Supporting Information for "Unprecedented spring 2020 ozone depletion in the context of 20 years of measurements at Eureka, Canada"

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Introduction

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- Text S2 describes the comparisons SLIMCAT trace gas columns with measured values.

- Figure S1 shows the HF columns from the Bruker FTIR, as well as the normalized time series of ozone, NO_2, HCl, $ClONO_2$, and HNO_3.

- Figure S2 shows the absolute differences between SLIMCAT active ozone and ozone measurements from the GBS, SAOZ, Bruker FTIR, and Brewer instruments.

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- Figure S4 shows the absolute differences between SLIMCAT HCl, $ClONO_2$, and HNO_3, and the Bruker FTIR measurements.

- Figure S5 shows the relative differences between SLIMCAT HCl, $ClONO_2$, and HNO_3, and the Bruker FTIR measurements.

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Text S1. Normalizing with HF

The Bruker FTIR HF retrievals use the same settings (following NDACC recommendations) as described for the other trace gases in the main text. The mean uncertainty of the HF columns for the measurement period is 3.5%, and the mean DOFS is 2.8. We use the HF columns as a tracer of vertical motion in the vortex (after Lindenmaier et al., 2012). HF displays a significant increasing trend in the Arctic stratosphere (e.g. Griffin et al., 2017), likely due to an increase in its source gases. To estimate the HF trend for the Bruker FTIR, we calculated yearly averages of HF columns outside the vortex (after Griffin et al., 2017), using the vortex criteria described in the main text. The yearly averages were then fitted using a robust fitting method, as described in Bognar et al. (2019). The line of best fit indicates a statistically significant increase of 4.33 ± 2.46 molec cm^{-1} yr^{-1} (2.1 ± 1.2 % yr^{-1} relative to 2007) in the yearly mean HF columns for 2007-2019 (no out-of-vortex measurements in 2020). To correct for this trend, the line of best fit (yearly values) was subtracted from all HF data, using 2007 as the baseline.

To remove the impact of some of the dynamical effects, we normalized trace gas column with the nearest trend-corrected HF measurement (within a ± 2 hour time window). The time series of HF and the normalized time series of ozone, NO_2, HCl, $ClONO_2$, and HNO_3 are shown in Figure S1, and discussed in the main text. The effect of the trend correction is most evident in the HF time series. Uncorrected HF columns for 2020 are much higher than those in 2011, while the trend-corrected columns are similar for both years. The interpretation of the ratio time series does not change as a function of the trend correction.

Text S2. SLIMCAT comparisons

To compare trace gas columns from SLIMCAT to measured column values, the 6-hourly SLIMCAT output was linearly interpolated to the individual measurement times. For the comparisons, the mean absolute difference was calculated as

$$\Delta_{abs} = \frac{1}{N}\sum_{i=1}^{N}(MODEL_i - MEAS_i), \quad (1)$$

and the mean relative differences were calculated as

$$\Delta_{rel} = \frac{1}{N}\sum_{i=1}^{N}\frac{(MODEL_i - MEAS_i)}{MEAS_i} \times 100\%, \quad (2)$$

where $MEAS_i$ and $MODEL_i$ are the individual measurements and corresponding SLIMCAT values, respectively. The reported uncertainty in the figures and in the text is the standard deviation of the differences. Differences were calculated separately for measurements inside and outside the vortex, using the vortex criteria for the measurements described in the main text. Since the instruments generally look south from Eureka, most measurements that sample inside the vortex correspond to times when the vertical profile directly over Eureka is also inside the vortex. For the ozone loss estimates in 2011 and 2020, vortex criteria were tested explicitly for SLIMCAT columns as well (see main text). In the following we present comparisons of SLIMCAT ozone, HCl, $ClONO_2$, and HNO_3 to GBS, SAOZ, Bruker FTIR and Brewer measurements, using all measurements from each instrument. SLIMCAT simulates OClO, BrO and NO_2 (the other trace gases of interest) as well, but comparison of these results is not straightforward given the large diurnal variation of each trace gas, and the coarse temporal resolution (6 hours) of the SLIMCAT output.

Figures S2 and S3 show absolute and relative differences between SLIMCAT active ozone and measured ozone as a function of time of year and vortex location. SLIM-

CAT generally agrees well with (and slightly underestimates) measurements inside the vortex. The mean relative differences are 1.4%, -3.9%, -8.9%, and -4.0% for the GBS, SAOZ, Bruker FTIR and Brewer data, respectively. The changes in the mean relative differences are consistent with the agreement between the various instruments (Bognar et al., 2019). The SLIMCAT results show the largest deviations when compared to Bruker FTIR measurements in late February. This peak is in large part the consequence of differences in spatial sampling. The SLIMCAT columns correspond to vertical profiles above Eureka, while the Bruker FTIR measurements have ground footprints of hundreds of km in the early spring due to large SZA. The the largest deviations between SLIMCAT and the Bruker FTIR occur for SZA>87°, which is expected given that the Bruker FTIR line-of-sight reaches 16 km altitude (the approximate lower boundary of the peak ozone concentrations) on average 150 km away from Eureka for such large SZA. Excluding measurements with SZA>87° reduces the late February differences, and so the mean relative differences improve to -8.2±3.9%. Comparisons to the GBS and SAOZ datasets do not show significant seasonal differences, likely due to longer stratospheric pathlengths for the ZSL-DOAS measurements. Brewer measurements inside the vortex are mostly restricted to 2020, and so the mean differences should be interpreted with caution.

SLIMCAT active ozone outside the vortex generally overestimates the measurements. A consistent offset of 10-11% (40-45 DU) is apparent between inside and outside comparisons across all instruments. Comparisons to Microwave Limb Sounder (MLS, on board NASA's Aura satellite) data indicate that this difference is already present at the start of the winter. The difference is likely related to model dynamics, and not to the springtime ozone depletion chemistry.

Figures S4 and S5 show absolute and relative differences between SLIMCAT HCl, $ClONO_2$, and HNO_3 and Bruker FTIR measurements, as a function of time of year and vortex location. HCl comparisons show similarly good agreement inside and outside the vortex, but with a significant early-season slope in the in-vortex differences. This is largely the result of the sampling issues discussed above. Excluding Bruker FTIR measurements with SZA>87°, the comparisons inside the vortex improve to -1.8% (from -4.0% when including all measurements). Measurement SZA does not have a significant impact on comparisons outside the vortex, likely because of the more uniform HCl background (Fig. 2c in the main text). $ClONO_2$ comparisons indicate very good agreement inside the vortex (0.6%), while SLIMCAT significantly overestimates $ClONO_2$ outside the vortex. It should be noted that $ClONO_2$ columns outside the vortex are generally small (Fig. 2d in the main text), and so the relative differences are large. $ClONO_2$ differences appear related to lower stratospheric temperature, with increasing differences for increasing temperatures (slope of $\sim$1.1% K^{-1} for differences inside the vortex). SLIMCAT generally underestimates HNO_3 both inside and outside the vortex (by 18.2% and 11.0%, respectively). This is expected given the simple equilibrium denitrification scheme included in the model. The scatter in the differences increases as temperatures approach T_{NAT}. The large spike in the comparisons around 18 March corresponds to the record low HNO_3 columns measured by the Bruker FTIR in 2020 (Fig. 2e in the main text).

The SLIMCAT comparisons presented here are in broad agreement with Lindenmaier et al. (2012), who compared SLIMCAT data to Bruker FTIR measurements for 2011. The differences between measurements inside and outside the vortex are consistent, and the underestimation of HNO_3 is present in both studies. Direct comparisons are difficult, however, since the model simulations in Lindenmaier et al. (2012) used an older version of SLIMCAT, with lower resolution and different reanalysis input.

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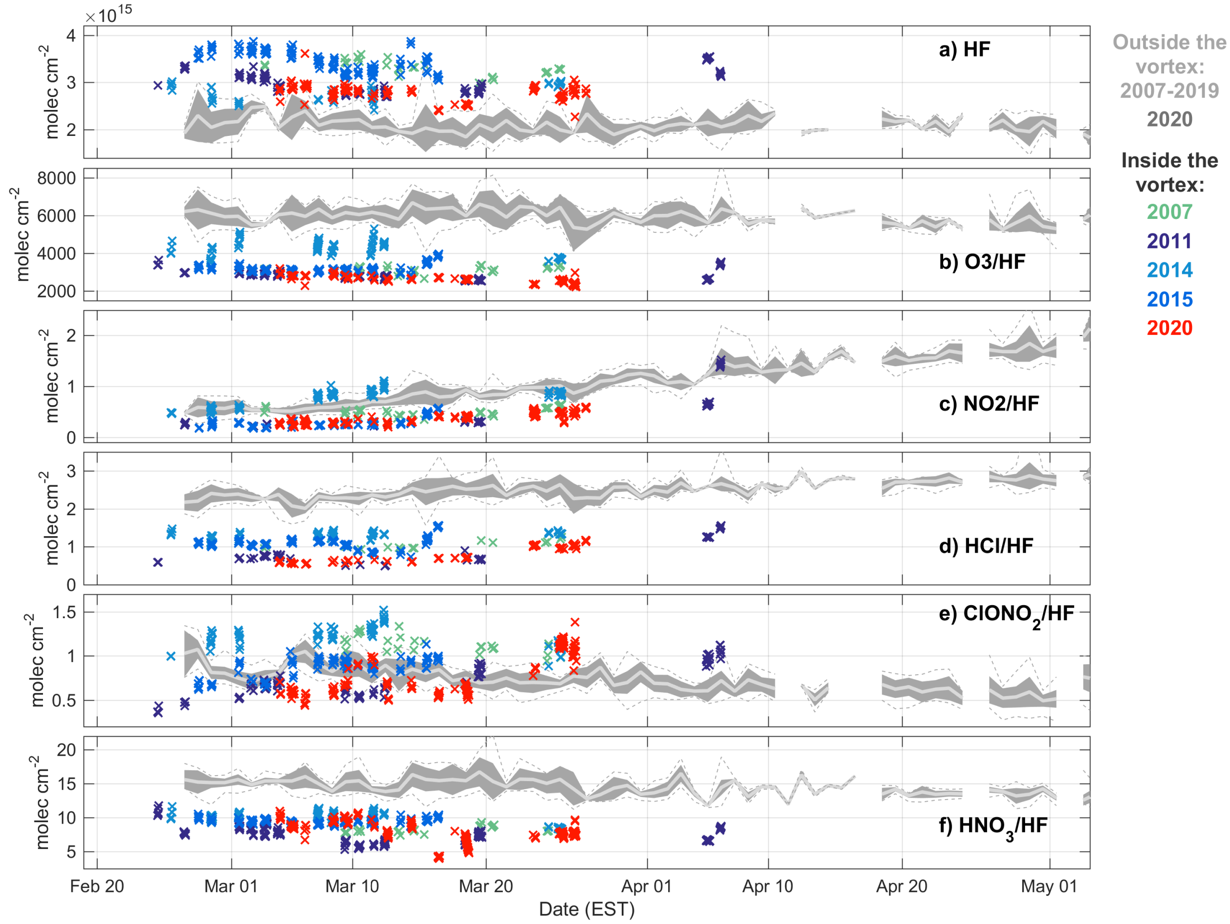

Figure S1. **a)** Measurements of HF columns from the Bruker FTIR. Measurements of **b)** ozone, **c)** NO_2, **d)** HCl, **e)** $ClONO_2$, and **f)** HNO_3, normalized by the HF columns. NO_2 columns were scaled to local noon prior to normalization. Measurements outside the vortex (up to 2019) are represented by the gray shaded area (daily mean and standard deviation) and the gray dashed lines (daily minima and maxima). The colored datapoints represent measurements inside the vortex, in years when the vortex was located above Eureka for a substantial part of the measurement period.

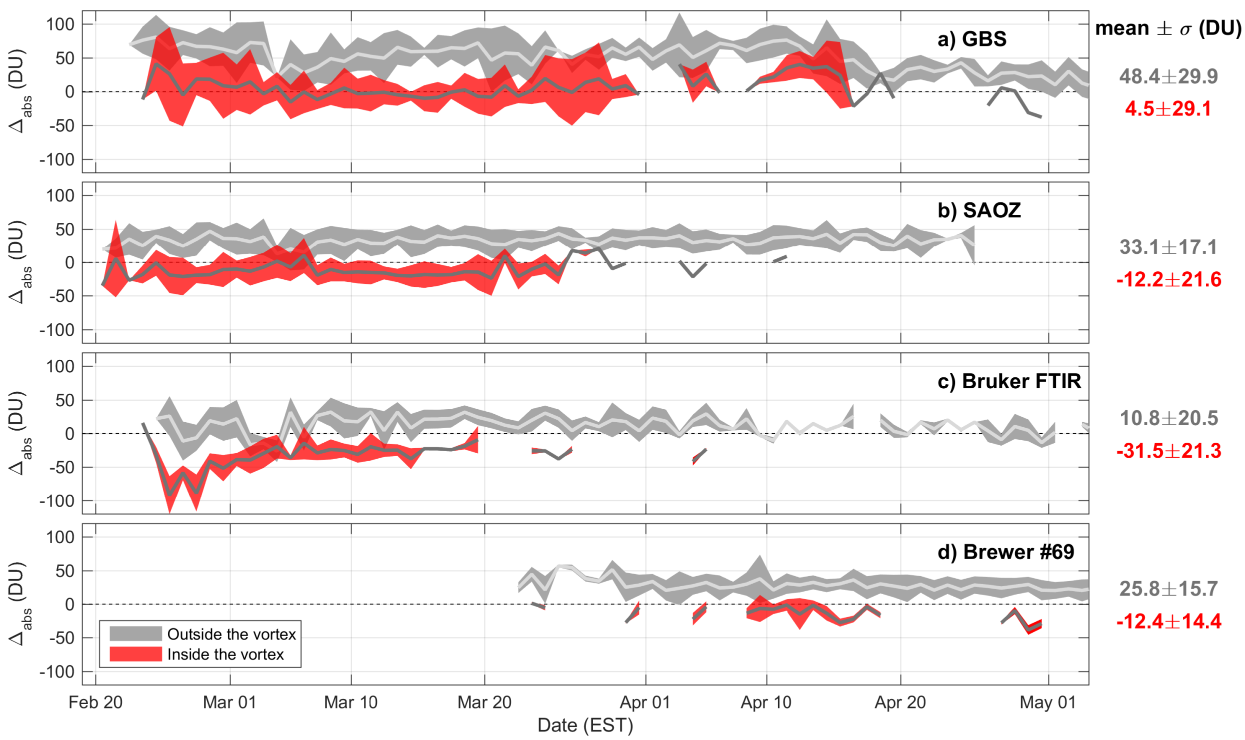

Figure S2. Absolute differences between SLIMCAT active ozone and measurements of ozone from **a)** GBS, **b)** SAOZ, **c)** Bruker FTIR and **d)** Brewer instruments, for all years with available data. The solid lines and shaded areas show daily mean and corresponding standard deviation from all available years. Measurements inside the vortex are shown by the red shading, while out-of-vortex measurements are shown in gray. Standard deviations are only plotted if more than two measurements are available for the given day. The overall mean absolute differences (and corresponding standard deviations) are indicated on the right for measurements inside and outside the vortex.

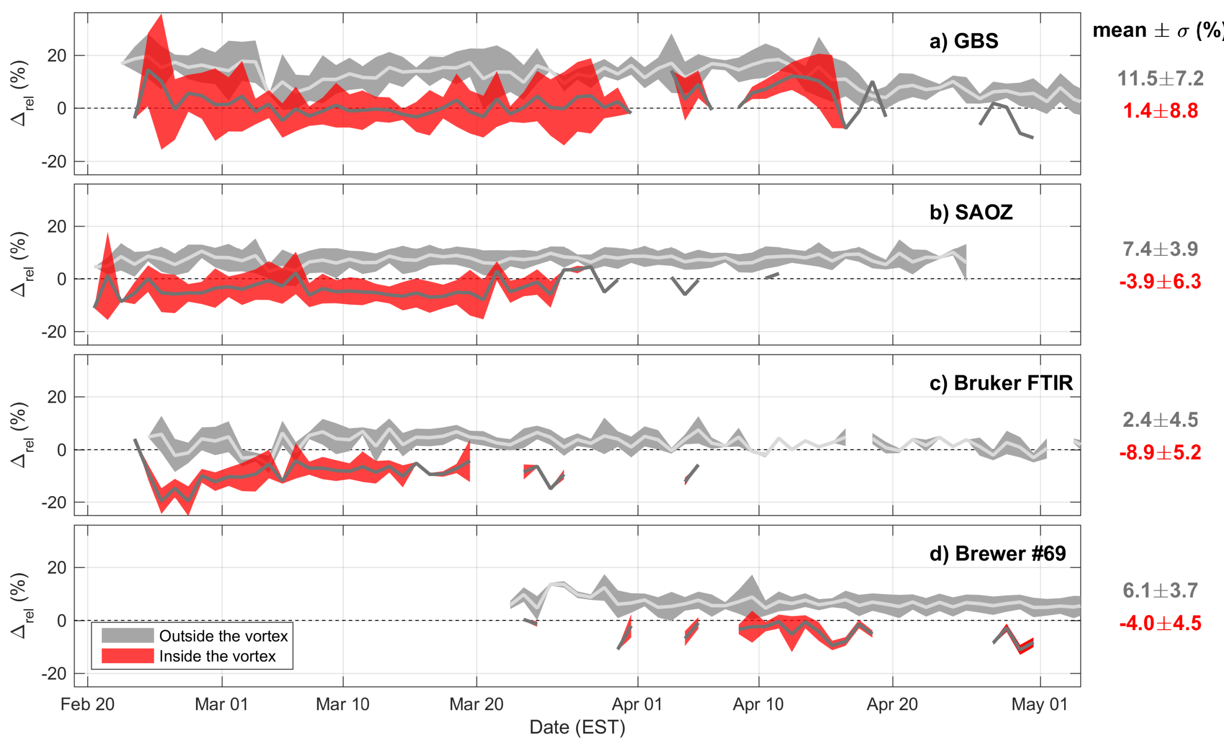

Figure S3. As for Figure S2, with relative ozone differences.

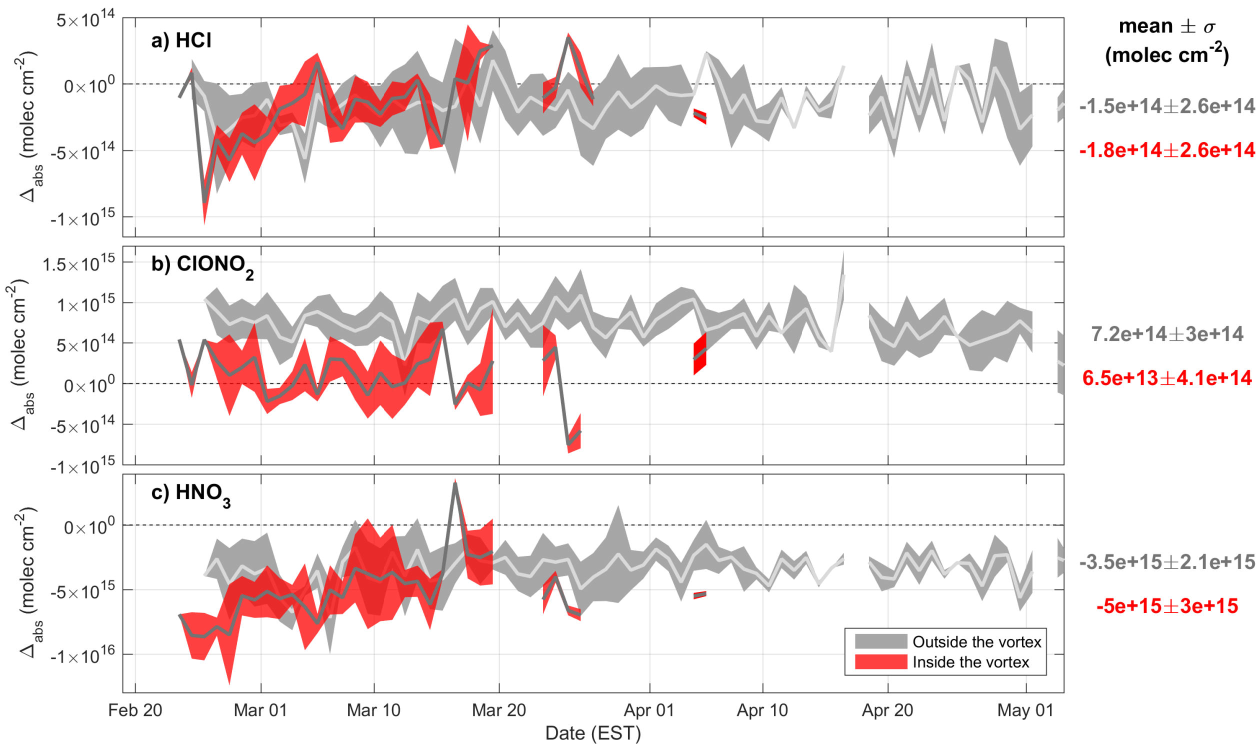

Figure S4. As for Figure S2, with SLIMCAT vs Bruker FTIR absolute differences for **a)** HCl, **b)** $ClONO_2$, and **c)** HNO_3. Note that the y-axis limits are different for each trace gas.

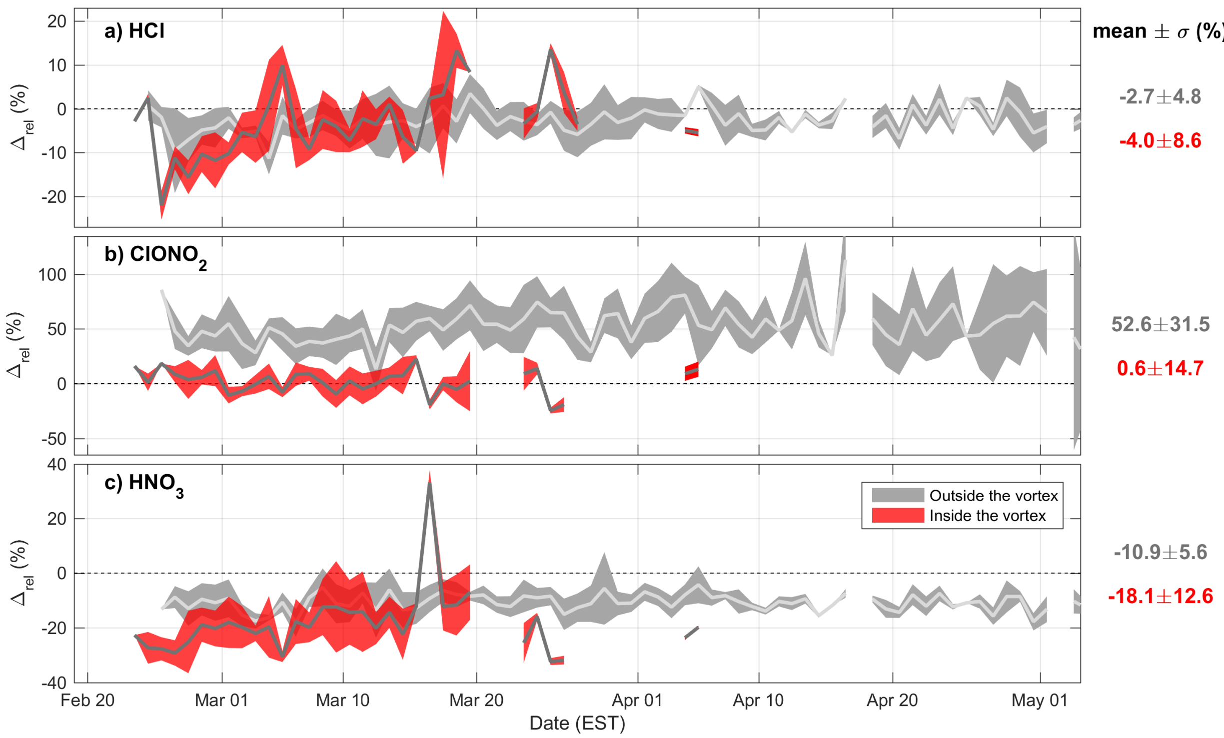

Figure S5. As for Figure S2, with SLIMCAT vs Bruker FTIR relative differences for **a)** HCl, **b)** $ClONO_2$, and **c)** HNO_3. Note that the y-axis limits are different for each trace gas.