the focus of intense research activities (Jimenez et al., 2009). The complexity of the atmospheric (organic) aerosol system places great demands on measurement techniques and instrumentation (Hallquist et al., 2009).”

- 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.

The sentence was changed to “[...] organosulfates and organonitrates may also contribute [...]”.

- 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.

On p. 27393, line 23, and p. 27399, line 20, “chemical composition” was changed to “dry chemical composition”.

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

Since those correlations are presented in the supplementary information of the DAURE overview paper (Pandolfi, M., Querol, X., Alastuey, A., Jimenez, J. L., Cusack, M., Reche, C.,

Pey, J., Mohr, C., DeCarlo, P. F., Ortega, A., Day, D., Prevot, A. S. H., Baltensperger, U., Artiñano, B., Baldasano, J. M., Jorba, O., Burkhardt, J., Hansel, A., Schallhart, S., Müller, M., Metzger, M., Saarikoski, S., Cubison, M. J., Ng, S., Lorente, J., Nemitz, E., Di Marco, C., Peñuelas, J., Sicard, M., Comeron, A., Amato, F., Moreno, T., Viana, M., Pérez, N., Moreno, N., Seco, R., Filella, I., Llusà, J., Piot, M., and Pay, M. T.: Sources and origin of PM in the Western Mediterranean Basin: An overview of the DAURE campaign, in preparation, 2011.), they are not included in the present manuscript.

- 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.*

To refrain from a qualitative statement, p. 27396, lines 12-19 were rephrased as following: “[...] and the HOA time series show a similar trend. However, there are spikes in the HOA time series not caught by the aethalometer model. This can be due to a lower time resolution of the aethalometer compared to the AMS or peaks in the organics time series assigned to HOA without or with lower concurrent BC emissions (i. e. possible variations in HOA/BC emission ratios of various vehicles or other fossil sources in the area). Removing 18 data points from spikes in the HOA time series (see Fig. S24) increases the R^2 value from initially 0.17 to 0.52.”

- 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?)*

“traffic” was replaced by “road traffic” throughout the manuscript when ship emissions were specifically excluded.

- 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.*

“is linear” was changed to “is assumed to 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.*

“first-generation” was changed to “early-generation”.

- 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.*

We agree that different AMSs can yield somewhat different mass spectral signatures. The coefficients q , a , b , c , and d were calculated based on PMF analyses of measurements by 3 different instruments. They are given with \pm one standard deviation, the values of which vary between 18 and 100% of the coefficient value. To consider this important point, the last sentence of this section was changed as following: “The application of this approach to other

datasets and the comparison of coefficient values will be of great interest, as discrepancies can also be influenced by the variation of mass spectral patterns of different instruments.”

Technical corrections

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

O/C was changed throughout the manuscript to O:C.

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

The sentence was rewritten to “A growing number of measurements in very diverse environments (e. g. Aiken et al., 2009; Huang et al., 2010) and increasing resolution of the AMS (DeCarlo et al., 2006) continuously enhance our knowledge of OA factors.”

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

“specially” was changed 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.

The Wiedensohler et al., 2011 reference was added to the reference list and the reference list was checked 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.

The sentence was changed to “[...] indicate that the temporal evolution of both components is driven by similar processes such as primary emissions, rapid chemical formation, and the diurnal cycles of land/sea breeze and boundary layer height rather than gas-to-particle partitioning based on volatility characteristics.”

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

“m/z” was italicized.

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

“emissions site” was changed to “emission sites”.

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

“relied” was changed to “related”.

References

<http://www.eol.ucar.edu/projects/milagro/>, access: August 13.

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