Supporting Information

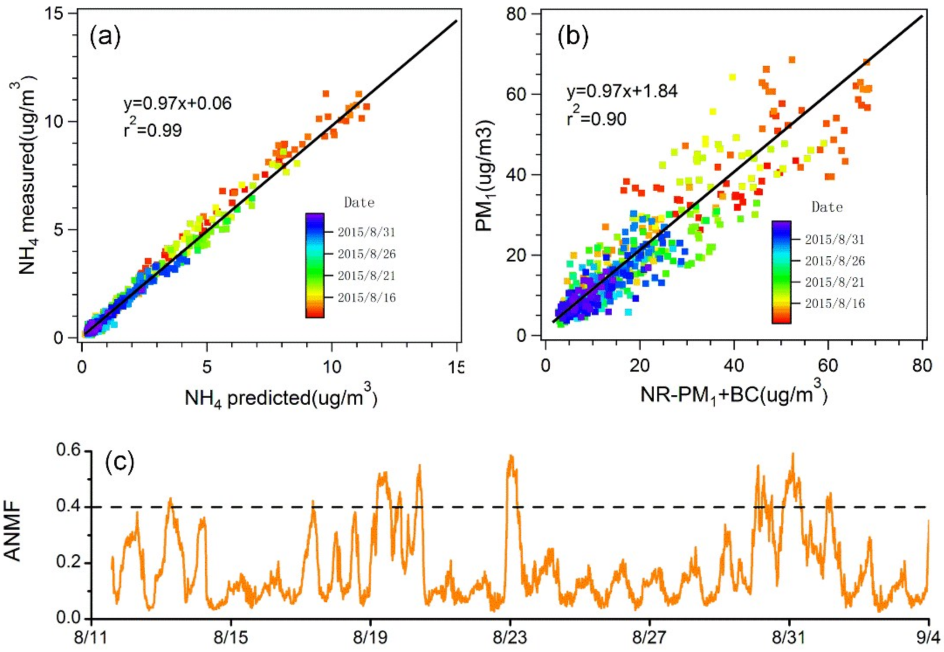

Figure S1. Scatterplots of **(a)** measured NH_4^+ versus predicted NH_4^+, **(b)** constructed PM_1 mass concentration versus total PM_1 mass measured by PM714 monitor, **(c)** temporal variations of ammonium nitrate mass fraction (ANMF).

Selection of the optimal PMF solution

PMF analysis based on the ACSM mass spectra was performed for 1 to 8 factors. Figure S2a showed Q/Q_{exp} as a function of the number of factors P. According to Ulbrich et al.(2009), values of $Q/Q_{exp} >1$ indicate underestimation of the errors or variability in the factor profiles that cannot be simply modeled as the sum of the given number of components. On the contrary, $Q/Q_{exp} <1$ means that the errors of the input data have been overestimated. As presented in Figure S2a, Q/Q_{exp} decreased with the increasing number of factors. In the two-factor solution, Q/Q_{exp} showed a large decrease from 1.37 to 0.88 compared to the one-factor solution, implying that the additional factor explained significantly more of the variation in the data. However, Factor 1 in the two-factor solution appeared to be a mixture of HOA and COA based on its diurnal cycles and mass spectra profiles. Therefore, more OA factors need to be considered. When the factor number increased to three, Factor 1 in the two-factor solution was separated into two new factors, which were identified as HOA and COA. Another factor was characterized as OOA. When the number of factors changed from four to eight, no obvious decrease of Q/Q_{exp} was observed and the mass spectra of new factors could not be well explained. For example, in the four-factor solution, a new factor which showed only an obvious peak at m/z 43 was separated. No similar mass spectra profile has been observed for organic aerosol from emission sources. Thus the three-factor solution was chosen as the optimal solution. Further, the rotational ambiguity of the three-factor solution was explored by varying FPEAK between -1.0 and +1.0 (step: 0.1). Lower Q/Qexp values can indicate a better fit to the dataset and is used as one criterion for selecting a suitable solution (Ulbrich et al., 2009). As shown in Figure 2b, the lowest Q/Qexp was obtained at approximately 0. Therefore, FPEAK = 0 was chosen as the best solution.

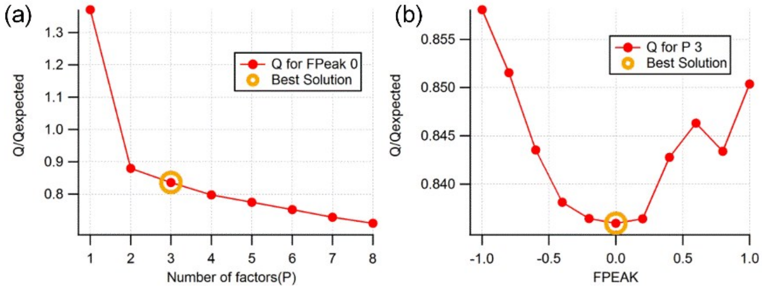

Figure S2. (a) $Q/Q_{expected}$, where Q is the sum of the squared scaled residuals over the entire dataset, plotted against the number of factors used in the positive matrix factorization (PMF) solution. (b) $Q/Q_{expected}$ plotted against the rotational forcing parameter (FPEAK) for solutions with three factors.

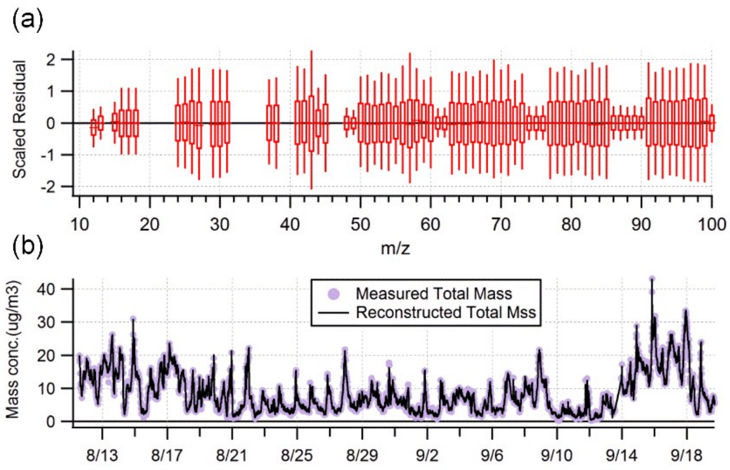

Figure S3. (a) The box and whiskers plot showing the distributions of scaled residuals for each m/z, (b) Time series of the measured organic mass and the reconstructed organic mass.

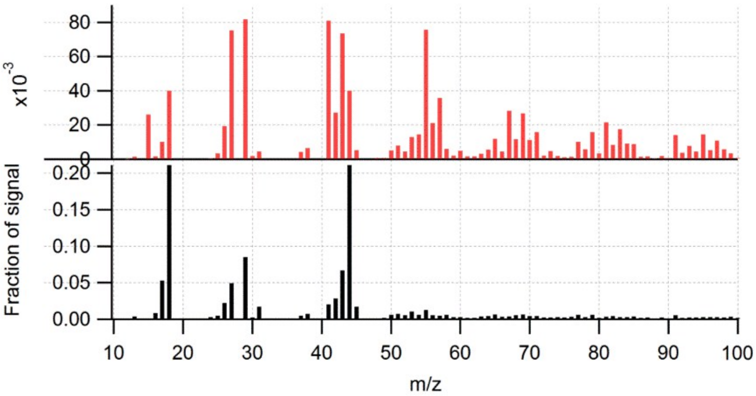

Figure S4. Mass spectra profiles of the two-factor solution.

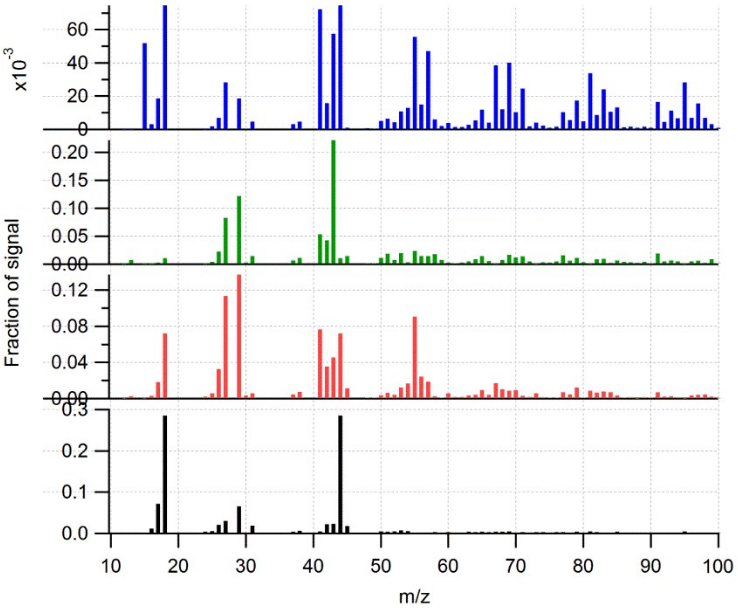

Figure S5. Mass spectra profiles of the four-factor solution.

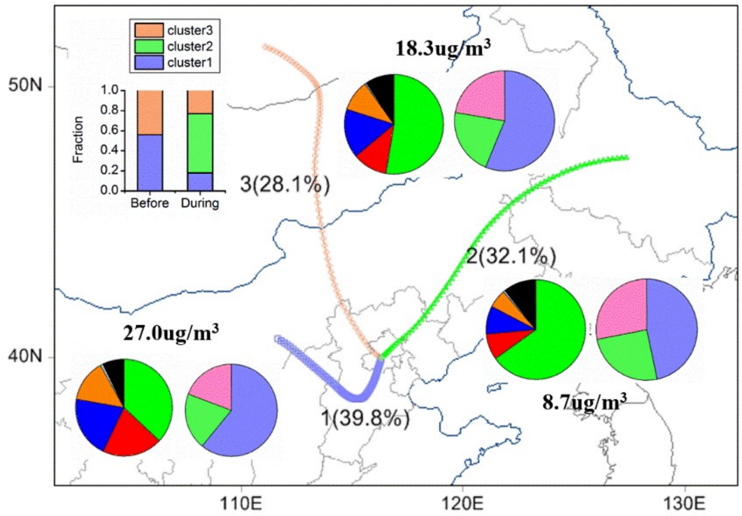

Figure S6. The back trajectory clustering analysis associated with the corresponding average PM_1 mass and composition during the campaign. The inserted bar graph shows the directional variation of BTs before and during the parade.