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First simultaneous measurements of peroxyacetyl nitrate (PAN) and ozone at Nam Co in the central Tibetan Plateau: impacts from the PBL evolution and transport processes

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Indirect calibration of PAN measurements

To obtain acceptable results using the indirect calibration method, we need two assumptions. First, the ambient concentration of CCl_4 at the observation site should be nearly constant during the measurement period. Second, whatever the ECD sensitivity changes with varying environmental conditions, the changes in relative responses of the ECD to PAN and CCl_4 should be the same during the period of consideration. In polluted areas, the first assumption is inapplicable simply because there is large spatial and temporal variation of CCl_4 emission. Even at the regional background site often impacted by polluted air masses, the CCl_4 concentration could be highly varying (Yao et al., 2010). However, CCl_4 is believed to be well mixed to a large scale in clean area air due to negligible emission and long lifetime (42 ± 12 years), thus its concentration at remote sites can be treated as constant in a short period (Simmonds et al., 1998). Based on this idea, Wang et al. (2000) suggested using CCl_4 as an internal reference in the preparation of standard gas mixtures. The second assumption is prerequired for any application of similar indirect calibration and normally applicable if the ECD sensitivity is stable with a GC run. On the basis of the two assumptions above, the ratio between the PAN and CCl_4 signals is used as the key quantity for correcting the PAN data. Therefore, the corrected PAN concentration is eventually determined by following expression:

$$C'_{PAN} = (C_{PAN} \times S'_{PAN} \times S_{CCl4})/(S_{PAN} \times S'_{CCl4}) \,, \qquad (1)$$

where, C'_{PAN} and C_{PAN} are the concentrations of ambient and standard PAN, respectively; S'_{PAN} and S_{PAN} are the PAN signals of air sample and standard sample, respectively; and S'_{CCl4} and S_{CCl4} are the CCl_4 signals of the air sample and the surrogate CCl_4 signal of the calibration, respectively. Since the standard sample did not contain CCl_4, the CCl_4 signal of the air sample prior to a calibration was taken as the surrogate of CCl_4 signal for the calibration run (S'_{CCl4}). This may introduce additional uncertainty to the PAN data as the ECD sensitivity may change from run to run. However, the change of the ECD sensitivity should be minor between consecutive runs within relative short time. Therefore, equation (1) is acceptable in our indirect calibration.

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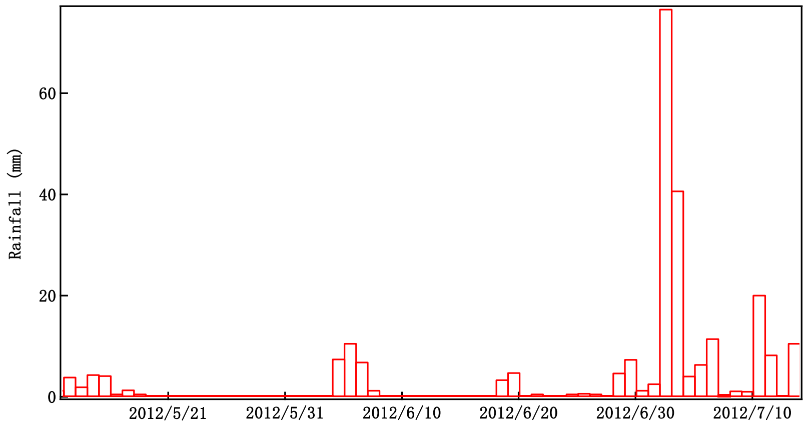

Figure S1 Daily rainfall during the observation period in 2012.

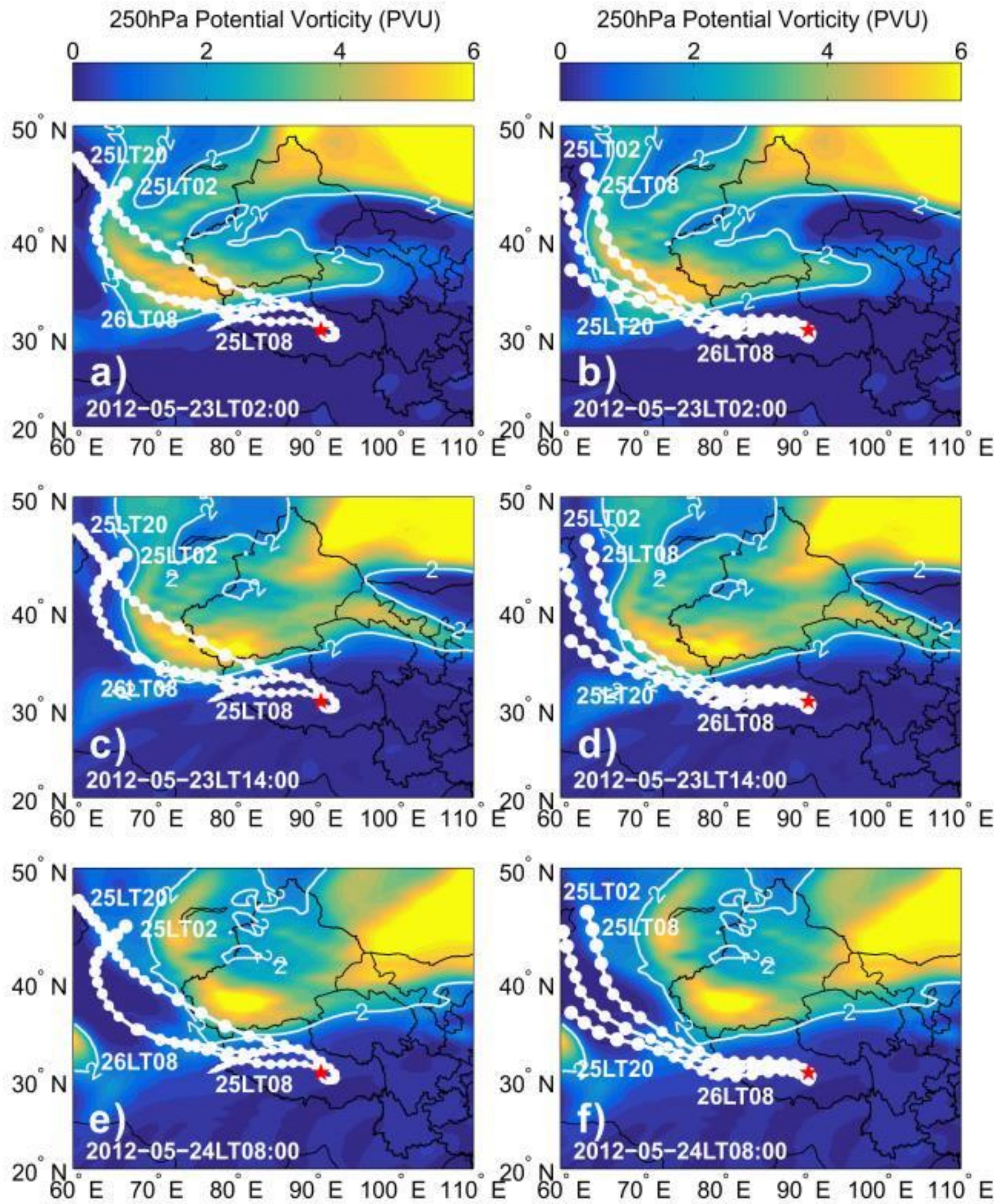

Figure S2 The 250 hPa potential vorticity fields at three timepoints during 23-24 May 2012 and back trajectories of air masses arriving at 500 m (left) and 1500 m (right) above ground of the NMC site (red star) during 25-26 May 2012.

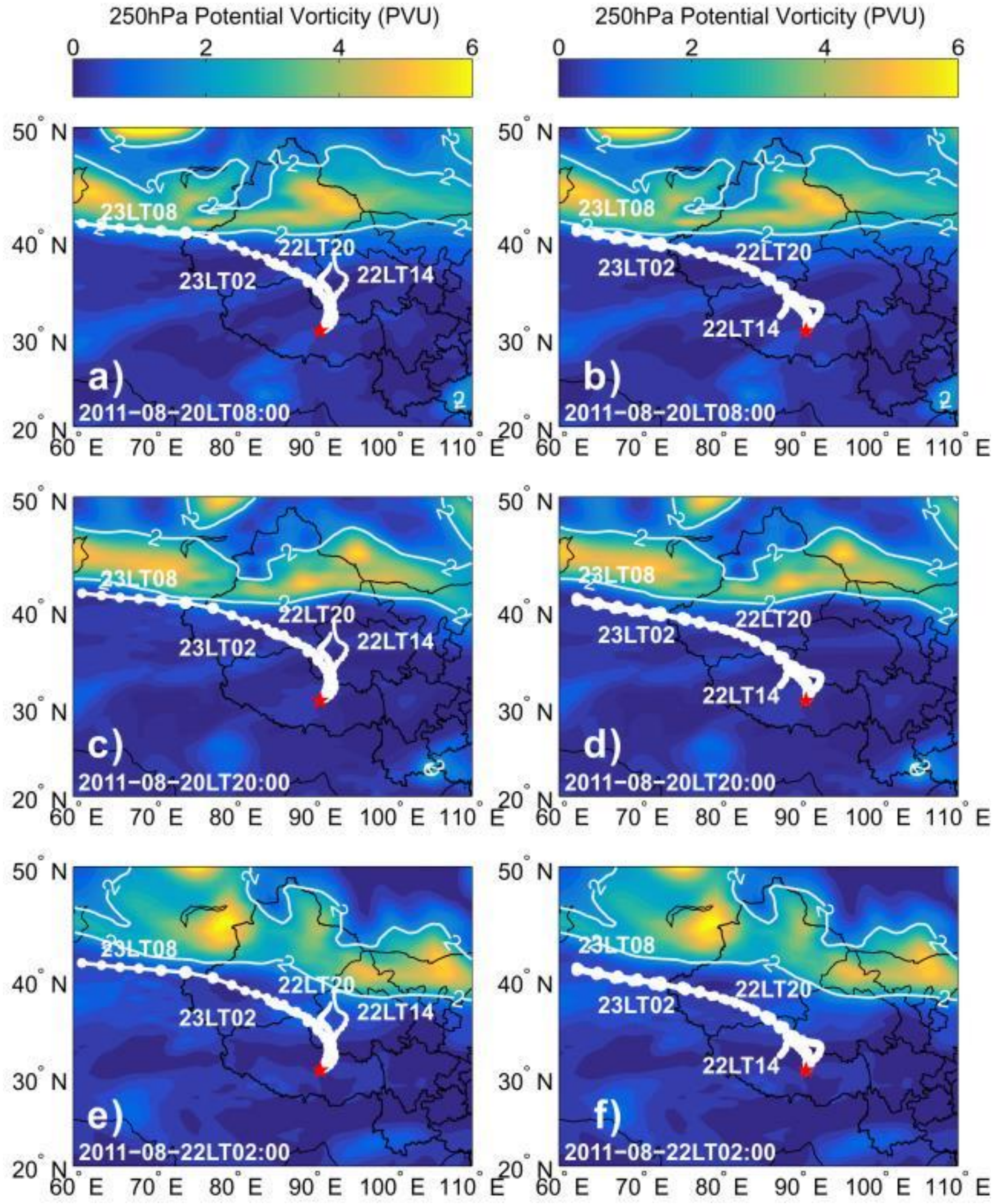

Figure S3 Same Figure S2, but for 22-23 August 2011.

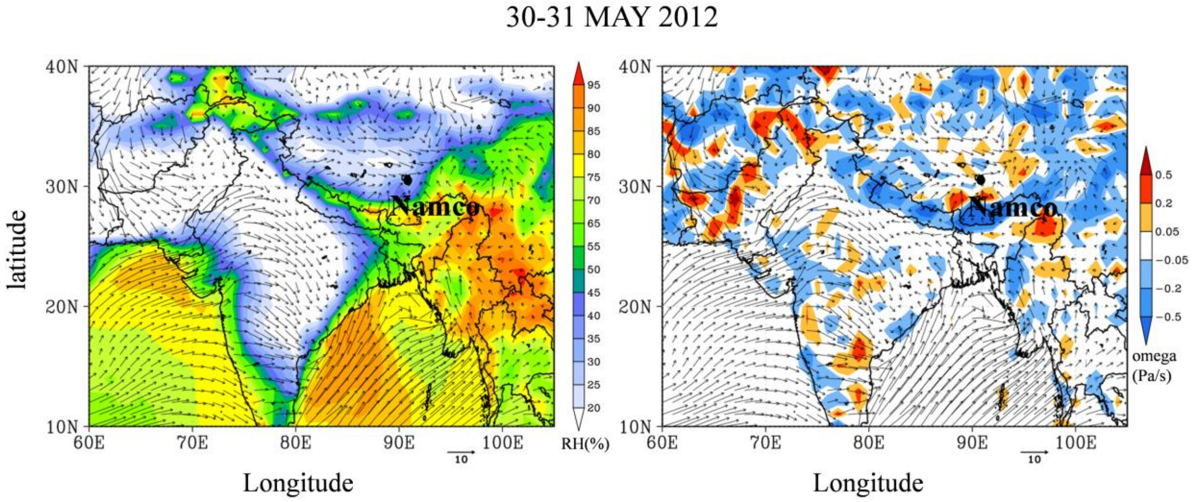

Figure S4 Average fields of wind, relative humidity (a) and ω (b) at sigma=0.995 for the periods 30-31 May 2012.

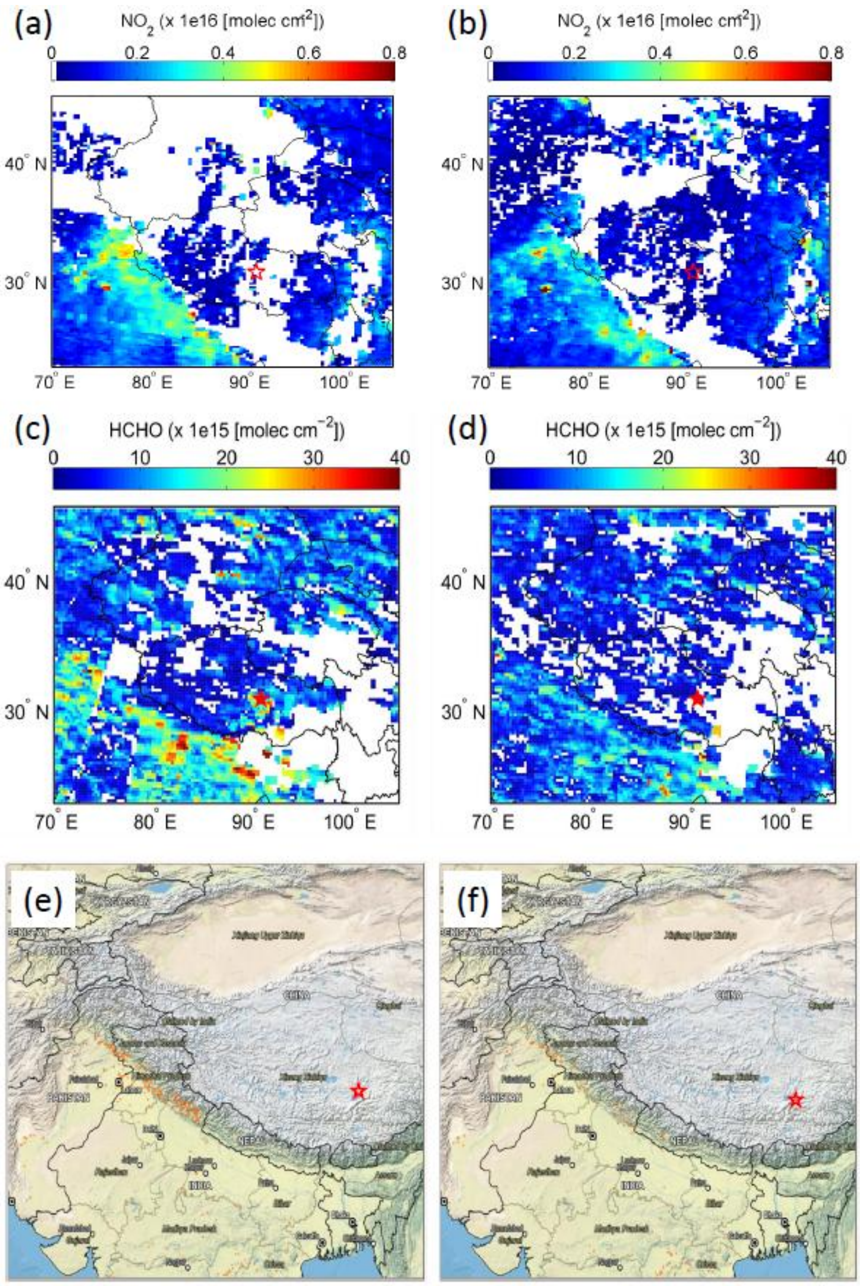

Figure S5 Average column densities of tropospheric NO_2 (a,b) and HCHO (c,d), and maps with fire spots (e,f) for the periods 1-3 (a,c,e) and 4-6 June 2012 (b,d,f). Daily tropospheric NO_2 data are from the OMI observations and made available by NASA (https://daac.gsfc.nasa.gov/datasets). Daily tropospheric HCHO are from GOME-2 observations and provided by the Tropospheric Emission Monitoring Internet Service (TEMIS) at The Royal Netherlands Meteorological Institute (KNMI), The Netherlands (http://www.temis.nl/index.php). Fire spots maps present the fire locations (orange dots) observed by MODIS and are produced by NASA's Web Fire Mapper (https://firms.modaps.eosdis.nasa.gov/firemap/).

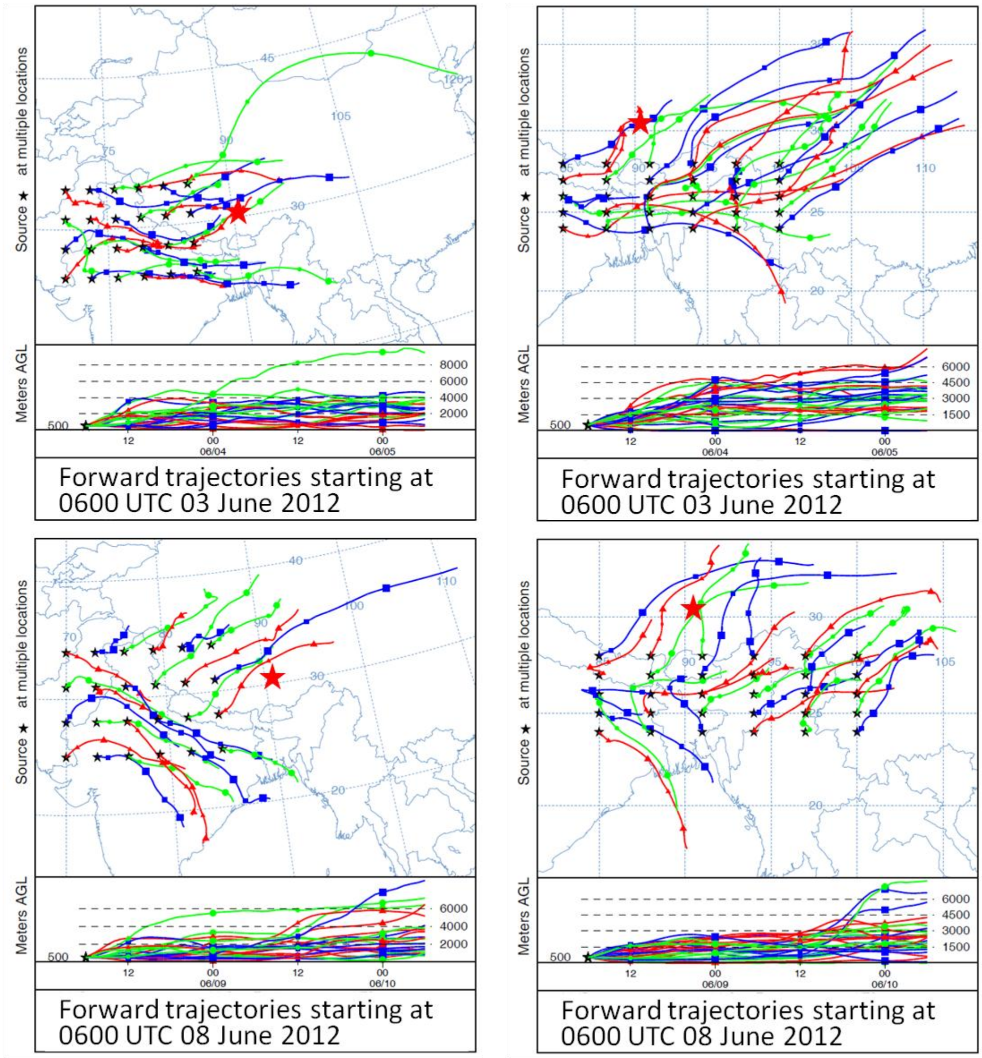

Figure S6 Matrices of 48-h air mass forward trajectories starting at 0600 UTC 3 June 2012 (upper panel) and 0600 UTC 8 June 2012 (bottom panel) from the domains west and south of the NMC site (red star). The online HYSPLIT model (https://ready.arl.noaa.gov/HYSPLIT_traj.php; Stein et al., 2015; Rolph et al., 2017) were used to produce the trajectory matrices. The starting height of the trajectories is 500 m above ground level.

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