

1 *Supplementary Information for:*

2 **Springtime Nitrogen Oxide-Influenced Chlorine Chemistry in the Coastal Arctic**

3
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Summary (24 pages with 1 table, 2 text sections, and 13 figures)

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- **Figure S11:** Vertical potential temperature measurements from daily soundings at Utqiagvik.
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- **Figure S13:** ClNO₂ and N₂O₅ mole ratios, air temperature, and solar radiation measurements during the three polluted periods.

S1. Chemical ionization mass spectrometer (CIMS) operation

Table S1: Overview of CIMS, O₃, and NO_x measurements and figures of merit

CIMS-Specific Parameters						
Species	Ions Quantified (m/z) ^a	3σ LOD (ppt)	3σ LOD 10 min avg. (ppt)	Measurement Uncertainty % + LOD, (ppt)	In-field Sensitivity (Hz ppt ⁻¹) ^b	Relative Sensitivity (to Cl ₂ or Br ₂)
ClO	³⁵ ClO ⁻ m/z 178	2	1	47% + 2		0.3 ± 0.1 ^c
HNO ₃ (after Apr 8) ^f	IHNO ₃ ⁻ m/z 190	19	10	30% + 19		3.8 ± 0.9 ^d
Cl ₂	³⁵ Cl ³⁷ Cl ⁻ m/z 199	0.8	0.5	22% + 0.8	14 – 22	
HO ₂ NO ₂	IHO ₂ NO ₂ ⁻ m/z 206	15	8	30% + 15		0.025 ± 0.001 ^e
ClNO ₂	³⁵ ClNO ₂ ⁻ m/z 208	0.3	0.2	22% + 0.3		5.6 ± 0.6 ^d
N ₂ O ₅ (after May 8) ^g	IN ₂ O ₅ ⁻ m/z 235	1	0.8	24% + 1		1.3 ± 0.1 ^d
BrCl	⁸¹ Br ³⁵ Cl ⁻ and ⁷⁹ Br ³⁷ Cl ⁻ m/z 243	3	2	39% + 3		1.6 ± 0.5 ^d
Br ₂	⁷⁹ Br ⁸¹ Br ⁻ m/z 287	1	0.8	29% + 1	36 – 67	
Additional Measurements						
Species	Instrument	LOD		Dates available		
O ₃	2B Instruments, Model 205	1 ppb		Mar 4 – May 20		
NO	Thermo Scientific, Model 42i	0.4 ppb		Mar 11-20, Mar 25-Apr 2, May 11-19		
NO ₂	MAX-DOAS ^{ref 1–3}	2.5 x 10 ¹⁵ molec. cm ⁻²		Mar 4 – May 20		

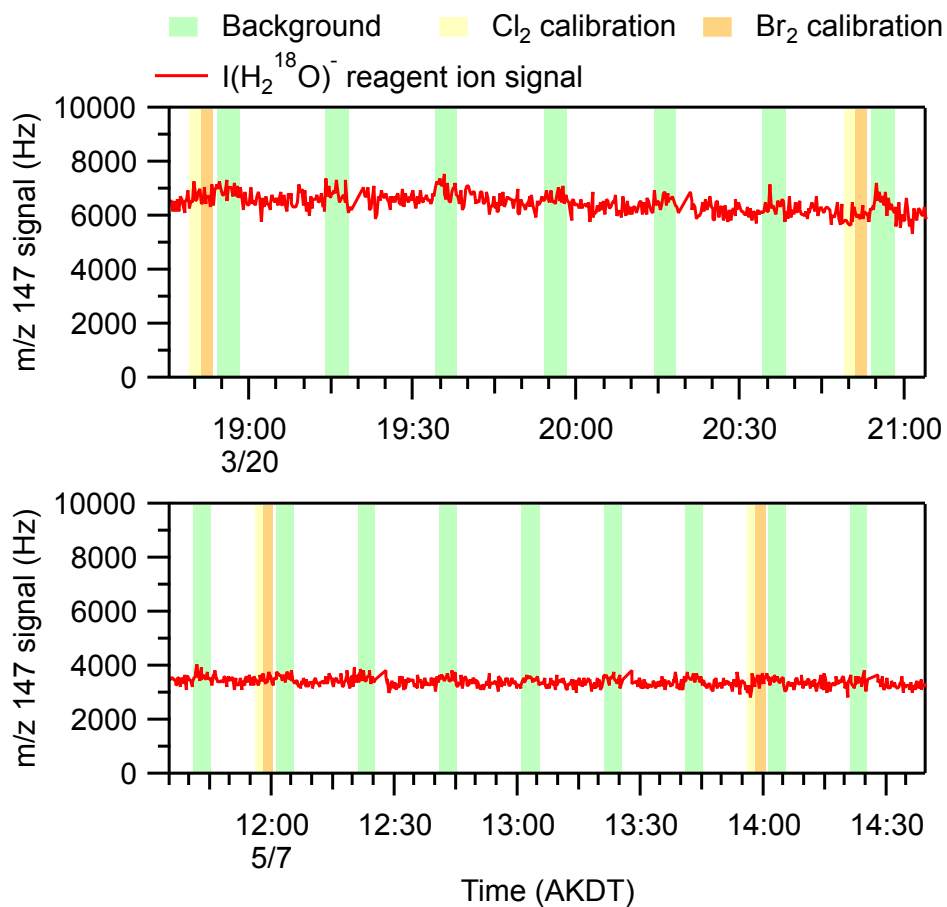
^aAll dwell times were 500 ms except for IHNO₃⁻ at m/z 190, which was 200 ms.

^bRange is mean ± calculated uncertainty for the campaign.

^cRelative to Cl₂ (m/z 197). ^dRelative to Cl₂ (m/z 199). ^eRelative to Br₂ (m/z 287).

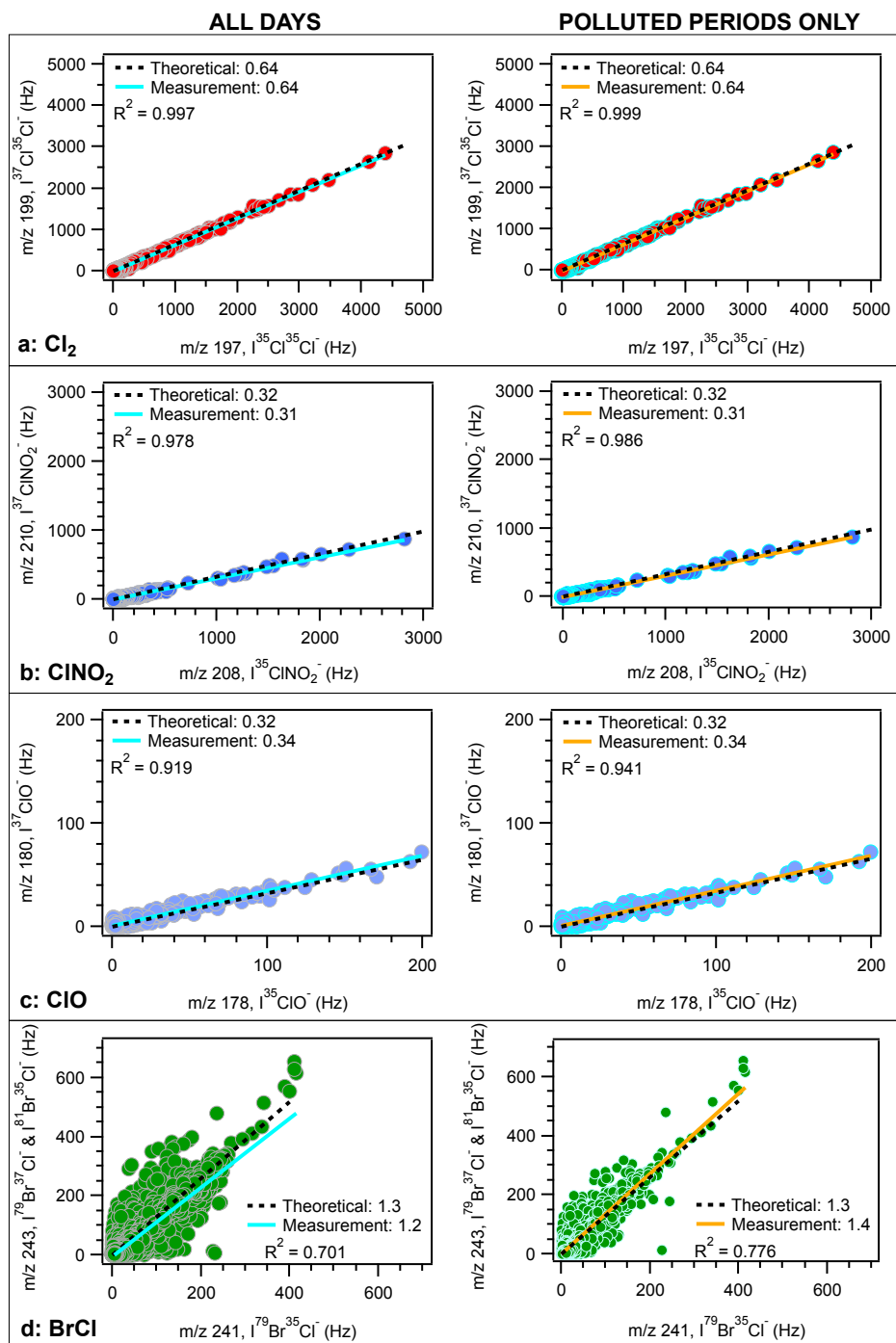
^fDue to high instrument backgrounds, only HNO₃ data from after Apr 8 are presented (**Figure S12**)

^gFor N₂O₅ quantified by hourly mass scans (Mar 4 – May 8), the LOD was estimated as 12 ppt.



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63 **Figure S1:** Reagent ion signal, monitored at m/z 147 as $\text{I}(\text{H}_2^{18}\text{O})^-$, during ambient sampling,
 64 backgrounds, and calibrations at two example periods during the campaign.



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66 **Figure S2:** 10 min averaged measured signals (background subtracted) of (A) $m/z\ 199$ vs $m/z\ 197$,
 67 (B) $m/z\ 210$ vs $m/z\ 208$, (C) $m/z\ 180$ vs $m/z\ 178$, and (D) $m/z\ 243$ vs $m/z\ 241$ showing isotopic
 68 ratios used to identify Cl_2 , ClNO_2 , ClO , and BrCl , respectively, from March 4 – May 20, 2016 (*left*
 69 *column*) and during the three polluted periods only (*right column*). The black dashed lines are the

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70 theoretical isotopic ratios and the solid lines are the measured ratios. For ClO in (C), all data
71 observed to be impacted by a mass interference at m/z 180 are not included here.

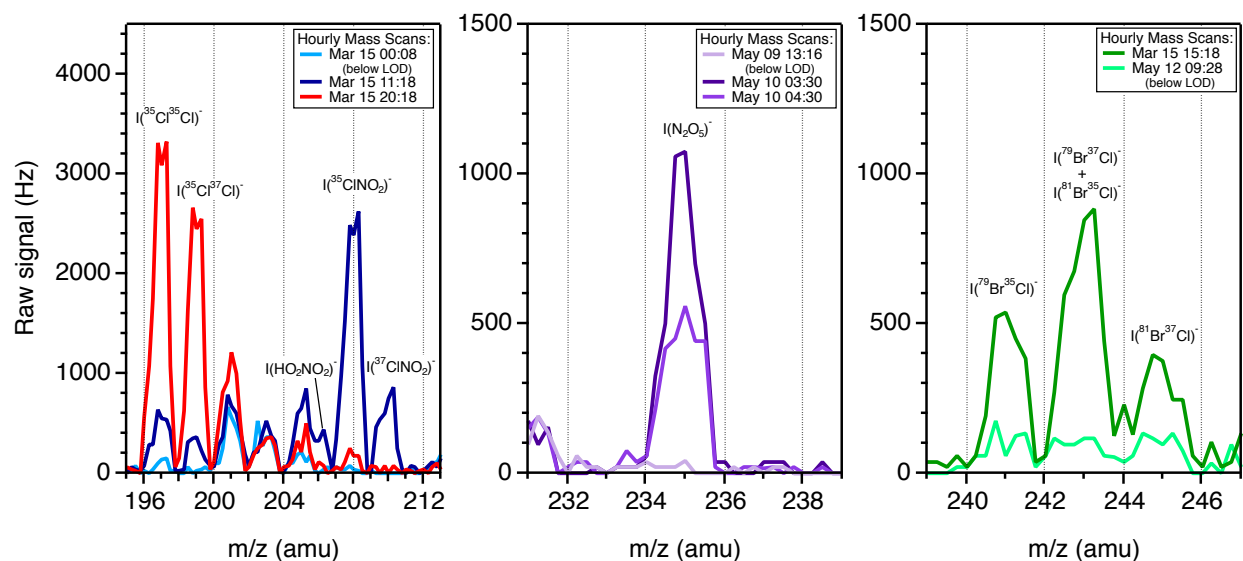
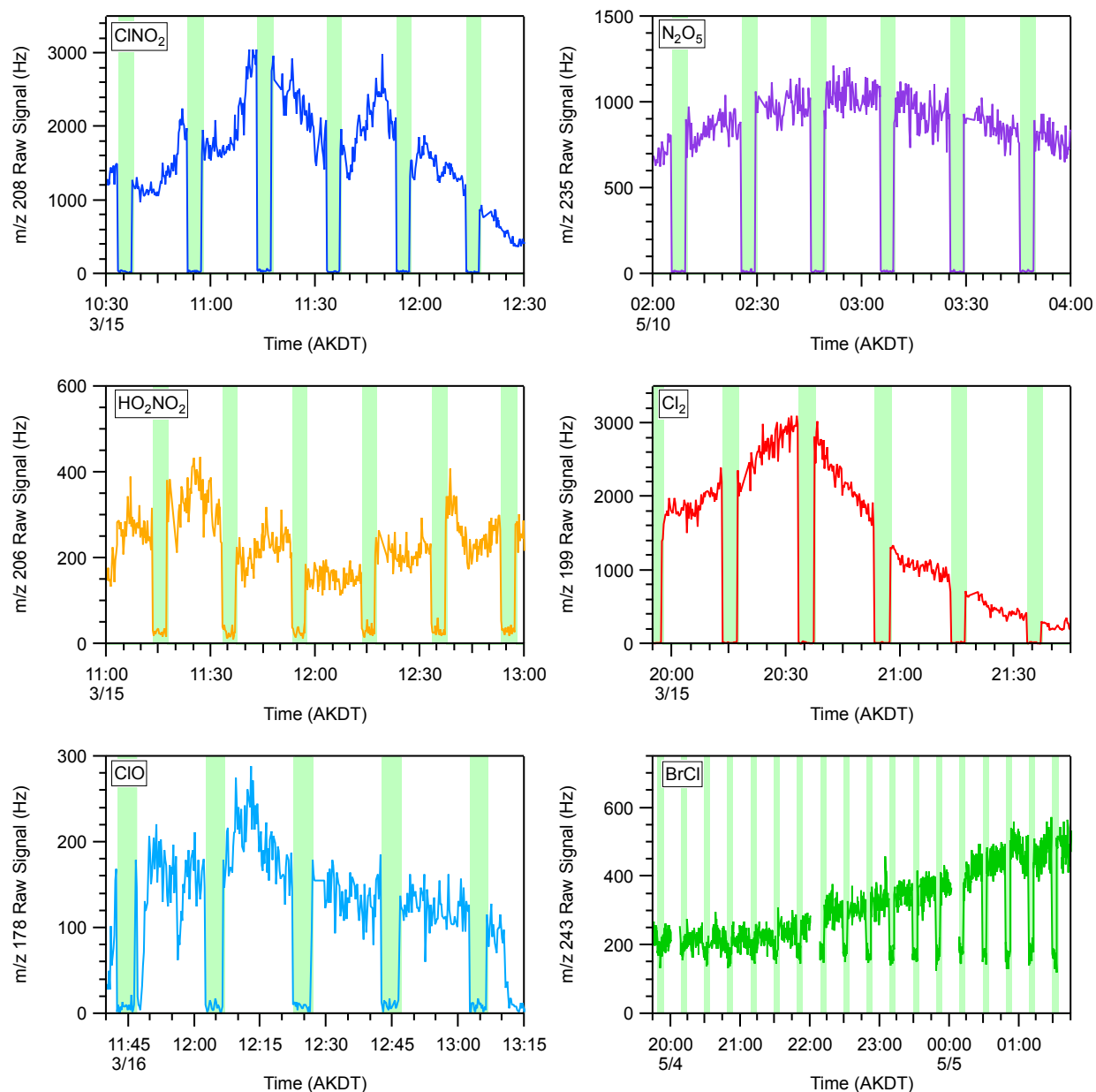


Figure S3: Representative mass spectra (at m/z 195-213, m/z 231-239, and m/z 239-247) comparing when Cl_2 , HO_2NO_2 , ClNO_2 , N_2O_5 , and BrCl were abundant versus times when signals were below detection limits (noted as “(below LOD)” in plot legends). All mass scans used a 0.25 amu step size and 100 ms dwell time.



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79 **Figure S4:** Performance of the glass wool scrubber during high-abundance periods of CINO₂,
 80 N₂O₅, HO₂NO₂, Cl₂, ClO, and BrCl, with background periods shown by green shading. A constant
 81 interference was present from m/z 241 – m/z 245 that was not removed by the scrubber, leading to
 82 the elevated signal during backgrounds for BrCl. However, these background signals were not

83 consistent with BrCl isotopic ratios and therefore did not impact the scrubber's ability to remove
84 BrCl.

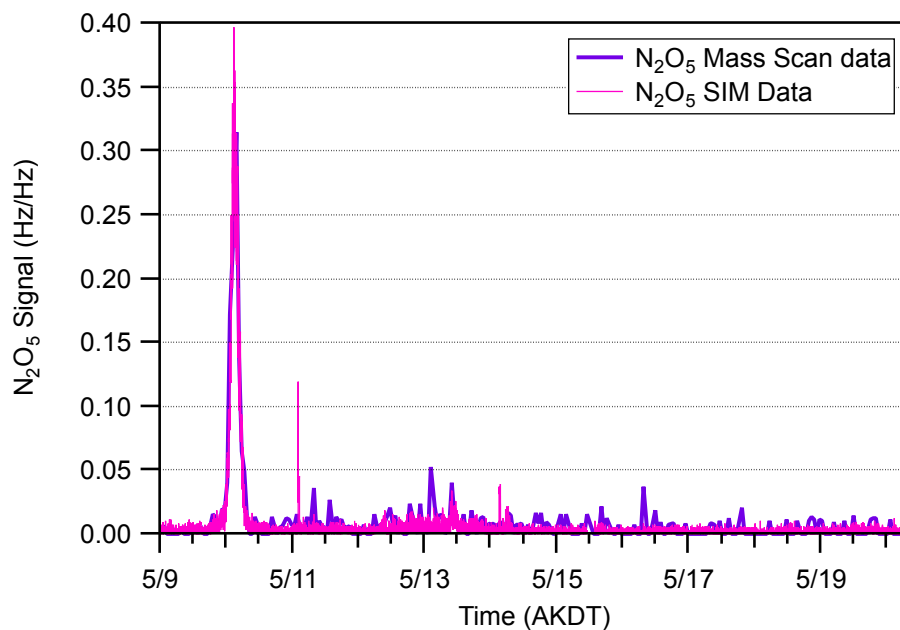


Figure S5: Comparison of N_2O_5 mass scan and selected ion monitoring (SIM) data, during the period of overlap from May 9-20, 2016. The estimated N_2O_5 mass scan LOD of 12 ppt was estimated using the SIM background period (4 min, LOD = 1 ppt), while accounting for the shortened mass scan dwell time (100 ms). Since the signal variation is assumed to be dominated by counting statistics,⁴ we account for the difference in dwell times by taking the square root of this factor (the 100 ms mass scan is a factor of 80 shorter than the total dwell time during the 4 minute SIM background period) to estimate the mass scan LOD.

S2. Identification of halogen species using isotopic ratios

The measured isotopic ratios of the halogen species were used to confirm their identities. Signals of two isotopes for the same species (e.g., $\text{I}^{35}\text{Cl}^{37}\text{Cl}^-$ vs. $\text{I}^{35}\text{Cl}^{35}\text{Cl}^-$ for Cl_2), averaged to 10 minutes, were plotted against each other and the slope of a linear regression was compared to the theoretical isotopic ratio (**Figure S2**). We report the following measured ratios, in comparison to the theoretical ratios, for all days of the campaign: For m/z 199 vs m/z 197, the measured ratio of 0.64 ($R^2 = 0.997$) confirmed the identity of Cl_2 (theoretical = 0.64). For m/z 210 vs m/z 208, the measured ratio of 0.31 ($R^2 = 0.978$) confirmed the identity of ClNO_2 (theoretical = 0.32). For m/z 180 vs m/z 178, the measured ratio of 0.34 ($R^2 = 0.919$) confirmed the identity of ClO (theoretical = 0.32). A mass interference at m/z 180 ($\text{I}^{37}\text{ClO}^-$) was observed, which resulted in ClO not being quantifiable from March 25 – April 30, when the ratio of m/z 180 to m/z 178 ($\text{I}^{35}\text{ClO}^-$) varied by >30%. For m/z 243 vs m/z 241, the measured ratio of 1.2 ($R^2 = 0.701$) confirmed the identity of BrCl (theoretical = 1.3).

S3. Offline calibrations of ClNO₂, N₂O₅, ClO, BrCl, HO₂NO₂, and HNO₃

S3.1. ClNO₂

Nitryl chloride (ClNO₂) was calibrated using a modified procedure of *Thaler et al.*⁵ In the laboratory, ClNO₂ was synthesized by passing 0 – 50 ppb Cl₂ over a 1.0 mL bed of 0.01 M NaNO₂ solution. The synthesized ClNO₂ was then passed through a 300°C quartz tube oven, where it was completely thermally dissociated (complete loss of ClNO₂ signal at *m/z* 208 and 210). The resulting NO₂ was quantified using a custom NO_x analyzer⁶ equipped with a blue light-emitting diode to photochemically reduce NO₂ to NO for measurement.⁷ The ClNO₂ sensitivity (at *m/z* 208) relative to Cl₂ (at *m/z* 199) was determined to be 5.6 ± 0.6 .

S3.2. N₂O₅

Dinitrogen pentoxide (N₂O₅) was calibrated using a modified procedure of *Bertram et al.*⁸ N₂O₅ was synthesized by combining flows of NO₂ and O₃ in a 61 cm long, 2.2 cm diameter PFA tube with a 1.5 min residence time (flow rate of 2.7 cm³ s⁻¹). N₂O₅ was quantified using the custom NO_x analyzer described in Section S3.1^{6,7} by measuring the amount of NO₂ reacted upon the addition of O₃, assuming two moles of NO₂ are reacted to form one mole of N₂O₅ (R7-R8). Loss of NO₂ due to excess NO₃ formation was negligible. For NO₂ at 40 ppb within the PFA tube during the calibration, the [NO₃] / [N₂O₅] ratio was calculated to be 0.04. The N₂O₅ sensitivity (at *m/z* 235) relative to Cl₂ (at *m/z* 199) was determined to be 1.3 ± 0.1 .

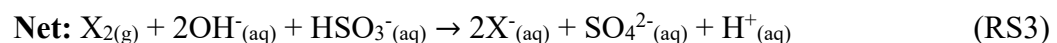
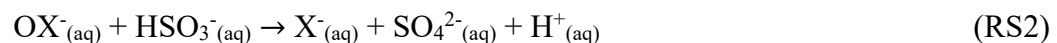
S3.3. ClO

The relative sensitivity of 0.3 ± 0.1 for chlorine monoxide (ClO) (*m/z* 178) / Cl₂ (*m/z* 197) determined by *Custard et al.*⁹ was applied for the ambient measurements for this study. For this calibration, ClO was synthesized in a quartz flow tube via the photolysis of Cl₂ with excess O₃ (90

ppb) and ~5 ppb of NO₂.⁹ The steady-state ClO concentration was determined using the measured photolysis rate of Cl₂ and measurements of Cl₂ and NO₂ mole ratios.⁹

S3.4. BrCl

Bromine chloride (BrCl) was synthesized by passing Cl₂ over a 1 mL bed of 1 M NaBr solution in a glass reaction vessel, similar to *Huff et al.*¹⁰ The output of the reaction vessel was predominately BrCl, Br₂, and the remaining Cl₂, as measured by CIMS. It is likely the observed Br₂ was a product of a subsequent reaction of BrCl and bromide (Br⁻).¹⁰ The reaction output was bubbled into 10 mL of sodium bicarbonate/sodium metabisulfite buffer solution in a glass impinger for 40 min, allowing the following reactions with the molecular halogens to occur (RS1 –RS3, where X = Cl or Br):



The total chloride concentration of the buffer solution following bubbling of the reaction output was measured using a Dionex ICS-2100 ion chromatograph. A blank solution (N₂ bubbled only) was also collected as a background measurement, and the background Cl⁻ (accounting for ~4-10%) was subtracted from the total measured Cl⁻ in the reaction output solution. Following the background subtraction, the Cl⁻ resulting from the remaining Cl₂ was subtracted from the total measured Cl⁻ using the Cl₂ signal measured by CIMS in parallel. The BrCl was then quantified as the remaining chloride in the solution following subtraction of the chloride from unreacted Cl₂ and the blank (background Cl⁻). The headspace of the impinger during the bubbling period was monitored with CIMS, and no Cl₂, BrCl, or Br₂ was observed; therefore, the output of the reaction

vessel was sufficiently captured by the buffer solution. The resulting BrCl sensitivity (at m/z 243) relative to Cl₂ (at m/z 199) was determined to be 1.6 ± 0.5 .

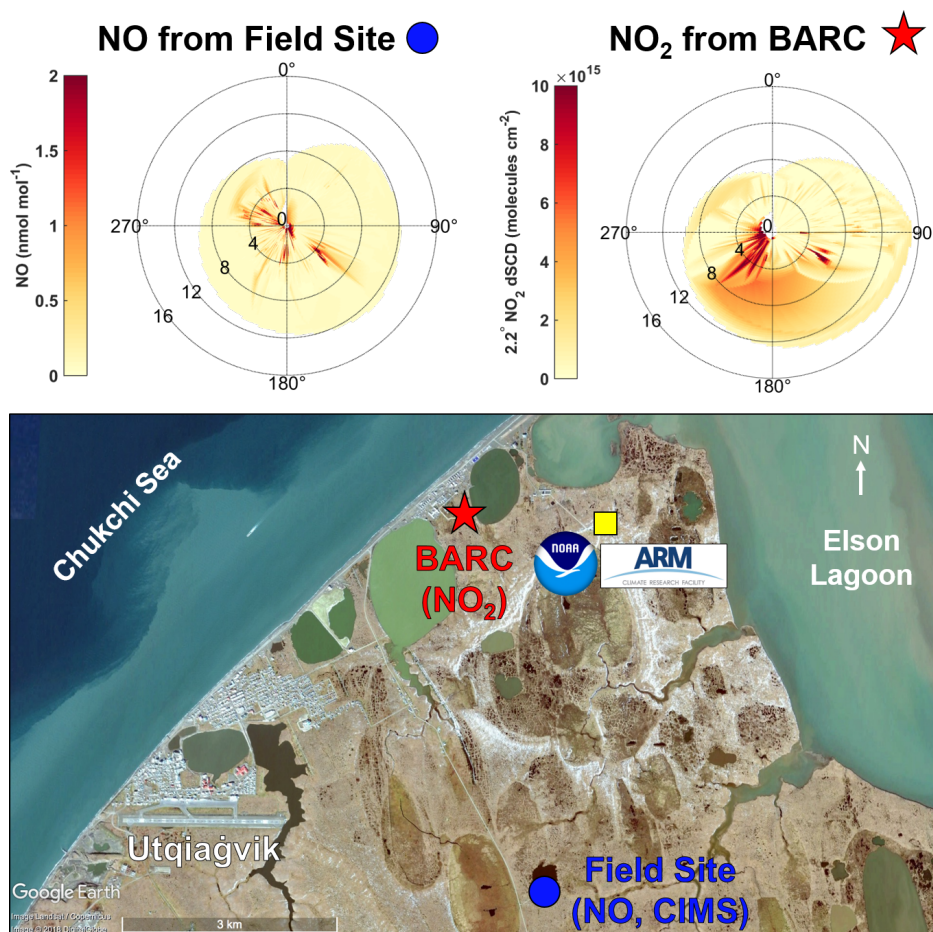
S3.5. HO₂NO₂

Peroxynitric acid (HO₂NO₂) was synthesized following *Appelman and Gosztola*¹¹ and was obtained in the gas-phase by bubbling N₂ through an impinger containing the synthesized peroxynitric acid in a solution of hydrogen peroxide on ice. The HO₂NO₂ was quantified using the NO₂ generated from its thermal decomposition at 140°C within a 91 cm Teflon tube, and it was assumed that the thermal decomposition of each HO₂NO₂ molecule led to the formation of one NO₂ molecule. The NO₂ was monitored using a custom chemiluminescence NO_y instrument, constructed following previous methods^{12–14} and calibrated using a NO₂ standard cylinder (Praxair).

S3.6. HNO₃

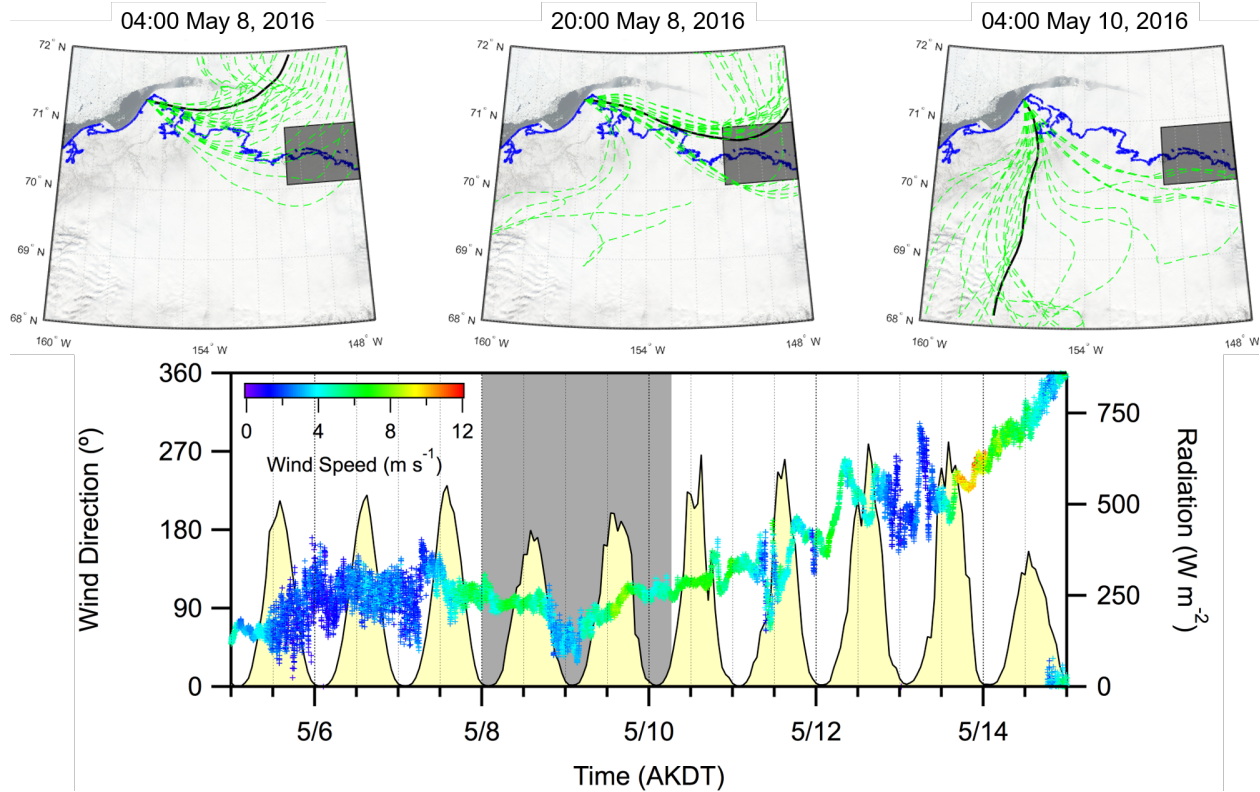
Nitric acid (HNO₃) was calibrated in the laboratory by adding to the CIMS inlet flow 0.2 L min⁻¹ of HNO₃ (in N₂) from a 50 ng min⁻¹ HNO₃ permeation source (VICI Metronics, Inc.). The HNO₃ mole ratio was varied from 0.8 – 9.7 ppb to obtain a calibration curve. The HNO₃ permeation rate was confirmed by bubbling the HNO₃ flow into 4 mL of micropure water, and the resulting nitrate concentration was quantified using a Dionex ICS-2100 ion chromatograph. The HNO₃ sensitivity (at m/z 190, IHNO₃⁻) relative to Cl₂ (at m/z 199) was determined to be 3.8 ± 0.9 .

169 **S4. Additional data figures**



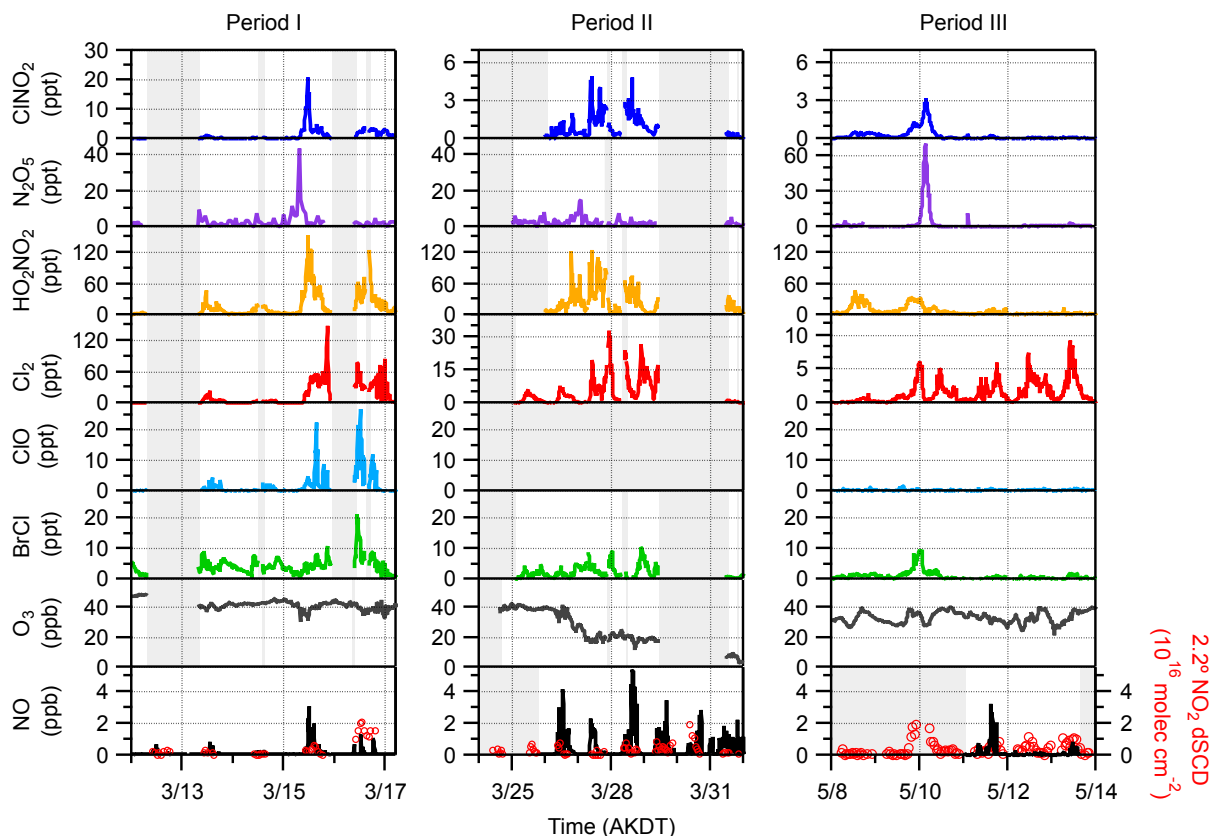
170
171 **Figure S6:** Wind rose plots, colored by NO mole ratios (*top-left*) and NO₂ differential slant column
172 densities (dSCDs, *top-right*) and a map of the region (*bottom*). *In situ* NO was measured at the
173 main field site (blue circle), located ~5 km ESE of Utqiagvik, and NO₂ measured from the Barrow
174 Arctic Research Center (BARC, red star), ~5 km NW of Utqiagvik. From March 4 – 17, wind
175 direction (°) and wind speeds (m s⁻¹) measured at the NOAA Barrow Observatory (yellow square),
176 with wind measurements occurring at the main field site for the remainder of the study. The ARM
177 Research Facility is located next to the NOAA Observatory (yellow square).

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179

180 **Figure S7:** (*top panels*) Representative HYSPLIT 72-hour backward air mass trajectories (black
 181 lines) arriving at Utqiagvik at 04:00 AKDT May 8 (*left*), 20:00 AKDT May 8 (*middle*), and 04:00
 182 AKDT May 10 (*right*). Green dashed lines are ensemble members of the trajectories, which
 183 represent uncertainty. Portions of the trajectories and ensemble members travel through the gray-
 184 shaded region representing the North Slope of Alaska and Prudhoe Bay Oilfields.¹⁵ For reference,
 185 the full period of “Prudhoe Bay Influence”, defined as times when air mass trajectories traveled
 186 through the gray-shaded region, was determined to be 00:00 AKDT May 8 to 05:00 AKDT May
 187 10. (*lower panel*) Wind direction, wind speed, and solar radiation observations from May 5 – 15,
 188 2016, with the gray shading representing the Prudhoe Bay-influenced period determined from the
 189 HYSPLIT trajectories.



190

191 **Figure S8:** Observations of CINO₂, N₂O₅, HO₂NO₂, Cl₂, ClO, BrCl, O₃, NO, and NO₂ during the

192 three episodes of enhanced chlorine chemistry (March 12-16, March 24-31, and May 8-13, 2016)

193 near Utqiagvik, AK. Gray shading denotes periods without CIMS, O₃, or NO data available due to

194 site power loss and use for other experiments. During Period II, ClO could not be quantified due

195 to a mass interference.

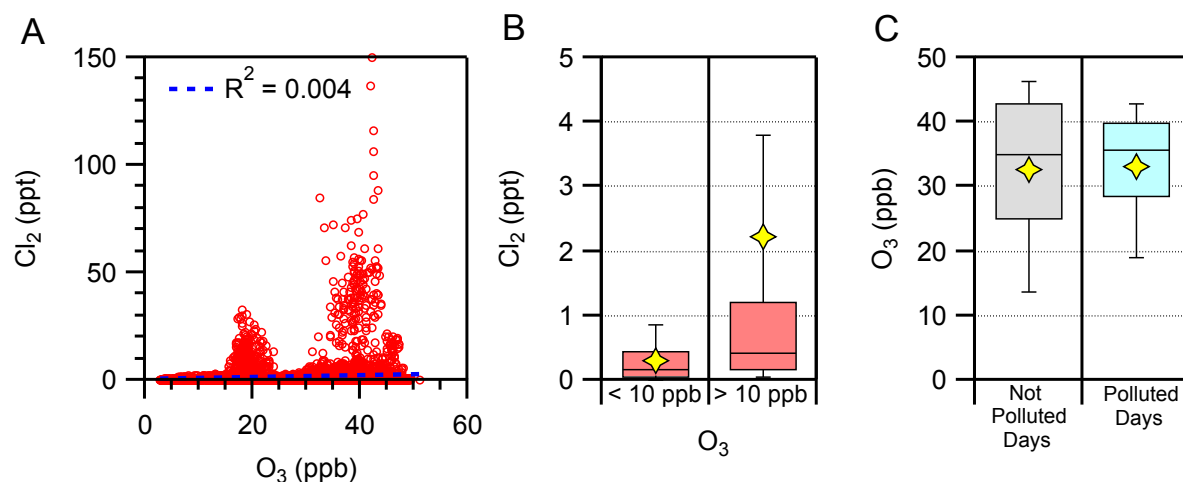
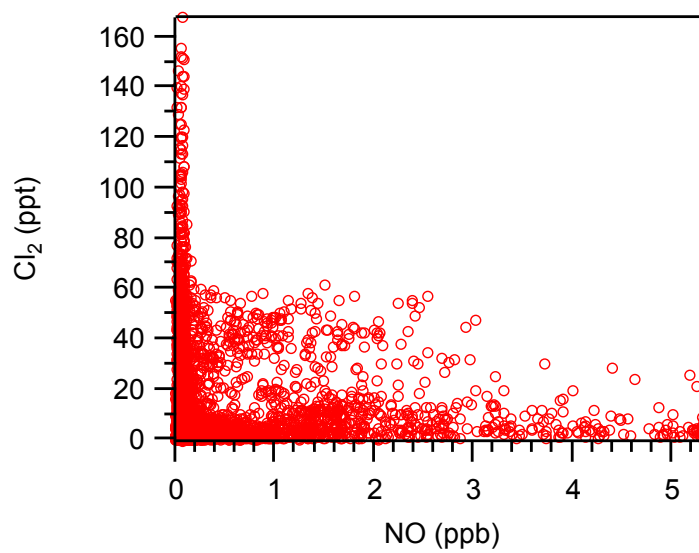


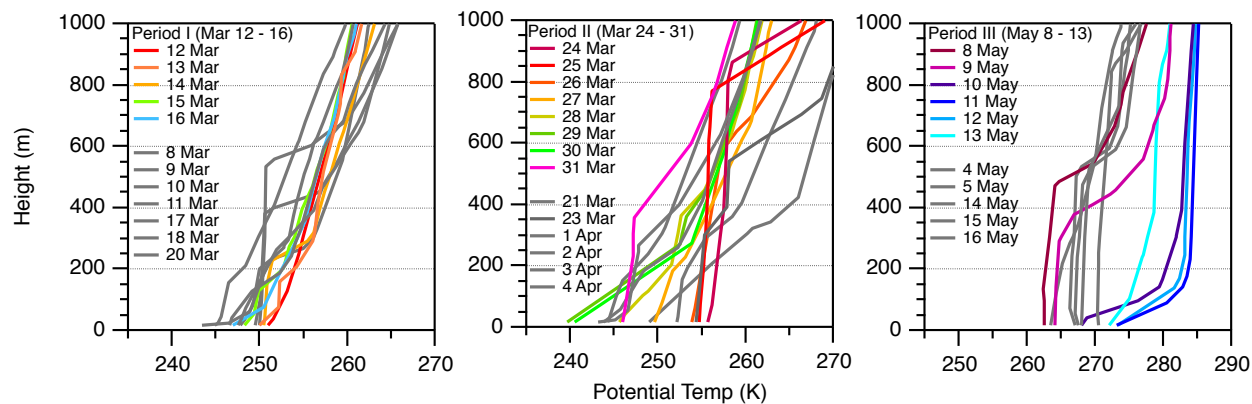
Figure S9: (A) Cl_2 mole ratios vs O_3 mole ratios (10 min averaged). Box and whisker plots show the distributions (90th/10th and 75th/25th percentiles, and medians) of (B) Cl_2 mole ratios, binned by O_3 mole ratios less or greater than 10 ppb, and (C) O_3 mole ratios, binned by Not Polluted and Polluted days (defined in Section 2.2 and used in Figure 2B). Yellow stars represent average mole ratios for the given populations. Elevated Cl_2 was observed when O_3 was near background levels (typically > 20 ppb), and Cl_2 was near zero when O_3 was depleted (< 10 ppb), as observed previously.^{9,16} Both Polluted and Not Polluted periods experienced nearly the same levels of O_3 , on average, during the campaign, showing that the levels of NO_x , rather than O_3 were likely responsible for the differences in Cl_2 mole ratios observed.



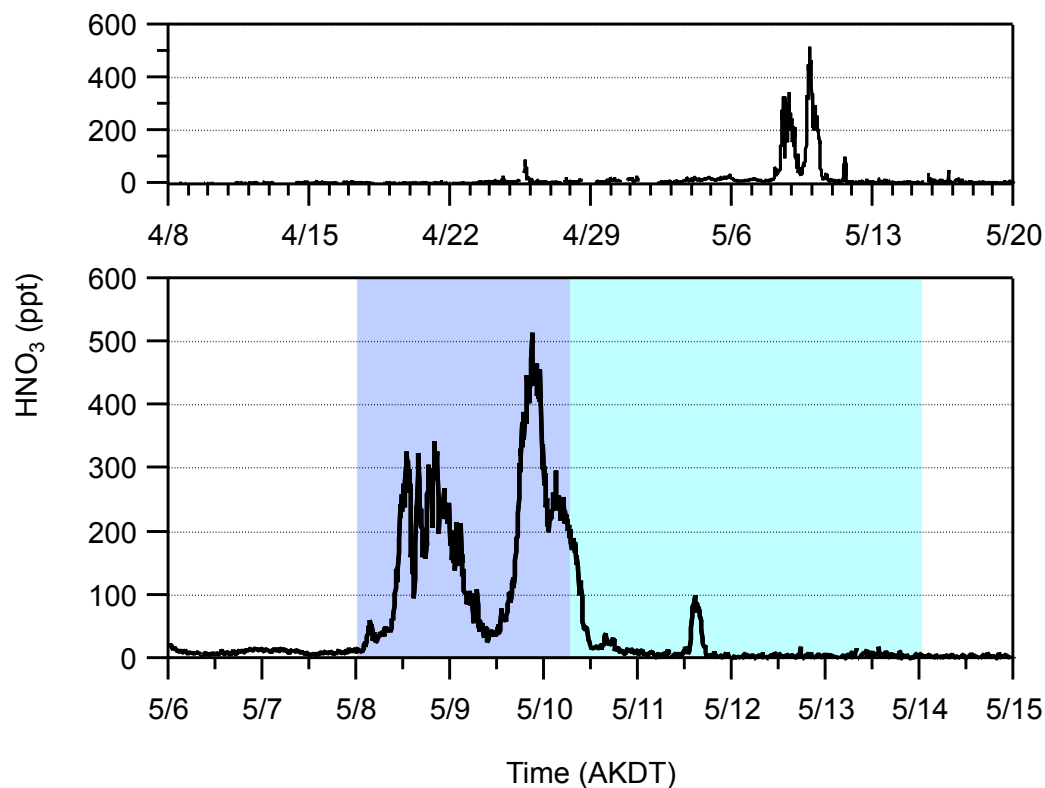
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208 **Figure S10:** 1 min averaged Cl_2 observations versus 1 min averaged NO observations ($R^2 =$

209 0.004) during PHOXMELT. Note that the NO instrument LOD is 0.4 ppb.



210
 211 **Figure S11:** Vertical potential temperature measurements from daily soundings at the PABR
 212 station in Utqiagvik. All times are 12:00 UTC (03:00 AKST). Soundings from the three polluted
 213 periods are shown with colored traces, and soundings from the surrounding days are shown with
 214 gray traces. All data are available at <http://weather.uwyo.edu/upperair/sounding.html>.



215
 216 **Figure S12:** 10 min averaged mole ratios of HNO_3 during (*top panel*) April 8 – May 20 and
 217 (*bottom panel*) Period III (blue shading), including the period influenced by the North Slope of
 218 Alaska oilfields (purple shading). The HNO_3 removal efficiency of the glass wool scrubber during
 219 backgrounds was 40-60%; therefore, the HNO_3 mole ratios shown are an underestimate.

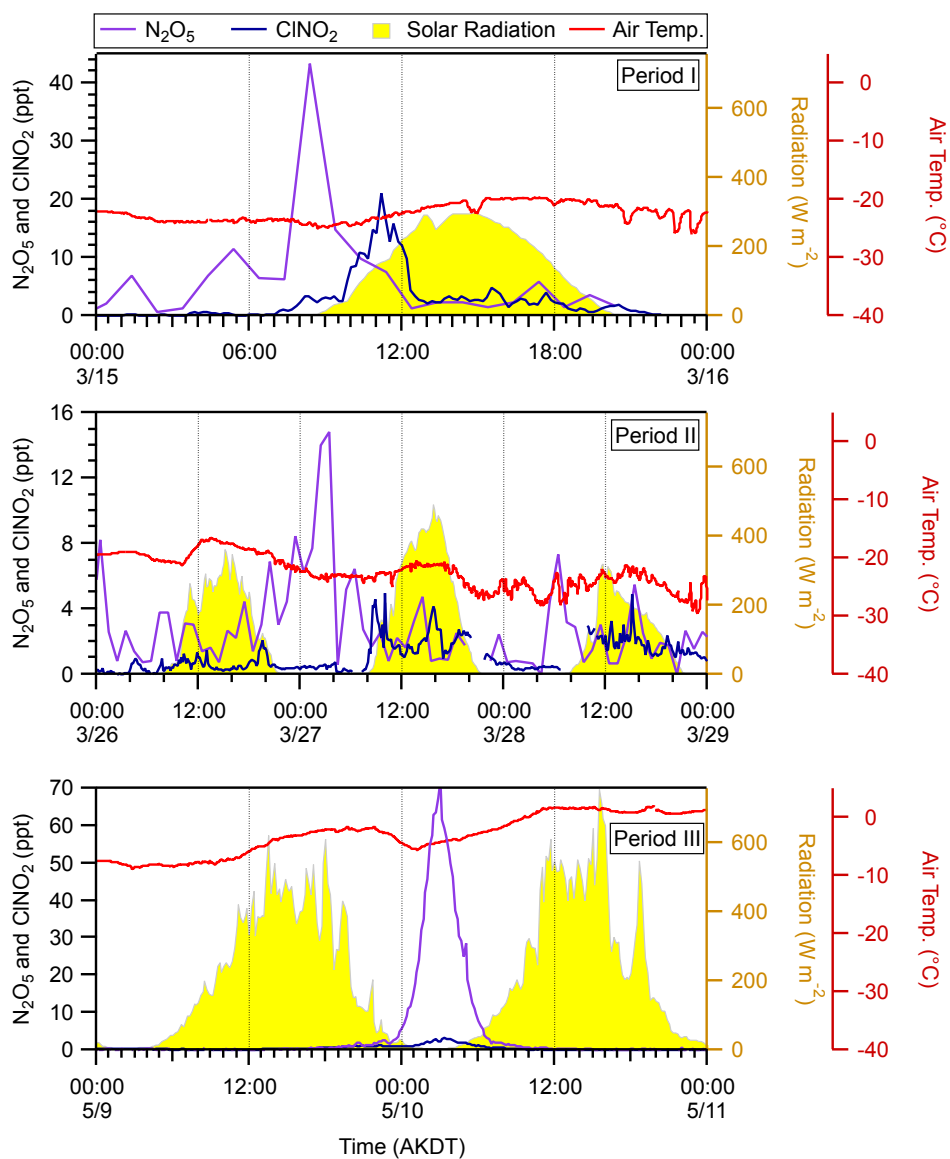


Figure S13: Mole ratios of N_2O_5 and ClNO_2 during Period I (*top panel*), Period II (*middle panel*) and Period III (*bottom panel*), shown with measurements of solar radiation and air temperature. All ClNO_2 mole ratios were 10 min averaged, and N_2O_5 mole ratios were hourly mass scans for Periods I and II and 10 min averaged for Period III.

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