

Supplement of

**Long-range transport biomass burning emissions to the Himalayas:
insights from high-resolution aerosol mass spectrometry**

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Text

Text S1. Estimation of organic nitrates

Organic nitrates, which are produced either by photochemical (OH-initiated) or nocturnal (NO₃-initiated) oxidation reactions of anthropogenic and biogenic volatile organic compounds (VOCs), are important components of secondary organic aerosol in the troposphere (Farmer et al., 2010). Model studies have suggested that photochemical ON production accounts for a substantial fraction of SOA functionalization (Camredon et al., 2007).

The total nitrates measured by the HR-ToF-AMS is the nitrate functionality (–ONO₂), which may consist of both inorganic and organic nitrates. However, the direct measurements of particle-phase organic nitrates are not available currently with the HR-ToF-AMS measurements. For solving this restriction, an estimation method based on the NO⁺/NO₂⁺ ratio (denoted as NO_x⁺ ratio method hereafter) in the AMS mass spectra is used widely to separate the measured total nitrates into nitrate functionality from inorganic and organic nitrates Farmer et al. (2010). Previous laboratory studies have shown that the NO_x⁺ ratios are substantially different for organic nitrates and inorganic nitrate (NH₄NO₃), namely the ratios for organic nitrates are typically 2–3 times higher than those for NH₄NO₃ (Fry et al., 2009; Bruns et al., 2010; Farmer et al., 2010; Boyd et al., 2015). Based on the difference in NO_x⁺ ratios, Farmer et al. (2010) have proposed an estimation method to calculate the concentrations of NO⁺ and NO₂⁺ fragments from organic nitrate by the following equations.

$$\text{NO}_{2,\text{ON}} = \frac{\text{NO}_{2,\text{meas}} \times (R_{\text{meas}} - R_{\text{AN}})}{R_{\text{ON}} - R_{\text{AN}}} \quad (1)$$

$$\text{NO}_{\text{ON}} = R_{\text{ON}} \times \text{NO}_{2,\text{ON}} \quad (2)$$

where the subscripts meas, AN, and ON are denoted as concentrations of NO⁺ and NO₂⁺ fragments or NO_x⁺ ratios (R_{meas} , R_{AN} and R_{ON}) from ambient measurement, pure ammonium nitrate and organic nitrates, respectively.

In this study, the R_{AN} values were determined from ionization efficiency (IE) calibrations using ammonium nitrate particles at the beginning, the middle, and the end of the campaigns, with an average value of 3.08 ± 0.15 , which was within the range reported in the literature. Although the R_{ON} value is more difficult to determine due to its variance between instruments and precursor volatile organic compounds, a $R_{\text{ON}}/R_{\text{AN}}$ ratio which has been revealed to be instrument independent was determined widely for different organic nitrates in the literature, with values ranging from 2.08 for isoprene to 3.99 for β -pinene (Bruns et al., 2010; Fry et al., 2013; Boyd et al., 2015). Thus, the lower and upper values of R_{ON} can be estimated by multiplying R_{AN} value determined from IE calibrations with $R_{\text{ON}}/R_{\text{AN}}$ mentioned above. Finally, the lower and upper limits of R_{ON} were calculated to be 6.41 and 12.29, respectively, to estimate the amount of organic nitrates in this study. Noting that this method is relatively rough to estimate the organic nitrates based on the HR-ToF-AMS data, the direct and accurate measurement of organic nitrates warrants

further study.

Most of the measured ambient $\text{NO}^+/\text{NO}_2^+$ ratios are within the range of the calibrated value (i.e., R_{AN} , 3.08) and the estimated upper value (i.e., $R_{\text{ON, upper}}$, 12.29) and with an average value of 4.43 ± 1.39 , which make the estimation of organic nitrates at QOMS possible. The time series of the measured total nitrate concentrations and $\text{NO}^+/\text{NO}_2^+$ ratios as well as the estimated lower and upper nitrate functionality from organic nitrates in this study are displayed in Fig. S8, while the diurnal cycle of mass concentration of the estimated organic nitrate is showed in Fig. S9. Finally, the average lower and upper nitrate functionality from organic nitrates are estimated to be 0.031 and 0.048 $\mu\text{g m}^{-3}$, respectively, which contributed 15–23 % of the total measured nitrate, suggesting that organic nitrates are significant components in aerosol particles at QOMS. This result is comparable with other studies based on AMS data. For example, Xu et al. (2015) has estimated that the nitrate functionality from organic nitrates could contribute about 63–100 and 10–20% of the total measured nitrates in summer and winter in the southeastern USA, respectively. Zhu et al. (2016) also estimated the nitrate functionality from organic nitrates in a southern China background site, which was calculated to be 15–22 % of the total measured nitrates.

Tables

Table S1. Summary of the average mass concentrations of total PM₁ and chemical compositions measured with the Aerodyne AMS instruments at various remote sites worldwide.

Dataset Location	PM ₁	Organics	Sulfate	Nitrate	Ammonium	References
Mt. Jungfrauoch (3580 m a.s.l.), Switzerland	0.55	0.25	0.17	0.04	0.05	Fröhlich et al. (2015)
Mt. Jungfrauoch (3580 m a.s.l.), Switzerland	2.24	1.20	0.51	0.20	0.33	Zhang et al. (2007)
Mt. Whistler (2182 m a.s.l.), Canada	1.91	1.05	0.58	0.05	0.23	Sun et al. (2009)
Mt. Bachelor (2.8 km a.s.l.), USA	15.10					Zhou et al. (2017)
Mace Head, Ireland	1.53	0.50	0.75	0.05	0.23	Zhang et al. (2007)
Hyytiala, Finland	2.04	1.20	0.31	0.24	0.28	Zhang et al. (2007)
Storm Peak, USA	2.11	0.90	0.59	0.22	0.40	Zhang et al. (2007)
Duke Forest, USA	2.82	1.30	1.10	0.10	0.32	Zhang et al. (2007)
Chebogue, Canada	2.91	1.60	0.89	0.08	0.34	Zhang et al. (2007)
Okinawa Island, Japan	7.89	1.70	4.70	0.15	1.30	Jimenez et al. (2009)
Fukue Island, Japan	12.03	5.03	4.80	0.56	1.57	Takami et al. (2005)
Jeju Island, Korea	10.66	4.30	3.90	0.76	1.70	Jimenez et al. (2009)

Table S2. Coefficient of determination (R^2) between time series of OA factors and other aerosol species. Values indicating the three stronger correlations for each components are in bold.

R^2	BBOA	NOA	MO-OOA	BBOA + NOA	NOA + MO-OOA
Sulfate	0.39	0.46	0.65	0.48	0.70
Nitrate	0.75	0.51	0.64	0.82	0.72
Ammonium	0.68	0.54	0.70	0.78	0.78
Chloride	0.62	0.46	0.40	0.70	0.51
Sulfate + Nitrate	0.58	0.52	0.70	0.67	0.77
Sulfate + Nitrate + Chloride	0.59	0.53	0.70	0.68	0.77
BC	0.72	0.57	0.66	0.82	0.76
C ₂ H ₄ O ₂ ⁺	0.91	0.48	0.48	0.96	0.59
C ₃ H ₅ O ₂ ⁺	0.87	0.52	0.56	0.94	0.67
Organic nitrates	0.45	0.62	0.40	0.58	0.57

Figures

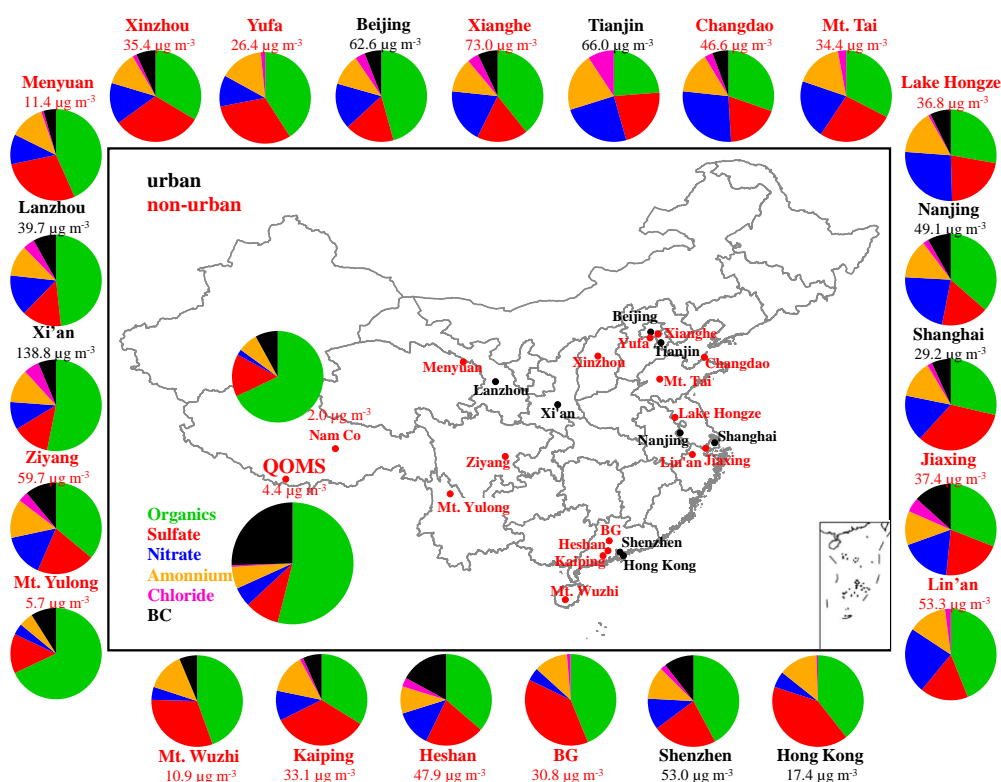


Fig. S1. Summary of the average mass concentrations and chemical compositions of PM_{10} measured with the Aerodyne AMS and ACSM instruments at various sites in China. Colors for the site labels indicate the type of sampling location as urban (black) or non-urban (red) site. Colors for the each piechart represents chemical compositions as organics (green), sulfate (red), nitrate (blue), ammonium (orange), chloride (purple), and black carbon (black). Detailed data for each site can refer to studies conducted in Beijing (Huang et al., 2010; Sun et al., 2010; Sun et al., 2012; Jiang et al., 2013; Sun et al., 2013; Sun et al., 2014; Zhang et al., 2014a; Chen et al., 2015; Sun et al., 2015; Sun et al., 2016b), Tianjin (Zhang et al., 2012), Xi'an (Wang et al., 2014), Lanzhou (Xu et al., 2014; Xu et al., 2016; Zhang et al., 2017), Nanjing (Tang et al., 2014; Zhang et al., 2015b; Wang et al., 2016a), Shanghai (Huang et al., 2012), Shenzhen (Yao et al., 2010; He et al., 2011), Hong Kong (Li et al., 2015; Sun et al., 2016a), Yufa (Gunthe et al., 2011), Xianghe (Sun et al., 2016c), Xinzhou (Wang et al., 2016b), Changdao (Hu et al., 2013), Mt. Tai (Zhang et al., 2014b), Lake Hongze (Zhu et al., 2016), Jiaxing (Huang et al., 2013), Lin'an (Zhang et al., 2015a; Zhang et al., 2015c), Menyuan (Du et al., 2015), Ziyang (Hu et al., 2016), Mt. Yulong (Zheng et al., 2017), BG (Xiao et al., 2011), Heshan (Gong et al., 2012), Kaiping (Huang et al., 2011), Mt. Wuzhi (Zhu et al., 2016), Nam Co (Xu et al., 2017), and QOMS in this study, respectively.

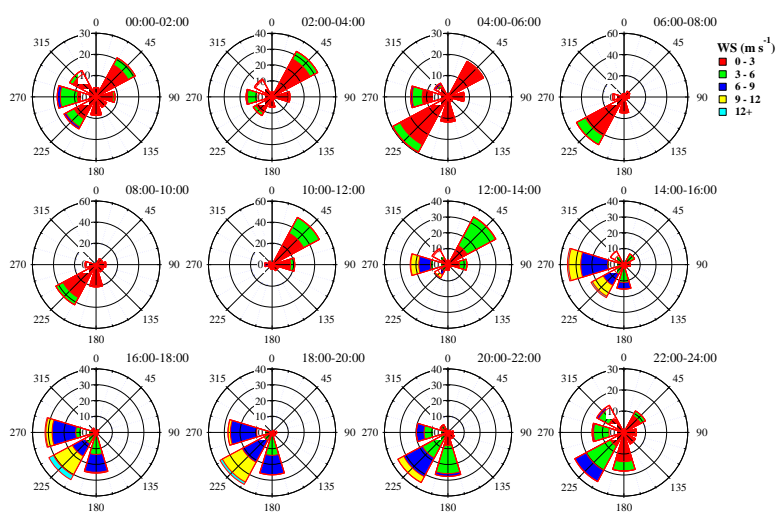


Fig. S2. Two-hour internal average wind rose plots for the entire period of 2016 Everest study.

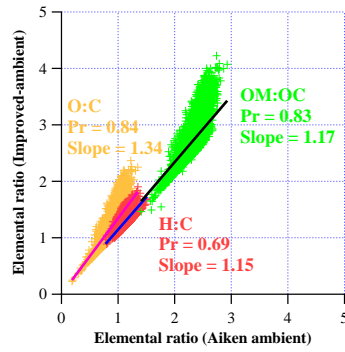


Fig. S3. Scatter plot of values calculated with “Improved-ambient” method versus that with “Aiken ambient” method

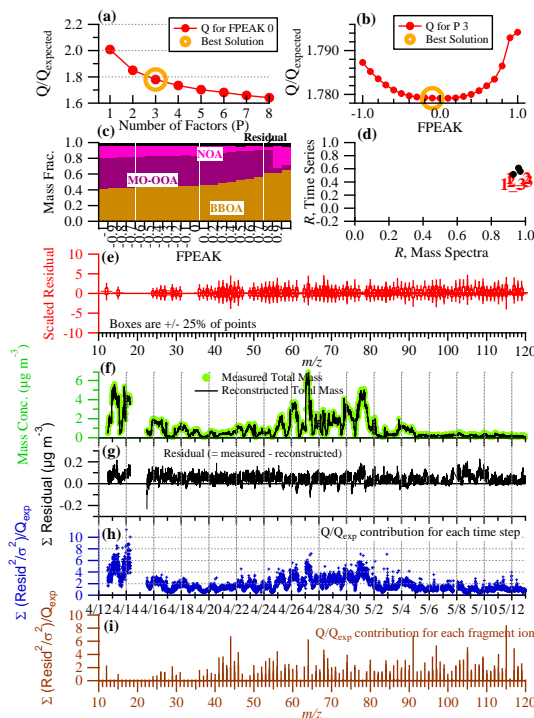


Fig. S4. Summary of key diagnostic plots of the PMF results: (a) Q/Q_{exp} as a function of number of factors (p) selected for PMF analysis. For the best solution (three factors solution): (b) Q/Q_{exp} as a function of FPEAK, (c) fractions of OA factors vs. FPEAK, (d) correlations among PMF factors, (e) the box and whiskers plot showing the distributions of scaled residuals for each m/z , (f) time series of the measured organic mass and the reconstructed organic mass (BBOA + Local-OOA + LRT-OOA), (g) variations of the residual (= measured - reconstructed) of the fit, (h) the Q/Q_{exp} for each point in time, and (i) the Q/Q_{exp} values for each ion.

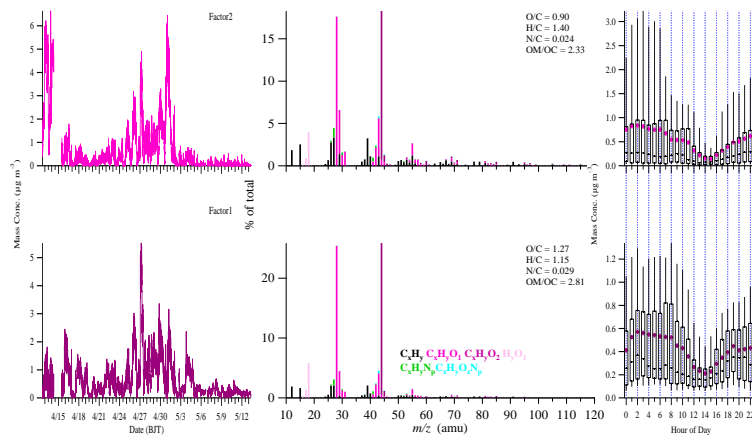


Fig. S5. Two factors solution analyzed by PMF analysis.

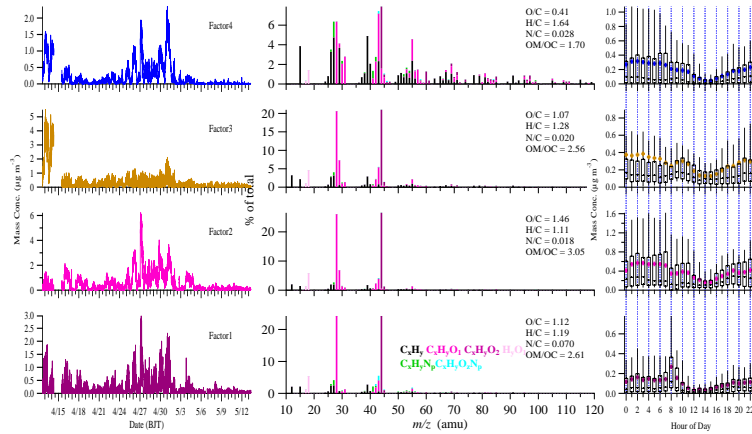


Fig. S6. Four factors solution analyzed by PMF analysis.

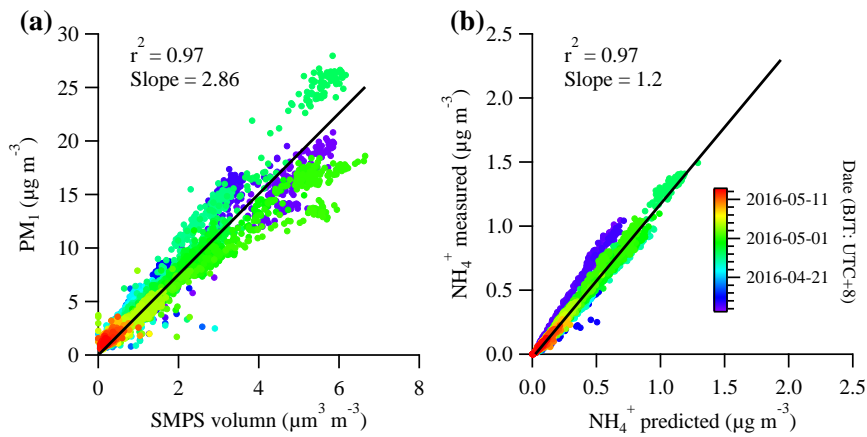


Fig. S7. Scatterplot of (a) PM_1 mass concentrations (NR- PM_1 + BC) versus particle volumes calculated from the SMPS by assuming spherical particles and (b) mass concentrations of measured NH_4^+ versus predicted NH_4^+ . The predicted NH_4^+ concentrations were calculated based on the SO_4^{2-} , NO_3^- , and Cl^- concentrations from the AMS and assumed full neutralization of these anions by NH_4^+ . Both the two black solid lines indicate the linear regression lines.

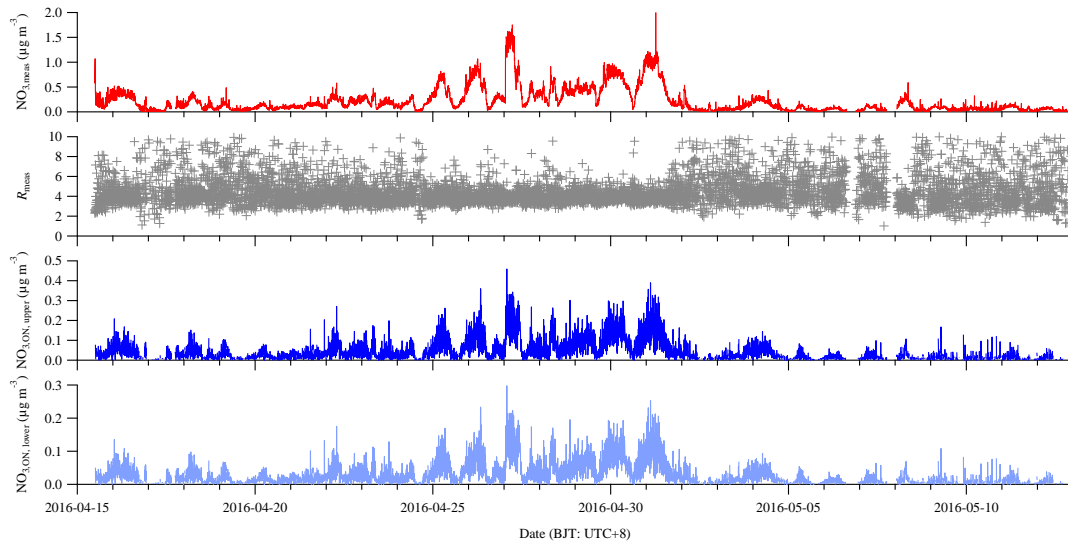


Fig. S8. The time series of the measured total nitrate concentrations and NO^+/NO_2^+ ratios as well as the estimated lower and upper organic nitrates.

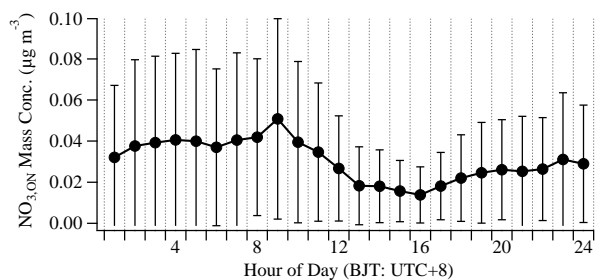


Fig. S9. The diurnal cycles of the estimated organic nitrates.

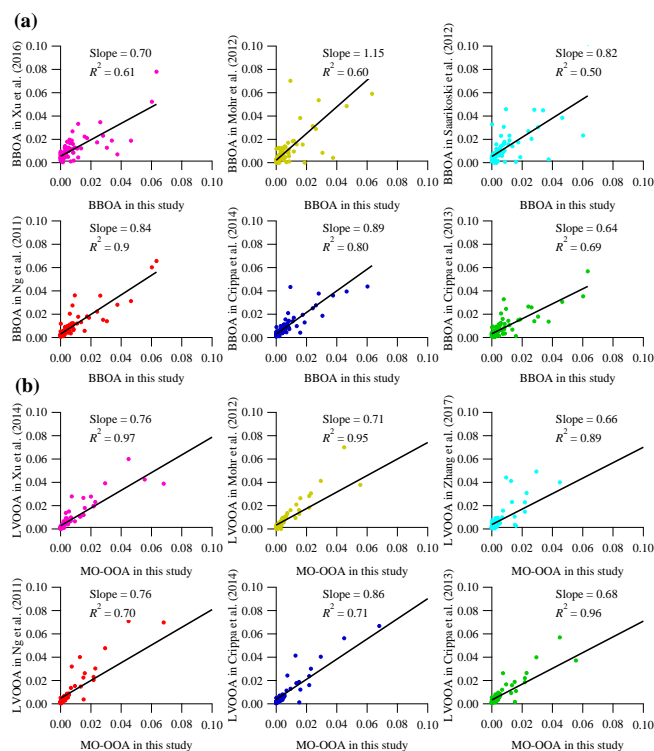


Fig. S10. Scatter plots of the high-resolution mass spectrums of BBOA (ignoring high signals on m/z 28 and 44) and MO-OOA in this study with those mass spectrums determined from other studies.

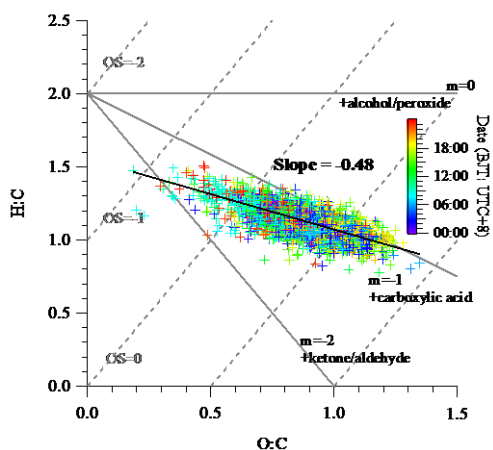


Fig. S11. Van Krevelen diagram for OA with linear fit. The measured OA data points are colored by time of the day.

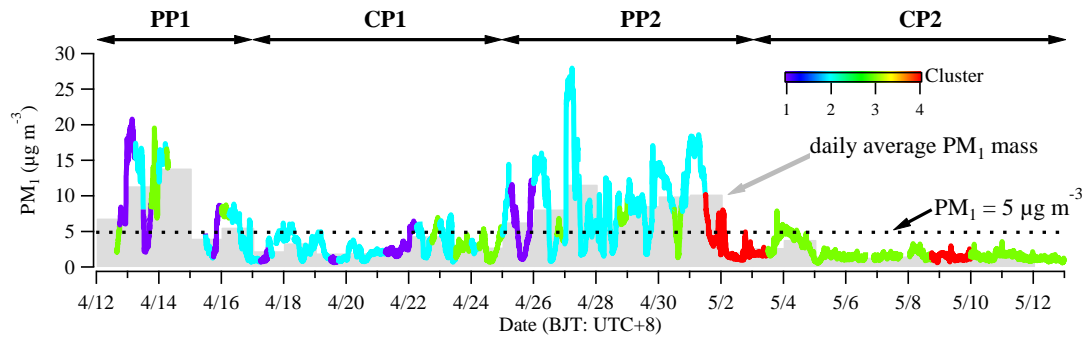


Fig. S12. Time series of 5-min PM_{10} mass concentrations colored by different back trajectory clusters (line) and daily average PM_{10} mass concentrations (grey histogram) for the entire study. The broken line indicate PM_{10} mass concentrations equal to $5 \mu g m^{-3}$. PP1 and PP2 are identified as two polluted periods while CP1 and CP2 are clear periods.

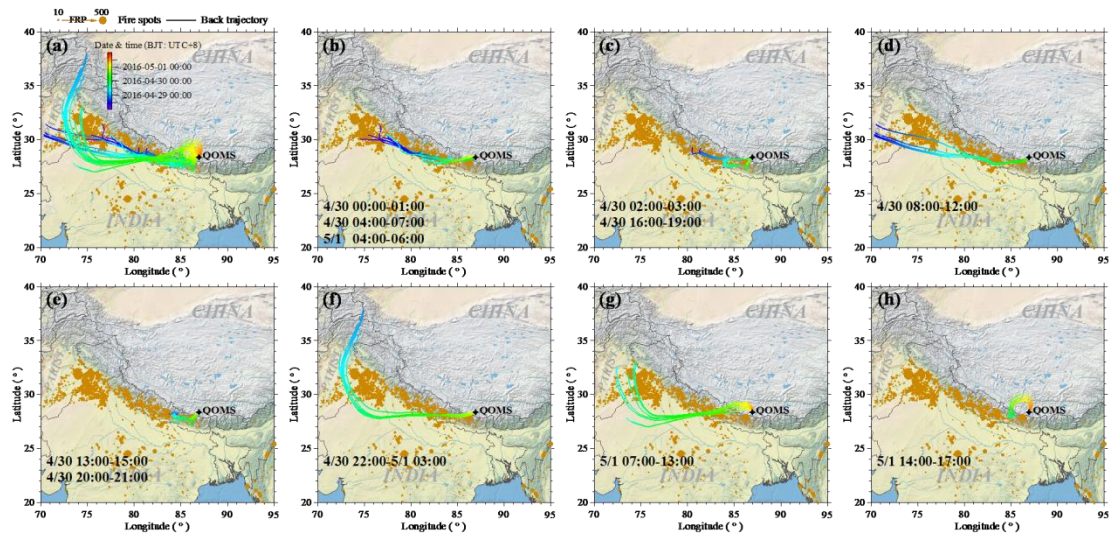


Fig. S13. The 48-h back trajectories calculated every 1 h and MODIS fire hotspots information for (a) the entire case study period from 30 April at 00:00 to 1 May at 18:00, and (b-h) different clusters during various time periods which have been marked in each figures.

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