

**Aerosol Characterization over the North China Plain: Haze Life Cycle and Biomass Burning Impacts in Summer**

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**Introduction**

Figures S1 – S5 show aerosol particle acidity, the inter-comparison between SMPS and ACSM measurements, diurnal cycles of PM<sub>1</sub> species, diurnal cycles of OA factors, diurnal cycles of PM<sub>1</sub> species// $\Delta$ CO, and fire spots from MODIS satellites. Figures S7 – S13 present the key diagnostic plots for the evaluation of the PMF solution.

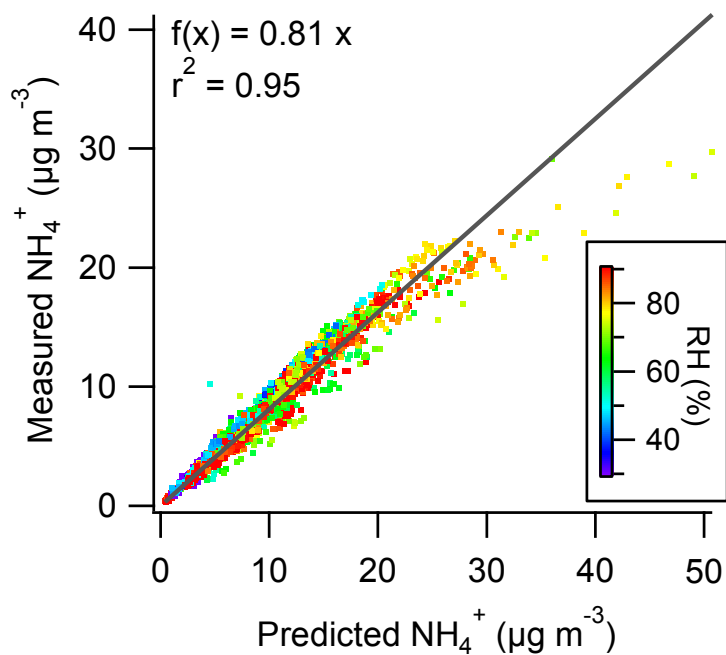


Figure S1. Scatter plot of measured  $\text{NH}_4$  to predicted  $\text{NH}_4$  ( $=\text{SO}_4/96 \times 36 + \text{NO}_3/62 \times 18 + \text{Cl}/35.5 \times 18$ ) that requires to fully neutralize  $\text{SO}_4$ ,  $\text{NO}_3$ , and  $\text{Cl}$  [Zhang *et al.*, 2007].

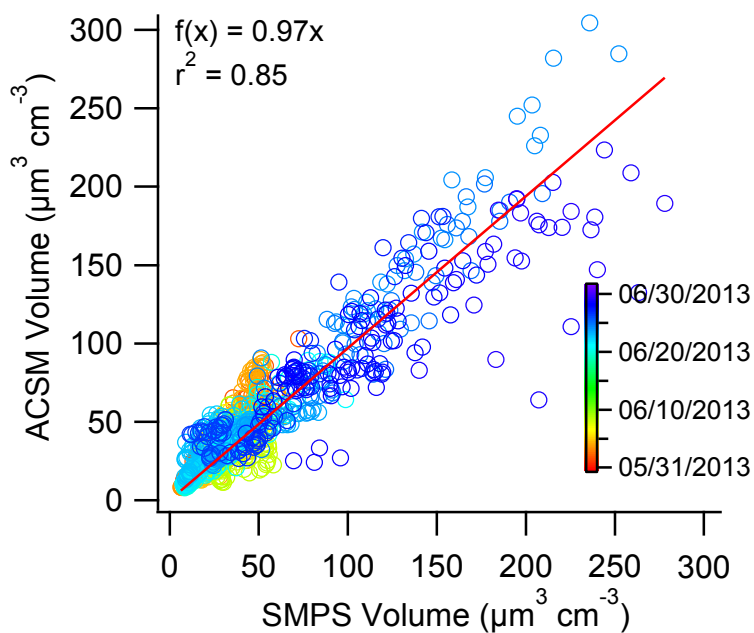


Figure S2. A comparison of the ACSM volume concentrations with those from the SMPS measurements (14 – 760 nm). The ACSM volume concentrations were calculated using the estimated densities on the basis of chemical composition of  $\text{PM}_{10}$  [Salcedo *et al.*, 2006].

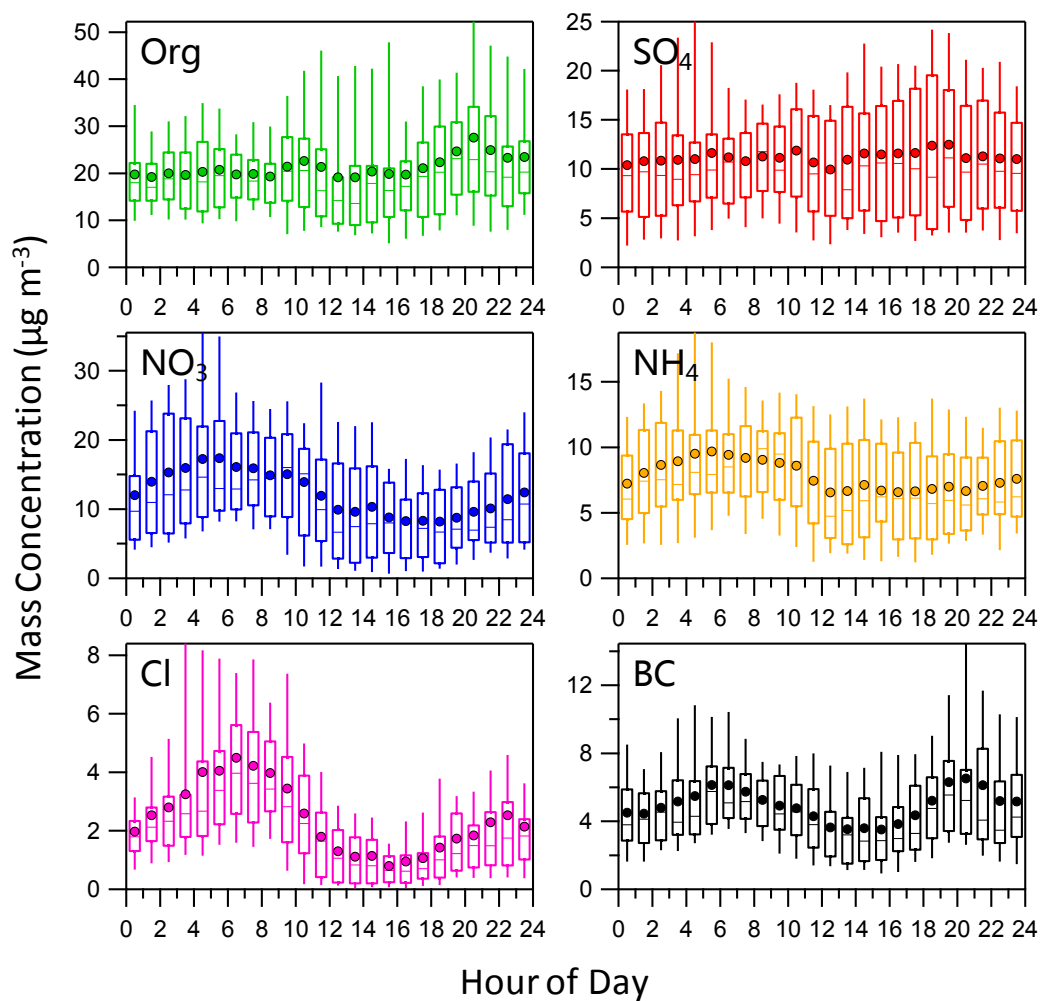


Figure S3. Diurnal cycles of aerosol species during NBB period. The mean (solid circles), median (horizontal line), 25th and 75th percentiles (lower and upper box), and 10th and 90th percentiles (lower and upper whiskers) are shown for each box.

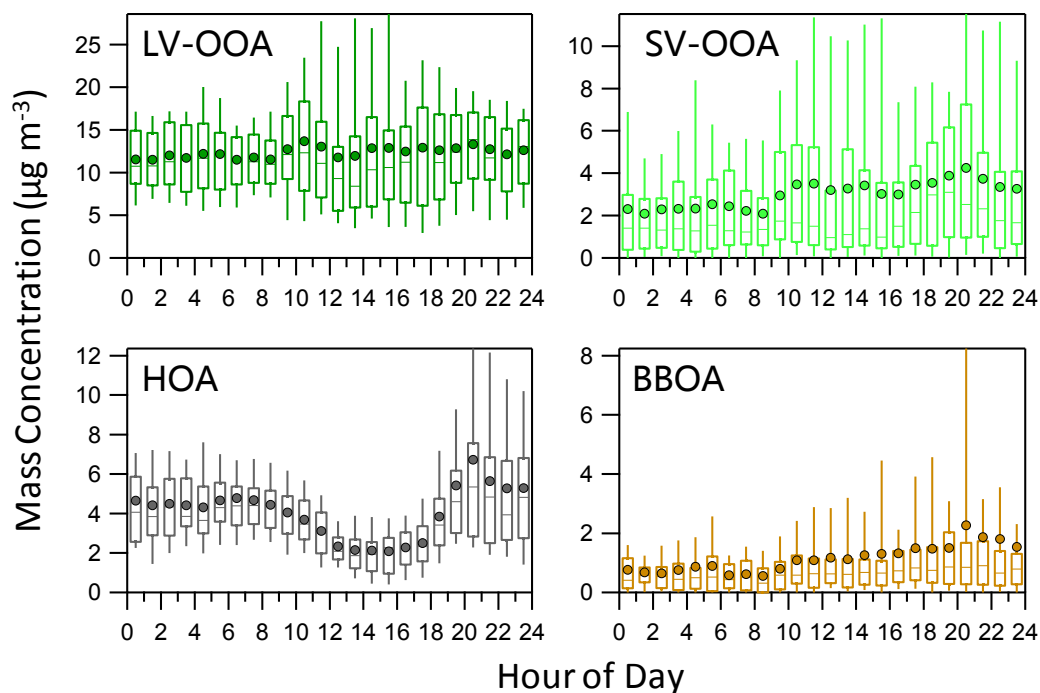


Figure S4. Diurnal cycles of OA factors during NBB period. The mean (solid circles), median (horizontal line), 25th and 75th percentiles (lower and upper box), and 10th and 90th percentiles (lower and upper whiskers) are shown for each box.

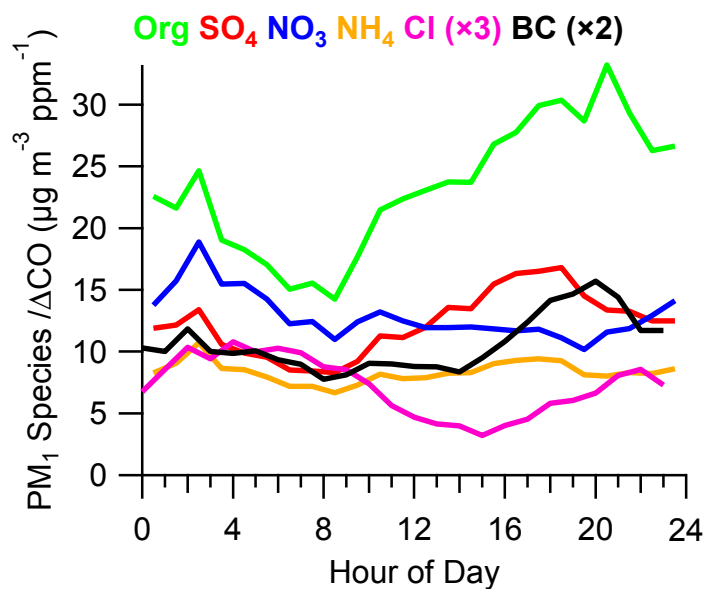


Figure S5. Diurnal cycles of submicron aerosol species/ $\Delta$ CO.  $\Delta$ CO was calculated by subtracting the background concentration of CO (0.544 ppm) which was determined as the average of the lowest 5% data.



Figure S6. Fire spots detected by the MODIS Terra and Aqua satellites during the BB period (June 23 – 29, 2013). We acknowledge the use of FIRMS data and imagery from the Land, Atmosphere Near real-time Capability for EOS (LANCE) system operated by the NASA/GSFC/Earth Science Data and Information System (ESDIS) with funding provided by NASA/HQ.

### **Evaluation and Selection of PMF Solutions.**

PMF analysis of ACSM organic aerosol spectra identified four OA factors, including hydrocarbon-like OA (HOA), biomass burning OA (BBOA), semi-volatile oxygenated OA (SV-OOA), and low volatility OOA (LV-OOA). The mass spectral profiles and time series of the four OA factors are shown in Figure 5. The two-factor solution resolved a primary OA (POA) and a secondary OA (SOA) factor. As indicated in Figure S7, the mass spectrum of POA was mixed with various primary sources, e.g., traffic and biomass burning. The three-factor solution resolved two SOA factor and a POA factor (Figure S8). Still, the traffic related HOA and BBOA cannot be separated. As a result, the four-factor solution which can be reasonably interpreted was chosen in this study. Figure S9 presents a detailed summary of key diagnostics plots for the four-factor solution. After a careful evaluation the mass spectral profiles and time series (Figure S10), the PMF solution at  $f_{\text{Peak}} = 0.8$  was chosen in this study.

As shown in Figure 2, the concentration of organics was significantly elevated during the BB period. Therefore, for a better evaluation of the PMF solution, PMF analysis was also performed on the OA during the NBB period. Three factors, including a HOA, a SV-OOA, and a LV-OOA were identified. The comparisons of mass spectral profiles and time series with those from PMF analysis of the entire dataset are shown in Figures S11-S13. The PMF results were overall consistent. The mass spectral profiles of HOA, SV-OOA and LV-OOA from PMF analysis of the NBB dataset were highly similar to those from PMF analysis of the entire dataset, and their time trends also tracked well. These results confirmed the rationale for the selection of 4-factor solution in this study. It should be noted that the HOA concentration from PMF analysis of the NBB dataset was overall higher than that from PMF analysis of the entire dataset, whereas SV-OOA was reversed. The reason was likely due to the influences of BBOA that was not resolved during the NBB period although it contributed a small fraction (Figure 3d).

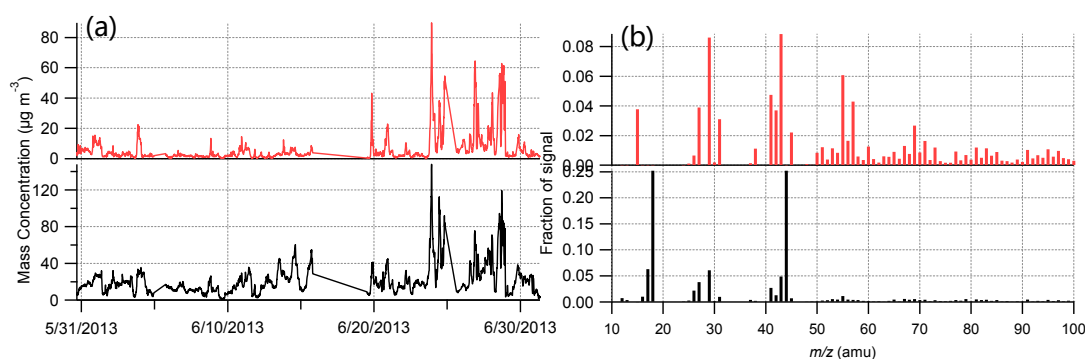


Figure S7. (a) Time series and (b) mass spectral profiles of 2-factor PMF solution.

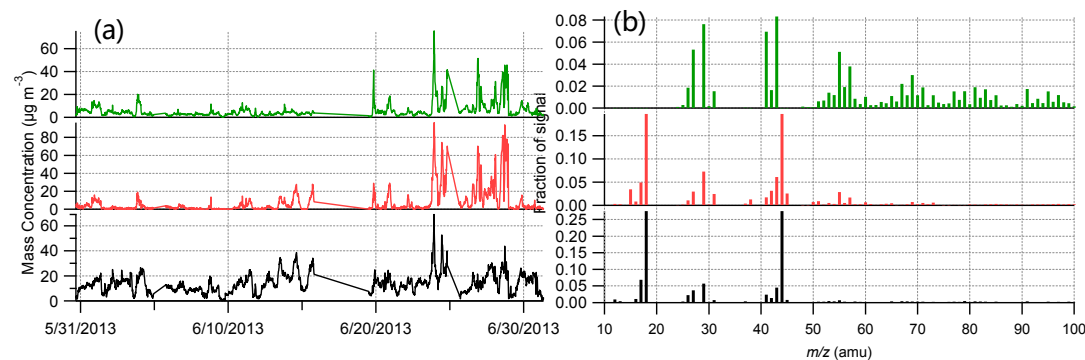


Figure S8. (a) Time series and (b) mass spectral profiles of 3-factor PMF solution.

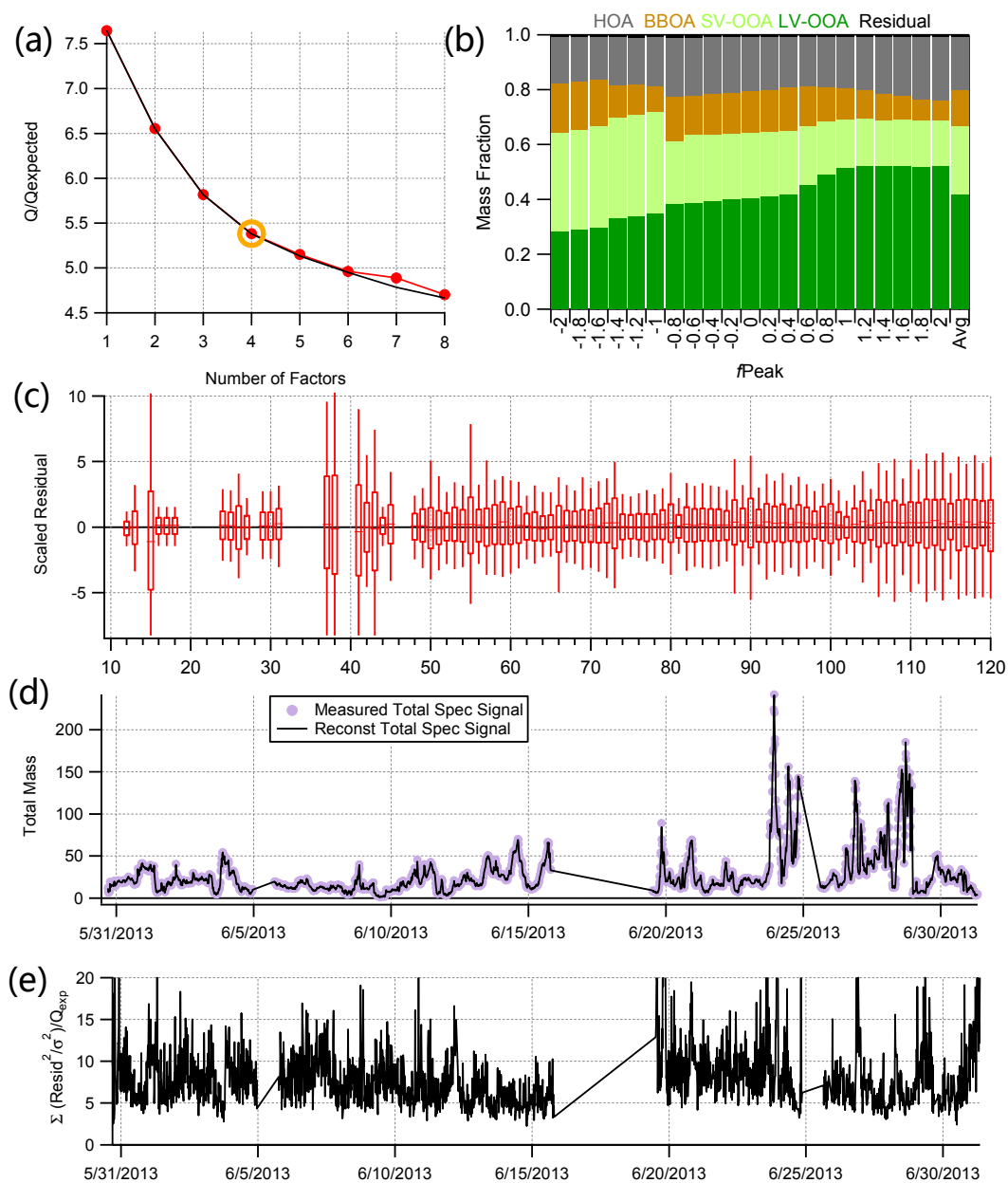


Figure S9. Summary of key diagnostic plots of the ACSM PMF results for 4-factor solution: (a)  $Q/Q_{\text{exp}}$  as a function of number of factors, (b) mass fraction of four OA factors as a function of  $f_{\text{PEAK}}$ , (c) the box and whiskers plot showing the distributions of scaled residuals for each  $m/z$ , (d) a comparison of the measured mass with the PMF reconstructed mass, (e) time series of  $Q/Q_{\text{exp}}$  for each point in time.

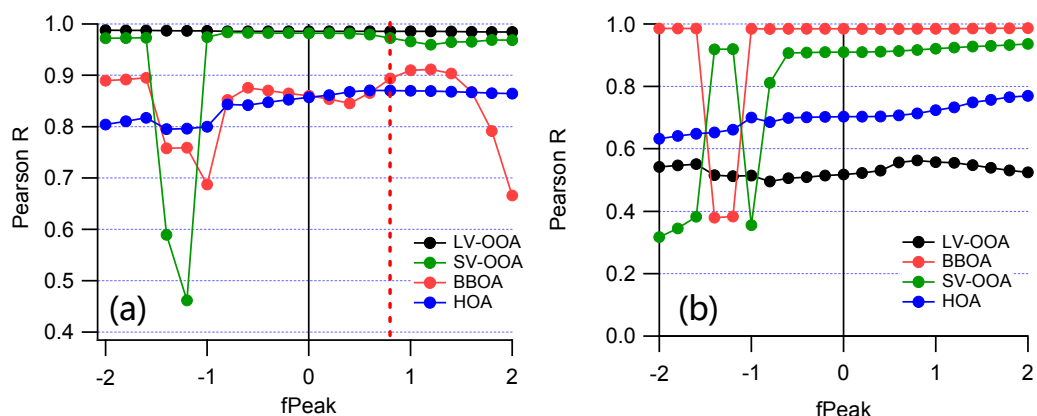


Figure S10. (a) mass spectral correlations of LV-OOA vs. fulvic acid [Zhang *et al.*, 2005], SV-OOA vs. OOA [Ng *et al.*, 2011], BBOA vs. Paris BBOA [Crippa *et al.*, 2013], and HOA vs. diesel exhaust aerosol [Canagaratna *et al.*, 2004], (b) time series correlations of LV-OOA vs.  $\text{SO}_4$ , SV-OOA vs.  $m/z$  43, BBOA vs.  $m/z$  60, and HOA vs. BC, as a function of fPeak.

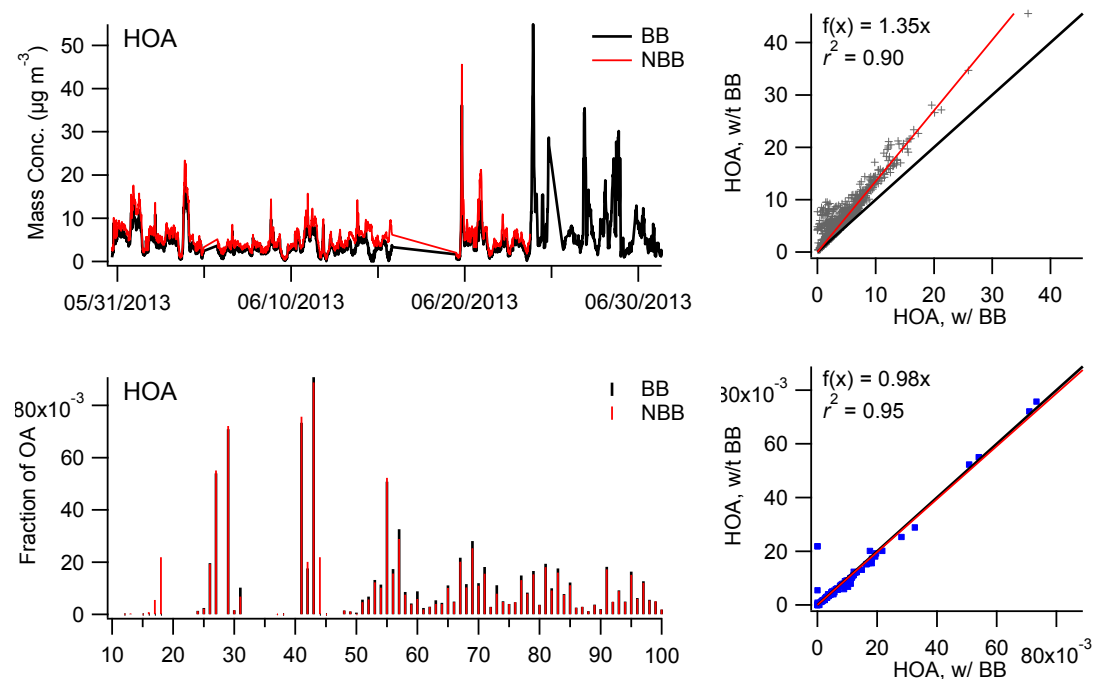


Figure S11. Comparisons of the mass spectrum and the time series of HOA from PMF analysis of the entire dataset and that by excluding the BB events.



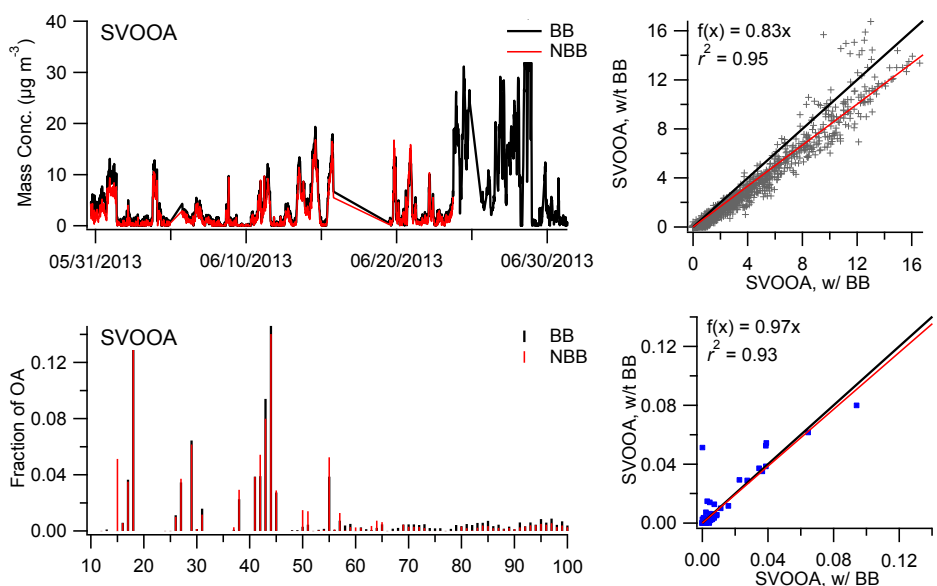


Figure S12. Comparisons of the mass spectrum and the time series of SV-OOA from PMF analysis of the entire dataset and that by excluding the BB events.

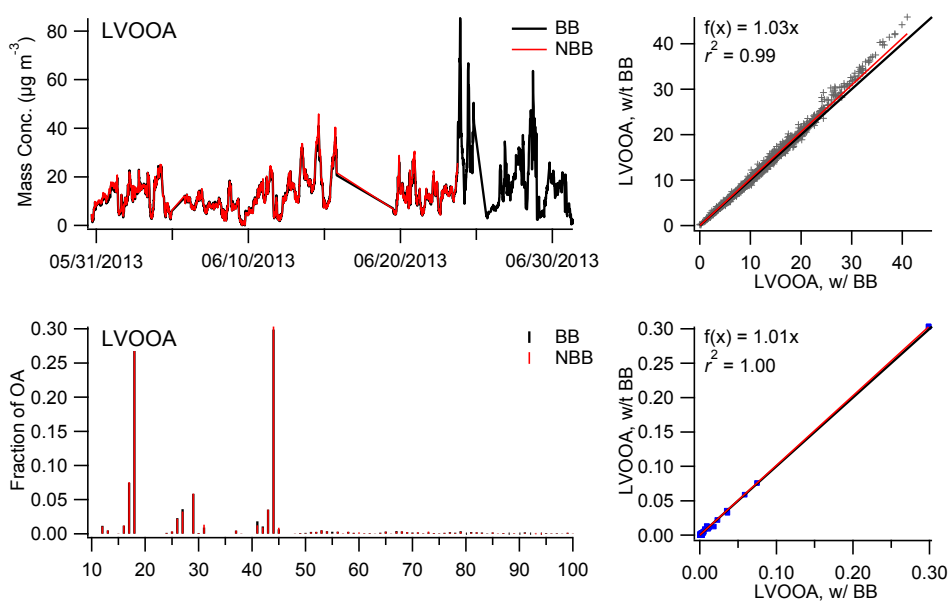


Figure S13. Comparisons of the mass spectrum and the time series of LV-OOA from PMF analysis of the entire dataset and that by excluding the BB events.

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