# The contributions of chemistry and transport to low Arctic ozone in March 2011 derived from Aura MLS Observations

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### Abstract

Stratospheric and total columns of Arctic O<sub>3</sub> (63-90°N) in late March 2011 averaged 320 and 349 DU, respectively. These values are 74 DU lower than averages for the previous 6 years. We use Aura MLS O<sub>3</sub> observations to quantify the roles of chemistry and transport and find there are two major reasons for low O<sub>3</sub> in March 2011: heterogeneous chemical loss and a late final warming that delayed the resupply of O<sub>3</sub> until April. Daily vortex-averaged partial columns in the lowermost stratosphere (p > 133 hPa) and middle stratosphere (p < 29 hPa) are unaffected by local heterogeneous chemistry and show a near total lack of transport into the vortex between late January and late March, contributing to the observed low column. The lower stratospheric (LS) column (133-29 hPa) is affected by both heterogeneous chemistry and transport. Low interannual variability of Aura MLS O<sub>3</sub> columns and temperature inside the Arctic vortex (2004-2011) shows that the transport contribution to vortex O<sub>3</sub> in fall and early winter is nearly the same each year. The descent of MLS N<sub>2</sub>O vortex profiles in 2011 provides an estimate of O<sub>3</sub> transported into the LS column during late winter. By quantifying the role of transport we determine that PSC-driven chemical loss causes 80 (±10) DU of vortex-averaged O<sub>3</sub> loss by late March 2011. Without heterogeneous chemical loss, March 2011 vortex O<sub>3</sub> would have been ~40 DU lower than normal due to the late final warming and resupply of O<sub>3</sub> which did not occur until April.

## 1. Introduction

Measurements made by the Ozone Measuring Instrument (OMI) and the Microwave Limb Sounder (MLS) on the NASA Aura satellite, averaged over March 20-26, 2011, showed mean Arctic O<sub>3</sub> (63-90°N) of 349 DU (total column) and 320 DU (stratospheric column), respectively. Figure 1 shows the OMI and MLS column measurements from March 24, 2011; both instruments show regions of less than 250 DU. The MLS Arctic mean O<sub>3</sub> displayed in Figure 1c shows considerable interannual (IA) variability in late winter, however, the low columns observed in 2011 (red) are in sharp contrast with the previous 6 years. MLS Arctic mean stratospheric column and OMI total column O<sub>3</sub> from 2005-2010 averaged 74 DU

higher - 394 DU and 423 DU, respectively - during this same one week period in late March. Why were late March 2011 Arctic  $O_3$  column means nearly 20% lower than recent years?

The 2011 Arctic vortex was unusual in its longevity and in the strength of its mixing barrier [Manney et al., 2011]. The strong, long-lasting vortex allowed lower stratospheric temperatures to be maintained below the threshold of chlorine activation from December to late March. Meteorology allowing such low temperatures to persist for more than 3 months is typical of the Antarctic and has not previously been observed in the Arctic. Although recent cold Arctic springs, notably 1997, 2000, 2003, and 2005, had strong March vortices and sufficiently low temperatures that produced daily March minimum O<sub>3</sub> columns as low as ~320 DU (averaged poleward of 63° equivalent latitude), they did not have prolonged periods of PSC formation (WMO, 2011). The lengthy period of low temperatures in 2011 led to a much greater degree of denitrification and subsequent ozone loss than has been previously observed in the Arctic [Manney et al., 2011; Sinnhuber et al., 2011].

Winters with above average stratospheric wave activity have a warm, disturbed vortex while winters with weak wave-driving have a cold and longer-lasting vortex, with well-known impacts on Arctic March temperatures [Newman et al., 2001] and column  $O_3$  [Chipperfield and Jones, 1999; Tegtmeier et al., 2008]. Greater poleward and downward transport of ozone in warm winters leads to higher levels of March ozone. In cold winters, not only is the vertical transport (descent) of  $O_3$  weaker, but the increased strength of the vortex creates a barrier to meridional transport that keeps high ozone outside the vortex. The converse is true for warm winters. The relationship between dynamical variability and  $O_3$  was demonstrated by Randel et al. (2002), who showed the link between changes in stratospheric wave driving (i.e., EP flux) and ozone transport to high latitudes in spring, concluding that hemispheric ozone trends were caused in part by the wave driving trends from 1979-2000. Chipperfield and Jones (1999) also observed this relationship and concluded that dynamical variability in the 1990's was a much greater contributor to IA variability in March Arctic  $O_3$  than chemical loss. Mueller et al. (2008) showed that some of the effects of dynamical variability on column  $O_3$  could be reduced by calculating daily column  $O_3$  means poleward of an equivalent latitude rather than a geometric latitude, thus increasing the correlation between March Arctic  $O_3$  and ozone loss.

Cold winters can have lower March ozone not only due to increased frequency of PSCs and weaker wave-driven transport but because transport or resupply of higher  $O_3$  to the Arctic by the final warming is delayed. Herein lies the difficulty in quantifying chemical  $O_3$  loss: there is no 'baseline' or typical Arctic March column  $O_3$ . We cannot quantify Arctic  $O_3$  loss caused by PSCs without quantifying  $O_3$  transport

into the vortex. This can be an especially large problem in years such as 1997 and 2011, which were cold enough to have significant chemical loss but did not have a final warming until April. The final warming can supply  $\sim$ 50 DU O<sub>3</sub> to the Arctic.

In this paper we use Aura MLS  $O_3$  and  $N_2O$  observations to quantify transport into the Arctic vortex in 2011 and deduce the heterogeneous chemical loss. We examine Arctic  $O_3$  behavior between October and April for the years 2004-2011 to characterize column  $O_3$  variability. Analysis of vortex partial column  $O_3$  above and below the levels of PSC occurrences demonstrates the effects of reduced transport on those parts of the vortex column. Analysis of MLS LS temperatures and column  $O_3$  shows there is remarkably little IA variability in transport inside the Arctic vortex before a warming occurs, allowing us to estimate the early winter LS column available for chemical loss in 2011. The descent of MLS  $N_2O$  vortex profiles provides a means to calculate the small effect of transport on LS vortex  $O_3$  in late winter. By the quantifying the transport contributions to vortex  $O_3$ , we determine the upper bound for chemical  $O_3$  loss averaged over the vortex. These results are discussed in the context of other estimates of chemical loss in 2011.

### 2. Observations

This study uses Aura MLS v3.3 level 3 gridded temperature,  $O_3$ , and  $N_2O$  on pressure surfaces between October 2004 and March 2011 (Livesey et al., 2011). Due to an instrument anomaly the MLS instrument did not make observations between March 27 and April 18, 2011. MLS v3.3  $O_3$  and temperature data are reported on a high vertical resolution grid with 12 pressures per decade;  $N_2O$  is reported on 6 pressure levels per decade. Livesey et al. (2011) report the  $2\sigma$  accuracy of MLS v3.3  $O_3$  columns as 4%. For MLS  $N_2O$  observations, the  $2\sigma$  accuracy in the lower stratosphere is reported as 14% (Lambert et al., 2007). Aura OMI total column  $O_3$  data are used to calculate 63-90°N averages. OMI is a UV/visible backscatter instrument that make measurements only in the sunlit globe, thus the Arctic is poorly sampled until mid-February. The OMI column  $O_3$  mean bias error is reported as less than 2% (Anton et al., 2009).

In this study we calculate MLS columns using data poleward of  $54^{\circ}$ N, from 268 to 0.46 hPa, and evaluate them inside and outside the vortex during a 7-year period. The lowest MLS  $O_3$  level used, 268 hPa, is generally near the tropopause inside the vortex but below the tropopause outside at high northern latitudes. Nash et al. (1996) showed that the location of the steepest potential vorticity gradient on a potential temperature surface can be used to define the vortex edge. We find that PV=4

(10<sup>-5</sup> K m<sup>2</sup> kg<sup>-1</sup> s<sup>-1</sup>) on the 500 K surface is consistently located in the region of steep gradients for all winters studied. We use daily potential vorticity fields from the GEOS-MERRA assimilation (Rienecker et al.., 2011) to identify whether an MLS observation is inside or outside the Arctic vortex. The location of the PV-based edge varies with height but only one edge definition can be used when evaluating column quantities. The PV=4 (500K) vortex edge definition used here is closely aligned with the location of large PV gradients in the lower stratosphere (400-450K) and in the middle stratosphere (600-700K). Ozone from 400-700K represents roughly three quarters of the stratospheric column.

3. Interannual variability of Arctic column Ozone

## 3.1 Variability Inside and Outside the Vortex

The polar vortex creates a strong barrier to horizontal mixing, thus polar transport processes and their variability affect vortex and nonvortex  $O_3$  differently. Figure 2 shows daily vortex- and nonvortex-averaged column  $O_3$  from October to March, calculated using the daily location of PV=4 on the 500K surface to define the vortex edge. The daily means are area-weighted and use all MLS  $O_3$  observations poleward of 54°N from 2004-2011. The 54°N lower latitude limit was chosen in order to include the vortex during wave events when it is pushed well off the pole. The Arctic mean column depends on the means of the vortex and nonvortex columns as well as the fractional area of each region. Figure 2c shows the time series of vortex area as a percentage of the Arctic (54°-90°N).

Figure 2 shows that column O<sub>3</sub> increases during fall and winter in all years both inside and outside the vortex; this is a well-understood consequence of poleward transport of O<sub>3</sub> from its low latitude source region by the Brewer-Dobson circulation (e.g., Newman et al., 2001; Randel et al., 2002). Arctic columns outside the vortex have increased IA variability from mid-December through March, while vortex O<sub>3</sub> variability is quite low until late January; after this point a major warming can cause a large, rapid increase in vortex O<sub>3</sub> and even the disappearance (breakdown) of the vortex (e.g., 2006, 2009, and 2010). Figure 2 shows that large contributions to Arctic mean column variability, as seen in Figure 1, include vortex area and column amounts outside the vortex beginning in early winter, and vortex column amounts after mid-January.

The formation of the vortex in fall is governed by radiative processes at a time when seasonal wave activity is weak. IA variability of vortex  $O_3$  in fall is therefore low because there is little IA variability in the radiative and photochemical processes controlling it (Kawa et al., 2002). Vortex temperatures reflect the low IA variability in early season transport. The solid color lines in Figure 3a shows the daily means

of vortex temperatures at 68 hPa for 7 recent Arctic winters. The dashed (dotted) lines show the 10<sup>th</sup> and 90<sup>th</sup> (25<sup>th</sup> and 75<sup>th</sup>) percentiles of all daily observed vortex temperatures for 7 years. Figure 3b shows the daily means and distributions for all Arctic 68 hPa temperatures, 54°-90°N. While temperatures over the entire Arctic have large IA variability – the 10<sup>th</sup> and 90<sup>th</sup> percentiles show a range over more than 30K in mid-January - the same period inside the vortex shows a range of ~15K and the daily means (colored lines) are tightly clustered. In the absence of a large wave event such as a major warming, vortex temperatures in fall and early winter are remarkably similar each year, producing descent and downward transport of the O<sub>3</sub> column that is radiatively driven (Kawa et al., 2002). In addition, Kawa et al. (2002) report that vortex O<sub>3</sub> profiles in November have low IA variability, thus the combination of low IA variability in both vortex O<sub>3</sub> profiles and descent rates leads to low IA variability in vortex column O<sub>3</sub>. Starting in late January, wave driving increases and so do the ranges of vortex temperature, mean column O<sub>3</sub>, and area (Figs. 3a, 2a and 2c). The observed MLS vortex temperatures show low IA variability from fall through early winter at all levels from 146-21 hPa.

# 3.2 Partial Stratospheric Columns Inside the Vortex

Arctic vortex column O<sub>3</sub> after mid-January shows large variability due to differences in transport (i.e., warmings) and the degree of PSC-driven O₃ loss. To separate the effects of chemistry and transport we examine the time series of partial columns. PSC-forming temperatures in the Arctic vortex typically occur on potential temperatures surfaces ranging from 370-550K (Rex et al., 2006; Tegtmeier et al., 2008), approximately 120-30 hPa. Temperature conditions for PSC formation occur infrequently in the lowermost stratosphere (LMS) (p > 120 hPa) or the middle/upper stratosphere (MS) (p < 30 hPa). Analysis of partial ozone columns in the LMS and MS allows us to assess how transport affects much of the stratospheric column during fall and winter without the complication of heterogeneous chemistry. We compute partial columns based on the pressure difference between the top and bottom edges of a box where the MLS-reported pressure level is the midpoint. The top and bottom edge pressures are calculated as the midpoint in log-pressure between each level and the one above (below) it. The lower stratospheric (LS) column where PSCs may form spans 133-29 hPa, which includes all midpoint pressure levels 121-32 hPa. It is approximately coincident with the column from 370-550 K, which encompasses the range of potential temperatures used in other studies of Arctic O<sub>3</sub> depletion (e.g., Rex et al., 2006; Tegtmeier et al., 2008, Manney et al., 2011). Heterogeneous chemical losses are likely to take place only in this vortex partial column.

Figure 4 shows the time series for MLS LMS (287-133 hPa) and MS (29-0.4 hPa) column  $O_3$  inside the vortex for 7 Arctic winters. Fall and early winter show nearly the same, gradually increasing column amounts in all years, consistent with low total stratospheric column variability shown in Figure 2a and the radiative control discussed in Section 3.1. After mid-January, the LMS and MS columns, which have no chemical loss, show increased variability due to IA transport differences. From 2005 to 2010, the LMS and MS  $O_3$  columns also increase after mid-January. Vortex means increase substantially in 2006, 2009, and 2010 due to major midwinter warmings. In each of these years the vortex is destroyed but reforms before the end of March.

The LMS and MS O<sub>3</sub> columns show little to no increase during February and March 2011. The MS partial column observed in late March is essentially unchanged from late January. The LMS column shows a small increase that is less than the other years. The lack of increase is consistent with LMS and MS temperatures which hold steady during this period, indicating little cooling or diabatic descent. Potential vorticity gradients show that the stratospheric vortex remains strong down to ~350K, prohibiting horizontal transport of high (midlatitude) O<sub>3</sub> across the vortex edge. The calculated LMS vortex column includes midpoint pressure level data at 261 hPa which may at time be in the troposphere. While there will be no transport barrier in this case, the contribution of this layer's tropospheric O<sub>3</sub> to the LMS partial column is a few DU. The average late winter LMS column for 2005-2010 is 9 DU higher than 2011, and the 6-year average of the MS column is 15 DU higher. The sum of these two partial columns is shown in Figure 4c and highlights just how different transport was in 2011. Unlike the previous years, LMS and MS columns in 2011 indicate that transport was insignificant for almost half the vortex column in late winter.

By first separating Arctic  $O_3$  measurements into columns inside and outside the vortex, and then into partial columns in the vortex above and below the levels of PSC-formation, we have isolated and quantified the effects of transport on roughly half of the Arctic column  $O_3$ . We next examine the LS  $O_3$  column inside the vortex and separate the effects of transport and chemistry for the winter of 2011.

## 4. Separating the effects of transport and chemistry in the LS vortex O<sub>3</sub> column

Figure 4 showed that the absence of late winter  $O_3$  transport in the middle and lowermost stratosphere distinguishes 2011 from the other years and suggests that any  $O_3$  transported into the LS vortex column will likely be small. We quantify transport into the LS vortex during late winter by examining the changes in MLS  $N_2O$  vortex profiles. Figure 5a shows the descent of MLS  $N_2O$  vortex

average profiles on isentropic surfaces from early February (Jan. 29-Feb. 4 average) to late March (Mar. 20-26 average); the  $2\sigma$  uncertainties are shown by the dashed lines. Note that the 15K descent is statistically significant at the  $2\sigma$  level at 550K, but only to  $1\sigma$  below. The profile differences in the LS (400-550K, or ~100-32 hPa) indicate a net diabatic descent of 15K ( $\pm$  15K) over the 50 day period, or ~0.3 K potential temperature/day In this regard the 2011 Arctic vortex is much like the Antarctic. Late winter Antarctic LS vortex descent rates calculated by Rosenfield et al. (1994) are very similar to those calculated here for the Arctic, but are much smaller than those calculated in the same study for the Arctic. Figure 5b shows the effect of 15K descent of the Feb. 1 vortex  $O_3$  profile, which would result in a LS column increase of 14 DU. This calculation cannot be applied to the columns above or below because  $N_2O$  is not retrieved in the LMS and MS vertical gradients are nearly zero. The Arctic LS vortex maintained strong PV gradients at the edge throughout this time and the mean vortex  $N_2O$  profile did not increase indicating that meridional transport of midlatitude air into the vortex (higher  $N_2O$ ) was insignificant.

Figure 6 shows LS vortex column O<sub>3</sub> for 2004-2011. As discussed in Section 3.1, transport is primarily radiatively controlled at high latitudes in fall and early winter, resulting in low IA variability of the O<sub>3</sub> columns. For all years shown, the maximum LS vortex O<sub>3</sub> column in mid to late January is ~175-190 DU. After mid-January, vortex O<sub>3</sub> can increase due to a sudden warming (e.g., 2006, 2009, and 2010) or decrease due to chemical losses (e.g., 2005, 2007, 2008, and 2011). The late January O<sub>3</sub> maximum is reached before the onset of significant chemical loss. Notice that the LS vortex columns in early December 2010 (red) were near the top of the data envelope while by late January 2011 they were on the low side. Given that PSCs and high ClO were observed in Dec. 2010 and Jan. 2011 (Manney et al., 2011), it is possible that the change in the 2010-2011 data from the high to the low side of the 7 years is due to early season chemical loss. Calculations of heterogeneous chemical O<sub>3</sub> loss using the GMI model with MERRA meteorological fields show that a 12 DU vortex-averaged loss occurs during December (2 DU) and January (10 DU). Balis et al. (2011) used a chemical transport model (CTM) integrated with forecast meteorological fields to quantify chemical O<sub>3</sub> loss, obtaining a similar value for vortex-averaged loss of 13 DU for January 2011.

The observed late January 2011 LS vortex average column  $O_3$  is 183 DU. Using this number and the model calculations of early season loss (~12 DU), we estimate ~195 DU as a reasonable upper bound for the (no loss) 2011 LS vortex column  $O_3$  at this point in the season. The observed changes in vortex  $N_2O$  profiles suggest that the late winter LS ozone column increased by 14 DU. By late March 2011 the

observed LS vortex mean column was 129 DU. Using these values and estimated  $1\sigma$  uncertainties, we obtain a vortex-averaged total chemical  $O_3$  loss of

$$183 (\pm 3.7) + 12 (\pm 5) + 14 (\pm 7) - 129 (\pm 2.6) DU = 80 (\pm 10) DU.$$

The area-weighted probability distribution functions shown in Figure 7 illustrate how analysis of partial  $O_3$  columns separates the effects of the chemistry and transport. The top panels show that the means and distributions of the total and LS vortex columns experience large and similar changes. The bottom panels show that the means and distributions of the columns above and below the levels of activated chlorine are nearly identical in late January and late March, confirming the lack of transport previously discussed. The change in the total stratospheric column in late winter corresponds to the change in the LS column (54 DU). Together, these panels show that chemical loss has been isolated in the LS column. Including the effects of early season chemical loss and late winter LS transport, we calculate the total heterogeneous chemical  $O_3$  loss for the winter of 2011 (up to March 26) of 80 DU. This is slightly higher than the 65 DU calculated by Balis et al. (2011) using a CTM for the period 1 Jan-31 Mar 2011.

In the absence of chemical loss the stratospheric vortex column mean in late March 2011 would have been only 371 DU (maximum 80 DU estimated chemical loss + 291 DU observed by MLS). This 'no loss' vortex column estimate is 30-40 DU lower than observed in the 3 recent years that had an early major warming and few PSCs: 408 DU (2006), 402 DU (2009), and ~400 DU (2010, vortex gone by late March). And although three other recent years had late March stratospheric column  $O_3$  as low or lower than 2011 - 371 DU (2005), 340 DU (2007), and 356 DU (2008) – all three were estimated to have 80 DU or more in PSC-driven losses (WMO, 2011), bringing their pre-loss columns to more than 400 DU. In the absence of chemistry, all six recent years would have late March vortex mean column  $O_3$  that was at least 40 DU higher than 2011. The reason for this difference is the lack of any significant wave activity, including a final warming, by the end of March 2011.

## 5. The final warming in April 2011

The March Arctic and October Antarctic total column  $O_3$  means are often used to illustrate interhemispheric differences in polar  $O_3$  loss (WMO, 2011). The Antarctic shows large decreases in  $O_3$  since 1980 in the 40-yr time series shown in Fig. 2-8 of the WMO Report (2011), while in the Arctic a smaller  $O_3$  decline is partially obscured by large IA variability. The large IA variability is mostly driven by dynamical variability [Chipperfield and Jones, 1999; Newman et al., 2001], but chemical loss plays an

important role and is correlated with cold winters, which have weaker transport and lower  $O_3$  regardless of chemical loss. Ozone is transported to the Arctic during fall and winter, and the breakdown of the Arctic vortex results in a large, late winter contribution to that transport. For example, data from 2005-2010 presented in Figure 1c show typical late winter increases of ~50 DU since January. The March Arctic mean usually includes some contribution from the final warming, but occasionally the vortex remains intact and over the pole at the end of March. In such cases the late winter transport contribution to high spring  $O_3$  has not year occurred, resulting in unusually low March Arctic column  $O_3$  (e.g., 1997 (Coy et al., 1997)).

Figures 1a and 1b show OMI and MLS column O₃ on March 24, 2011. The vortex is large and strong on this date. Columns inside the vortex are low, in part, due to chemical O₃ loss, and a significant fraction of the seasonal poleward transport of high O<sub>3</sub> has not yet occurred. Figure 8a shows total column O<sub>3</sub> measured by OMI on April 10, 2011, as the breakdown occurs. The vortex, with column O<sub>3</sub> of <300 DU, is pushed off the pole while high O<sub>3</sub> air with columns >440 DU moves over it. Figure 8b shows time series of OMI daily column O<sub>3</sub> averaged over 63-90°N from Feb. 10 to April 30, 2005-2011. Except for 2005 and 2011 (blue and red), Arctic daily column means are above 400 DU before the end of February. In 2005 the late winter transport of high O<sub>3</sub> occurs in mid-March (blue) as the vortex breaks down; this produces mean March Arctic column for 2005 that is lower than the other years, not just because of chemical loss that took place, but because of the late timing of the transport. The final warming in 2011 begins in early April and the associated large O₃ transport is not part of the March average. By mid-April 2011, the OMI Arctic column O<sub>3</sub> appears quite 'normal' compared to the previous 6 years. The use of April instead of March mean Arctic O<sub>3</sub> would reduce much of the IA variability seen in Arctic  $O_3$  in the past 30 years (e.g., Tegtmeier et al., 2008; WMO, 2011), however it would be difficult to assess O<sub>3</sub> losses because most years the ozone-depleted vortex air is dispersed into the midlatitudes by April. The situation in the Antarctic is quite different, where IA variability in the timing of chemical losses or the vortex breakdown rarely contributes to the October mean  $O_3$ . Chemical  $O_3$  loss in Arctic cannot be accurately assessed without quantitative consideration of transport.

## 6. Discussion and Summary

Low IA variability inside the Arctic vortex in early winter combined with the exceptionally weak polar transport in the winter of 2011 provide the basis for using Aura MLS  $O_3$  and  $N_2O$  observations to separate the contributions of chemistry and transport to the low  $O_3$  columns observed in March 2011. Mean March Arctic  $O_3$  has large IA variability that complicates efforts to separate the effects of

chemistry and transport. We show that  $Arctic O_3$  variability comes from columns outside the vortex (outside PSC-forming regions), vortex area, and the timing of the final warming that transports high levels of  $O_3$  poleward. Column  $O_3$  inside the vortex shows little IA variability for the winters of 2005-2011 prior to the first warming of the season. These data allow us to estimate the amount of  $O_3$  in the LS vortex column before large chemical losses began in early February. The analysis of vortex  $O_3$  behavior presented here is made possible by the continuous three-dimensional coverage of the polar region by the MLS instrument.

We quantify the effects of chemistry and transport on vortex column  $O_3$  by separately evaluating the behavior of the LS column, which can be affected by heterogeneous chemistry and transport, and that of the LMS and MS columns, which are mainly affected by transport. In contrast to the behavior observed in 2005-2010, the LMS and MS 2011 columns show a near total lack of transport into the vortex between late January and late March. These two partial columns are 15-35 DU lower in late March than any of the previous 6 years. Using the descent observed in vortex  $N_2O$  profiles between late January and late March, we estimate that vertical transport brought 14 ( $\pm 7$ ) DU into the LS vortex  $O_3$  column during the period of PSC-driven chemical loss.

We estimate the total LS vortex column O<sub>3</sub> available for chemical loss by summing the observed late January LS column, the estimated early season O<sub>3</sub> loss, and the late winter O<sub>3</sub> transport inferred from N<sub>2</sub>O. The MLS vortex-averaged LS column O<sub>3</sub> was 183 DU in late January. Manney et al. [2011] state that small O<sub>3</sub> losses occurred in January based on MLS observations of enhanced vortex CIO after mid-December. We estimate that ~12 DU O<sub>3</sub> loss occurred by the end of January. Based on the 15K descent observed in MLS vortex mean N<sub>2</sub>O profiles, we estimate that late winter descent in the strongly isolated vortex contributed 14 DU to the LS O<sub>3</sub> column. This results in a maximum late winter LS column of 209 DU in the absence of heterogeneous chemical loss. Adding to this the late March LMS and MS columns, 61 and 101 DU respectively, we estimate that the vortex mean stratospheric column in the absence of heterogeneous chemistry would have been 371 DU. This 'no loss' Arctic column is very similar to pre-ozone hole (i.e., 'no loss') total columns observed in the Antarctic in the 1970's (WMO, 2011), underscoring the Antarctic-like meteorology of the 2011 Arctic vortex.

The observed vortex-averaged column for late March is 291 DU (Figure 7a). This represents a PSC-driven loss of 80 ( $\pm$ 10) DU averaged over the vortex, slightly more than the 65 DU calculated for PSC-driven loss by a CTM simulation (Balis et al., 2011). The chemical loss of 120 DU averaged over the vortex reported by Manney et al. (2011) was calculated by subtracting the 2011 MLS O<sub>3</sub> data from daily

Arctic column  $O_3$  observations averaged over 1979-2010. This 31-year mean  $O_3$  reflects the Arctic climatological mean meteorology, which includes the final warming occurring by late March. The 2011 Arctic meteorology was quite atypical with its final warming in April. Sinnhuber et al. (2011) report vortex losses of 120 DU by early April based on model calculations with a passive  $O_3$  tracer. Their simulated vortex  $N_2O$  compared to MIPAS observations showed a problem with insufficient descent or vortex isolation or both, indicating that  $O_3$  transport would not be accurately represented. If 120 DU  $O_3$  were lost in the 2011 vortex, the MLS  $O_3$  data indicate that transport of an additional ~40 DU into the LS vortex would be required between late January and late March. If the  $O_3$  arrived by descent then the  $N_2O$  late March profile would show about four times the observed descent (i.e, 60K descent). Meridional transport of 40 DU  $O_3$  into the vortex is unlikely as the March LS vortex maintained low  $N_2O$  and strong PV gradients. In this study we estimate a late winter vortex transport contribution of 14 DU. The additional 40 DU  $O_3$  required to produce a 120 DU  $O_3$  loss is approximately equal to the transport contribution of a final warming.

The final warming that resupplies  $O_3$  to high latitudes is an essential component of seasonal transport in the Arctic, and resupply is not necessarily complete in some years – or even begun – by the end of March (e.g., 1997 and 2011). OMI data have excellent coverage of the Arctic in March and April, demonstrating that long-lasting low temperature was not the only unusual feature of this year's Arctic stratosphere – the corollary to the long-lasting vortex is the late timing of the resupply, which began after April 1. An estimate of the amount of chemical  $O_3$  loss inside the vortex based on a multi-year average of late March conditions assumes that resupply has occurred. When it has not, this inappropriate baseline exaggerates chemical  $O_3$  loss. Heterogeneous chemical  $O_3$  loss in the Arctic was large in 2011, ~80 DU averaged over the vortex, but the timing of the final warming is an equally important part of the story of the March 2011 Arctic  $O_3$ . The Arctic  $O_3$  loss in 2011 may be the largest ever observed, but it is only half the amount lost in the Antarctic, where losses inside the vortex average ~150 DU (Tilmes et al., 2006).

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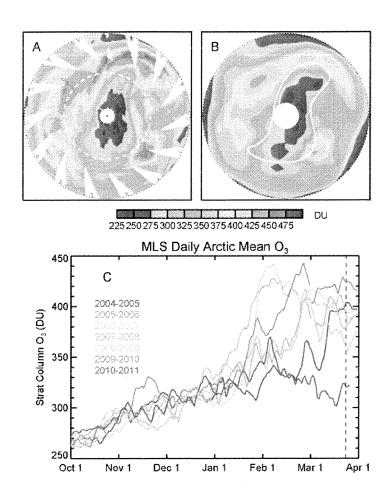


Figure 1. a) OMI Total Column  $O_3$  in the Arctic, March 24, 2011. The white dashed line is  $63^\circ N$ . b) MLS stratospheric column  $O_3$  on March 24, 2011. The white solid line shows the vortex edge (PV=4 x  $10^{-5}$  K m<sup>2</sup> kg<sup>-1</sup> s<sup>-1</sup>). The difference between a) and b) is the tropospheric column, ~25-35 DU. c) Daily MLS Arctic Mean ( $63^\circ$ - $88^\circ N$ ) stratospheric column  $O_3$  time series from Oct 1 to April 1 for seven fall/winter seasons, 2004-2011. March  $24^{th}$  is indicated by the dashed line.

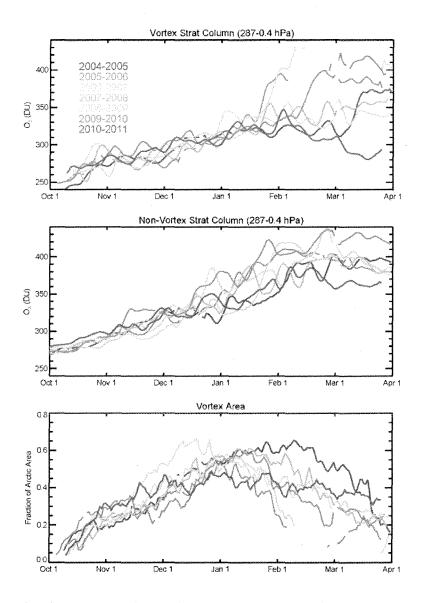


Figure 2. a) Daily MLS stratospheric column  $O_3$  averaged inside the Arctic vortex for 2004-2011, b) daily MLS stratospheric column  $O_3$  outside the vortex. Daily averages for a) and b) are calculated from MLS observations 54-88°N in order to capture all vortex observations during warmings. c) Fraction of the area poleward of 54°N occupied by the Arctic vortex. The vortex is defined by areas with PV≥4 on the 500K surface.

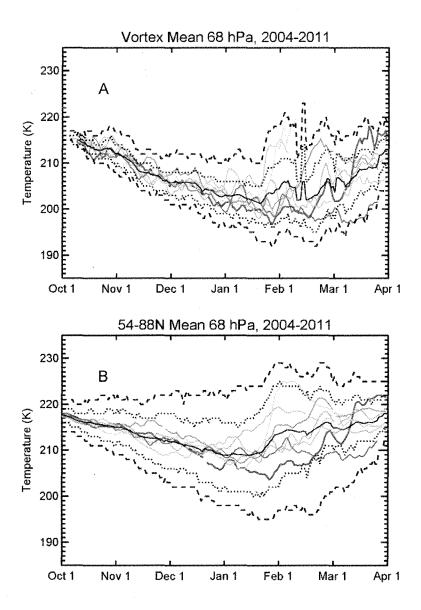


Figure 3. Daily MLS 68 hPa temperatures for 7 fall/winter seasons (colored lines). a) daily averages inside the vortex, and b) daily averages for the Arctic, 54-88°N. Heavy black line is the average of 7 years of daily averages. Dotted lines are the 25<sup>th</sup> and 75<sup>th</sup> percentiles of all daily temperatures observed for all years. Dashed lines are for the 10<sup>th</sup> and 90<sup>th</sup> percentiles.

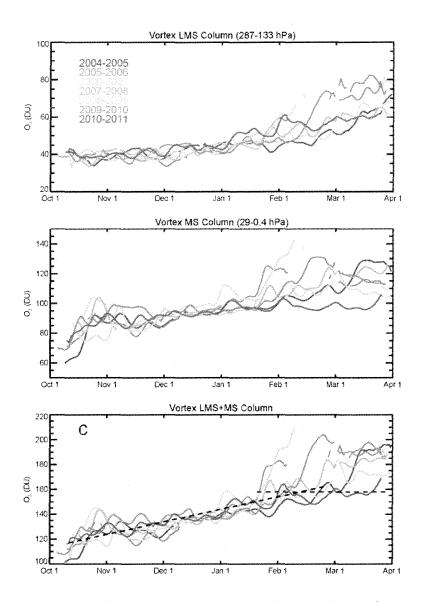


Figure 4. Daily MLS partial column  $O_3$  averaged inside the vortex for 7 fall/winter seasons. a) The lowermost stratosphere (LMS) column, 287-133 hPa, b) the middle/upper stratosphere (MS) columns, 29-0.4 hPa, and c) the sum of the LMS and MS partial columns. The dashed lines show the increasing trend in 2010-2011 columns before February and the lack of increase in February and March.

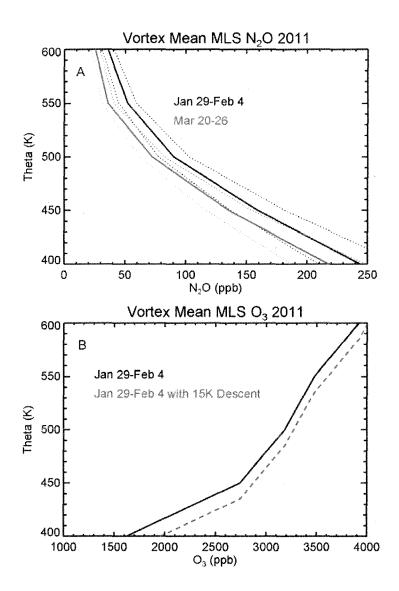


Figure 5. a) Vortex-average  $N_2O$  profiles near Feb. 1 (Jan. 29-Feb. 4 average, black) and near March 23 (Mar. 20-26 average, red). The  $N_2O$  profile descends 15K in the lower stratosphere (400-550K) during this period. B) Vortex-averaged  $O_3$  profile near Feb. 1 (black) and the same profile shifted downward by 15K to estimate the effect of descent on  $O_3$  (red). These changes produce a LS column increase of 14 DU.

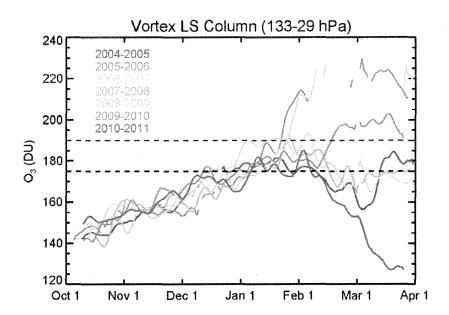


Figure 6. Daily MLS lower stratosphere vortex column  $O_3$ , 133-29 hPa. The dashed lines show the approximate range of LS column amounts observed in mid-late January for all years, excluding major warming (2006 and 2009). The difference between the late January and late March columns in 2011 is 183-129 DU (54 DU).

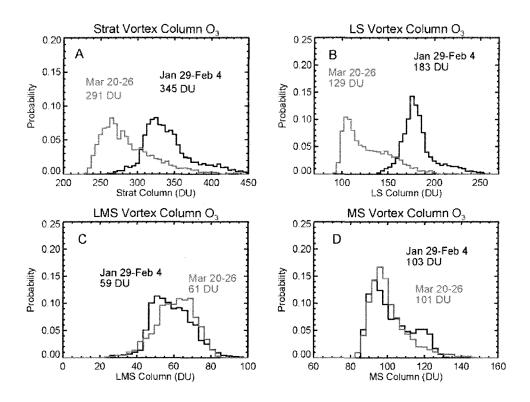
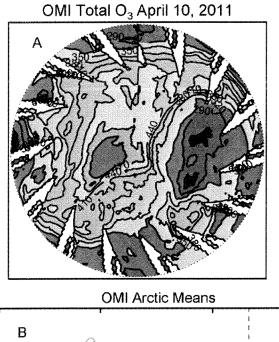


Figure 7. Comparison of probability distribution functions of MLS vortex column  $O_3$  at the end of January (Jan 29-Feb 4, 2011, black) before late winter chemical loss with MLS vortex columns after the period of large chemical loss (Mar. 20-26, red). There are no MLS observations from March 27 to April 18, 2011. The stratospheric and lower stratospheric columns show the same change, 54 DU. The LMS and MS columns show very little change to their means or distributions.



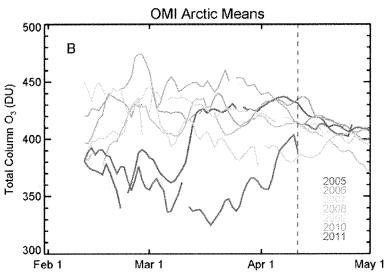


Figure 8. a) OMI total column  $O_3$  on April 10, 2011. The final warming has begun. The vortex, where columns are < 320 DU, has been pushed off the pole and replaced by high column  $O_3$  (>400 DU) from the midlatitudes. b) Daily OMI Arctic mean total column  $O_3$  (63-90°N) for 7 winter/spring seasons. OMI data are only shown after February  $10^{th}_3$  when observations first reach 70°N. In all years except 2005 and 2011, Arctic column  $O_3$  is above 400 DU almost daily in February and March. The 2005 vortex persisted until early March, after which Arctic mean columns exceeded 400 DU. The 2011 vortex broke down much later than the other years shown, but mean columns exceeding 400 DU are reached by mid-April.