

The referee's comments are presented followed by our responses in *italic* script.

Anonymous Referee #1

This manuscript introduces a method aiming at accounting for (correcting for) the real variability observed in time series of stratospheric tracers. More specifically, simultaneous measurements of hydrogen chloride (HCl, the main reservoir of stratospheric chlorine) and nitrous oxide (a long-lived source of nitrogen) are used in conjunction, so that the variability of stratospheric N₂O is used to remove the one of HCl, assuming they have the same origins, here circulation and transport. The ultimate aim is to determine HCl trends unaffected by atmospheric variability, useful for the verification of the effectiveness of the Montreal Protocol in the stratosphere.

It is claimed that this method is more appropriate than those using proxies for representing multiyear dynamical variabilities resulting from, e.g., the QBO, ENSO. In the present case, the implementation of the method to simultaneous measurements of HCl and N₂O by the Aura/MLS instrument results in the determination of significant HCl decreases in the lower stratosphere over the 2004-2016 time period (about 13 years) which are in agreement with the evolution of total organic chlorine at the surface, when accounting for the mean age of stratospheric air. In contrast, direct HCl trends are mostly not significant over this decadal time scale.

One can foresee that the method presented in this study will be used in future trend evaluations, for HCl, but also for other target gases relevant to ozone depletion and recovery, when trying to reconcile tropospheric and stratospheric trends, . . . provided that simultaneous measurements of tracers with similar response to atmospheric dynamical variability or other influences are available.

Therefore, I recommend publication of this study, after consideration of the suggestions indicated below.

Major comments

In order to allow precise implementations of this approach or replication of the method in future studies, it would be good to have available a description on how the "time-series regression" is actually performed (by simple ratioing or more elaborated ways). I believe this would not be possible with the current version of the text.

See comment after next paragraph.

Another aspect which is not described is the evaluation of the uncertainties affecting the various trends. It is stated in caption of Fig. 2 that "the 2σ uncertainty in that fit includes consideration of auto-correlation in the time series". Various approaches have been used in recent papers such as to account for auto-correlation in the data sets, particularly for studies of ozone recovery. But we do not know how the uncertainty ranges were determined in the present instance, while visual inspection of the HCl and N₂O time series suggests that auto-correlation might be quite significant. It would be good to know how the authors accounted for auto-correlation. A brief description of the statistical evaluation of the confidence intervals should be added. A citation might be relevant if the current method has been used before.

Have added a short paragraph explaining the time series model and the estimate of trend uncertainty including an estimate for the increase in uncertainty due to auto-correlation of the residuals. We have added a citation to Weatherhead et al. [1998] where the method is described in detail.

Specific comments and remarks

There is a mismatch between the main text and the captions for Fig. 2 and 3. It is indicated in the text that anomalies or deviations are shown. But it looks like the captions correctly state that deseasonalized time series are shown for HCl and N₂O. Several portions of the main body text need to be amended accordingly, or the captions and figures 2 and 3 updated.

Changed “anomalies” to “time series” in the text

Minor comments or typos

-Page 1/line 30: suggest changing to “... a few years for them to reach. . .”
We prefer to spelling out “CFCs” rather than “them”

-Page 1/line 37: Jungfraujoch is misspelt
fixed

-Page 1/line 38: suggest changing to “. . .from in situ surface measurements. . .”
done

-Page 2/line 14: might be good to indicate why N₂O is a relevant target for this purpose (and/or add a reference)
We feel that this paper explains why N₂O is a relevant target. This is particularly evident in Figure 3 where we show the covariance of the time series of N₂O and HCl. This is one of the main points of the paper.

-Page 3/line 1: I don’t think that there is a comparison between the N₂O products from MLS, only the drift as a function of altitude is given
This has been reworded to avoid the ambiguity..

-Page 4/line 24: “deseasonalized” is misspelt
fixed

-Page 5/line3-line5: this sentence needs to be reworded
Has been reworded to to indicate that stratospheric chlorine is expected to have changed due to changes in chlorine-containing source gases.

-Page 5/line 16: “HCl anomaly time series as in Figures 2 and 3”; true if Fig. 2 and 3 are updated accordingly
Reworded to indicate that it is same as Figures 2 and 3 with the mean removed.

-Page 6/line28-30: a good reference is needed here, to introduce the concept of age of air, and showing that a 3 years delay is appropriate
Added a reference to the review paper by Waugh and Hall. We do not feel the need to explain age of air as it is explained in detail in Waugh and Hall and is only a minor point in the present paper.

-Page 7/figure 5: perhaps provide approximate altitude information on the right scale?
Added words in caption to indicate approximate altitude.

-Page 7/line 23: suggest replacing “showed” by ‘reported’
Changed

-Page 7/line 24: suggest replacing “linear trend” by “linear decrease”
Changed

-Page 8/line 36: [Carpenter and Reimann, 2014] instead of 2015
Fixed

-Page 9/line 8: 2014 instead of 2015
Fixed

Anonymous Referee #2

This paper presents a nice perspective on observed variability and trends in northern mid-latitude stratospheric HCl. It describes an approach whereby dynamical influences on that variability can be accounted for through consideration of a trace gas such as N₂O, which shares many of the dynamical influences as HCl but experiences different chemical processes.

While I recognize the value of the method described, and am keen to see it appear in the literature, I am concerned the uncertainties ascribed to some of the numbers found/used by the authors are on the optimistic side. I wonder if more complete assessment of these uncertainties might lead to a reduction in the reported "significance" of the result and this assessment might then suggest that a softening of some of the wording is merited. I also have a concern as to whether the ordering of the operations in their method is appropriate, and whether more robust results might be obtained if it were reversed. Both of these topics are expanded upon below.

The standard of English is reasonably high, but it would clearly have benefited from a more careful read through by the authors as there are several parts that are erroneously and/or ambiguously worded. I've endeavored to identify some of these, but fear I may have overlooked some others.

Major concerns

My concern about the ordering of steps in the method is as follows. I would have thought that it would have been better to "correct" the N₂O for both the likely MLS drift and the surface growth rate before using it as an explanatory variable in the HCl analysis rather than, as appears to be the case, after. My sense is that this would lead to a corrected N₂O variable that would do a better job of explaining the dynamical influences on the HCl, enabling a clearer trend to be obtained. The results may be little different in the end, but my sense is that the study would be better expressed in that manner. If nothing else, the authors would do well to enact that alternative formulation and comment on the difference it makes to the result (even if they chose not to show it in the end). It might make sense to include an actual algebraic expression for the fit and the various corrections. This would make for an easier description for the various terms involved and their uncertainties.

Actually, to correct the MLS drift we fit the difference between the bands with a linear trend plus a seasonal cycle in the mean and a seasonal cycle in the trend. In the end, we simply took the linear trend portion of the fit to the difference between the two bands. We did not feel that the knowledge of the drift, and how to extrapolate it, was good enough to justify more than just a simple linear correction. Using the simple linear trend means that it makes no difference to the end result where in the process we apply the correction. The uncertainty in the fit trend was obtained in the same way as all of the linear trend uncertainty estimates in the paper, from the standard deviation multiplied by a factor from the Weatherhead et al. reference to account for autocorrelation of the residuals. We have added a short explanation of this reasoning.

My more major concern relates to the uncertainties quoted for some of the results. This is particularly important given the extent to which many of them are only just statistically significant (using the authors' 2-sigma threshold). Firstly, it is clear that the level-to-level variations in the bottom line results are mostly driven by the reported N₂O 190/640 drift ($r=0.75$ between it and the result) rather than by the observed HCl trend ($r=-0.24$). That is to say, the results are affected more by the "correction" than by the actual input (the latter being the HCl trend with the N₂O fit term included). Accordingly, this correction deserves particular scrutiny. The degree of level-to-level changes in this drift term is large compared to the uncertainty quoted on many of the individual drifts. Arguably, the standard deviation (1-sigma=1.5%/decade) of these different estimates would be just as valid a measure of the uncertainty in any or all of them. Indeed it might have been just as valid to chose to use the multi-level-mean drift as the value for all levels, given the uncertainty introduced by the inherent assumptions being made. Foremost among those assumptions is the

one that the N₂O drifts seen in the first part of the MLS mission are the same as those expected in the post-2013 period, when the 640 GHz N₂O product is unavailable. I would have thought that the uncertainties derived here might need to be inflated in some way to account for this. Might more information be gained through consideration of other MLS products measured in the same period? Fundamentally, I think more information is needed here (including from the MLS team) on these uncertainties and their validity.

We do appreciate this concern. Our goal was to emphasize the concept of modeling variability in measurement time series by using the variability of another measured constituent. We have added a statement to this effect in the abstract and a paragraph at the end of the paper to mention these concerns and emphasize the conceptual focus of the paper.

My second concern on the uncertainty relates to the 0.05%/decade (2-sigma) uncertainty quoted on the impact of N₂O emissions. Firstly, the use of a constant 2.8%/decade trend at all altitudes here strikes me as highly simplistic. There are factors such as changes in age of air (and its spectrum) that surely come into play and might lead to variations. Similarly, the use of a 3-year lag at all altitudes seems overly simplistic. I grant that these issues may only have a small impact, and they may be very hard to quantify from the measurements available. Thus, the use of a constant value may well be justified in that light. However, I find it hard to believe that, in the face of those issues, the 0.05%/decade 2-sigma uncertainty estimate is an appropriate one.

Since the surface N₂O trend is close to linear, the time delay has little impact on the results and contributes little to the uncertainty.

If nothing else, I would urge the authors to validate this number through, for example, examination of CCM runs (to which this team has ready access). Quantifying the degree to which the modeled 45N N₂O timeseries at different pressure levels tracks the surface trend would provide a useful measure of this uncertainty.

Actually we first realized the possibility of this approach from model results. The model has better correlations than the data. We feel that the measurements shown in Figure 3 with the high degree of correlation between HCl and N₂O clearly illustrate that these quantities are correlated in the atmosphere. Citing details from model results would not add much to the discussion.

This issue is perhaps tied up with the ordering one discussed above, as the use of N₂O as an explanatory variable for the sought-after HCl trends may absorb these factors to some extent (though I haven't thought this through fully). In my mind all these issues argue that a more complete exploration of their methods, their inherent assumptions, and the uncertainties therein should be included in the manuscript.

We understand that this is simplistic. The idea was to keep from overcomplicating the analysis to keep from obscuring the main point about the concept of using measured constituent variability in place of standard proxies for dynamical variability. We have added sentences in the abstract and in the conclusion that broaden the point a bit by pointing out that even if you do not use the second constituent (N₂O in this case) as a direct proxy, you do gain important information by examining the time series of other species that co-vary with the one you are considering (HCl in this case). This information is particularly important when dynamics may be the cause of the apparent "trend". Strahan et al. (2011, JGR) showed that N₂O and mean age have a linear relationship up to 30 hPa in the midlatitudes. This means that age spectrum variations are not important to the midlatitude N₂O used in the 32-68 hPa range of this study. That paper also showed mean ages of 2-4 years in this region, which is why a 3-year lag between the surface and the lower stratosphere was chosen.

More minor points

— Page 1

Line 19: "Statistically" -> "Statistical"
fixed

Line 25: "altitude" -> "vertical" (as you're using pressures rather than altitudes in words that follow).
Changed to "vertical pressure"

Line 27: "... amount of inorganic stratospheric chlorine. This marker can be ..." to avoid the ambiguity about whether it is the HCl or the inorganic chlorine that "can be measured from the ground and from satellites".
fixed

Line 33: Commas needed after "showed" and "measurements"
fixed

Line 34: "Inorganic chlorine" is more than just HCl and ClONO₂, though granted the others may be minor. Or is the point that Rinsland et al. only measured those two species and argued that they are the bulk inorganic chlorine. Please clarify.
added clarification

Line 37: Jungfraujoch misspelt
fixed

Line 38: "during the early 2000s. This was followed by an increase in the HCl column over Jungfraujoch from ..." to avoid the ambiguity about whether it is the HCl or the source gases (the most recent things being discussed) being referred to.
clarified

Lines 39-43: The way this is worded, it seemingly ignores the fact that Mahieu et al. also looked at this signal in MLS data (as embodied in the GOZCARDS dataset). Please reword accordingly.
added clarification

— Page 2

Line 3: Quote the latitude of Jungfraujoch in the caption.
done

Also, some redundancy, as you say the MLS data is a 100-10hPa column in one sentence and then talk about it being a partial column (without the numbers) later on.

Lines 10-15: Again, please be sure your wording is consistent with the use to which Mahieu et al. put MLS data.

Line 10: "results from simulations using the SLIMCAT model driven by..."
fixed

Line 21: July 2004 doesn't sound like "late 2004" to me.
changed to July

Line 23: "altitude" -> "vertically resolved", given that the vertical coordinate is pressure.
changed to "vertical"

Line 26: "has little change since" -> "shows little change from"
fixed

Line 30: Perhaps put "band 14" in quotes as it's jargon that's not explained earlier (and is presumably covered in the references given earlier in the paragraph).
removed

Lines 32-34: Please clarify, has the N₂O product been "redefined" since the release of v4.2, or was the redefinition part of v4.2 from the outset?
We have replaced the paragraph describing the N₂O product with a new paragraph that more explicitly explains the situation.

Line 34: Unless I've misunderstood, it's part of MLS that has "deteriorated" is it not? Starting at some point during the mission. The way this is worded it sounds like the MLS data files are somehow deteriorating with time (like food going off in the refrigerator) regardless of the time at which the observations were made. Please reword more precisely. —

replaced entire paragraph with what we hope is a clearer discussion

Page 3

Line 1: "next" -> "following" sounds better to me. Line 2: Are the "640 channel" measurements also from the v4.2 dataset or from some earlier version?

As stated above we have rewritten the description to be more explicit about the data products.

Lines 1-6: This would presumably be a good place to have a discussion about the validity of assuming that the pre-2013 drifts are representative of the post-2013 observations. (Or possibly on page 6, see later).

Added a description later in the discussion of Table 1.

Figure 2: The way you've drawn this, with the shaded envelope being narrow at the left hand edge is not an accurate depiction of the manner in which the regression is capturing in the uncertainty in the fit. The way it's shown it implies that the regression is constrained to have a fixed value at $t=t_0$, which is not the case (unless you specifically performed such a fit, which I doubt). I suggest you leave the envelope off to avoid this potential for confusion (I don't see a more accurate but clear way to depict this uncertainty graphically). The caption will need to be updated to match.

Thanks for pointing this out. We have replaced Figures 2 and 4 with new versions showing the shaded area coming to a point through the middle. This better represents the actual meaning of the regression uncertainty.

Line 9: Actually isn't this "mixing ratio" rather than "concentration"? (sorry to be picky)

fixed

Line 10: Actually the dashed line doesn't look that "heavy" to me.

Agreed and fixed

Line 18/19: "...are shown as a percentage deviation..." sounds better to me.

fixed

Line 19: Define "seasonal mean", is it three-monthly averages (DJF, MAM etc.) or monthly averages?

Improved the description to describe that we removed the seasonal cycle, while retaining the mean and have plotted the percent deviation of the residual from the mean.

Line 22: "look at" -> "examine" sounds more scientific to me.

fixed

— Page 4

Line 7: Perhaps "effects" -> "cycles"?

Decided to stay with "effects". We think of the seasonal effect as a cycle. The QBO is an oscillation having an irregular frequency, especially for the last few years.

Lines 23-26: Add "MLS" before "HCl" (line 23) and "N₂O" (line 24) and then delete "from MLS measurements of each constituent."

Changed and added "measurements" after each.

Line 24: add "a" before "deseasonalized"?

fixed

— Page 5

Line 2: "determined by" -> "that due to" Figure 4: As with figure 2, I suggest you remove the "flared" red shading (and update caption accordingly).

Also replaced Figure 4 same change as with Figure 2.

- Page 6 Line 1. The point about the "raw" and "Trend with N₂O fit" being similar at the higher altitudes is a good one and makes geophysical sense to me. However, this then exposes a weakness in the authors' arguments and methods, in that the N₂O drift and surface N₂O trend terms add significantly to the "final" result, moving it far from the "raw" original. If dynamical variability is indeed "relatively small" at these altitudes then why do these modifying terms get the same "weight" at these upper levels as they do lower down where dynamical variability is significant? There seems to be some kind of inconsistency here that needs thought.

The 'trend with N₂O fit' and drift rate only have significant effects on the net trend at pressures 22-68 hPa. At altitudes above 22 hPa, Cly is nearing its maximum stratospheric value and thus becomes insensitive to dynamical variability. Note that at 10 and 15 hPa the 'trend with N₂O fit' is balanced by the surface trend – an indication of the reduced sensitivity to N₂O (dynamics) here.

— Page 6

Lines 5-13: This is the other place where it would be good to talk about the validity of assuming pre- and post-2013 N₂O drifts are consistent.

We have added some clarifying discussion of this problem here and in the conclusions.

Table 1 caption: Suggest that you delete "with 2-sigma uncertainties" on line 18 and instead say at the end of the caption something like: "All uncertainties are quoted at 2-sigma".

done

Lines 25-30: This is where some discussion of age-of-air and related issues would clearly go.

This paragraph has been rewritten to be clearer and we have added a basic reference to age-of-air (Waugh and Hall). The age-of-air issue is extremely minor in this case.

— Page 8

Lines 12-14: Again, this point is seemingly at odds with the "final" results for the higher altitudes.

This statement refers to the effect of using the N₂O time series as a proxy for dynamical variability. The raw trend and corrected trend in columns 1 and 2 of Table 1 are approximately equal. The corrections do lead to a difference in the "final" results.

Line 29: "kkm" typo.

fixed

Lines 34-41: Doesn't the age-of-air spectrum come into this issue too? In any case, it would be best to "show your working" as to how the -4.9% estimate is arrived at here.

Considering the age-of-air spectrum here would be over complicating this simple estimate of the slope of chlorine expected during this time period. We have changed the statement to an estimate of -5%/decade because the original 4.9%/decade overstated the significant figures. Our main point here is that we obtained a reasonable result by applying the N₂O as a proxy.

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

3
4 Richard S. Stolarski
5 Johns Hopkins University

6
7 Anne R. Douglass, Susan E. Strahan
8 NASA Goddard Space Flight Center
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 2σ 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.

Richard Stolarski 3/9/18 3:45 PM

Deleted: ly

23
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.

Richard Stolarski 3/9/18 3:46 PM

Deleted: altitude

Richard Stolarski 3/9/18 3:50 PM

Deleted: that

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.

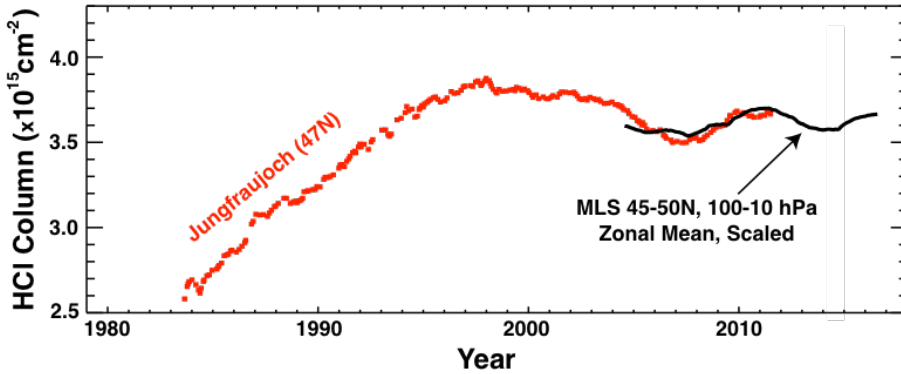
Richard Stolarski 3/9/18 3:44 PM

Deleted: r

Microsoft Office User 3/12/18 2:31 PM

Deleted: then

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).



7
 8
 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**

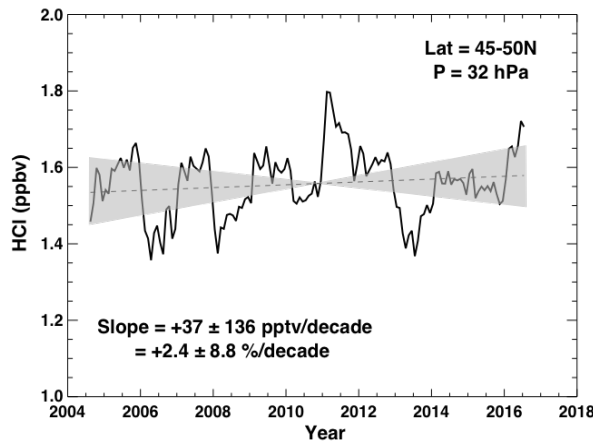
25
 26 | We use the MLS HCl [and N₂O data together](#) to test whether chlorine is decreasing in
 27 the stratosphere as expected from adherence to the provisions of the Montreal
 28 Protocol. MLS was launched on the Aura Satellite in [July, 2004](#) [Waters et al., 2006]
 29 and continues to operate 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.

32
 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

- Richard Stolarski 3/14/18 6:08 PM
Deleted: will
- Richard Stolarski 3/9/18 4:01 PM
Deleted: late
- Richard Stolarski 3/14/18 6:09 PM
Deleted: 7
- Richard Stolarski 3/9/18 4:02 PM
Deleted: altitude
- Richard Stolarski 3/9/18 4:03 PM
Deleted: has little change since

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.



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.

27
 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

Richard Stolarski 3/9/18 1:49 PM

Deleted: is

Richard Stolarski 3/9/18 1:51 PM

Deleted: band 14

Richard Stolarski 3/9/18 2:09 PM

Deleted: For N₂O we use the version 4.2 redefined standard product that uses signals from the band 3 (190-GHz channel); the standard product was redefined because the product from band 12 (640 GHz channel) deteriorated and the band was shut off in August 2013. In the next section we will compare this redefined standard to the measurements from the 640-GHz channel and correct for the drift observed between these two channels during the time that both were yielding N₂O measurements. The 190-GHz N₂O data are stated to be useful in the 68-0.46 hPa range. We will thus restrict our analyses to pressure levels between 68 and 10 hPa in this paper where both the HCl and N₂O measurements are useful.

Richard Stolarski 3/9/18 3:22 PM

Deleted:

Richard Stolarski 3/9/18 5:01 PM

Deleted: heavy

Richard Stolarski 3/13/18 3:13 PM

Deleted: anomalies

Richard Stolarski 3/13/18 3:14 PM

Deleted: in the

Richard Stolarski 3/13/18 3:14 PM

Deleted: monthly-mean measurements

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 ±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.

Microsoft Office User 3/12/18 2:45 PM
Deleted: the

Microsoft Office User 3/12/18 2:45 PM
Deleted: pressure level

Richard Stolarski 3/9/18 8:11 PM
Deleted: in percentage deviation

Richard Stolarski 3/13/18 3:15 PM
Deleted: from the seasonal mean of the entire data set

Richard Stolarski 3/9/18 8:21 PM
Deleted: o look at

Richard Stolarski 3/10/18 10:12 PM
Deleted: anomalies

Richard Stolarski 3/14/18 6:18 PM
Deleted: an interaction between the two effects

Richard Stolarski 3/14/18 6:11 PM
Deleted: anomalies

Richard Stolarski 3/14/18 6:13 PM
Deleted: s, or anomalies,

Richard Stolarski 3/14/18 6:19 PM
Deleted: from MLS measurements

Richard Stolarski 3/14/18 6:14 PM
Deleted: anomaly

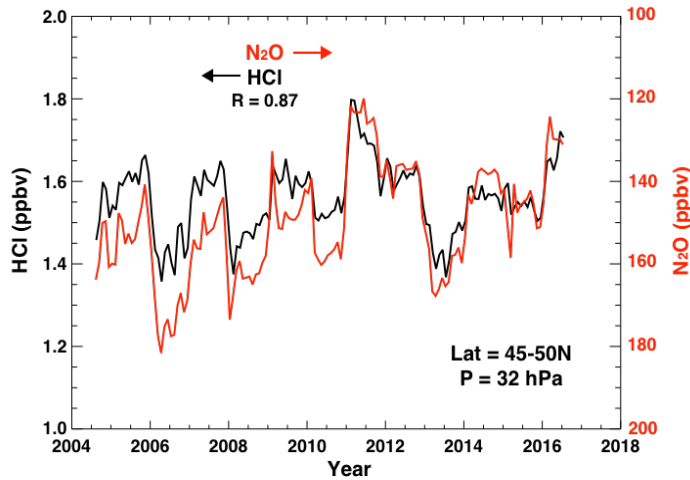


Figure 3: Deseasonalized time series of [MLS HCl measurements](#) (black curve, same as Figure 2) and deseasonalized time series of [MLS N₂O measurements](#) plotted with reverse scale on right side of figure (red curve) for the latitude band 45-50N at 32 hPa pressure level.

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

The trend that we are trying to isolate and confirm for the HCl time series is determined by the change in abundance of chlorine-containing halocarbons driven by the provisions of the Montreal Protocol on ozone-depleting substances.

[Inorganic chlorine in the stratosphere](#) is expected to have decreased since 2000 in response to the decreases in the chlorine-containing source gases. N₂O, on the other hand, is known to be increasing at a rate of about 2.8%/decade [NOAA GMDL data updated from Elkins and Dutton [2009] available at ftp://ftp.cmdl.noaa.gov/hats/n2o/combined/HATS_global_N2O.txt]. Our approach is to use the N₂O time series at each altitude, such as that shown in Figure 3, as an explanatory variable in a time-series regression to remove the dynamical variability from the HCl time series. Trends calculated for HCl in this time-series regression are then corrected for the underlying trend in N₂O.

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

Richard Stolarski 3/9/18 8:32 PM

Deleted: from MLS measurements of each constituent.

Richard Stolarski 3/14/18 6:14 PM

Deleted: Anomalies

Richard Stolarski 3/10/18 10:24 PM

Deleted: The chlorine-containing source gases

Richard Stolarski 3/10/18 10:24 PM

Deleted: are

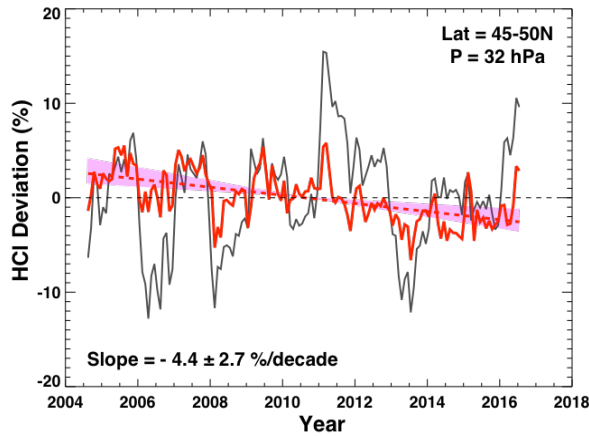


Figure 4: HCl anomaly time series as in Figures 2 and 3 with the mean removed (black) and the residual time series after regression to N₂O time series (red). Red dashed line is linear fit to residual series with 2σ uncertainty bounds indicated by shaded area.

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

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

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

Richard Stolarski 3/9/18 3:35 PM

Deleted:

Richard Stolarski 3/10/18 10:30 PM

Deleted:

Richard Stolarski 3/10/18 10:30 PM

Deleted: as

Richard Stolarski 3/10/18 10:30 PM

Deleted: with

Richard Stolarski 3/10/18 10:31 PM

Deleted: fit to

Microsoft Office User 3/12/18 2:56 PM

Deleted: is

Microsoft Office User 3/12/18 2:56 PM

Deleted: that has a slope of

Richard Stolarski 3/14/18 6:20 PM

Deleted: is relatively small. The

Richard Stolarski 3/14/18 6:22 PM

Deleted: at the lower levels is

Richard Stolarski 3/14/18 7:36 PM

Deleted: N. Livesey

Richard Stolarski 3/14/18 7:35 PM

Deleted: MLS team

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

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

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

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

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

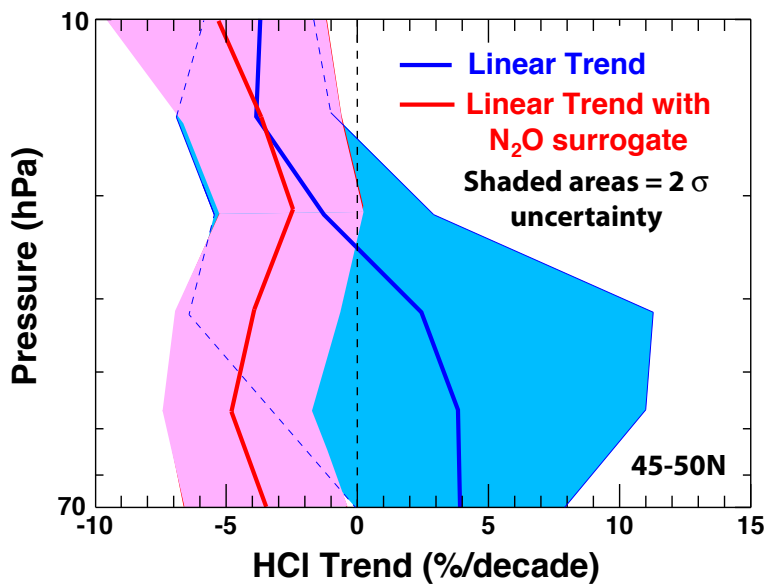
Richard Stolarski 3/14/18 8:03 PM

Deleted: in %/decade with 2σ uncertainties

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
9 and 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.](#)

Microsoft Office User 3/12/18 4:06 PM
Deleted: good



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

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.

Richard Stolarski 3/10/18 10:51 PM

Deleted: showed

Richard Stolarski 3/10/18 10:52 PM

Deleted: trend

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.

Richard Stolarski 3/14/18 7:38 PM

Deleted: the

Richard Stolarski 3/14/18 7:38 PM

Deleted: is less

Richard Stolarski 3/13/18 3:23 PM

Deleted: significant.

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.

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.

Richard Stolarski 3/10/18 10:55 PM

Deleted: 5

Richard Stolarski 3/13/18 3:44 PM

Deleted: k

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.

Richard Stolarski 3/13/18 3:49 PM

Deleted: 4.9

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.

21 | References:

22 |
23 | Brown, A.T., M.P. Chipperfield, C. Boone, C. Wilson, K.A. Walker, and P.F. Bernath,
24 | [2011], Trends in atmospheric halogen containing gases since 2004, *J. Quant.*
25 | *Spectrosc. Radiat. Transfer*, 112 (16), 2552-2566, doi:
26 | 10.1016/j.jqsrt.2011.07.005.

27 | Carpenter, L.J. and S. Reimann [2014], Lead Authors, Scientific Assessment of Ozone
28 | Depletion: 2014, Chapter 1: Update on Ozone-Depleting Substances (ODSs)
29 | and Other Gases of Interest to the Montreal Protocol
30 | Global Ozone Research and Monitoring Project—Report No. 55.

Richard Stolarski 3/10/18 10:55 PM

Deleted: 5

31 |
32 | Elkins, J.W, and G.S. Dutton [2009], Nitrous oxide and sulfur hexafluoride [in 'State
33 | of the Climate in 2008']. *Bull. Amer. Meteor. Soc.*, 90 S38-S39.

34 |
35 | Froidevaux, L., N.J. Livesey, W.G. Read, R.J. Salawitch, J.W. Waters, B. Drouin, I.A.
36 | MacKenzie, H.C. Pumphrey, P. Bernath, C. Boone, R. Nassar, S. Montzka, J.
37 | Elkins, D. Cunnold, and D. Waugh [2006], Temporal decrease in upper
38 | atmospheric chlorine, *Geophys. Res. Lett.*, 33, L23812, doi:
39 | 10.1029/2006GL027600

40 |
41 | Hubert, D., J.C. Lambert, T. Verhoelst, J. Granville, A. Keppens, J.L. Baray, A.E.
42 | Bourassa, U. Cortesi, D.A. Degenstein, L. Froidevaux, S. Godin-Beekmann, K.W.
43 | Hoppel, B.J. Johnson, E. Kyrola, T. Leblanc, G. Lichtenberg, M. Marchand, C.T.
44 | McElroy, D. Murtagh, H. Nakane, T. Portafaix, R. Querel, J.M. Russell, J.
45 | Salvador, H.G. Smit, K. Stebel, W. Steinbrecht, K.B. Strawbridge, R. Stubi, D.P.J.
46 | Swart, P.J. Daan, G. Taha, D.W. Tarasick, A.M. Thompson, J. Urban, J.A.E. van

- 1 | [Gijssels, R. Van Malderen, P. von der Gathen, K.A. Walker, E. Wolfram, and J.M.](#)
2 | [Zawodny. \(2016\). Ground-based assessment of the bias and long-term](#)
3 | [stability of 14 limb and occultation ozone profile data records. *Atmospheric*](#)
4 | [Measurement Techniques](#), 9(6), 2497–2534. [https://doi.org/10.5194/amt-9-](https://doi.org/10.5194/amt-9-2497-2016)
5 | [2497-2016](#).
- 6 |
- 7 | Jones, A., J. Urban, D.P. Murtagh, C. Sanchez, K.A. Walker, N.J. Livesey, L. Froidevaux,
8 | and M.L. Santee [2011], Analysis of HCl and ClO time series in the upper
9 | stratosphere using satellite data sets, *Atmos. Chem. Phys.*, 11 (11), 5321-5333,
10 | doi: 10.5194/acp-11-5321-2011
- 11 |
- 12 |
- 13 | Kohlhepp, R., R. Ruhnke, M.P. Chipperfield, M. De Maziere, J. Notholt, S. Barthlott, R.L.
14 | Batchelor, R.D. Blatherwick, T. Blumenstock, M.T. Coffey, P. Demoulin, H. Fast,
15 | W. Feng, A. Goldman, D.W.T. Griffith, K. Hamann, J.W. Hannigan, F. Hase, N.B.
16 | Jones, A. Kagawa, I. Kaiser, Y. Kasai, O. Kirner, W. Kouker, R. Lindenmaier, E.
17 | Mahieu, R.L. Mittermeier, B. Monge-Sanz, I. Morino, I. Murata, H. Nakajima, M.
18 | Palm, C. Paton-Walsh, U. Raffalski, T. Reddmann, M. Rettinger, C.P. Rinsland, E.
19 | Rozanov, M. Schneider, C. Senten, C. Servais, B.-M. Sinnhuber, D. Smale, K.
20 | Strong, R. Sussmann, J.R. Taylor, G. Vanhaelewyn, T. Warneke, C. Whaley, M.
21 | Wiehle, and S.W. Wood [2012], Observed and simulated time evolution of HCl,
22 | ClONO₂, and HF total column abundances, *Atmos. Chem. Phys.*, 12 (7), 3527-
23 | 3556, doi: 10.5194/acp-12-3527-2012,
- 24 |
- 25 | Livesey, Nathaniel J., William G. Read, Paul A. Wagner, Lucien Froidevaux, Alyn
26 | Lambert, Gloria L. Manney, Luis F. Milla'n Valle, Hugh C. Pumphrey, Michelle
27 | L. Santee, Michael J. Schwartz, Shuhui Wang, Ryan A. Fuller, Robert F. Jarnot,
28 | Brian W. Knosp, Elmain Martinez [2017], Jet Propulsion Laboratory Report
29 | JPL D-33509 Rev. B, "Earth Observing System (EOS) Aura Microwave Limb
30 | Sounder (MLS) Version 4.2x-3.0: Level 2 data quality and description
31 | document".
- 32 |
- 33 | Mahieu, E., M.P. Chipperfield, J. Notholt, T. Reddmann, J. Anderson, P.F. Bernath, T.
34 | Blumenstock, M.T. Coffey, S.S. Dhomse, W. Feng, B. Franco, L. Froidevaux, D.W.
35 | T. Griffith, J.W. Hannigan, F. Hase, R. Hossaini, N.B. Jones, I. Morino, I. Murata,
36 | H. Nakajima, M. Palm, C. Paton-Walsh, J.M. Russell III, M. Schneider, C. Servais,
37 | D. Smale, and K.A. Walker [2014], Recent Northern Hemisphere stratospheric
38 | HCl increase due to atmospheric circulation changes, *Nature*, 515, 104-107,
39 | doi=10.1038/nature13857.
- 40 |
- 41 | Nassar, R., P. F. Bernath, C. D. Boone, C. Clerbaux, P. F. Coheur, G. Dufour, L.
42 | Froidevaux, E. Mahieu, J. C. McConnell, S. D. McLeod, D. P. Murtagh,
43 | C. P. Rinsland, K. Semeniuk, R. Skelton, K. A. Walker, and R. Zander [2006], A
44 | global inventory of stratospheric chlorine in 2004, *J. Geophys. Res.* 111,
45 | D22312, doi:10.1029/2006JD007073.
- 46 |

1 Rinsland, C. P., et al. [2003], Long-term trends of inorganic chlorine from ground-
2 based infrared solar spectra: Past increases and evidence for stabilization, J.
3 Geophys. Res., 108 (D8), 4252, doi:10.1029/2002JD003001, 2003.
4
5 Strahan, S. E., L. D. Oman, A. R. Douglass, and L. Coy (2015), Modulation of Antarctic
6 vortex composition by the quasi-biennial oscillation, Geophys. Res. Lett., 42,
7 doi:10.1002/2015GL063759.
8
9 Waters, J.W., L. Froidevaux, R.S. Harwood, R.F. Jarnot, H.M. Pickett, W.G. Read, P.H.
10 Siegel, R.E. Cofield, M.J. Filipiak, D.A. Flower, J.R. Holden, G.K. Lau, N.J. Livesey,
11 G.L. Manney, H.C. Pumphrey, M.L. Santee, D.L. Wu, D.T. Cuddy, R.R. Lay, M.S.
12 Loo, V.S. Perun, M.J. Schwartz, P.C. Stek, R.P. Thurstans, M.A. Boyles, S.
13 Chandra, M.C. Chavez, G-S. Chen, B.V. Chudasama, R. Dodge, R.A. Fuller, M.A.
14 Girard, J.H. Jiang, Y. Jiang, B.W. Knosp, R.C. LaBelle, J.C. Lam, K.A. Lee, D. Miller,
15 J.E. Oswald, N.C. Patel, D.M. Pukala, O. Quintero, D.M. Scaff, W.V. Snyder, M.C.
16 Tope, P.A. Wagner, and M.J. Walch (2006), The Earth Observing System
17 Microwave Limb Sounder (EOS MLS) on the Aura satellite, IEEE Trans. Geosci.
18 Remote Sensing 44, no. 5, doi:10.1109/TGRS.2006.873771.
19
20 [Waugh, D. and T. Hall \(2002\), Age of stratospheric air: Theory, observations, and](#)
21 [models, Rev. Geophys. 40, 1-26, doi: 10.1029/2000RG000101](#)
22
23
24 [Weatherhead, E.C., G.C. Reinsel, G.C. Tiao, X-L. Meng, D. Choi, W-K. Cheang, T. Keller, J.](#)
25 [DeLuisi, D.J. Wuebbles, J.B. Kerr, A.J. Miller, S.J. Oltmans, and J.E. Frederick](#)
26 [\(1998\), Factors affecting the detection of trends: Statistical considerations](#)
27 [and applications to environmental data, J. Geophys. Res. 103, 17149-17161.](#)
28
29 Zander, R., M.R. Gunson, C.B. Farmer, C.P. Rinsland, F.W. Irion, and E. Mahieu [1992],
30 The 1985 chlorine and fluorine inventories in the stratosphere based on
31 ATMOS observations at 30-degrees north latitude, J. Atmos. Chem. 15, 171-
32 186.
33
34
35
36