

1 *Supporting Information of*

2 **Nocturnal production of N<sub>2</sub>O<sub>5</sub> and ClNO<sub>2</sub> in Delhi: driving factors and impacts**

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25 **This PDF file includes:**

26 Text S1 to S3

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30

31 **Supplementary Text**

32 **Text. S1 Details of field measurements in Delhi**

33 **1.1 FIGAERO-I-CIMS configurations**

34 The reagent ions ( $I^-$  and  $I\cdot H_2O^-$ ) were produced by passing  $\sim 2$  lpm ultrahigh purity  
35 (UHP)  $N_2$  via a  $CH_3I$  permeation tube and through a Po-210 ion source, where it  
36 interacted with the sampled air or thermally desorbed molecules from  $PM_{2.5}$  in the ion-  
37 molecular reactor (IMR). A constant 10 sccm of  $H_2O$ -saturated UHP was added directly  
38 into the IMR to reduce the sensitivity variability with ambient RH. The IMR pressure  
39 was maintained at 200-250 mbar during gas sampling and 120-140 mbar during  $PM_{2.5}$   
40 desorption. The relatively lower pressure during the  $PM_{2.5}$  measurement was likely  
41 caused by the resistance and associated pressure drop across the filters. Considering the  
42 differences in IMR pressures, we conducted sensitivity calibrations for the gas- and  
43 particle-phase measurements separately under conditions similar to the field  
44 observations in Delhi. The average mass resolution was  $\sim 3500$  at  $m/z$  127.

45 For the gas-phase measurements, ambient air was drawn at 3 lpm through a 4-m-  
46 long PFA tube (inner diameter, 5 mm, residence time 1.6 s) with 2 lpm pumped away,  
47 and the remaining 1 lpm was diluted with 1 lpm UHP  $N_2$  before entering the IMR to  
48 avoid reagent ions titration by high levels of pollutants (e.g.,  $HNO_3$ ) in the ambient air.  
49 The inlet was flushed with  $\sim 2$  lpm UHP  $N_2$  during the first and last two minutes of each  
50 20-min gas-phase measurement to obtain background signals, while ambient sampling  
51 was performed during the remaining period. The signals from the second zeroing period  
52 were used as the background, since those from the first zeroing period may have been  
53 influenced by carryover desorption signals (especially for sticky compounds) from the  
54 preceding particle-phase measurement (Fig. S5). The background-subtracted and  
55 dilution ratio (a factor of 2) corrected ion signals were subsequently averaged to 1-hour  
56 intervals. For  $PM_{2.5}$  measurements, ambient air was drawn at 3 lpm through a 4-m-long  
57 copper tube (inner diameter, 5 mm) fitted with a  $PM_{2.5}$  cyclone during the gas-sampling  
58 period, with 2 lpm discarded and 1 lpm collected on a Teflon filter for 20 min. The filter  
59 was then moved to the desorption position, where it was thermally desorbed and carried  
60 into the IMR under a 2 lpm flow of UHP  $N_2$ . The  $N_2$  flow was gradually ramped from  
61 room temperature to 200 °C in 20 min, soaked at 200 °C for 20 min, and finally cooled  
62 back to room temperature in 10 min. The particle phase signal was integrated over the  
63 temperature ramping and soaking period during filter desorption. The baseline was  
64 determined from linear fitting of the signal between the onset of heating and the end of  
65 soaking and was integrated and subtracted from the integrated desorption signals.

66 **1.2. Sensitivity calibrations of  $N_2O_5$ ,  $CINO_2$  and related species**

67  $N_2O_5$  and  $CINO_2$ .  $N_2O_5$  was prepared by mixing  $O_3$  with excessive  $NO_2$  to promote

68 the conversion of  $\text{NO}_3$  to  $\text{N}_2\text{O}_5$ . The generated  $\text{N}_2\text{O}_5$  concentrations were determined by  
69 the change in  $\text{O}_3$  following the addition of  $\text{NO}_2$ , and different  $\text{N}_2\text{O}_5$  levels was achieved  
70 via varying  $\text{O}_3$  concentration (Bertram et al., 2009). The produced  $\text{N}_2\text{O}_5$  was further  
71 diluted with humidified zero air to approximate Delhi RH conditions before entering  
72 the CIMS.  $\text{ClNO}_2$  was produced by passing a known amount of  $\text{N}_2\text{O}_5$  through a wetted  
73  $\text{NaCl}$ -slurry placed in a Teflon tube, assuming a unit conversion from  $\text{N}_2\text{O}_5$  to  $\text{ClNO}_2$   
74 (Finlayson-Pitts, 2003). All the tubings are covered by aluminum foil to avoid  $\text{NO}_3$   
75 photolysis and were flushed with dry zero air overnight before calibration to minimize  
76 potential  $\text{N}_2\text{O}_5$  hydrolysis on the tubing surfaces. No water-dependent sensitivity  
77 correction was applied for  $\text{N}_2\text{O}_5$  and  $\text{ClNO}_2$ , as no significant sensitivity variation was  
78 observed in the range of  $\text{H}_2\text{O}\cdot\text{I}^-/\text{I}^-$  ratios during the field measurements in Delhi. The  
79 calibration curve for  $\text{N}_2\text{O}_5$  and  $\text{ClNO}_2$  are shown in **Fig. S19a-b**.

80 *HCl and chloroacetic acid.* A certified 10 ppm HCl gas cylinder (Linde Specialgas)  
81 was used as the standard source, which was diluted with zero air to different mixing  
82 ratios before introducing to the CIMS (**Fig. S19c**). Notably, we observed a significant  
83 decrease of the HCl sensitivity at high  $\text{H}_2\text{O}\cdot\text{I}^-/\text{I}^-$  ratios (**Fig. S19d**), likely due to the  
84 competing effects of  $\text{H}_2\text{O}$  for clustering with iodide anion and that the reaction of HCl  
85 with  $\text{H}_2\text{O}\cdot\text{I}^-$  is thermodynamically less favorable compared to reacting with iodide  
86 anion. The slight increase in HCl sensitivity with increasing  $\text{H}_2\text{O}\cdot\text{I}^-/\text{I}^-$  ratio at low RH  
87 levels may be related to the stabilization of  $\text{HCl}\cdot\text{I}^-$  clusters by water vapors. Similar  
88 patterns have been reported for other acids in a previous study (Lee et al., 2014).  
89 Gaseous chloroacetic acid was generated by placing solid chloroacetic acid in a glass  
90 vial submerged in a 30 °C water bath. The emission rate, determined by gravimetric  
91 weight loss analysis, was 177.3 ng/min. No significant  $\text{H}_2\text{O}$  dependence of chloroacetic  
92 acid sensitivity was observed.

93 *Levoglucosan.* A certain amount (10~90 ng) of levoglucosan dissolved in acetone  
94 solutions was deposited on the filter of the CIMS, and then the droplet underwent the  
95 same thermal desorption cycle as in the field measurements. The sensitivity for  
96 levoglucosan was derived as the integrated background-subtracted signals divided by  
97 the deposited mass.

98 **1.3. Supporting measurements and data quality control**

99 Meteorological factors, i.e., T, RH, wind speed, and wind direction, were measured  
100 by an automated weather station (AWS) (Davis Vantage Pro 2, Davis Instruments  
101 Corporation, USA). Trace gases, including  $\text{O}_3$ , NO and  $\text{NO}_2$ , CO, and  $\text{SO}_2$  were  
102 measured by on-line gas analyzers (ECOTECH Serinus). For the long-term (2017~2024)  
103 hourly observation data obtained from R.K. Puram station, we applied rigorous quality

104 controls following recent studies (Vohra et al., 2025;Xie et al., 2024) with a few  
 105 modifications, including (1) Remove duplicates: In sequences of five or more  
 106 consecutive identical hourly values, only the first value out of the sequence is retained,  
 107 (2) Remove outliers: For a 24-h running window, flag any observation as an outlier if  
 108 its absolute difference from the median exceeds three times the Median Absolute  
 109 Deviation (MAD, median distance between each observation and the median of all  
 110 observations), (3) Remove constant data: In a 24-h running window, remove constant  
 111 data values with coefficient of variation (ratio of standard deviation to the mean) less  
 112 than 5%, (4) Validation of NO, NO<sub>2</sub> measurements by comparing with the measured  
 113 NO<sub>x</sub>: consider NO and NO<sub>2</sub> are reported correctly in unit of  $\mu\text{g}/\text{m}^3$  and assess if NO<sub>x</sub>  
 114 calculated from these is within 2% of reported NO<sub>x</sub> + 2.5 ppb. If not, the measured NO,  
 115 NO<sub>2</sub>, and NO<sub>x</sub> data were omitted.

## 116 **Text. S2 Estimation of the N<sub>2</sub>O<sub>5</sub> uptake coefficient**

### 117 **2.1 Inapplicability of the steady-state method**

118 The steady-state approximation of the NO<sub>2</sub>-NO<sub>3</sub>-N<sub>2</sub>O<sub>5</sub> system is a widely applied  
 119 method (Brown et al., 2003) for estimating the values for the NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> sinks  
 120 (thereby  $\gamma_{N_2O_5}$ ) in various atmospheric environments (Brown et al., 2006; Wang et al.,  
 121 2017b; Wang et al., 2017a). This method assumes that production rate of NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub>  
 122 equals the sum of the loss rates during a certain period of the night (Eq. 1), and therefore  
 123  $k_{NO_3}$  and  $k_{N_2O_5}$  can be determined as the slope and intercept, respectively, of the  
 124 linear fitting between  $\tau_{N_2O_5}^{-1}$  and  $\frac{1}{K_{eq}[NO_2]}$  (Eq. 2). However, we found no  
 125 significant positive even negative correlation between  $\tau_{N_2O_5}^{-1}$  and  $\frac{1}{K_{eq}[NO_2]}$  during  
 126 the nighttime throughout the campaign (an example is shown in **Fig. S20**). This is likely  
 127 attributed to the intense and variable NO<sub>x</sub> emissions in Delhi (avg. nighttime hourly  
 128 NO<sub>x</sub> mixing ratios of  $92 \pm 60$  ppb) which preclude the system from approaching steady-  
 129 state on the time-scale of a night. As is shown in **Fig. 2a** of the main text, N<sub>2</sub>O<sub>5</sub>  
 130 presented a strong negative dependence on NO. The high concentrations and large  
 131 fluctuations of NO dominated the variation of N<sub>2</sub>O<sub>5</sub>, thereby decoupling the lifetime of  
 132 N<sub>2</sub>O<sub>5</sub> from  $\frac{1}{K_{eq}[NO_2]}$ .

$$133 \quad k_{NO_2+O_3}[NO_2][O_3] = k_{NO_3}[NO_3] + k_{N_2O_5}[N_2O_5] \quad (Eq. 1)$$

$$134 \quad K_{eq} = 2.7 \times 10^{-27} \times \exp(11000/T)$$

135 Substitute  $[NO_3] = \frac{[N_2O_5]}{K_{eq}[NO_2]}$  to Eq. 1, we get

136 
$$\tau_{N_2O_5}^{-1} = \frac{k_{NO_2+O_3}[NO_2][O_3]}{[N_2O_5]} = \frac{k_{NO_3}}{K_{eq}[NO_2]} + k_{N_2O_5} \quad (Eq. 2)$$

137 Where  $k_{NO_3}$  and  $k_{N_2O_5}$  are the pseudo-first-order loss rate constants of  $NO_3$  and  $N_2O_5$ ,  
 138 respectively,  $k_{N_2O_5} = \frac{\bar{c}\gamma}{4} S_a$ ,  $\tau_{N_2O_5}^{-1}$  is the inverse of the  $N_2O_5$  steady-state liftime.

139 **2.2 Validation of the RH parameterization method**

140 Cl $NO_2$  yield indicates the branching ratio of  $H_2ONO_2^+$  reacting with  $H_2O$  (R4) and  
 141 chloride (R5), which is theoretically determined by the respective reaction rates shown  
 142 in Eq. 3(Bertram and Thornton, 2009).

143 
$$f_{ClNO_2} = \frac{k_{R5}[H_2ONO_2^+][Cl^-]}{k_{R5}[H_2ONO_2^+][Cl^-] + k_{R4}[H_2ONO_2^+][H_2O]} = \frac{1}{1 + \frac{k_{R4}}{k_{R5}} \frac{[H_2O]}{[Cl^-]}} \quad (Eq. 3)$$

144 Where  $[H_2O]$  and  $[Cl^-]$  are concentrations in the aqueous phase of particles derived  
 145 from thermodynamic models,  $k_{R5}$  and  $k_{R4}$  are reaction rate coefficients of Reaction  
 146 R5 and R4. The experimentally determined values for  $\frac{k_{R5}}{k_{R4}}$  is  $483 \pm 175$  from the work  
 147 of Bertram and Thornton(Bertram and Thornton, 2009) and  $836 \pm 32$  from Behnke et  
 148 al(Behnke et al., 1997). Due to the lack of aerosol inorganic composition (e.g.,  $NH_4^+$ ,  
 149  $NO_3^-$ , and  $SO_4^{2-}$ ) measurements during the 2023 campaign, we estimated the range of  
 150  $[Cl^-]/[H_2O]$  ratio from January to March in 2022 Delhi when ACSM measurements are  
 151 available(Ali et al., 2025) and the measured chloride level was similar to the campaign  
 152 in 2023. The estimated nocturnal (18:00~06:00)  $[Cl^-]/[H_2O]$  ratio ranged from  $1.8 \times 10^-5$   
 153 to 0.76 with an average of 0.07, which mostly fell in the region where the  
 154 corresponding Cl $NO_2$  yield close to 1 (**Fig. S13a**). We therefore tested the validity of  
 155 the RH parameterization method by simulating the average nocturnal variation of  
 156 Cl $NO_2$  constrained with the parameterized gamma, observed  $N_2O_5$  mixing ratios and  
 157 aerosol surface area concentrations, with the Cl $NO_2$  yield set to 1. The results showed  
 158 that the simulation largely overestimated the production of Cl $NO_2$  from 20:00 to 00:00  
 159 (**Fig. S13b**) when the absolute chloride concentrations were relatively low (**Fig. 1d**).  
 160 By comparison, when constraining Cl $NO_2$  yield to 0 for the chloride-deficient period  
 161 (20:00 to 00:00) and 1 for the chloride-sufficient period (0:00 to 6:00), the modelled  
 162 Cl $NO_2$  evolution well tracked the observations (**Fig. S13b**). The overall consistency  
 163 between modelling and observations suggest that the measured Cl $NO_2$  can largely  
 164 explained by the locally heterogeneous uptake of  $N_2O_5$  on chloride-rich particles in  
 165 Delhi and that the RH-parameterized  $\gamma_{N_2O_5}$  is a reasonable estimate, indicating the key  
 166 role of RH and thereby aerosol liquid water content in driving  $N_2O_5$  uptake in Delhi.  
 167 Nevertheless, comprehensive aerosol composition measurements or direct

168 measurements of N<sub>2</sub>O<sub>5</sub> reactivity (Bertram et al., 2009; Li et al., 2025) in future studies  
169 are required to enable better quantification and understanding N<sub>2</sub>O<sub>5</sub> uptake processes  
170 in Delhi.

171 **Text. S3 Estimation of ClNO<sub>2</sub> in the residual layer**

172 The nocturnal boundary layer typically comprised a surface layer and an overlying  
173 residual layer (RL), where the RL was isolated from ground emissions and preserved  
174 the chemical composition of the well-mixed boundary layer from the late-afternoon. To  
175 estimate the maximum ClNO<sub>2</sub> produced in the RL, we used the ground-observed  
176 average N<sub>2</sub>O<sub>5</sub> mixing ratios and aerosol surface area prior to sunset (16:00~18:00),  
177 assuming  $\gamma_{N_2O_5}=0.1$  and  $f_{ClNO_2}=1$  (Eq. 4). The calculation indicated that N<sub>2</sub>O<sub>5</sub> was  
178 fully consumed within 1 hour.

$$179 [ClNO_2]_{RL} = \int_{t1}^{t2} \frac{\bar{c}\gamma_{N_2O_5}}{4} S_a [N_2O_5]_{obs} f_{ClNO_2} dt \quad (\text{Eq. 4})$$

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197 **Table S1 Summary of nocturnal peak and average (shown in the brackets) N<sub>2</sub>O<sub>5</sub>**  
 198 **and CINO<sub>2</sub> mixing ratios measured around the world. The unit is ppt.**

Location	Period	Type	N <sub>2</sub> O <sub>5</sub>	CINO <sub>2</sub>	ref.
India Institute of Technology, Delhi	2023.2.23-3.14	urban inland	556 (16)	1340 (116)	this study
India Institute of Technology, Delhi	2019.1.11-2.5	urban inland	203 (4)	724 (48)	(Haslett et al., 2023)
Gulf of Mexico, Huston, Texas	2006.7-9	urban coastal	750	1200	(Osthoff et al., 2008)
National Oceanic and Atmospheric Administration, Boulder, Colorado	2009.2.11-2.25	urban inland	1500	440	(Thornton et al., 2010)
University of Utah, Salt Lake City, Utah	2015.12-2016.2	urban inland	1520 (76)		(Baasandorj et al., 2017)
University of Michigan, Ann Arbor, Michigan	2016.2.1-3.10	urban inland		220	(McNamara et al., 2020)
Toronto	2021.1.11-1.25	urban inland		300	(Wang et al., 2023)
Kensington	2012.7 -8	urban inland	1700	724 (84)	(Bannan et al., 2015)
Manchester	2014.10-11	urban inland		506	(Priestley et al., 2018)
Frankfurt Observatory (825 m a.s.l.)	2011.8-9	rural inland	3000	800	(Phillips et al., 2012)
China, Wangdu	2014.6-7	rural inland	500	2070	(Tham et al., 2016)
China, Wangdu	2017.12	rural inland	1000	1400	(Xia et al., 2021)
China, Wangdu	2023.2.10-3.5	rural inland		3600	(Chen et al., 2025)
China, mountain in Hong Kong (957 a.s.l.)	2013.11-12	mountain site	7700	4700	(Wang et al., 2016)
China, Heshan	2017.1.2-1.15	urban coastal	3000	8300	(Chen et al., 2023)

200 **Table S2 Campaign-averaged  $\text{NO}_3\cdot$  reactivity (unit:  $\text{s}^{-1}$ ) during the 2019 and 2023**  
 201 **campaign.**

	$\text{NO}_3 + \text{NO}$	$\text{NO}_3 + \text{hv}$	$\text{N}_2\text{O}_5$ hete.	$\text{NO}_3 + \text{VOCs}$
2019.1~2	50.175	0.031	0.462	0.081
2023.2~3	15.585	0.043	0.091	0.081*

202 \*assumed to be the same as those observed in 2019.

203 **Table S3 Average emission factor (g/kg fuel burned) of K, HCl, HCN, CO, and  $\text{SO}_2$**   
 204 **from biomass, biofuel, and garbage burning processes and vehicles.**

	K	HCl	HCN	$\text{SO}_2$	CO	HCl/CO	ref
Peat	0.004~0.0 15	-	1.00	-	38	-	(Andrea e, 2019)
Crop residues	0.48	0.18	0.42	0.8	77	$2.3 \times 10^{-3}$	(Andrea e, 2019)
Rice straw	-	0.44	0.37	1.3	60	$7.3 \times 10^{-3}$	(Stockw ell et al., 2014)
Biomass burning							
Wheat straw	-	0.47	0.10	0.7	39	$1.2 \times 10^{-2}$	(Stockw ell et al., 2014)
Tropical forest	0.32	0.13	0.44	0.8	104	$1.3 \times 10^{-3}$	(Andrea e, 2019)
Temperate forest	0.18	0.04	0.64	0.7	112	$3.5 \times 10^{-4}$	(Andrea e, 2019)
Dung	0.09	0.04	1.30	0.7	89	$4.3 \times 10^{-4}$	(Andrea e, 2019)
Biofuels without dung	0.13	0.08	0.39	0.5	84	$8.9 \times 10^{-4}$	(Andrea e, 2019)
Biofuel burning							
Charcoal	0.75	0.11	-	0.6	207	$5.3 \times 10^{-4}$	(Andrea e, 2019)
Wood	-	0.02	0.54	bld	77	$2.4 \times 10^{-4}$	(Stockw ell et al., 2016)

Garbage burning	Garbage burning	0.02	2.80	0.43	0.5	66	$4.2 \times 10^{-2}$	(Andrea e, 2019)
	Mixed plastic garbage	-	2.30	0.43	bld	85	$2.7 \times 10^{-2}$	(Stockw ell et al., 2016)
	Two- wheeled vehicles	-	bld	0.68	bld	761	-	(Stockw ell et al., 2016)
Vehicles	Agricultur al diesel irrigation pumps	-	bld	0.12	bld	17	-	(Stockw ell et al., 2016)

205 Note: bld indicates below the limit of detection.

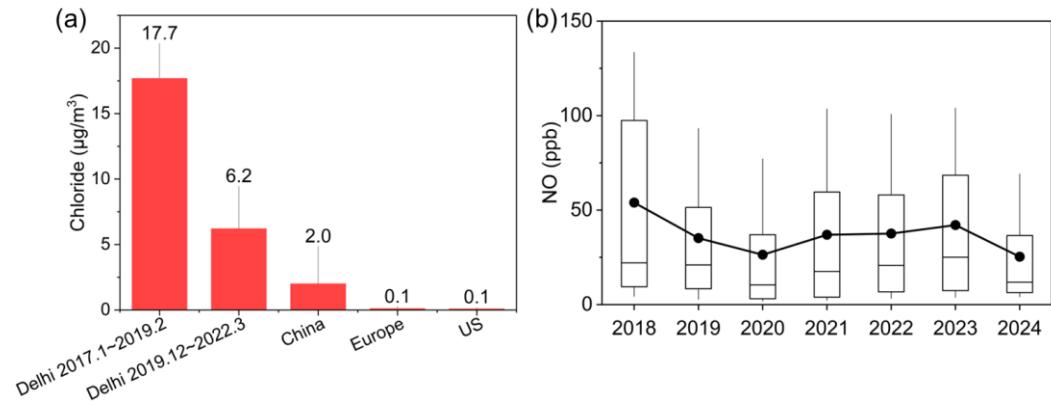
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207 **Table S4 Summary of the bulk PM composition measured at IITD during winter**  
208 **from 2017 to 2022. The unit is  $\mu\text{g}/\text{m}^3$ .**

Period	PM	Cl	$\text{NO}_3$	$\text{SO}_4$	$\text{NH}_4$	Org	ref
2017.1~2017.3	$\text{PM}_1$	14.2	16.1	15.8	15.7	81.0	(Gani et al., 2019)
2017.12~2018.3	$\text{PM}_1$	18.9	19.6	11.1	15.9	99.9	(Gani et al., 2019)
2018.1.17~1.19 2018.2.5~3.11	$\text{PM}_{2.5}$	20.4	13.3	12.0	14.3	65.3	(Lalchandani et al., 2021)
2019.1.11~2.5	$\text{PM}_1$	17.3	20.8	18.0	23.7	80.0	(Haslett et al., 2023)
2019.12.1~2020.1.5	$\text{PM}_{2.5}$	5.7	14.5	11.4	9.5	60.9	(Ali et al., 2023)
2020.2.1~3.20	$\text{PM}_1$	4.9	10.6	9.7	8.9	47.9	(Mandariya et al., 2024)
2020.12.15~12.31	$\text{PM}_{2.5}$	11.3	27.2	13.7	25.7	111.5	(Faisal et al., 2025)

2020.12.15~2021.2.23	PM <sub>2.5</sub>	4.9	19.8	15.5	15.7	96.2	(Ali et al., 2023)
2021.1.1~2021.2.28	PM <sub>2.5</sub>	10.1	16.1	12.1	19.0	87.0	(Faisal et al., 2025)
2022.1.12~3.31	PM <sub>2.5</sub>	4.5	15.3	14.0	12.9	86.2	(Ali et al., 2025)

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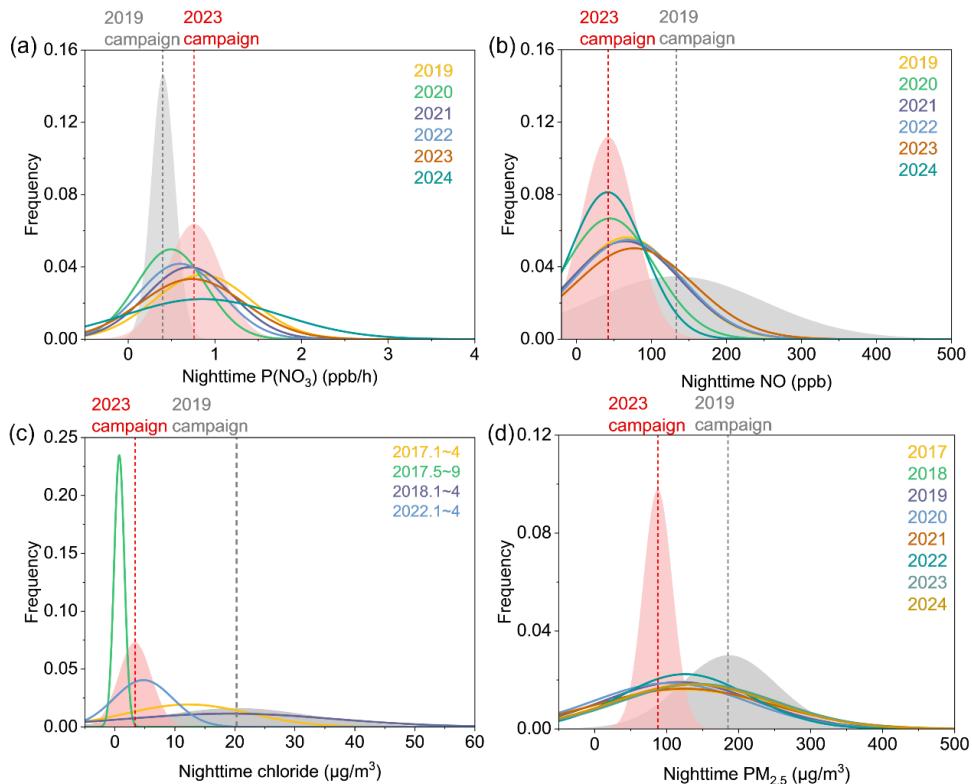
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212 **Fig. S1 (a) Comparison of the observed chloride levels globally. (b) Box plots of**  
213 **the annual daily averages of NO concentrations in Delhi.** The chloride  
214 concentrations in Delhi (compiled from Table S4), China (Xia et al., 2021;Tham et al.,  
215 2018;Xia et al., 2020), Europe (Eger et al., 2019;Lanz et al., 2010), and US (Mielke et  
216 al., 2013;Sarwar et al., 2012) are campaign-average values retrieved from previous  
217 studies. The whiskers in (a) represent standard deviation. The NO concentrations shown  
218 in (b) were obtained from the R.K. Puram monitoring station. The boxes represent the  
219 interquartile range with the whiskers extending to the 10<sup>th</sup> and 90<sup>th</sup> percentiles. The dot  
220 and horizontal line within each box denote the mean and median values, respectively.

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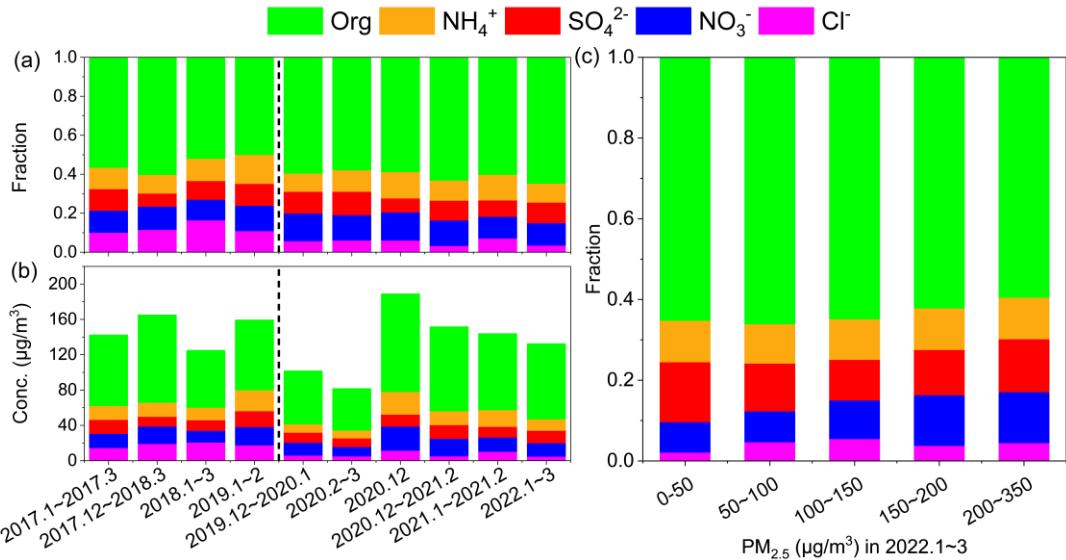
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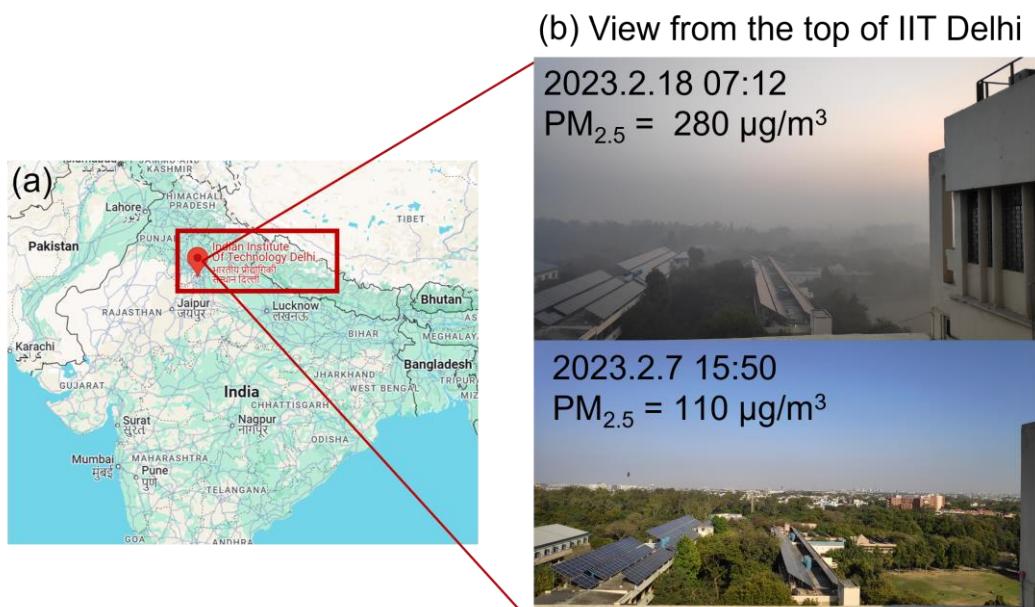
225 **Fig. S2 Representativeness of the two field-campaigns in Delhi.** Gaussian-fitted  
 226 frequency distributions of nightly (a)  $P(NO_3)$ , (b) NO, (c) chloride, and (d)  $PM_{2.5}$   
 227 concentrations from 2017 to 2024. The nighttime is defined as 20:00~04:00 throughout  
 228 the years. The data for  $P(NO_3)$ , NO, and  $PM_{2.5}$ , are obtained from the R.K. Puram  
 229 monitoring station. Chloride concentrations were unavailable at the station and are  
 230 retrieved from previous studies (Gani et al., 2019; Ali et al., 2025). The gray and red  
 231 shaded area denote the respective pollutant distribution during the 2019 and 2023  
 232 campaign and the dashed lines indicate the campaign averages.

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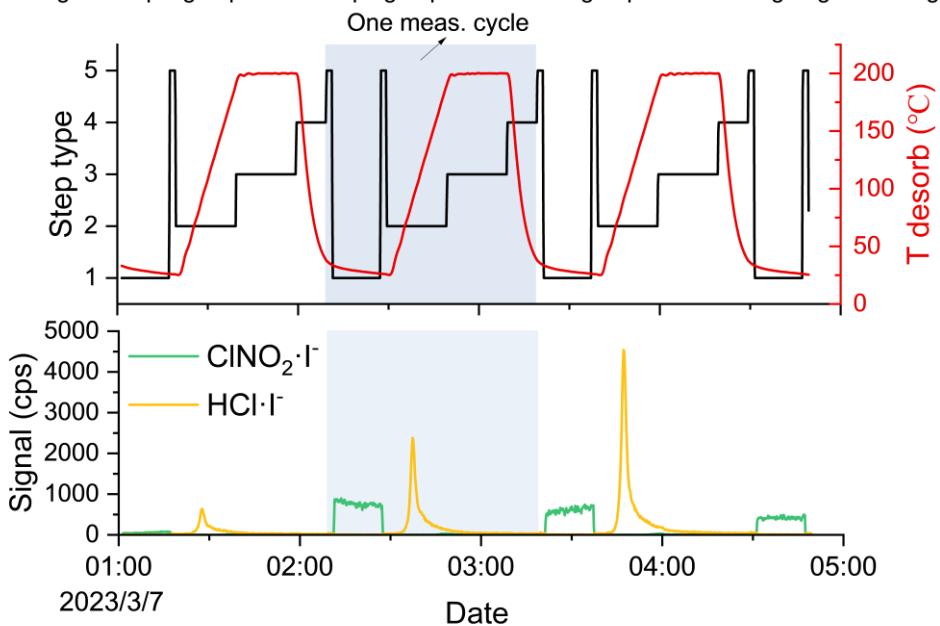
235 **Fig. S3 (a-b) Temporal trends in bulk PM composition measured at IITD during**  
 236 **winter from 2017 to 2022. (c) Variations of the composition fractions under**  
 237 **varying  $\text{PM}_{2.5}$  ranges.** The original data for (a-b) was compiled from previous studies  
 238 based on ACSM or AMS measurements and shown in **Table S3.** The dashed vertical  
 239 line in (a-b) indicates the end of 2019. The  $\text{PM}_{2.5}$  composition data in (c) was retrieved  
 240 from Ali et al (Ali et al., 2025). and the corresponding  $\text{PM}_{2.5}$  mass concentrations were  
 241 obtained from the nearby R.K. Puram station.



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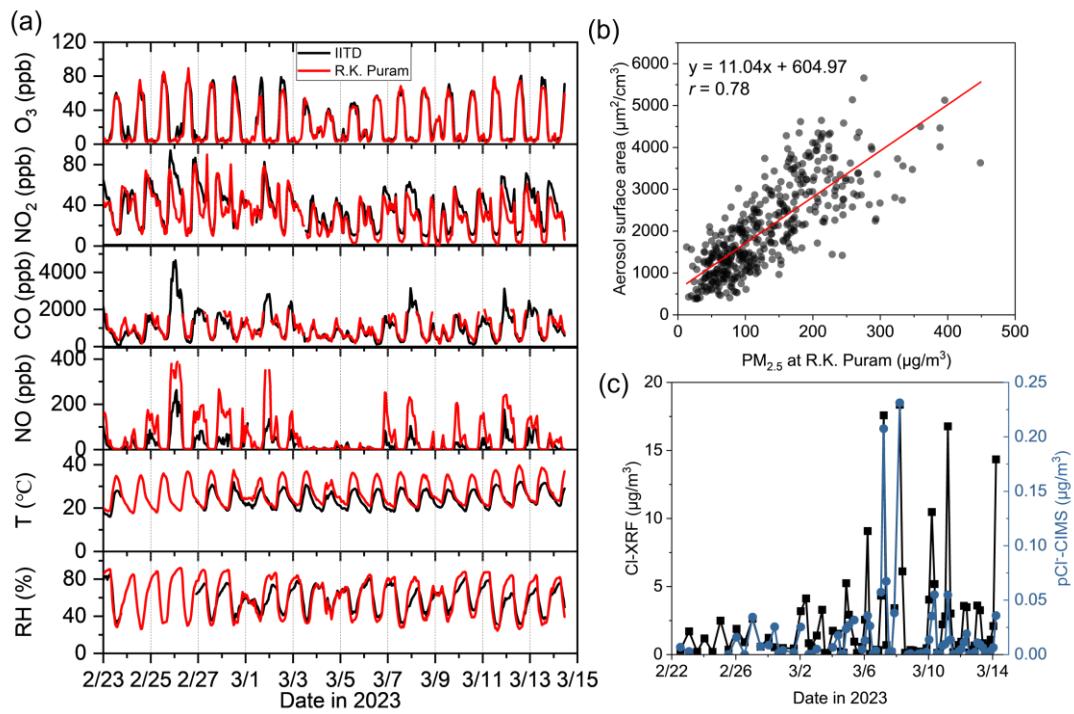
243 **Fig. S4 (a) The locations and (b) a zoom-in view of the sampling site.** The two photos  
 244 in (b) were taken during the early morning and late afternoon hours, respectively. Map  
 245 data © Google Maps 2026.

1: gas sampling 2: particle ramping 3: particle soaking 4: particle cooling 5: gas zeroing



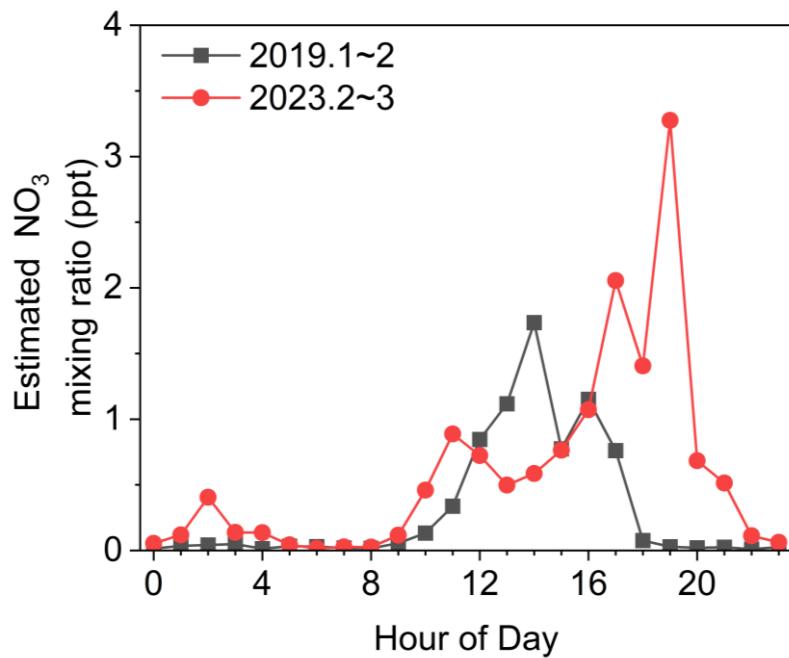
246

247 **Fig. S5 An example of the measurement cycle.**



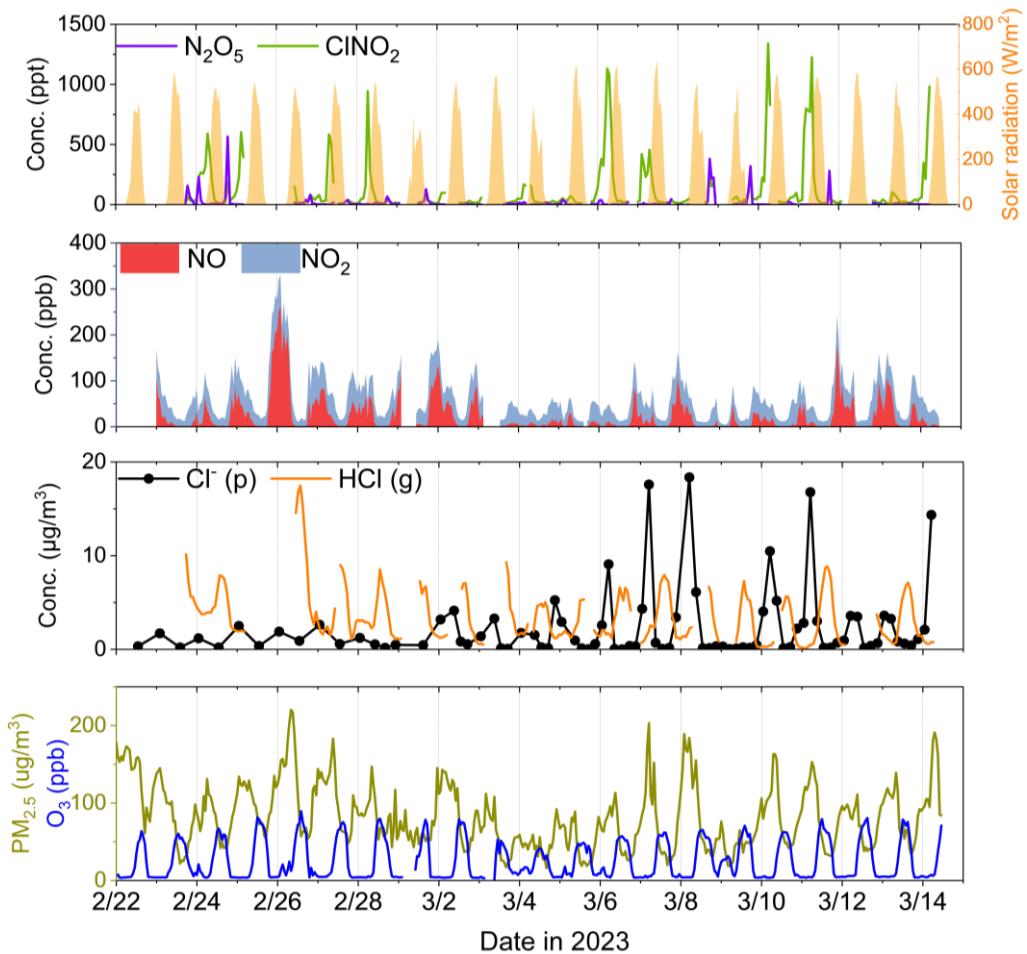
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249 **Fig. S6 Intercomparison of the measured species and estimation of aerosol surface**  
 250 **area. (a) Comparison of the time series of trace gases and meteorological parameters**  
 251 **measured at IIT Delhi and the R.K. Puram station in 2023 campaign. (b) Correlation**  
 252 **between the measured aerosol surface area at IIT Delhi(Gani et al., 2020) and  $\text{PM}_{2.5}$  at**  
 253 **R.K. Puram station from February to March in 2018. (c) Time series of particulate Cl**  
 254 **measured by XRF and CIMS.**



255

256 **Fig. S7 estimated  $\text{NO}_3$ - mixing ratios.** The  $\text{NO}_3$  mixing ratio was estimated by using  
 257 the observed  $\text{N}_2\text{O}_5$  and  $\text{NO}_2$  mixing ratio and the temperature-dependent equilibrium  
 258 constant between  $\text{NO}_2$ ,  $\text{NO}_3$ , and  $\text{N}_2\text{O}_5$ .



259

260 **Fig. S8 Overview of the field observations.** Time series of  $\text{N}_2\text{O}_5$ ,  $\text{ClNO}_2$ , solar  
 261 radiation, NO,  $\text{NO}_2$ , gaseous  $\text{HCl}$ ,  $\text{PM}_{2.5}$ , and  $\text{O}_3$ , and Cl element concentrations in  
 262  $\text{PM}_{2.5}$ .

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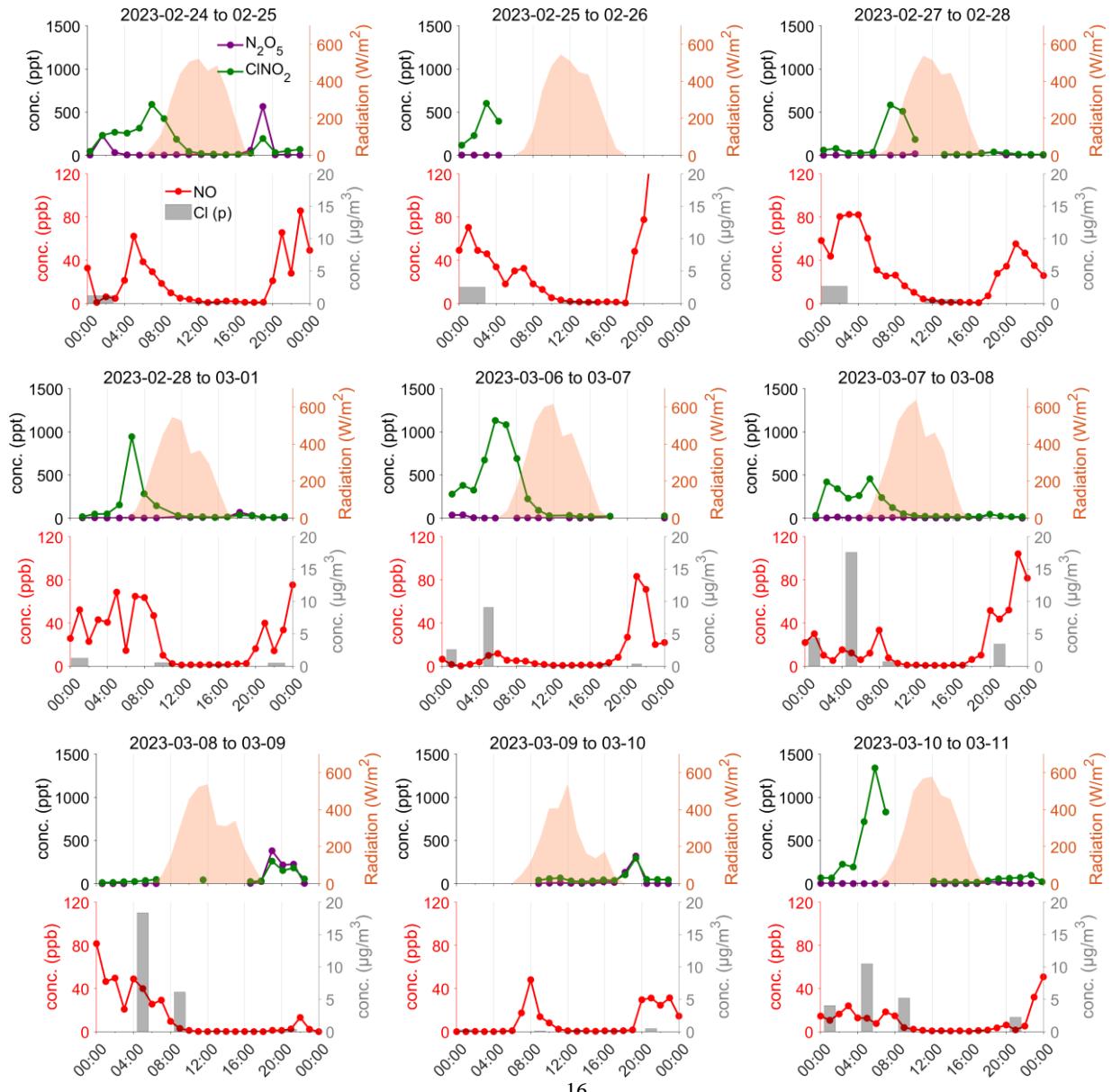
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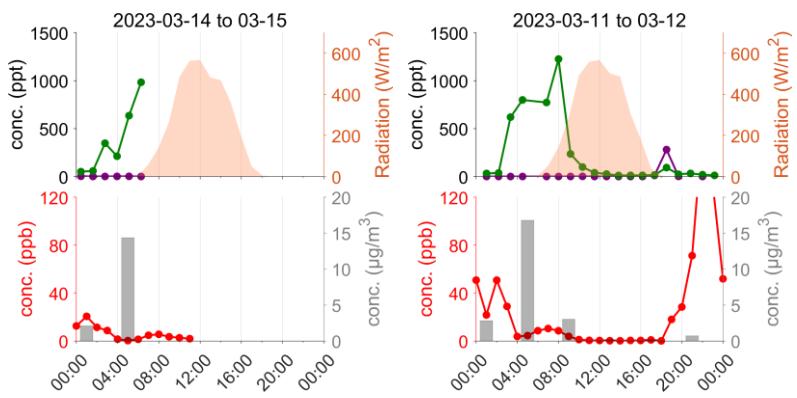
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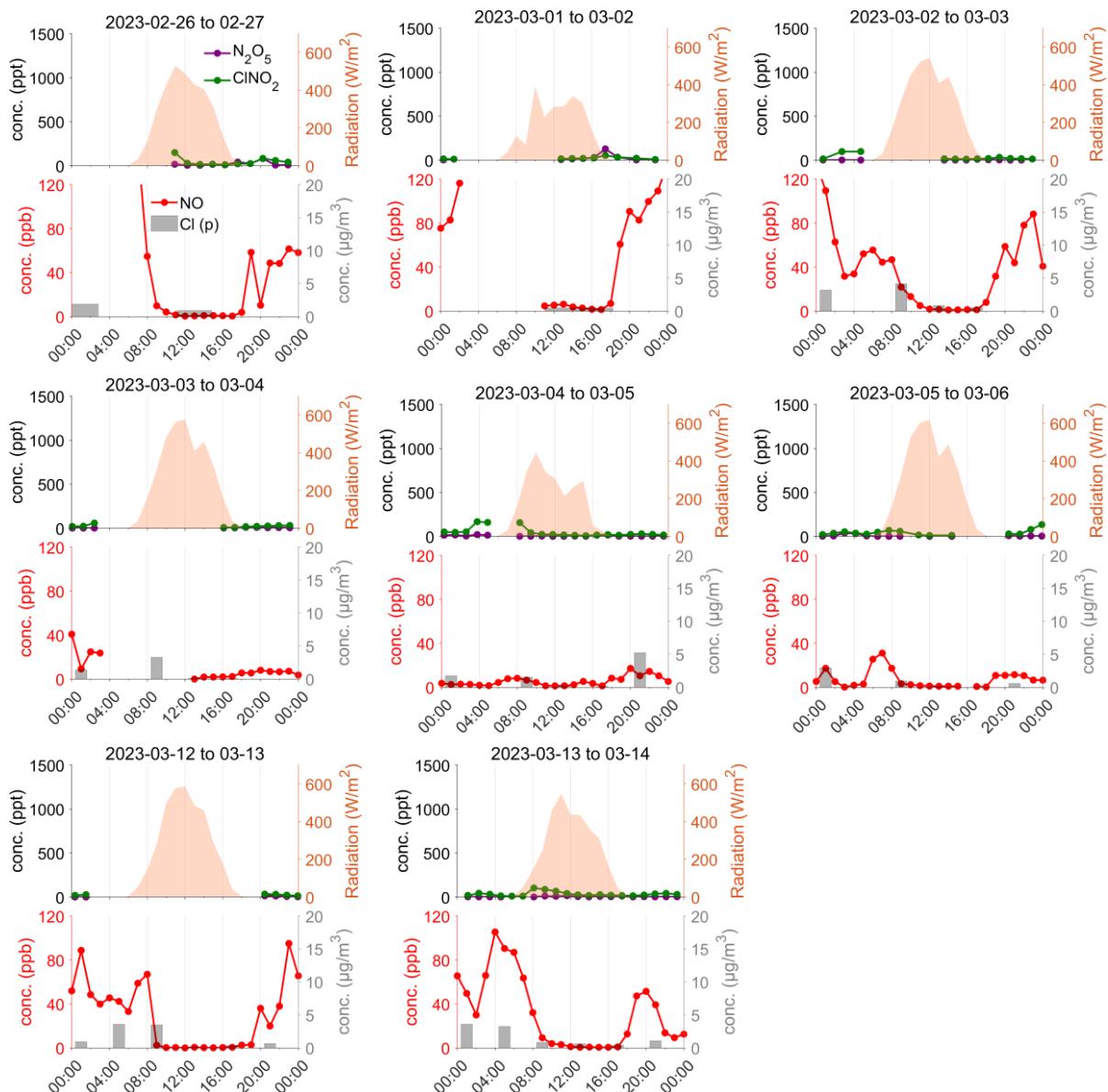
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271 **Fig. S9 Dependence of nocturnal  $\text{ClNO}_2$  on chloride and NO during the (a) 2023  
272 and (b) 2019 campaign.**  $\text{ClNO}_2$  and NO were stratified by chloride concentration  
273 intervals. Boxes and whiskers represent 25<sup>th</sup> and 75<sup>th</sup>, 10<sup>th</sup> and 90<sup>th</sup> percentiles,  
274 respectively. The dots and lines inside the box denote average and median values.



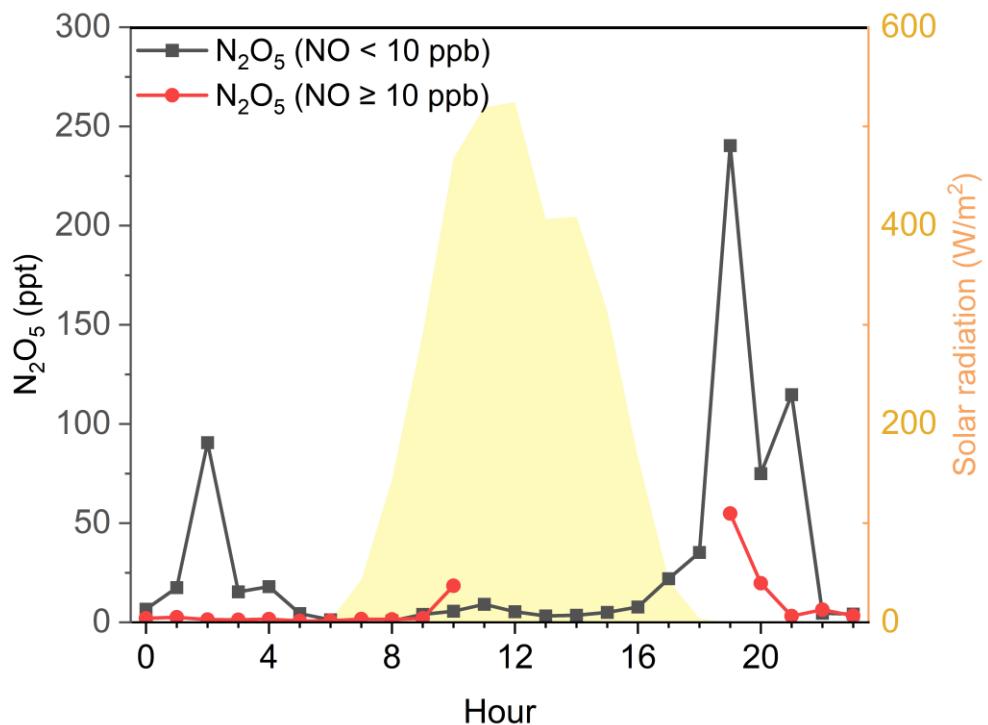


275 **Fig. S10 Day by day plots of the enhanced cases observed in 2023.** An enhanced  
 276 case is identified by a pronounced nighttime increase in either  $\text{N}_2\text{O}_5$  or  $\text{ClNO}_2$ .  
 277



278  
 279 **Fig. S11 Day by day plots of the non-enhanced cases observed in 2023.** A non-

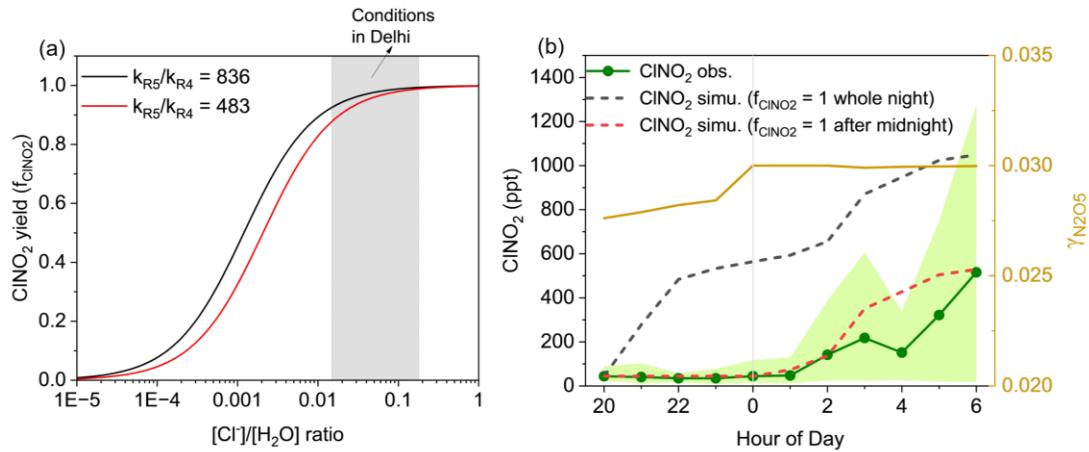
280 enhanced case is identified when neither  $\text{N}_2\text{O}_5$  nor  $\text{ClNO}_2$  shows a pronounced  
281 nighttime increase.



282  
283 **Fig. S12 Average diurnal variations of (a)  $\text{N}_2\text{O}_5$  under low- and high-NO conditions**  
284 **(b)  $\text{PM}_{2.5}$ , total measured gaseous and particulate organics in 2023.** A 10 ppb of NO  
285 threshold was applied to ensure statistically robust and comparable datasets. As all NO  
286 concentrations were below 10 ppb between 11:00 and 18:00, no  $\text{N}_2\text{O}_5$  data are available  
287 for the high-NO condition during this period. The right axis in (b) shows the average  
288 solar radiation in 2023.

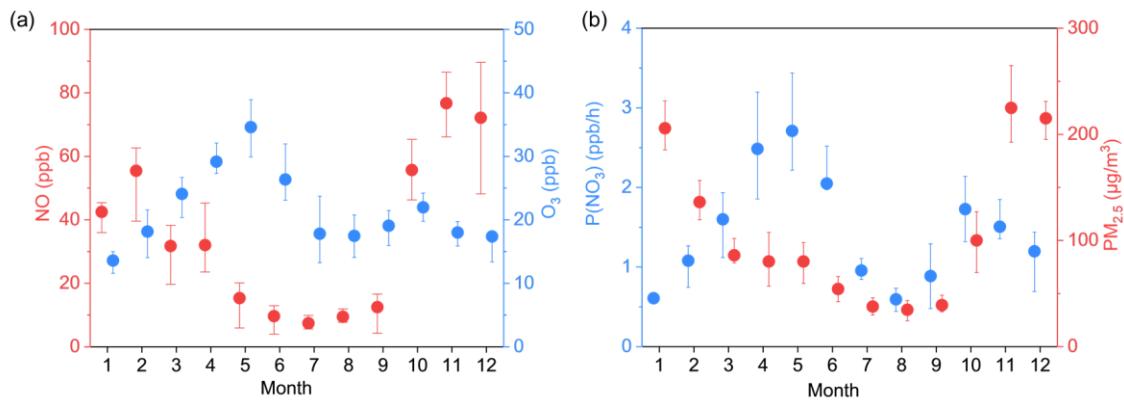
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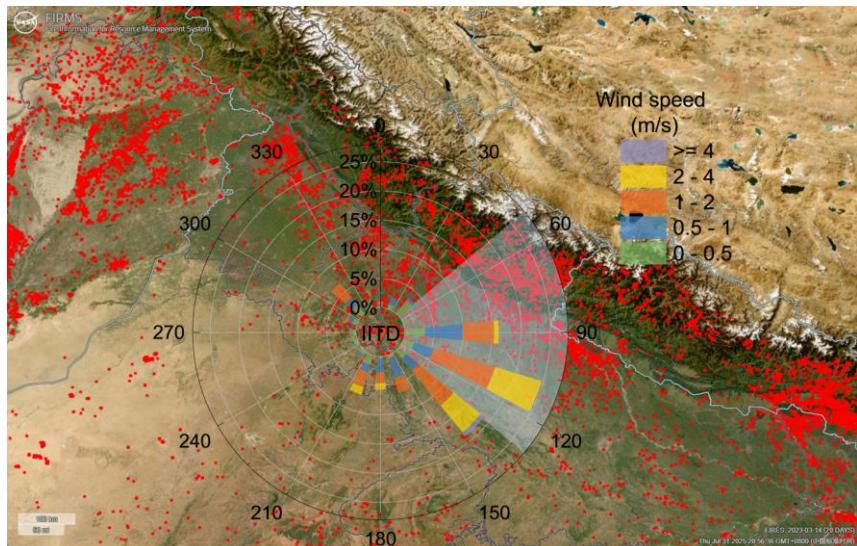
291

292 **Fig. S13 (a) Parameterization of ClNO<sub>2</sub> yield and (b) simulation of the nocturnal**  
 293 **ClNO<sub>2</sub> variations.** The lines in (a) correspond to ClNO<sub>2</sub> yield calculated using Eq. 3  
 294 with different reported reaction rate constant ratios. The grey shaded area indicates the  
 295 10<sup>th</sup> and 90<sup>th</sup> percentiles of the calculated [Cl<sup>-</sup>]/[H<sub>2</sub>O] ratios using ISOROPPIA from  
 296 January to March of 2022 in Delhi(Ali et al., 2025). The green shaded area in (b)  
 297 represents the 10<sup>th</sup> and 90<sup>th</sup> percentiles of the observed average nocturnal ClNO<sub>2</sub> mixing  
 298 ratios. The dashed lines in (b) are the simulated ClNO<sub>2</sub> mixing ratios and the average  
 299 RH parameterized N<sub>2</sub>O<sub>5</sub> uptake coefficient is shown in the right axis of (b).



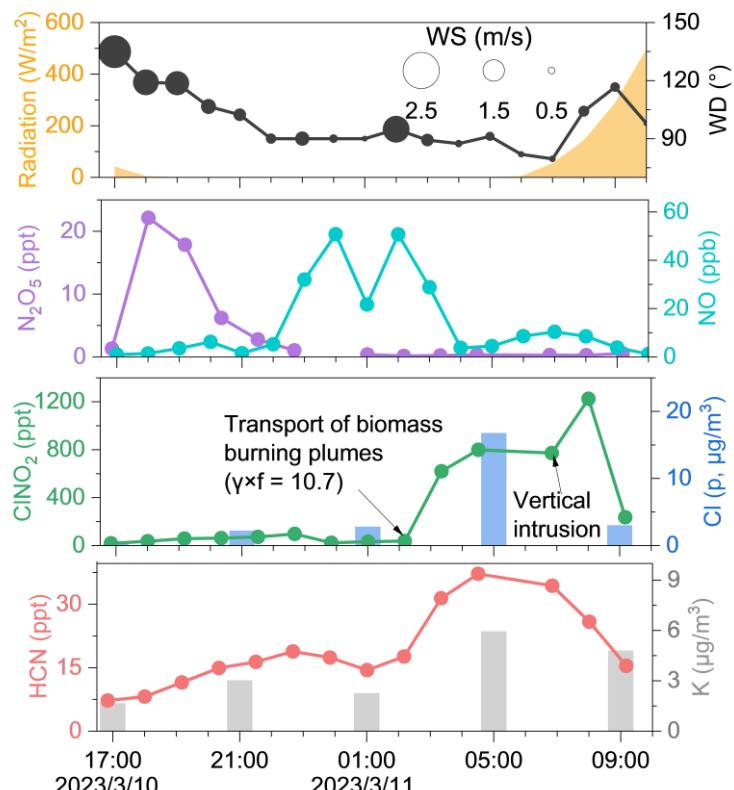
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301 **Fig. S14 Monthly variations of NO, O<sub>3</sub>, P(NO<sub>3</sub>), and PM<sub>2.5</sub> in Delhi.** Average  
 302 monthly variations of (a) NO and O<sub>3</sub> and (b) P(NO<sub>3</sub>) and PM<sub>2.5</sub> from 2017 to 2024. The  
 303 dots represent the averages and the whiskers denote the 25<sup>th</sup> to 75<sup>th</sup> percentile ranges.  
 304 All data were obtained from the R.K. Puram national monitoring station.



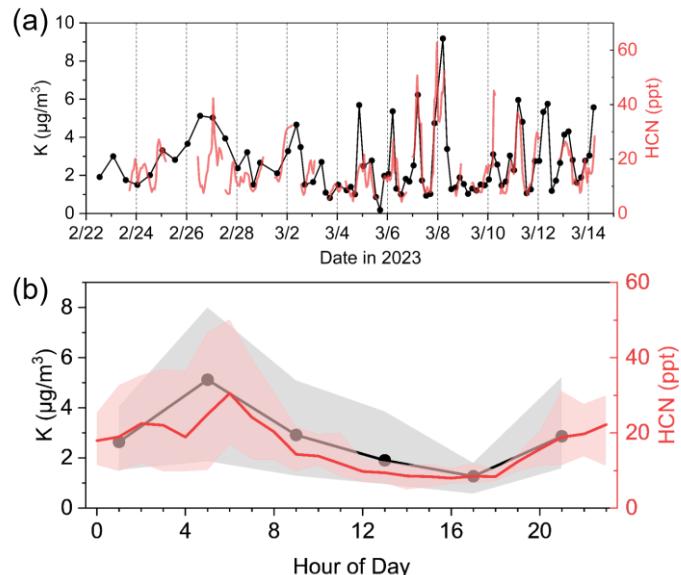
305

306 **Fig. S15 Satellite observations of active fire hotspots and wind rose plot during the**  
 307 **2023 campaign.** The map of the fire-points was downloaded from <https://earthdata.nasa.gov/firms>. The measurement site (IIT Delhi) was indicated as IITD. The  
 308 light blue shaded area indicates the dominant wind-direction (50~130°) when the  
 309 extremely high values of the uptake parameter ( $\gamma \times f$ ) exceeding 0.1 were measured.  
 310



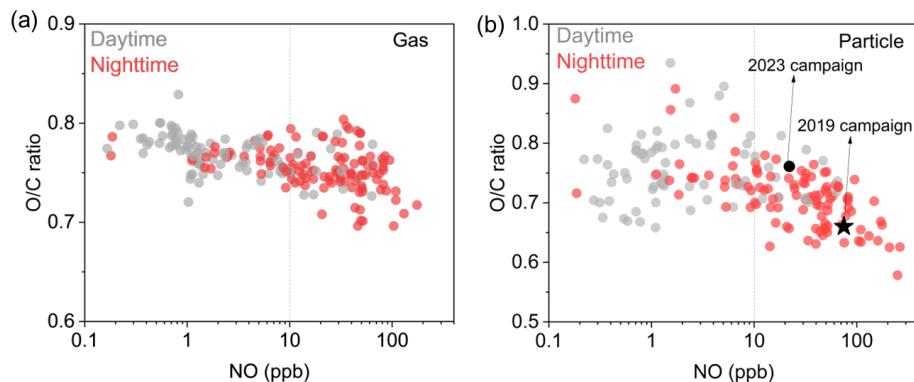
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312 **Fig. S16 A case study indicating the influence of transport on the observed ClNO<sub>2</sub>**  
 313 **levels.** An exceptionally high  $\gamma \times f$  value (10.7) observed at 2023/3/11 2:10 is also noted.



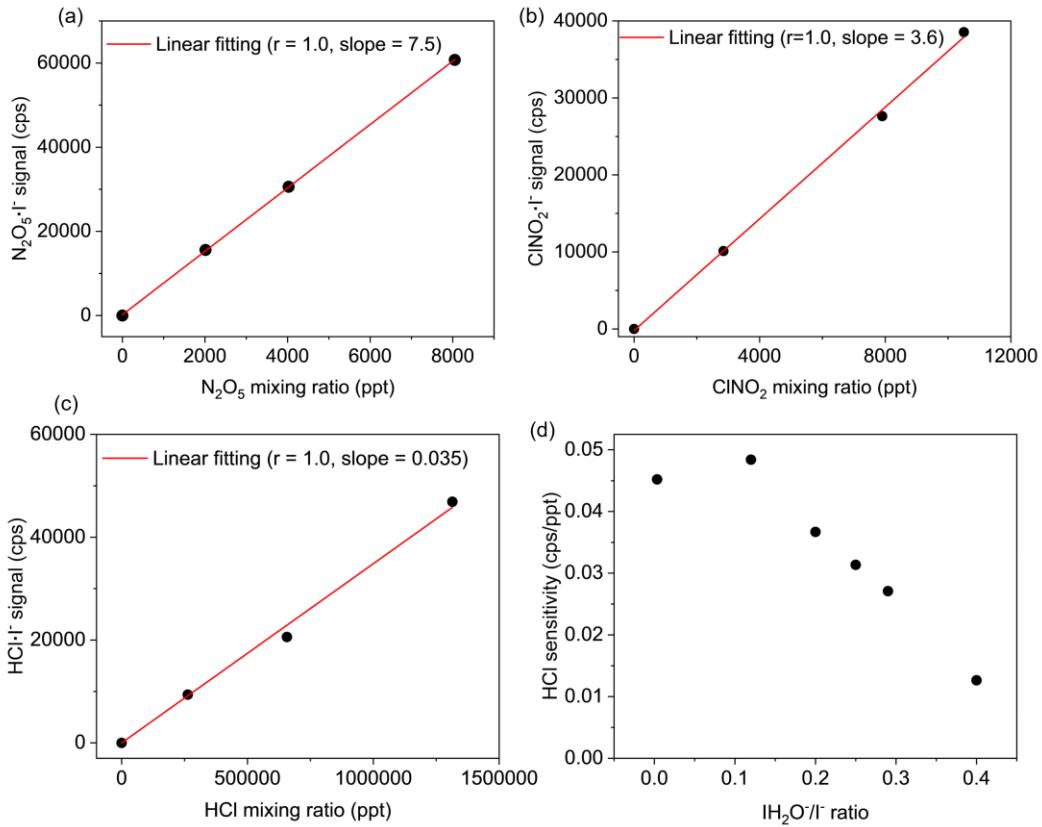
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315 **Fig. S17 (a) Time series and (b) average diurnal pattern of element K and gaseous**  
 316 **HCN during the 2023 campaign.** The shaded areas indicates 10<sup>th</sup> and 90<sup>th</sup> percentiles  
 317 and the lines represent the campaign-averaged values. Hourly CIMS measurements  
 318 were shown for HCN. The highest K and HCN concentrations on 3/7 and 3/8 were  
 319 likely driven by large-scale bonfires involving wood and leaves burning during the  
 320 Holika Dahan festival (3/6 16:00~3/8).



321

322 **Fig. S18 NO dependence of the gaseous and particulate bulk O/C ratio in Delhi.**  
 323 Scatter plots showing the relationship between ambient NO levels and concentration-  
 324 weighted bulk O/C ratios in the (a) gas and (b) particle phases.



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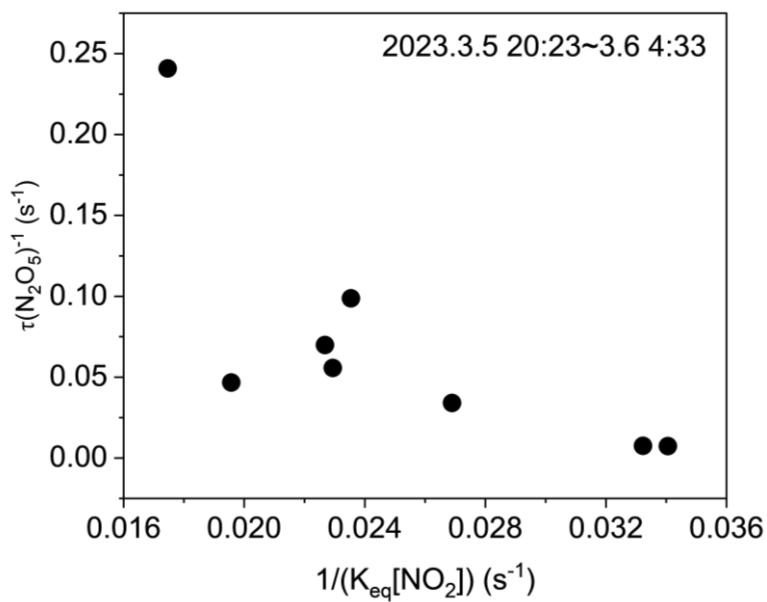
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**Fig. S19 Sensitivity calibration of gaseous (a)  $\text{N}_2\text{O}_5$ , (b)  $\text{ClNO}_2$ , and (c)  $\text{HCl}$  and (d) water dependency of  $\text{HCl}$  sensitivity.** Multi-point calibrations for  $\text{N}_2\text{O}_5$  and  $\text{ClNO}_2$  were performed under RH conditions corresponding to an  $\text{IH}_2\text{O}^-/\text{I}^-$  ratio of  $0.42 \pm 0.02$ , while the calibration curve at an  $\text{IH}_2\text{O}^-/\text{I}^-$  ratio of 0.19 was shown for  $\text{HCl}$ .



330

331

**Fig. S20 An example of the steady-state analysis of the  $\text{NO}_2\text{-NO}_3\text{-N}_2\text{O}_5$  system.**

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