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Supplement of

Atmospheric submicron aerosol composition and particulate organic nitrate formation in a boreal forestland–urban mixed region

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S1. Experimental

S1.1 Trajectory analysis

To investigate the aerosol regional source on the measurement site, 48 h back trajectories at 500m arrival height above ground level were computed every 2h using Hybrid Single Particle Lagragian Integrated Trajectory model (HYSPLIT-4) (Draxler, R.R. and Rolph, G.D., 2013).

S2. Results and discussion

S2.1 PMF factor solution

Fig. S1 shows Positive Matrix Factorization (PMF) key diagnostics in this study. Five-factor solution Fpeak +0.1 were selected. Fig S2 shows the time series and profiles of PMF five-factor solution at Fpeak=+0.1. Factors 3 and 4 produced meaningful time series and mass profiles but we could not find other gas or particle phase observations that correlate with them during the measurement. Thus these two factors were merged to generate a new factor by a mass-weighted combination. After merging, the four factors are NIA (nitrate inorganic aerosol), LVOOA (low-volatile oxygenated OA), SVOOA (semi-volatile oxygenated OA), HOA (hydrocarbon-like organic aerosol). In four-factor solution the factor 4 from 5-factor solution was divided between factor 5 and factor 2 and made HOA factor meaningless. In six-factor solution the LVOOA from five-factor solution was split into LVOOA1 (Factor 3) and LVOOA2 (Factor 4) without reasonable reason and doesn't give any better interpretation on the data (Fig. S4). Thus a five-factor solution was selected.

The rotational ambiguity of five-factor solution was explored by varying Fpeak between -1.0 and +1.0. The choice of Fpeak <-0.1 gives periods of zeros in factor 4 that don't correspond to any events in observations. Increasing Fpeak to +0.1 improves the corrections of SVOOA and NIA to nitrate compared to Fpeak value of 0 whilst it does not affect the correlations of other factors to the external tracers. Thus Fpeak = 0.1 was selected (Fig. S5).

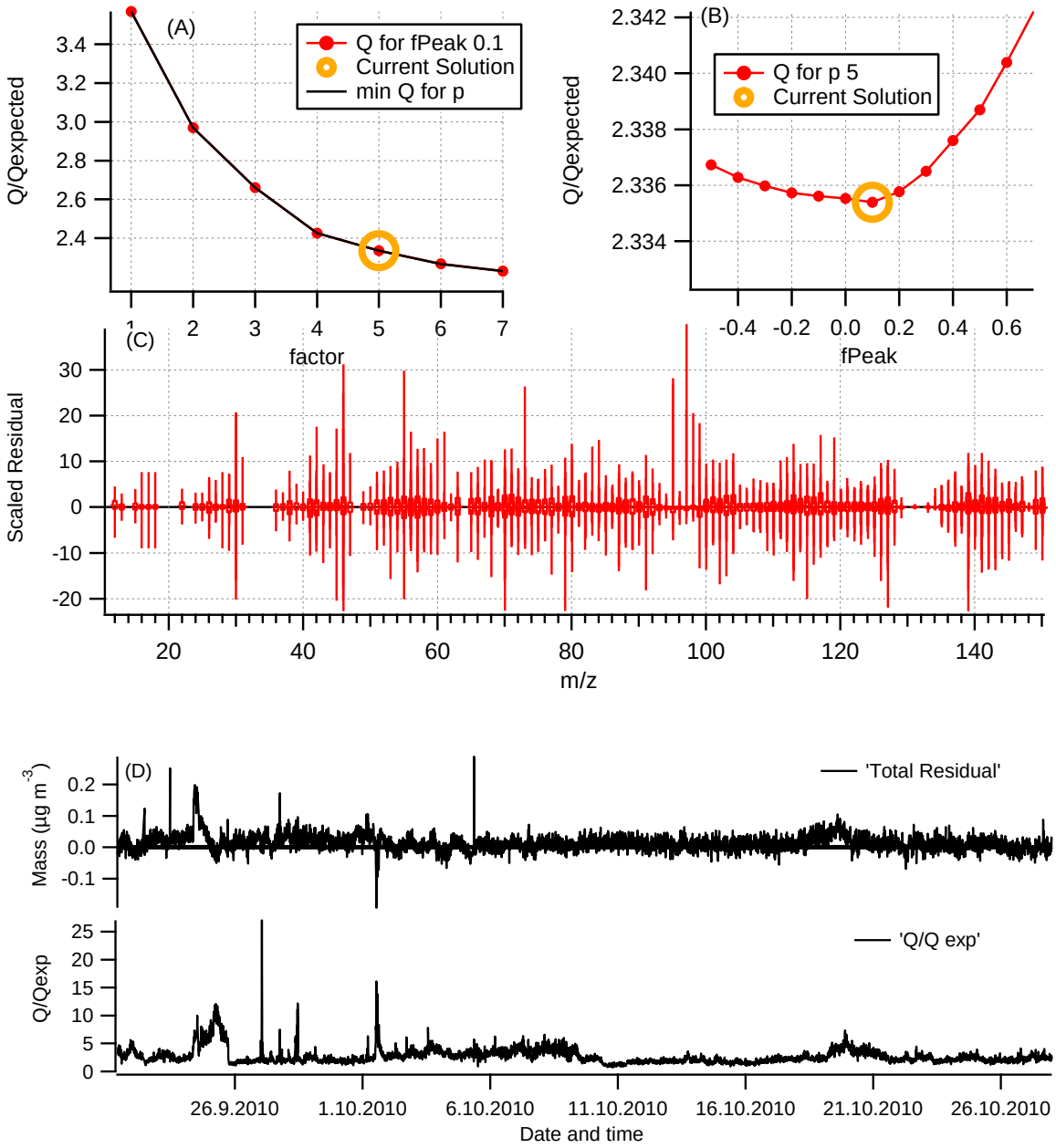


Figure S1 PMF key diagnostics plots: (A) $Q/Q_{expected}$ varies as function of PMF factor at fPeak 0.1; (B) $Q/Q_{expected}$ varies as function of rotational ambiguity; (C) Scaled residual for each mass; (D) time series of the total residual and Q/Q expected contribution for every point during this study. For more details on PMF and the interpretation of these plots see Ulbrich et al. (2009).

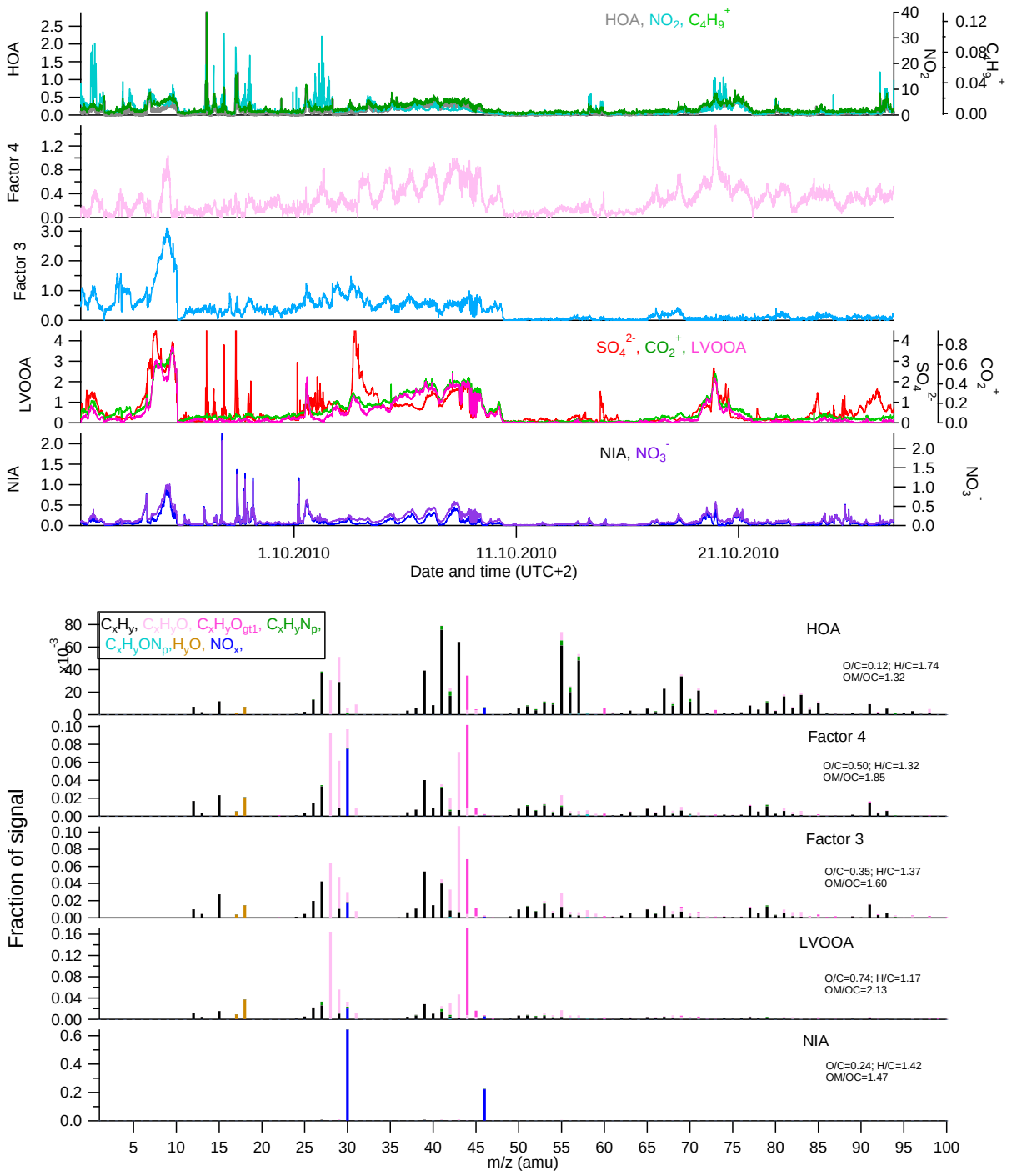


Figure S2. Time series and mass profiles by PMF analysis at five-factor solution. Factor 3 and 4 were merged to generate a new factor by a mass-weighted combination, which results are reported in the paper. HOA, hydrocarbon-like organic aerosol; LVOOA, low-volatile oxygenated OA; NIA, nitrate inorganic aerosol

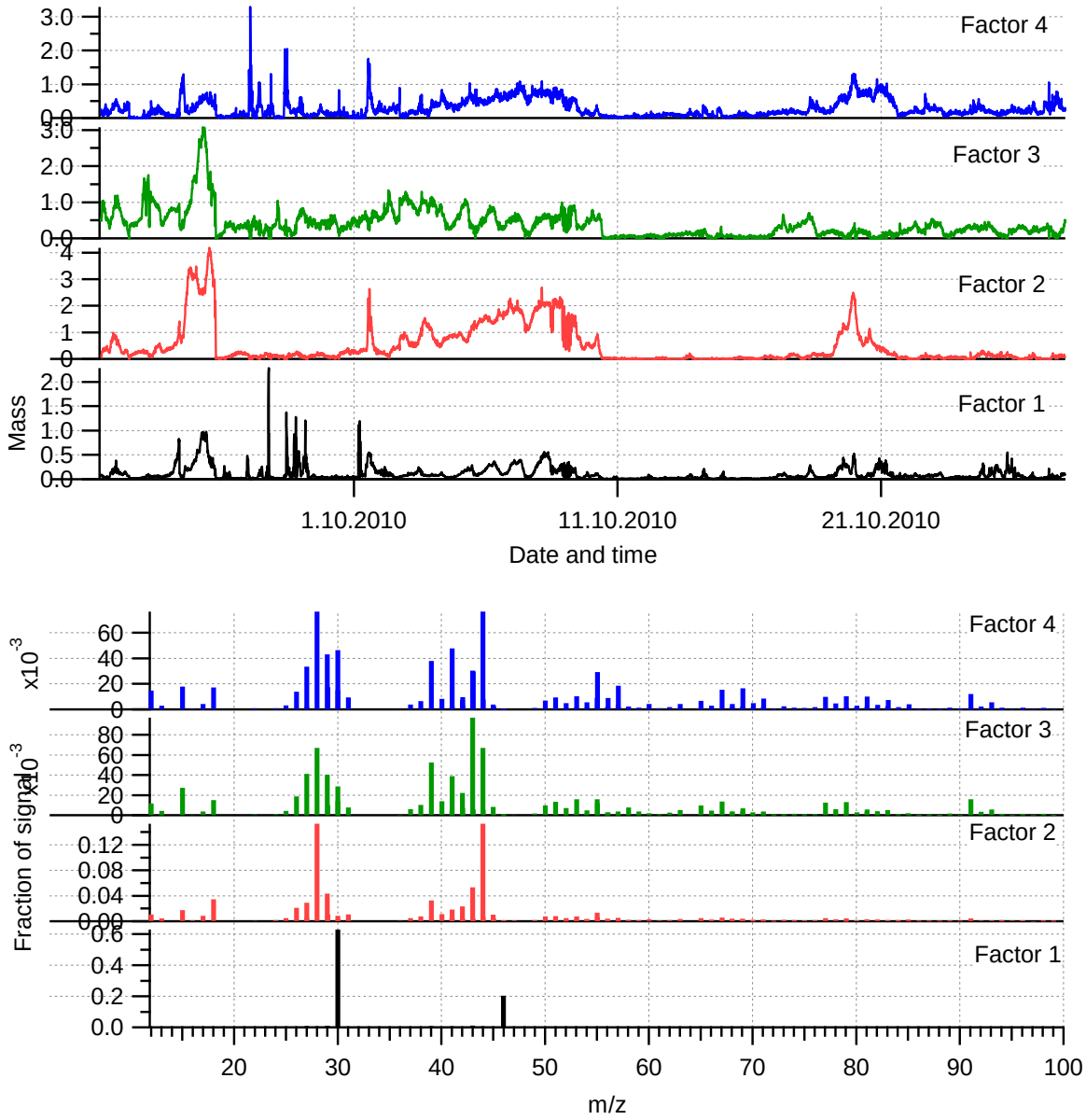


Figure S3. Time series and mass profiles by PMF analysis at four-factor solution with fPeak =+0.1.

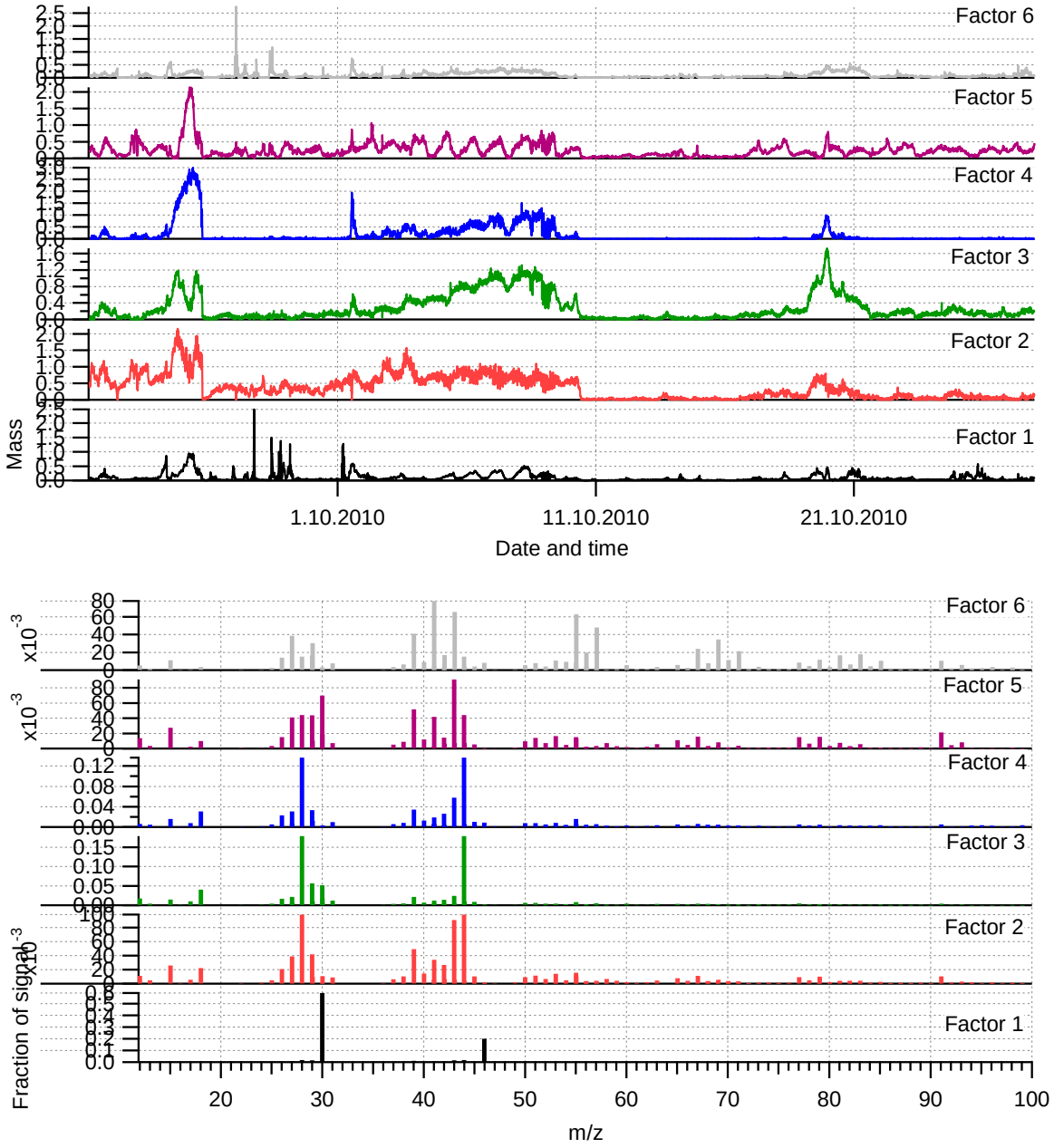


Figure S4. Time series and mass profiles by PMF analysis at six-factor solution with fPeak =+0.1.

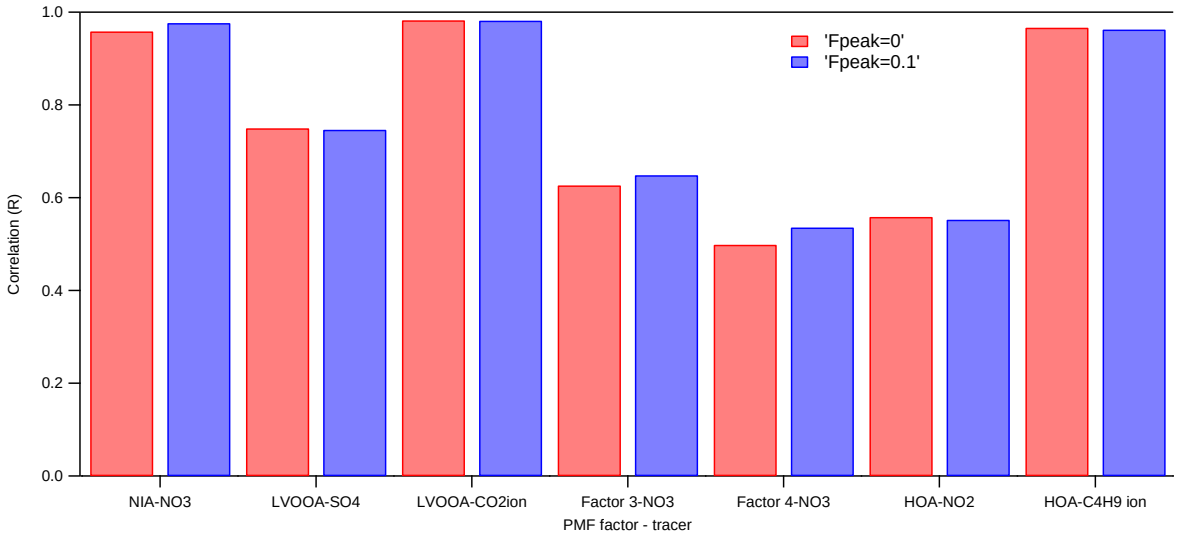


Figure S5. Correction coefficients between the PMF factors and tracers by varying Fpeak from 0 to +0.1.

S2.2 Other supplementary plots

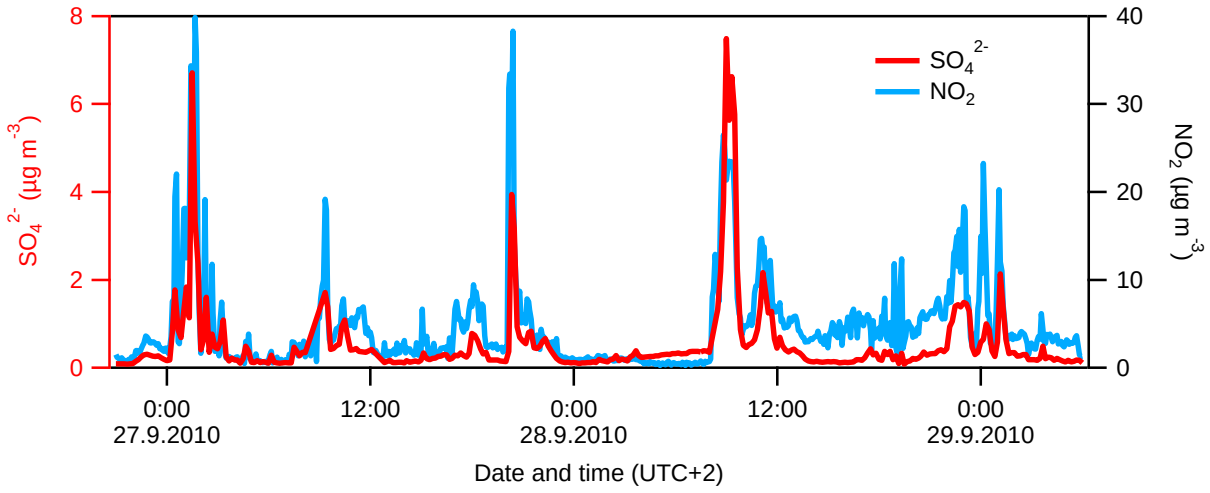


Figure S7. Comparisons of time series of SO_4^{2-} and NO_2 during the primary aerosol emitting days, which is shown in the gray bar in Figure 1. Good correlation of SO_4^{2-} with NO_2 suggests that the SO_4^{2-} is primary in nature during this period.

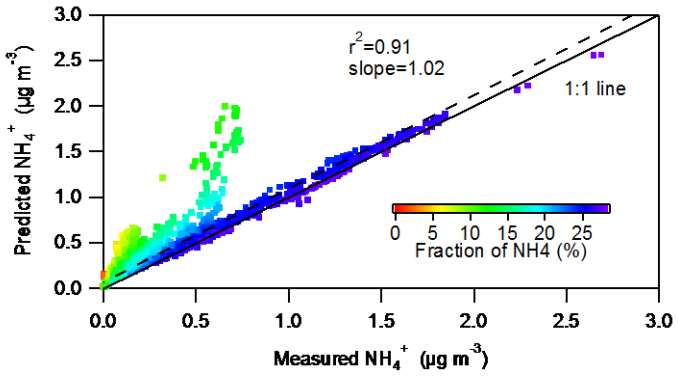


Figure S8. Predicted NH_4^+ (assuming fully neutralized aerosol) vs measured NH_4^+, colored by the mass fraction of measured NH_4^+ to the sum of SO_4^{2-}+ NO_3^-+Cl^-. The predicted NH_4^+ was determined by $NH_{4,pre}^+$ = 18×(2× SO_4^{2-}/96+ NO_3^- /62+ Cl^-/35.5), where NH_4^+, SO_4^{2-}, NO_3^- and Cl^- represent the mass concentrations (in μg m^{-3}) of the species and the denominators correspond to their molecular weights. The factor 18 is the molecular weight of NH_4^+ (Zhang et al., 2007).

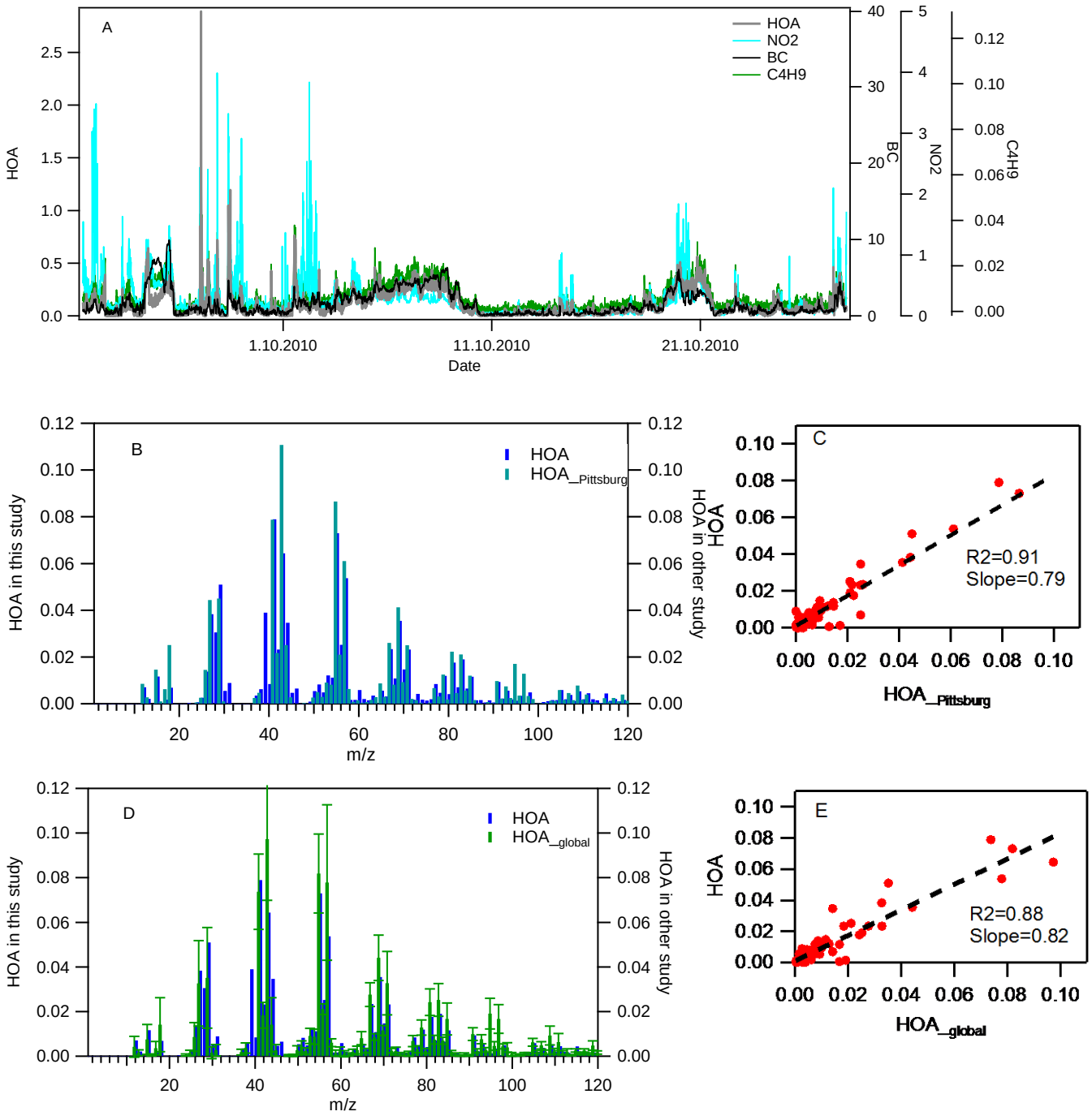
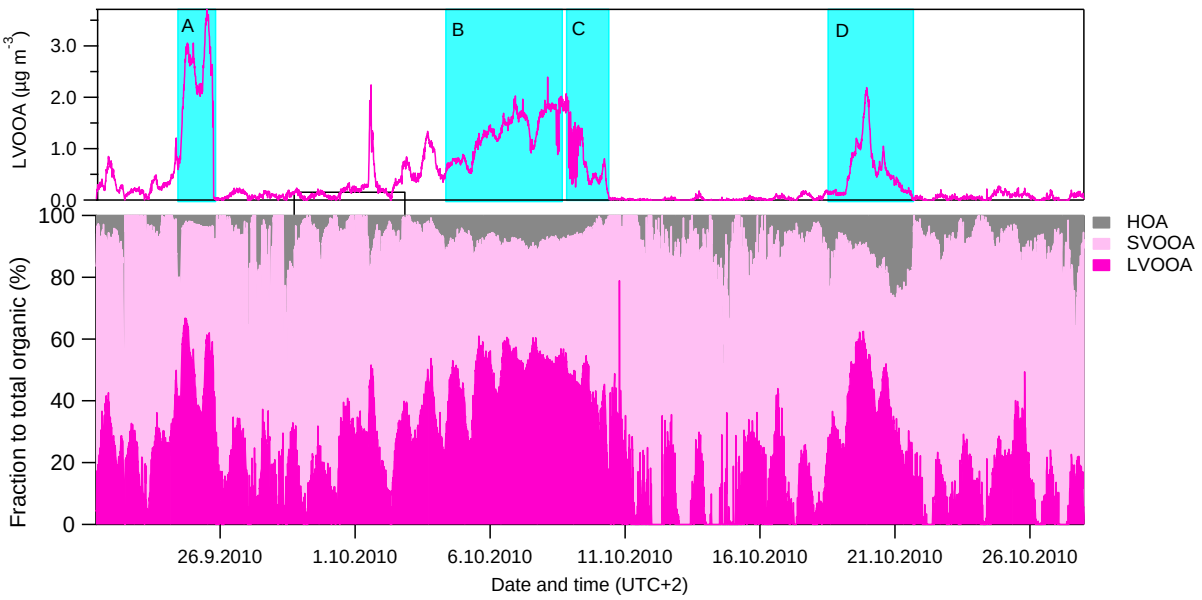
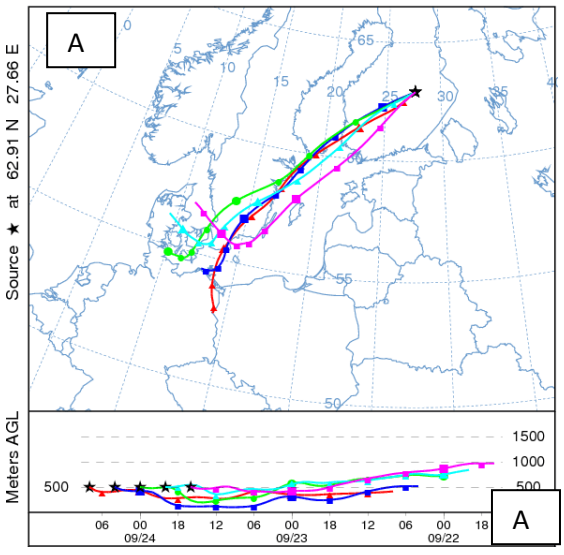
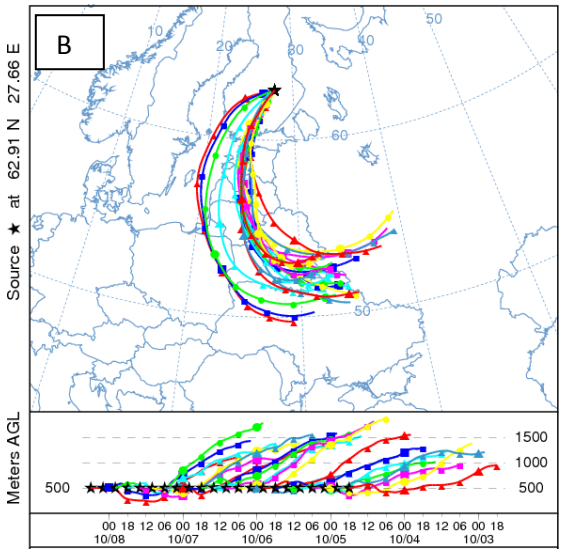
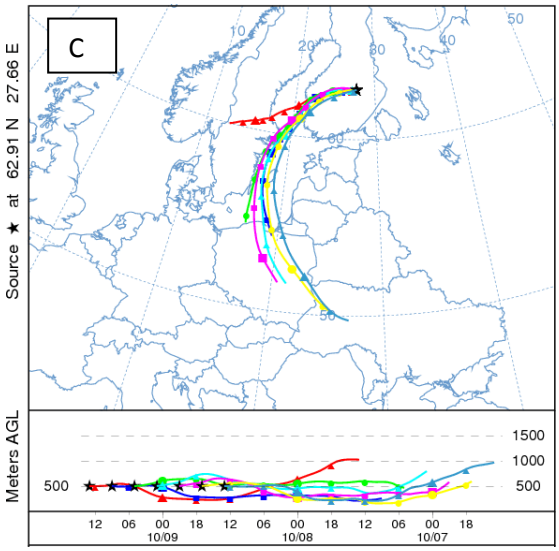


Figure S9. (A) Correlations of HOA time series with tracers; (B) Comparison of HOA mass spectrum from HOA determined in Pittsburg (Ulbrich et al., 2009); (C) a scatter plot of HOA mass spectrum between this study and Pittsburg; (D) Comparison of HOA mass spectrum from HOA determined on a global scale (Ng et al., 2010); (E) a scatter plot of HOA mass spectrum between this study and on a global scale;


A
B
C
D
LVOOA (µg m-3)
3.0
2.0
1.0
0.0
100
80
60
40
20
0
Fraction to total organic (%)
HOA
SVOOA
LVOOA
26.9.2010
1.10.2010
6.10.2010
11.10.2010
16.10.2010
21.10.2010
26.10.2010
Date and time (UTC+2)


A
Source ★ at 62.91 N 27.66 E
Meters AGL
1500
1000
500
500
06 00 18 12 06 00 18 12 06 00 18
09/24 09/23 09/22
A


B
Source ★ at 62.91 N 27.66 E
Meters AGL
1500
1000
500
500
00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18
10/08 10/07 10/06 10/05 10/04 10/03


C
Source ★ at 62.91 N 27.66 E
Meters AGL
1500
1000
500
500
12 06 00 18 12 06 00 18 12 06 00 18
10/09 10/08 10/07

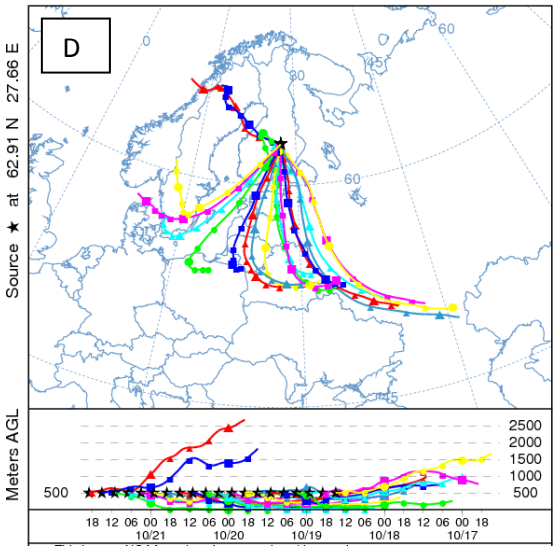

D
Source ★ at 62.91 N 27.66 E
Meters AGL
2500
2000
1500
1000
500
500
18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18
10/21 10/20 10/19 10/18 10/17

Figure S10. Top panel: time series of LVOOA and mass fractions of LVOOA to total organic. Four periods on the LVOOA plumes were selected for the back trajectory studies marked by blue bars. Bottom panel: Back trajectory analysis on the sources of LVOOA. The trajectories were conducted on the four plumes of LVOOA time series, showing LVOOA were from South Finland, south Sweden, central Europe.

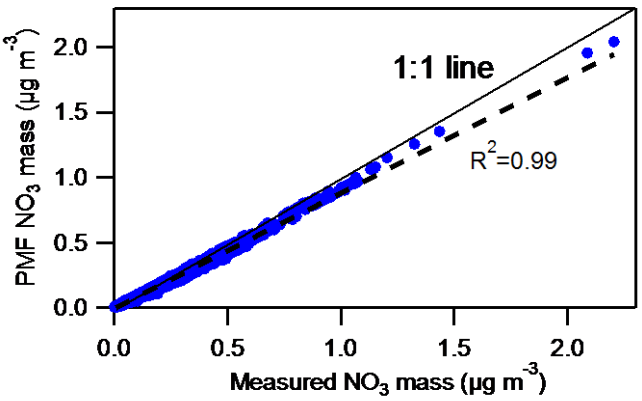


Figure S11. Comparisons of mass concentrations of nitrate aerosols between the fitted by PMF and the measured by AMS.

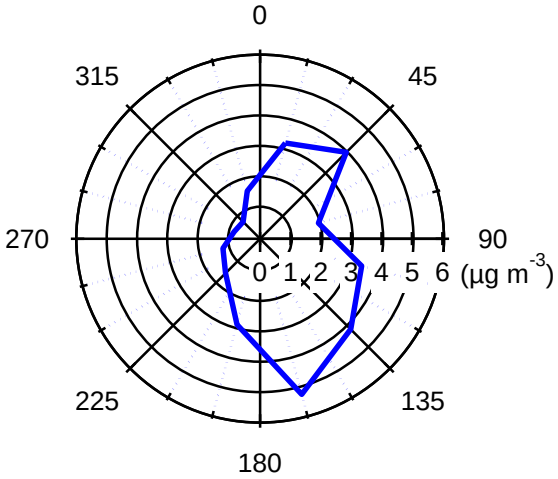


Figure S12. Wind rose for NO_2.

References

Draxler, R.R., and Rolph, G.D.: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://ready.arl.noaa.gov/HYSPLIT.php). NOAA Air Resources Laboratory, Silver Spring, MD, 2003.

Zhang, Q., Jimenez, J. L., Worsnop, D. R., and Canagaratna, M.: A case study of urban particle acidity and its influence on secondary organic aerosol, Environ. Sci. Technol., 41, 3213–3219, 2007.