

1 **Using Satellite Measurements of N₂O to remove dynamical variability from HCl**
2 **measurements**

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9
10 *Abstract:*

11
12 Column HCl measurements show deviations from the expected slow decline
13 following the regulation of chlorine-containing compounds by the Montreal Protocol.
14 We use the simultaneous measurements of N₂O and HCl by the MLS instrument on
15 the Aura satellite to examine this problem. We find that the use of N₂O
16 measurements at a specific altitude to represent the impact of dynamical variability
17 on HCl results in a derived linear trend in HCl that is negative (ranging from -
18 2.5%/decade to 5.3%/decade) at all altitudes between 68 hPa and 10 hPa. These
19 trends are at or near 2 σ statistical significance at all pressure levels between 68 hPa
20 and 10 hPa. This shows that analysis of simultaneous measurements of several
21 constituents is a useful approach to identify small trends from data records that are
22 strongly influenced by dynamical interannual variability.

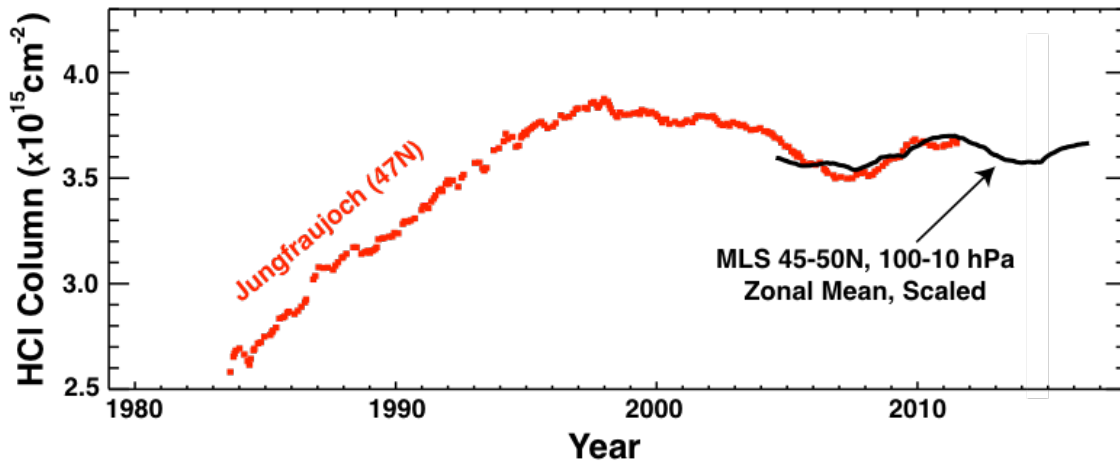
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24 **I. Introduction**

25
26 HCl is the primary constituent of inorganic chlorine in the stratosphere, comprising
27 75-80% of the inorganic chlorine in the vertical pressure range from 68 hPa to 10
28 hPa [Zander et al., 1992; Nassar, et al. 2006]. As such it provides a convenient
29 marker for the total amount of inorganic stratospheric chlorine. This marker can be
30 measured from the ground as a total column amount and from satellites as a vertical
31 profile. The column amount of HCl is expected to follow the behavior of the
32 concentration of the organic sources of chlorine as measured at the surface (e.g.
33 CFCs) with a time delay of a few years for the CFCs to reach the stratosphere where
34 they are converted to inorganic chlorine compounds.

35
36 Rinsland et al. [2003], using the NDACC record of ground-based column
37 measurements of HCl and ClONO₂ (~1990 – 2002), showed that their total
38 stratospheric burden had leveled out by approximately 1995. These two gases
39 comprise most of stratospheric inorganic chlorine outside the winter polar vortices.
40 The next step would be to observe the expected decrease in inorganic chlorine.
41 Recently Mahieu et al. [2014] have shown that, in fact, the measured HCl column
42 over Jungfraujoch decreased more rapidly than expected from in-situ ground-based
43 measurements of source gases during the early 2000's. The rapid decrease in
44 measured HCl column was followed by an increase from about 2007 to 2010 even as
45 the chlorine gases were decreasing. We show, as Mahieu et al. [2014] indicated,

1 that the Microwave Limb Sounder (MLS) measurements of the lower stratospheric
2 column of HCl also decrease and increase in concert with the Jungfraujoch data,
3 followed by a decrease from 2011 to mid 2013 and a subsequent increase from that
4 time to the present (see Figure 1).

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9 Figure 1: Total column measurements of HCl above Jungfraujoch (latitude = 46.5°N)
10 smoothed with a 3-year running mean as shown by Mahieu et al. [2014] (red curve). Also
11 shown are the 3-year running mean smoothed zonal mean of measurements of the lower
12 stratospheric column of HCl (100-10 hPa) from the MLS instrument on Aura for the latitude
13 band from 45 to 50N (black curve). The MLS measurements are of partial column and have
14 been scaled upward to match the Jungfraujoch FTIR measurements for better visual
15 comparison.

16

17 Mahieu et al. [2014] use results from model simulations with the SLIMCAT model
18 driven by ERA-Interim meteorological fields from the European Centre for Medium-
19 Range Weather Forecasts (ECMWF) to suggest that variability in the stratospheric
20 circulation causes the accelerated decrease and the unexpected increase in HCl
21 column. We will explore this explanation using measurements of N₂O from MLS as a
22 measure of this variability in circulation.

23

24 2. MLS Data: HCl and N₂O

25

26 We use the MLS HCl and N₂O data together to test whether chlorine is decreasing in
27 the stratosphere as expected from adherence to the provisions of the Montreal
28 Protocol. MLS was launched on the Aura Satellite in July 2004 [Waters et al., 2006]
29 and continues to operate in 2018. The record is now more than 13 years in length
30 with vertical profiles of HCl, HNO₃, N₂O and many other species measured globally
31 on a daily basis.

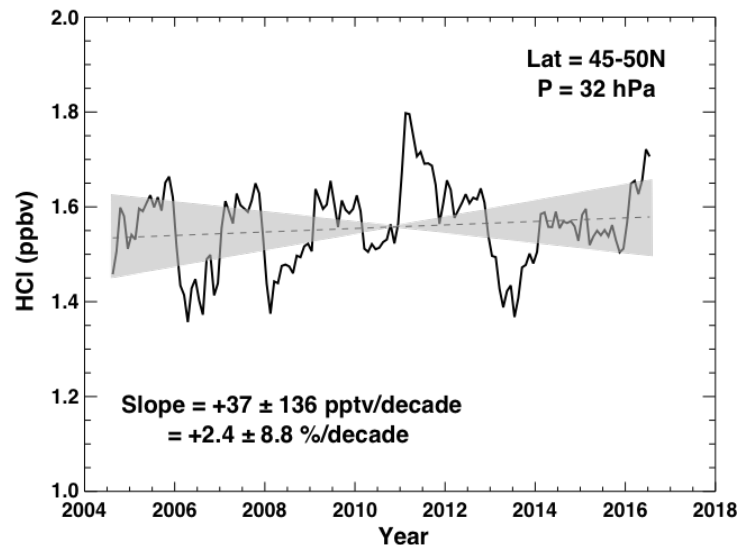
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33 For HCl we use the version 4.2 product, measured by the 640 GHz receiver, that
34 shows little change from the previous version 3 products. According to the MLS

1 data quality document [Livesey et al., 2017] the useful range for HCl measurements
2 is from 100 to 0.32 hPa. However, the useful data for trends are limited to pressures
3 greater than 10 hPa due to the insufficient reliability of the retrievals in the upper
4 stratosphere. The possibility of a temporal drift in the HCl product was evaluated by
5 comparing the drift between the O₃-240 and O₃-640 (L. Froidevaux, personal
6 communication, 2017). Because the ozone products had a drift of ≤0.1%/yr, other
7 measurements obtained with the 640 GHz receiver, e.g., HCl and N₂O, are expected
8 to be comparably stable. An evaluation of the O₃-240 GHz product compared to
9 correlative satellite and ground-based measurements shows no evidence of a
10 temporal drift (Hubert et al., 2016).

11
12 For N₂O we use the version 4.2 data product from 190 GHz receiver because the N₂O
13 640 GHz data set ends in summer 2013 due to failure of the N₂O primary band. The
14 MLS measurements with the 190 GHz receiver have been found to have a temporal
15 drift relative to the 640 GHz receiver (N. Livesey, personal communication, 2017).
16 Later in this section we will compare N₂O-190 and N₂O-640 measurements to
17 correct for the drift observed between these two channels prior to 2013. The 190-
18 GHz N₂O data are stated to be useful in the 68-0.46 hPa range. We will thus restrict
19 our analyses to pressure levels between 68 and 10 hPa in this paper where both the
20 HCl and N₂O-190 measurements are useful.

21



22

23 Figure 2: Deseasonalized monthly-mean MLS measurements of HCl concentration at 32 hPa.
24 Measurements are area-weighted between 45°N and 50°N. The dashed line is a linear least
25 squares fit to the data and the shaded area indicates the 2σ uncertainty in that fit including
26 consideration of auto-correlation in the time series.

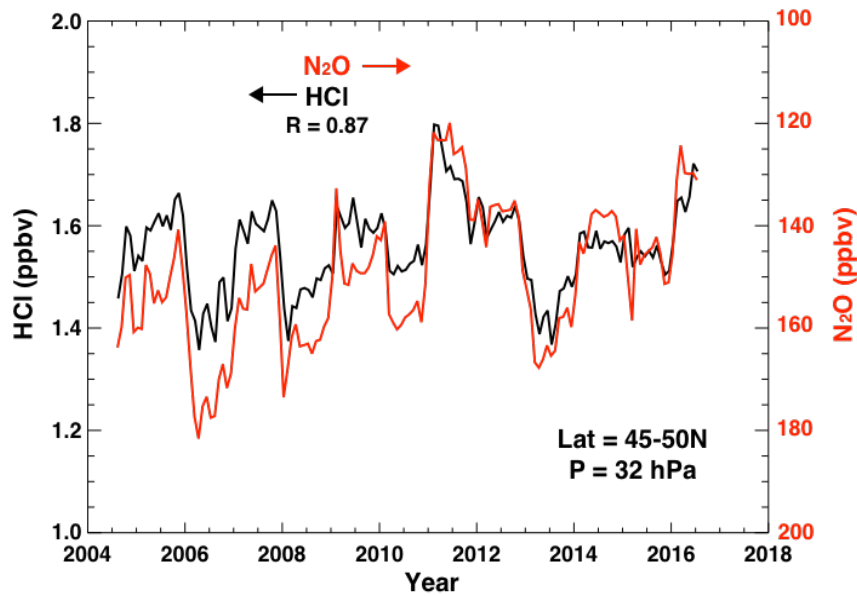
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28 To better understand the variations in HCl column amounts at 47°N observed by
29 Mahieu et al. [2014] we begin with consideration of the MLS measurements of the
30 HCl profile at specific pressure levels in the stratosphere. For example, Figure 2
31 shows the deseasonalized monthly mean measurements by MLS of HCl averaged

1 between 45°N and 50°N latitude at 32 hPa. The data are shown as mixing ratio after
2 removal of the repeating seasonal cycle. The data clearly show deviations of as
3 much as $\pm 10\%$ with significant auto-correlation.

4
5 One way to examine the HCl time series shown in Figure 2 is to attempt to “explain”
6 the variance by fitting to various measures of dynamical variability such as the
7 Quasi-Biennial Oscillation (QBO) or El-Niño/Southern Oscillation (ENSO). This
8 method may remove much of the dynamical variance but has at least two potential
9 problems: 1) the fitting parameters may only remove part of the dynamical
10 variability because of incomplete representation of that variability and 2) they may
11 over-represent the variability because of correlation between parameters. Either of
12 these problems could lead to difficulties in separating real trends from apparent
13 trends in the residual over short time scales such as the 12 years of data since 2004.
14 For example, the impact of the QBO on southern mid-latitude composition depends
15 on the QBO phase during early (southern) winter [Strahan et al. 2015]. The
16 resulting dynamical variability is not easily represented by fitting a QBO plus a
17 seasonal term in a statistical model, because the actual variability depends on the
18 QBO phase during a particular season. We have attempted to model the HCl time
19 series with standard proxies for QBO and other effects with unsatisfactory results.

20
21 We use a different method to remove dynamical variability in the HCl data set,
22 taking advantage of simultaneous measurements of another species made by the
23 MLS instrument on the Aura satellite. The observed deviations from the seasonal
24 variation of HCl and other constituents including N₂O are the result of dynamical
25 variability acting on mixing ratio gradients. These gradients may be vertical,
26 horizontal, or a combination of both. If two constituents have gradients in the same
27 or opposite directions, the impact of dynamic variability will be to cause deviations
28 that are either correlated or anti-correlated with each other depending on the sign
29 of the gradients. An example is shown in Figure 3 where we plot the deseasonalized
30 HCl mixing ratios at 32 hPa for the latitude band 45-50N as in Figure 2 and the
31 deseasonalized N₂O mixing ratios on a reverse scale for the same latitude band and
32 pressure level. The correlation coefficient is -0.87 between these two time series. A
33 similar correlation, with opposite sign, is found between HCl and HNO₃ data from
34 MLS.

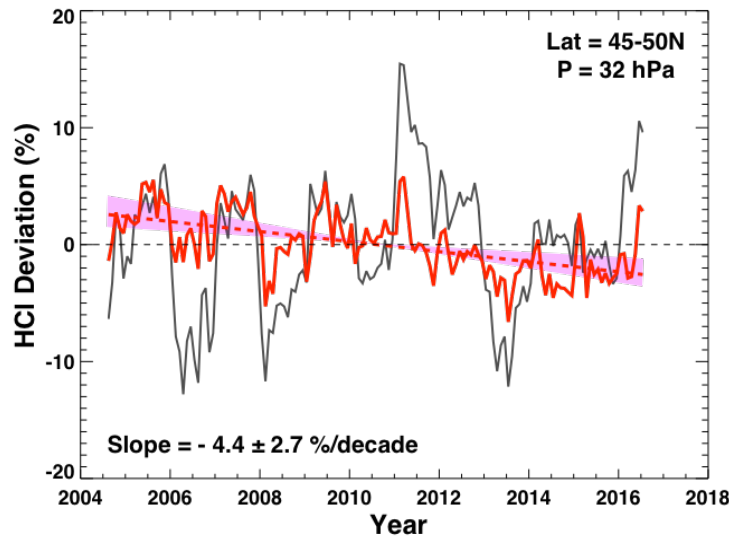


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2 Figure 3: Deseasonalized time series of MLS HCl measurements (black curve, same as Figure
3 2) and deseasonalized time series of MLS N₂O measurements plotted with reverse scale on
4 right side of figure (red curve) for the latitude band 45-50N at 32 hPa pressure level.

5
6 **3. Time Series Analysis: Using N₂O Measurements as a Fitting Parameter**

7
8 The trend that we are trying to isolate and confirm for the HCl time series is
9 determined by the change in abundance of chlorine-containing halocarbons driven
10 by the provisions of the Montreal Protocol on ozone-depleting substances.
11 Inorganic chlorine in the stratosphere is expected to have decreased since 2000 in
12 response to the decreases in the chlorine-containing source gases. N₂O, on the other
13 hand, is known to be increasing at a rate of about 2.8%/decade [NOAA GMDL data
14 updated from Elkins and Dutton [2009] available at
15 ftp://ftp.cmdl.noaa.gov/hats/n2o/combined/HATS_global_N2O.txt]. Our approach
16 is to use the N₂O time series at each altitude, such as that shown in Figure 3, as an
17 explanatory variable in a time-series regression to remove the dynamical variability
18 from the HCl time series. Trends calculated for HCl in this time-series regression
19 are then corrected for the underlying trend in N₂O.

20
21 The time series method used is a simple regression with two fitting terms, a linear
22 trend and the N₂O time series. Uncertainties are estimated by calculating the
23 standard deviation of the residual time series after removing the fit to linear trend
24 plus the N₂O time-series and then multiplying the result by the factor
25 $((1+\Phi)/(1-\Phi))^{1/2}$ to obtain the uncertainty including auto-regression (see
26 Weatherhead et al. [1998]) where Φ is the auto-correlation lag 1 coefficient.



1
2 Figure 4: HCl anomaly time series as in Figures 2 and 3 with the mean removed (black) and
3 the residual time series after regression to N₂O time series (red). Red dashed line is linear
4 fit to residual series with 2σ uncertainty bounds indicated by shaded area.

5
6 The result of using N₂O as a fitting parameter for the 32 hPa MLS time series for HCl
7 is shown in Figure 4. The solid red line in the figure is the residual time series after
8 fitting, which takes advantage of the substantial covariance and shows significantly
9 reduced variability. The resulting trend shown by the red dashed line is $- 4.4 \pm 2.7$
10 (2σ) %/decade. The HCl trend at this pressure level is now negative and
11 statistically significant at more than the 3σ level. The same procedure has been
12 carried out at each of the pressure levels for MLS retrievals. The result is shown the
13 third column of Table 1.

14
15 The second column of Table 1 shows the raw trend obtained from the MLS HCl
16 measurements. We can see that the raw trend is essentially the same as the N₂O-
17 fitted trend at the two highest levels (10 and 15 hPa) where inorganic chlorine is
18 relatively insensitive to dynamical variability due to its small vertical gradient. At
19 higher pressures, where inorganic chlorine has a larger vertical gradient, the trend
20 is substantially different when using N₂O as a fitting parameter. Using N₂O as a
21 fitting parameter changes the lower level trends from positive to negative with a
22 substantial reduction in the uncertainty.

23
24 A further problem alluded to earlier in this section is that MLS now uses the 190-
25 GHz band for its standard N₂O product because the 640-GHz band is no longer
26 usable. The problem arises because the 190-GHz band displays a drift in N₂O
27 measurements with respect to the 640-GHz band [L. Froidevaux, pers. commun.].
28 Since the 640-GHz band was found to be stable during its operational period, we use
29 the drift rate of the 190-GHz band with respect to the 640-GHz band calculated by
30 the time-independent drift rates for the time period in which they both were
31 operational (2004-2012). We assume that the drift rate computed over the overlap

1 time period continues through the end of the data record. This assumption is
 2 subject to an uncertainty that we have no way of evaluating. The results for the
 3 computation of the drift between the 190-GHz band with respect to the 640-GHz
 4 band are shown in the fourth column of Table 1 with estimated uncertainties in the
 5 fit.

6
 7 We chose to show the trend resulting from the fit to the N₂O product before
 8 adjusting it for the drift between the two N₂O bands and then following that with a
 9 correction for the N₂O drift. Because we are making a simple straight-line trend
 10 correction with no time variability except the trend, the result is the same as that
 11 obtained by first correcting the N₂O product time series and using that result as a
 12 fitting parameter to the HCl data.

Pressure Level (hPa)	Raw HCl Trend (%/dec)	Trend with N ₂ O fit (%/dec)	N ₂ O 190/640 drift (%/dec)	Surf N ₂ O Trend (%/dec)	Final HCl Trend (%/dec)
10	-3.7±2.0	-3.0±2.0	-5.1±3.7	+2.8±0.05	-5.3±4.2
15	-3.8±3.1	-3.5±1.9	-2.9±2.2	+2.8±0.05	-3.6±2.9
22	-1.3±4.2	-3.8±2.3	-1.5±1.3	+2.8±0.05	-2.5±2.6
32	+2.4±8.8	-4.4±2.7	-2.2±1.4	+2.8±0.05	-3.8±3.0
46	+3.8±7.2	-2.3±2.3	-5.2±1.8	+2.8±0.05	-4.7±2.9
68	+3.9±4.0	-1.3±2.7	-5.0±1.6	+2.8±0.05	-3.5±3.1

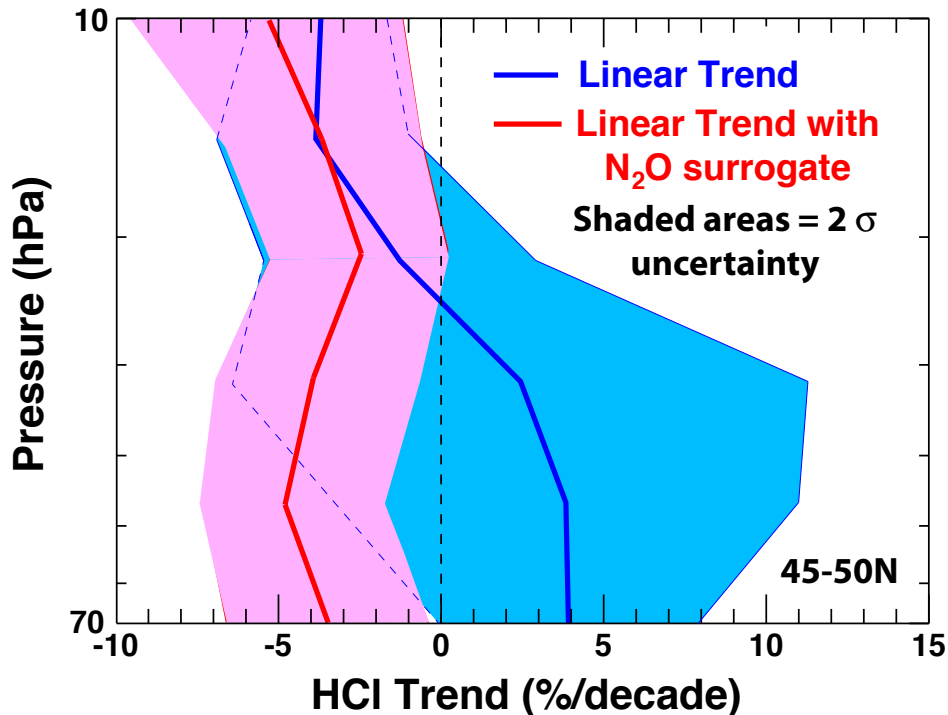
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 15 Table 1. HCl trends derived from MLS data at 6 pressure levels. Column 1 gives the
 16 pressure level. Column 2 gives the raw trend derived directly from the MLS HCl
 17 measurements in %/decade. Column 3 gives the HCl trend derived using MLS N₂O
 18 measurements as an explanatory variable. Column 4 gives the derived trend in the 190-GHz
 19 channel of MLS N₂O measurements relative to the 640-GHz channel during the time of their
 20 overlap. Column 5 gives the slope of the NOAA global surface measurements of N₂O over
 21 the period from 2001 to 2012. Column 6 gives the overall resulting trend obtained by
 22 combining the information in columns 3 to 5. All uncertainties are quoted at 2σ.

23
 24 Finally, we note that the surface levels of N₂O increased by about 2.8%/decade. We
 25 used the “global” nitrous oxide data reported at the NOAA ESRL Global Monitoring
 26 Division web site (<https://www.esrl.noaa.gov/gmd/hats/combined/N2O.html>). We
 27 fit a trend to the data between 2001 and 2012 to represent the ground values that
 28 should be seen in the stratosphere about 3 years later in the MLS data. The time
 29 delay represents the approximate mean age of air in the lower stratosphere (see e.g.
 30 Waugh and Hall [2002]). Since N₂O has been increasing at a nearly constant rate, the
 31 choice of time period for estimating its trend does not lead to a significant
 32 uncertainty.

33
 34 The final result is obtained by summing the trends in columns 3 to 5 of Table 1 and
 35 is shown in column 6 of the table. The uncertainties were obtained by using the root
 36 sum of squares (RSS) of the uncertainties in columns 3 to 5. This final result is also
 37 shown in Figure 5. The blue solid line indicates the trend result obtained from a

1 linear fit to the deseasonalized residuals with no attempt to account for dynamical
2 variability (column 2 of Table 1). The red solid curve is the result when the N₂O
3 time series is used as a dynamical surrogate, with corrections for drift and surface
4 trends, in the fitting procedure (column 6 of Table 1). The shaded areas represent
5 2σ uncertainties in the linear trends.

6
7 Note in Figure 5 that the use of N₂O as a surrogate for dynamical variability reduces
8 the uncertainty in the calculated trend for all of the pressure levels between 15 hPa and
9 68 hPa where the MLS data for both HCl and N₂O are considered to be suitable
10 for trend analysis. Figure 5 also shows that the calculated linear trend in HCl using
11 the N₂O surrogate with corrections is negative at all pressure levels and is
12 significantly negative (2σ) at all levels from 68 hPa to 10 hPa with the exception of
13 22 hPa where it has nearly 2σ significance. These results are consistent with the
14 observed decrease in organic chlorine species at the surface.



17
18 Figure 5: Linear trend in HCl concentrations determined from MLS measurements between
19 70 and 10 hPa (approximately 20 to 30 km altitude) for the latitude band of 45-50N. The
20 blue line is the trend determined from the raw deseasonalized data. The red curve is the
21 trend determined while including the N₂O time series as an explanatory variable. The
22 shaded areas represent 2σ uncertainties for each.

23 24 25 4. Conclusion

1 In their paper, Mahieu et al. [2014] reported that total column HCl measured at
2 Jungfraujoch showed significant variation from the expected simple linear decrease.
3 We have attempted to quantitatively evaluate the effect of dynamical variability on
4 the concentrations of HCl in the stratosphere by using N₂O measurements since late
5 2004 made by the Aura MLS instrument. Since both HCl and N₂O have
6 concentration gradients (horizontal and vertical) that are acted upon by dynamical
7 processes to create inter-annual variability, we have used the variability of N₂O
8 concentrations determined from MLS observations as a measure of the dynamical
9 variability that should be expected in HCl concentrations. We suggest that this
10 method more reliably removes the real atmospheric variability than does the use of
11 other proxies.

12
13 We have shown that using an N₂O surrogate in trend analysis of the MLS HCl time
14 series results in a trend that is negative at all measured levels from 68 hPa upward
15 to 10 hPa and that these negative trends are 2 σ statistically significant or nearly so.
16 The N₂O surrogate had little effect at the upper two levels of 10 and 15 hPa where
17 inorganic chlorine is less sensitive to dynamical variability. The surrogate had
18 significant impact on the derived trends lower in the stratosphere where the
19 inorganic chlorine vertical gradient is larger and most of the HCl column resides.

20
21 Previous data-based estimates of HCl decrease in the stratosphere include
22 Froidevaux et al. [2006], Jones et al. [2011], Brown et al. [2011], and Kohlhepp et al.
23 [2012]. All of these studies had to consider the issues we have discussed in this
24 paper, namely the contribution of dynamic variability to the apparent trend. In each
25 case, the shortness of the data record was a significant limitation to the
26 interpretation of potential trends due to the decrease in tropospheric organic
27 chlorine sources.

28
29 The results from these authors are summarized in the 2014 Ozone Assessment
30 Report [Carpenter and Reimann, 2014]. Specifically Froidevaux et al. [2006]
31 derived a trend for the 50-65 km altitude range of -8%/decade from MLS data for
32 the years 2004-2006. Jones et al. [2011] derived a trend of -5%/decade using
33 HALOE and ACE FTS data between 35 and 45 km from 1997 to 2008 at midlatitudes.
34 Brown et al. [2011] deduced a trend of -7%/decade for the 50 to 54 km range from
35 ACE FTS data from 2004 to 2010. Finally, Kohlhepp et al. [2012] analyzed the total
36 column HCl data from 17 NDACC FTIR stations for the years 2000 to 2009, obtaining
37 trends that ranged from -4 to -16%/decade depending on station.

38
39 The best comparison for evaluating our results is considering the change in the
40 organic chlorine sources at the surface. The 2014 ozone assessment, Chapter 1,
41 [Carpenter and Reimann, 2015] estimates changes in the tropospheric available
42 organic chlorine of -6%/decade from 2000-2004 followed by -4.6%/decade from
43 2004-2008 and -4%/decade from 2008-2012. Assuming a 3-5 year delay between
44 changes in the tropospheric source gases for the stratospheric chlorine implies an

1 average change from 2004 to 2016 of about -5 %/decade, in agreement with our
2 estimate from MLS data within the uncertainty bounds.

3
4 These results indicate the potential power of using the time series of measurements
5 of one constituent to understand, and possibly remove, the dynamical variability in
6 another constituent. In the case we have presented, we had to apply a drift
7 correction to the MLS N₂O data to get the best estimate of trend. We have attempted
8 to make an estimate of the uncertainty in this drift correction and include it in the
9 estimate of the overall uncertainty in the trend calculation. Although the drift
10 correction was a large enough fraction of the overall trend to cause some worry
11 about the results, we assert that this does not diminish the value of the concept of
12 using the dynamical variability of a other measured constituents to provide
13 complementary information about trends and variability of the constituent whose
14 possible trends are under consideration.

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