

1 Supplementary information of manuscript

2 **Concurrent photochemical whitening and darkening of ambient**
3 **brown carbon**

4 Qian Li¹, Dantong Liu^{1*}, Xiaotong Jiang¹, Ping Tian², Yangzhou Wu¹, Siyuan Li¹, Kang Hu¹, Quan Liu³,
5 Mengyu Huang², Ruijie Li², Kai Bi², Shaofei Kong⁴, Deping Ding²

6 ¹Department of Atmospheric Science, School of Earth Science, Zhejiang University, Hangzhou, 310027, China

7 ²Beijing Key Laboratory of Cloud, Precipitation and Atmospheric Water Resources, Beijing Meteorological Service, Beijing,
8 100089, China.

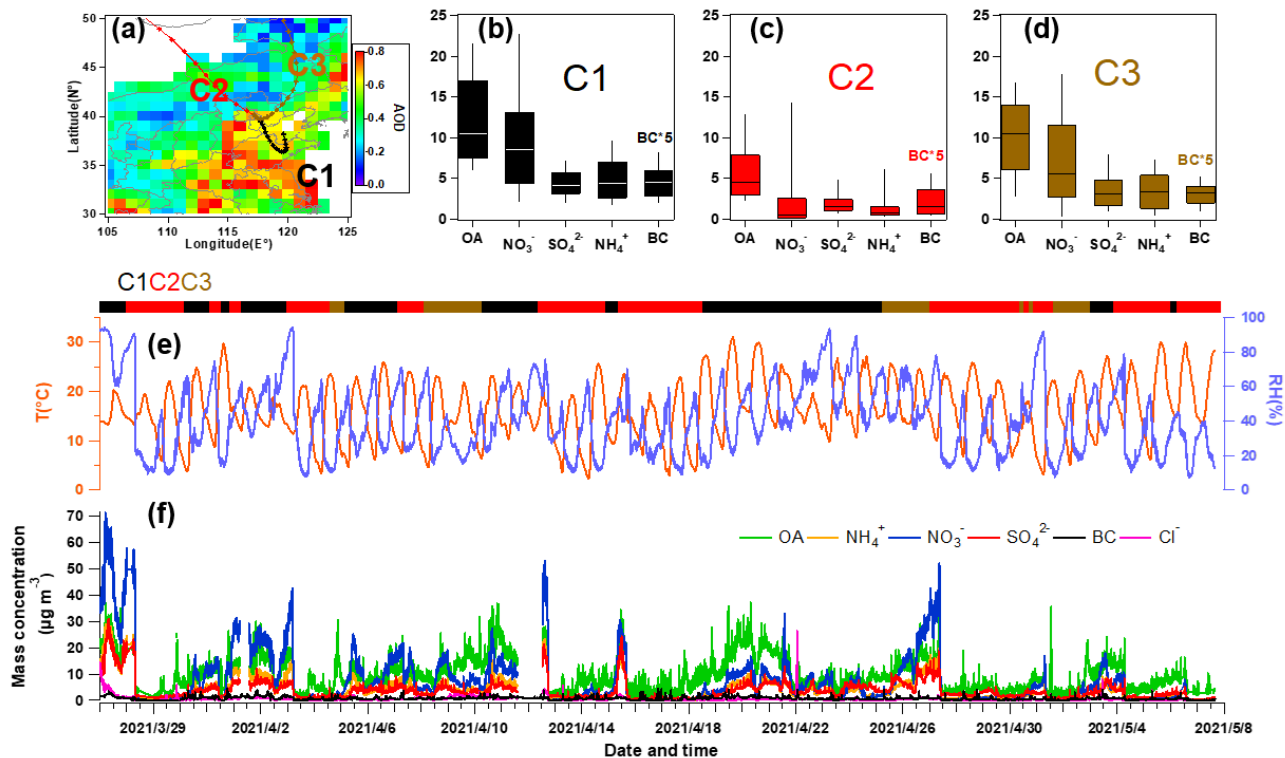
9 ³State Key Laboratory of Severe Weather & Key Laboratory of Atmospheric Chemistry of CMA, Chinese Academy of
10 Meteorological Sciences, Beijing, 100081, China

11 ⁴Department of Atmospheric Science, School of Environmental Science, China University of Geosciences, Wuhan, 430074,
12 China

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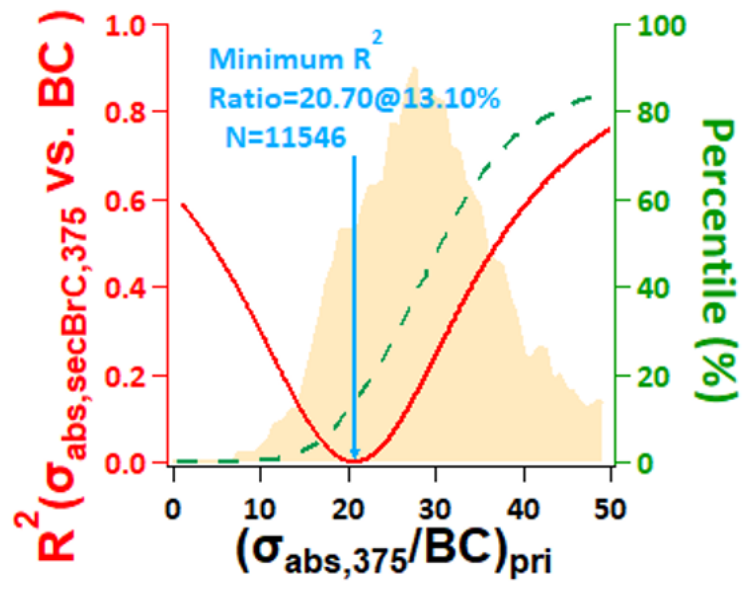
14 *Correspondence to:* Dantong Liu (dantongliu@zju.edu.cn)

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 17 **Figure S1. (a) Clustered back-trajectories for the past 72 hours during the experiment with markers denoting 12h intervals. (b-d)**
 18 **Statistics for the concentrations of key aerosol compositions from each cluster. The whiskers, box boundaries and lines in box denote**
 19 **the 10th/90th percentiles, 25th/75th percentiles and the median, respectively. (e) Time series of RH and T, (f) mass concentrations of**
 20 **key aerosol compositions.**

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23 Figure S2. Minimum R-square analysis to obtain the $(\sigma_{\text{abs},375}/BC)_{\text{pri}}$.

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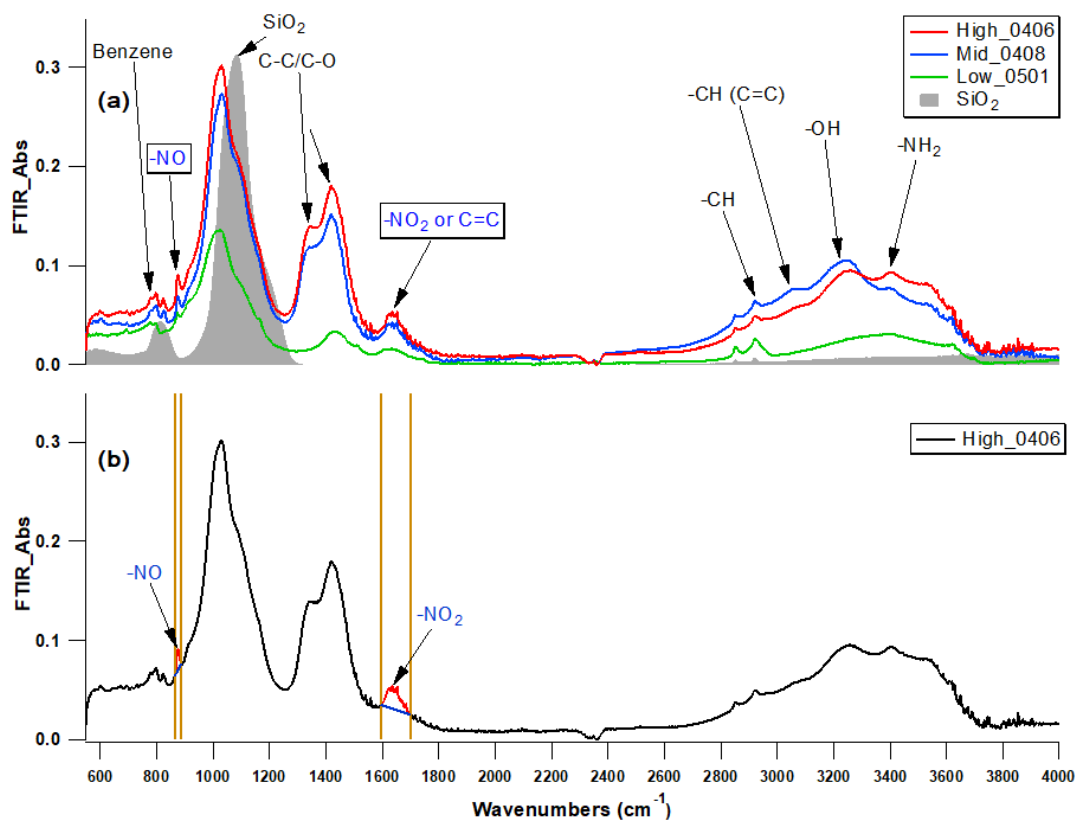
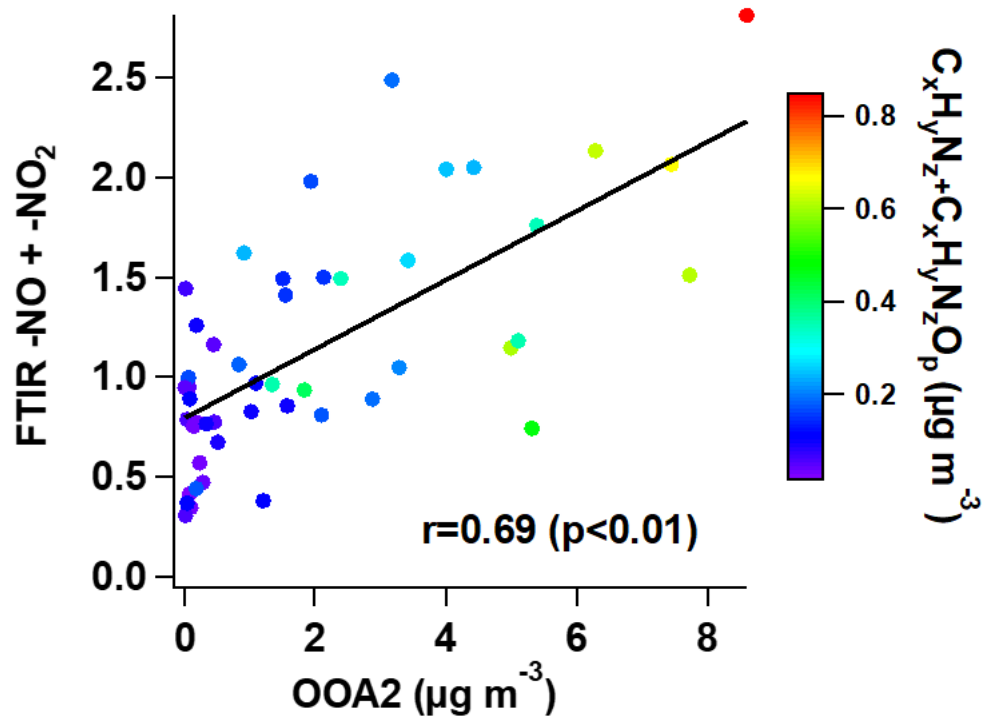


Figure S3. (a) The FTIR absorbance spectrum for the blank and filter samples during three days with different pollution levels. The main functional groups in the FTIR spectra are indicated. (b) Method of peak extraction for the -NO and -NO₂ bond.

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Figure S4. Relationship between the FTIR absorption of -NO, -NO₂ bond and the OOA2, colored by the C_xH_yN_z and C_xH_yN_zO_p fragments measured by the AMS.

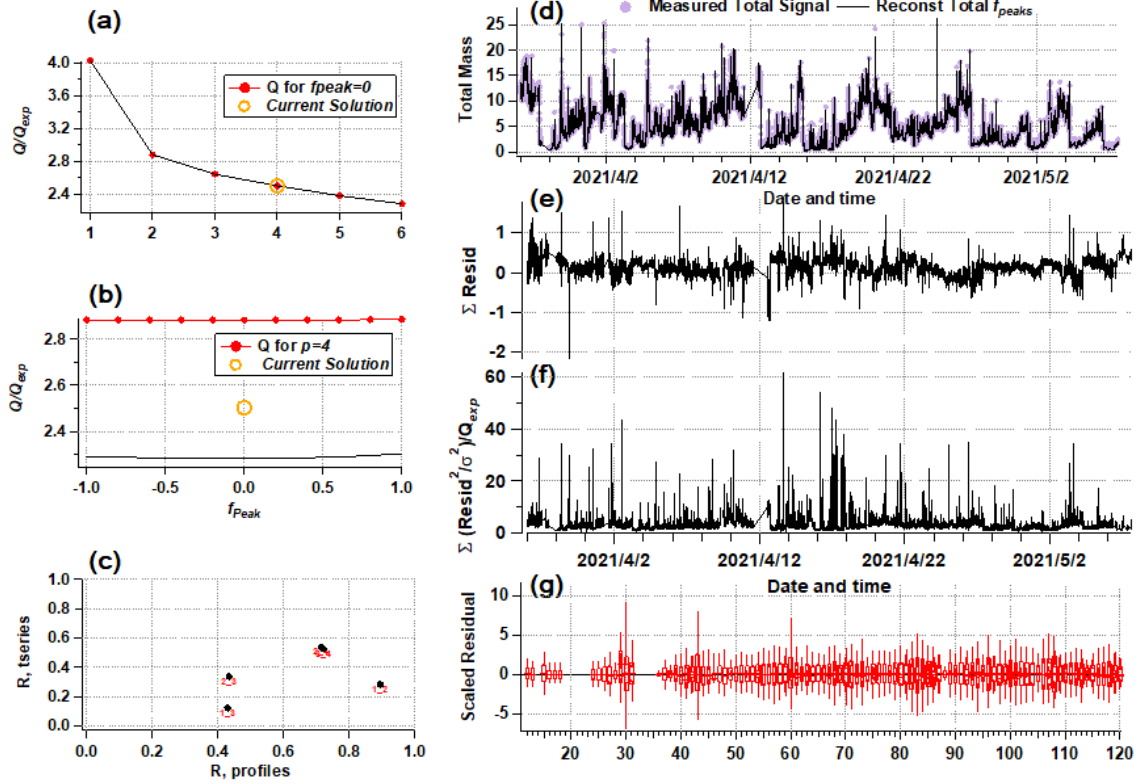
33 **Text S1. PMF Analysis from the HR-ToF-AMS**

34 The HR-ToF-AMS was operated in V mode with high sensitivity. High resolution mass spectra of organic matrix for m/z 12-
35 120 were analyzed with PMF2 algorithm (Paatero and Tapper, 1994), following the data processing and factors selecting
36 steps (Ulbrich et al., 2009; Zhang et al., 2011). The two criteria of the ratio of the scaled residuals (Q/Q_{exp}) and the rotational
37 parameter (f_{peak}) to select the best model number of factors. The f_{peak} parameter represents the rotational sensitivity of the
38 solution sets, and the range of the f_{peak} parameter from -1 to 1 with steps = 0.2 in our study. Factor numbers from 1 to 6 were
39 selected to run in the PMF model.

40 The key diagnostic plots of 4 to 6-factor solutions are shown in Figure S5.1-3. For example, a large decrease in Q/Q_{exp} with
41 the addition of a factor indicates that the additional factor is able to explain a significant fraction of the variation (Figure
42 S5.1a). The presence of time-dependent structure in the residual time series suggests the need for additional factor for better
43 fitting (Figure S5.1e). After a detailed evaluation of time series, mass spectral profiles, diurnal variations, and correlations
44 with external tracers, the 5-factor solution with $f_{peak} = 0$ ($Q/Q_{exp} = 2.38$) was chosen.

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4 factors

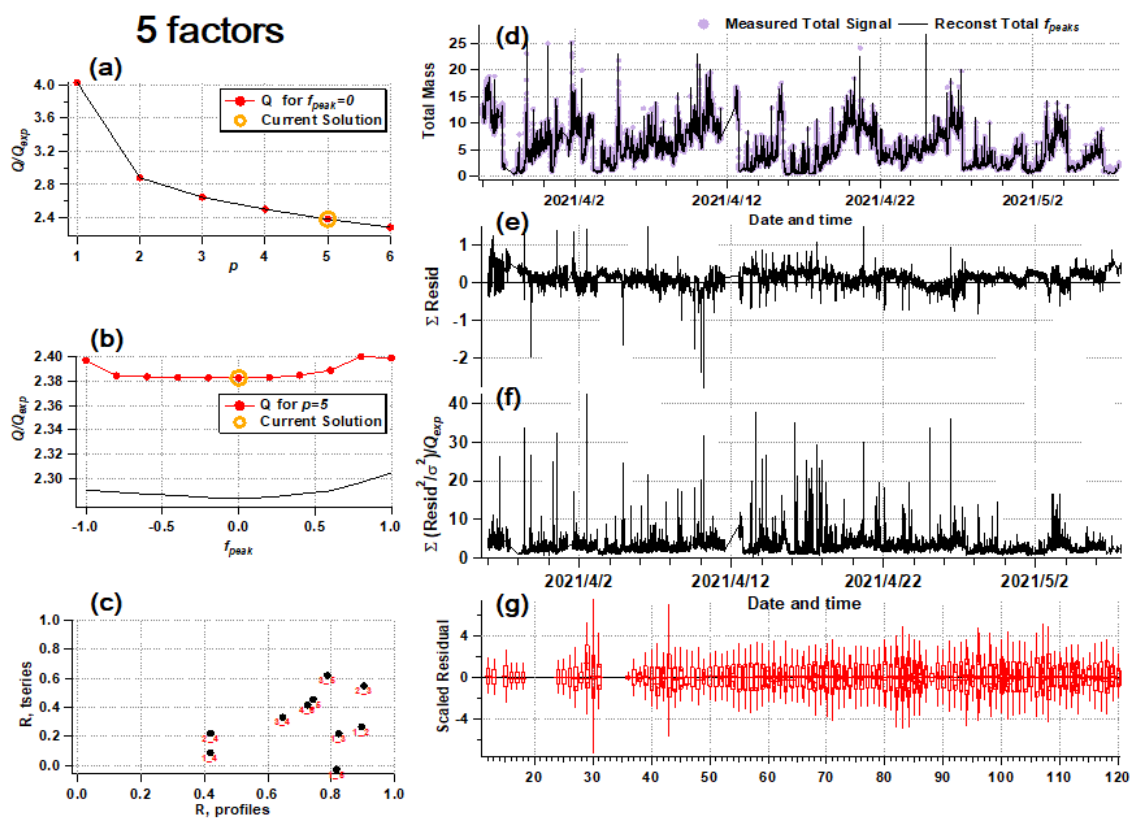


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Figure S5.1 Diagnostic plots of the 4 factors of PMF analysis on OA mass spectral matrix for the spring observation.

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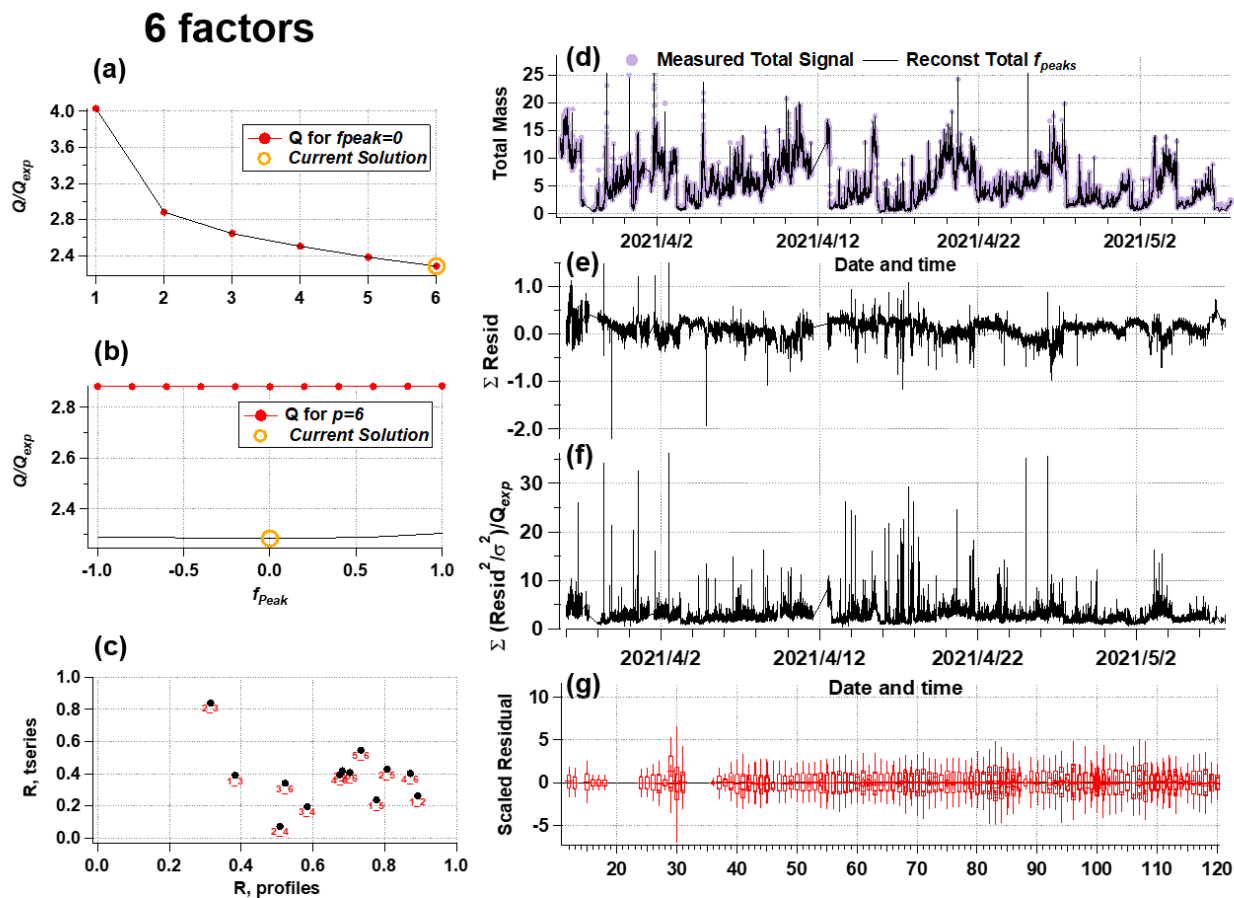


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Figure S5.2 Diagnostic plots of the 5 factors of PMF analysis on OA mass spectral matrix for the spring observation.

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54 **Figure S5.3** Diagnostic plots of the 6 factors of PMF analysis on OA mass spectral matrix for the spring observation.

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56 References

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