Supplement

S1. Description of the WRF performance

The wind fields were computed with the Weather Research and Forecasting (WRF) model version 3.1.1. The model was run in four two-way nested domains at horizontal resolutions of 36 km, 12 km, 4 km and 1 km, with 101×101, 109×109, 121×121, 201×201 grids for domains 1, 2, 3 and 4, respectively. A two-way nested run is a run where multiple domains at different grid resolutions are run simultaneously and communicate with each other: The coarser domain provides boundary values for the nest, and the nest feeds its calculation back to the coarser domain. Both domains had 33 terrain-followed vertical sigma levels. The NCEP FNL reanalyses data (1^{o} horizontal resolution) and Global Telecommunications System (GTS) data were used to provide the initial and boundary conditions for WRF simulation. The MODIS high resolution satellite remote sensing data were used to initialise the land surface parameters for the NOAH land surface model. The physics options were chosen as follows: the Mellor-Yamada-Janjic (MYJ) boundary layer scheme, the Kain-Fritsch (new Eta) scheme only for domain 1 and 2, the WSM3 microphysics scheme, the RRTM longwave and the Dudhia shortwave scheme. The start times of simulations were 20:00 (LT) of the days before mobile observation. Outputs were achieved every 30 minutes (1 hour) for domain 4. After the spin-up period, the model could generate the reliable high resolution (i.e., 1 km) wind field.

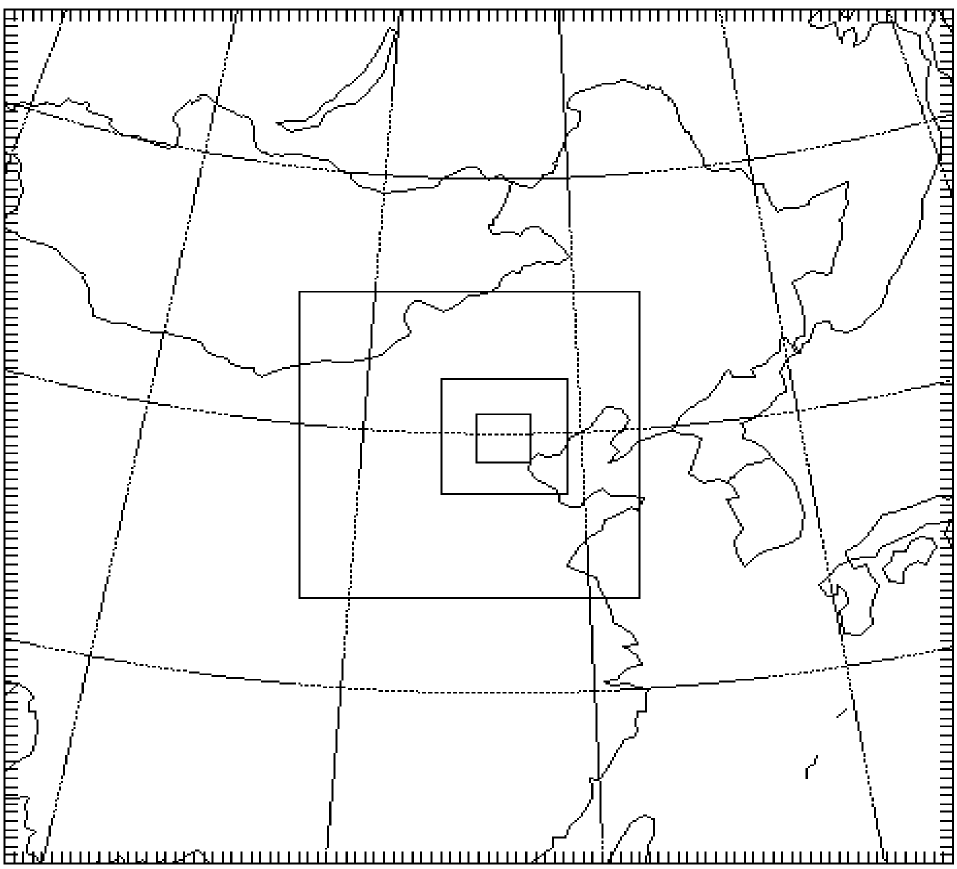

Figure S1. WRF domains 1-4.

S2. Comparisons between observed and predicted wind speed and directions

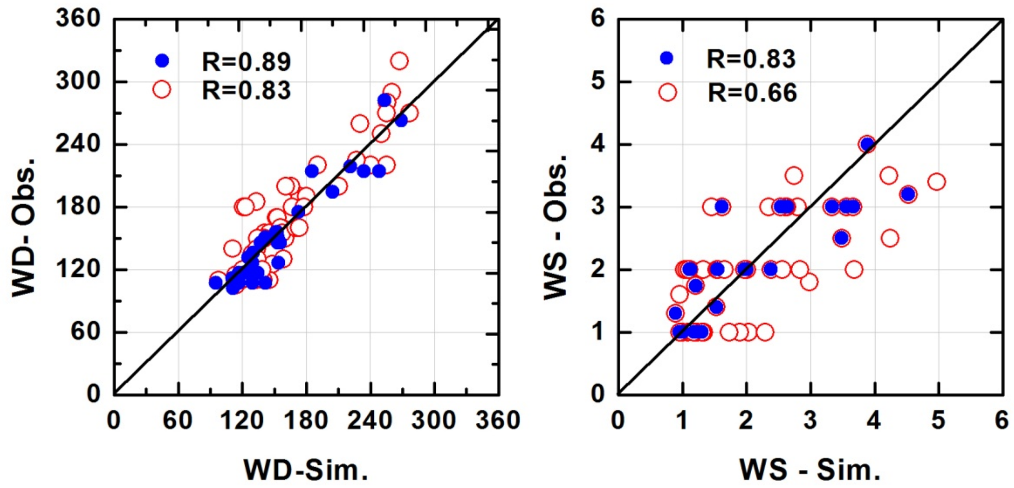

Figure S2 Correlations between observed and simulated wind speed and directions by CAMS stations and WRF model. The blue dots show the CAMS sites in rural areas and the red circles show all the sites from CAMS.

Fig. S2 shows the comparison between the modeling results and the observations from the meteorological stations during the measurement periods. It was found that the correlation coefficient between the predicted and observed wind direction was 0.83 and increased to 0.89 when the urban sites were excludedThe agreement for the wind speed in all sites was slightly low(R=0.66) but was good when only the urban sites involved. This suggested that WRF can generate generally reliable spatial distributions of wind fields for further analysis.

S3. WRF PBL and lidar extinction coefficient comparison

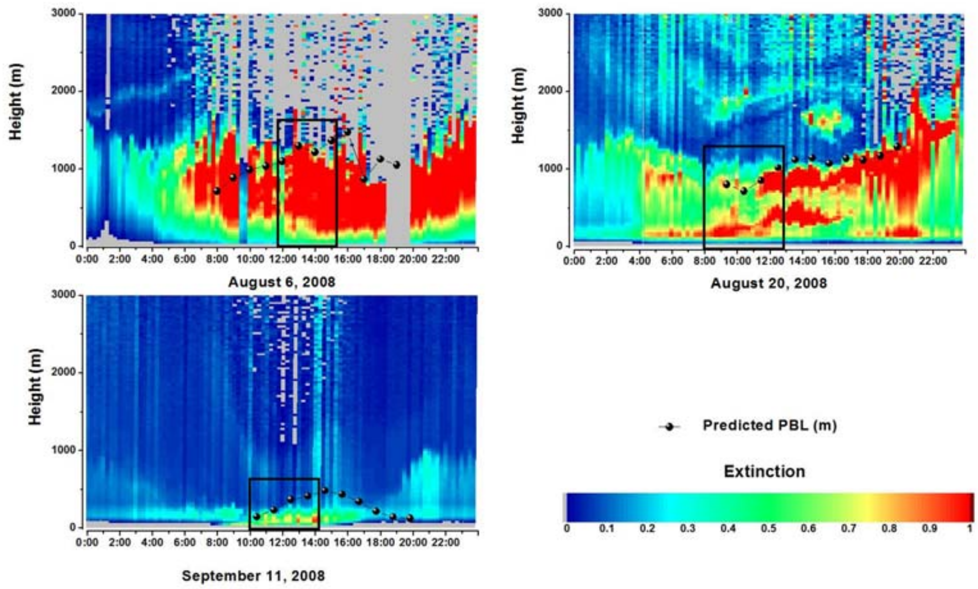

Figure S3 Comparisons of the WRF predicted PBL variations with lidar retrieved extinction coefficients. The lidar measurements were conducted at the the institute of atmospheric physics, Chinese Academy of Sciences (39°58'34.60"N, 116°22'40.39"E) during the Olympic period. Three days (6 August, 20 August and 11 September, 2008) were chosen for the comparison with the model calculated PBL. The mobile samplings were conducted between 12:00-14:50 (6 August), 7:58-12:50 (20 August) and 10:09-14:37 (11 September) which have been marked by black boxes. A good agreement can be found during the mobile monitoring period.

S4. The PBL distrubutions during the measurement periods

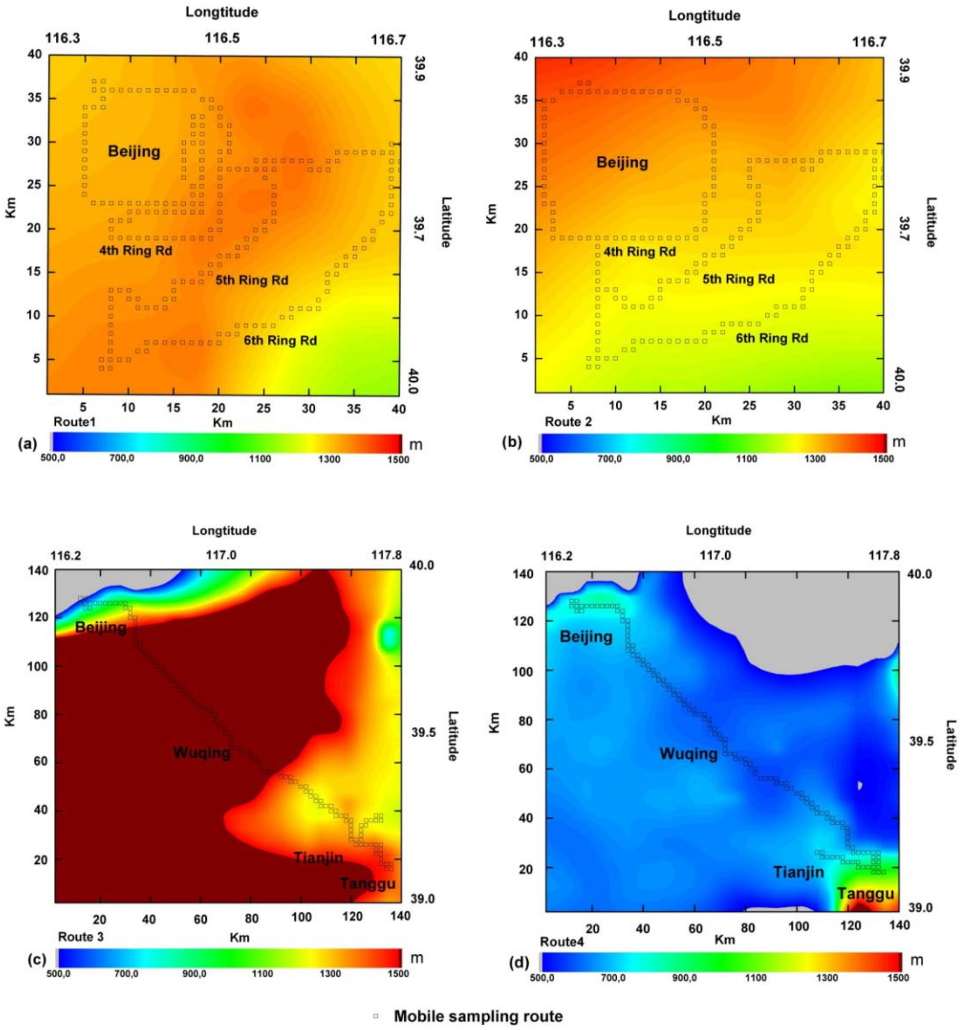

Figure S4. The PBL distributions on August 20, September 3, 4 and 11, 2008 during the measurement period.

Hourly PBL distributions during the measurement period were averaged as shown in Fig. S4. It is clear that the PBL decreased from Beijing to Tianjin on August 20, September 3 and 4 when the southly wind dominated. The height of the mixing boundary layer in Beijing was approximately 1350m and was 1000m in Tianjin area. Whereas high PBL was only observed Tianjin area on September 11 about 1400 m when wind direction from north area. The height of PBL in most of the area was below 1000 m.

S5. The principle of calculating the SO_2 flux using forumla (1)

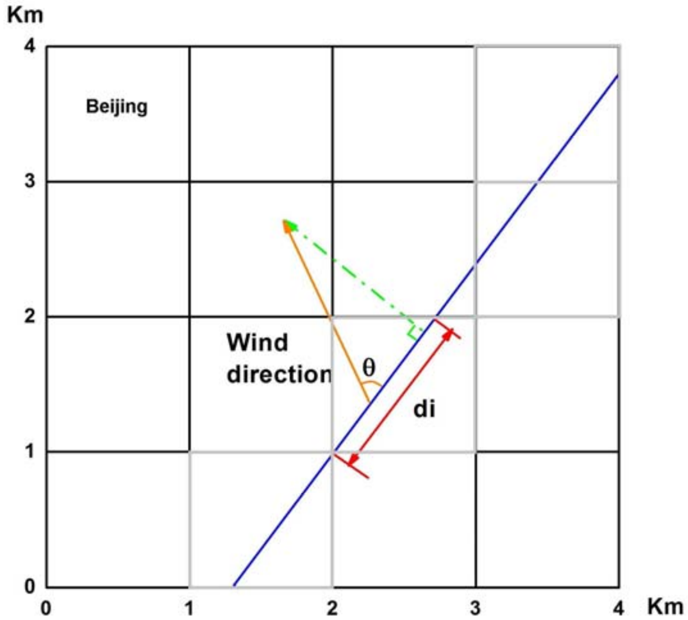

Fig. S5. The principle of calculating the SO_2 flux using formula (1). The blue line shows the segment of mobile measurements routes across the blocks (1×1 km^2, grey color). The angle □ shows the angle between the wind speed and the driving route in the ith cell. The orange arrow line shows the WRF predicted wind vectors in the ith cell. Therefore, the green arrow line shows the wind vectors which are used for the flux calculations. The di indicates the distance of the traverse path within the ith cell.