Supplement Information of

Reduced volatility of aerosols from surface emission to the top of planetary boundary layer

Quan Liu[1,3], Dantong Liu[2,*], Yangzhou Wu[2], Kai Bi[1], Wenkang Gao[4], Ping Tian[1], Delong Zhao[1], Siyuan Li[2], Chenjie Yu[5], Yunfei Wu[6],Kang Hu[2], Shuo Ding[2], Qian Gao[1], Fei Wang[1], Hui He[1,7], Mengyu Hang[1,7], Deping Ding[1]

[1] Beijing Weather Modification Office, Beijing 100089, China

[2] Department of Atmospheric Sciences, School of Earth Sciences, Zhejiang University, Hangzhou, Zhejiang, 310027, China

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

[4] State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

[5] Centre for Atmospheric Sciences, School of Earth and Environmental Sciences, University of Manchester, Manchester M13 9PL, UK.

[6] Key Laboratory of Middle Atmosphere and Global Environment Observation (LAGEO), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

[7] Field experiment base of cloud and precipitation research in North China, China Meteorological Administration, Beijing, 101200, China

Corresponding author: Dantong Liu (dantongliu@zju.edu.cn)

AMS data analysis

Standard AMS data analysis software packages (SQUIRREL 1.59D and PIKA 1.19D) were used to deconvolve mass spectrum and obtain mass concentrations of chemical components. The AMS collection efficiency (CE), which accounts for the incomplete detection of aerosol species due to particle bounce at the vaporizer and/or the partial transmission of particles by the lens (Canagaratna et al., 2007), is significantly modulated by particle phase (Matthew et al., 2008). In this study, we used a CE correction following the principle developed by Middlebrook et al. (2012).(Middlebrook et al., 2012)

High-resolution mass spectra of OA for m/z 12-150 at both sites were analyzed by positive matrix factorization (PMF) with PMF2.exe algorithm (Paatero and Tapper, 1994), following the data-processing and factors-selecting steps given by Ulbrich et al. (2009) and Zhang et al. (2011).(Ulbrich et al., 2009;Zhang et al., 2011) The PMF-derived mass spectra and time series of organic components at both sites are shown in Fig. S4 and S5, while the detailed diagnostic plots of the PMF results are also presented in Fig. S2 and S3. As a result, five factors were resolved from the surface OA, including three primary OA (POA), that is, hydrocarbon-like OA (HOA) from traffic emissions, cooking-related OA (COA), and coal combustion OA (CCOA), and two secondary OA (SOA), that is, semi-volatile oxidized OA (SV-OOA) and low-volatility oxygenated OA (LV-OOA). Note that the COA factor was not resolved from the mountain OA. Like the results here, the high similarity of mass spectra (mainly for hydrocarbon ion series) between HOA and COA (Fig. S4) may lead to the difficult discrimination between both factors for the processed OA. Previous studies also tended to mainly resolve the COA factor in urban environment close to sources rather than for more ageing environment (Zhang et al., 2011;Mohr et al., 2012). The HOA factor at mountain may therefore involve some of the transported cooking sources from the surface while it is unable to be clearly distinguished.

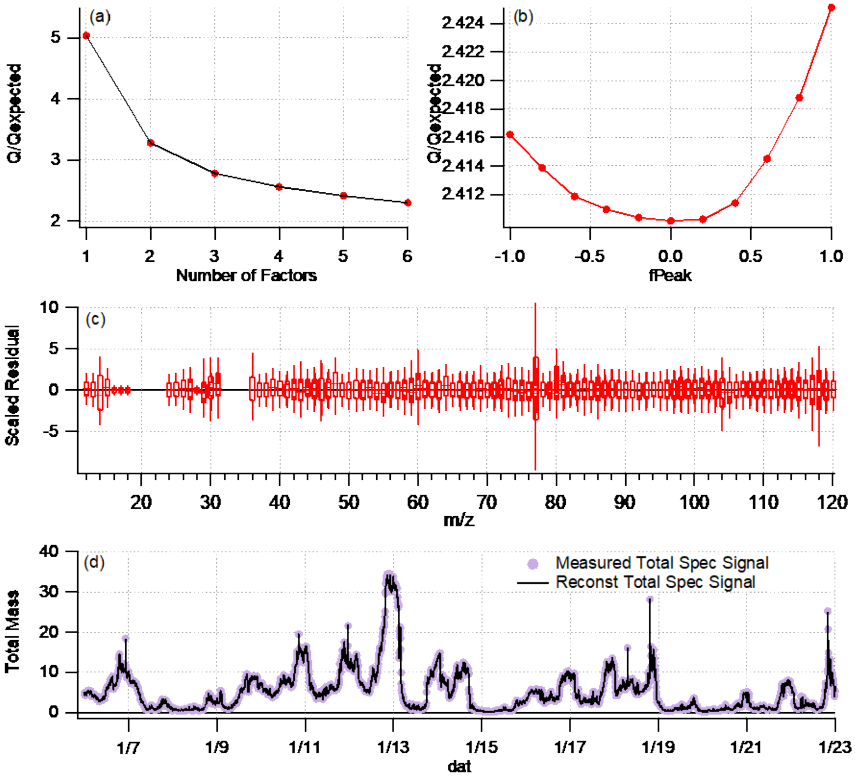

Fig. S1 PMF diagnostic plots of the surface site: (a) $Q/Q_{expected}$ varied as a function of the number of factors, (b) $Q/Q_{expected}$ varied as a function of *f*peak, (c) scaled residual for each m/z, (d) time series of measured and PMF reconstructed organic mass concentrations.

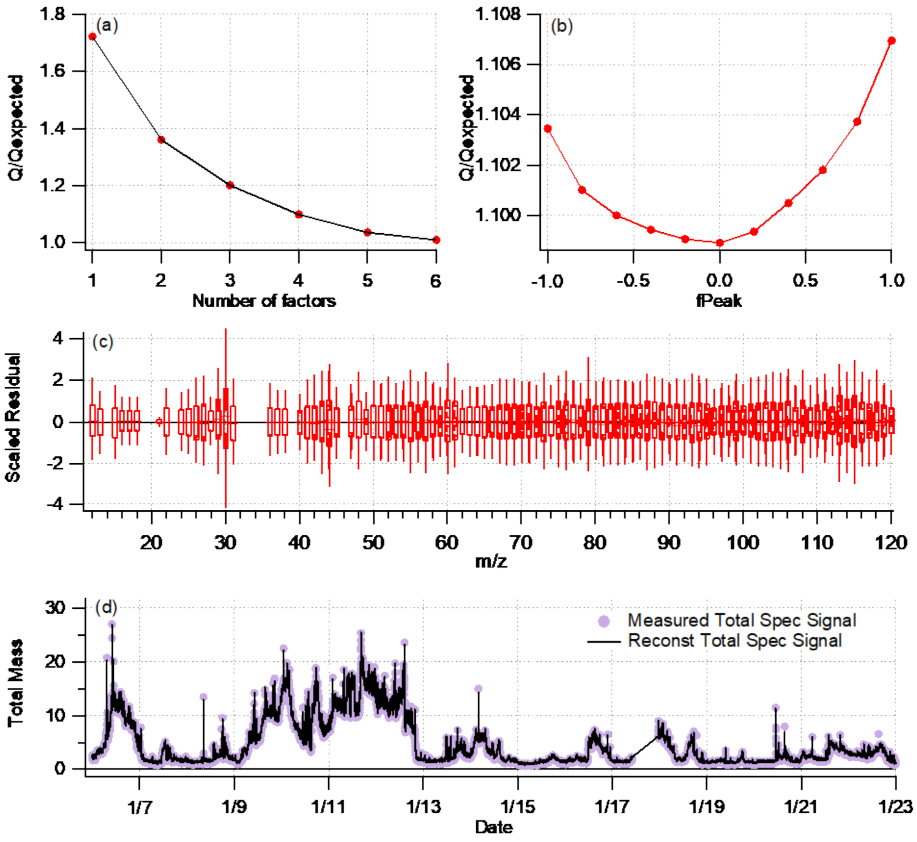

Fig. S2 PMF diagnostic plots of the mountain site: (a) $Q/Q_{expected}$ varied as a function of the number of factors, (b) $Q/Q_{expected}$ varied as a function of *f*peak, (c) scaled residual for each m/z, (d) time series of measured and PMF
55 reconstructed organic mass concentrations.

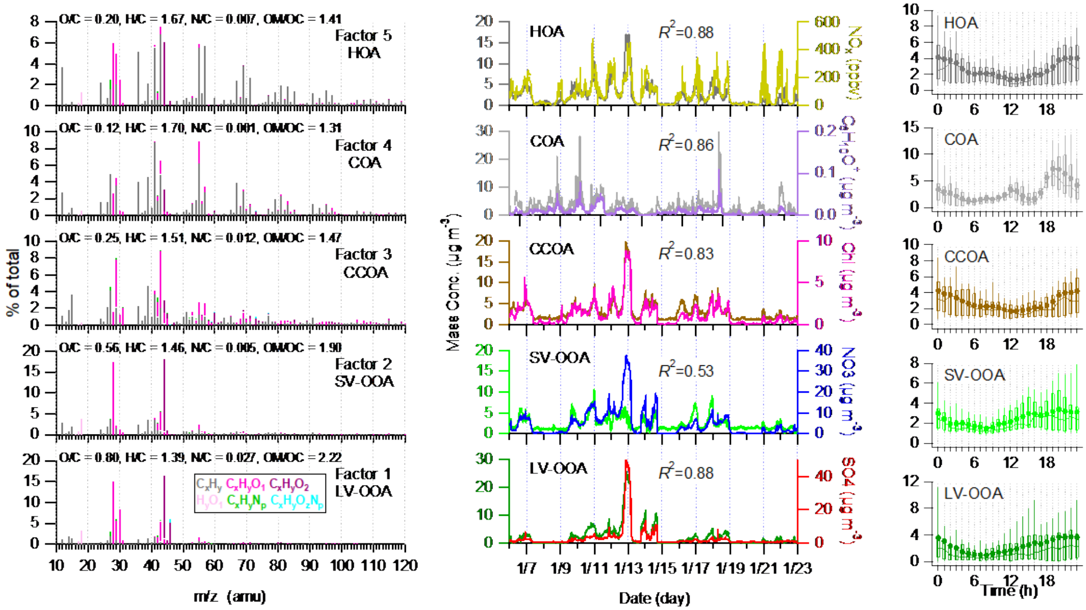

Fig. S3 Mass spectra (left column), time series (middle column), and diurnal variations (right column) of organic factors derived from the data of the surface site. Time series of corresponding tracers are given in the middle column for comparisons.

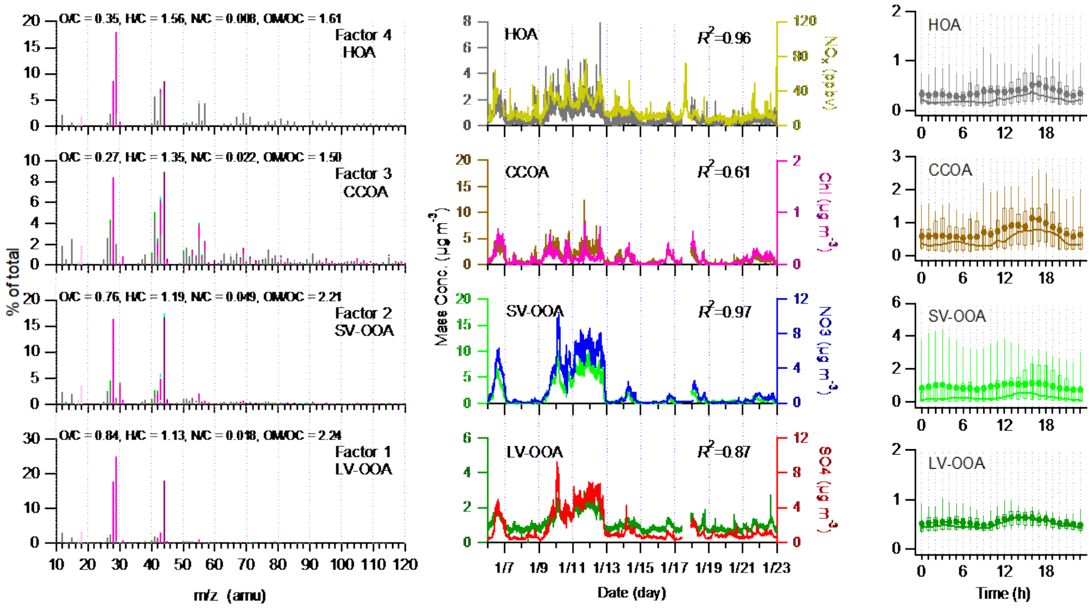

Fig. S4 Mass spectra (left column), time series (middle column), and diurnal variations (right column) of organic factors derived from the data of the mountain site. Time series of corresponding tracers are given in the middle column for comparisons.

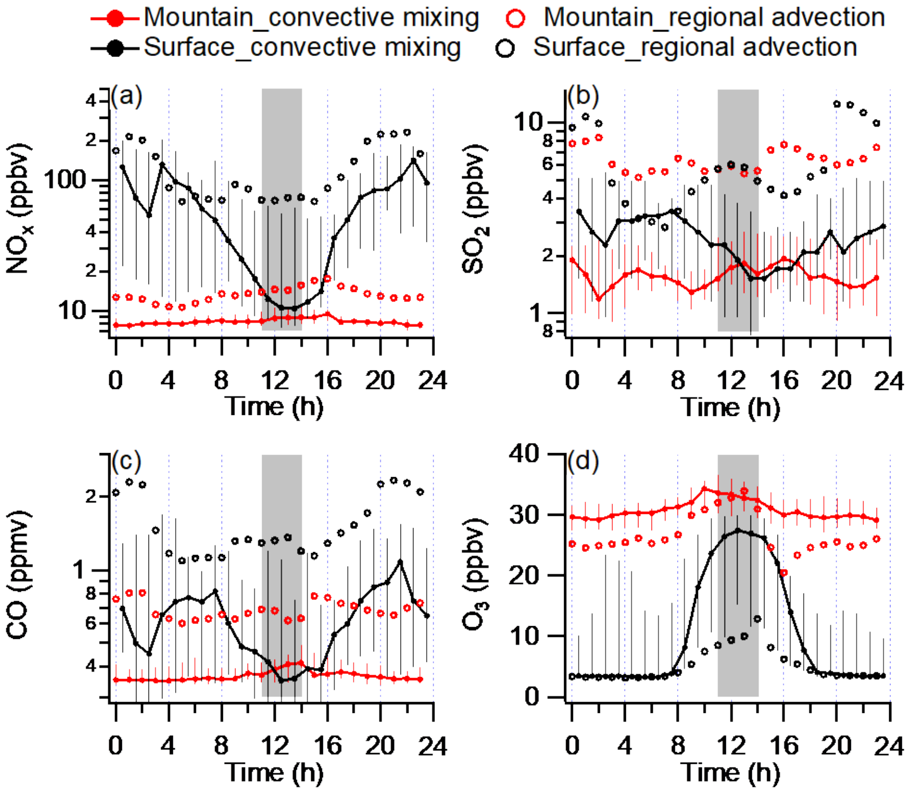

Fig. S5 Diurnal variations of (a) NO_x, (b) SO_2, (c) CO, and (d) O_3 during the observation. Black and red colors represent the surface and mountain sites respectively. Solid circles and error bars show the median, 75th, and 25th percentiles in CM period. Circle markers show the mean value in RA period.

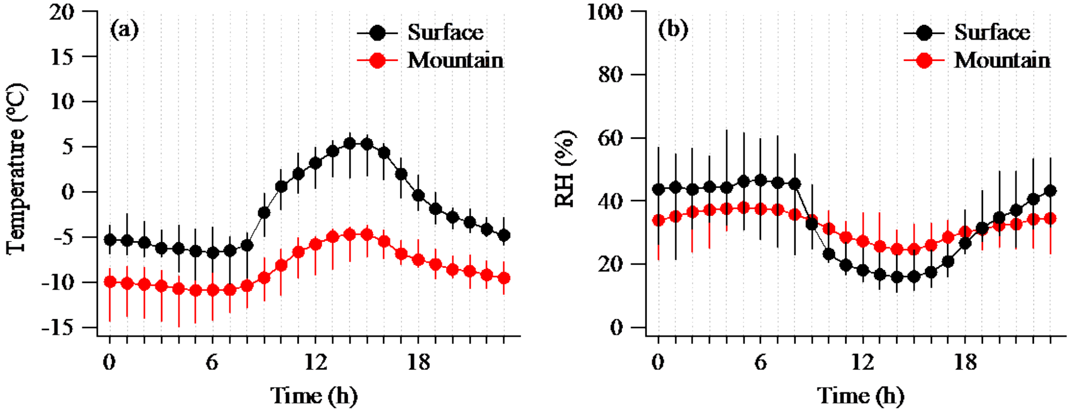

Fig. S6 Diurnal variations of (a) Temperature, and (b) relative humidity (RH) during the observation. Black and red 80 colors represent the surface and mountain sites respectively. Solid circles show the mean value with error bars of 75th, and 25th percentiles.

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