



*Supplement of*

**Measurement report: Three-year characteristics of sulfuric acid in urban Beijing and derivation of daytime sulfuric acid proxies applicable to inland sites**

**Yishuo Guo et al.**

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## 9 Section S1. Measurement at Hyytiälä site

10 The boreal forest SMEAR II station is located at Hyytiälä, southern Finland (Hari and Kulmala, 2005; Hari et al.,  
11 2013). The datasets used in this study are from 8<sup>th</sup> March to 13<sup>th</sup> August in 2018.

12 Sulfuric acid was measured by a CI-API-TOF mass spectrometry using nitric acid as reagent ions at the top of a  
13 35 m tower. The calibration coefficient of sulfuric acid during the measurement period was  $3.2 \times 10^9$  molec cm<sup>-3</sup>.  
14 UVB radiation was also measured at the same height as sulfuric acid by a radiometer (Solar Light SL501A). The  
15 particle number distributions of 3 - 1000 nm aerosols were obtained by a twin differential mobility particle sizer  
16 (DMPS) (Aalto et al., 2001) at 8 m in height above ground, and was then used to calculate the condensation sink  
17 (CS) according to the method from Kulmala et al. (Kulmala et al., 2001; Kulmala et al., 2012; Kerminen et al., 2001)  
18 Besides, meteorological parameters and trace gases were continuously monitored at various heights (4.2, 8.4, 16.8,  
19 33.6, 50.4, 67.2, 101, and 125 m) on a 126 m mast. Air temperature was measured by PT-100 resistance thermometer,  
20 and air relative humidity (RH) was monitored by RH sensor (Rotronic HygroMet MP102H with HygroClip HC2S3,  
21 Rotronic AG, Switzerland). SO<sub>2</sub> mixing ratio was monitored using an SO<sub>2</sub> fluorescence analyzer (Model 43i,  
22 Thermo, USA). The temperature, RH and SO<sub>2</sub> measured at 33.6 m were utilized to match the height of sulfuric acid  
23 and UVB.

## 24 Section S2. Estimation of hourly PM<sub>2.5</sub> for Hyytiälä site

25 The mass concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> of Hyytiälä were continuously measured with the filter-sampled  
26 Impactor, but they only had a resolution of 1–3 days. Meanwhile, the PM<sub>10</sub> was additionally monitored by a SHARP  
27 instrument with hourly resolution. Therefore, it is possible to obtain the hourly PM<sub>2.5</sub> data from hourly PM<sub>10</sub>. We  
28 then explore the relationship between PM<sub>2.5</sub> and PM<sub>10</sub> from Impactor measurement. And as shown in Figure S12B,  
29 PM<sub>2.5</sub> had a very good linear correlation (R = 1.0) with PM<sub>10</sub> and the scaling factor between them was 0.8. Thus,  
30 we calculated the hourly PM<sub>2.5</sub> based on the equation of  $PM_{2.5} = 0.8 \times PM_{10}$ .

## 31 Section S3. Uncertainty analysis of sulfuric acid proxies

32 According to equation (4), the uncertainty of OH-CS based proxy can be calculated as follows:

$$33 E_{\text{Proxy}_{\text{OH,CS}}} = \sqrt{(-0.7)^2 \cdot (E_T)^2 + (E_{\text{SO}_2})^2 + (E_{\text{OH}})^2 + (-1)^2 \cdot (E_{\text{CS}})^2}$$

34 The relationship between OH radical and UVB is  $\text{OH} = 6.14 \times 10^6 \cdot \text{UVB}$ . Then the uncertainty of replacing OH  
35 radical by UVB is:

$$36 E_{\text{UVB-substitution}} = \sqrt{(E_{\text{UVB-measure}})^2 + (E_{\text{UVB-to-OH}})^2 + (E_{\text{OH-modelling}})^2}$$

37 The relationship between CS and PM<sub>2.5</sub> is  $\text{CS} = 2.67 \times 10^{-3} \cdot \text{PM}_{2.5}^{2/3}$ . Then the uncertainty of replacing CS by PM<sub>2.5</sub>  
38 is:

$$39 E_{\text{PM}_{2.5}\text{-substitution}} = \sqrt{(2/3)^2 \cdot (E_{\text{PM}_{2.5}\text{-measure}})^2 + (E_{\text{PM}_{2.5}\text{-to-CS}})^2 + (E_{\text{CS}})^2}$$

40 The accuracies of measuring temperature (Vaisala) and PM<sub>2.5</sub> (ThermoFisher) are usually smaller than 1% , and  
 41 therefore can be ignored. The accuracies of measuring SO<sub>2</sub> (ThermoFisher) and UVB (KIPP&ZONEN) are around  
 42 1% and 5% respectively. The uncertainty of the model calculations of OH radical is approximately 40% (Tan et al.,  
 43 2017;Ma et al., 2019;Ma et al., 2022). The underestimation of CS from DMPS measurement is around 11.7%  
 44 (Figure S14, averaged relative errors of three periods). The relative error of using UVB to substitute OH radical is  
 45 86.5% (Figure S7A), and the relative error of using PM<sub>2.5</sub> to substitute CS is 28.9% (Figure S7B). Bring those  
 46 uncertainties of parameters into the above equations, we can get:

$$47 \quad E_{\text{UVB-substitution}} = \sqrt{(E_{\text{UVB-measure}})^2 + (E_{\text{UVB-to-OH}})^2 + (E_{\text{OH-modelling}})^2} = \sqrt{(5\%)^2 + (86.5\%)^2 + (40\%)^2} = 95.4\%$$

$$48 \quad E_{\text{PM}_{2.5}\text{-substitution}} = \sqrt{(E_{\text{PM}_{2.5}\text{-to-CS}})^2 + (E_{\text{CS}})^2} = \sqrt{(28.9\%)^2 + (11.7\%)^2} = 31.2\%$$

$$49 \quad E_{\text{Proxy}_{\text{OH,CS}}} = \sqrt{(E_{\text{SO}_2})^2 + (E_{\text{OH}})^2 + (E_{\text{CS}})^2} = \sqrt{(1\%)^2 + (40\%)^2 + (11.7\%)^2} = 41.7\%$$

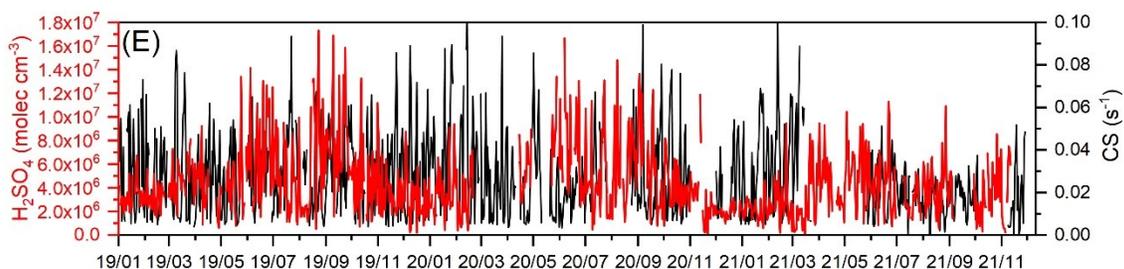
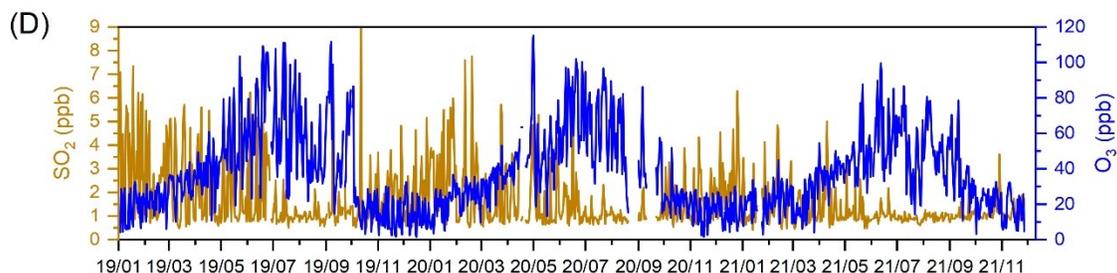
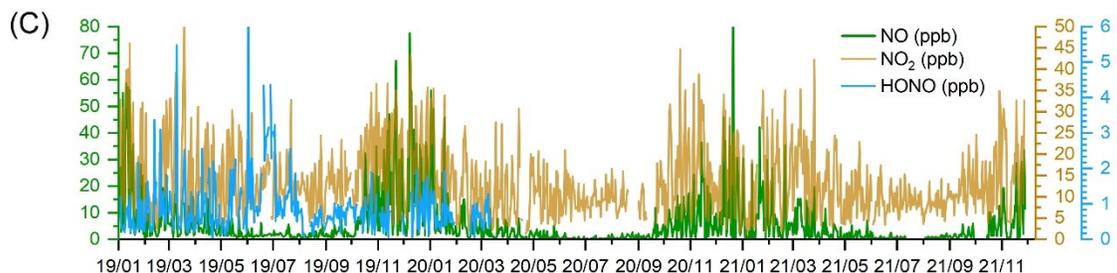
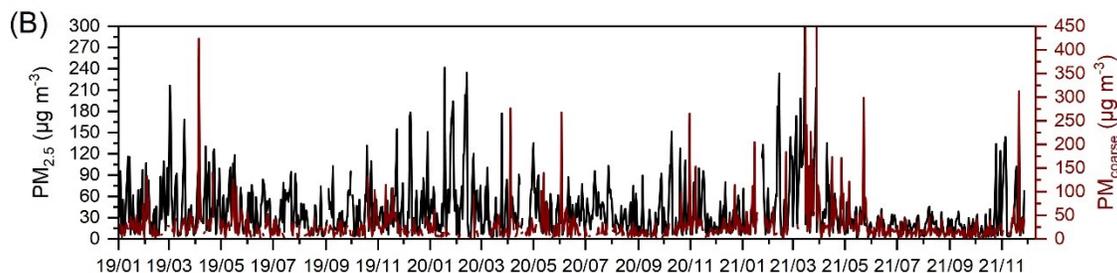
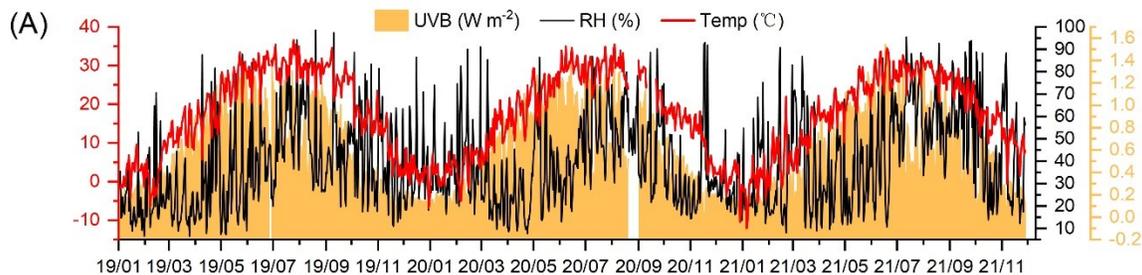
$$50 \quad E_{\text{Proxy}_{\text{UVB,CS}}} = \sqrt{(E_{\text{SO}_2})^2 + (E_{\text{UVB-substitution}})^2 + (E_{\text{CS}})^2} = \sqrt{(1\%)^2 + (95.4\%)^2 + (11.7\%)^2} = 96.1\%$$

$$51 \quad E_{\text{Proxy}_{\text{UVB,PM}_{2.5}}} = \sqrt{(E_{\text{SO}_2})^2 + (E_{\text{UVB-substitution}})^2 + (E_{\text{PM}_{2.5}\text{-substitution}})^2} = \sqrt{(1\%)^2 + (95.4\%)^2 + (31.2\%)^2} = 100.4\%$$

52 In summary, the uncertainties of OH-CS, UVB-CS and UVB-PM<sub>2.5</sub> based proxies are estimated to be 41.7%, 96.1%  
 53 and 100.4%, respectively.

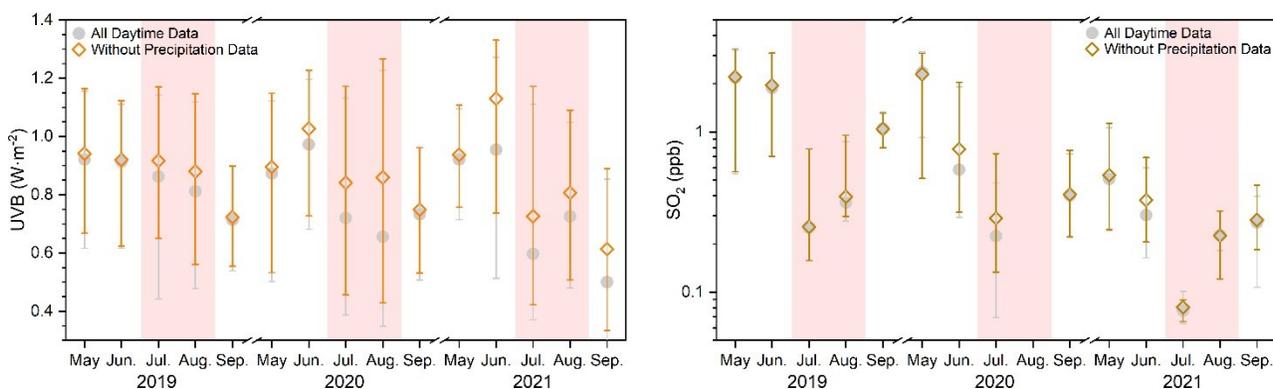
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55 **Figures**

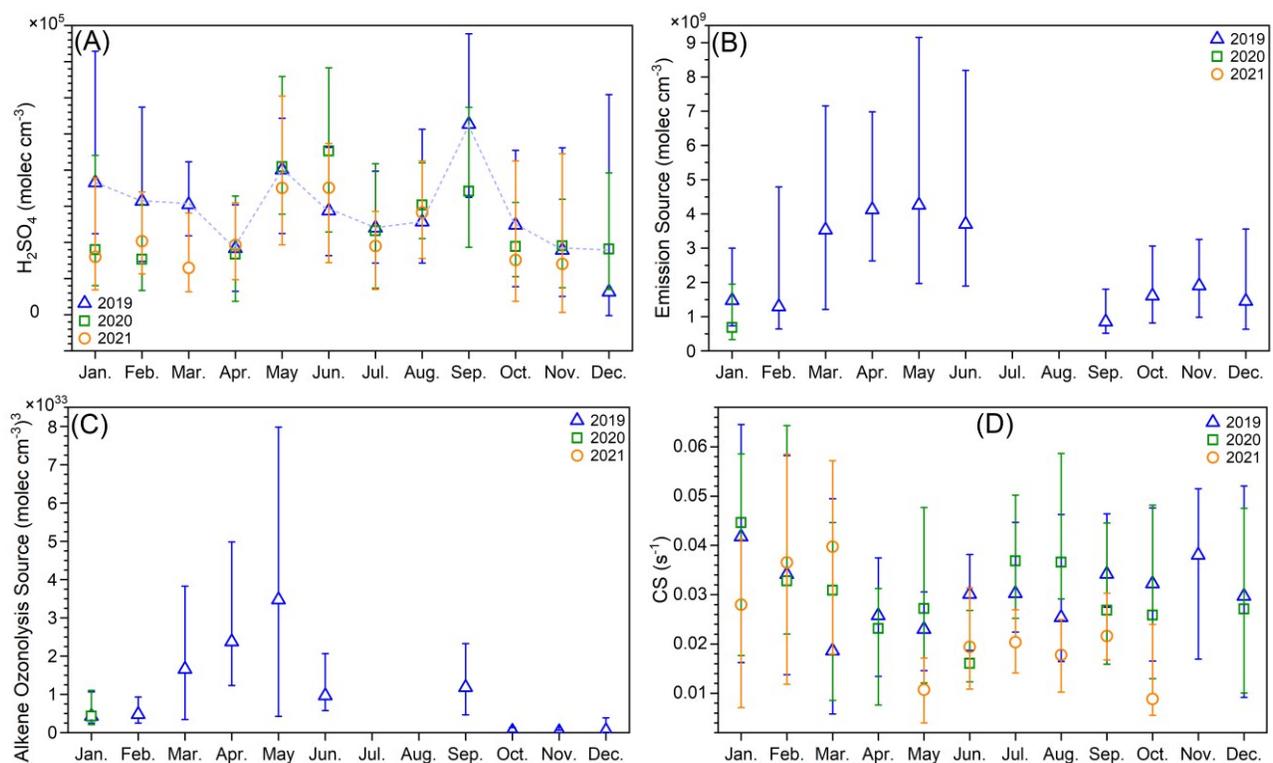


61 **Figure S1.** Three-year (from 2019 to 2021) time variations of (A) UVB, RH and temperature, (B)  $\text{PM}_{2.5}$  and  $\text{PM}_{\text{coarse}}$ , (C) NO,  
 62  $\text{NO}_2$  and HONO, (D)  $\text{SO}_2$  and  $\text{O}_3$ , and (E) sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and condensation sink (CS) during daytime (10:00-14:00).  
 63 The labels on x-axes are in “year/month” format.

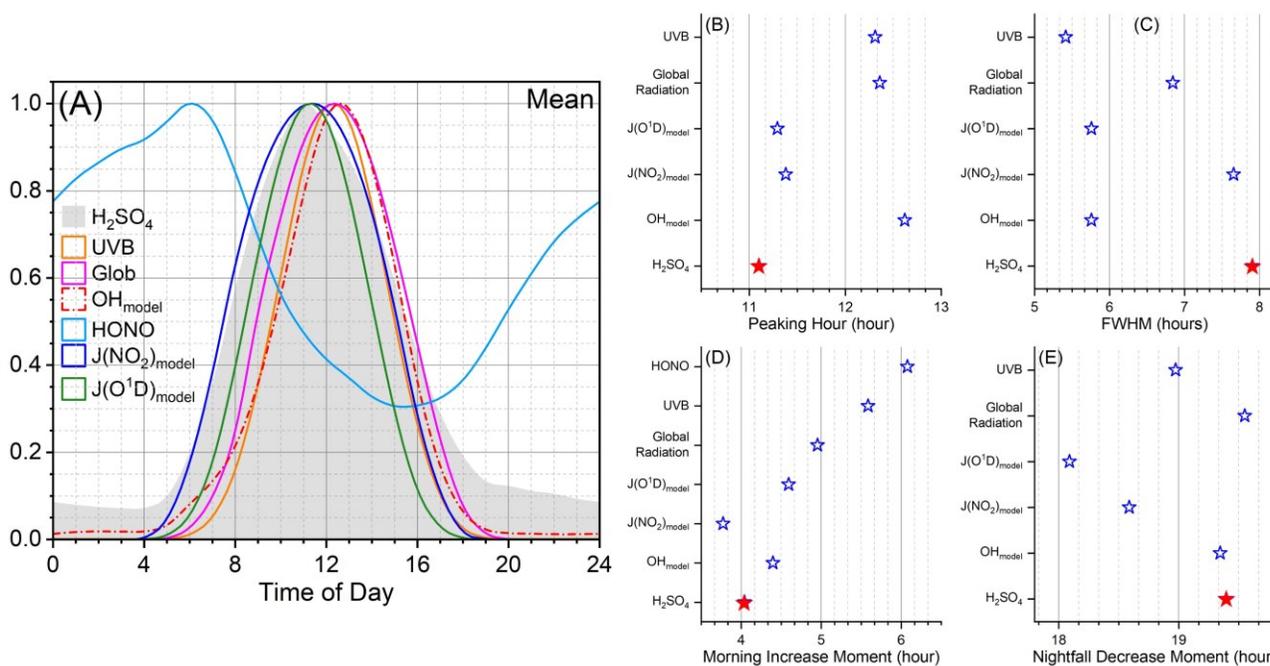
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66 **Figure S2.** Monthly variations of UVB and SO<sub>2</sub> during daytime (10:00-14:00) in May, June, July, August and September in  
67 2019, 2020 and 2021. The gray circles and orange diamonds represents datasets of all and without precipitation moments,  
68 respectively. The up line, middle marker and bottom line stand for upper quartile, median and lower quartile values respectively.  
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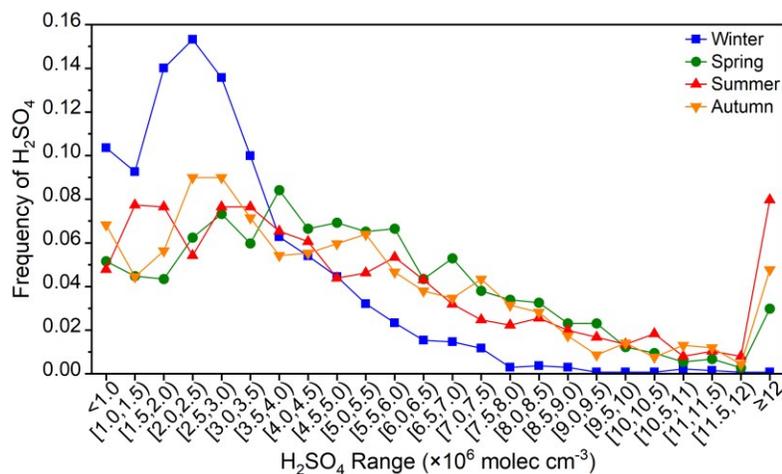


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72 **Figure S3.** Three-year (from 2019 January to 2021 November) monthly variations of (A) H<sub>2</sub>SO<sub>4</sub> concentration, (B) emission  
73 source of H<sub>2</sub>SO<sub>4</sub> (Emission Source= $[\text{Benzene}] \left(\frac{\text{WS}}{1 \text{ m s}^{-1}}\right)^{1.398} \left(\frac{\text{CS}}{0.01 \text{ s}^{-1}}\right)^{-1.404}$  (Yang et al., 2021)), (C) alkene ozonolysis source of  
74 H<sub>2</sub>SO<sub>4</sub> (Alkene Ozonolysis Source= $[\text{SO}_2][\text{O}_3][\text{Alkene}]$  (Guo et al., 2021)) and (D) condensation sink (CS) during nighttime  
75 (22:00-02:00 next day). Blue triangles, green squares and orange circles represent data in 2019, 2020 and 2021, respectively.  
76 The up line, middle marker and bottom line stand for upper quartile, median and lower quartile values, respectively.  
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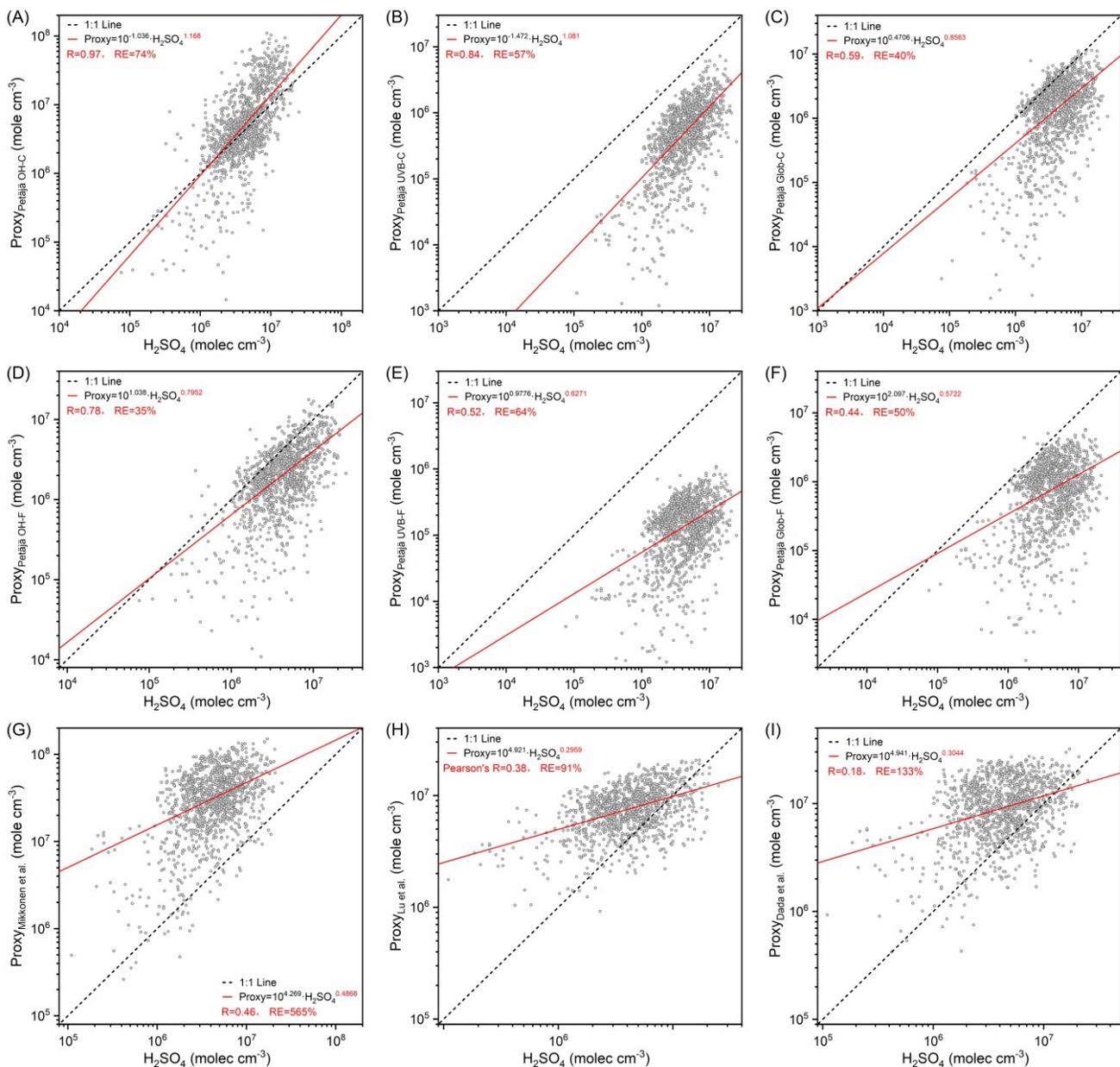
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**Figure S4.** (A) Mean diurnal variations of H<sub>2</sub>SO<sub>4</sub>, UVB, global radiation (Glob), modelled OH radical (OH<sub>model</sub>), HONO, modelled photolysis rate of NO<sub>2</sub> (J(NO<sub>2</sub>)<sub>model</sub>) and modelled photolysis rate of O<sub>3</sub> to generate O(<sup>1</sup>D) (J(O<sup>1</sup>D)<sub>model</sub>) of 2019. The diurnal curves are depicted in normalized values. Values of (B) peaking hour, (C) full width at half maximum (FWHM), (D) morning increase moment, and (E) nightfall decrease moment from the curves of figure (A). Except from HONO, morning increase moment is when the diurnal curve starts increasing rapidly in the morning, and the nightfall decrease moment is when the diurnal curve turns from rapid to slow decreases at nightfall. For HONO, the “morning increase moment” is when HONO starts declining rapidly in the morning.

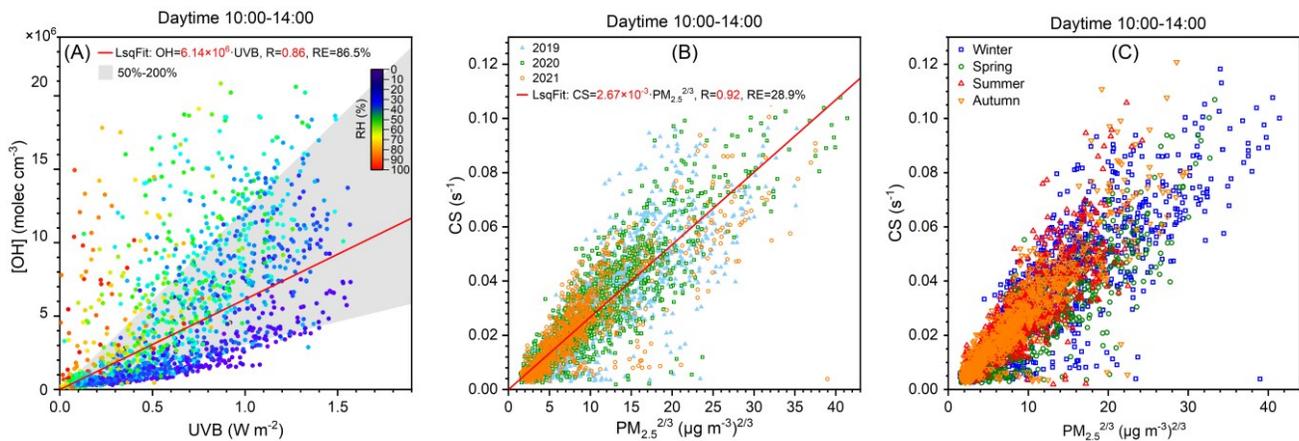


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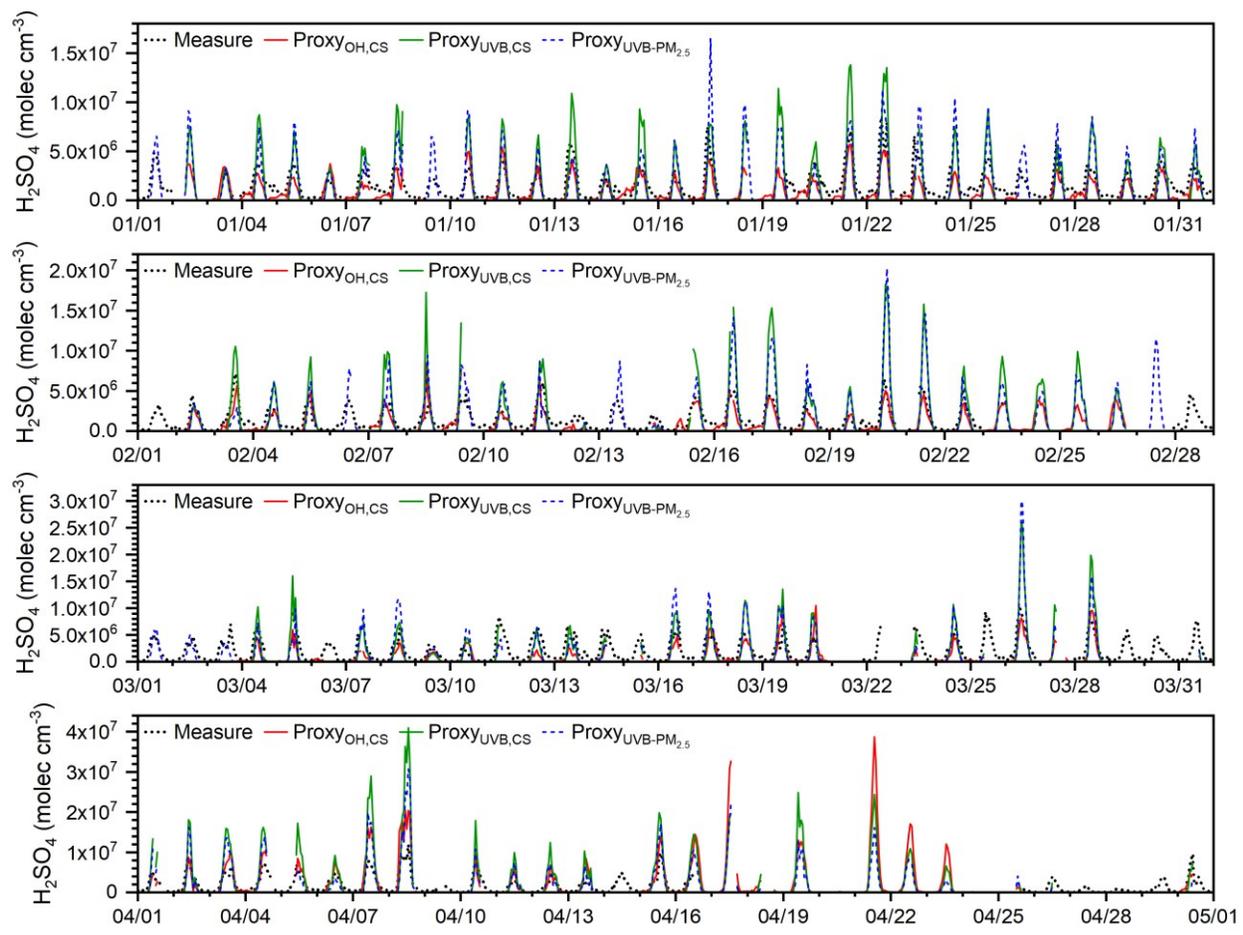
**Figure S5.** Distribution of daytime (10:00-14:00) sulfuric acid concentration in four seasons during three years (2019 to 2021).



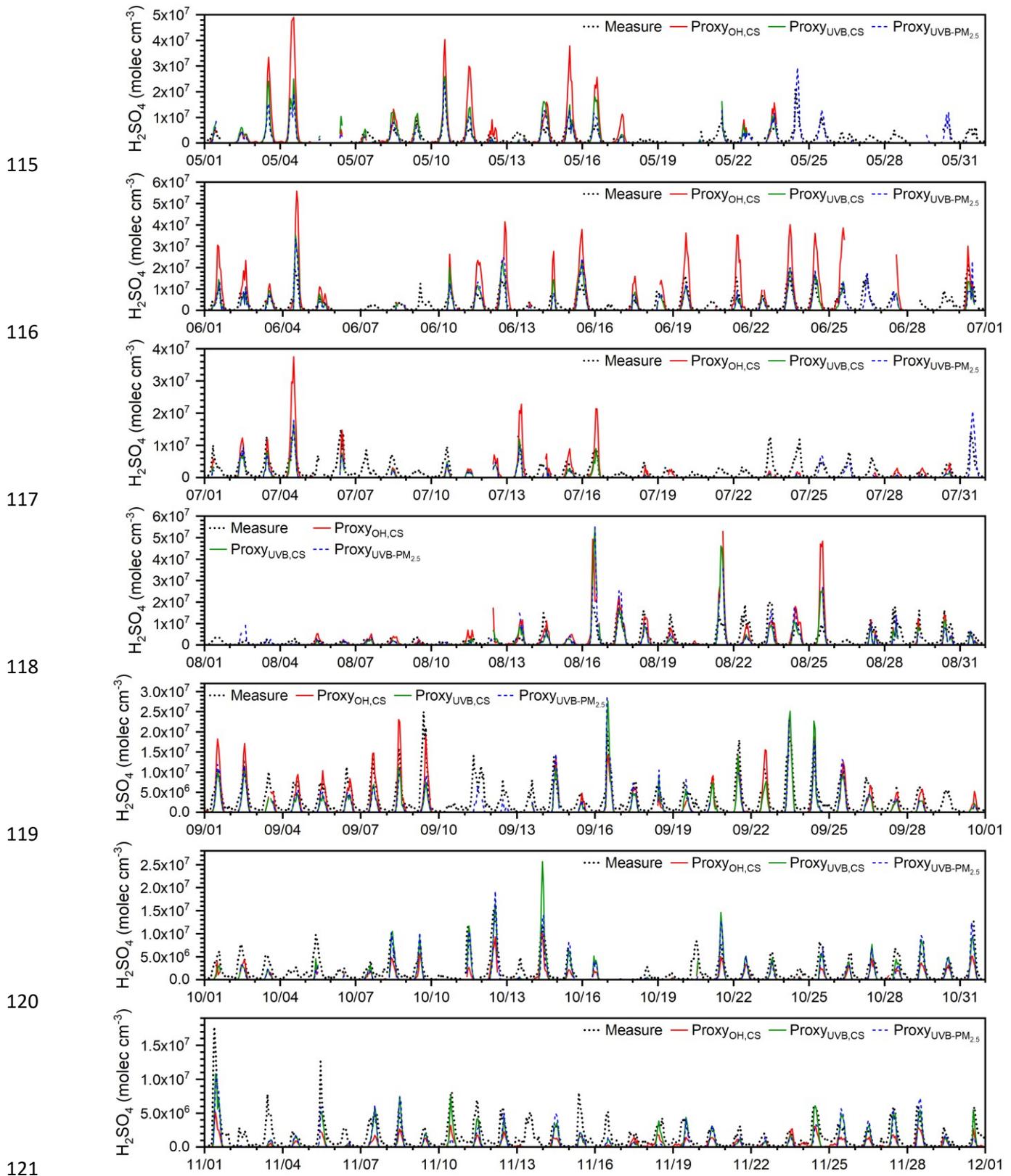
**Figure S6.**  $\text{H}_2\text{SO}_4$  proxy suggested by (A) Petäjä et al., 2009 using OH radical with constant pre-factor  $k_1$ , (B) Petäjä et al., 2009 using UVB with constant pre-factor  $k_2$ , (C) Petäjä et al., 2009 using global radiation with constant pre-factor  $k_3$ , (D) Petäjä et al., 2009 using OH radical with fitted pre-factor  $k_1$ , (E) Petäjä et al., 2009 using UVB with fitted pre-factor  $k_2$ , (F) Petäjä et al., 2009 using global radiation with fitted pre-factor  $k_3$ , (G) Mikkonen et al., 2011 with equation N5, (H) Lu et al., 2019 with equation N7, and (I) Data et al., 2020 with the result of Figure 7(b) vs. measured  $\text{H}_2\text{SO}_4$  during daytime (10:00-14:00) in 2019. In each plot, the black dashed lines are 1:1 lines, and the red lines are the least square fit lines between proxy and measured  $\text{H}_2\text{SO}_4$ . Corresponding functions of the least square fit, correlation coefficients (R), and the relative error (RE) between proxies and measured  $\text{H}_2\text{SO}_4$  are also shown in the legend.

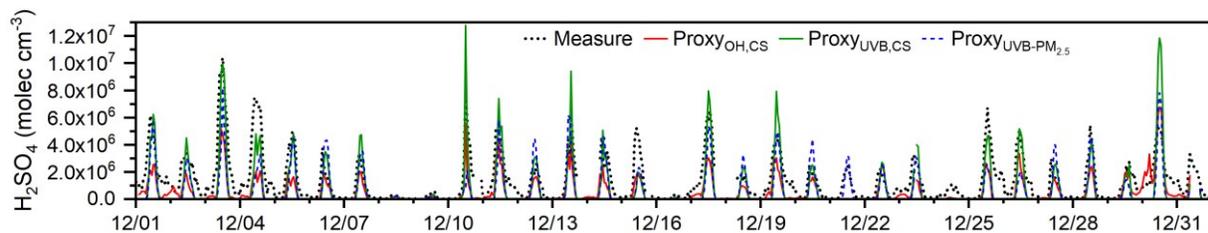


103 **Figure S7.** (A) Relationship between modelled OH radical and UVB colored by RH during daytime (10:00-14:00) in 2019. (B)  
 104 and (C) Relationship between condensation sink (CS) and  $PM_{2.5}^{2/3}$  during daytime from 2019 to 2021. In figure B, light blue  
 105 triangle, hollow green square and hollow orange circle are for 2019, 2020 and 2021, respectively. In figure C, blue square,  
 106 green circle, red upper triangle and orange lower triangle are for winter, spring, summer and autumn, respectively. Red lines  
 107 are the least square fit lines between two parameters. The corresponding functions, correlation coefficients (R) and relative  
 108 errors (RE) are shown in the legend. Gray area in figure A denotes 50% to 200% of the least square fitting.  
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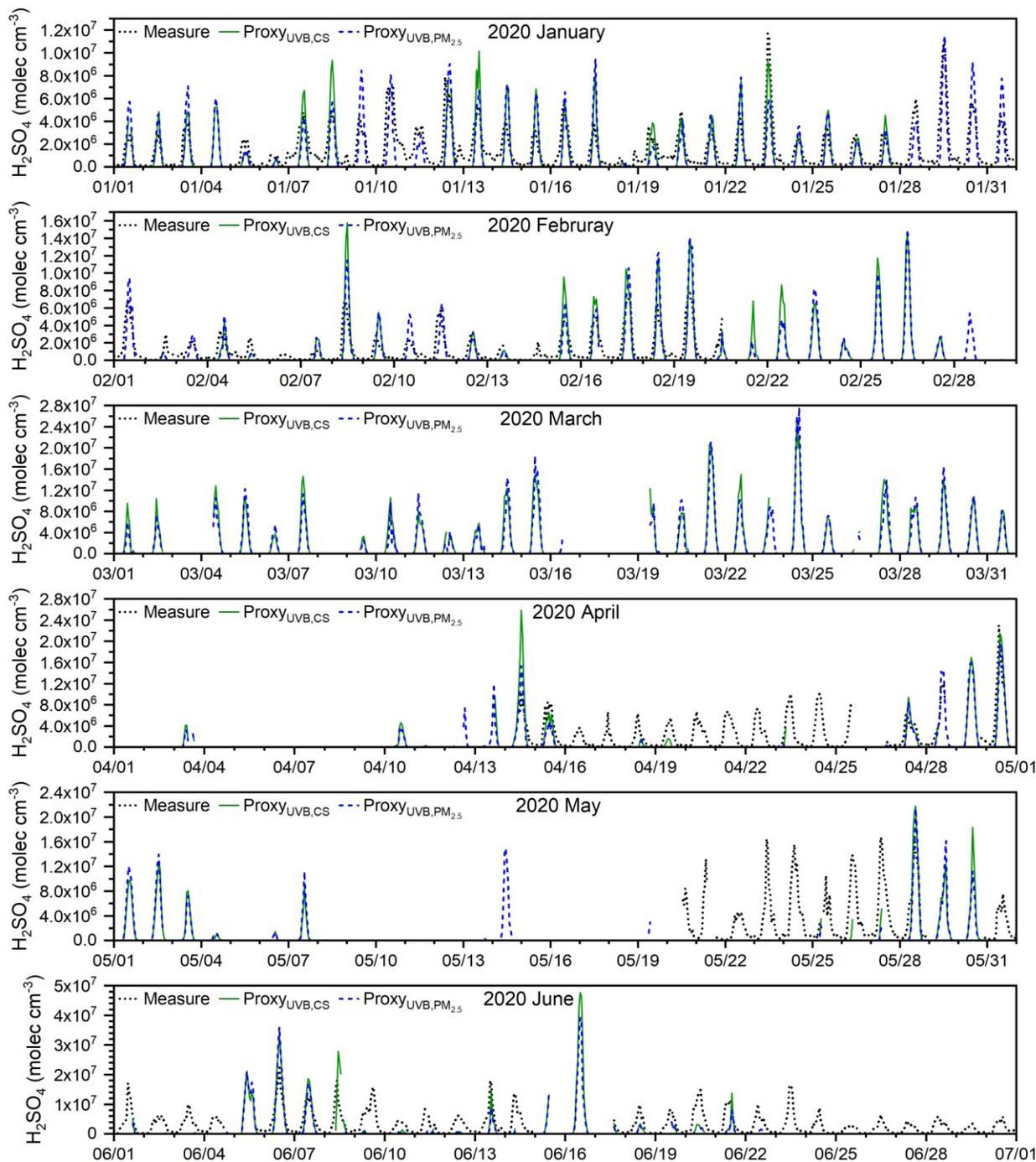


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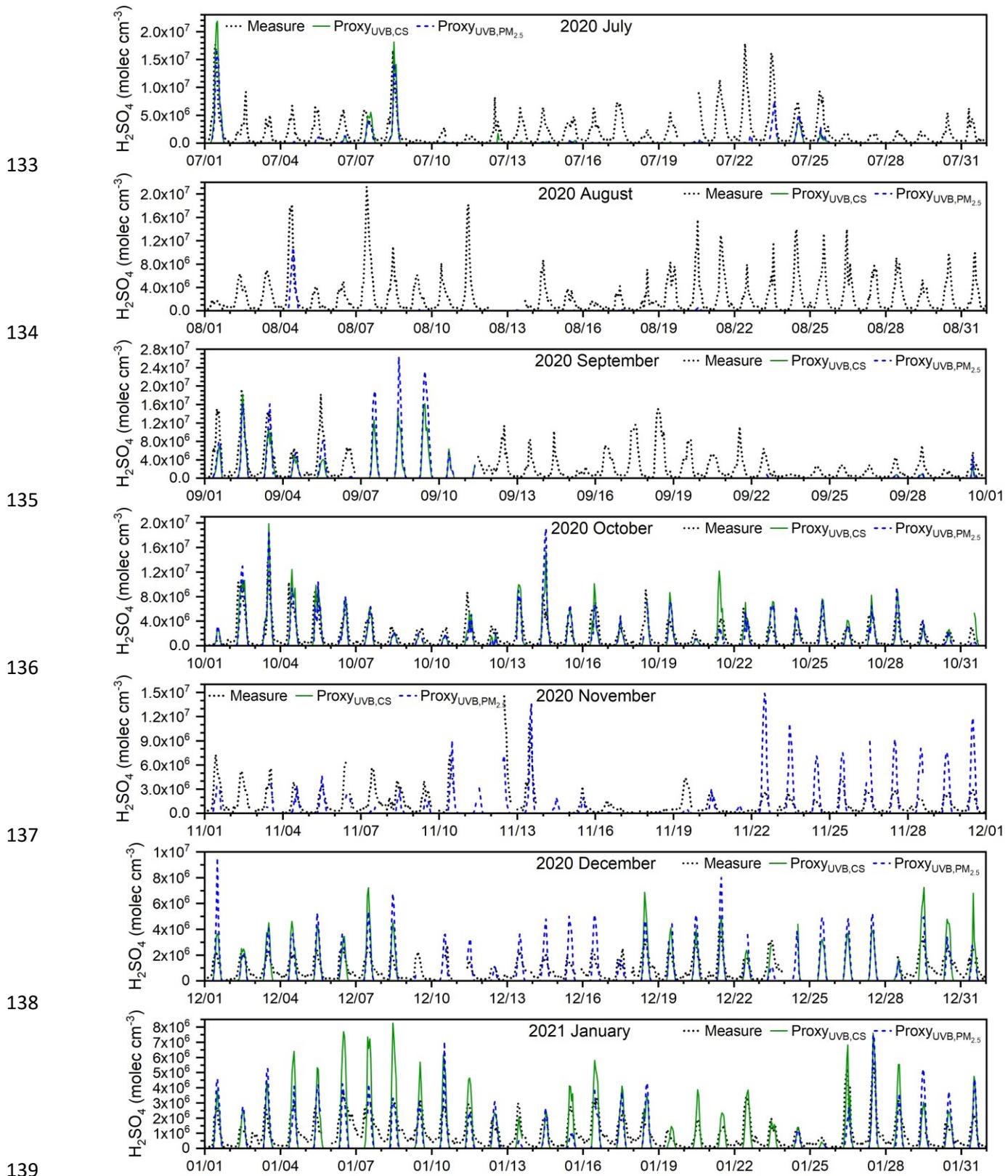


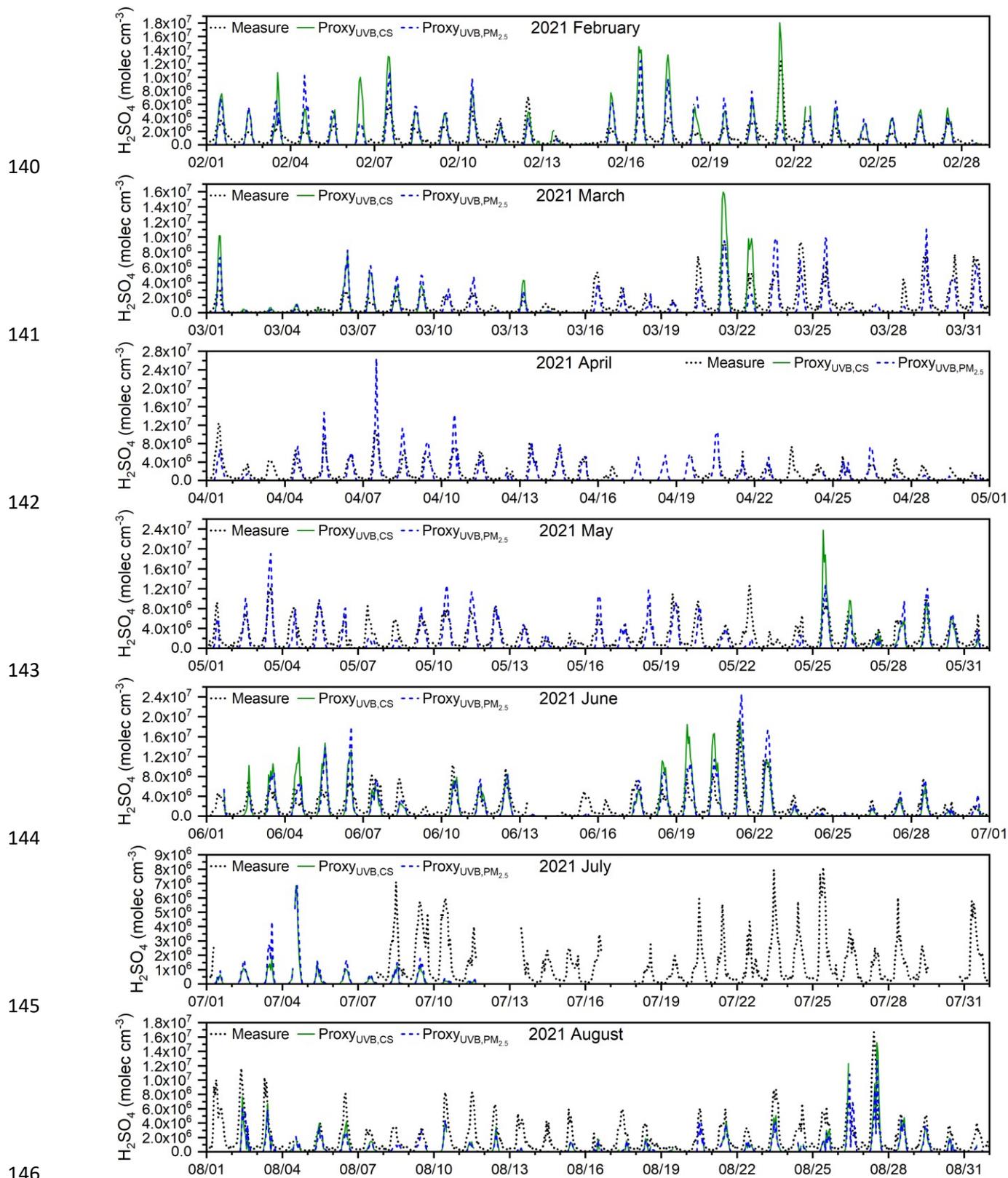


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 123 **Figure S8.** Time variation of measured sulfuric acid (Measure, black dashed line), OH-CS based proxy (Proxy<sub>OH,CS</sub>, red line),  
 124 UVB-CS based proxy (Proxy<sub>UVB,CS</sub>, green line) and UVB-PM<sub>2.5</sub> based proxy (Proxy<sub>UVB,PM<sub>2.5</sub></sub>, blue dashed line) of sulfuric acid  
 125 in 2019.  
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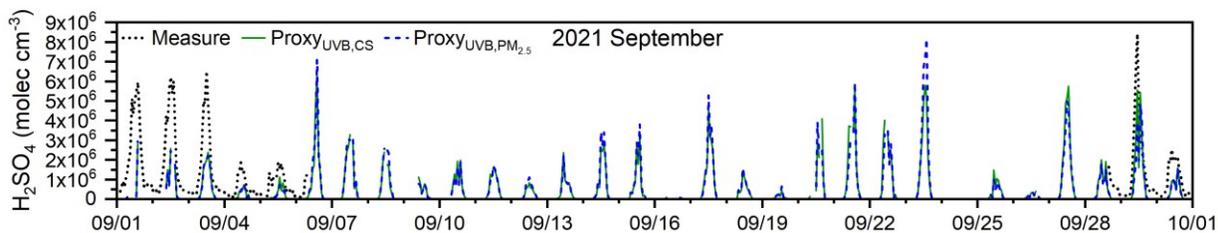


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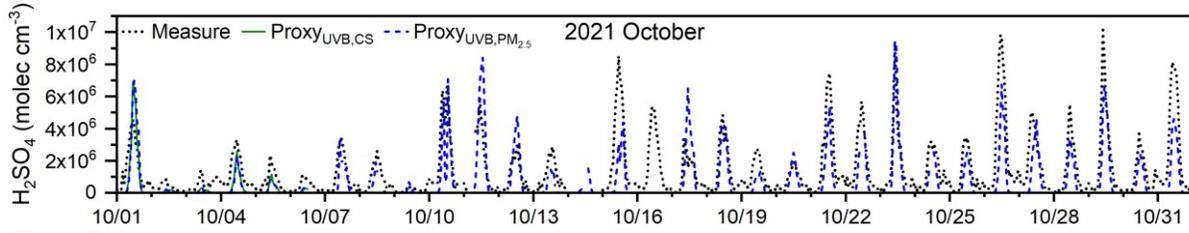




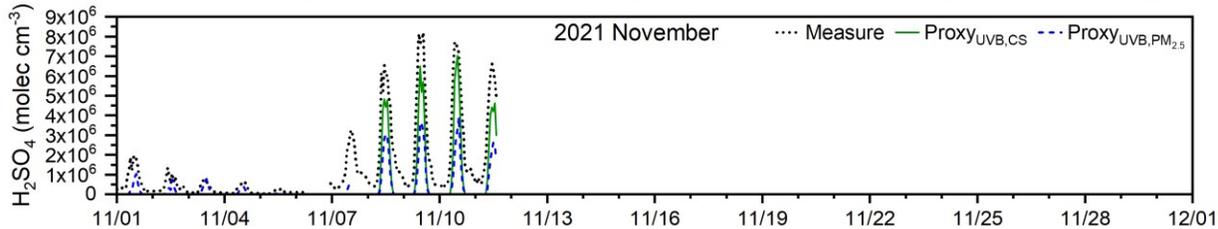
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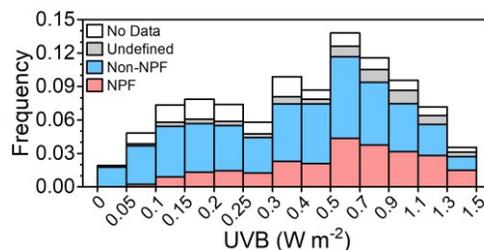
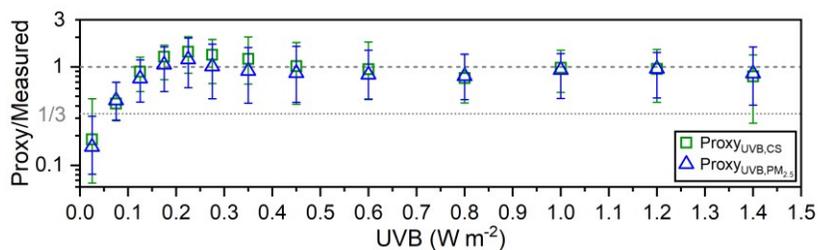
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**Figure S9.** Time variations of measured sulfuric acid (Measure, black dashed line), UVB-CS based proxy (Proxy<sub>UVB,CS</sub>, green line) and UVB-PM<sub>2.5</sub> based proxy (Proxy<sub>UVB,PM<sub>2.5</sub></sub>, blue dashed line) of sulfuric acid in 2020 and 2021.

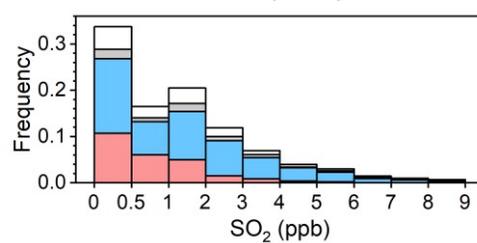
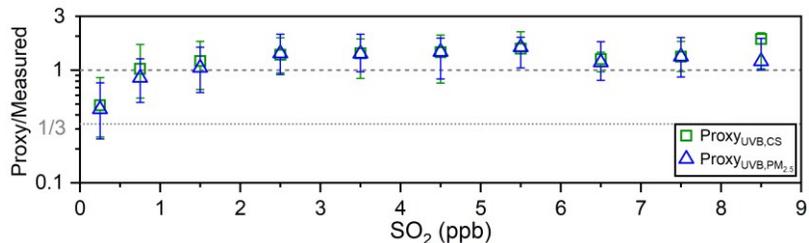
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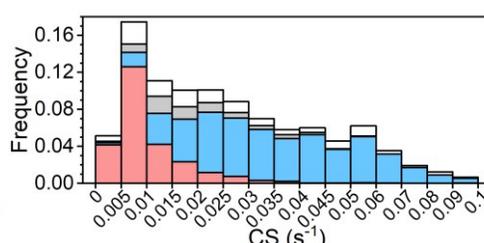
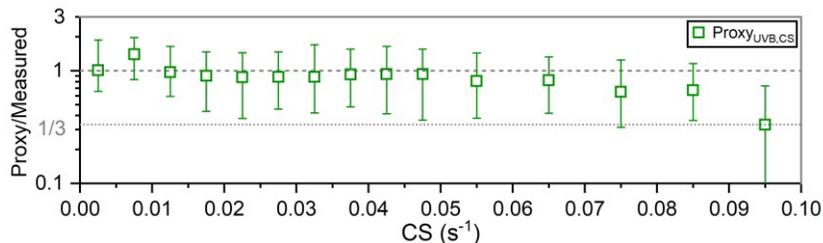
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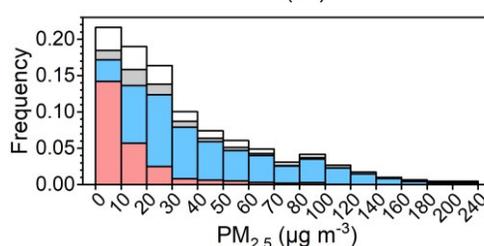
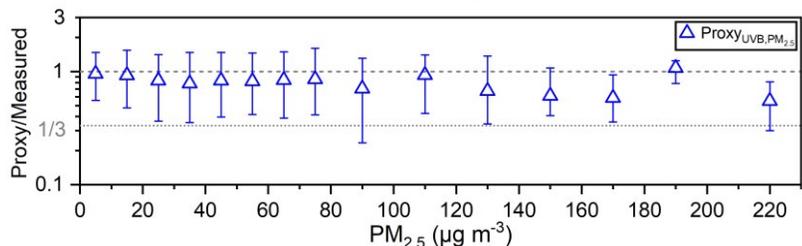
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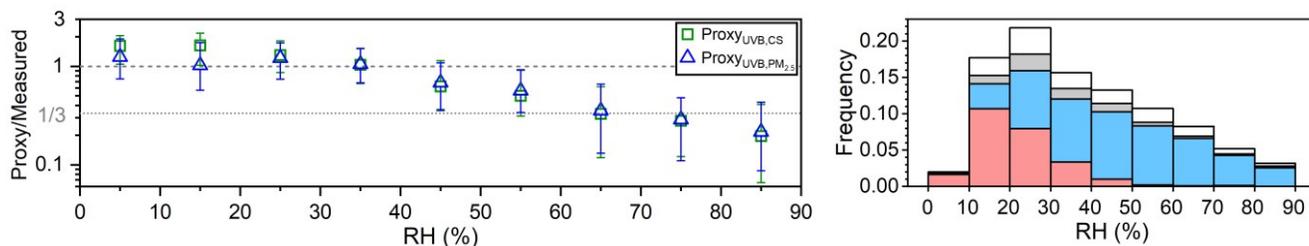


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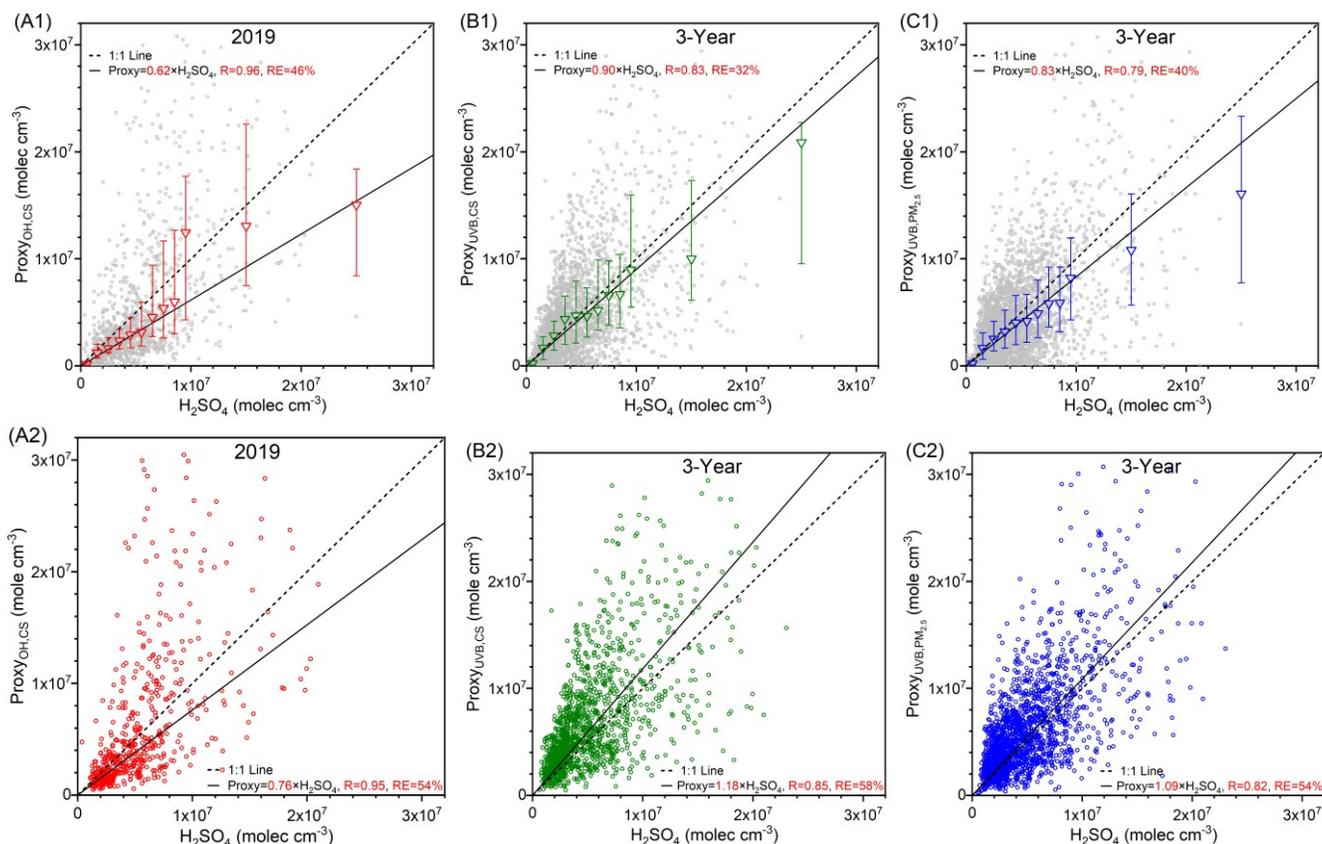


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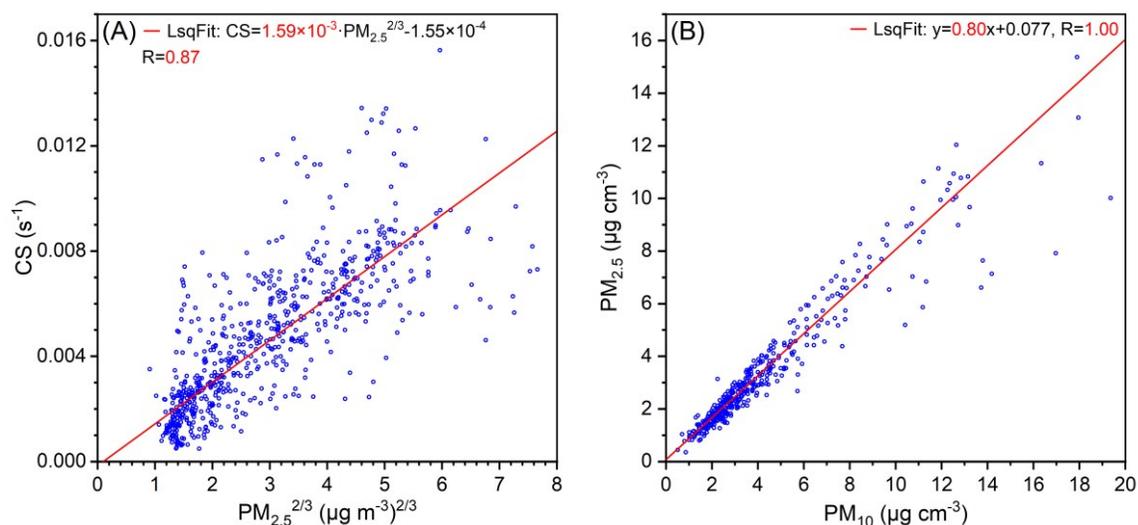




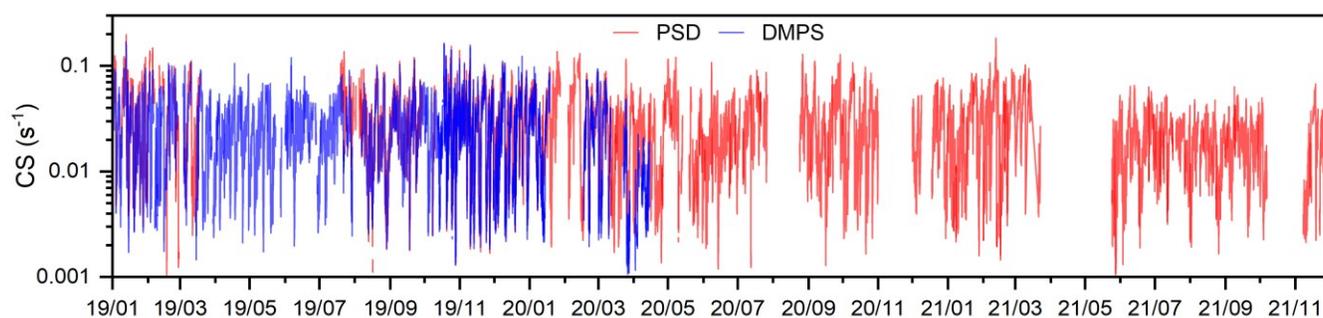
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 158 **Figure S10.** Left: The ratios of sulfuric acid concentrations estimated by proxies in this study to the measured one  
 159 (Proxy/Measured) vs. UVB, SO<sub>2</sub>, CS, PM<sub>2.5</sub> and RH during daytime (10:00-14:00) of three years. Different colored markers  
 160 represent different proxies. The up line, middle marker and bottom line stand for upper quartile, median and lower quartile  
 161 values respectively. Right: Frequency distributions of corresponding parameters classified by “NPF”, “Non-NPF”,  
 162 “Undefined”, and “No Data” periods.  
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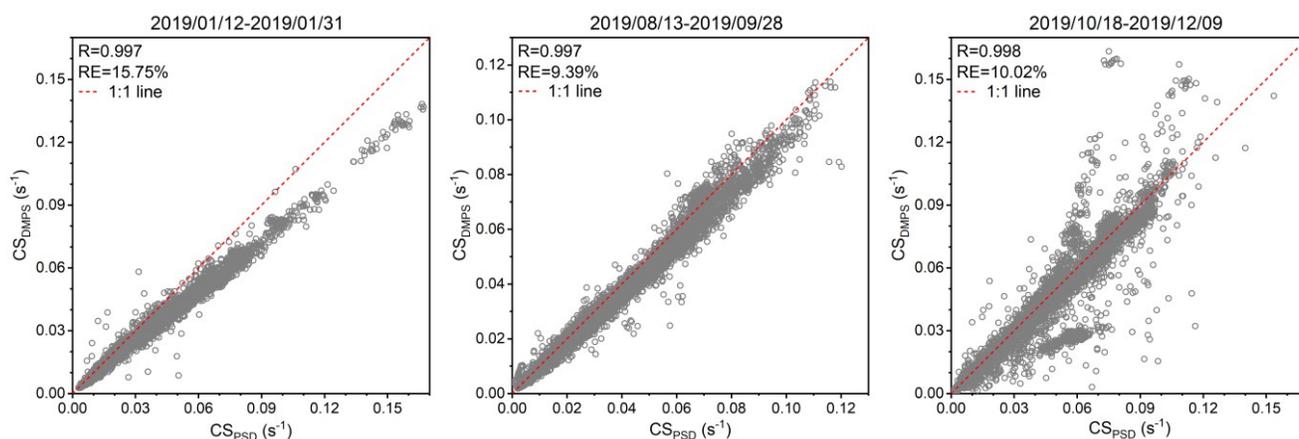
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 166 **Figure S11.** Sulfuric acid concentrations estimated by proxies vs. measured concentration during daytime (10:00-14:00) for  
 167 (A1) and (A2) Proxy<sub>OH,CS</sub> in 2019, (B1) and (B2) Proxy<sub>UVB,CS</sub> in 3 years, and (C1) and (C2) Proxy<sub>UVB,PM<sub>2.5</sub></sub> in 3 years. The  
 168 black dashed lines are 1:1 lines, and the black lines are the distance weighted least square fits between proxy and measured  
 169 sulfuric acid. Corresponding functions of the fits, correlation coefficients (R) and relative errors (RE) are shown in the legend.  
 170 For the first row, datasets include all daytime data. The triangle marker represents the binned data, where the up line, middle  
 171 marker and bottom lines stand for upper quartile, median and lower quartile, respectively. For the second row, datasets only  
 172 cover the optimal parameter ranges. Optimal parameter ranges for Proxy<sub>OH,CS</sub>:  $4 \times 10^5 < [\text{OH}] < 1.2 \times 10^7$  molec cm<sup>-3</sup>,  $0.015 <$   
 173  $\text{CS} < 0.07$  s<sup>-1</sup>,  $\text{SO}_2 > 0.5$  ppb and  $\text{RH} < 60\%$ . Optimal parameter ranges for Proxy<sub>UVB,CS</sub>:  $\text{UVB} > 0.1$  W m<sup>-2</sup>,  $\text{CS} < 0.07$  s<sup>-1</sup>,  $\text{SO}_2 >$   
 174  $0.5$  ppb and  $\text{RH} < 60\%$ . Optimal parameter ranges for Proxy<sub>UVB,PM<sub>2.5</sub></sub>:  $\text{UVB} > 0.1$  W m<sup>-2</sup>,  $\text{PM}_{2.5} < 200$  μg m<sup>-3</sup>,  $\text{SO}_2 > 0.5$  ppb  
 175 and  $\text{RH} < 60\%$ .  
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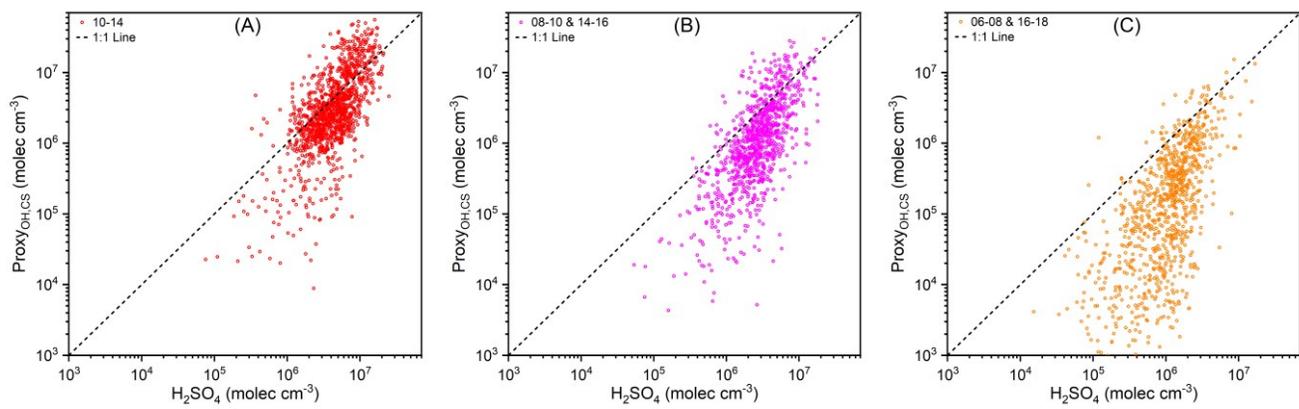
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 178 **Figure S12.** (A) Relationship between condensation sink (CS) and  $PM_{2.5}^{2/3}$  for Hyytiälä site during daytime (10:00–14:00) from  
 179 8<sup>th</sup> March to 13<sup>th</sup> Aug. 2018. (B) Relationship between daily  $PM_{2.5}$  and daily  $PM_{10}$  for Hyytiälä site from 2017 to 2019. In both  
 180 two plots, the red lines are the least square fit lines between two parameters. Corresponding functions of the fits and correlation  
 181 coefficients (R) are also shown in the legend.  
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 184 **Figure S13.** Three-year (from 2019 to 2021) time variations of condensation sink (CS) calculated from the particle number  
 185 size distribution of PSD and DMPS. The labels on x-axes are in “year/month” format.  
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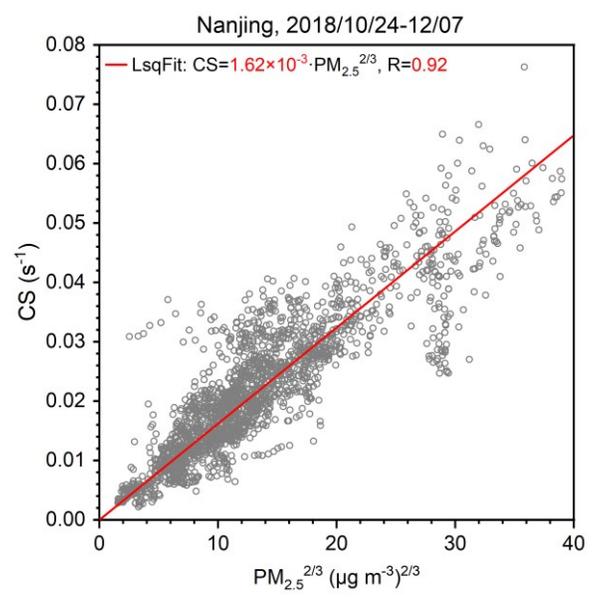


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 188 **Figure S14.** Correlation between condensation sinks (CS) calculated from DMPS with size ranges of 6–840 nm and PSD with  
 189 size ranges of 3 nm–10  $\mu$ m during three periods with continuous and stable measurement. The correlation coefficients (R) and  
 190 relative errors between them are listed in the legend.  
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**Figure S15.** OH-CS based proxy vs. measured sulfuric acid during daytime in 2019 for (A) 10:00–14:00, (B) 08:00–10:00 and 14:00–16:00, as well as (C) 06:00–08:00 and 16:00–18:00. The black dashed lines are 1:1 lines.



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**Figure S16.** Relationship between condensation sink (CS) and  $PM_{2.5}^{2/3}$  for Nanjing site (Nie et al., 2022) from 24<sup>th</sup> October to 7<sup>th</sup> December, 2018.

200 **Tables**

201 **Table S1.** Precipitation frequency during daytime (10:00-14:00) in May, June, July, August and September in 2019, 2020 and  
 202 2021.

Month	2019	2020	2021
May	4%	14%	8%
June	5%	15%	25%
<b>July</b>	<b>19%</b>	<b>21%</b>	<b>37%</b>
<b>August</b>	<b>13%</b>	<b>20%</b>	<b>26%</b>
September	5%	12%	24%

203 **Table S2.** Monthly concentration of H<sub>2</sub>SO<sub>4</sub> (molec cm<sup>-3</sup>) during nighttime (22:00-02:00 next day) from 2019 to 2021. “NaN”  
 204 means there is no data available.  
 205

Month	2019			2020			2021		
	Median	25th	75th	Median	25th	75th	Median	25th	75th
January	4.7×10 <sup>5</sup>	3.2×10 <sup>5</sup>	8.3×10 <sup>5</sup>	2.8×10 <sup>5</sup>	1.8×10 <sup>5</sup>	5.4×10 <sup>5</sup>	2.6×10 <sup>5</sup>	1.7×10 <sup>5</sup>	4.8×10 <sup>5</sup>
February	4.1×10 <sup>5</sup>	2.5×10 <sup>5</sup>	6.7×10 <sup>5</sup>	2.5×10 <sup>5</sup>	1.7×10 <sup>5</sup>	4.1×10 <sup>5</sup>	3.0×10 <sup>5</sup>	2.1×10 <sup>5</sup>	4.4×10 <sup>5</sup>
March	4.0×10 <sup>5</sup>	3.2×10 <sup>5</sup>	5.2×10 <sup>5</sup>	NaN	NaN	NaN	2.3×10 <sup>5</sup>	1.6×10 <sup>5</sup>	3.8×10 <sup>5</sup>
April	2.8×10 <sup>5</sup>	1.6×10 <sup>5</sup>	4.0×10 <sup>5</sup>	2.7×10 <sup>5</sup>	1.4×10 <sup>5</sup>	4.3×10 <sup>5</sup>	2.9×10 <sup>5</sup>	2.0×10 <sup>5</sup>	4.1×10 <sup>5</sup>
May	5.0×10 <sup>5</sup>	3.3×10 <sup>5</sup>	6.4×10 <sup>5</sup>	5.1×10 <sup>5</sup>	3.8×10 <sup>5</sup>	7.6×10 <sup>5</sup>	4.5×10 <sup>5</sup>	2.9×10 <sup>5</sup>	7.0×10 <sup>5</sup>
June	3.9×10 <sup>5</sup>	2.6×10 <sup>5</sup>	5.6×10 <sup>5</sup>	5.5×10 <sup>5</sup>	3.3×10 <sup>5</sup>	7.8×10 <sup>5</sup>	4.5×10 <sup>5</sup>	2.4×10 <sup>5</sup>	5.7×10 <sup>5</sup>
July	3.4×10 <sup>5</sup>	2.4×10 <sup>5</sup>	5.0×10 <sup>5</sup>	3.3×10 <sup>5</sup>	1.7×10 <sup>5</sup>	5.2×10 <sup>5</sup>	2.9×10 <sup>5</sup>	1.7×10 <sup>5</sup>	3.9×10 <sup>5</sup>
August	3.6×10 <sup>5</sup>	2.4×10 <sup>5</sup>	6.1×10 <sup>5</sup>	4.0×10 <sup>5</sup>	3.1×10 <sup>5</sup>	5.2×10 <sup>5</sup>	3.8×10 <sup>5</sup>	2.6×10 <sup>5</sup>	5.3×10 <sup>5</sup>
September	<b>6.3×10<sup>5</sup></b>	4.2×10 <sup>5</sup>	8.8×10 <sup>5</sup>	4.4×10 <sup>5</sup>	2.9×10 <sup>5</sup>	6.7×10 <sup>5</sup>	NaN	NaN	NaN
October	3.5×10 <sup>5</sup>	1.8×10 <sup>5</sup>	5.6×10 <sup>5</sup>	2.9×10 <sup>5</sup>	2.1×10 <sup>5</sup>	4.1×10 <sup>5</sup>	2.5×10 <sup>5</sup>	1.4×10 <sup>5</sup>	5.3×10 <sup>5</sup>
November	2.8×10 <sup>5</sup>	1.5×10 <sup>5</sup>	5.6×10 <sup>5</sup>	2.9×10 <sup>5</sup>	1.7×10 <sup>5</sup>	4.2×10 <sup>5</sup>	2.4×10 <sup>5</sup>	1.1×10 <sup>5</sup>	5.5×10 <sup>5</sup>
December	<b>1.6×10<sup>5</sup></b>	9.8×10 <sup>4</sup>	7.1×10 <sup>5</sup>	2.8×10 <sup>5</sup>	1.7×10 <sup>5</sup>	4.9×10 <sup>5</sup>	NaN	NaN	NaN

206 **Table S3.** Mean, standard deviation (Std), median, lower quartile (25th) and upper quartile (75th) of sulfuric acid concentrations  
 207 from measurement and estimated by proxies from literatures during the time window of 10:00-14:00 in 2019 (1<sup>st</sup> January to  
 208 31<sup>st</sup> December). The unit of concentration is molec cm<sup>-3</sup>.  
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Parameters		Mean	Std	Median	25th	75th
Measured H <sub>2</sub> SO <sub>4</sub>		4.9×10 <sup>6</sup>	3.5×10 <sup>6</sup>	3.9×10 <sup>6</sup>	2.5×10 <sup>6</sup>	6.3×10 <sup>6</sup>
H <sub>2</sub> SO <sub>4</sub> Proxy	Proxy Petäjä OH-C	1.0×10 <sup>7</sup>	1.4×10 <sup>7</sup>	4.7×10 <sup>6</sup>	2.5×10 <sup>6</sup>	1.1×10 <sup>7</sup>
	Proxy Petäjä OH-F	2.8×10 <sup>6</sup>	2.6×10 <sup>6</sup>	2.1×10 <sup>6</sup>	1.2×10 <sup>6</sup>	3.5×10 <sup>6</sup>
	Proxy Petäjä UVB-C	7.7×10 <sup>5</sup>	8.0×10 <sup>5</sup>	5.5×10 <sup>5</sup>	2.7×10 <sup>5</sup>	1.0×10 <sup>6</sup>
	Proxy Petäjä UVB-F	1.9×10 <sup>5</sup>	1.5×10 <sup>5</sup>	1.6×10 <sup>5</sup>	7.2×10 <sup>4</sup>	2.7×10 <sup>5</sup>
	Proxy Petäjä Glob-C	2.1×10 <sup>6</sup>	1.8×10 <sup>6</sup>	1.7×10 <sup>6</sup>	7.4×10 <sup>5</sup>	3.0×10 <sup>6</sup>
	Proxy Petäjä Glob-F	1.1×10 <sup>6</sup>	9.0×10 <sup>5</sup>	9.0×10 <sup>5</sup>	3.9×10 <sup>5</sup>	1.6×10 <sup>6</sup>
	Proxy Mikkonen et al.	3.9×10 <sup>7</sup>	2.6×10 <sup>7</sup>	3.4×10 <sup>7</sup>	1.9×10 <sup>7</sup>	5.2×10 <sup>7</sup>
	Proxy Lu et al.	8.3×10 <sup>6</sup>	3.7×10 <sup>6</sup>	7.8×10 <sup>6</sup>	5.6×10 <sup>6</sup>	1.0×10 <sup>7</sup>
Proxy Dada et al.	1.1×10 <sup>7</sup>	5.8×10 <sup>6</sup>	9.7×10 <sup>6</sup>	6.0×10 <sup>6</sup>	1.4×10 <sup>7</sup>	

210 **Table S4.** Mean, standard deviation (Std), median, lower quartile (25th) and upper quartile (75th) of sulfuric acid concentrations  
 211 from measurement and estimated by proxies in this study during daytime (10:00-14:00). The concentrations in 2019, 2020,  
 212 2021 and 3 years are summarized respectively. The unit of concentration is molec cm<sup>-3</sup>.  
 213

Year	Parameters	Mean	Std	Median	25th	75th
2019	[H <sub>2</sub> SO <sub>4</sub> ] <sub>2019</sub>	4.9×10 <sup>6</sup>	3.5×10 <sup>6</sup>	3.9×10 <sup>6</sup>	2.5×10 <sup>6</sup>	6.3×10 <sup>6</sup>
	Proxy <sub>OH,CS</sub>	6.0×10 <sup>6</sup>	8.2×10 <sup>6</sup>	2.8×10 <sup>6</sup>	1.5×10 <sup>6</sup>	6.3×10 <sup>6</sup>
3-Year	[H <sub>2</sub> SO <sub>4</sub> ] <sub>3-Year</sub>	4.4×10 <sup>6</sup>	3.3×10 <sup>6</sup>	3.5×10 <sup>6</sup>	2.1×10 <sup>6</sup>	5.8×10 <sup>6</sup>
	Proxy <sub>UVB,CS</sub>	5.1×10 <sup>6</sup>	5.5×10 <sup>6</sup>	3.6×10 <sup>6</sup>	1.5×10 <sup>6</sup>	6.9×10 <sup>6</sup>
	Proxy <sub>UVB,PM<sub>2.5</sub></sub>	4.5×10 <sup>6</sup>	4.7×10 <sup>6</sup>	3.2×10 <sup>6</sup>	1.3×10 <sup>6</sup>	6.1×10 <sup>6</sup>

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**Table S5.** Performance statistics for the comparison between simulated (SIM) and observed (OBS) concentrations of NO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub>, and PM<sub>2.5</sub> at BUCT site during the study period (2019.1.1 – 2020.3.15).

Variables	NO <sub>2</sub> (ppb)	SO <sub>2</sub> (ppb)	PM <sub>2.5</sub> (μg/m <sup>3</sup> )	O <sub>3</sub> (ppb)
Mean OBS	19.5	1.6	27.7	45.2
Mean SIM	17.6	1.9	28.6	40.5
Normalized Mean Bias	-10%	33%	3%	-11%
Normalized Mean Error	63%	88%	66%	57%

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**Table S6.** Performance statistics for the comparison between simulated (SIM) and observed (OBS) concentrations of NO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub>, and PM<sub>2.5</sub> at national monitoring sites in China in 2019.

Variables	NO <sub>2</sub> (ppb)	SO <sub>2</sub> (ppb)	PM <sub>2.5</sub> (μg/m <sup>3</sup> )	O <sub>3</sub> (ppb)
Mean OBS (μg/m <sup>3</sup> )	29.3	11.3	39.2	93.3
Mean SIM (μg/m <sup>3</sup> )	26.2	8.5	34.3	92.9
Normalized Mean Bias	-11%	-25%	-12%	0%
Normalized Mean Error	20%	27%	20%	16%

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**Table S7.** Performance statistics for the comparison between simulated (SIM) and observed (OBS) concentrations of NO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub>, and PM<sub>2.5</sub> at national monitoring sites in China in 2020.

Variables	NO <sub>2</sub> (ppb)	SO <sub>2</sub> (ppb)	PM <sub>2.5</sub> (μg/m <sup>3</sup> )	O <sub>3</sub> (ppb)
Mean OBS (μg/m <sup>3</sup> )	26.0	10.0	34.5	91.6
Mean SIM (μg/m <sup>3</sup> )	21.7	6.9	31.0	83.9
Normalized Mean Bias	-17%	-31%	-10%	-8%
Normalized Mean Error	28%	38%	22%	19%

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**Table S8.** Monthly concentration of SO<sub>2</sub> (ppb) during daytime (10:00-14:00) from 2019 to 2021. “NaN” means there is no data available.

Month	2019			2020			2021			Annual decline using median value / %
	Median	25th	75th	Median	25th	75th	Median	25th	75th	
January	3.00	1.51	5.71	3.63	1.94	5.15	1.50	0.94	2.66	-24.9
February	2.07	0.95	4.41	1.75	0.51	3.20	2.03	1.17	3.00	-1.0
March	3.61	0.78	5.40	1.21	0.45	2.67	1.07	0.63	1.90	-35.2
April	2.08	0.79	3.54	1.88	0.47	3.94	1.02	0.36	2.08	-25.4
May	2.18	0.55	3.22	2.36	0.92	3.15	0.51	0.24	1.07	-38.3
June	1.88	0.70	3.10	0.58	0.29	1.91	0.30	0.16	0.60	-41.9
July	0.25	0.16	0.79	0.22	0.07	0.48	0.08	0.06	0.10	-34.7
August	0.36	0.28	0.87	NaN	NaN	NaN	0.23	0.18	0.32	-18.9
September	1.05	0.79	1.33	0.40	0.22	0.73	0.27	0.11	0.40	-37.0
October	0.77	0.31	1.70	0.68	0.46	1.49	0.35	0.25	0.59	-27.2
November	1.43	0.55	2.43	1.46	0.45	2.85	0.32	0.26	0.58	-39.0
December	1.46	0.75	2.30	1.75	0.98	2.66	NaN	NaN	NaN	19.3
Average										-25.4

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227 **References**

- 228 Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm, J., Mäkelä, J. M., Hoell, C., O’ Dowd, C. D., Hansson, H.-C., Väkevä, M.,  
229 Koponen, I. K., Buzorius, G., and Kulmala, M.: Physical characterization of aerosol particles during nucleation events, *Tellus*  
230 *B: Chemical and Physical Meteorology*, 53, 344-358, 10.3402/tellusb.v53i4.17127, 2001.
- 231 Guo, Y., Yan, C., Li, C., Ma, W., Feng, Z., Zhou, Y., Lin, Z., Dada, L., Stolzenburg, D., Yin, R., Kontkanen, J., Daellenbach,  
232 K. R., Kangasluoma, J., Yao, L., Chu, B., Wang, Y., Cai, R., Bianchi, F., Liu, Y., and Kulmala, M.: Formation of nighttime  
233 sulfuric acid from the ozonolysis of alkenes in Beijing, *Atmospheric Chemistry and Physics*, 21, 5499-5511, 10.5194/acp-21-  
234 5499-2021, 2021.
- 235 Hari, P., and Kulmala, M.: Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II), *Boreal Environment*  
236 *Research*, 10, 315-322, 2005.
- 237 Hari, P., Nikinmaa, E., Pohja, T., Siivola, E., Bäck, J., Vesala, T., and Kulmala, M.: Station for Measuring Ecosystem-  
238 Atmosphere Relations: SMEAR, in: *Physical and Physiological Forest Ecology*, edited by: Hari, P., Heliövaara, K., and  
239 Kulmala, L., Springer Netherlands, Dordrecht, 471-487, 2013.
- 240 Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X., Xu, Z., Chen, L., Li,  
241 Y., Che, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q., Liu, B., Chai, F., Davis, S. J., Zhang, Q., and  
242 He, K.: Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China, *Natl Sci*  
243 *Rev*, 8, nwaal37, 10.1093/nsr/nwaa137, 2021.
- 244 Kerminen, V.-M., Pirjola, L., and Kulmala, M.: How significantly does coagulation limit atmospheric particle  
245 production?, *Journal of Geophysical Research: Atmospheres*, 106, 24119-24125, <https://doi.org/10.1029/2001JD000322>, 2001.  
246 CUV5 and SUV Series Ultraviolet Radiometers: <https://www.kippzonen.com/Product/363/SUV5-Total-UV-Radiometer>.
- 247 Kulmala, M., Maso, M. D., Mäkelä, J. M., Pirjola, L., Väkevä, M., Aalto, P., Mikkilainen, P., Hämeri, K., and O’ Dowd, C.  
248 D.: On the formation, growth and composition of nucleation mode particles, *Tellus B: Chemical and Physical Meteorology*, 53,  
249 479-490, 10.3402/tellusb.v53i4.16622, 2001.
- 250 Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H. E., Lehtipalo, K., Dal Maso, M., Aalto, P. P., Junninen, H.,  
251 Paasonen, P., Riipinen, I., Lehtinen, K. E. J., Laaksonen, A., and Kerminen, V.-M.: Measurement of the nucleation of  
252 atmospheric aerosol particles, *Nature Protocols*, 7, 1651-1667, 10.1038/nprot.2012.091, 2012.
- 253 Ma, X., Tan, Z., Lu, K., Yang, X., Liu, Y., Li, S., Li, X., Chen, S., Novelli, A., Cho, C., Zeng, L., Wahner, A., and Zhang, Y.:  
254 Winter photochemistry in Beijing: Observation and model simulation of OH and HO<sub>2</sub> radicals at an urban site, *Science of The*  
255 *Total Environment*, 685, 85-95, <https://doi.org/10.1016/j.scitotenv.2019.05.329>, 2019.
- 256 Ma, X., Tan, Z., Lu, K., Yang, X., Chen, X., Wang, H., Chen, S., Fang, X., Li, S., Li, X., Liu, J., Liu, Y., Lou, S., Qiu, W.,  
257 Wang, H., Zeng, L., and Zhang, Y.: OH and HO<sub>2</sub> radical chemistry at a suburban site during the EXPLORE-YRD campaign  
258 in 2018, *Atmospheric Chemistry and Physics*, 22, 7005-7028, 10.5194/acp-22-7005-2022, 2022.
- 259 Nie, W., Yan, C., Huang, D. D., Wang, Z., Liu, Y., Qiao, X., Guo, Y., Tian, L., Zheng, P., Xu, Z., Li, Y., Xu, Z., Qi, X., Sun,  
260 P., Wang, J., Zheng, F., Li, X., Yin, R., Dallenbach, K. R., Bianchi, F., Petäjä, T., Zhang, Y., Wang, M., Schervish, M., Wang,  
261 S., Qiao, L., Wang, Q., Zhou, M., Wang, H., Yu, C., Yao, D., Guo, H., Ye, P., Lee, S., Li, Y. J., Liu, Y., Chi, X., Kerminen,  
262 V.-M., Ehn, M., Donahue, N. M., Wang, T., Huang, C., Kulmala, M., Worsnop, D., Jiang, J., and Ding, A.: Secondary organic  
263 aerosol formed by condensing anthropogenic vapours over China’s megacities, *Nature Geoscience*, 15, 255-261,  
264 10.1038/s41561-022-00922-5, 2022.
- 265 Tan, Z., Fuchs, H., Lu, K., Hofzumahaus, A., Bohn, B., Broch, S., Dong, H., Gomm, S., Häsel, R., He, L., Holland, F., Li,  
266 X., Liu, Y., Lu, S., Rohrer, F., Shao, M., Wang, B., Wang, M., Wu, Y., Zeng, L., Zhang, Y., Wahner, A., and Zhang, Y.:  
267 Radical chemistry at a rural site (Wangdu) in the North China Plain: Observation and model calculations of OH, HO<sub>2</sub> and RO<sub>2</sub>  
268 radicals, *Atmospheric Chemistry and Physics*, 17, 663-690, 10.5194/acp-17-663-2017, 2017.
- 269 TEOM 1405-D Ambient Particulate Monitor: [https://www.thermofisher.cn/order/catalog/product/TEOM1405DF?SID=srch-](https://www.thermofisher.cn/order/catalog/product/TEOM1405DF?SID=srch-srp-TEOM1405DF)  
270 [srp-TEOM1405DF](https://www.thermofisher.cn/order/catalog/product/TEOM1405DF).
- 271 Thermo Scientific Model 43i-TLE: <https://www.thermofisher.cn/order/catalog/product/43ITLE?SID=srch-hj-43i-TLE>.
- 272 Relative Humidity and Temperature Probe HMP4: [https://www.vaisala.com/en/products/instruments-sensors-and-other-](https://www.vaisala.com/en/products/instruments-sensors-and-other-measurement-devices/instruments-industrial-measurements/hmp4)  
273 [measurement-devices/instruments-industrial-measurements/hmp4](https://www.vaisala.com/en/products/instruments-sensors-and-other-measurement-devices/instruments-industrial-measurements/hmp4).

274 Yan, C., Shen, Y., Stolzenburg, D., Dada, L., Qi, X., Hakala, S., Sundström, A. M., Guo, Y., Lipponen, A., Kokkonen, T.,  
275 Kontkanen, J., Cai, R., Cai, J., Chan, T., Chen, L., Chu, B., Deng, C., Du, W., Fan, X., He, X. C., Kangasluoma, J., Kujansuu,  
276 J., Kurppa, M., Li, C., Li, Y., Lin, Z., Liu, Y., Liu, Y., Lu, Y., Nie, W., Pulliainen, J., Qiao, X., Wang, Y., Wen, Y., Wu, Y.,  
277 Yang, G., Yao, L., Yin, R., Zhang, G., Zhang, S., Zheng, F., Zhou, Y., Arola, A., Tamminen, J., Paasonen, P., Sun, Y., Wang,  
278 L., Donahue, N. M., Liu, Y., Bianchi, F., Daellenbach, K. R., Worsnop, D. R., Kerminen, V. M., Petäjä, T., Ding, A., Jiang, J.,  
279 and Kulmala, M.: The effect of COVID-19 restrictions on atmospheric new particle formation in Beijing, *Atmos. Chem. Phys.*  
280 *Discuss.*, 2022, 1-24, 10.5194/acp-2021-1079, 2022.  
281 Yang, L., Nie, W., Liu, Y., Xu, Z., Xiao, M., Qi, X., Li, Y., Wang, R., Zou, J., Paasonen, P., Yan, C., Xu, Z., Wang, J., Zhou,  
282 C., Yuan, J., Sun, J., Chi, X., Kerminen, V.-M., Kulmala, M., and Ding, A.: Toward building a physical proxy for gas-phase  
283 sulfuric acid concentration based on its budget analysis in polluted Yangtze River Delta, East China, *Environmental Science*  
284 *& Technology*, 55, 6665-6676, 10.1021/acs.est.1c00738, 2021.  
285 Zhang, R., Zhang, Y., Lin, H., Feng, X., Fu, T.-M., and Wang, Y.: NO<sub>x</sub> Emission Reduction and Recovery during COVID-19  
286 in East China, *Atmosphere*, 11, 10.3390/atmos11040433, 2020.  
287