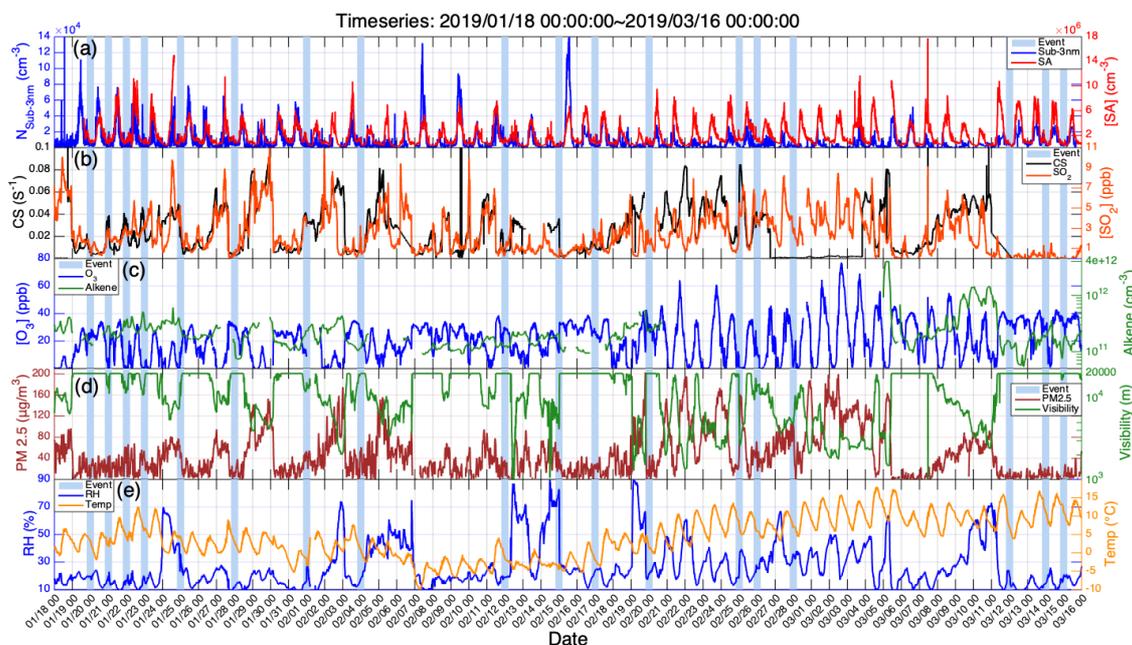


Supplement of Formation of Nighttime Sulfuric Acid from the Ozonolysis of Alkenes in Beijing

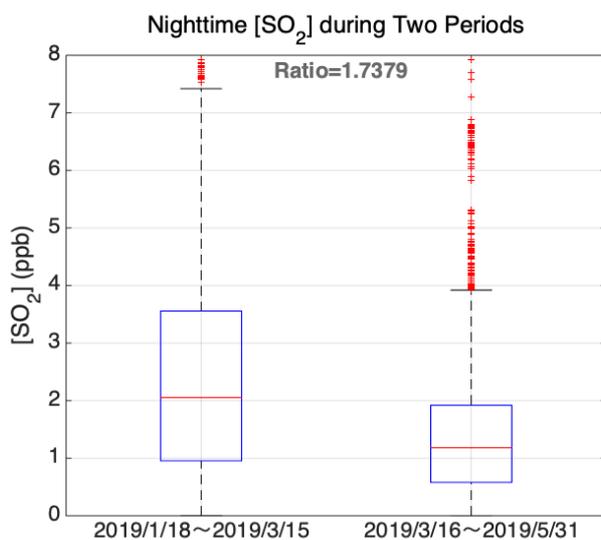
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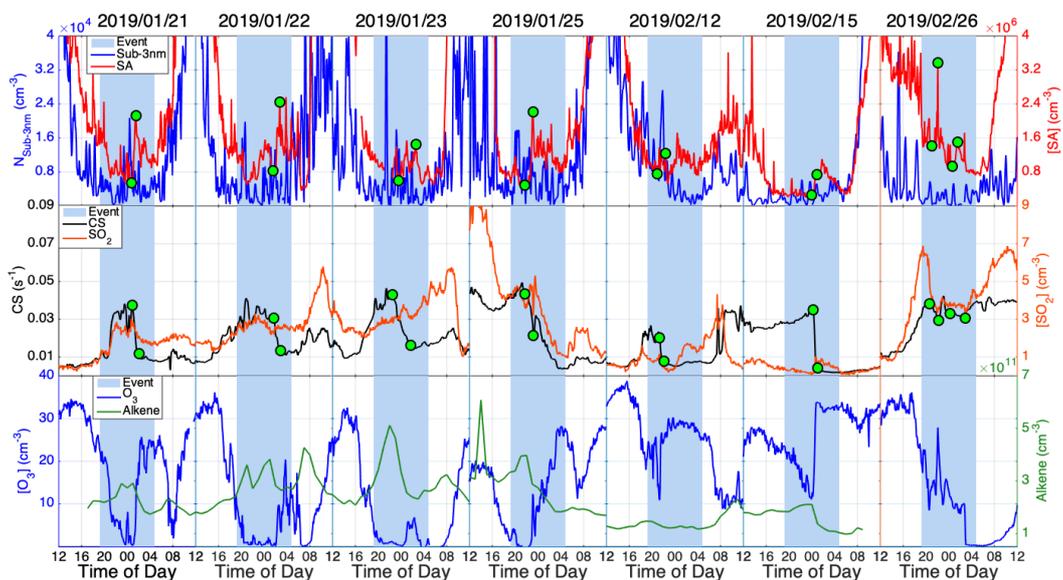
Figures



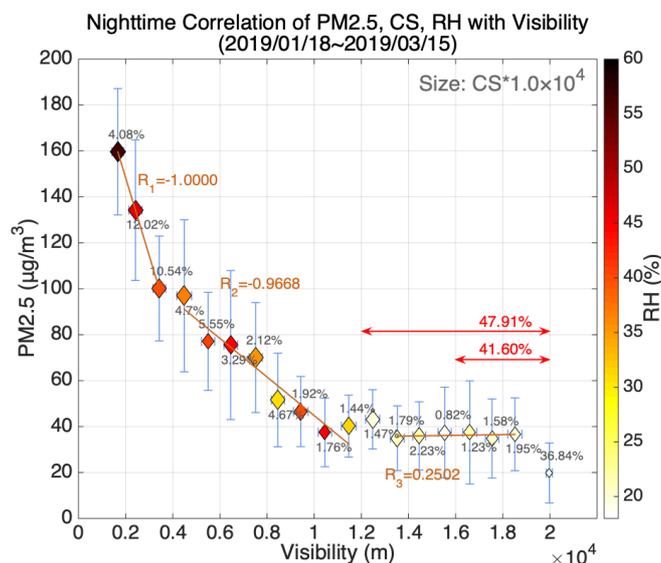
10 **Fig. S1.** Overview of different parameters measured between 18th January and 16th March 2019 for (a) sulfuric acid (SA) concentration and particle number concentration of sub-3-nm particles ($N_{\text{Sub-3nm}}$, measured by PSM), (b) condensation sink (CS) and SO_2 concentration, (c) concentration of O_3 and Alkenes, (d) $\text{PM}_{2.5}$ and visibility, and (e) relative humidity (RH) and temperature. The light blue bars represent nighttime SA event cases.



15 **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.



20 **Fig. S3.** Daily time-series of different parameters on nighttime SA event days when SA cases occurred under SO₂ increase conditions. The first row: N_{sub-3nm} and SA concentration, the second row: CS and SO₂ concentration, and the third row: concentration of O₃ and Alkene. The increase starting points and maximum value points of SA concentration as well as the corresponding SO₂ concentration at the same moments is marked by cyan dots.



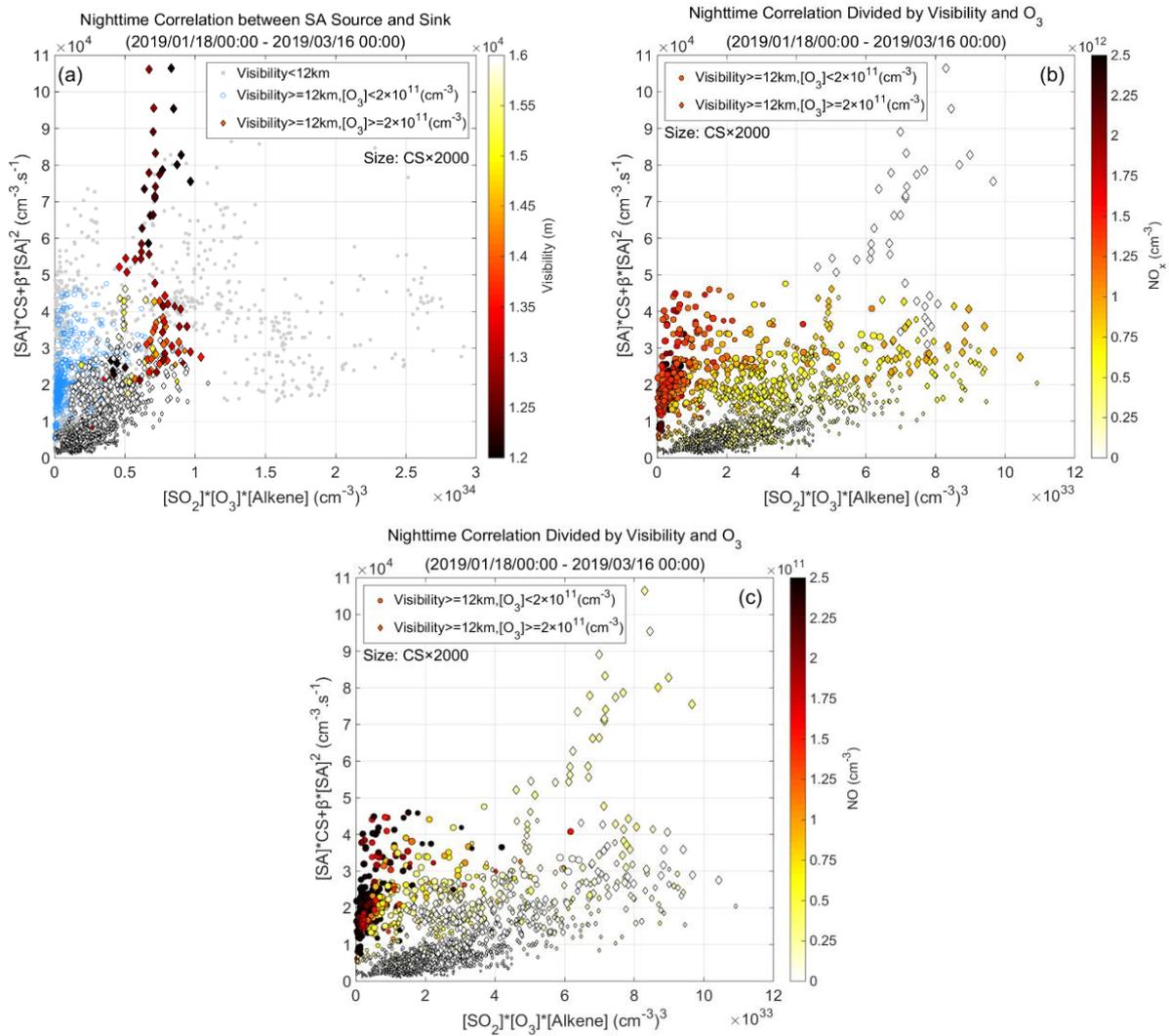
25 **Fig. S4.** Nighttime correlation between PM_{2.5} and visibility colored by RH and sized by CS*1.0×10⁴. Note that the data points are based on data averaged and binned into different visibility ranges instead of the original, high time resolution data. And the error bars are the standard deviation of all data points in each bin.

Cleanliness is kind of an ambiguous concept, and one may judge cleanliness by PM_{2.5} while another may judge by visibility or NO_x. Therefore, we did some efforts to determine the final parameter used to represent cleanliness. Fig.S5 shows the correlation of PM_{2.5}, CS, RH with visibility. It can be seen that with the increase of visibility, PM_{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_{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_{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_{2.5} and visibility can be further divided into the following 3 groups: a. visibility < 4.0 km (heavy polluted conditions): visibility and PM_{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 $k_1 = -0.0339$ $\mu\text{g}/\text{m}^4$; b. $4.0 \text{ km} \leq \text{visibility} < 12.0 \text{ km}$ (mildly polluted conditions): visibility and $\text{PM}_{2.5}$ also have a negative linear correlation with $R_2 = -0.9688$, but the decrease rate of visibility with $\text{PM}_{2.5}$ reduces to $k_2 = -0.0084$ $\mu\text{g}/\text{m}^4$; c. visibility $\geq 12.0 \text{ km}$ (clean conditions): $\text{PM}_{2.5}$ stays constant with varying visibility values, which means that when $\text{PM}_{2.5}$ is smaller than $40 \mu\text{g}/\text{m}^3$ 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.

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Fig. S5. Nighttime correlation between the source term ($[\text{SO}_2] \cdot [\text{O}_3] \cdot [\text{Alkene}]$) and sink term ($[\text{SA}] \cdot \text{CS} + \beta \cdot [\text{SA}]^2$) of SA under pseudo-steady-state for (a) with all data points divided by $[\text{O}_3]$ and visibility, (b) and (c) with data points having visibility larger than 12.0 km divided by $[\text{O}_3]$ and colored by NO_x and NO respectively.

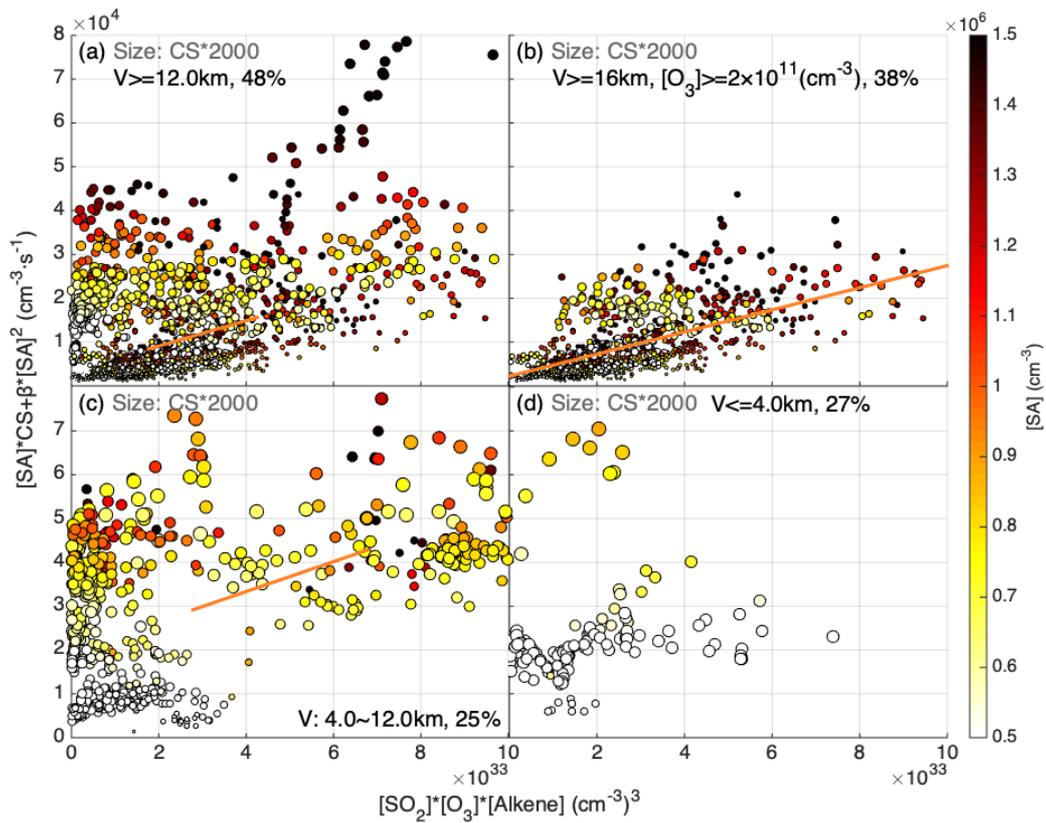


Fig. S6. Nighttime correlation between the source term ($[\text{SO}_2] [\text{O}_3] [\text{Alkene}]$) and sink term ($[\text{SA}] \text{CS} + \beta [\text{SA}]^2$) of SA under pseudo-steady-state for (a) Clean-1 condition, (b) Clean-2 condition, (c) mildly polluted condition and (d) heavy polluted condition.

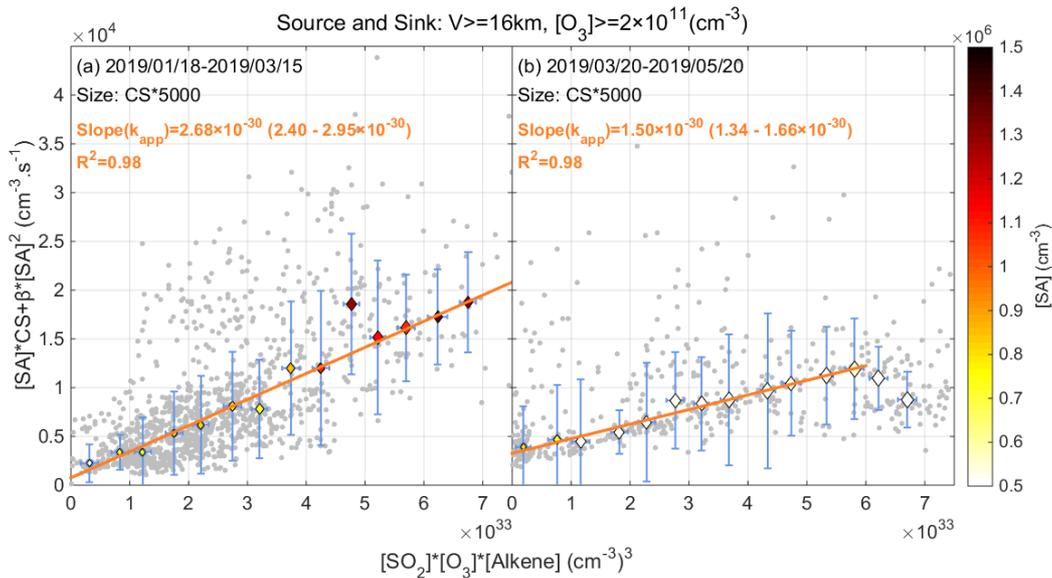


Fig. S7. Nighttime correlation between the source term ($[\text{SO}_2] [\text{O}_3] [\text{Alkene}]$) and sink term ($[\text{SA}] \text{CS} + \beta [\text{SA}]^2$) under clean conditions for (a) during 2019/01/18-2019/03/15 and (b) during 2019/03/20-2019/05/20. The gray dots are original, high time resolution 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.

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Tables

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 ₂ Increase Case	Other Cases
2019.01.20	1			1
2019.01.21	1	1		
2019.01.22	1	1		
2019.01.23	1	1		
2019.01.25	1	1		
2019.01.28	1			1
2019.02.01	1			1
2019.02.04	1		1	
2019.02.12	2	1	1	
2019.02.15	1	1		
2019.02.17	1		1	
2019.02.20	1		No CS data	
2019.02.25	1			
2019.02.26	2	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%*)

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