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Nitrate-driven urban haze pollution during summertime over the North China Plain

Haiyan Li et al.

Correspondence to: Qiang Zhang (qiangzhang@tsinghua.edu.cn) and Kebin He (hekb@tsinghua.edu.cn)

Text S1. Positive matrix factorization of organic aerosol

Positive matrix factorization (PMF) analysis was performed for 1 to 8 factors with the rotational parameter (FPEAK) varying from -1 to 1 (step = 0.2) for both the Beijing and Xinxiang measurements. Detailed information on how to select the PMF factors can be found in Tables S1-2 and Figs. S2-7. Finally, a four-factor solution with FPEAK = 0 and a three-factor solution with FPEAK = 0 were chosen as the optimal solutions for Beijing and Xinxiang, respectively. The mass spectra, time series, diurnal variations, and correlations with external tracers of the organic aerosol (OA) factors are evaluated in Figs. S2-7.

Table S1. Detailed investigation of the PMF solutions of the Beijing measurements.

Factor number	FPEAK	Q/Q_{exp}	Solution Description
1	0	1.11	Too few factors, large residuals at time periods and key *m/z*'s
2	0	0.73	Too few factors, large residuals at time periods and key *m/z*'s
3	0	0.69	Too few factors (mixed HOA and COA, SV-OOA, and LV-OOA). Factors are mixed to some extent based on the time series and spectra.
4	**0**	**0.67**	**Optimum choices for PMF factors (HOA, COA, SV-OOA, and LV-OOA). Time series and diurnal variations of the PMF factors are consistent with the external tracers. The spectra of the four factors are consistent with the source spectra in the AMS spectra database.**
5-8	0	0.65-0.61	Factor split. For example, when factor num. = 5, HOA was split into two factors with similar spectra but different time series. When factor num. = 6, there is an extra split factor from SV-OOA.

Table S2. Detailed investigation of the PMF solutions of the Xinxiang measurements.

Factor number	FPEAK	Q/Q_{exp}	Solution Description
1	0	1.24	Too few factors, large residuals at time periods and key *m/z*'s
2	0	1.02	Too few factors (mixed HOA and SV-OOA, and LV-OOA). Factors are mixed to some extent based on the time series and spectra.
3	0	0.95	**Optimum choices for PMF factors (HOA, SV-OOA, and LV-OOA). Time series and diurnal variations of the PMF factors are consistent with the external tracers. The spectra of the three factors are consistent with the source spectra in the AMS spectra database.**
4-8	0	0.84-0.92	Factor split. For example, when factor num. = 5, HOA was split into two factors with similar spectra but different time series. When factor num. = 6, there is an extra split factor from SV-OOA.

33 **Table S3-1.** Summary of the location, sampling period, and references of field measurements during summertime discussed in this study.

Dataset Name	Site type	Sampling period	References
Beijing, 2006	urban	7/9/2006-7/21/2006	Sun et al. (2010)
Beijing, 2008	urban	7/24/2008-9/20/2008	Huang et al. (2010)
Beijing, 2011	urban	6/26/2011-8/28/2011	Sun et al. (2012)
Beijing, 2012	urban	7/29/2012-8/29/2012	Hu et al. (2017)
Beijing, 2015	urban	6/30/2015-7/27/2015	this study
Beijing, China	rural/remote	8/10/2006-9/9/2006	Gunthe et al. (2011)
Xinxiang, China	urban	8/8/2017-8/25/2017	this study
Xinzhou, China	urban downwind	7/17/2014-9/5/2014	Wang et al. (2016)
Xianghe, China	urban downwind	6/1/2013-6/30/2013	Sun et al. (2016)
Nanjing, China	urban	6/1/2013-6/15/2013	Zhang et al. (2015)
Shanghai, China	urban	5/15/2010-6/10/2010	Huang et al. (2012)
Jiaxing, China	urban downwind	6/29/2010-7/15/2010	Huang et al. (2013)
Back Garden, China	rural/remote	7/12/2006-7/30/2006	Xiao et al. (2011)
Hongkong, China	urban downwind	9/1/2011-9/29/2011	Li et al. (2015)
Tokyo, Japan, 2003	urban	7/24/2003-8/13/2003	Takegawa, et al. (2006)
Tokyo, Japan, 2004	urban	7/26/2004-8/15/2004	Miyakawa et al. (2008)
Tokyo, Japan, 2008	urban	7/28/2008-8/29/2008	Xing et al. (2011)
Wakayama, Japan	rural/remote	8/20/2010-8/30/2010	Han et al. (2014)
Gwangju, Korea	urban	7/23/2012–8/6/2012	Park et al. (2013)
BNL, NY	rural/remote	7/15/2011-8/15/2011	Zhou et al. (2016)

Queens, NY, 2001	urban	6/30/2001-8/5/2001	Drewnick et al., (2010)
Queens, NY, 2009	urban	7/13/2009-8/3/2009	Sun et al. (2011)
Pinnacle State Park, NY	rural/remote	7/18/2004-8/6/2004	Bae et al. (2007)
Look Rock, TN	rural/remote	6/1/2013–9/21/2013	Budisulistiorini et al. (2016)
Centreville, AL	rural/remote	6/1/2013-7/15/2013	Xu et al. (2015)
Yorkville, GA	rural/remote	6/26/2012-7/20/2012	Xu et al. (2015)
Atlanta, GA	urban	6/20/2012-9/21/2012	Budisulistiorini et al. (2016)
Rocky Park, CO	rural/remote	7/2/2010-8/31/2010	Schurman et al. (2015)
Sacramento, CA	urban downwind	6/2/2010-6/28/2010	Setyan et al. (2012)
Riverside, CA	urban	7/15/2005-8/15/2005	Docherty et al. (2011)
London, UK	urban downwind	7/30/2003-8/6/2003	Cubison et al. (2006)
Paris, France	urban downwind	7/1/2009-7/31/2009	Crippa et al. (2013)
Zurich, Switzerland	urban downwind	7/14/2005-8/4/2005	Lanz et al. (2010)
Melpitz, Germany	rural/remote	5/23/2008-6/9/2008	Poulain et al. (2011)
Prague, Czech Republic	urban downwind	6/20/2012-7/31/2012	Kubelova et al. (2015)
Mt. Cimone, Italy	rural/remote	6/11/2012-7/11/2012	Rinaldi et al. (2015)
Crete, Greece	rural/remote	8/16/2012-9/31/2012	Bougiatioti et al. (2014)
Patras, Greece	urban downwind	6/8/2012-6/27/2012	Kostenidou et al. (2015)
Athens, Greece	urban downwind	7/12/2012-7/26/2012	Kostenidou et al. (2015)
Hyytiälä, Finland	rural/remote	7/12/2012-8/12/2010	Corrigan et al. (2013)

Table S3-2. Summary of the average mass concentrations of aerosol species in submicron particles during the summer observations discussed in this study (unit: μg m^{-3}).

Dataset Name	PM type	PM Conc.	Org Conc.	SO_4 Conc.	NO_3 Conc.	NH_4 Conc.	Chl Conc.	BC Conc.
Beijing, 2006	NR-PM_1	80.0	28.0	20.0	17.6	12.8	1.1	
Beijing, 2008	PM_1	63.1	23.9	16.8	10.0	10.0	0.5	1.9
Beijing, 2011	NR-PM_1	50.0	20.0	9.0	12.5	8.0	0.5	
Beijing, 2012	PM_1	37.5	12.5	9.7	6.3	5.4	0.3	3.2
Beijing, 2015	PM_1	35.0	12.2	6.3	8.4	4.3	0.4	3.1
Beijing, China	NR-PM_1	26.2	9.8	7.8	2.7	5.2	0.5	
Xinxiang, China	PM_1	64.2	18.0	14.4	16.5	12.2	0.6	2.3
Xinzhou, China	PM_1	35.4	11.7	11.5	5.1	4.2	0.5	2.4
Xianghe, China	PM_1	73.0	28.5	13.1	14.6	8.8	2.9	5.1
Nanjing, China	PM_1	38.5	15.0	4.6	8.9	6.2	0.4	3.1
Shanghai, China	PM_1	29.2	8.4	9.7	4.8	3.9	0.5	2.0
Jiaxing, China	PM_1	32.9	10.6	5.9	8.3	4.1	1.0	3.0
Back Garden, China	NR-PM_1	30.0	13.2	11.4	1.5	3.6	0.3	
Hongkong, China	NR-PM_1	15.6	4.1	8.7	0.4	2.4	0.01	
Tokyo, Japan, 2003	NR-PM_1	12.7	5.7	3.2	1.0	1.8	0.09	
Tokyo, Japan, 2004	PM_1	33.0	14.5	9.1	1.5	4.0	0.3	3.6
Tokyo, Japan, 2008	NR-PM_1	10.6	5.6	3.4	0.2	1.4	0.05	
Wakayama, Japan	NR-PM_1	4.0	1.8	1.6	0.04	0.5	0.01	
Gwangju, Korea	NR-PM_1	8.7	4.9	2.4	0.4	0.9	0.06	

BNL, NY	PM_1	12.6	8.1	3.0	0.2	1.0	0.01	0.1
Queens, NY, 2001	NR-PM_1	10.6	2.7	3.9	0.8	1.4	0.03	
Queens, NY, 2009	PM_1	11.7	6.3	2.8	0.5	1.3	0.03	0.7
Pinnacle State Park, NY	NR-PM_1	12.3	5.7	4.9	0.4	1.3	0.01	
Look Rock, TN	NR-PM_1	8.4	5.3	2.1	0.3	0.7	0.01	
Centreville, AL	NR-PM_1	7.4	5.0	1.9	0.1	0.4	0.01	
Yorkville, GA	NR-PM_1	16.1	11.2	3.5	0.3	1.1	0.03	
Atlanta, GA	NR-PM_1	8.8	6.1	1.5	0.4	0.7	0.01	
Rocky Park, CO	NR-PM_1	5.1	3.8	0.8	0.2	0.2	0.05	
Sacramento, CA	PM_1	3.0	2.4	0.3	0.1	0.1	0.003	0.05
Riverside, CA	PM_1	20.5	9.1	3.5	4.4	2.5	0.09	0.9
London, UK	NR-PM_1	5.4	2.5	1.7	0.5	0.7	0.04	
Paris, France	PM_1	4.5	2.2	1.3	0.2	0.3	0.02	0.6
Zurich, Switzerland	NR-PM_1	9.6	6.5	1.4	0.8	0.1		
Melpitz, Germany	PM_1	11.6	6.8	2.5	0.6	0.9	0.02	0.6
Prague, Czech Republic	NR-PM_1	8.3	4.2	2.0	0.8	1.2	0.1	
Mt. Cimone, Italy	NR-PM_1	4.5	2.8	0.9	0.3	0.4	0.05	
Crete, Greece	PM_1	9.2	3.2	4.0	0.2	1.3		0.6
Patras, Greece	PM_1	8.6	3.8	3.3	0.1	0.9		0.5
Athens, Greece	PM_1	14.2	6.6	5.3	0.2	1.4		0.7
Hyytiälä, Finland	PM_1	6.7	4.4	1.3	0.2	0.4		0.3

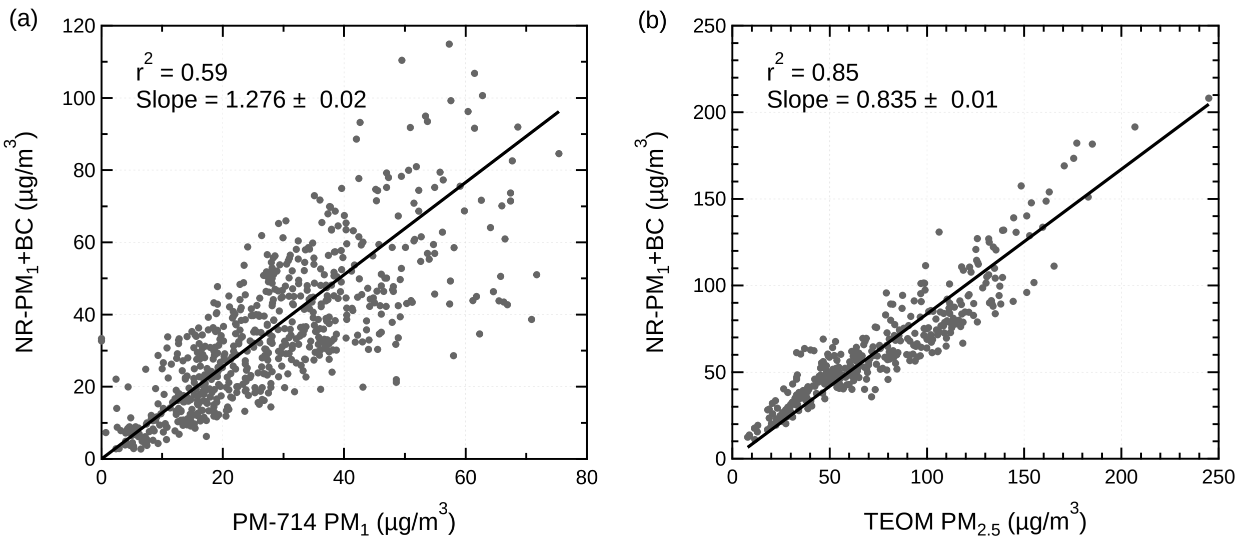


Figure S1. Scatter plots of (a) the mass concentrations of NR-PM_1 plus BC vs. total PM_1 measured by PM-714 in Beijing, and (b) the mass concentrations of NR-PM_1 plus BC vs. total $PM_{2.5}$ measured by TEOM in Xinxiang.

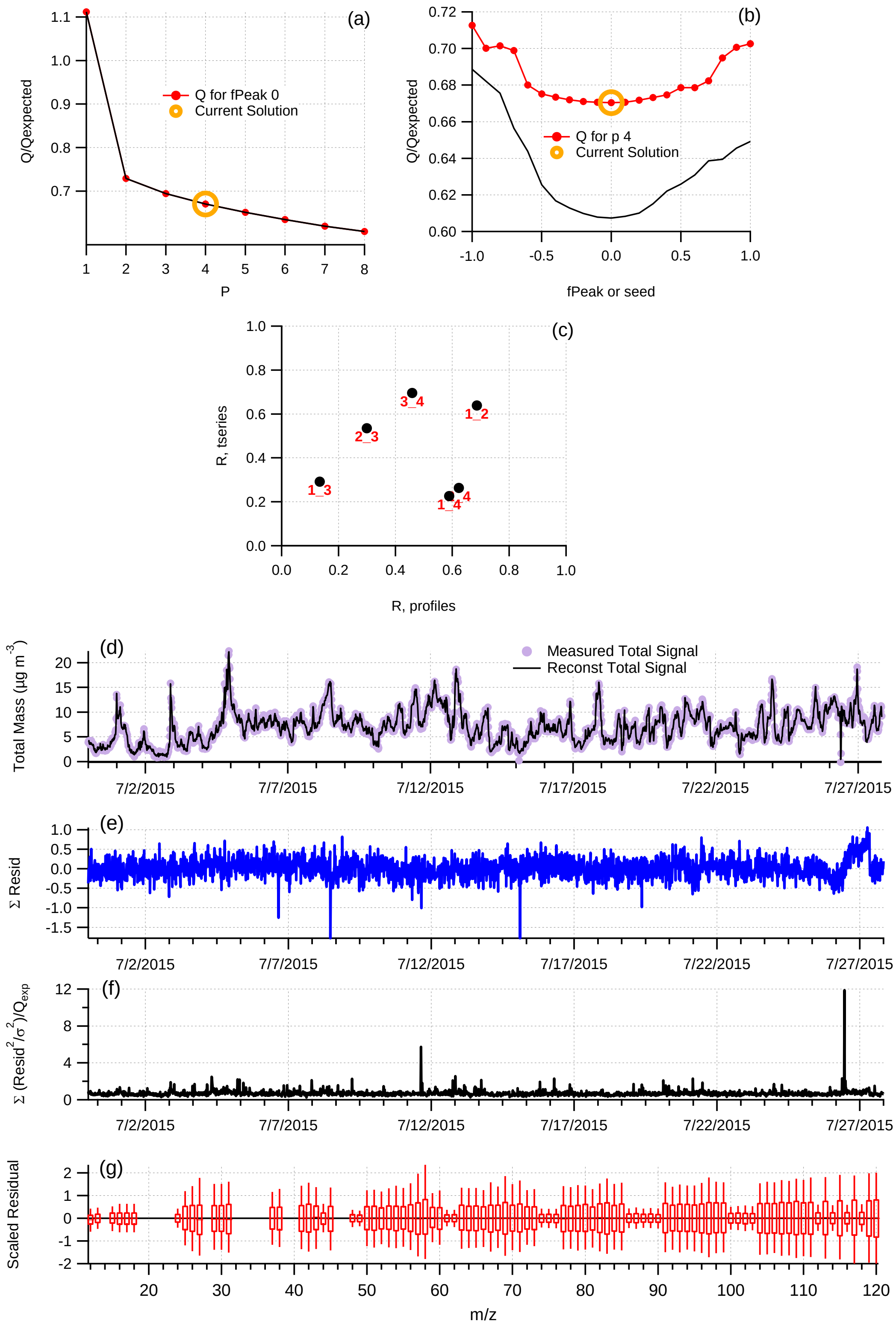


Figure S2. Summary of the key diagnostic plots of the chosen 4-factor solution from PMF analysis of the Beijing measurements.

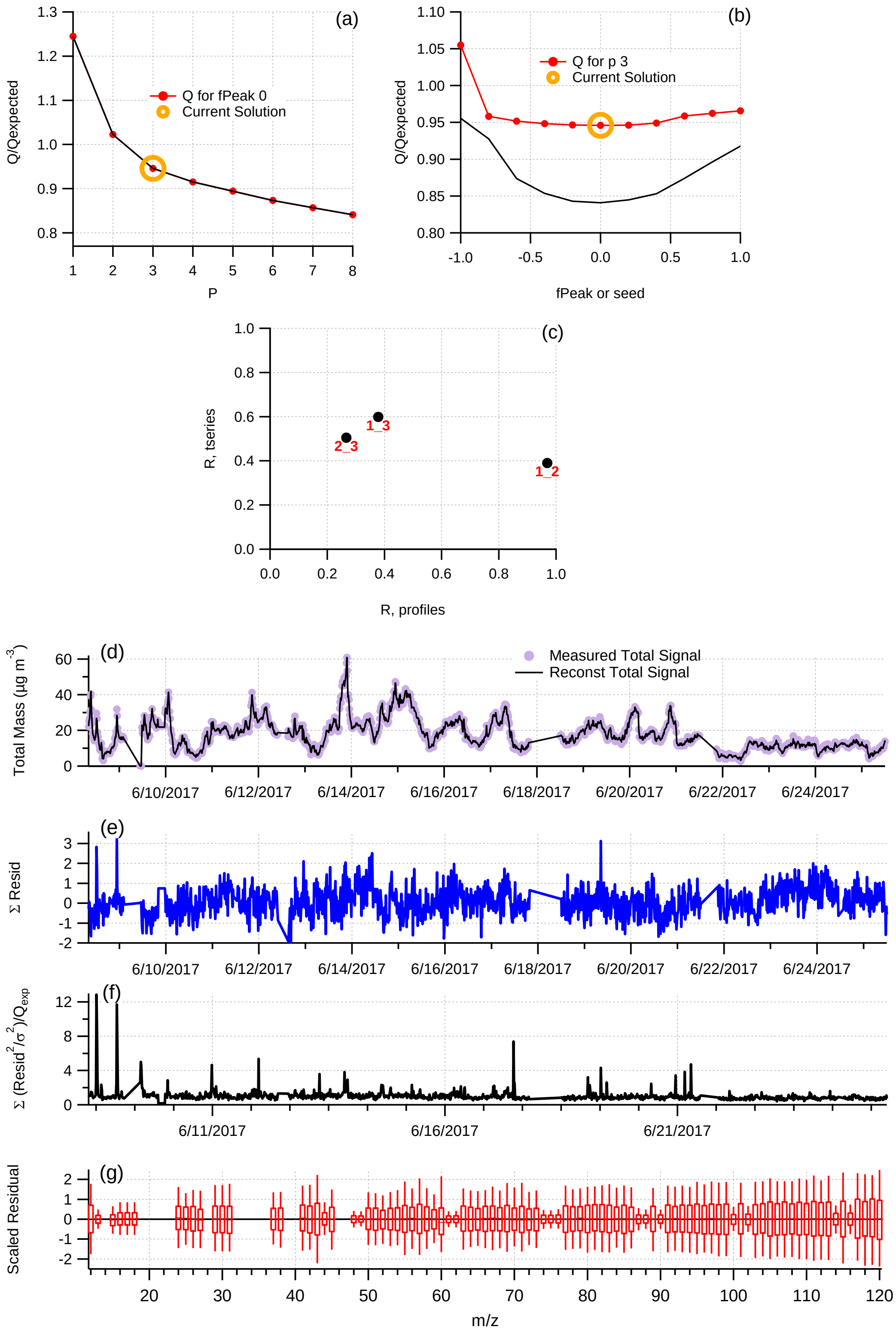


Figure S3. Summary of the key diagnostic plots of the chosen 3-factor solution from PMF analysis of the Xinxiang measurements.

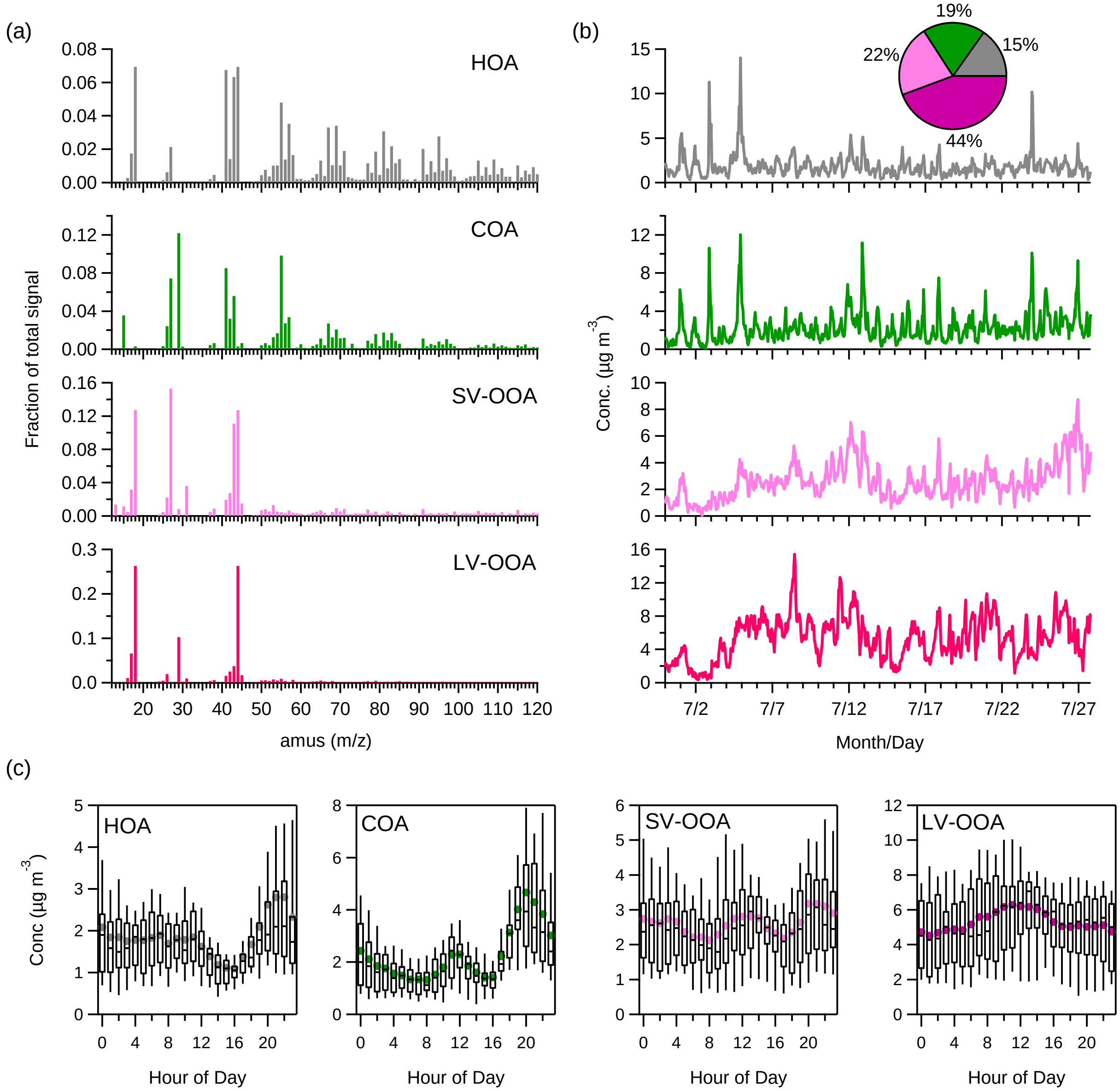
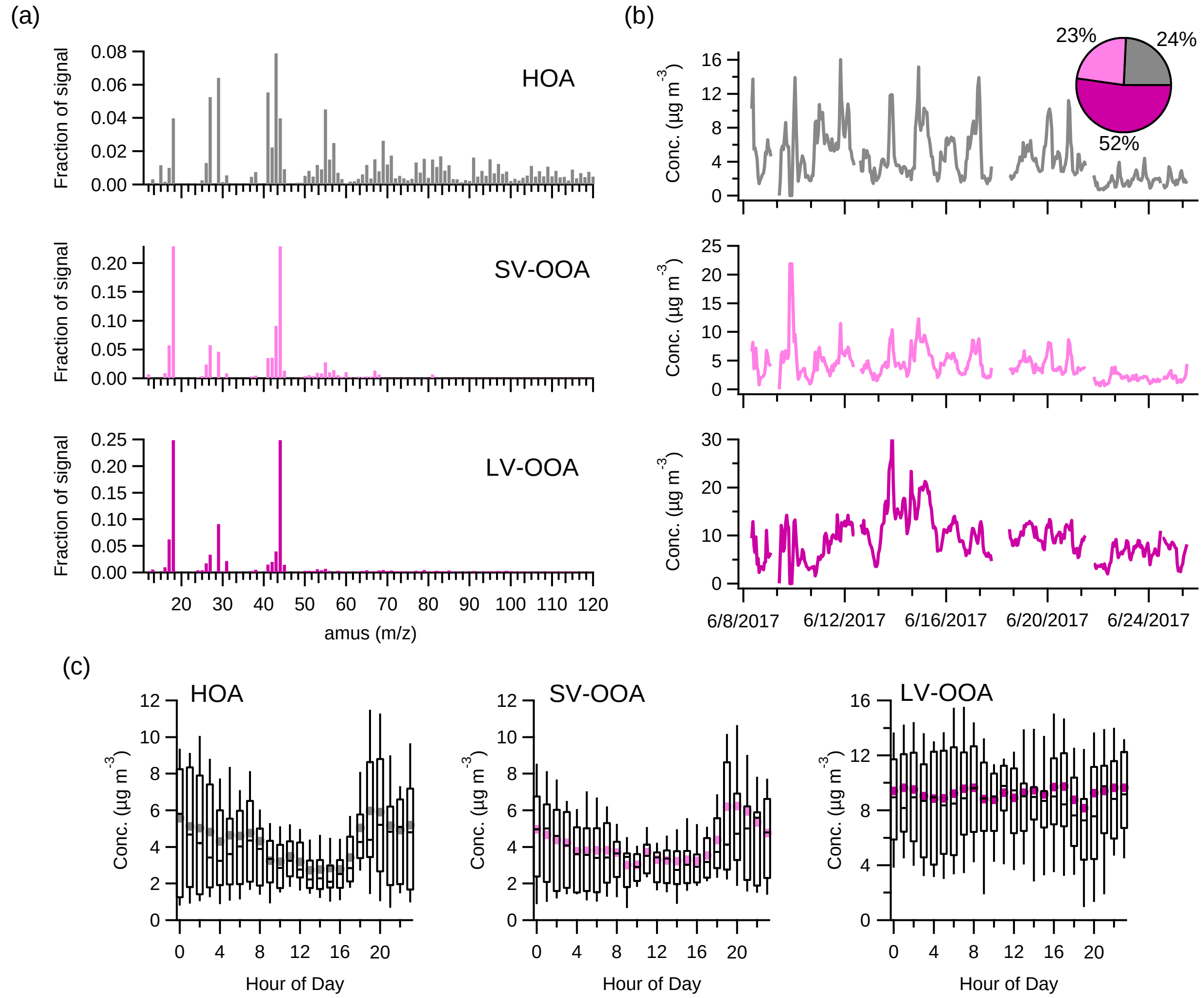


Figure S4. (a) The mass spectra, **(b)** time series, and **(c)** diurnal variations of the four-factor PMF solution of the Beijing measurements.



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56 **Figure S5. (a)** The mass spectra, **(b)** time series, and **(c)** diurnal variations of the four-factor PMF
57 solution of the Xinxiang measurements.

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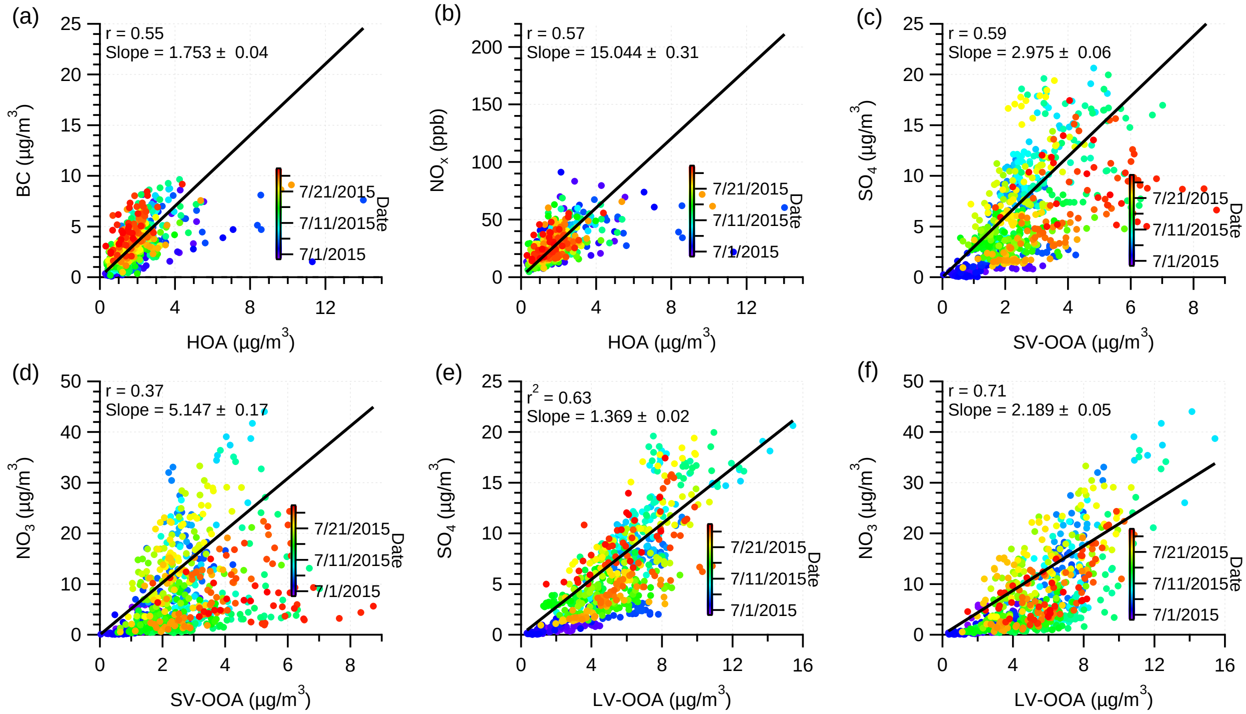


Figure S6. Scatter plots of **(a)** HOA vs. BC, **(b)** HOA vs NO_x, **(c)** SV-OOA vs. sulfate, **(d)** SV-OOA vs nitrate, **(e)** LV-OOA vs. sulfate, and **(f)** LV-OOA vs. nitrate of the Beijing measurements.

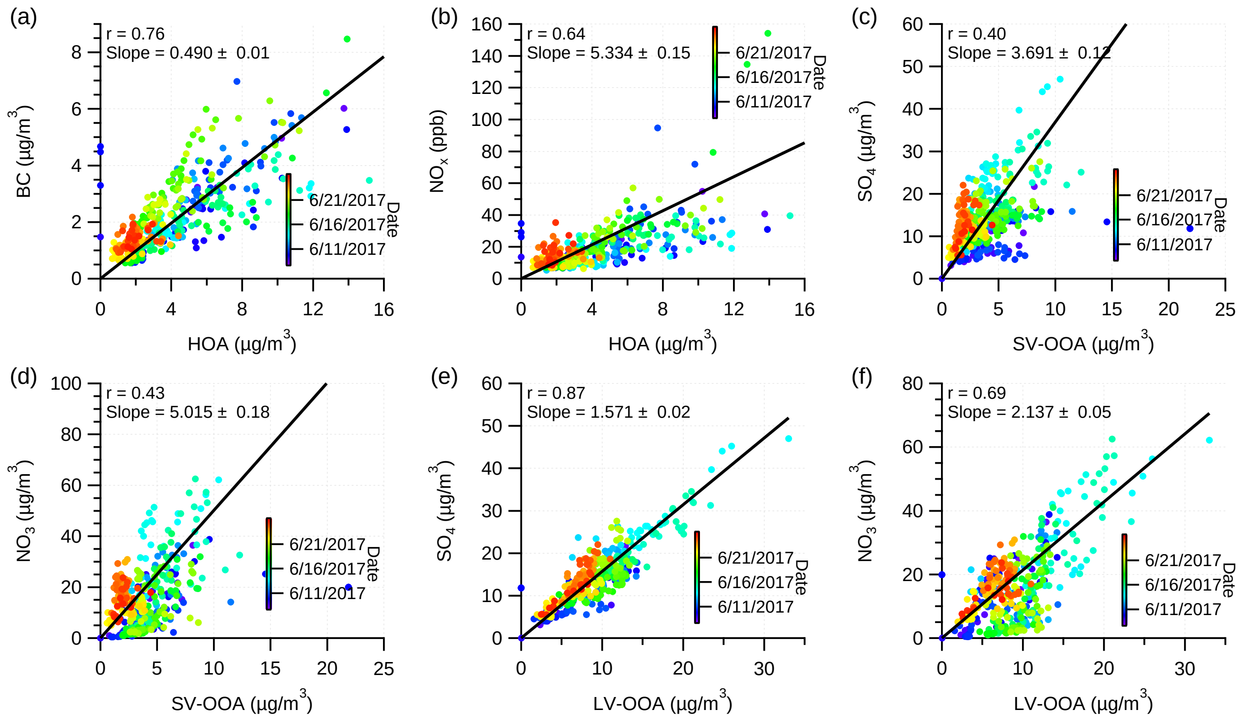


Figure S7. Scatter plots of **(a)** HOA vs. BC, **(b)** HOA vs NO_x, **(c)** SV-OOA vs. sulfate, **(d)** SV-OOA vs nitrate, **(e)** LV-OOA vs. sulfate, and **(f)** LV-OOA vs. nitrate of the Xinxiang measurements.

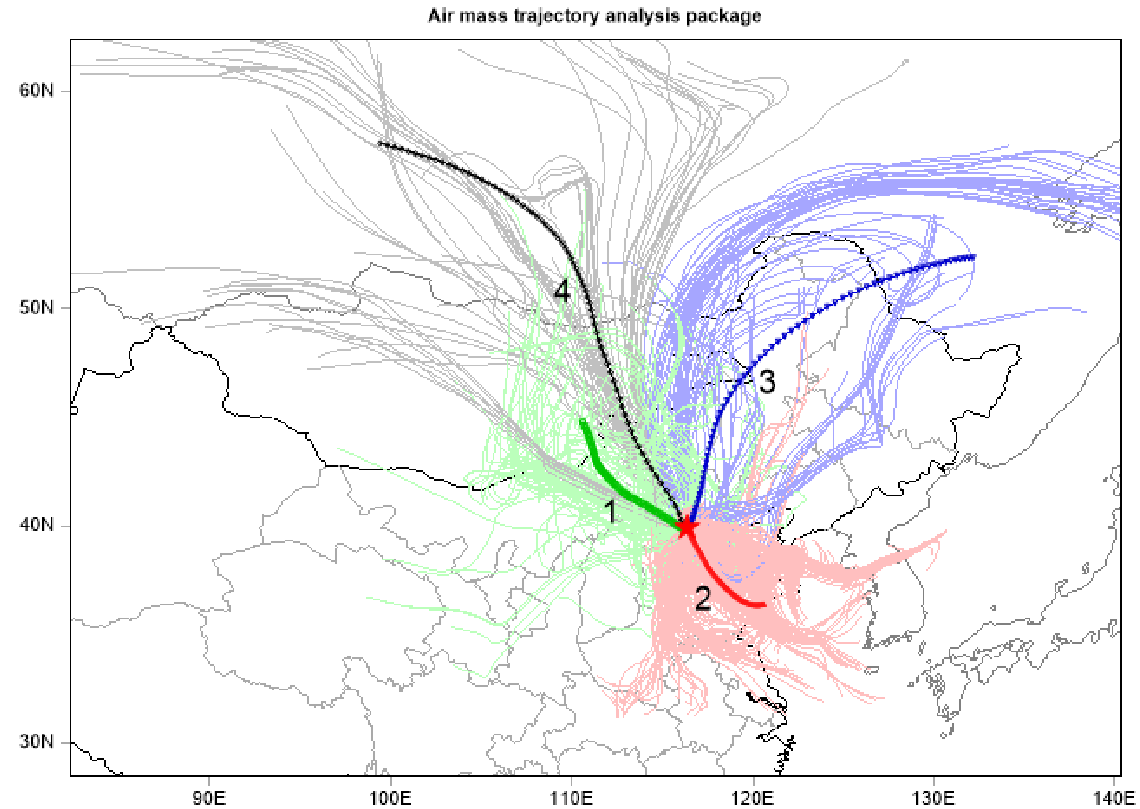


Figure S8. Back trajectories of air masses arriving in Beijing every hour, with the four clusters determined using the inbuilt function in the HYSPLIT model.

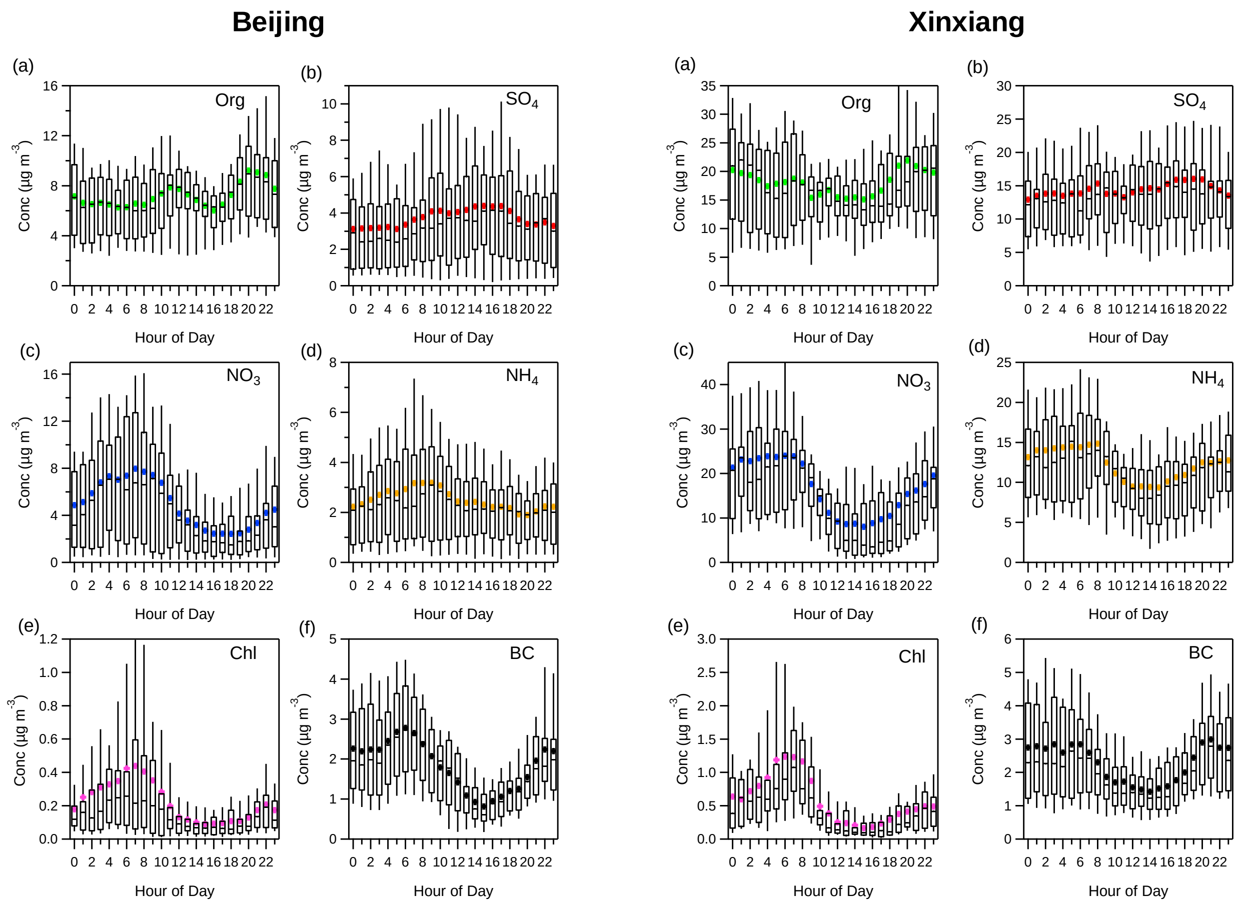


Figure S9. Diurnal variations of **(a)** organics, **(b)** sulfate, **(c)** nitrate, **(d)** ammonium, **(e)** chloride, and **(f)** BC in Beijing and Xinxiang.

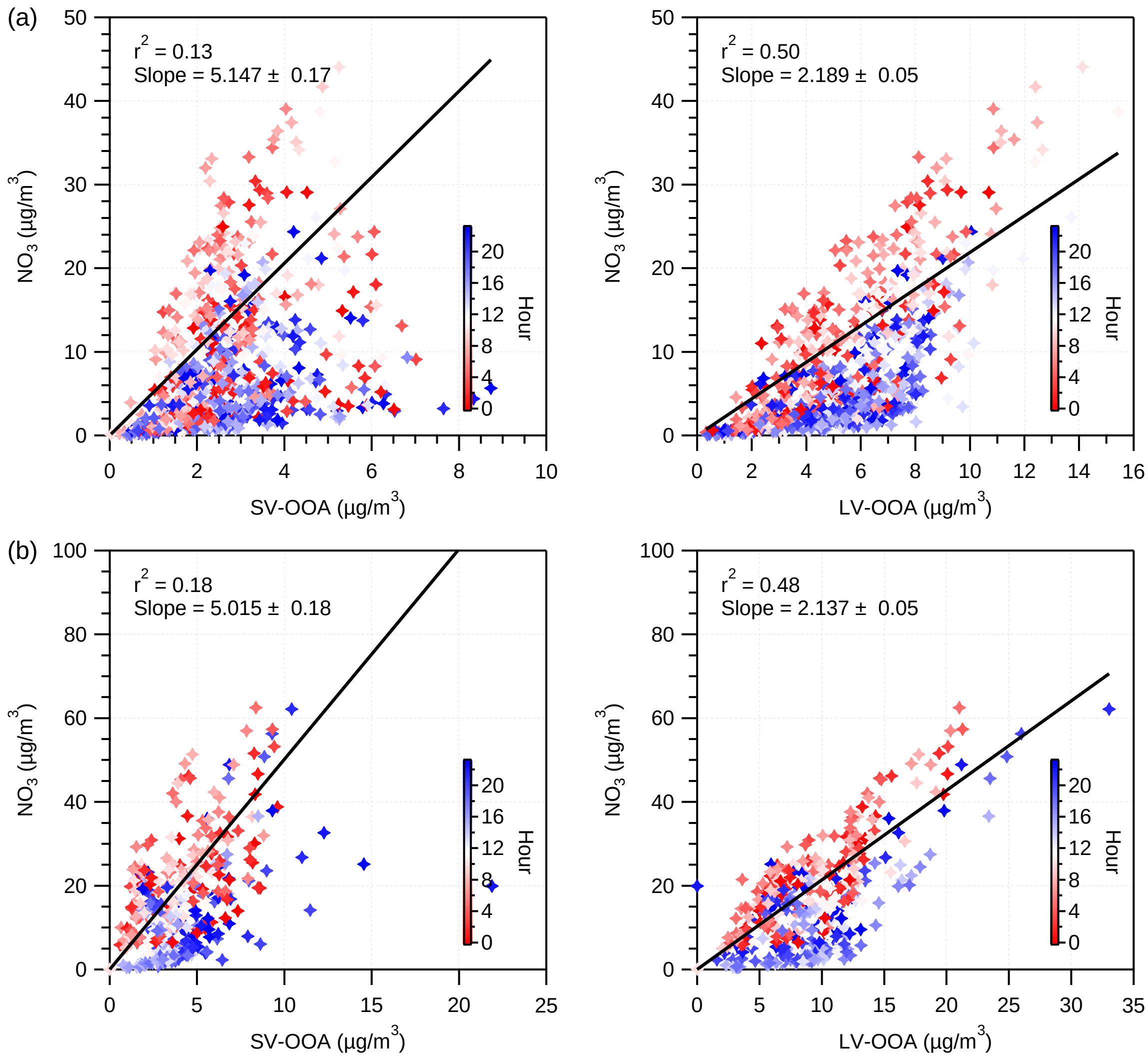
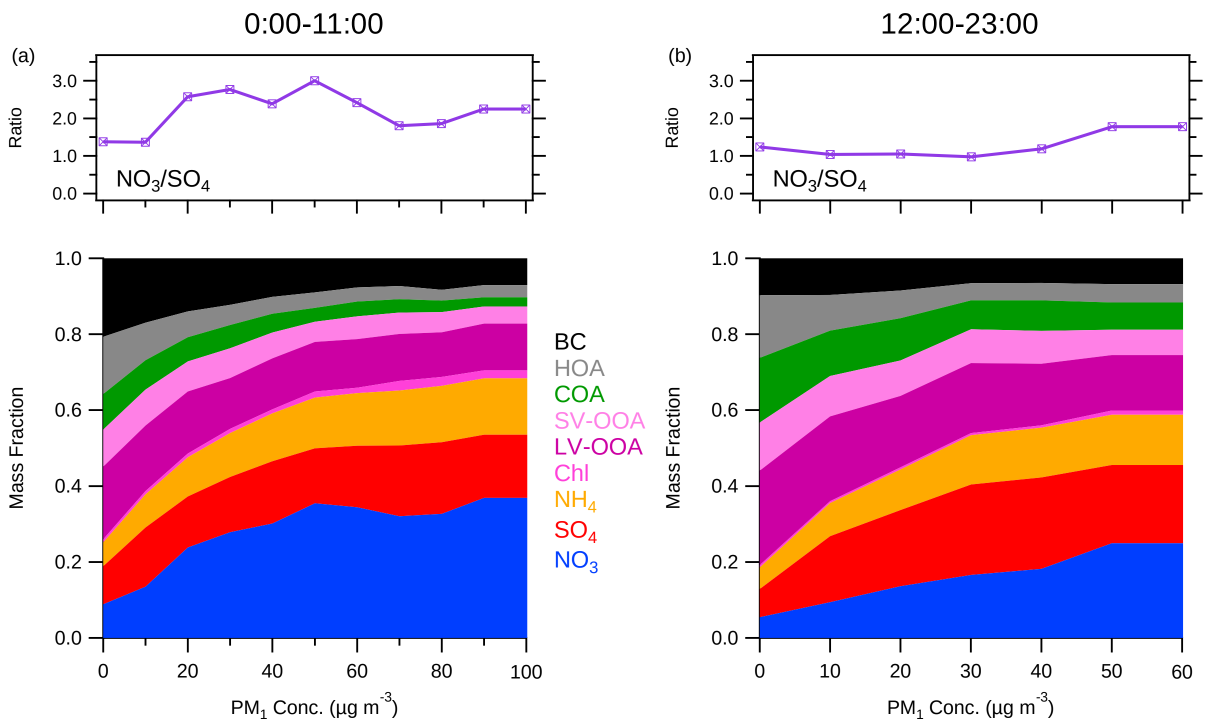


Figure S10. Scatterplots of nitrate vs. SV-OOA and nitrate vs. LV-OOA colored by the hour of the day, in **(a)** Beijing and **(b)** Xinxiang.

80 

81 **Figure S11.** Variations in the mass fractions of aerosol species and nitrate/sulfate mass ratio as a
82 function of total PM_1 mass loadings for the periods **(a)** 0:00 – 11:00 and **(b)** 12:00 -23:00 in Beijing.

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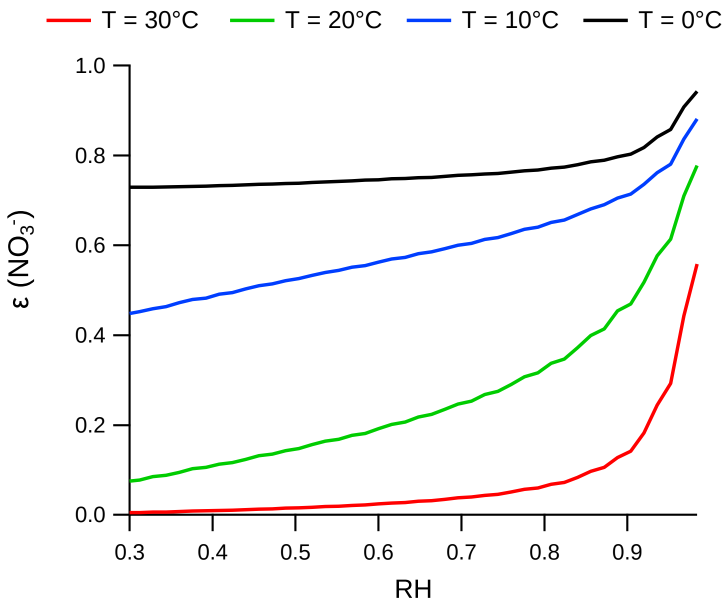

Figure S12. Simulated ε(NO_3^-) by the ISORROPIA-II model at 0 ℃, 10 ℃, 20 ℃, and 30 ℃ with varying RH conditions.

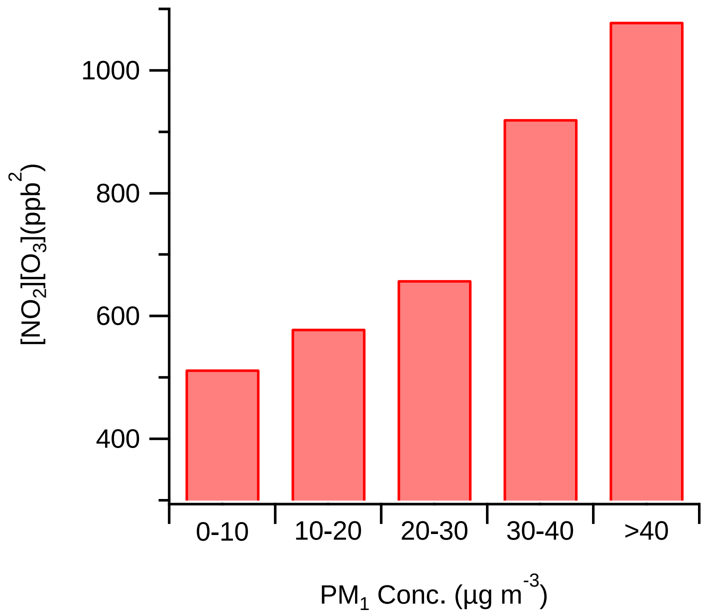

Figure S13. Variations in $[NO_2][O_3]$ at 0:00 as a proxy for the nighttime formation of HNO_3 for different PM_1 concentration bins.

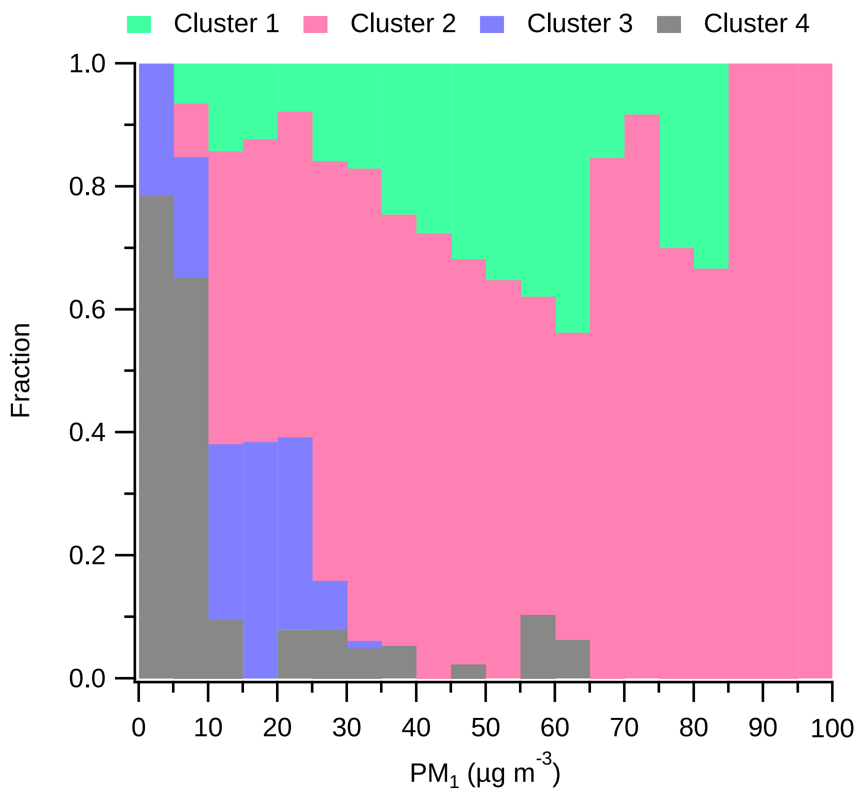

Figure S14. Distribution of the four clusters resolved in this study as a function of PM_1 concentration.

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