

1 *Supporting Information of*

2 **Enhanced nocturnal N₂O₅-ClNO₂ chemistry in Delhi under lower-NO conditions**

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Supplementary Text

Text. S1 Details of field measurements in Delhi

1.1 FIGAERO-I-CIMS configurations

The reagent ions (I^- and $\text{I} \cdot \text{H}_2\text{O}^-$) were produced by passing ~ 2 lpm ultrahigh purity (UHP) N_2 via a CH_3I permeation tube and through a Po-210 ion source, where it interacted with the sampled air or thermally desorbed molecules from $\text{PM}_{2.5}$ in the ion-molecular reactor (IMR). A constant 10 sccm of H_2O -saturated UHP was added directly into the IMR to reduce the sensitivity variability with ambient RH. The IMR pressure was maintained at 200-250 mbar during gas sampling and 120-140 mbar during $\text{PM}_{2.5}$ desorption. The relatively lower pressure during the $\text{PM}_{2.5}$ measurement was likely caused by the resistance and associated pressure drop across the filters. Considering the differences in IMR pressures, we conducted sensitivity calibrations for the gas- and particle-phase measurements separately under conditions similar to the field observations in Delhi. The average mass resolution was ~ 3500 at m/z 127.

For the gas-phase measurements, ambient air was drawn at 3 lpm through a 4-m-long PFA tube (inner diameter, 5 mm, residence time 1.6 s) with 2 lpm pumped away. An impactor with a cut-off of $\text{PM}_{2.5}$ was placed at the gas inlet to reduce large particle deposited in the sampling system. The bypass flow was utilized to ensure a short residence time (~ 1.6 s) while maintaining a laminar flow (Reynolds number = $840 < 2300$) within the tubing, thereby suppressing both wall loss and gas-particle reactions within the inlet. The remaining 1 lpm was diluted with 1 lpm UHP N_2 before entering the IMR to avoid reagent ions titration by high levels of pollutants (e.g., HNO_3) in the ambient air.

For background measurements, the inlet was flushed with ~ 2 lpm UHP N_2 during the first and last two minutes of each 20-min gas-phase measurement to obtain background signals, while ambient sampling was performed during the remaining period. Continuous and automated background measurements were achieved with this flow rate configuration. The signals from the second zeroing period were used as the background, since those from the first zeroing period may have been influenced by carryover desorption signals (especially for sticky compounds) from the preceding particle-phase measurement (Fig. S5). The background-subtracted and dilution ratio (a factor of 2) corrected ion signals were subsequently averaged to 1-hour intervals.

For $\text{PM}_{2.5}$ measurements, ambient air was drawn at 3 lpm through a 4-m-long copper tube (inner diameter, 5 mm) fitted with a $\text{PM}_{2.5}$ cyclone during the gas-sampling period, with 2 lpm discarded and 1 lpm collected on a Teflon filter for 20 min. The filter was then moved to the desorption position, where it was thermally desorbed and carried

into the IMR under a 2 lpm flow of UHP N₂. The N₂ flow was gradually ramped from room temperature to 200 °C in 20 min, soaked at 200 °C for 20 min, and finally cooled back to room temperature in 10 min. The particle phase signal was integrated over the temperature ramping and soaking period during filter desorption. The baseline was determined from linear fitting of the signal between the onset of heating and the end of soaking and was integrated and subtracted from the integrated desorption signals.

1.2. Sensitivity calibrations of N₂O₅, ClNO₂ and related species

N₂O₅ and ClNO₂. N₂O₅ was prepared by mixing O₃ with excessive NO₂ to promote the conversion of NO₃ to N₂O₅. The generated N₂O₅ concentrations were determined by the change in O₃ following the addition of NO₂, and different N₂O₅ levels was achieved via varying O₃ concentration (Bertram et al., 2009). The produced N₂O₅ was further diluted with humidified zero air to approximate Delhi RH conditions before entering the CIMS. ClNO₂ was produced by passing a known amount of N₂O₅ through a wetted NaCl-slurry placed in a Teflon tube, assuming a unit conversion from N₂O₅ to ClNO₂ (Finlayson-Pitts, 2003). All the tubings are covered by aluminum foil to avoid NO₃ photolysis and were flushed with dry zero air overnight before calibration to minimize potential N₂O₅ hydrolysis on the tubing surfaces. The calibration curve for N₂O₅ and ClNO₂ are shown in Fig. S19a-b. No water-dependent sensitivity correction was applied for N₂O₅ and ClNO₂, as no significant sensitivity variation was observed in the range of H₂O·I⁻/I⁻ ratios during the field measurements in Delhi (Fig. S19c-d).

HCl and chloroacetic acid. A certified 10 ppm HCl gas cylinder (Linde Specialgas) was used as the standard source, which was diluted with zero air to different mixing ratios before introducing to the CIMS (Fig. S19e). Notably, we observed a significant decrease of the HCl sensitivity at high H₂O·I⁻/I⁻ ratios (Fig. S19f), likely due to the competing effects of H₂O for clustering with iodide anion and that the reaction of HCl with H₂O·I⁻ is thermodynamically less favorable compared to reacting with iodide anion. The slight increase in HCl sensitivity with increasing H₂O·I⁻/I⁻ ratio at low RH levels may be related to the stabilization of HCl·I⁻ clusters by water vapors. Similar patterns have been reported for other acids in a previous study (Lee et al., 2014). Gaseous chloroacetic acid was generated by placing solid chloroacetic acid in a glass vial submerged in a 30 °C water bath. The emission rate, determined by gravimetric weight loss analysis, was 177.3 ± 0.8 ng/min. No significant H₂O dependence of chloroacetic acid sensitivity was observed. It is noted that HCl was calibrated in the concentration range about 2~3 orders of magnitude higher than the ambient level. Nevertheless, given the strong linearity of the multipoint calibration at high HCl concentrations, and the verified linear response of the CIMS at lower concentrations for N₂O₅ and ClNO₂, we assume that the calibrated HCl sensitivity is applicable to

ambient measurements.

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

1.3. R.K. Puram station data quality control

Hourly pollutant measurements from the R.K. Puram station were obtained to examine the long-term pollutant trends in Delhi. We applied rigorous data quality controls following recent studies (Vohra et al., 2025; Xie et al., 2024) with a few modifications, including (1) Remove duplicates: In sequences of five or more consecutive identical hourly values, only the first value out of the sequence is retained, (2) Remove outliers: For a 24-h running window, flag any observation as an outlier if its absolute difference from the median exceeds three times the Median Absolute Deviation (MAD, median distance between each observation and the median of all observations), (3) Remove constant data: In a 24-h running window, remove constant data values with coefficient of variation (ratio of standard deviation to the mean) less than 5%, (4) Validation of NO, NO₂ measurements by comparing with the measured NO_x: consider NO and NO₂ are reported correctly in unit of µg/m³ and assess if NO_x calculated from these is within 2% of reported NO_x + 2.5 ppb. If not, the measured NO, NO₂, and NO_x data were omitted.

Text. S2 Estimation of the N₂O₅ uptake coefficient

2.1 Inapplicability of the steady-state method

The steady-state approximation of the NO₂-NO₃-N₂O₅ system is a widely applied method (Brown et al., 2003) for estimating the values for the NO₃ and N₂O₅ sinks (thereby $\gamma_{N_2O_5}$) in various atmospheric environments (Brown et al., 2006; Wang et al., 2017b; Wang et al., 2017a). This method assumes that production rate of NO₃ and N₂O₅ equals the sum of the loss rates during a certain period of the night (Eq. 1), and therefore k_{NO_3} and $k_{N_2O_5}$ can be determined as the slope and intercept, respectively, of the linear fitting between $\tau_{N_2O_5}^{-1}$ and $\frac{1}{K_{eq}[NO_2]}$ (Eq. 2). However, we found no significant positive even negative correlation between $\tau_{N_2O_5}^{-1}$ and $\frac{1}{K_{eq}[NO_2]}$ during the nighttime throughout the campaign (an example is shown in Fig. S20). This is likely attributed to the intense and variable NO_x emissions in Delhi (avg. nighttime hourly NO_x mixing ratios of 92±60 ppb) which preclude the system from approaching steady-state on the time-scale of a night. As is shown in Fig. 2a of the main text, N₂O₅

presented a strong negative dependence on NO. The high concentrations and large fluctuations of NO dominated the variation of N₂O₅, thereby decoupling the lifetime of N₂O₅ from $\frac{1}{K_{eq}[NO_2]}$.

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

$$K_{eq} = 5.5 \times 10^{-27} \times \exp(10724/T)$$

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

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

Where k_{NO_3} and $k_{N_2O_5}$ are the pseudo-first-order loss rate constants of NO₃ and N₂O₅, respectively, $k_{N_2O_5} = \frac{\bar{c}\gamma}{4} S_a$, $\tau_{N_2O_5}^{-1}$ is the inverse of the N₂O₅ steady-state lifetime.

2.2 Validation of the RH parameterization method

ClNO₂ yield indicates the branching ratio of H₂ONO₂⁺ reacting with H₂O (R4) and chloride (R5), which is theoretically determined by the respective reaction rates shown in Eq. 3 (Bertram and Thornton, 2009).

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

Where [H₂O] and [Cl⁻] are mole concentrations in the aqueous phase of particles derived from thermodynamic models, k_{R5} and k_{R4} are reaction rate coefficients of Reaction R5 and R4. The experimentally determined values for k_{R5}/k_{R4} is 483±175 from the work of Bertram and Thornton (Bertram and Thornton, 2009) and 836±32 from Behnke et al (Behnke et al., 1997). Due to the lack of aerosol inorganic composition (e.g., NH₄⁺, NO₃⁻, and SO₄²⁻) measurements during the 2023 campaign, we estimated the range of [Cl⁻]/[H₂O] ratio from January to March in 2022 Delhi when ACSM measurements were available (Ali et al., 2025), where the chloride level was similar to the campaign in 2023. The estimated nocturnal (18:00~06:00) [Cl⁻]/[H₂O] ratio ranged from 1.8×10⁻⁵ to 0.76 with an average of 0.07, which mostly fell in the region where the corresponding ClNO₂ yield close to 1 (Fig. S13a). We therefore tested the validity of the RH parameterization method by simulating the average nocturnal variation of ClNO₂ constrained with the parameterized gamma, observed N₂O₅ mixing ratios and aerosol surface area concentrations, with the ClNO₂ yield set to 1. The results showed that the simulation largely overestimated the production of ClNO₂ from 20:00

to 00:00 (Fig. S13b) when the absolute chloride concentrations were relatively low (Fig. 1d). By comparison, when constraining ClNO₂ yield to 0 for the chloride-deficient period (20:00 to 00:00) and 1 for the chloride-sufficient period (0:00 to 6:00), the modelled ClNO₂ evolution well tracked the observations (Fig. S13b). The overall consistency between modelling and observations suggest that the measured ClNO₂ can largely explained by the locally heterogeneous uptake of N₂O₅ on chloride-rich particles in Delhi and that the RH-parameterized $\gamma_{N_2O_5}$ is a reasonable estimate, indicating the key role of RH and thereby aerosol liquid water content in driving N₂O₅ uptake in Delhi. Nevertheless, comprehensive aerosol composition measurements or direct measurements of N₂O₅ reactivity (Bertram et al., 2009; Li et al., 2025) in future studies are required to enable better quantification and understanding N₂O₅ uptake processes in Delhi.

Table S1 Summary of the bulk PM composition measured at IITD during winter from 2017 to 2022. The unit is $\mu\text{g}/\text{m}^3$.

Period	PM	Cl	NO ₃	SO ₄	NH ₄	Org	ref
2017.1~2017.3	PM ₁	14.2	16.1	15.8	15.7	81.0	(Gani et al., 2019)
2017.12~2018.3	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	PM _{2.5}	20.4	13.3	12.0	14.3	65.3	(Lalchandani et al., 2021)
2019.1.11~2.5	PM ₁	17.3	20.8	18.0	23.7	80.0	(Haslett et al., 2023)
2019.12.1~2020.1.5	PM _{2.5}	5.7	14.5	11.4	9.5	60.9	(Ali et al., 2023)
2020.2.1~3.20	PM ₁	4.9	10.6	9.7	8.9	47.9	(Mandariya et al., 2024)
2020.12.15~12.31	PM _{2.5}	11.3	27.2	13.7	25.7	111.5	(Faisal et al., 2025)
2020.12.15~2021.2.23	PM _{2.5}	4.9	19.8	15.5	15.7	96.2	(Ali et al., 2023)
2021.1.1~2021.2.28	PM _{2.5}	10.1	16.1	12.1	19.0	87.0	(Faisal et al., 2025)
2022.1.12~3.31	PM _{2.5}	4.5	15.3	14.0	12.9	86.2	(Ali et al., 2025)

Table S2 Campaign-averaged NO₃⁻ reactivity (unit: s⁻¹) during the 2019 and 2023 campaign.

	NO ₃ + NO	NO ₃ + hv	N ₂ O ₅ hete.	NO ₃ + VOCs
2019.1~2	50.175	0.031	0.462	0.081
2023.2~3	15.585	0.043	0.091	0.081*

*It is assumed to be the same as those observed in 2019.

Table S3 Summary of nocturnal peak and average (shown in the brackets) N₂O₅ and ClNO₂ mixing ratios measured around the world. The unit is ppt.

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Location	Period	Type	N ₂ O ₅	CINO ₂	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, Houston, 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)

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208 **Table S4 Average emission factor (g/kg fuel burned) of K, HCl, HCN, CO, and SO₂**
209 **from biomass, biofuel, and garbage burning processes and vehicles.**

		K	HCl	HCN	SO ₂	CO	HCl/CO	ref
Biomass burning	Peat	0.004~ 0.015	-	1.00	-	38	-	(Andreae, 2019)
	Crop residues	0.48	0.18	0.42	0.8	77	2.3×10 ⁻³	(Andreae, 2019)
	Rice straw	-	0.44	0.37	1.3	60	7.3×10 ⁻³	(Stockwell et al., 2014)
	Wheat straw	-	0.47	0.10	0.7	39	1.2×10 ⁻²	(Stockwell et al., 2014)
	Tropical forest	0.32	0.13	0.44	0.8	104	1.3×10 ⁻³	(Andreae, 2019)
	Temperate forest	0.18	0.04	0.64	0.7	112	3.5×10 ⁻⁴	(Andreae, 2019)
	Dung	0.09	0.04	1.30	0.7	89	4.3×10 ⁻⁴	(Andreae, 2019)
	Biofuels without dung	0.13	0.08	0.39	0.5	84	8.9×10 ⁻⁴	(Andreae, 2019)
	Charcoal	0.75	0.11	-	0.6	207	5.3×10 ⁻⁴	(Andreae, 2019)
	Wood	-	0.02	0.54	bld	77	2.4×10 ⁻⁴	(Stockwell et al., 2016)
Garbage burning	Garbage	0.02	2.80	0.43	0.5	66	4.2×10 ⁻²	(Andreae, 2019)
	Mixed plastic garbage	-	2.30	0.43	bld	85	2.7×10 ⁻²	(Stockwell et al., 2016)
Vehicles	Two-wheeled vehicles	-	bld	0.68	bld	761	-	(Stockwell et al., 2016)
	Agricultural irrigation pumps	-	bld	0.12	bld	17	-	(Stockwell et al., 2016)

210 Note: bld indicates below the limit of detection.

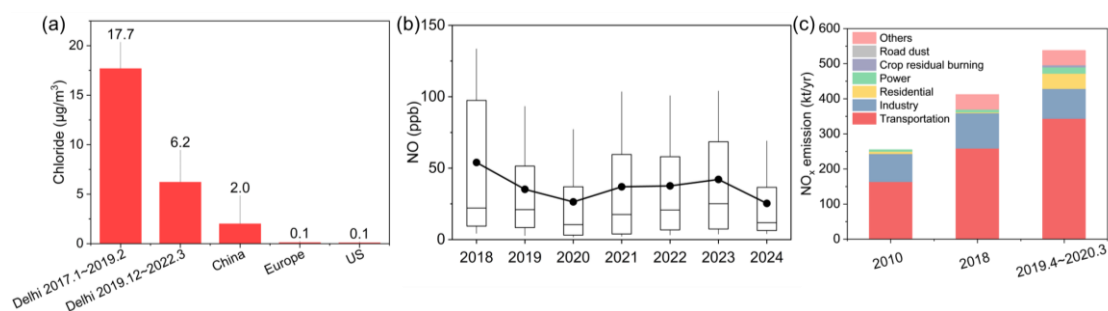


Fig. S1 Atmospheric chloride and NO concentrations and NO emissions in Delhi.

(a) Comparison of the observed chloride levels globally. (b) Box plots of the annual daily averages of NO concentrations in Delhi. (c) NO_x emissions in Delhi from the year of 2010(2010), 2018(Indian Institute of Tropical Meteorology, 2018) and 2019.4~2020.3(Sahu et al., 2023). The chloride concentrations in Delhi (compiled from Table S1), China (Xia et al., 2021;Tham et al., 2018;Xia et al., 2020), Europe (Eger et al., 2019;Lanz et al., 2010), and US (Mielke et al., 2013;Sarwar et al., 2012) are campaign-average values retrieved from previous studies. The whiskers in (a) represent standard deviation. The NO concentrations shown in (b) were obtained from the R.K. Puram monitoring station. The boxes represent the interquartile range with the whiskers extending to the 10th and 90th percentiles. The dot and horizontal line within each box denote the mean and median values, respectively.

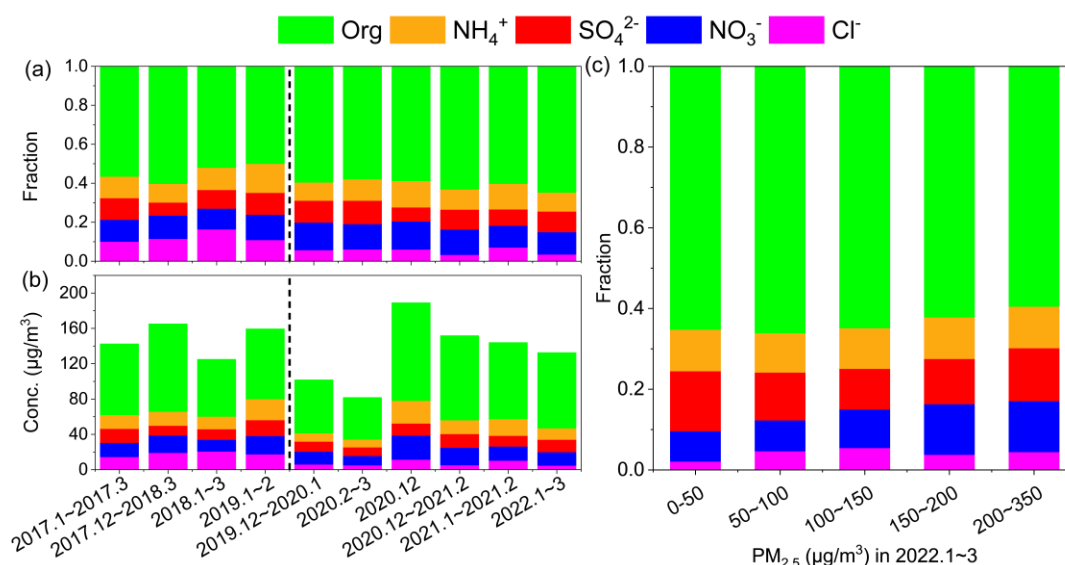


Fig. S2 (a-b) Temporal trends in bulk PM composition measured at IITD during winter from 2017 to 2022. (c) Variations of the composition fractions under varying PM_{2.5} ranges. The original data for (a-b) was compiled from previous studies based on ACSM or AMS measurements and shown in Table S1. The dashed vertical line in (a-b) indicates the end of 2019. The PM_{2.5} composition data in (c) was retrieved from Ali et al(Ali et al., 2025). and the corresponding PM_{2.5} mass concentrations were obtained from the nearby R.K. Puram station.

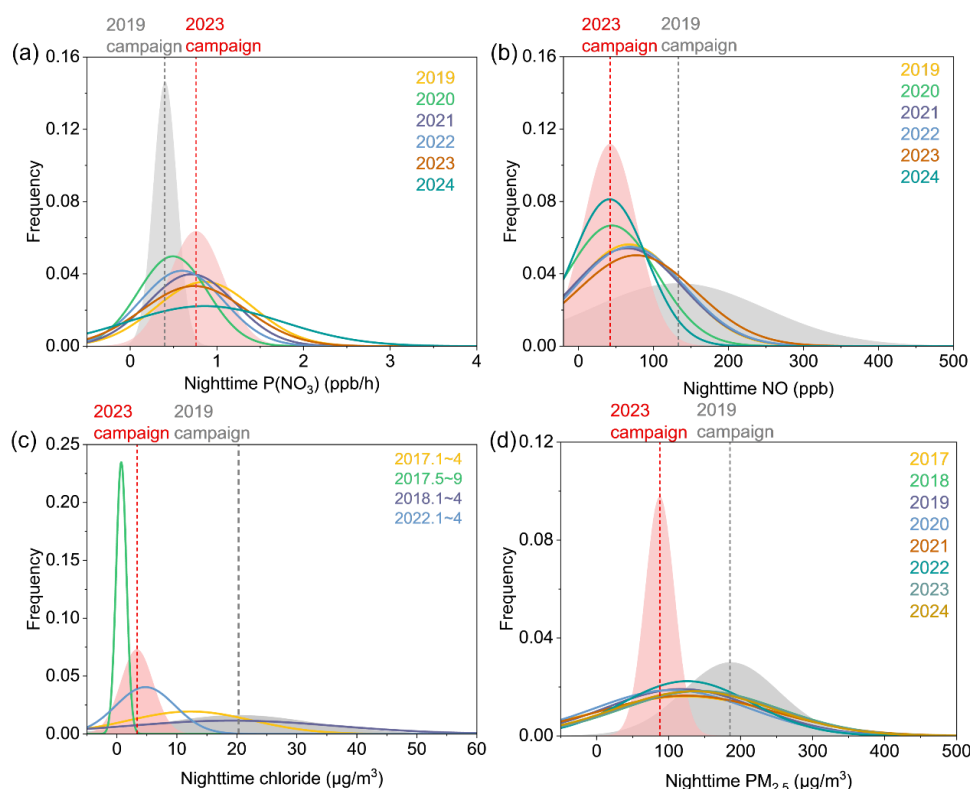


Fig. S3 Representativeness of the two field-campaigns in Delhi. Gaussian-fitted frequency distributions of nightly (a) $P(NO_3)$, (b) NO, (c) chloride, and (d) $PM_{2.5}$ concentrations from 2017 to 2024. The nighttime is defined as 20:00~04:00 throughout the years. The data for $P(NO_3)$, NO, and $PM_{2.5}$, are obtained from the R.K. Puram monitoring station. Chloride concentrations were unavailable at the station and are retrieved from previous studies (Gani et al., 2019; Ali et al., 2025). The gray and red shaded area denote the respective pollutant distribution during the 2019 and 2023 campaign and the dashed lines indicate the campaign averages.

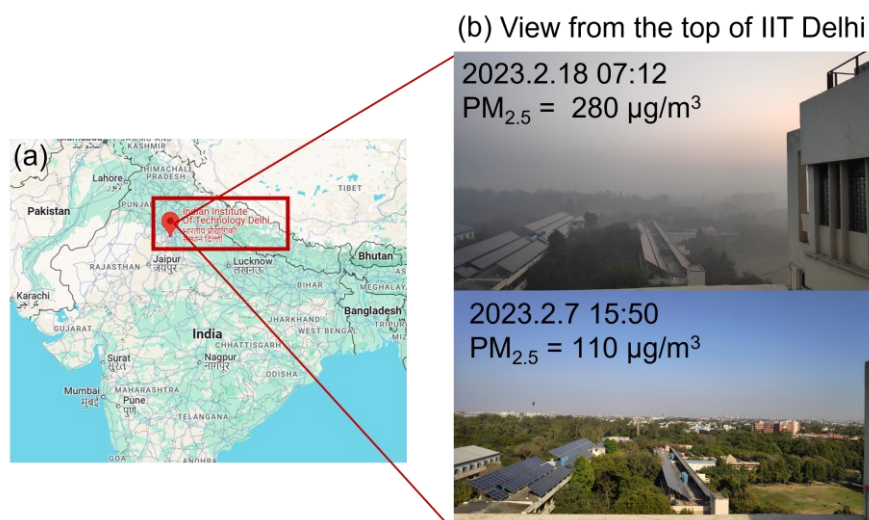


Fig. S4 (a) The locations and (b) a zoom-in view of the sampling site. The two photos in (b) were taken during the early morning and late afternoon hours, respectively.

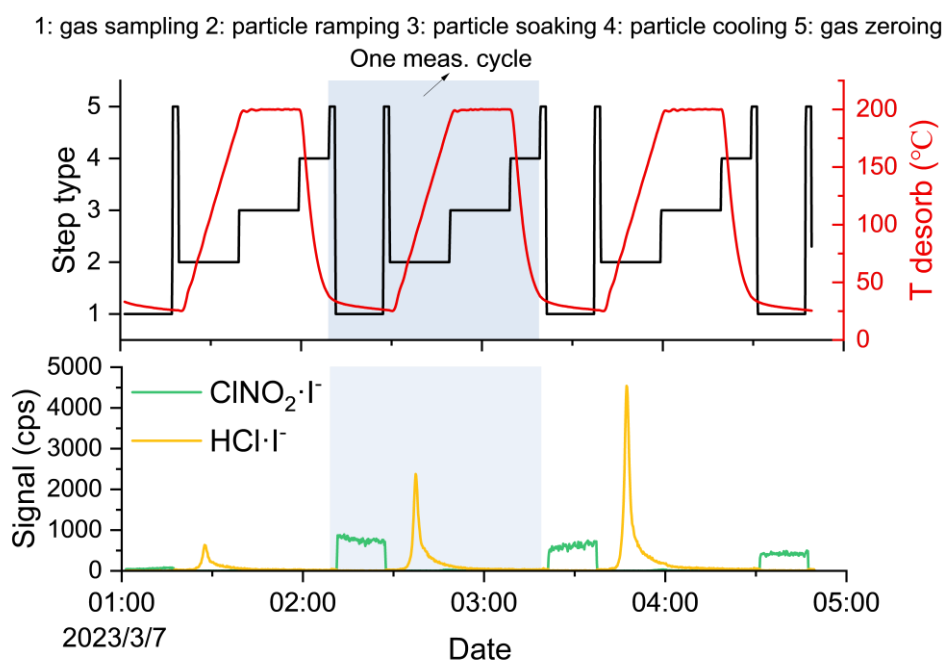


Fig. S5 An example of the measurement cycle.

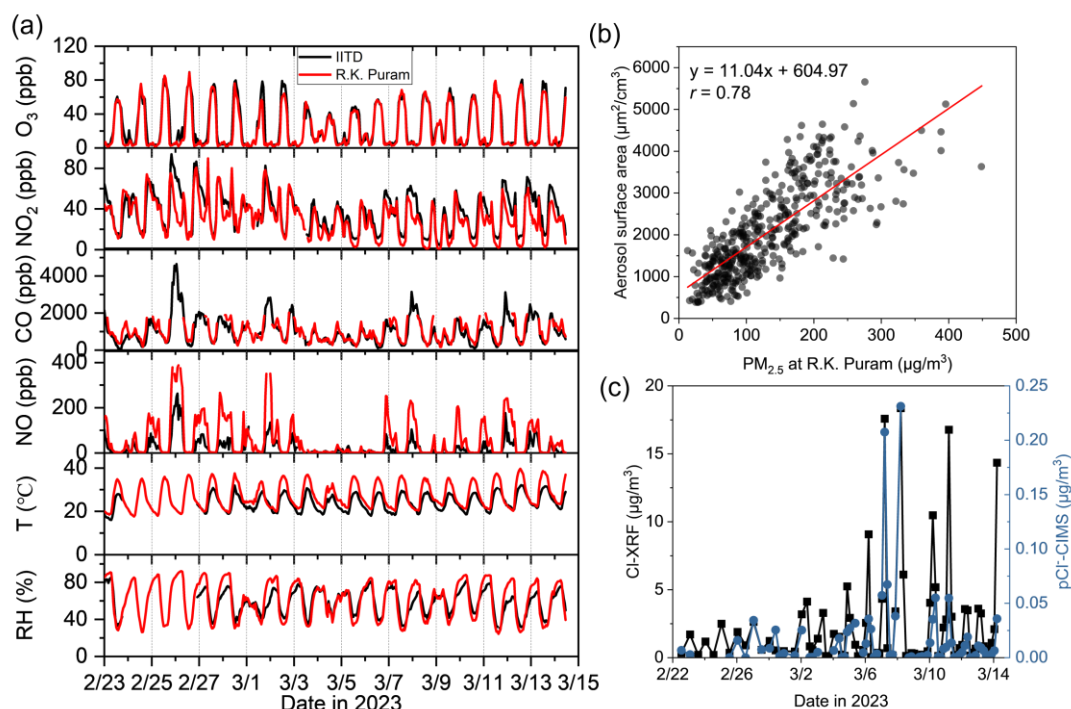


Fig. S6 Intercomparison of the measured species and estimation of aerosol surface area. (a) Comparison of the time series of trace gases and meteorological parameters measured at IIT Delhi and the R.K. Puram station in 2023 campaign. (b) Correlation between the measured aerosol surface area at IIT Delhi(Gani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

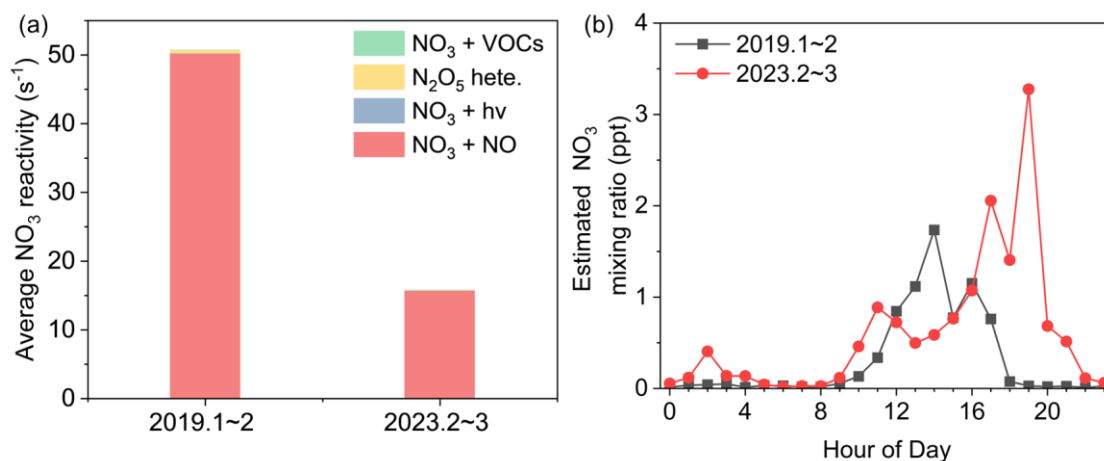
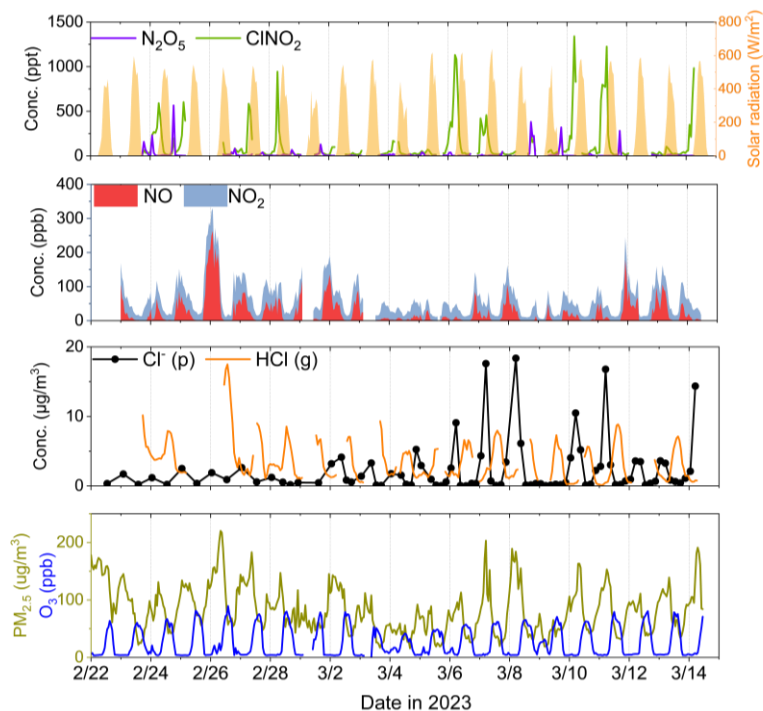
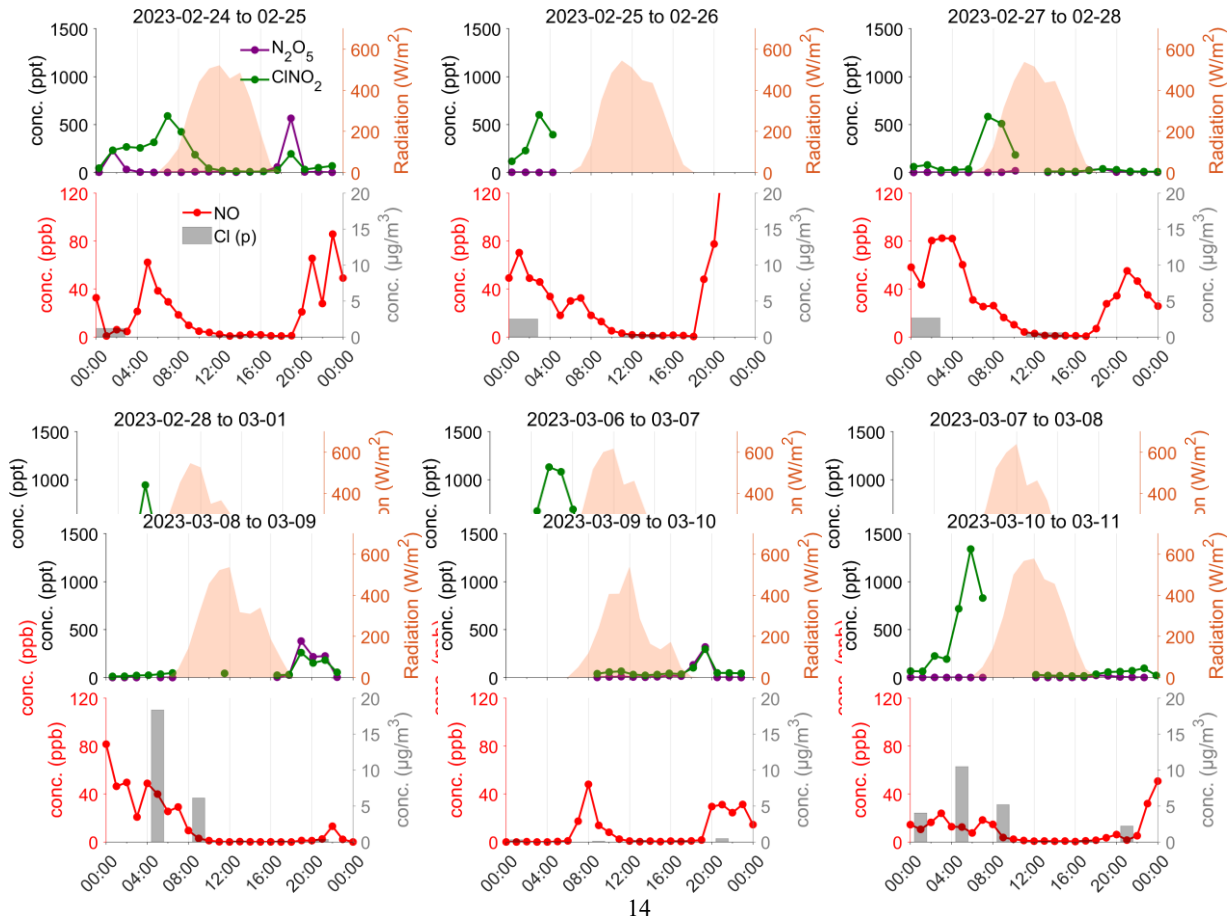


Fig. S7 Loss of NO₃· and NO₃· mixing ratios. (a) Campaign-averaged NO₃· reactivity and (b) diurnal pattern of estimated NO₃· mixing ratios. The NO₃ mixing ratio was estimated by using the observed N₂O₅ and NO₂ mixing ratio and the temperature-dependent equilibrium constant between NO₂, NO₃, and N₂O₅.



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260 **Fig. S8 Overview of the field observations.** Time series of N_2O_5 , ClNO_2 , solar
 261 radiation, NO , NO_2 , gaseous HCl , $\text{PM}_{2.5}$, and O_3 , and Cl element concentrations in
 262 $\text{PM}_{2.5}$.



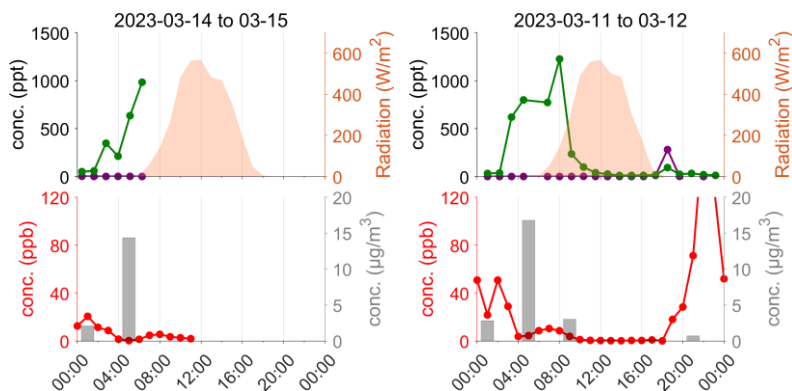


Fig. S9 Day by day plots of the enhanced cases observed in 2023. An enhanced case is identified by a pronounced nighttime increase in either N_2O_5 or ClONO_2 .

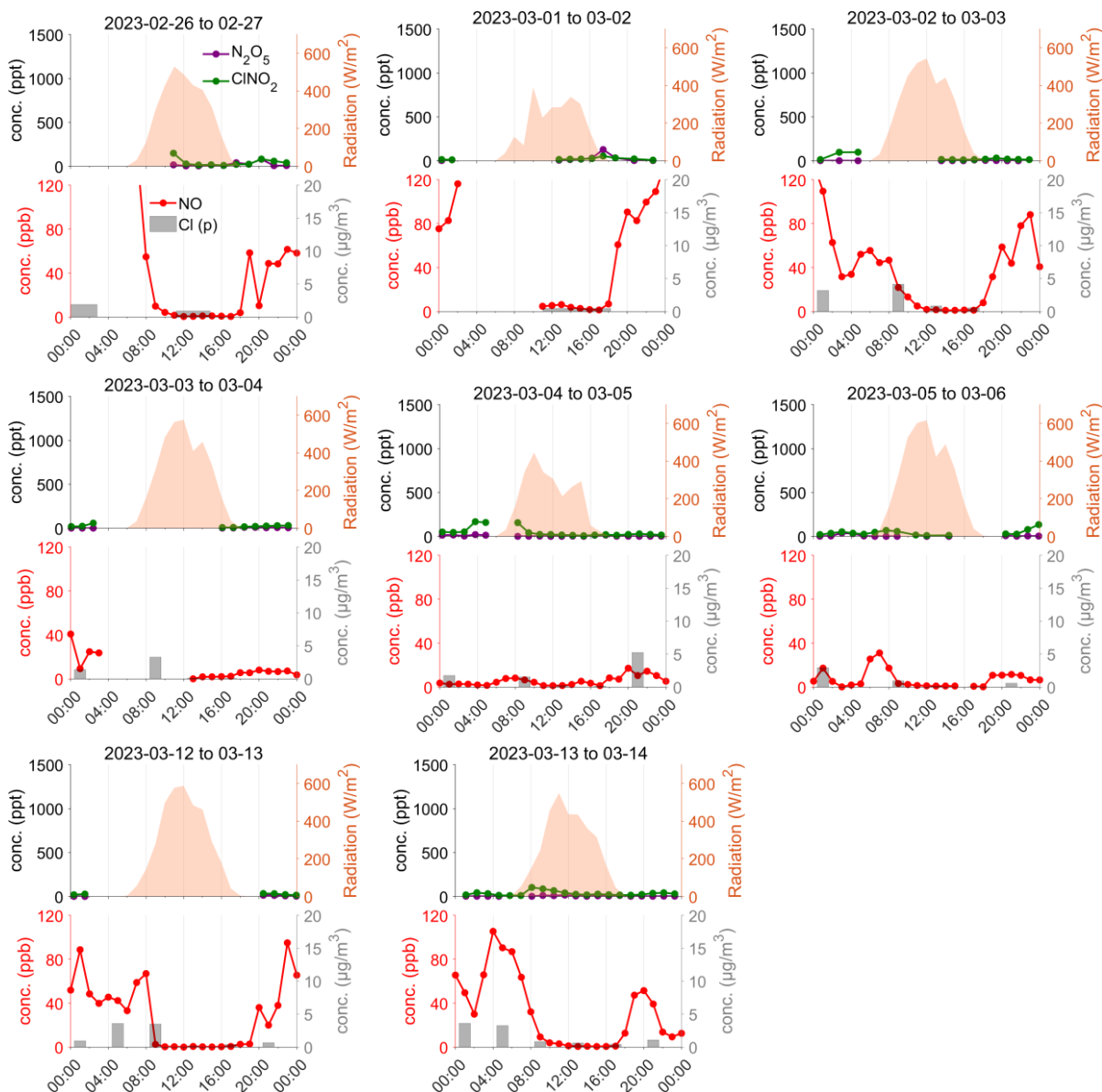


Fig. S10 Day by day plots of the non-enhanced cases observed in 2023. A non-

enhanced case is identified when neither N_2O_5 nor ClNO_2 shows a pronounced nighttime increase.

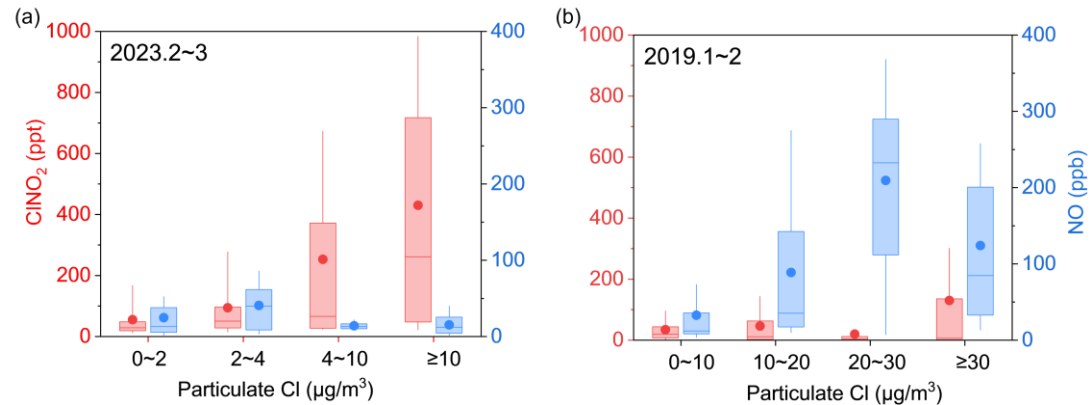


Fig. S11 Dependence of nocturnal ClNO_2 on chloride and NO during the (a) 2023 and (b) 2019 campaign. ClNO_2 and NO were stratified by chloride concentration intervals. Boxes and whiskers represent 25th and 75th, 10th and 90th percentiles, respectively. The dots and lines inside the box denote average and median values.

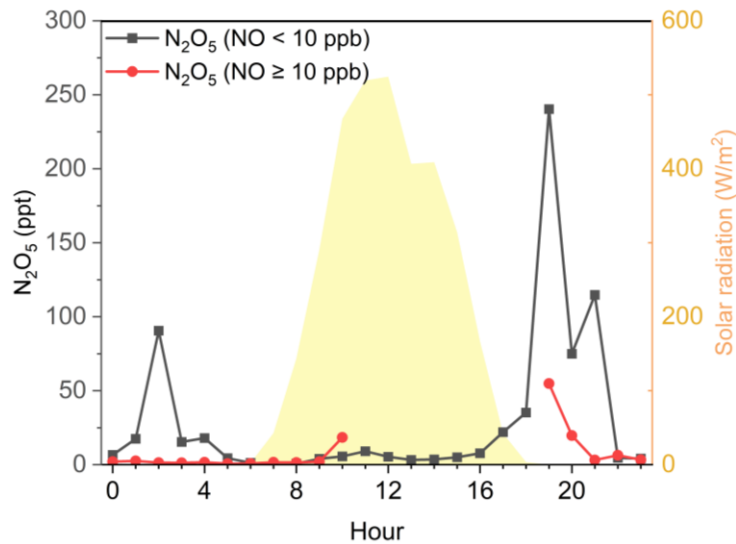


Fig. S12 Average diurnal variations of (a) N_2O_5 under low- and high-NO conditions (b) $\text{PM}_{2.5}$, total measured gaseous and particulate organics in 2023. A 10 ppb of NO threshold was applied to ensure statistically robust and comparable datasets. As all NO concentrations were below 10 ppb between 11:00 and 18:00, no N_2O_5 data are available for the high-NO condition during this period. The right axis in (b) shows the average solar radiation in 2023.

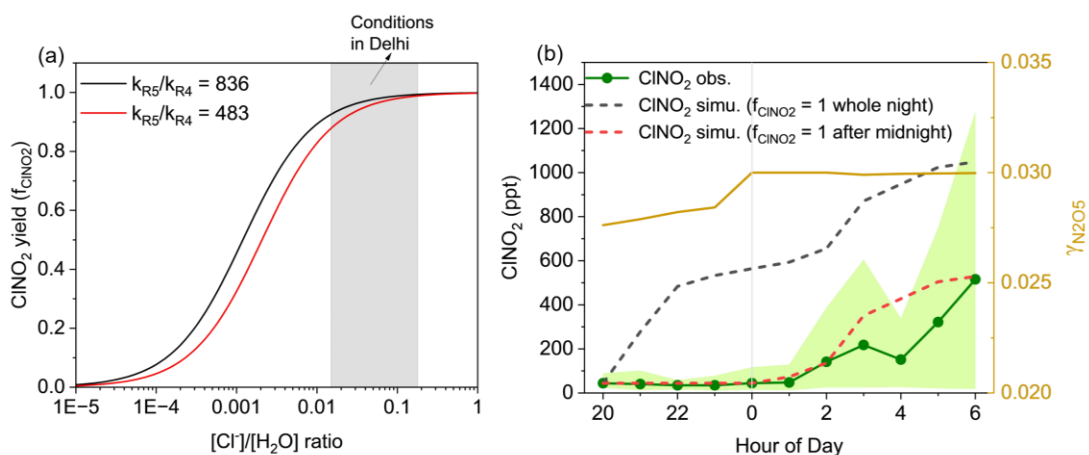


Fig. S13 (a) Parameterization of ClNO_2 yield and (b) simulation of the nocturnal ClNO_2 variations. The lines in (a) correspond to ClNO_2 yield calculated using Eq. 3 with different reported reaction rate constant ratios. The grey shaded area indicates the 10th and 90th percentiles of the calculated $[\text{Cl}^-]/[\text{H}_2\text{O}]$ ratios using ISOROPPIA from January to March of 2022 in Delhi (Ali et al., 2025). The green shaded area in (b) represents the 10th and 90th percentiles of the observed average nocturnal ClNO_2 mixing ratios. The dashed lines in (b) are the simulated ClNO_2 mixing ratios and the average RH parameterized N_2O_5 uptake coefficient is shown in the right axis of (b).

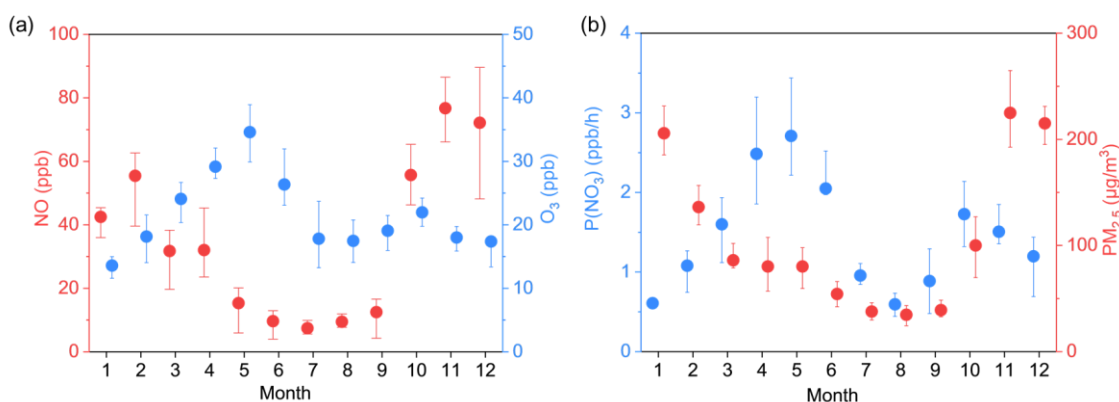


Fig. S14 Monthly variations of NO, O₃, P(NO_3), and PM_{2.5} in Delhi. Average monthly variations of (a) NO and O₃ and (b) P(NO_3) and PM_{2.5} from 2017 to 2024. The dots represent the averages and the whiskers denote the 25th to 75th percentile ranges. All data were obtained from the R.K. Puram national monitoring station.

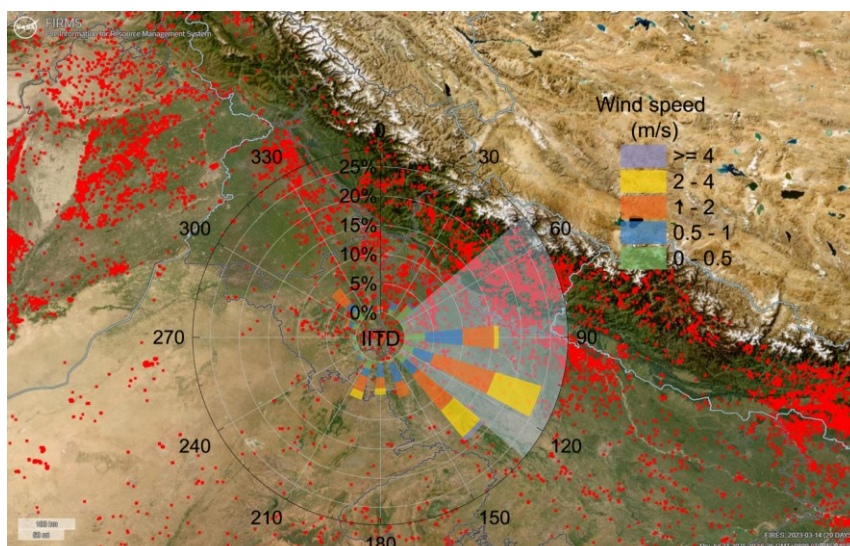


Fig. S15 Satellite observations of active fire hotspots and wind rose plot during the 2023 campaign. The map of the fire-points was downloaded from <https://earthdata.nasa.gov/firms>. The measurement site (IIT Delhi) was indicated as IITD. The light blue shaded area indicates the dominant wind-direction ($50\sim130^\circ$) when the extremely high values of the uptake parameter ($\gamma \times f$) exceeding 0.1 were measured.

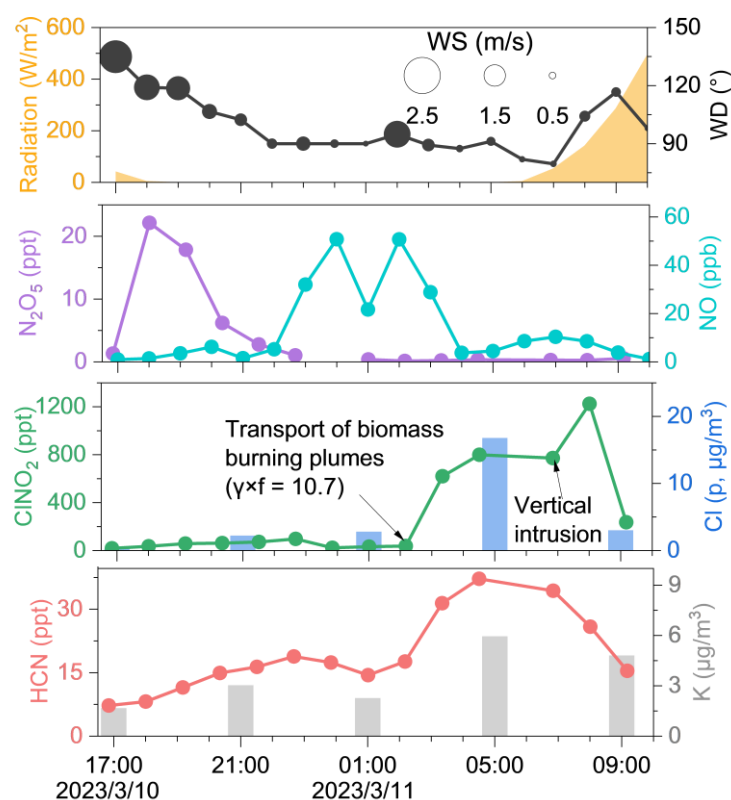


Fig. S16 A case study indicating the influence of transport on the observed ClNO_2 levels. An exceptionally high $\gamma \times f$ value (10.7) observed at 2023/3/11 2:10 is also noted.

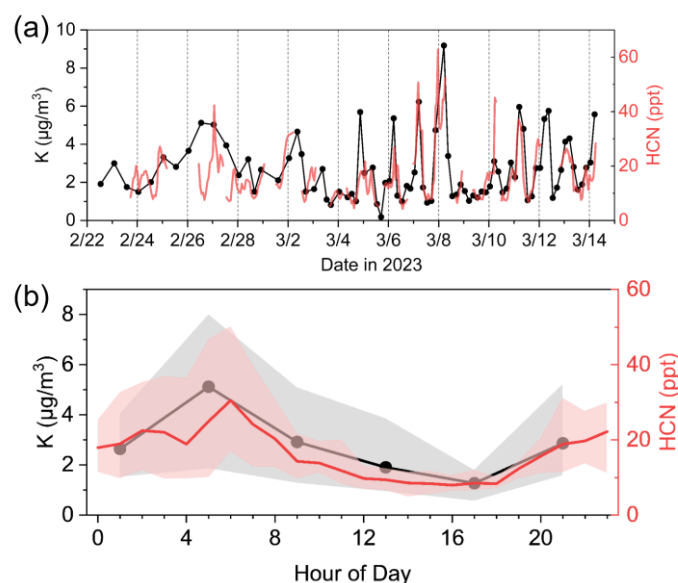


Fig. S17 (a) Time series and (b) average diurnal pattern of element K and gaseous HCN during the 2023 campaign. The shaded areas indicates 10th and 90th percentiles and the lines represent the campaign-averaged values. Hourly CIMS measurements were shown for HCN. The highest K and HCN concentrations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

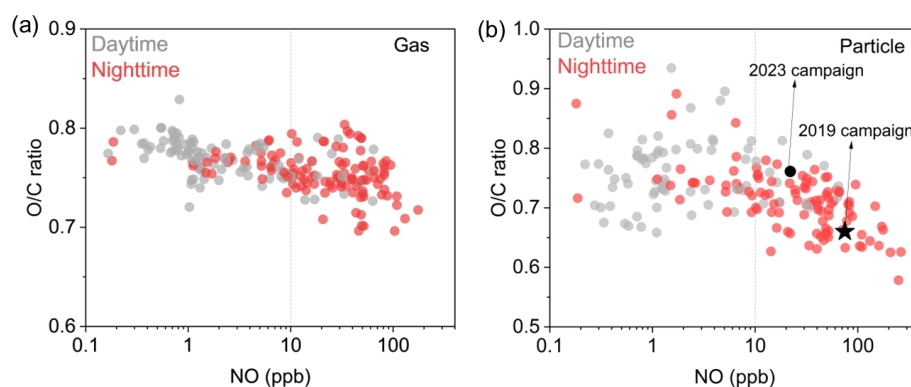


Fig. S18 NO dependence of the gaseous and particulate bulk O/C ratio in Delhi. Scatter plots showing the relationship between ambient NO levels and concentration-weighted bulk O/C ratios in the (a) gas and (b) particle phases.

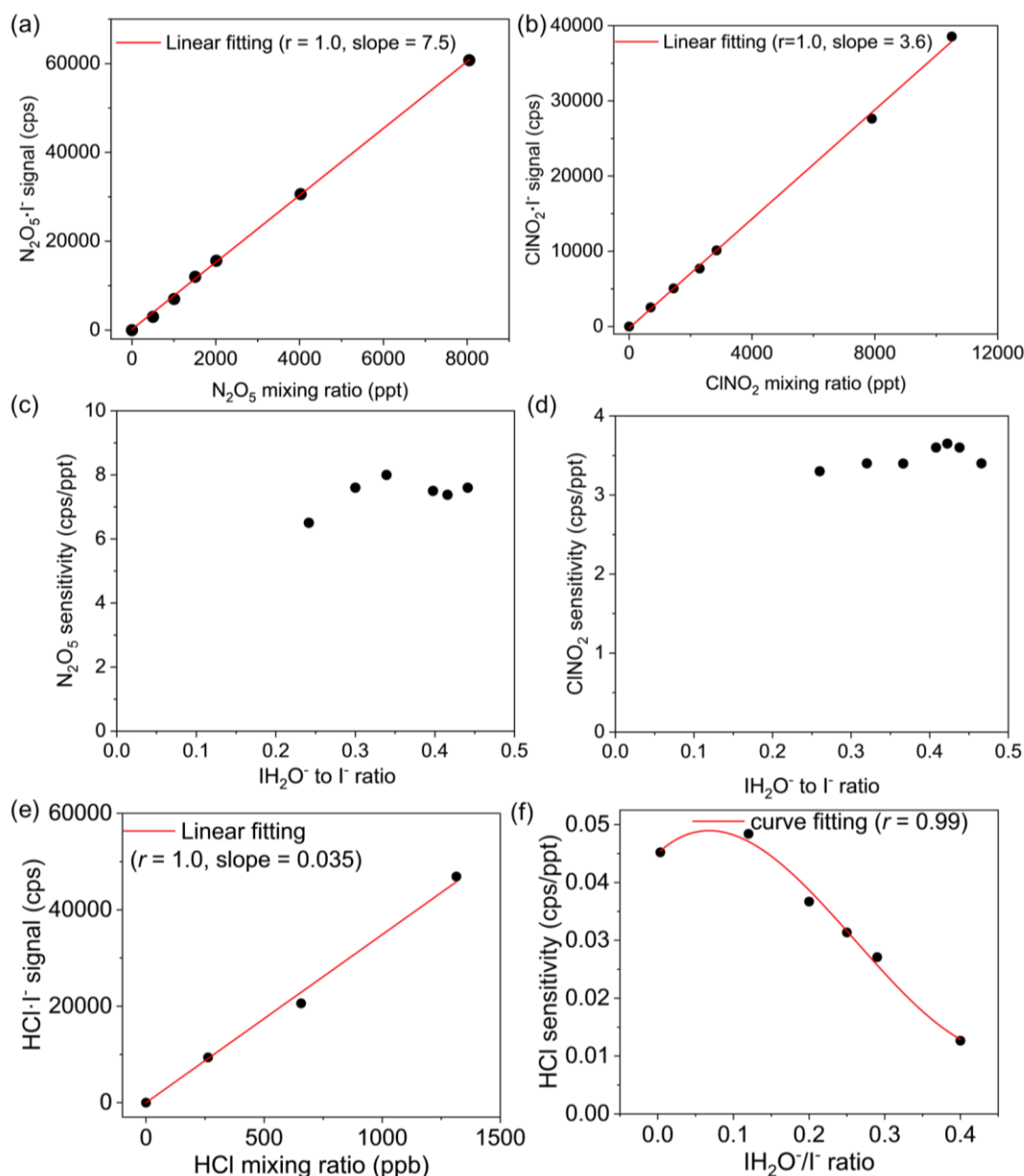


Fig. S19 Sensitivity calibrations for gaseous (a) N_2O_5 , (b) ClNO_2 , and (f) HCl, and the water dependence of the sensitivities of (c) N_2O_5 , (d) ClNO_2 , and (f) HCl. Multi-point calibrations for N_2O_5 and ClNO_2 were performed under RH conditions corresponding to an IH_2O^- to I^- ratio of 0.42 ± 0.02 , while the calibration curve at an $\text{I} \cdot \text{H}_2\text{O}^-$ to I^- ratio of 0.19 was shown for HCl.

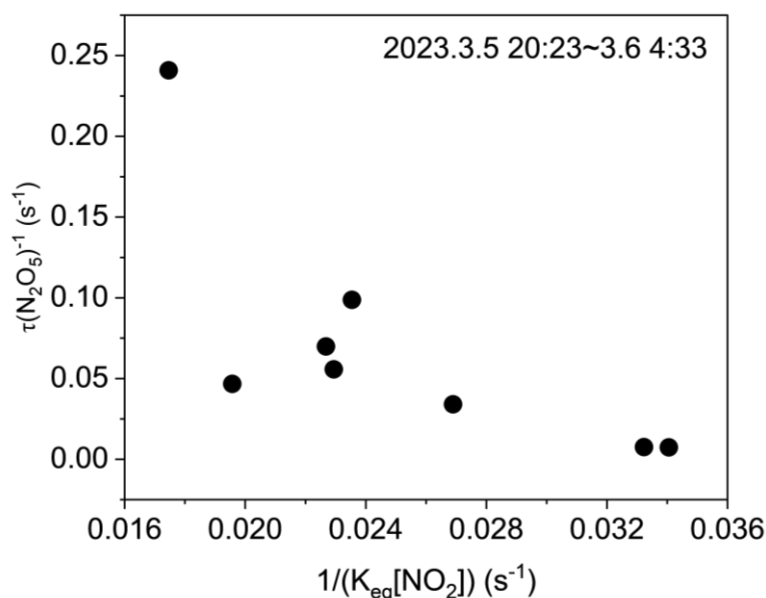


Fig. S20 An example of the steady-state analysis of the NO₂-NO₃-N₂O₅ system.

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