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Supplement of

Characterization of fresh and aged organic aerosol emissions from meat charbroiling

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1. PMF analysis for smog chamber Experiments 1-4

PMF using PET (Ulbrich et al., 2009) was applied to the AMS results of each of the 4 experiments separately. The selection of the solution was based on the characteristics of their mass spectra and corresponding time series. The model residuals for the 1 to 3 factors solutions are illustrated in Figures S1-S4. Moving from 1 to 2 factors resulted in a significant reduction of the residuals. The 3-factor solution residuals were slightly lower than those of the 2-factor solution but the improvement was marginal. The 3-factor solution is discussed in a subsequent section.

The corresponding $Q/Q_{exp}$ versus the number of the factors is shown in Figures S5-S8. For the 2 factor solution we selected an $f_{peak}$ value in the range in which the solution was stable. Figures S9-S12 depict the $Q/Q_{exp}$ versus the $f_{peak}$ value for each experiment. The stable $f_{peak}$ regions for Experiments 1-4 were -2.0-0.4, -0.2-1.0, -1.0-1.0 and 0.0-1.0 respectively and the selected $f_{peak}$ values were -0.6, 0.0, -0.4 and 0.0.

In all four cases the 3-factor solution resulted in a fresh, an intermediate and an aged factor. The corresponding time series and mass spectra are shown in Figures S13 and S16. Moving to 4 factors one of the factors was split into two parts. Figure S17 and S18 shows the time series and the mass spectra of the 4-factor PMF solution for experiment 2, where the intermediate factor was split into two factors with similar time series and mass spectra.
Figure S1. Model residuals $E = X - GF$ calculated using the PMF evaluation tool, PET (Ulbrich et al., 2009) for Experiment 1. Comparison between: (a) 1-factor (purple lines) and 2-factor (pink lines) PMF solution and (b) 2-factor (pink lines) and 3-factor (black lines) solution. The residuals decreased significantly moving from 1 to 2 factors.
Figure S2. Model residuals $E = X - GF$ calculated using the PMF evaluation tool, PET (Ulbrich et al., 2009) for Experiment 2. Comparison between: (a) 1-factor (purple lines) and 2-factor (pink lines) PMF solution and (b) 2-factor (pink lines) and 3-factor (black lines) solution.
Figure S3. Model residuals $E = X - GF$ calculated using the PMF evaluation tool, PET (Ulbrich et al., 2009) for Experiment 3. Comparison between: (a) 1-factor (purple lines) and 2-factor (pink lines) PMF solution and (b) 2-factor (pink lines) and 3-factor (black lines) solution.
Figure S4. Model residuals $\mathbf{E} = \mathbf{X} - \mathbf{GF}$ calculated using the PMF evaluation tool, PET (Ulbrich et al., 2009) for Experiment 4. Comparison between: (a) 1-factor (purple lines) and 2-factor (pink lines) PMF solution and (b) 2-factor (pink lines) and 3-factor (black lines) solution.
Figure S5. $Q/Q_{\text{exp}}$ versus the number of the PMF factors for Experiment 1.

Figure S6. $Q/Q_{\text{exp}}$ versus the number of the PMF factors for Experiment 2.
**Figure S7.** $Q/Q_{\text{exp}}$ versus the number of the PMF factors for Experiment 3.

**Figure S8.** $Q/Q_{\text{exp}}$ versus the number of the PMF factors for Experiment 4.
Figure S9. $Q/Q_{\text{exp}}$ for $f_{\text{peak}}$ values from -2 to 2 for the 2-factor solution for Experiment 1. The stable solution corresponds to $f_{\text{peak}}$ values between -2.0 and 0.4. We selected $f_{\text{peak}} = -0.6$.

Figure S10. $Q/Q_{\text{exp}}$ for $f_{\text{peak}}$ values from -2 to 2 for the 2-factor solution for Experiment 2. The solution is stable for $f_{\text{peak}}$ values between -0.2 and 1. We selected $f_{\text{peak}} = 0.0$. 

Figure S11. $Q/Q_{\text{exp}}$ for $f_{\text{peak}}$ values from -2 to 2 for the 2-factor solution for Experiment 3. The solution is stable for $f_{\text{peak}}$ values between -1.0 and -1.0. We selected $f_{\text{peak}}=-0.4$.

Figure S12. $Q/Q_{\text{exp}}$ for $f_{\text{peak}}$ values from -2 to 2 for the 2-factor solution for Experiment 4. The solution is stable for $f_{\text{peak}}$ values between 0.0 and 1.0. We selected $f_{\text{peak}}=0.0$. 
Figure S13. PMF factor time series for Experiments: (a) 1 and (b) 2 for the 3-factor solution.
Figure S14. PMF factor time series for Experiments: (a) 3 and (b) 4 for the 3-factor solution.
Figure S15. PMF factor mass spectra for Experiments: (a) 1 and (b) 2 for the 3-factor solution.
Figure S16. PMF factor mass spectra for Experiments: (a) 3 and (b) 4 for the 3-factor solution.
Figure S17. Time series of the concentrations of the PMF factors for Experiment 2 for the 4-factor solution. The intermediate factor has been split into two factors with similar time series.
Figure S18. PMF factor mass spectra for Experiment 2 for the 4-factor solution. The intermediate factor has been split into two factors with similar mass spectra.
2. PMF analysis for the ambient measurements

Figure S19 depicts the model residuals for solutions with 1 to 5 factors using PET (Ulbrich et al., 2009). There is an important decrease in the residuals moving from 3 to 4 factors, but there is little change between 4 and 5 factors. Thus a 4 factor solution was selected.

The $Q/Q_{\text{exp}}$ versus the number of the factors is illustrated in Figure S20. Figures S21 depicts the $Q/Q_{\text{exp}}$ versus $f_{\text{peak}}$. The solution was quite stable for $f_{\text{peak}}$ values from -0.6 to 0.8 and therefore we selected $f_{\text{peak}}=0.0$.

Moving to 5 factors the otBB-OA factor was split into two similar factors. Figure S22 and S23 show the corresponding mass spectra and time series for this case.
Figure S19. Model residuals $E = X - GF$ calculated using the PMF evaluation tool, PET (Ulbrich et al., 2009) for the ambient measurements. Comparison between: (a) 1-factor (purple lines) and 2-factor (pink lines) PMF solutions, (b) 2-factor (pink lines) and 3-factor (black lines) solutions, (c) 3-factor (black lines) and 4-factor (red lines) solutions, and (d) 4-factor (red lines) to 5-factor (green lines) solutions. The residuals decreased significantly moving from 3 to 4 factors, but there was no significant change from 4 to 5 factors.
Figure S20. $Q/Q_{\text{exp}}$ versus the number of factors for the ambient measurements.

Figure S21. $Q/Q_{\text{exp}}$ for $f_{\text{peak}}$ values -2 to 2 for a 4 factor solution for the ambient measurements. The solution was stable for $f_{\text{peak}}$ values between -0.6 and 0.8. We selected $f_{\text{peak}}=0.0$. 
Figure S22. The PMF mass spectra for 5-factor solution which resulted in two otBB-OA factors with similar spectra.
Figure S23. The time series of the 5 factors for the corresponding solution in which otBB-OA was split into two factors with similar quite noisy time series.
3. PMF analysis for the ambient measurements using ME-2

ME-2 (Canonaco et al., 2013) was also used for the analysis of the AMS field data using the HR organic mass spectra. First we examined the 4-factor solution without using an external factor profile. The resulting four factors were almost identical to those derived using PMF/PET. Figure S25 shows the comparison of the corresponding time series resulting from the two algorithms. The $R^2$ were higher than 0.99. Figure S26 depicts the four ME-2 spectra. The $R^2$ between the corresponding mass spectra using PET was again higher than 0.99 ($\theta<2$ degrees).

After this test ME-2 was used again applying a constrained solution for the HOA using the HOA mass spectrum of Kostenidou et al. (2013) with $a=0.1$. The corresponding four time series are shown in Figure S27. The OOA, otBB-OA and COA times series did not change ($R^2>0.99$) for all practical purposes, but the HOA times series had a lower correlation ($R^2=0.79$) with the unconstrained HOA solution. Figure S28 shows the four mass spectra using the constrained solution, which are practically the same with the unconstrained factors ($R^2>0.99$, $\theta<2$ degrees).
Figure S24. $Q/Q_{\text{exp}}$ for 20 iterations for the 4-factor solution for the ambient measurements. The solution is quite stable.
Figure S25. Time series of the 4 factors using PMF/PET (solid lines) and ME-2 (dotted lines). The results are almost identical with $R^2 > 0.99$. 
Figure S26. Mass spectra of the four factors using ME-2. They are quite similar to those derived using PMF/PET with $\theta<2$ degrees ($R^2>0.99$).
Figure S27. Time series of the four factors using ME-2 for an unconstrained solution (dotted lines) and a constrained solution (solid lines). The results were almost the same for the OOA, otBB-OA and COA factors. However, for the HOA there were small differences.
Figure S28. Mass spectra of the four factors using a constrained solution in ME-2. The resulting 4 mass spectra were practically the same with those of the unconstrained solution ($R^2 > 0.99$, $\theta < 2$ degrees).
4. OA concentration change during the experiments

(a) 1st experiment
- Concentration (µg m⁻³) over time (h)
- Dilution step
- UV 'on'

(b) 2nd experiment
- Concentration (µg m⁻³) over time (h)
- UV 'on'

(c) 3rd experiment
- Concentration (µg m⁻³) over time (h)
- +O₃

(d) 4th experiment
- Concentration (µg m⁻³) over time (h)
- UV 'on'
Figure S29. Measured and wall loss corrected OA concentration for the five experiments.