Supplement of Insights into particulate matter pollution in the North China Plain during wintertime: local contribution or regional transport?

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The supplement provides description about model description, model evaluation, and the calculation of average wind direction during the study episode.

24 Section S1 Parameterization of the heterogeneous oxidation of SO$_2$ involving aerosol water

The SO$_2$ heterogeneous reaction parameterization is used in this study in which the SO$_2$ oxidation in aerosol water by O$_2$ catalyzed by Fe$^{3+}$ is limited by mass transfer resistances in the gas-phase and the gas-particle interface.

$$ S(IV) + \frac{1}{2} O_2 + Fe^{3+} \rightarrow S(VI) $$

When the solution pH is between 5.0 and 7.0, the oxidation reaction is second order in dissolved iron and first order in S(IV) and can be expressed as follows (Seinfeld and Pandis, 2006):

$$ -\frac{d[S(IV)]}{dt} = 1 \times 10^{-3} [S(IV)] \quad 5.0 < pH < 6.0 $$

$$ -\frac{d[S(IV)]}{dt} = 1 \times 10^{-4} [S(IV)] \quad pH \sim 7.0 $$

where [S(IV)] is the sulfite (S(IV)) concentration. The measured SO$_2$ mass accommodation coefficient on aqueous surfaces is around 0.1 (Worsnop et al., 1989). Due to sufficient NH$_3$ and presence of mineral dust in the atmosphere in northern China, the calculated pH in aerosol water is between 5.0 and 7.0 (Cao et al., 2013). The SO$_2$ uptake coefficient on aerosol water surface is estimated to be about $10^4$–$10^5$ if the sulfite oxidation is catalyzed by Fe$^{3+}$. The sulfate heterogeneous formation from SO$_2$ is therefore parameterized as a first-order irreversible uptake by aerosols, with a reactive uptake coefficient of $0.5 \times 10^{-4}$, assuming that there is enough alkalinity to maintain the high iron-catalyzed reaction rate:
\[ -\frac{d[SO_2]}{dt} = -\left(\frac{1}{4} \gamma_{SO_2} v_{SO_2} A_W\right)[SO_2] \]

where \([SO_2]\) is the SO\(_2\) concentration, \(A_W\) is the aerosol water surface area, \(\gamma_{SO_2}\) is the SO\(_2\) reactive uptake coefficient, and \(v_{SO_2}\) is the SO\(_2\) thermal velocity. The aerosol hygroscopic growth is directly predicted by ISORROPIA (Version 1.7) in the model, and the aerosol water surface area is scaled from the calculated wet aerosol surface area using the third-moment of aerosol species.

**Section S2 Model Evaluation**

**Section S2.1 Air pollutants simulations in different cities in the NCP**

Considering that there are many monitoring sites in the NCP, scatter plots of observed and simulated PM\(_{2.5}\), O\(_3\), NO\(_2\), SO\(_2\) and CO concentrations for all sites in Beijing, Tianjin, Hebei, Henan, Shandong, Shanxi, Jiangsu, and Anhui from 05 December 2015 to 04 January 2016 have also been provided in Figures S4 to S8, respectively. Except Anhui, the correlation coefficients between observed and simulated PM\(_{2.5}\) concentrations are generally larger than 0.70 (Figure S4). The model also performs well in simulating the O\(_3\) concentration in the NCP, with correlation coefficients generally larger than 0.80 (Figure S5). The NO\(_2\) concentration in the NCP is also simulated reasonably, with correlation coefficients generally ranging from 0.70 to 0.80 (Figure S6). Considering that the SO\(_2\) is mainly emitted from point sources, which is more sensitive to meteorological conditions, the model has difficulties in simulating the SO\(_2\) concentration, with correlation coefficients generally less than 0.60 (Figure S7). In addition to Tianjin and Shanxi, the CO concentration is also reasonably reproduced, with correlation coefficient larger than 0.70 (Figure S8).

**Section S2.2 Cloud properties**

Clouds are one of the most important factors affecting the solar radiation reaching the ground. The daily cloud fraction (CF) used in this study was retrieved from Terra- and Aqua-Moderate Resolution Imaging Spectroradiometer (MODIS) level 2 products. Figure S13
presents the scatter plot of the daily retrieved and simulated CF averaged in the NCP from 05 December 2015 to 31 December 2015. Generally, the simulated daily average CF correlates well with that retrieved, with a correlation coefficient of 0.69. The simulated average CF over the NCP during the episode is 52.8%, lower than the MODIS retrieved 78.4%. Numerical models still have difficulties in representing accurately clouds in terms of microphysical processes, cloud morphologies, occurrence and dissipation. In addition, many uncertainties also significantly impact CF retrievals, such as the satellite’s view zenith angle, cloud microphysics assumptions, namely cloud phase, particle size and shape, et al. (An and Wang, 2015; Platnick et al., 2017; Zeng et al., 2012; Li et al., 2014). Therefore, it is still difficult to validate cloud simulations using the satellite cloud products.

Section S3 Calculation of the average wind direction

The wind direction simulated in this study is calculated using the U (the velocity toward east) and V (the velocity toward north) component at a specific grid point over the simulation domain and the average wind direction is calculated based on the average U and V.
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Table S1 SOA yield scenarios using a four-product basis set with saturation concentrations of 1, 10, 100, and 1000 µg m$^{-3}$ at 298 K.

| SOA Precursors | Aerosol Yield* | Aerosol Yield | Molecular Weight |
|----------------|----------------|---------------|------------------|
|                | Low-NOx Parameterization | High-NOx Parameterization | (g mol$^{-1}$) |
|                | 1   | 10  | 100 | 1000 | 1   | 10  | 100 | 1000 |                 |
| ALK4           | 0.000 | 0.038 | 0.000 | 0.000 | 0.000 | 0.075 | 0.000 | 0.000 | 120 |
| ALK5           | 0.000 | 0.150 | 0.000 | 0.000 | 0.000 | 0.300 | 0.000 | 0.000 | 150 |
| OLE1           | 0.001 | 0.005 | 0.038 | 0.150 | 0.005 | 0.009 | 0.060 | 0.225 | 120 |
| OLE2           | 0.003 | 0.026 | 0.083 | 0.270 | 0.023 | 0.044 | 0.129 | 0.375 | 120 |
| ARO1           | 0.003 | 0.165 | 0.300 | 0.435 | 0.075 | 0.225 | 0.375 | 0.525 | 150 |
| ARO2           | 0.002 | 0.195 | 0.300 | 0.435 | 0.075 | 0.300 | 0.375 | 0.525 | 150 |
| CRES           | -   | -   | -   | -   | -   | -   | -   | -   | -   |
| ISOP           | 0.001 | 0.023 | 0.015 | 0.000 | 0.009 | 0.030 | 0.015 | 0.000 | 136 |
| TERP           | 0.012 | 0.122 | 0.201 | 0.500 | 0.107 | 0.092 | 0.359 | 0.600 | 180 |

*The SOA yields are based on an assumed density of 1.5 g cm$^{-3}$. 
Table S2 Parameters used to treat partitioning of POA emissions.

|                         | 0.01 | 0.1 | 1   | 10  | 10^2 | 10^3 | 10^4 | 10^5 | 10^6 |
|-------------------------|------|-----|-----|-----|------|------|------|------|------|
| **C** at 298K (µg m^-3) |      |     |     |     |      |      |      |      |      |
| Fraction of emissions   | 0.03 | 0.06| 0.09| 0.14| 0.18 | 0.30 | 0.40 | 0.50 | 0.80 |
| Emission Phase (Particle:P;Gas:G) | P | P | P | P | P | G | G | G |
| Molecular Weight (g mol^-1) | 250 | 250 | 250 | 250 | 250 | 250 | 250 | 250 | 250 |
| ΔH (kJ mol^-1) (Robinson et al., 2007) | 112 | 106 | 100 | 94  | 88  | 82  | 76  | 70  | 64  |
| ΔH (kJ mol^-1) (Grieshop et al., 2009) | 77  | 73  | 69  | 65  | 61  | 57  | 54  | 50  | 46  |
Supplement Figure Captions

Figure S1 Model simulation domain and designation of source regions. The black filled circles with number denote the meteorological sites used in this study in the NCP. 1: Beijing; 2: Tianjin; 3: Shijiazhuang; 4: Jinan; 5: Zhengzhou; 6: Hefei; 7: Nanjing.

Figure S2 (a) Geopotential heights and (b) the mean sea level pressures with wind vectors during the study episode from 05 December 2015 to 04 January 2016.

Figure S3 Comparison of observed (black dots) and simulated (solid red lines) diurnal profiles of near-surface (a) pressure, (b) temperature, (c) wind speed, and (d) wind direction averaged at monitoring sites in the NCP from 05 December 2015 to 04 January 2016.

Figure S4 Scatter plot of hourly simulated and observed PM$_{2.5}$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.

Figure S5 Scatter plot of hourly simulated and observed O$_3$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.

Figure S6 Scatter plot of hourly simulated and observed NO$_2$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.

Figure S7 Scatter plot of hourly simulated and observed SO$_2$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.

Figure S8 Scatter plot of hourly simulated and observed CO concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.

Figure S9 Pattern comparisons of simulated (color contours) vs. observed (colored circles) near-surface mass concentrations of PM$_{2.5}$ at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016. The black arrows indicate simulated surface winds.

Figure S10 Pattern comparisons of simulated (color contours) vs. observed (colored circles) near-surface mass concentrations of O$_3$ at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016. The black arrows indicate simulated surface winds.

Figure S11 Pattern comparisons of simulated (color contours) vs. observed (colored circles) near-surface mass concentrations of NO$_2$ at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016. The black arrows indicate simulated surface winds.

Figure S12 Pattern comparisons of simulated (color contours) vs. observed (colored circles) near-surface mass concentrations of SO$_2$ at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016. The black arrows indicate simulated surface winds.

Figure S13 Scatter plot of the MODIS retrieved and simulated daily cloud fraction averaged
in the NCP from 05 December 2015 to 31 December 2015.

Figure S14 Spatial distribution of average PM$_{2.5}$ contributions from Beijing at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016.

Figure S15 Spatial distribution of average PM$_{2.5}$ contributions from Tianjin at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016.

Figure S16 Spatial distribution of average PM$_{2.5}$ contributions from Hebei at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016.

Figure S17 Spatial distribution of average PM$_{2.5}$ contributions from Henan at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016.

Figure S18 Spatial distribution of average PM$_{2.5}$ contributions from Shandong at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016.

Figure S19 Spatial distribution of average PM$_{2.5}$ contributions from Shanxi at (a) 00:00 BJT, (b) 04:00 BJT, (c) 08:00 BJT, (d) 12:00 BJT, (e) 16:00 BJT, and (e) 20:00 BJT averaged from 05 December 2015 to 04 January 2016.

Figure S20 Vertical profiles of average PM$_{2.5}$ contribution from local (red line) and non-local (black line) emissions in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, and (f) Shanxi from 05 December 2015 to 04 January 2016.

Figure S21 Spatial distribution of average near-surface PM$_{2.5}$ contributions from (a) residential, (b) transportation, (c) industry, (d) power, and (e) agriculture emission sectors from 05 December 2015 to 04 January 2016.

Figure S22 Average (a) decrease of surface temperature (TSFC), (b) increase of relative humidity (RH), and (c) percentage decrease of PBLH caused by the BC transported from the south of 32°N, as a function of the near-surface PM$_{2.5}$ concentration in the NCP during daytime from 05 December 2015 to 04 January 2016.
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Figure S5 Scatter plot of hourly simulated and observed O$_3$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.
Figure S6 Scatter plot of hourly simulated and observed NO$_2$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.
Figure S7: Scatter plot of hourly simulated and observed SO$_2$ concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.
Figure S8 Scatter plot of hourly simulated and observed CO concentration in (a) Beijing, (b) Tianjin, (c) Hebei, (d) Henan, (e) Shandong, (f) Shanxi, (g) Anhui, and (h) Jiangsu during the episode from 05 December 2015 to 04 January 2016.
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