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## Severe 2011 ozone depletion assessed with 11 years of ozone, NO<sub>2</sub>, and OCIO measurements at 80°N

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[1] Unusually cold conditions in Arctic winter 2010/11 led to large stratospheric ozone loss. We investigate this with UV-visible measurements made at Eureka, Canada (80.05°N, 86.42°W) from 1999–2011. For 8–22 March 2011, OCIO was enhanced, indicating chlorine activation above Eureka. Ozone columns were lower than in any other year in the record, reaching minima of 237 DU and 247 DU in two datasets. The average NO<sub>2</sub> column inside the vortex, measured at visible and UV wavelengths, was  $46 \pm 30\%$  and  $45 \pm 27\%$  lower in 2011 than the average NO<sub>2</sub> column from previous years. Ozone column loss was estimated from two ozone datasets, using a modeled passive ozone tracer. For 12–20 March 2011, the average ozone loss was 27% and 29% (99 DU and 108 DU). The largest percent ozone loss in the 11-year record of 47% (250 DU and 251 DU) was observed on 5 April 2011. **Citation:** Adams, C., et al. (2012), Severe 2011 ozone depletion assessed with 11 years of ozone, NO<sub>2</sub>, and OCIO measurements at 80°N, *Geophys. Res. Lett.*, 39, L05806, doi:10.1029/2011GL050478.

### 1. Introduction

[2] In spring 2011, chemical ozone loss in the Arctic was comparable to that observed over Antarctica for the first time on record [Manney et al., 2011]. This resulted from an unusually prolonged period with a strong, cold polar vortex. Due to these persistent low temperatures, polar stratospheric

clouds (PSCs) were observed until mid-March and activated chlorine was observed until late March. This resulted in a record ozone loss [Balis et al., 2011; Manney et al., 2011].

[3] The polar vortex was above the Polar Environment Atmospheric Research Laboratory (PEARL), located at Eureka, Canada (80.05°N, 86.42°W) for a large part of spring 2011. A suite of instruments, operated by the Canadian Network for the Detection of Atmospheric Change (CANDAC), take continuous measurements at PEARL. We present results from four differential optical absorption spectroscopy (DOAS) instruments. DOAS instruments can measure under both clear and cloudy conditions and therefore present a more continuous timeseries of ozone and NO<sub>2</sub> than solar tracking Fourier Transform Infrared (FTIR) spectrometers. Furthermore, DOAS instruments can also measure OCIO, which is a good qualitative indicator of chlorine activation [Sessler et al., 1995]. However, quantification of ClO from OCIO measurements is difficult due to uncertainties in model predictions, particularly under strong chlorine activation [e.g., Oetjen et al., 2011]. Ozone, NO<sub>2</sub>, and OCIO measurements can be combined with stratospheric parameters in order to identify ozone depletion, chlorine activation, and denitrification within the polar vortex [e.g., Tornkvist et al., 2002; Tétard et al., 2009].

### 2. Datasets

[4] Measurements included in this study were taken by four ground-based DOAS instruments: the PEARL and University of Toronto ground-based spectrometers (GBSS) [Fraser et al., 2009] and two System D'Analyse par Observations Zenithales (SAOZ) instruments [Pommereau and Goutail, 1988]. These instruments are part of the Network for the Detection of Atmospheric Composition Change (NDACC) and their Eureka ozone and NO<sub>2</sub> datasets are described in detail by Adams et al. [2012]. Measurements from the two GBS (two SAOZ) instruments are nearly identical and therefore were combined to create a single GBS (SAOZ) dataset.

[5] The GBSSs are UV-visible Triax-180 triple-grating spectrometers, built by Instruments S.A. / Jobin Yvon Horiba, with cooled charge-coupled device detectors and a 2° field-of-view. The resolution varies from 0.2–2.5 nm and the wavelength range varies from 320–600 nm depending on the selected grating and target wavelength. GBS ozone and NO<sub>2</sub> columns were retrieved above Eureka in the spring for 1999–2011, except for 2001 and 2002. The SAOZ instruments are grating spectrometers, which measure in the 270–620 nm range with a 1.0-nm resolution and a 10° field-

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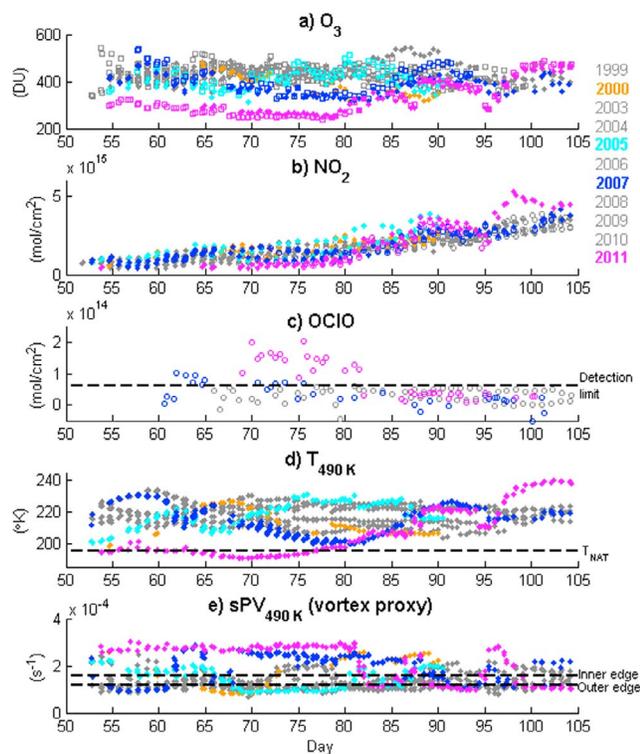
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**Figure 1.** Timeseries of measurements and dynamical parameters along the DOAS line-of-sight for 1999–2011 versus day of year. Year 2000 is shown in orange, 2005 in cyan, 2007 in blue, 2011 in magenta, and all other years are shown in grey. (a) Ozone total columns measured by the GBS (closed diamonds) and SAOZ (open squares). (b)  $\text{NO}_2$  partial columns (17 km to top of atmosphere) measured by GBS-vis (closed diamonds) and GBS-UV (open circles). (c) OCIO DSCDs measured by the GBS. (d)  $T_{490\text{K}}$  and (e)  $\text{sPV}_{490\text{K}}$ .

of-view and record spectra on uncooled 1024-pixel linear diode array detectors. SAOZ instruments took spring-time measurements at Eureka for 2005–2011.

[6] The DOAS (GBS and SAOZ) ozone measurements were analyzed in the 450–550 nm range, using the NDACC guidelines [Hendrick *et al.*, 2011].  $\text{NO}_2$  partial columns were retrieved for the GBS instruments in two different wavelength regions: 425–450 nm (GBS-vis) and 350–380 nm (GBS-UV), depending on the selected measurement grating. The GBS  $\text{NO}_2$  partial columns were calculated for 17 km to the top of the atmosphere for the validation of satellite partial column measurements, using AMFs that were set to zero below 17 km [Adams *et al.*, 2012]. SAOZ  $\text{NO}_2$  total columns were retrieved in the 410–510 nm range. The SAOZ  $\text{NO}_2$  total columns are qualitatively consistent with the GBS partial columns, but are not presented here because they cover a different altitude range. The SAOZ and GBS ozone and  $\text{NO}_2$  columns have been shown to agree well with other ground-based and satellite measurements [Fraser *et al.*, 2008, 2009; Adams *et al.*, 2012]. OCIO differential slant column densities (DSCDs) at solar zenith angle  $90^\circ$  were also retrieved from spring 2007, 2008, and 2011 GBS

spectra in the 350–380 nm range. The OCIO retrievals are described in the auxiliary material.<sup>1</sup>

[7] Derived meteorological products [Manney *et al.*, 2007] were calculated along the lines-of-sight of the DOAS instruments [Adams *et al.*, 2012] for 1999–2003 using the Met Office analysis and 2004–2011 using the GEOS-5.1.0/GEOS-5.2.0 analysis. Stratospheric temperatures and scaled potential vorticity (sPV) were interpolated to the 490-K potential temperature level ( $\sim$  ozone concentration maximum,  $\sim$ 19 km) and are referred to here as  $T_{490\text{K}}$  and  $\text{sPV}_{490\text{K}}$ . The inner and outer vortex edges are identified by  $\text{sPV}_{490\text{K}}$  values of  $1.6 \times 10^{-4} \text{ s}^{-1}$  and  $1.2 \times 10^{-4} \text{ s}^{-1}$ , respectively [Manney *et al.*, 2007].

### 3. Timeseries of Ozone, $\text{NO}_2$ , and OCIO

[8] The 1999–2011 timeseries of ozone,  $\text{NO}_2$ , OCIO,  $T_{490\text{K}}$ , and  $\text{sPV}_{490\text{K}}$  are shown in Figure 1. In 2000, 2005, 2007 and 2011, low ozone columns were measured above Eureka when the polar vortex was overhead. These years are shown in color, while the other measurement years are shown in grey. Low ozone coincides with low  $\text{NO}_2$ , low  $T_{490\text{K}}$ , and time periods when the instruments are sampling inside the polar vortex.

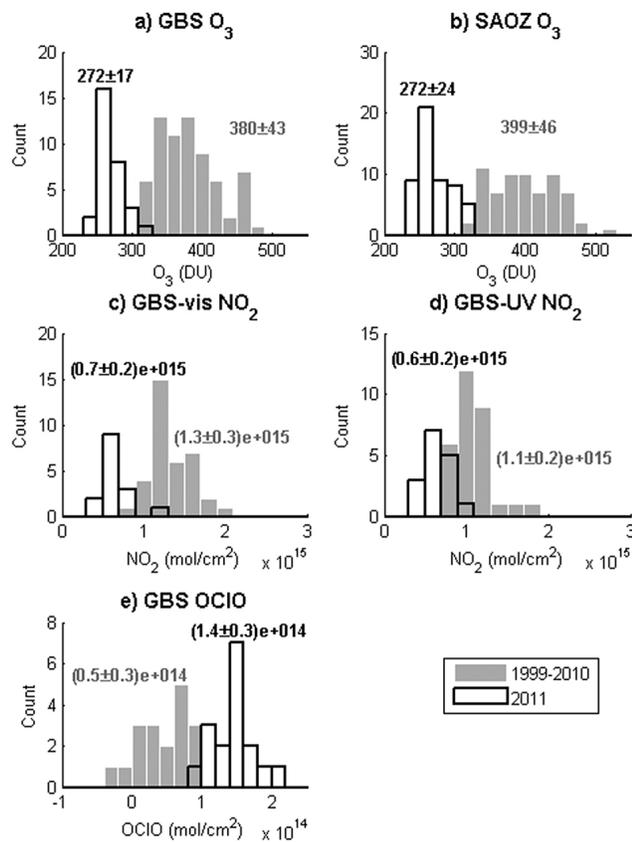
[9] For 23 February to 21 March 2011 (days 54–80), the DOAS instruments sample lower stratospheric air inside the polar vortex. OCIO DSCDs of  $0.8\text{--}2.0 \times 10^{14} \text{ mol/cm}^2$  are within the range of previous elevated OCIO measurements [e.g., Tornkvist *et al.*, 2002], suggesting chlorine activation. All elevated OCIO DSCDs, from 8–22 March 2011 (days 67–81) and 2–5 March 2007 (days 61–64), are measured inside the polar vortex when the high-latitude minimum temperature (calculated by Manney *et al.* [2011], not shown here) is below the threshold for PSC formation ( $T_{\text{NAT}}$ ). High OCIO measurements do not always correspond with local  $T_{490\text{K}} < T_{\text{NAT}}$  (Figure 1d), because the time-scale for vortex mixing ( $\sim$ 5–7 days) is smaller than the time-scale for chlorine deactivation ( $\sim$ weeks). During the period of elevated OCIO in 2011, ozone,  $\text{NO}_2$ , and  $T_{490\text{K}}$  all reach minima in the 11-year record, with ozone values of 247 DU (237 DU) measured by the GBS (SAOZ) on 18 March (day 77).

[10] After 22 March 2011 (day 81), the instruments primarily sample the lower stratosphere outside the polar vortex. During this period, ozone and  $\text{NO}_2$  increase to levels that are normal in the context of the 11-year data record. On 5 April (day 87) and 28 March (day 95), ozone and  $\text{NO}_2$  columns and  $T_{490\text{K}}$  decrease sharply, as the instruments sample air masses inside the vortex. After 5 April (day 95),  $T_{490\text{K}}$  and  $\text{NO}_2$  increase to maxima in the 11-year dataset. This increase is the subject of a companion study.

### 4. Dynamical and Chemical Contributions to Low Ozone

[11] As is evident in the DOAS timeseries (Figure 1), 2011 is extremely different from previous years. Ozone,  $\text{NO}_2$ , and OCIO measurements taken inside the polar vortex ( $\text{sPV}_{490\text{K}} > 1.6 \times 10^{-4} \text{ s}^{-1}$ ) for days 55–80 (24 February to 19/20 March) were selected to investigate this further. The time-period was limited in order to reduce the impact of

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2011GL050478.



**Figure 2.** Histograms of (a) GBS ozone, (b) SAOZ ozone, (c) GBS-vis  $\text{NO}_2$ , (d) GBS-UV  $\text{NO}_2$ , and (e) GBS OCIO. Measurements were taken inside the vortex for days 55–80 (24 February to 19/20 March), with 1999–2010 in grey and 2011 transparent with thick black lines.  $N \pm M$  denotes the average ( $N$ ) and  $1\sigma$  standard deviation ( $M$ ) in the measurements.

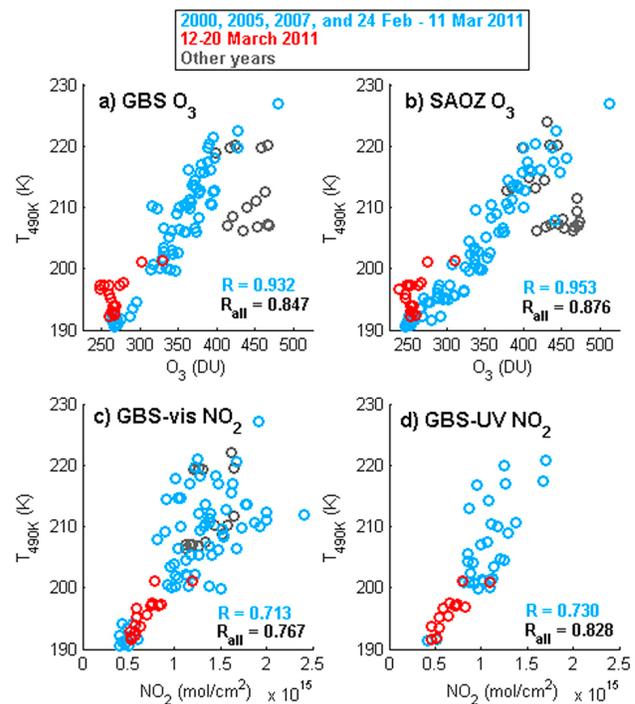
seasonal variation on the results.  $\text{NO}_2$  measurements were scaled to local solar noon using a photochemical model [McLinden *et al.*, 2000] initialized with temperature and ozone from Eureka ozonesonde profiles from the nearest available date.

[12] Figure 2 shows histograms of ozone,  $\text{NO}_2$ , and OCIO for 1999–2010 (grey) and 2011 (transparent with thick black outline). In 2011, the mean vortex ozone column measured by the GBS (SAOZ) is  $28 \pm 13\%$  ( $32 \pm 14\%$ ) lower than the mean column from other years, where the error denotes the  $1\sigma$  statistical uncertainty. Similarly, GBS-vis (GBS-UV)  $\text{NO}_2$  is  $46 \pm 30\%$  ( $45 \pm 27\%$ ) lower and GBS OCIO is three times higher in 2011 than in previous years.

[13] The unusual 2011 ozone and  $\text{NO}_2$  columns are a result of both chemistry and transport, which contribute approximately equally to year-to-year total ozone column variability in the Arctic [Tegtmeier *et al.*, 2008]. Figure 3 shows the correlation between vortex ozone/ $\text{NO}_2$  measurements and the local  $T_{490\text{K}}$ . Correlation between ozone and local lower stratospheric temperature has been observed in previous studies and points to replenishing of ozone through vertical descent, horizontal mixing across the vortex edge, and adiabatic compression of the column, which all increase with higher stratospheric temperatures (e.g., supplementary material of Manney *et al.* [2011], and references therein). In

the present study, the strongest correlation between ozone and  $T_{490\text{K}}$  was calculated when data were excluded from years with few vortex measurements above Eureka (grey) and 12–20 March 2011 (days 71–79, red). The outliers for years with few vortex measurements may result from errors in matching  $T_{490\text{K}}$  and  $\text{sPV}_{490\text{K}}$  to measurements both spatially and temporally when the vortex edge is near Eureka. For 12–20 March 2011, the ozone columns remain low, despite the rise in the local  $T_{490\text{K}}$ . This deviation from the correlation between  $T_{490\text{K}}$  and ozone suggests chemical depletion (supplementary material of Manney *et al.* [2011]).  $\text{NO}_2$  is also correlated with local lower stratospheric temperature, as has been observed in previous studies [e.g., Pommereau and Goutail, 1988; Dirksen *et al.*, 2011]. The correlation for  $\text{NO}_2$  is weaker than for ozone, likely due to the seasonal increase in  $\text{NO}_2$  as it is released from night-time reservoirs.

[14] Investigation of complementary datasets provides further evidence of chemical ozone depletion and denitrification above Eureka in 2011. For 9–18 March (days 68–77), low  $\text{HNO}_3$  and  $\text{ClONO}_2$  columns over Eureka were measured by the CANDAC Bruker FTIR [Lindenmaier *et al.*, 2012]. This suggests that the extremely low  $\text{NO}_2$  columns measured during the same period are not caused by conversion to  $\text{HNO}_3$  or  $\text{ClONO}_2$ . During this period of low  $\text{ClONO}_2$ ,  $\text{HNO}_3$ , and  $\text{NO}_2$ , OCIO DSCDs are elevated, reinforcing that chlorine remains activated. Furthermore, PSCs were measured above Eureka with the CANDAC



**Figure 3.** Correlation between  $T_{490\text{K}}$  and (a) GBS ozone, (b) SAOZ ozone, (c) GBS-vis  $\text{NO}_2$ , and (d) GBS-UV  $\text{NO}_2$ . Measurements were taken inside the vortex for days 55–80 (24 February to 19/20 March). Data are shown for 2000, 2005, 2007, and 24 February to 11 March 2011 (blue); 12–20 March 2011 (red); and other years (grey).  $R$  and  $R_{\text{all}}$  are correlation coefficients for data indicated by blue only and for all data in the figure, respectively.

Rayleigh-Mie-Raman Lidar between 8–18 March (days 67–77) [Lindenmaier et al., 2012]. These measurements agree with photochemical model runs in supplementary material of Manney et al. [2011], which indicate that prolonged denitrification by sedimentation of PSCs delayed chlorine deactivation, leading to the record ozone loss.

[15] In order to isolate chemical ozone depletion from dynamical features, the passive subtraction method [e.g., Manney et al., 1995; World Meteorological Organization, 2003; Feng et al., 2007] was employed using SLIMCAT [Chipperfield, 2006], a three-dimensional off-line chemical transport model. These ozone loss estimates are described in detail in the auxiliary material. The average ozone loss for 12–20 March 2011 was 27% (29%) or 99 DU (108 DU), as estimated from GBS (SAOZ) data. The maximum percent ozone loss in the 11-year data record was calculated from GBS (SAOZ) data on 5 April 2011 at 47% (47%) or 250 DU (251 DU). A similar maximum ozone loss of 266 DU was observed by Lindenmaier et al. [2012] on 5 April 2011 above Eureka.

## 5. Conclusion

[16] Unprecedentedly low ozone and NO<sub>2</sub> columns were measured in 2011 and correspond to elevated OClO, suggesting chlorine activation and ozone depletion. Vortex ozone and NO<sub>2</sub> total columns from 1999–2011 are correlated with the lower stratospheric temperature above Eureka, indicating that transport also contributes to the low ozone and NO<sub>2</sub> measurements. Using the SLIMCAT passive tracer model, a maximum percent ozone loss of 47% was observed on 5 April 2011.

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