Decline in Antarctic Ozone Depletion and Lower Stratospheric Chlorine Determined From Aura Microwave Limb Sounder Observations

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Abstract Attribution of Antarctic ozone recovery to the Montreal protocol requires evidence that (1) Antarctic chlorine levels are declining and (2) there is a reduction in ozone depletion in response to a chlorine decline. We use Aura Microwave Limb Sounder measurements of O3, HCl, and N2O to demonstrate that inorganic chlorine (Clly) from 2013 to 2016 was 223 ± 93 parts per trillion lower in the Antarctic lower stratosphere than from 2004 to 2007 and that column ozone depletion declined in response. The mean Clly decline rate, ~0.8%/yr, agrees with the expected rate based on chlorofluorocarbon lifetimes. N2O measurements are crucial for identifying changes in stratospheric Clly loading independent of dynamical variability. From 2005 to 2016, the ozone depletion and Clly time series show matching periods of decline, stability, and increase. The observed sensitivity of O3 depletion to changing Clly agrees with the sensitivity simulated by the Global Modeling Initiative chemistry transport model integrated with Modern Era Retrospective Analysis for Research and Applications 2 meteorology.

Plain Language Summary The Antarctic ozone hole is healing slowly because levels of the man-made chemicals causing the hole have long lifetimes. We use Microwave Limb Sounder (MLS) satellite data to measure O3 over Antarctica at the beginning of winter and then compare it to O3 near the end of winter to calculate depletion. During this period, nearly all O3 change is due to depletion. MLS also measures HCl, and when ozone levels are very low, nearly all the reactive chlorine species (Clly) are converted to HCl. Clly varies a lot from year to year from atmospheric motions. Fortunately, MLS measures nitrous oxide (N2O), a long-lived gas that also varies with the motions. Using the ratio of Clly to N2O, we find that there is less chlorine now than 9 years ago and that Clly has decreased on average about 25 parts per trillion per year (0.8%/yr). The O3 depletion we calculate from MLS data responds to changes in the Clly levels, and the ratio of change in ozone loss to the change in Clly matches model calculations. All of this is evidence that the Montreal Protocol is working—that the Clly is decreasing in the Antarctic stratosphere and the ozone destruction is decreasing along with it.

1. Introduction

Unambiguous observational evidence for a decrease in Antarctic ozone depletion attributable to the Montreal Protocol and its amendments remains elusive more than 20 years after ozone depleting substance (ODS) emissions ceased (World Meteorological Organization (WMO), 2015). The breakdown of chlorine-containing ODSs produces stratospheric inorganic chlorine (Clly). The extent of Antarctic ozone depletion each year depends primarily on Clly and temperature in the Antarctic vortex. Inorganic chlorine consists of long-lived species, HCl and ClONO2, and short-lived species such as Cl, ClO, and HOCl. Clly can be treated as a long-lived trace gas whose distribution is controlled by transport (Plumb & Ko, 1992). In the upper stratosphere where HCl represents nearly all Clly, satellite data analyses identified a global decrease in HCl from 1997 to 2000 (Anderson et al., 2000) and from 2004 to 2006 (Froidevaux et al., 2006). Rinland et al. (2003) used column measurements of ClONO2 and HCl to show that global inorganic chlorine levels had stabilized by 2001. But in the Antarctic lower stratosphere, both the detection of ozone recovery and its attribution to the Montreal Protocol remain challenging because year-to-year variations in ozone depletion are largely driven by temperature rather than chlorine variations, and there are no Clly observations to verify a decrease. Nedoluha et al. (2016) applied a temperature-sensitivity adjustment to column ClO measurements at 78°S to estimate the Antarctic Clly trend. Their analysis of ground-based measurements showed a decline since 1996 consistent with the expected decline, but it began earlier than expected and was not statistically significant.
They estimated a \(-0.5 \pm 0.4 \, (2\sigma) \, \%/\text{yr}\) Cl\(_y\) trend from Microwave Limb Sounder (MLS) ClO data 2004–2015. The expected Cl\(_y\) decline rate in the Antarctic lower stratosphere, controlled by long ODS lifetimes, is \(-0.8\%/\text{yr}\) (Newman et al., 2007; Strahan et al., 2014).

Several studies have argued for positive trends in Antarctic ozone based on multivariate analyses of the satellite O\(_3\) data record. Salby et al. (2011) found an increasing Antarctic O\(_3\) trend from 2000 to 2009, averaged between September and November, after accounting for dynamical variability in late winter. Hassler et al. (2011) questioned these results, noting that in October and November Antarctic O\(_3\) has large variability due to temperature and transport that is not driven by the late winter wave forcing used by Salby et al. (2011). The analysis of Kuttippurath et al. (2013) used a simulated passive O\(_3\) tracer along with the satellite O\(_3\) record to fit terms for dynamics and chemistry, finding a +1 Dobson unit (DU)/yr trend from 2000 to 2010. However, neither this study nor Salby et al. (2011) accounted for the significant interannual variability of vortex Cl\(_y\) caused by stratospheric transport variations (Strahan et al., 2014, hereafter referred to as S2014), which controls the total chlorine available for ozone loss. Based on their analysis of South Pole ozone sondes from 1986 to 2010, Hassler et al. (2011) expect that a signature of ozone recovery will be found in decreased ozone loss rates in August and September. Solomon et al. (2016) also recognize the difficulty of detecting O\(_3\) trends in October due to the large dynamical variability, and instead use the changes in September O\(_3\) to identify evidence for recovery. They used a specified dynamics simulation to attribute half of the O\(_3\) increase from 2000 to 2014 to declining halogens but were unable to attribute the other half.

These studies and others recognize that each year’s ozone depletion depends strongly on temperature. Antarctic temperature and its interannual variations, measured by satellite instruments, are well represented in meteorological analyses (Lawrence et al., 2015). Year-to-year temperature variations have a much larger impact on ozone loss and the size of the ozone hole than do annual changes in Cl\(_y\) (Newman et al., 2004). Lacking observations that verify declining Cl\(_y\) levels in the Antarctic LS, attribution studies often rely on an assumed steady rate of Cl\(_y\) decline. S2014 showed that Antarctic Cl\(_y\) does not decline monotonically. They used MLS N\(_2\)O to show that year-to-year variations in Antarctic Cl\(_y\) levels can be up 10 times greater than the expected 20–22 parts per trillion (ppt)/yr decline rate. Without measurements of a dynamical tracer to account for this variability, a decade of Cl\(_y\) decrease (~200 ppt) is required for 95% confidence that Antarctic Cl\(_y\) has decreased since the previous decade.

This study tackles the attribution problem by using MLS measurements of N\(_2\)O, O\(_3\), and HCl from 2004 to 2016 to identify conditions under which HCl represents nearly 100% of Cl\(_y\) in the Antarctic lower stratosphere. MLS O\(_3\) data are used to estimate seasonal Antarctic O\(_3\) depletion based on vortex-averaged O\(_3\) changes between July and mid-September when transport and dynamical variabilities are relatively low. Most satellite O\(_3\) measurements are made by UV instruments (e.g., Total Ozone Mapping Spectrometer and Ozone Monitoring Instrument), but MLS measures microwave emissions, making it uniquely suited for quantifying high-latitude ozone depletion during winter. Using the MLS HCl, N\(_2\)O, and O\(_3\) data sets, we show that Cl\(_y\) has declined with respect to a steady dynamical reference (N\(_2\)O) and that from 2005 to 2016 changes in vortex-averaged column O\(_3\) depletion correspond to the Cl\(_y\) changes. While the findings of multivariate regression studies of springtime Antarctic total ozone indicate the beginning of ozone recovery, uncertainties in measurements and in the statistical analyses used preclude the definitive conclusion that Antarctic stratospheric ozone is increasing due to declining ODSs (WMO, 2015). This study uses observations to determine a lower stratospheric Cl\(_y\) decline and commensurate change in O\(_3\) depletion inside the Antarctic ozone hole.

2. Data and Methods

2.1. Data Sets

We use version 4.2 MLS N\(_2\)O (190 GHz), HCl (640 GHz), and O\(_3\) (240 GHz) standard products from 2004 to 2016 in all analyses. The accuracies reported here are from the MLS V4.2 data quality document (Livesey et al., 2017). Temporal instability in any of the measurements affects the analysis results; thus, we emphasize the impact of drift in the following paragraphs.

We use O\(_3\) on pressures from 261 to 12 hPa to calculate partial columns. The reported column 2\(\sigma\) accuracy is 4%. An evaluation of the O\(_3\)-240 GHz product compared to correlative satellite and ground-based measurements shows no evidence of a temporal drift (Hubert et al., 2016). The drift between the O\(_3\)-240 and O\(_3\)-640
products was found to be $\leq 0.1\%$/yr (L. Froidevaux, personal communication, 2017); thus, other measurements obtained with the 640 GHz receiver, including HCl and N$_2$O, are expected to be comparably stable (N. Livesey, personal communication, 2017). The vertical range of the standard HCl product is 100–10 hPa; the reported $2\sigma$ accuracy is 200 ppt and MLS HCl data agree with ACE-FTS HCl measurements to within $\pm 5\%$ (Froidevaux et al., 2008).

We use the N$_2$O data product from 190 GHz receiver because the N$_2$O-640 data set ends in 2013. The N$_2$O-190 data on the 46 and 68 hPa levels used here have a $2\sigma$ reported accuracy of 10 and 22%, equivalent to $\sim 10$ ppb and $\sim 30$ ppb, respectively, in the Antarctic. The MLS measurements with the 190 GHz receiver have been found to have a drift relative to the 640 GHz receiver (N. Livesey, personal communication, 2017). N$_2$O-190 has a mean drift rate and $2\sigma$ uncertainty of $-0.7 (\pm 0.3)\%$/yr at the latitudes and pressures used here relative to N$_2$O-640 (L. Froidevaux, personal communication, 2017). N$_2$O has a well-measured surface growth rate of $-0.24\%$/yr (Elkins & Dutton, 2009); the combined measurement drift and growth rate is $-0.5 (\pm 0.3)\%$/yr. We correct for the drift and growth rate in the analysis so that Cl$_y$ change can be calculated using N$_2$O as a steady dynamical frame of reference (section 3.1).

We use potential vorticity and temperature from the Modern Era Retrospective Analysis for Research and Applications 2 (MERRA2) (Gelaro et al., 2017) to identify the vortex edge and calculate vortex-averaged quantities. The temperature fields are used to create an Antarctic temperature climatology for 1980–2016. MERRA2 fields were also used to integrate a simulation using the Global Modeling Initiative (GMI) chemistry transport model (CTM) for the Aura period (Strahan, Douglass, & Newman, 2013). This simulation is used to demonstrate the close relationship between HCl and Cl$_y$ under ozone-depleted conditions. Details are provided in the supporting information.

### 2.2. Estimates of Antarctic Vortex Winter Ozone Depletion

We calculate chemical depletion as the difference between vortex-averaged ozone partial columns in early and late winter in a way to minimize O$_3$ changes caused by dynamics. We identify the vortex edge by the maximum MERRA2 potential vorticity gradient constrained by the location of the wind jet maximum on the 450 K surface ($\sim 50–60$ hPa) (Nash et al., 1996). The O$_3$ partial columns are calculated only for MLS levels where depletion occurs, 261–12 hPa. MLS partial column O$_3$ varies by about $\pm 20$ DU each year in the early winter vortex prior to significant depletion. We remove this source of dynamical variability from the depletion estimate by subtracting the 1–10 July partial column O$_3$ (early winter) from the 11–20 September vortex-average (late winter). We average over each 10 day period to minimize the effect of short-term dynamical fluctuations on column O$_3$. Because declining Cl$_y$ will reduce O$_3$ loss rates in August and September (Hassler et al., 2011), this period is appropriate for detecting a change in depletion due to changing chlorine while minimizing dynamically-driven O$_3$ variability that increases in spring. Nedoluha et al. (2016) observed that column ClO variability increased rapidly after 17 September due to temperature variability. Dynamical activity influencing column O$_3$ increases after mid-September (e.g., Kramarova et al., 2014) leading to an increased variability in O$_3$ loss that comes from temperature rather than a change in Cl loading. The loss rates through mid-September do not control the size of the O$_3$ hole or the minimum column. The maximum area inside the 220 DU contour and the O$_3$ column minimum are found between late September and mid-October; they vary considerably due to temperature variability.

### 2.3. Determination of Cl$_y$ From HCl

Under certain conditions, nearly 100% of total inorganic chlorine is converted to HCl. From October to mid-November the vortex is generally quite strong and remains severely ozone-depleted and denitrified. Under these low O$_3$ conditions, the catalytic O$_3$ loss cycle involving ClO$_x$ ceases and Cl instead reacts with CH$_4$ to produce HCl. The low O$_3$ levels also increase the NO/NO$_2$ ratio, leading to greater Cl production via NO + ClO, followed by conversion of that Cl to HCl through reaction with CH$_4$ (Douglass et al., 1995; Prather & Jaffe, 1990). After a few days under these conditions, HCl is approximately equal to Cl$_y$.

To identify HCl measurements that represent Cl$_y$, we select data points with severely depleted O$_3$ (O$_3$ less than 300 ppb at 68 hPa and less than 500 ppb at 46 hPa). A simulation with the GMI CTM of the Aura period confirms that selecting HCl data conditioned on severely depleted O$_3$ accurately identifies locations where HCl equals 96–98% of Cl$_y$. Co-located N$_2$O data points are used to provide an independent dynamical
coordinate for assessing Cl\textsubscript{y} change (discussed below). The simulated HCl conditioned on low O\textsubscript{3} produces a compact tracer-tracer correlation with N\textsubscript{2}O as expected; see the supporting information for details.

3. The Decline in Antarctic O\textsubscript{3} Depletion and in Lower Stratospheric Cl\textsubscript{y}

3.1. The Decline in HCl (Cl\textsubscript{y}) With Respect to N\textsubscript{2}O

We use N\textsubscript{2}O as a reference coordinate to identify Cl\textsubscript{y} change that is independent of dynamical variability and attributable to declining ODSs. Dynamical variability has been shown to produce multyear “trends” in HCl that are unrelated to changes in stratospheric Cl\textsubscript{y} loading (Douglass et al., 2017; Mahieu et al., 2014). Woodbridge et al. (1995) and Schauf\textsubscript{fler} et al. (2003) found a compact relationship between N\textsubscript{2}O > 50 ppb and Cl\textsubscript{y} using a comprehensive set of airborne measurements of organic halocarbons in the polar lower stratosphere. S2014 took advantage of this compact relationship to infer Cl\textsubscript{y} variability inside the Antarctic vortex from MLS N\textsubscript{2}O observations. The absolute values of Cl\textsubscript{y} inferred by this method depend on the surface mixing ratios of chlorine-containing source gases, the Antarctic mean age spectrum, and fractional release rates of those source gases (Newman et al., 2007).

As described in section 2.3, we use co-located MLS O\textsubscript{3} and HCl measurements to identify points where the HCl measurement is nearly 100% of Cl\textsubscript{y}. The observations show a compact tracer-tracer relationship between HCl conditioned on low O\textsubscript{3} (hereafter indicated by HCl (Cl\textsubscript{y})) and co-located N\textsubscript{2}O in all years 2004–2016 (Figure 1), confirming the expected relationship between HCl and Cl\textsubscript{y} under low O\textsubscript{3} conditions (see the supporting information). Each distribution in Figure 1 is made from more than 9,000 N\textsubscript{2}O/HCl measurement pairs. For each year, observations were chosen from days with the highest HCl values and the fewest low HCl values; these conditions are found each year during a 1–2 week period between 1 October and 10 November. Data from 4 to 5 years are combined on each panel in order to fully sample the range of polar lower stratospheric N\textsubscript{2}O values. The solid line shows the mean values for the frequency distributions for N\textsubscript{2}O values 50–170 ppb (from the 46 and 68 hPa levels only). The Schauf\textsubscript{fler} et al. (2003) relationship (dashed line) has the same shape but is offset ~150 ppt below MLS values in Figure 1a. Some of the distributions’ breadth comes from the precision of the MLS measurements. However, the distribution of the N\textsubscript{2}O/HCl pairs is sharply peaked, demonstrating the compactness of the tracer-tracer relationship.

In GMI simulations there is greater scatter than seen in Figure 1 when other forms of Cl\textsubscript{y} are present (not shown). When analyzed separately, the slopes determined from the 46 and 68 hPa observations are the same where their mixing ratios overlap. The distributions were calculated using 10 ppb wide N\textsubscript{2}O bins, which were adjusted by −0.5%/yr in order to account for the N\textsubscript{2}O growth rate and the MLS measurement drift rate during this period.

The dashed line showing the Schauf\textsubscript{fler} fit is the same in all panels. While all of the points in Figure 1a sit above the dashed line, most are found below this line by 2013–2016 (Figure 1c), indicating a decline in HCl (Cl\textsubscript{y}) inside the Antarctic vortex. The compact correlation of N\textsubscript{2}O and HCl (Cl\textsubscript{y}) for 2008–2012 (Figure 1b) lies in between for N\textsubscript{2}O < 120 ppb. The declining HCl (Cl\textsubscript{y}) is attributable to declining source gases because dynamically driven transport variations do not alter the N\textsubscript{2}O/Cl\textsubscript{y} relationship. The GMI simulation shows similar decline rates for HCl and Cl\textsubscript{y} and that declines occur throughout the Antarctic at pressures where depletion occurs.

Figure 1d quantifies the 9 year HCl (Cl\textsubscript{y}) change (red) as the difference between the mean relationships in Figures 1a and 1c that were fit to a second-degree polynomial nearly identical to the Schauf\textsubscript{fler} curve except for the offset (blue and black dashed). The 2σ uncertainties in the HCl (Cl\textsubscript{y}) change, shown as red dotted lines, come almost entirely from uncertainty in the polynomial fits but above 120 ppb N\textsubscript{2}O the drift rate contributes 20–35% to the uncertainty. The mean decline over 9 years is 223 ± 93 ppt (2σ), or 25 ± 10 ppt/yr on the 46 and 68 hPa levels. The mean is weighted by the uncertainty in the HCl (Cl\textsubscript{y}) change for each N\textsubscript{2}O bin, and the polynomial fit uncertainties are greatest at the highest and lowest N\textsubscript{2}O bins.

The variation in HCl (Cl\textsubscript{y}) decline over the observed N\textsubscript{2}O range is not expected based on GMI model calculations. It may be caused by the drift rate’s dependence on the MLS averaging kernels and the steep N\textsubscript{2}O vertical gradients inside the vortex (N. Livesey, personal communication, 2017). The HCl (Cl\textsubscript{y}) change for N\textsubscript{2}O > 120 ppb (Figure 1d, red) is much more sensitive to the assumed N\textsubscript{2}O-190 drift rate than lower N\textsubscript{2}O
values and may explain the small HCl (Cl<sub>y</sub>) changes at higher N<sub>2</sub>O values shown in Figure 1b. Smaller-magnitude drift rates produce less variation in the decline. These results are equivalent to a mean Cl<sub>y</sub> decline of ~0.8%/yr and agree with the rate calculated by Newman et al. (2007) using ODS fractional releases and an Antarctic mean age of 5.5 yrs. The GMI-MERRA2 simulation in Figure S1c in the supporting information has a 26 ppt/yr decline rate (~0.9%/yr) at 68 and 46 hPa, with similar decline rates throughout the Antarctic lower stratosphere (see Figure S1 in the supporting information). The excellent agreement between these independent determinations increases our confidence in the MLS-derived results and affirms our understanding of the chemical and physical processes controlling stratospheric chlorine.

### 3.2. Vortex-Averaged Cl<sub>y</sub> From N<sub>2</sub>O Measurements

Ozone loss occurs throughout the Antarctic vortex and most loss occurs in September. To attribute changes in ozone depletion to changes in Cl<sub>y</sub> requires an estimate of the September lower stratospheric vortex-averaged Cl<sub>y</sub>. In this section we use the N<sub>2</sub>O/Cl<sub>y</sub> slope and Cl<sub>y</sub> decline rate determined in section 3.1 along with September vortex-averaged V4 N<sub>2</sub>O-190 to infer vortex-averaged Cl<sub>y</sub> from 2004 to 2016.
Both temperature and Cl$_y$ have large biennial variability. A power spectral analysis of the MERRA2 Antarctic temperatures for 1980–2016 finds the greatest power at the 2 year period; Baldwin and Dunkerton (1998) found no effect of the QBO phase on the Antarctic vortex winds or temperatures during winter. The Antarctic Cl$_y$ variability is driven by slowly declining ODSs and the QBO and has no known physical relationship to temperature variability. We apply a low-pass 1-2-1 filter to both time series to eliminate the high-frequency (i.e., ≤2 years) variability to reveal the lower frequency signal from slow ODS (Cl$_y$) decline. The resulting time series shown in Figure 3b reveal three distinct periods that show the response of O$_3$ depletion to changes in Cl$_y$. From 2005 to 2010, O$_3$ loss and Cl$_y$ decline together, while from 2010 to 2013, they are nearly constant and are at their lowest levels of the Aura period. After 2013, there is a dynamical increase in Cl$_y$ and a corresponding increase in O$_3$ depletion.

Figure 2 shows the 12 year time series of vortex-averaged N$_2$O on the 450 K isentropic surface, averaged from 1 to 20 September. The N$_2$O data are adjusted annually for the measurement drift and surface growth rates. The large interannual variability is due to composition variations that originated in the midlatitude middle stratosphere in the previous winter and are caused by the quasi-biennial oscillation (QBO) meridional circulation (Strahan et al., 2015). High N$_2$O from 2010 to 2013 was caused by a series of winter QBO easterlies from 600 to 700 K during the previous years (2009–2012), which produced anomalously high midlatitude N$_2$O. Winter QBO westerlies at this level in 2013 and 2015 led to low N$_2$O midlatitude anomalies, reversing the earlier upward trend. The vortex-averaged V3 N$_2$O-640 from S2014 (dashed line) is 10–15% lower but shows the same interannual variability as the V4 N$_2$O-190 analysis.

The Cl$_y$ time series in Figure 2 is calculated from the vortex-averaged N$_2$O and the initial N$_2$O/HCl slope (2004–2007), annually adjusted using the mean Cl$_y$ decline rate of 25 ppt/yr determined in section 3.1. Figure 2 shows that vortex-averaged Cl$_y$ decreases steadily from 2004 to 2010 with biennial variability. The QBO influence discussed above resulted in a period of anomalously low vortex Cl$_y$ from 2010 to 2013 followed by higher Cl$_y$ from 2013 to 2016. The downward N$_2$O trend from 2011 to 2016 indicates that increasing Cl$_y$ during this period is dynamically driven and explains why Cl$_y$ in 2016 is only 85 ppt lower than in 2004 in spite of the mean decline rate of 25 ppt/yr. Changes in Cl$_y$ on the 450 K surface (~50–60 hPa) impact the O$_3$ column because significant depletion occurs here (Hassler et al., 2011).

3.3. The Decline in Ozone Depletion Since 2005 and Attribution to Cl$_y$

We seek to identify changes in ozone depletion that occur in response to changes in Antarctic Cl$_y$. The MLS instrument is ideally suited for measuring changes in depletion because it observes ozone throughout the winter as far south as 82°. As previously described, to minimize the contribution from dynamics, we calculate the changes in vortex-averaged partial column O$_3$ between 1–10 July and 11–20 September. GMI simulations with and without heterogeneous chemistry show that more than 99% of ozone loss occurs in the partial column, 12–261 hPa, less than 10 DU of the loss occurs before July, and this period encompasses about 80% of the seasonal depletion.

Figure 3a shows each winter’s O$_3$ change (loss). (There are no MLS measurements in early July 2004.) The 1σ ozone loss uncertainties, shown as vertical lines, are calculated from the standard deviations of the 10 day vortex-averaged means in early July and mid-September. The interannual variations in ozone loss are large and are strongly modulated by temperature (Newman et al., 2004). To identify the coldest years, we calculate the anomalies, reversing the earlier upward trend. The vortex-averaged N$_2$O-190 from S2014 (dotted) is 10–15% lower than new V4 (N$_2$O-190) but has similar variability.

Figure 2. September vortex-averaged N$_2$O (black) and Cl$_y$ (red) on the 450 K surface. Cl$_y$ is calculated from vortex N$_2$O and the N$_2$O/Cl$_y$ relationship in Figure 1a, adjusted annually by the Cl$_y$ mean decline rate (see text). The V3 time series (N$_2$O-640) from S2014 (dotted) is 10–15% lower than new V4 (N$_2$O-190) but has similar variability.
Figure 3c shows GMI O₃ loss and vortex Clᵧ calculated using the same methods and filtering as in Figure 3b.

The sensitivity of ozone loss to Clᵧ obtained from MLS, dO₃ loss/dClᵧ, is nearly identical to that shown in the GMI simulation and equal to about 80 DU O₃ loss for every ppb of Clᵧ. The agreement between the observed and simulated sensitivity supports the conclusion that changes in MLS O₃ loss are in response to Clᵧ changes.

The trends of increasing Clᵧ in the last few years of Figures 3b and 3c are dynamically driven. Figure 1 shows that HCl (Clᵧ) has declined about 223 ppt over 9 years when examined in a framework independent of dynamical variability.

4. Conclusions

Inorganic chlorine, Clᵧ, was estimated each year by judiciously selecting MLS HCl measurements conditioned on low O₃ in midspring while the vortex remains strong. The results show that Antarctic HCl (Clᵧ) levels have decreased by 223 ± 93 ppt over a 9 year period, equivalent to an annual rate of 25 ± 10 ppt/yr (~0.8%/yr). The
use of N₂O as a reference coordinate is essential for quantifying the change in stratospheric Clᵥ loading independent of dynamical variability.

We used the MLS-derived Clᵥ decline rate along with vortex-averaged September N₂O to determine vortex Clᵥ on the 450 K surface from 2004 to 2016. The Clᵥ variability is driven by slowly declining ODSs and the QBO. Antarctic temperature and Clᵥ have large but physically unrelated biennial variability. With high-frequency variability removed, Aura MLS observations show evidence of changes in ozone depletion in response to Clᵥ changes from 2005 to 2016. From 2005 to 2010, depletion and Clᵥ decrease together, while from 2010 to 2013, both depletion and Clᵥ levels hold steady. After 2013, Clᵥ levels rise and there is a corresponding rise in depletion; N₂O reveals that the uptick in Clᵥ is caused by dynamics and not an unexpected increase in ODS loading. The observed changes in depletion and Clᵥ are consistent with the Clᵥ/Clᵥ sensitivity found in the GMI CTM simulation. A reduction in Antarctic ozone depletion attributable the Montreal Protocol can be seen over this 12 year period.

Antarctic ozone depletion calculated as the difference between July and mid-September vortex-averaged partial column O₃ is an optimal way to identify the impact of Clᵥ on depletion while minimizing dynamical contributions to O₃ change. Using MLS partial column O₃, we have shown that vortex-averaged O₃ loss decreased over the 12 year data record because lower stratospheric Clᵥ levels are lower. Although temperature continues to play the dominant role in interannual variations in depletion, depletion levels are responding to changes in Clᵥ. As Clᵥ continues to decline, the signal of recovery will become stronger.

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