Supplement of Formation of Nighttime Sulfuric Acid from the Ozonolysis of Alkenes in Beijing

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Figures

Fig. S1. Overview of different parameters measured between 18th January and 16th March 2019 for (a) sulfuric acid (SA) concentration and particle number concentration of sub-3nm particles (N_{sub-3nm}, measured by PSM), (b) condensation sink (CS) and SO$_2$ concentration, (c) concentration of O$_3$ and Alkenes, (d) PM$_{2.5}$ and visibility, and (e) relative humidity (RH) and temperature. The light blue bars represent nighttime SA event cases.

Fig. S2. Boxplot for SO$_2$ mixing ratio during nighttime in winter-heating-supply period (2019.01.18~2019.03.15) and non-heating-supply period (2019.03.16~2019.05.31). The middle line in the box is the median, the bottom and the top are the 25 and 75 percentiles, the whiskers are the 5 and 95 percentiles and the red points are the outliers. The dark gray value on the top is the ratio between median SO$_2$ values of two periods.
Fig. S3. Daily time-series of different parameters on nighttime SA event days when SA cases occurred under SO\textsubscript{2} increase conditions. The first row: \(N\textsubscript{sub-3nm}\) and SA concentration, the second row: CS and SO\textsubscript{2} concentration, and the third row: concentration of O\textsubscript{3} and Alkene. The increase starting points and maximum value points of SA concentration as well as the corresponding SO\textsubscript{2} concentration at the same moments is marked by cyan dots.

Fig. S4. Nighttime correlation between PM\textsubscript{2.5} and visibility colored by RH and sized by CS*1.0\times10\textsuperscript{4}. Note that the data points are based on data averaged and binned into different visibility ranges instead of the original, high time resolution data. And the error bars are the standard deviation of all data points in each bin.

Cleanliness is kind of an ambiguous concept, and one may judge cleanliness by PM\textsubscript{2.5} while another may judge by visibility or NO\textsubscript{x}. Therefore, we did some efforts to determine the final parameter used to represent cleanliness. Fig.S5 shows the correlation of PM\textsubscript{2.5}, CS, RH with visibility. It can be seen that with the increase of visibility, PM\textsubscript{2.5} decreases monotonically, and RH and CS also have a declining trend. Hence, visibility is a good candidate to represent cleanliness. Besides, in the visibility range of 12.0 km to 19.0 km, with the increase of visibility, PM\textsubscript{2.5} and RH do not vary too much, with CS slightly declining as well, which also implies that visibility is more sensitive than PM\textsubscript{2.5}, RH and CS. Thus, visibility indeed can be used to judge the cleanliness for this specific time period of this work.

It also can be found out that the correlation between PM\textsubscript{2.5} and visibility can be further divided into the following 3 groups: a. visibility < 4.0 km (heavy polluted conditions): visibility and PM\textsubscript{2.5} have a very good negative linear correlation...
with $R_1$ (correlation coefficient) = -1.000, and the decrease rate of visibility is rather fast with the slope of $k_1 = -0.0339 \, \mu g/m^4$; b. $4.0 \, km \leqslant visibility < 12.0 \, km$ (mildly polluted conditions): visibility and PM$_{2.5}$ also have a negative linear correlation with $R_2 = -0.9688$, but the decrease rate of visibility with PM$_{2.5}$ reduces to $k_2 = -0.0084 \, \mu g/m^4$; c. visibility $\geqslant 12.0 \, km$ (clean conditions): PM$_{2.5}$ stays constant with varying visibility values, which means that when PM$_{2.5}$ is smaller than 40 $\mu g/m^3$ during heating supply winter period, visibility will be more likely influenced by other factors. In total, data points under the clean conditions mentioned above take up 47.91% of all data points.

Fig. S5. Nighttime correlation between the source term ([SO$_2$·[O$_3$]·[Alkene]]) and sink term ([SA]·[CS+β]·[SA]$^2$) of SA under pseudo-steady-state for (a) with all data points divided by [O$_3$] and visibility, (b) and (c) with data points having visibility larger than 12.0 km divided by [O$_3$] and colored by NO$_x$ and NO respectively.
Fig. S6. Nighttime correlation between the source term ([SO$_2$]·[O$_3$]·[Alkene]) and sink term ([SA]·CS+β·[SA]$^2$) of SA under pseudo-steady-state for (a) Clean-1 condition, (b) Clean-2 condition, (c) mildly polluted condition and (d) heavy polluted condition.

Fig. S7. Nighttime correlation between the source term ([SO$_2$]·[O$_3$]·[Alkene]) and sink term ([SA]·CS+β·[SA]$^2$) under clean conditions for (a) during 2019/01/18-2019/03/15 and (b) during 2019/03/20-2019/05/20. The gray dots are original, high time resolution data, and the diamond points are based on data median averaged and binned to different source ranges instead of the original, high time resolution data. The error bars are the standard deviation of all data points in each bin.
### Table S1 Dates of Nighttime SA event and non-event days.

| Nighttime SA Event Day | Nighttime SA Non-event Day |
|------------------------|-----------------------------|
| 2019.01.20             | 2019.01.24                  |
| 2019.01.21             | 2019.01.30                  |
| 2019.01.22             | 2019.02.03                  |
| 2019.01.23             | 2019.02.06                  |
| 2019.01.25             | 2019.02.08                  |
| 2019.01.28             | 2019.02.11                  |
| 2019.02.01             | 2019.02.13                  |
| 2019.02.04             | 2019.02.21                  |
| 2019.02.12             | 2019.02.22                  |
| 2019.02.15             | 2019.02.27                  |
| 2019.02.17             | 2019.03.04                  |
| 2019.02.20             | 2019.03.06                  |
| 2019.02.25             | 2019.03.07                  |
| 2019.02.26             | 2019.03.09                  |
| 2019.02.28             | 2019.03.10                  |
| 2019.03.12             | 2019.03.11                  |
| 2019.03.14             |                             |
| 2019.03.15             |                             |

### Table S2 Features of Nighttime SA Event Cases on 18 Event Nights.

| Date       | Number of Nighttime SA Case | CS Decrease Case | SO\textsubscript{2} Increase Case | Other Cases       |
|------------|----------------------------|------------------|----------------------------------|-------------------|
| 2019.01.20 | 1                          |                  |                                  |                   |
| 2019.01.21 | 1                          |                  |                                  |                   |
| 2019.01.22 | 1                          |                  |                                  |                   |
| 2019.01.23 | 1                          |                  |                                  |                   |
| 2019.01.25 | 1                          |                  |                                  |                   |
| 2019.01.28 | 1                          |                  |                                  |                   |
| 2019.02.01 | 1                          |                  |                                  |                   |
| 2019.02.04 | 1                          |                  |                                  |                   |
| 2019.02.12 | 2                          | 1                |                                  |                   |
| 2019.02.15 | 1                          | 1                |                                  |                   |
| 2019.02.17 | 1                          |                  |                                  |                   |
| 2019.02.20 | 1                          |                  |                                  | No CS data        |
| 2019.02.25 | 1                          |                  |                                  |                   |
| 2019.02.26 | 2                          |                  |                                  |                   |
| 2019.02.28 | 1                          |                  |                                  | No CS data        |
| 2019.03.12 | 1                          |                  |                                  | No CS data        |
| 2019.03.14 | 1                          |                  |                                  | No CS data        |
| 2019.03.15 | 1                          |                  |                                  | No CS data        |

Total 15 with CS data (20 in total) 8 (53.33\%*) 4 (26.67\%*) 3 (20.00\%*)

* There are 5 days when CS data is not available and the statistical percentages in the brackets are based on the CS available cases.