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Impacts of meteorology and emissions on summertime surface ozone increases over central eastern China between 2003 and 2015

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1. The model simulated monthly-mean MDA8 O_3 in July 2004 and July 2014

To further confirm the conclusions drawn from the comparison between 2003 and 2015, we also conducted model simulations for July 2004 and July 2014. Figure S2 shows the comparison of observed versus simulated monthly-mean surface O_3 levels at six rural sites in July 2004. The model captures well the observed O_3 concentrations at Mt Tai, SDZ, and Mt Hua, with only minor bias (-1.6–4.0 ppbv). In comparison, the model tends to overestimate the O_3 levels at Mt Huang, Lin'an and Cape D'Aguilar. Figure S8 shows similar spatial distributions of MDA8 O_3 over CEC in July 2003, 2004, 2014 and 2015. In July 2004, the regions with MDA8 O_3 >75 ppbv moved to the south of North China Plain compared to July 2003, mostly due to the different atmospheric circulation patterns. The regional mean MDA8 O_3 in July 2004 is 67.8±6.2 ppbv, a little higher than that in July 2003 (65.5±7.9). The regional mean MDA8 O_3 in July 2014 is 74.8±9.8 ppbv, which is comparable to that in July 2015 (74.4±8.7 ppbv). We can find the significant increases of MDA8 O_3 from 2004 to 2014 as well as from 2003 to 2015. The different concentrations and spatial distributions of O_3 between 2003 and 2004 (as well as between 2014 and 2015) should be mostly due to the inter-annual variability in meteorological conditions. Overall, the modelling results in 2004 and 2014 supported the major conclusions derived from 2003 and 2015.

2. The model simulated monthly-mean MDA8 O_3 in August 2003 and August 2015

Figure S9 shows the spatial distributions of MDA8 O_3 in August 2003 and August 2015. In August 2003, there is no region with monthly-mean MDA8 O_3 >75 ppbv. In August 2015, the region with monthly-mean MDA8 O_3 >75 ppbv is comparable to that in July 2015, but the O_3 levels are generally lower than those in July 2015. The regional mean MDA8 O_3 in August 2003 and August 2015 are 63.4±4.9 and 73.8±5.0 ppbv, yielding an increase of 10.4 ppbv. The regional mean MDA8 O_3 during July-August of 2003 and 2015 are 64.5±6.4 and 74.1±6.8 ppbv, giving a comparable increase of 9.6 ppbv from 2003 to 2015. The difference in the regional mean MDA8 O_3 between July and August of 2003 is 2.1 ppbv, a little higher than that between July and August in 2015 (0.6 ppbv). Such levels are much lower than the difference between 2003 and 2015. Overall, the modelling results in August of 2003 and 2015 supported the major conclusions derived from July 2003 and July 2015.

3. Spatial distributions of the modelled O_3 precursors over CEC in July 2003 and 2015

The spatial distributions of the modelled NO_2 and NMVOCs over CEC in July 2003 and 2015 and their differences are shown in Figures S10 and S11, respectively. We found that both NO_2 and NMVOCs had increased significantly over CEC from July 2003 to July 2015. The spatial distribution is in accordance with the emission inventory in Figure S15, which shows high levels of NO_2 and NMVOCs in the eastern CEC and Sichuan basin. Comparing the results of 03E15M-03E03M and 15E03M-03E03M, we can find that the contribution from the emission change (15E03M-03E03M) is much higher than that from the meteorology change (03E15M-03E03M). This is as expected since O_3 precursors are primary pollutants and should be governed by the anthropogenic emissions. Furthermore, the O_3 precursor levels over the eastern CEC increased much higher than the western part. Overall, the modelling results for NO_2 and NMVOCs agree well with the results of O_3 in the main context.

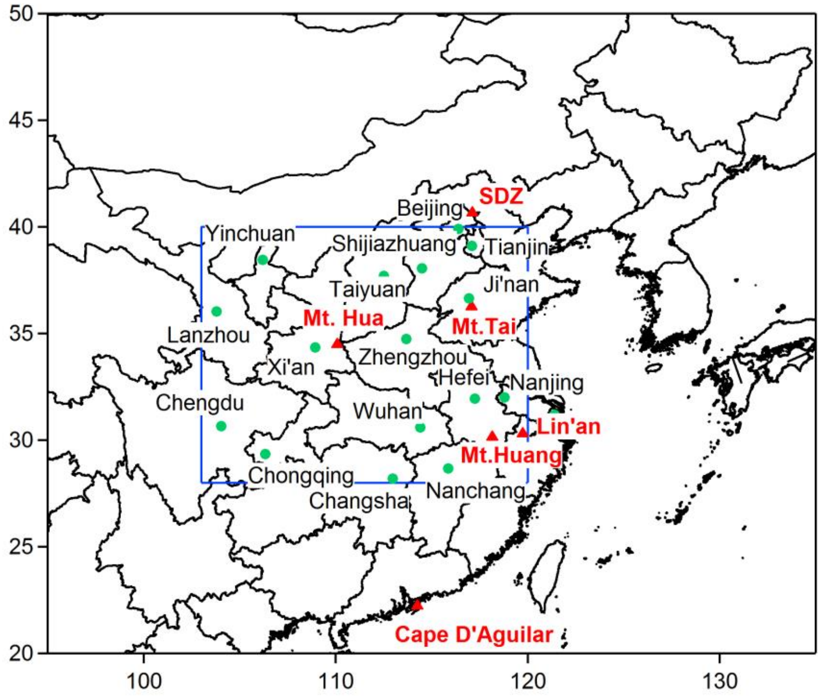

Figure S1. Locations of rural/regional baseline sites (red triangle) and the capital of each province in Central Eastern China (green dot). The blue rectangle represents the Central Eastern China region.

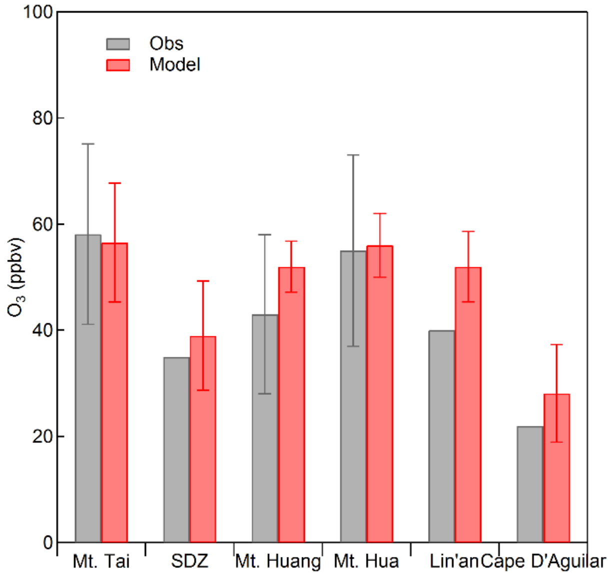

Figure S2. Comparison of observed versus simulated monthly-mean O_3 concentrations at the six rural sites in July 2004.

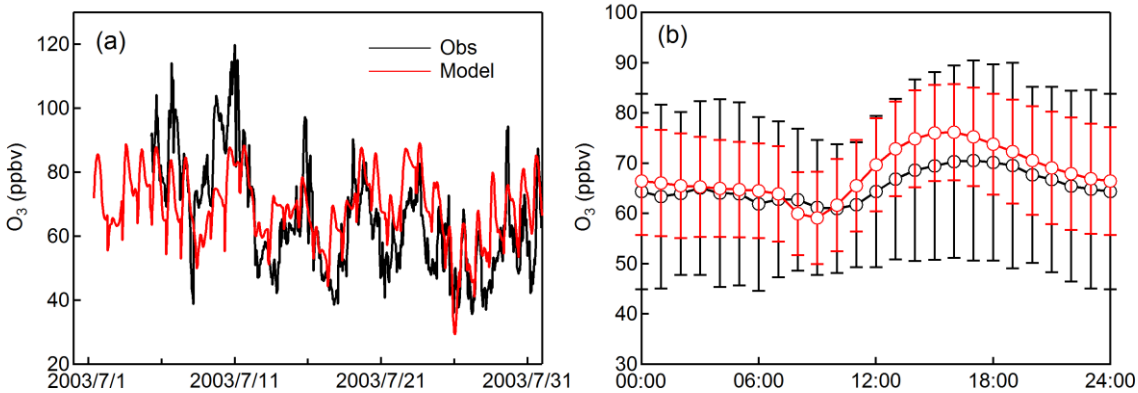

Figure S3. (a) Time series of observed and simulated O_3 concentrations at Mt Tai in July 2003. (b) Diurnal variations of observed and simulated O_3 at Mt Tai in July 2003. Error bars indicate the standard deviation of the mean.

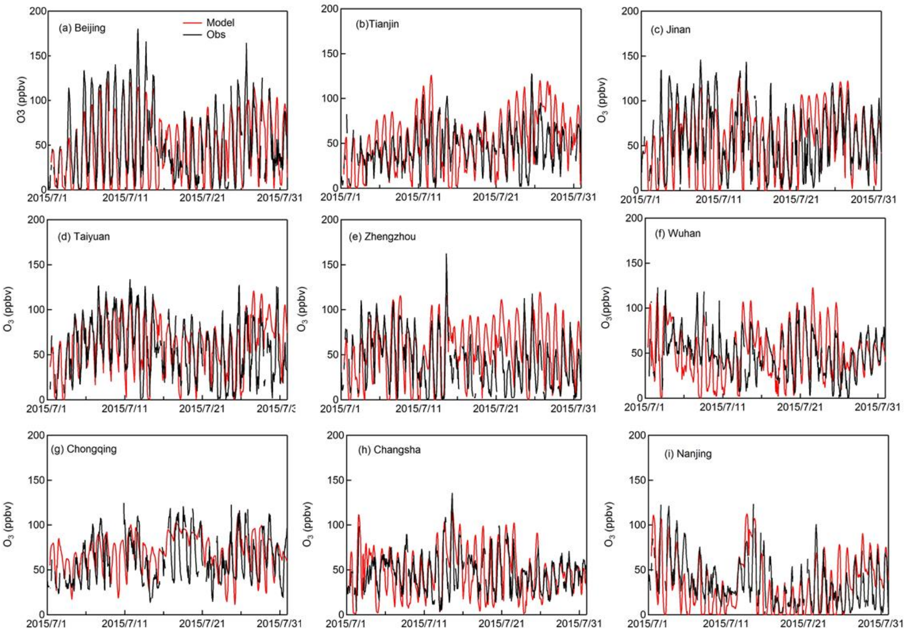

Figure S4. Time series of surface O_3 in July 2015 at nine air quality monitoring stations in different provinces: (a) Dingling in Beijing, (b) Tuanbowa in Tianjin, (c) Kegansuo in Ji'nan, Shandong, (d) Jinsheng in Taiyuan, Shanxi, (e) Ganli Shuiku in Zhengzhou, He'nan, (f) Zhuankou in Wuhan, Hubei, (g) Jinyunshan in Chongqing, (h) Hu'nan Shifan in Changsha, Hu'nan and (i) Xianlin in Nanjing, Jiangsu.

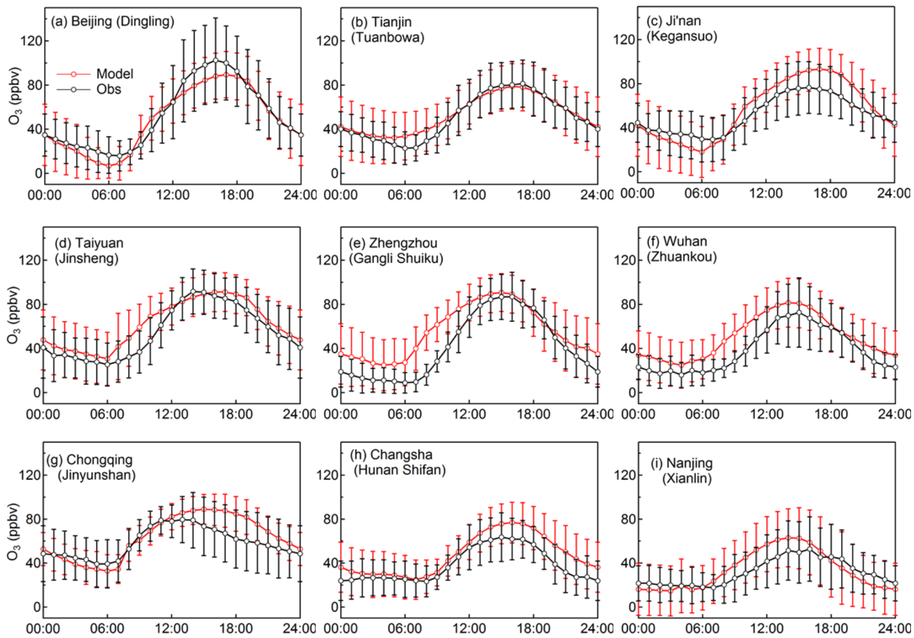

Figure S5. Observed and simulated monthly-mean diurnal variations of surface O_3 in July 2015 at representative air quality monitoring stations in nine cities.

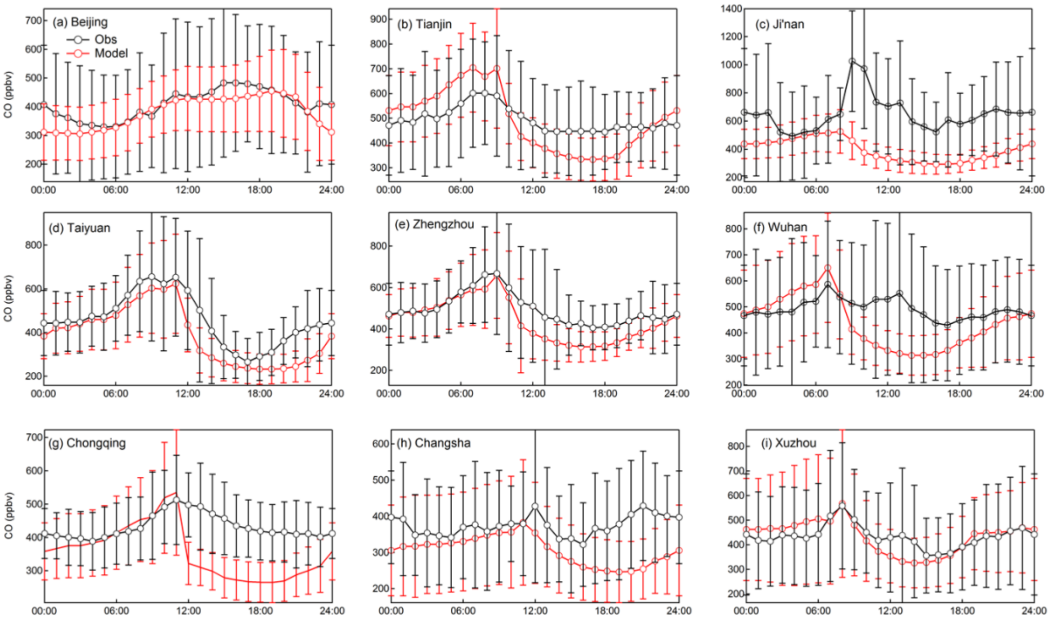

Figure S6. Observed and simulated monthly-mean diurnal variations of surface CO in July 2015 at representative air quality monitoring stations in nine cities.

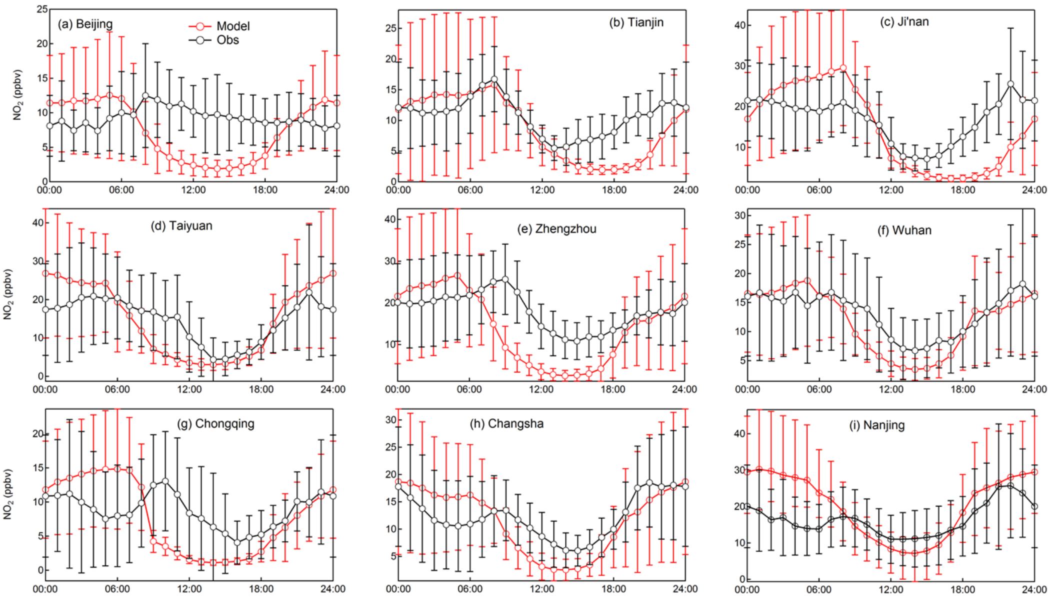

Figure S7. Observed and simulated monthly-mean diurnal variations of surface NO_2 in July 2015 at representative air quality monitoring stations in nine cities.

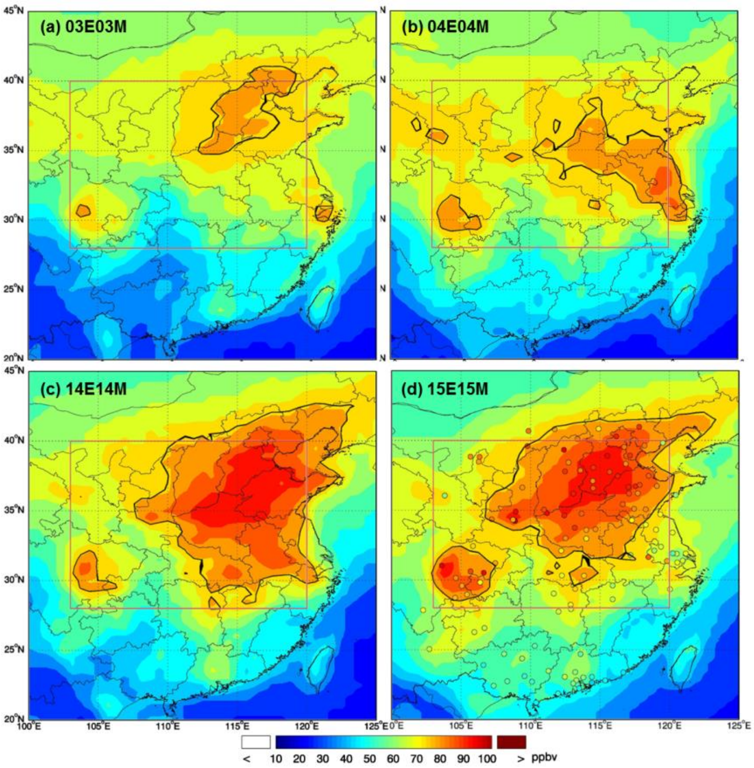

Figure S8. Monthly mean spatial distributions of surface MDA8 O_3 in July over East China. (a) 03E03M: 2003 standard simulation; (b) 04E04M: 2004 standard simulation; (c) 14E14M: 2014 standard simulation and (d) 15E15M: 2015 standard simulation. Black contours indicate the regions with MDA8 O_3 > 75 ppbv. Filled circles in (d) show the observed MDA8 O_3 at 115 sites of the network of Chinese National Environmental Monitoring Center in July 2015. The red rectangle represents the Central Eastern China region (CEC: 103°E-120°E, 28°N-40°N).

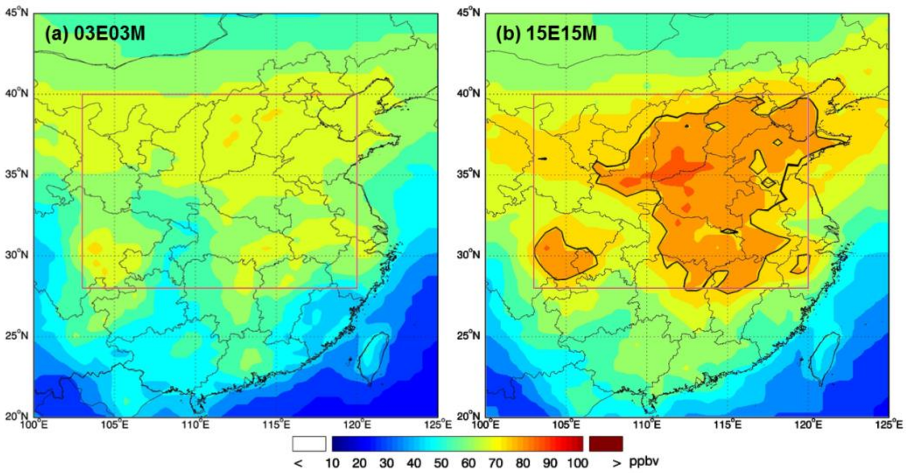

Figure S9. Monthly mean spatial distributions of surface MDA8 O_3 in August over East China. (a) 03E03M: 2003 standard simulation and (b) 15E15M: 2015 standard simulation. Black contours indicate the regions with MDA8 O_3 > 75 ppbv. The red rectangle represents the Central Eastern China region (CEC: 103°E-120°E, 28°N-40°N).

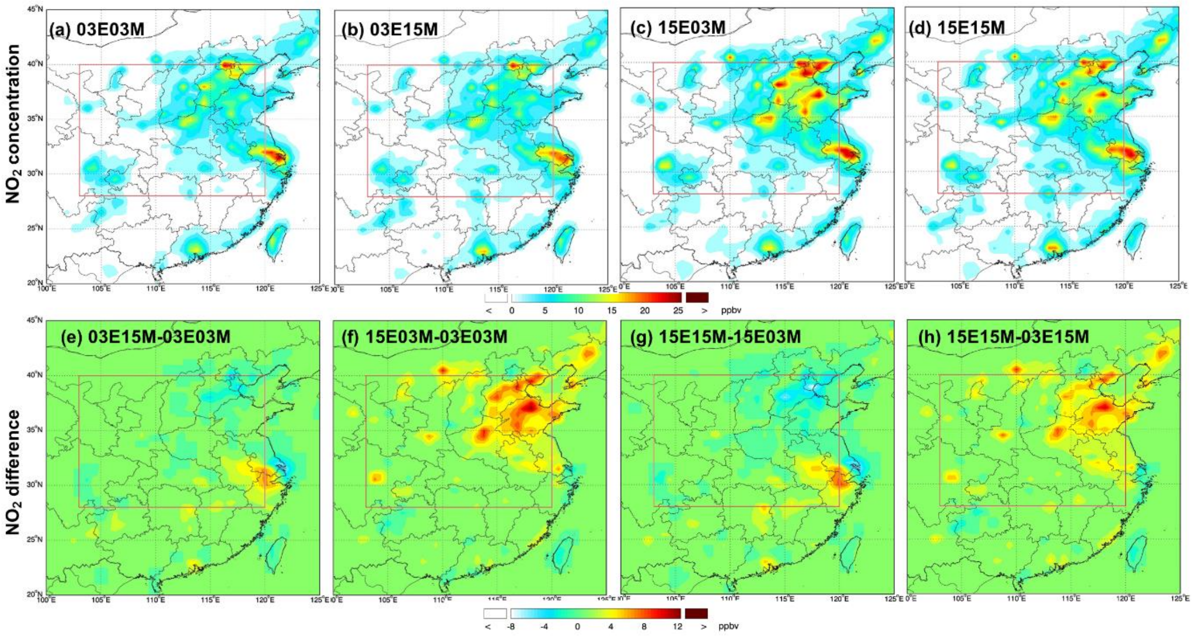

Figure S10. Monthly mean spatial distributions of surface NO_2 in July over CEC: (a)-(d) and the differences in NO_2 concentrations between these simulations: (e)-(h). The red rectangle represents the Central Eastern China region (CEC: 103°E-120°E, 28°N-40°N).

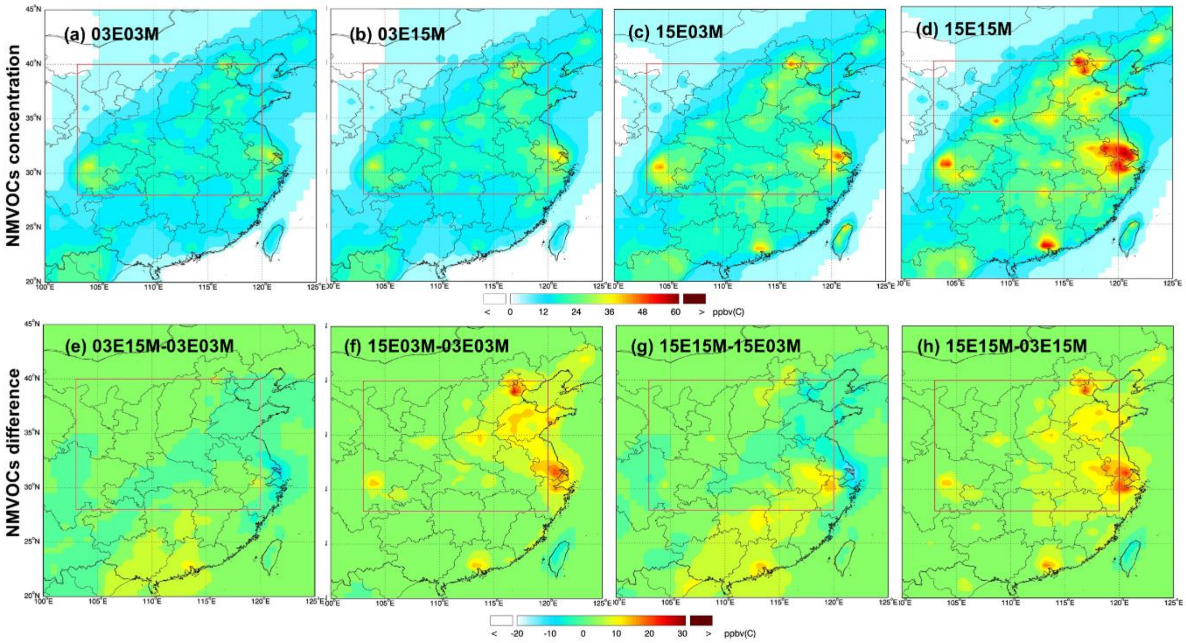

Figure S11. Monthly mean spatial distributions of surface NMVOCs in July over CEC: (a)-(d) and the differences in NMVOC concentrations between these simulations: (e)-(h). The concentrations of NMVOCs include: ALK4 (lumped >=C4 Alkanes), Isoprene, Acetone, Methyl Ethyl Ketone, Acetaldehyde, RCHO (lumped Aldehyde >=C3), PRPE (lumped >=C3 Alkenes), Formaldehyde, Hydroxyacetone and Glycoaldehyde. The red rectangle represents the Central Eastern China region (CEC: 103°E-120°E, 28°N-40°N).

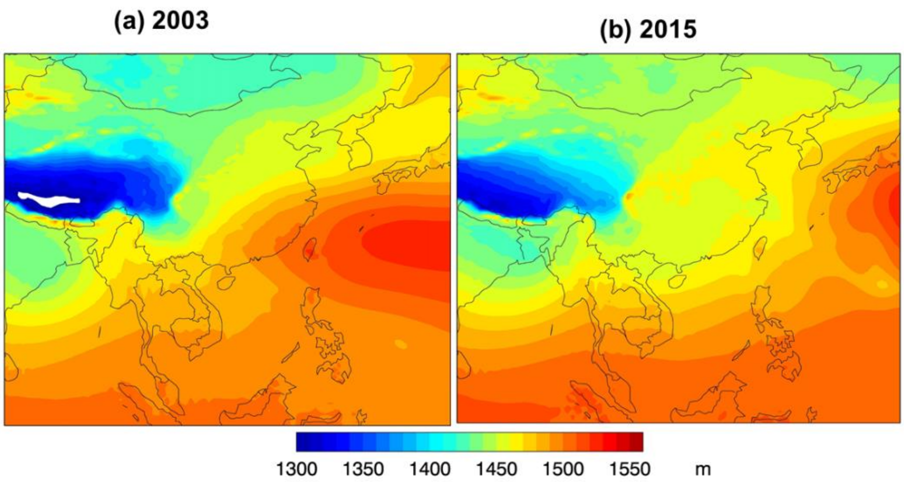

Figure S12. Monthly mean geopotential heights at 850 hPa in July of 2003 and 2015 from MERRA-2 meteorological field.

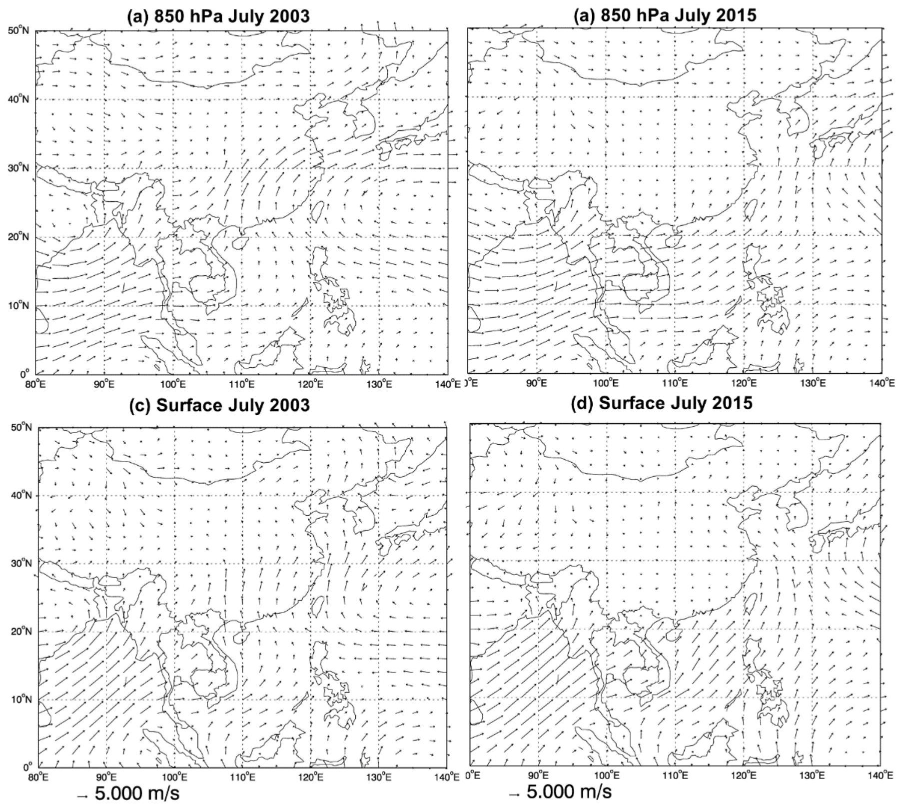

Figure S13. Monthly mean wind fields at 850 hPa and at surface in July of 2003 and 2015 based on the MERRA-2 meteorological field.

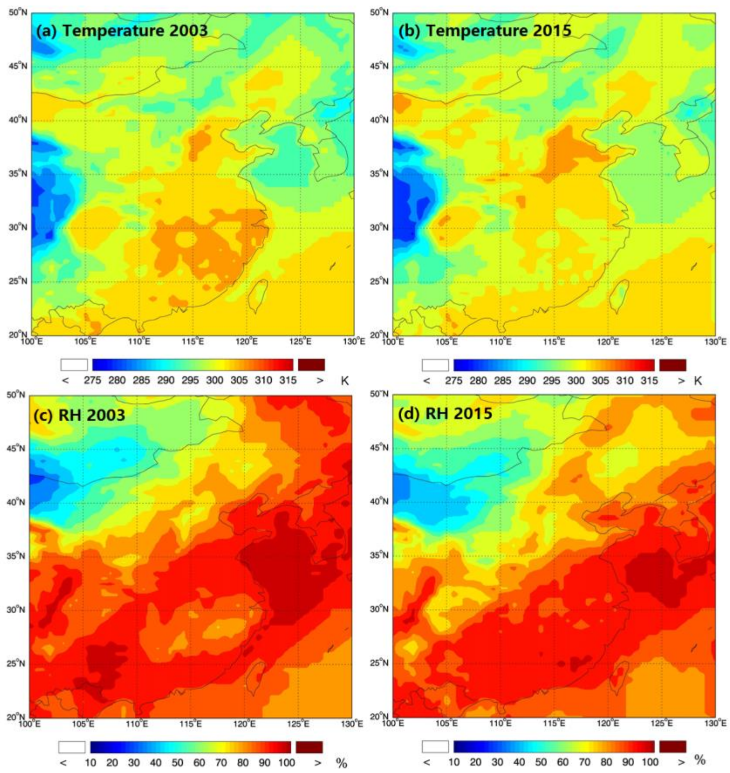

Figure S14. Monthly mean temperature and relative humidity on surface layer in July of 2003 and 2015 based on the MERRA-2 meteorological field.

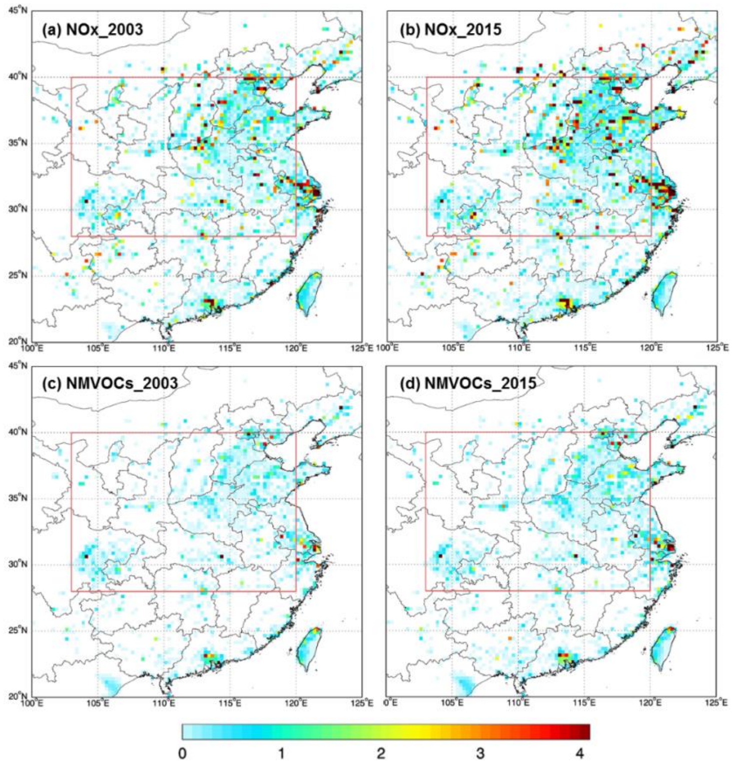

Figure S15. Anthropogenic emissions of NOx in (a) July 2003 and (b) July 2015 and NMVOCs in (c) July 2003 and (d) July 2015. Unit: Mg/Km^2/mon for NOx; Mg(C)/Km^2/mon for NMVOCs. The red rectangle represents the Central Eastern China region. The emission data in 2003 are taken from MEIC (2008) and scaled to 2003 by using annual scale factors from REAS-v2. The emission data in 2015 are taken from MEIC (2014) directly.

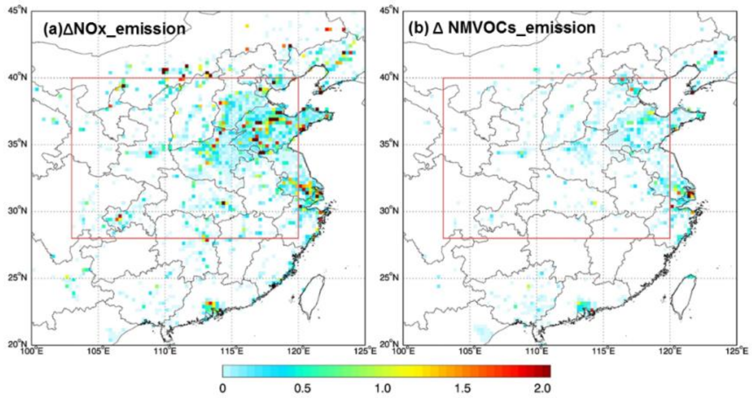

Figure S16. Spatial distributions of changes in NOx (a) and NMVOCs (b) emissions between July 2003 and July 2015 (2015-2003). Unit: Mg/Km^2/mon for NOx; Mg(C)/Km^2/mon for NMVOCs. The red rectangle represents the Central Eastern China region. The emission data in 2003 are taken from MEIC (2008) and scaled to 2003 by using annual scale factors from REAS-v2. The emission data in 2015 are taken from MEIC (2014) directly.