We have responded to all of the comments provided by both reviewers, which were very helpful in clarifying the manuscript. Our direct responses are indicated in red below. A manuscript with indicated changes has also been submitted.

Reviewer 1

ClO is a tricky molecule to deal with due to its strong diurnal and temperature dependent properties. The authors have done an excellent job in being able to look at Antarctic stratospheric ClO from the ground-based platform (ChlOE) at Scott-Base as well as using satellite data (MLS missions). Analyses using temperature data from MERRA were also performed to account for the relationship between stratospheric ClO and temperature around Scott-Base. As a result of an excellent intercomparison analysis, trends around the Scott-Base region were estimated which were found to be consistent with previous studies as well as providing up to date trends for the Antarctic region.
I recommend this paper for publication after the following revisions are made.
Thank you for the kind words and the recommendation. With very few exceptions we agree that all of the suggested changes improve the manuscript.

MAJOR

P3,L6: There are is one reference to the ClhOE instrument providing some more detailed information about its specifications and general performance. As it is the one of the main instruments focused upon here in this paper, there should be something written more that helps give the reader an idea of how well it performs. How does it validate to other measurements for example? On P2,L20 it is stated that Connor et al., compared MLS to ClhOE, maybe it would be good to provide a more detailed description in one or two lines of the results here in this paper.
We have added some more details about the history of ChlOE measurements in the previous paragraph, and have added from the Connor et al. (2007) the result that “comparisons of measurements taken within ±30 minutes of the MLS ascending orbit overpass showed agreement of 11±8% in the peak mixing ratios”.

P3, L14. It is stated ***“For these measurements we have defined day as the period from 3 hours after sunrise to 1 hour before sunset, and night as the period from 4 hours after sunset to 1 hour before sunrise.”***
Is there any particular reason as to why you chose these values? Do the results change significantly if using a larger/smaller range? The text needs some explanation as to why these values were chosen.
The values were empirically determined based on avoiding periods of rapid change in ClO. Explanatory text has been added.

Figure 2. I like this plot, but I think there needs to be more explanation for the ClO behavior over time. As I understand it, the small difference between day and night values at the beginning and end of the time period (at both peaks) are because of low ClO activation on a diurnal scale. Is this because there is no sunlight at the beginning of the period, whilst at the end of the period, there is only daylight? I think this needs to be explained a little clearer.
We have added some text here regarding the seasonal evolution of the partitioning between ClO, HCl, and $ClONO_2$. We have also clarified that this Figure would look similar if we showed MLS daytime measurements only.
Furthermore, I suggest adding a geometric altitude on the y-axis to stay consistent with Figure 1.
done

P5, L23: You mention you use a climatology in order to calculate anomaly ClO values. How is this climatology calculated? Do you use a simple mean, median, weighted mean, such as using the uncertainties of the measurements? Did you use all measurements or did you deal with outliers?, if so, how? A little more information is needed here to explain how the climatology was made.

We have clarified that the climatology is based on simple means with a 5-day smoothing. We do point out that ChlOE measurements are missing on some days due to poor tropospheric weather.

P7, L20: "**At the latitude of Scott Base (77.85ºS) we do not expect that during these dates any of the measurements have occurred outside of the vortex (except possibly in 2002, when the Antarctic stratosphere exhibited an unusual major warming)**".

Is there any way to actually confirm this? There are various tools that can be used to check where the vortex edge is estimated to be situated. One particular tool is Scaled Potential Vorticity (SPV) that has been provided for the MLS data using the GEOS-5 model (Manney et al., 2009). http://www.atmos-chem-phys.net/9/4775/2009/acp-9-4775-2009.pdf SPV values are provided for each MLS measurement. As MLS is used here in this paper, you should be able to get a reasonable idea as to what air mass Scott Base was situated in for these measurement periods. If you have possible access to this data, I would suggest trying to confirm this.

Unfortunately there were no MLS measurements in 2002, so the SPV provided for the MLS data does not help us. In other years Scott Base seems to be well within the vortex, but given that there are different vortex-edge definitions we are hesitant to make a definitive statement.

Figure 7: There is a good anti-correlation between T and ozone loss, which is seen clearly at most of the maxima and minima here. However, 2004 and 2005 there seems to be a lag between the two, is there any explanation for this?

We are certainly not aware of any physical mechanism which would provide for a lag between the two years.

P15, L12, I think one has to be a little careful concerning the turn-around of 1997. Most previous studies use this year, but it has never really been proven to be the actual turn-around, but best estimate (as far as I'm aware) for the upper stratosphere mid latitudes. I think it would be better to write 'about 1997' or 'around 1997'.

We certainly do not wish to indicate anything definitive about 1997. 'Around 1997' is certainly a better way to phrase this.

MINOR

P1, L17 : I would suggest entering the range of years analyzed here as well stating austral spring. .. "We present 20 years (1996-2015) of austral springtime..". Otherwise, the comments further down about August and September seem somewhat general and could be from any 20 year period. done

P1, L22: comma needed. "In order to study inter-annual differences, we focus.." done

P1,L22: comma needed. "By making better use of this relationship, we can …" done

P2, L14: Any reference that could be sited about the future installation at Mauna Loa?

This has been changed to indicate that we now have an instrument operating from Mauna Loa. Since we have just started taking measurement we have nothing to reference.

P3, L31: I suggest adding a temperature range/or minimum value for the term 'very cold Antarctic conditions' to which PSCs are formed. According to the WMO, PSCs form below 195 K. done

P4, L2: Comma needed. "because of the weak signal, the best ground-based.." done

P4, L3: Commas needed. "Measurements of this ClO peak, as made from the ChlOE3 done instrument at Mauna Kea, have been shown in Nedoluha et al. (2011) and Connor et al. (2013)."

P4, L5. Comma needed. "Since all of the measurements shown here 5 will be with the ChlOE1 instrument, we will henceforth refer to this instrument simply as ChlOE". done

Figure 1. Caption could read better as; “The retrieved day minus night ClO mixing ratio profile for September 4, 2011 (solid line), and the a priori profile for that day (dashed line), as a function of pressure (left y-axis) and geometric altitude (right y-axis)”. done

P5, L1: Comma needed. “Here, we use the..” done

P5, L8: I would suggest to use pressure as an altitude reference and put the geometric altitude in brackets as a reference. This then stay consistent with the paper up to this point. Hence; whatever pressure is associated with 23 km. done

P5, L15: I suggest rewording to; “Figure 3 shows measurements of day minus night ClO column from ChlOE during 2006 together with those from the coincident MLS (within ±2° latitude and ±15° longitude of Scott Base) measurements.” done

P7, L9: Spelling; “solar times of its measurements..” done

P7, L20: Comma needed; “At the latitude of Scott Base (77.85°S), we do not expect that” done

P8, L2: suggest adjusting to; “We therefore choose September 17 as the final day of the period for which we will compare interannual variations as it provides the best representation of the atmospheric state as a result of a lower variance”.

The previous sentence already notes the increase in variance that occurs if we extend to Sept. 22, so it seems repetitive to say this again.

P8, L5: Suggest adjusting to; “The choice of 5 August 28 as the first day for the comparisons provides us a 3-week period yielding an average of 16.4 daily measurements from ChlOE for each year.” done

P9, L3: Are these really anomalies in Fig. 5? Would that not imply fluctuation around zero? I thought you used the anomalies and added them back onto the climatology, hence yielding ”the annual average ClO column”?

The word “anomaly” has been using incorrectly in several places in the manuscript. As the reviewer suggests, we have replaced this with “annual average ClO column”, or some similarly appropriate phrase.

P9, L13: Comma needed; “For the 12 years of Aura MLS ClO column measurements, the correlation..” done

P9, L22: Commas needed; “The fraction of Cly, which is in the form of ClO, is sensitive..”

We changed “which” to “that” without adding commas.

P9, L23: Also referring to P3, L31, you state “very low temperatures”…A minimum value should be included. done

P9, L24: CALIPSO needed to be written in full before abbreviating “(Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations” done

P10, L2: I would suggest trying to quantify the amount of correlation. How much is “are quite well correlated..”. This provides the reader with his/her own interpretation of the relationship between the two variables.

Since this refers to a number of individual cases, it is difficult to quantify. We changed the wording to read “coincide with”.

P11, L6: Better suited to read "The annual average MERRA temperature anomalies at three pressure levels…" done

P11, L13: What does TIROS stand for? Television InfraRed Operational Sounder? Please write the full name first of the measuring instrument, before abbreviating.

We now provide the full name, Television Infrared Observation Satellite, in the text.

P11, L19: comma needed; "Based on the biases in MERRA temperatures indicated by the sonde data, we added 4.0K to the…" done

P11, L19-L20: I would reword this sentence slightly to confirm that the extra K are added to account for biases. "Due to the biases in MERRA temperatures indicated by the sonde data, we added 4.0K to the 20 hPa 1996-1998 MERRA temperatures and 2.1K to the 30 hPa temperatures to account for this offset". done

P12, L2: comma needed: "When we estimate chlorine trends in Section 5, it will be particularly important to have…" done

P12, L24: Please quantify the correlation value between MLS ClO Z.A. column and zonally average temperature; "showed the highest correlation" done

P15, L26: So "The linear trend in the annual August 28 to September 7 ChlOE ClO anomalies (those shown in Figure 5) is -1.1±0.4%/yr" is referring to years 1996 to 2015? Could you please just make that clear. It might be important to the reader as you state trend periods for previous analyses in your literature review.

Yes, this is a good point. We have added the years.

P16, L25: Please add 'respectively' "The calculated trends with REAN2 temperatures are -0.6±0.2% yr^{-1}, -1.4±0.9% yr^{-1}, and -0.5±0.4% yr^{-1} for zonal MLS, Scott Base 25 MLS, and ChlOE, respectively". done

References: Need some work to make them all consistent with ACP standards, as well as being in chronological order if under the same leading author.

Reviewer 2

This paper uses long-term observations from a ground-based instrument and satellite to study trends in ClO in the Antarctic lower stratosphere. The ClO observations are adjusted for temperature effects and trends in inorganic chlorine are derived. Overall, I think that the authors have a powerful dataset but more work is needed before the paper would be acceptable for ACP (i.e. major revisions). I think that they can address these points and a useful paper will result. My comments are summarized below.

The reviewer comments are primarily aimed at improving the presentation to clarify the goals and to provide some relevant background. We hope that our response to the comments adequately addresses these weaknesses in the original manuscript.

Major comments.

1. The aim of the study needs to be clarified. Polar ozone loss is a 'mature' topic and some simple qualitative results are not an advance. It seems that the aim is to use the ClO to derive underlying Cly trends. The motivation for this needs to be made up front.

The introduction has been extended to emphasize that we are studying trends. We would also like to point out that no results from this ground-based dataset have been published for many years, so in part the motivation really is simply to make the community aware of these unique long-term measurements.

Why can't we just observe Cly?

We now state explicitly that ClO has emission lines at microwave frequencies.

Why do we need to know what Cly is doing? Is there any implication for detection of polar ozone recovery?

Text has been added to the introduction to address these questions.

Would you expect the same Cly trend in different latitudes (which the abstract implies)?

Yes, we would expect similar trends in Cly at different latitudes, however it was not obvious how closely the magnitude would agree.

2. Effect of temperature on ClO. Temperature could affect ClO levels both by changing the extent of chlorine activation (conversion of HCl or ClONO2) or by changing the partitioning of ClOx. There is also the impact of short-term dynamics (vortex movement).

The abstract says that ClO is anti-correlated with T both on a daily and interannual timescale. You should explain the mechanisms for this. Specifically, I thought about how T changes might change ClOx partitioning, but T increases would increase ClO and decrease Cl2O2. Daytime ClOx is mostly ClO but there could be a small effect. In any case, it is up the authors to explain (and quantify) how T might affect ClOx so that the temperature correction can be seen to be robust.

We have added text, primarily in the introduction, discussing the importance of PSCs and in turn their dependence on temperature. While there are certainly other chemical mechanisms involved, we think that it is the link between PSCs and temperature which is the dominant driver of the anti-correlation between ClO and temperature. However, we have not run any model calculations and are therefore not in a position to present any detailed mechanism. The results shown in the manuscript rely completely on the phenomenological relationship which we calculate from the measurements.

We also reran the entire analysis chain using daytime MLS measurements only, and found that the results were very similar (agreement to within1σ). We added this result to the text near Figure 2. We also note that if we use nighttime measurements only a very week correlation with temperature (-0.22), and a statistically insignificant slope when comparing to ClO column and temperature as in Figure 8.

3. What about other atmospheric changes contributing to ClO trends? There is a paper by Solomon et al (literally just published) which argues for changes aerosol loading affecting polar ozone loss (and presumably ClO). How big an effect is that?

As Solomon et al. (2016) show, chemistry and dynamic dominates the variations, and it is the dynamical component which we have tried to remove with our temperature correction. They do show that aerosols from volcanoes have some effect on ozone loss, but this is well below our level of sensitivity. Solomon et al. suggest that volcanic aerosols caused a reduction in the "healing trend" of ~10% in ozone, but the error bars associated with our calculated Cly trends are well above 10% of the trend values.

4. Anti-correlation of ClO and T. Although this is expected qualitatively, I found it interesting how linear this is, especially for the larger scale MLS data. In fact, the scientific interest is not that it occurs but how strong this anti-correlation is. I think that this is what the abstract should emphasise.

We have added text to the abstract to emphasize the strong correlation and the calculation of a linear fit.

Other specific comments

Title: The current title does not give any indication of the scientific message of the paper. This should be modified to indicate what the 20 years of ClO data are used for. . .

We have added an emphasis on the implications of these measurements in the paper, but we have not changed the title.

Abstract: Lines 15-16. Give the dates covered by the data. done

Abstract. Line 29. Define Cly. done

Page 2. Line 6. What is the most important cycle? I know it is ClO + ClO in the Antarctic, but ClO is also involved in ClO + BrO, which is number two. So this is not clear.

We now specify the chemical cycle

Page 2. Section 1. I think this introduction needs a paragraph on the processes involved in polar ozone depletion where you can explain the role of temperature, PSCs, HCl as a reservoir etc. At present little bits of this information is used bit-by-bit in the results and overall it will be confusing to a non-expert.

We have added several sentences to the introduction, and have split the first paragraph into two, so that the first paragraph discusses the processes involved in polar ozone depletion, and the second provides information on ClO measurements.

Page 3. Line 8. Need to define ClOx. The normal definition is Cl + ClO +2Cl2O2, in which case ClO -> Cl2O2 is a repartitioning within ClOx, not a conversion from ClOx.

Yes, this was incorrectly expressed. It now reads: "all ClO … converts to Cl2O2".

Page 3. Line 15. Give altitude at which this SZA is sunset. done

Page 3. Line 31. Example of information which needs to be in an introductory paragraph on polar processes. Now in both places.

Page 4. Line 4. Where is Mauna Kea? I do know but this is another example of where the authors have not thought about the non-expert readers. done

Page 4. Line 5. Change 'will be' to 'are' – that tense fits the paper better. done

Page 4. Figure 1. Use (a), (b), (c) for the panels. The way the panels are laid out, it is not clear if there are two or three. done

Page 5. Line 24. Does the comparison change if you subsample the ChlOE data to match the MLS time period?

No, it makes almost no difference. We have now indicated this in the text.

Page 6. Figure 3. There are no error bars or estimate of uncertainty in the figure. What could be added to inform the comparison of the different datasets?

We provide here no error analysis of individual measurements. However, we do make use of the day-to-day scatter for each year (shown in this figure for 2006) to estimate (later in the paper) the uncertainties for the annual averages which are the main focus of this paper.

Page 6. Figure 3 caption. You need to say where this plot is for! I.e.the Scott Base station (with latitude details etc). Say that MLS is sampled at station. done

Page 7. Line 8. What does 'were consistent' mean quantitatively? done

Page 7. Line 15. This is a long way into the paper to state what the primary goal is! The introduction should state this (and it should be reflected in the title and abstract content).

This sentence has been moved to the introduction and a different sentence now introduces this Section.

Page 9. Figure 5 caption. This says both 'annual' and 'each year'. It is not an annual average.

The caption here, and in other places, has been rewritten to say "climatology plus annual average anomally".

Page 9.Line 10 onwards. It would be helpful to give the correlation coefficients on the plot (with a legend for the lines).

The correlation coefficients are now indicated on the plot.

Page 9. Line 17. Another example of background polar chemistry that should be stated earlier. . .

We have added some of this to the now much extended introduction.

Page 9. Line19. ‘3D single layer’? I think that SLIMCAT is a 3D model and that SLIMCAT is just a name, not an acronym. In any case, this paragraph does not say anything. What did this studies show which is relevant here?

This truly unnecessary paragraph has been eliminated.

Page 11. Figure 6 caption. Met. reanalyses should not be classed as ‘measurements’.

done

Page 12. Line 7. ‘or more properly’ – just say what it really is. Choose one way of saying it.

This awkward wording was introduced because correlations and anti-correlations are being compared. We now write “magnitude of the anti-correlation”.

Page 14. Line 8. Why not reduce power of 10 and remove some decimal places?

This notation is used because the measured columns themselves are given in units of 10^{15} cm^{-2}/K.

Page 17. Line 10. This is a Summary or Conclusions. It is the final section and it does not add any more discussion. The lack of a conclusions section gives the impression that in this draft the authors were clear about their main scientific message.

We agree with the reviewer, and the title of this Section has been changed to Summary.

20 Years of ClO Measurements in the Antarctic Lower Stratosphere

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Abstract. We present 20 years (1996-2015) of austral springtime measurements of chlorine monoxide (ClO) over Antarctica from the Chlorine Oxide Experiment (ChlOE1) ground-based millimeter wave spectrometer at Scott Base, Antarctica, as well 12 years (2004-2015) of ClO measurements from the Aura Microwave Limb Sounder (MLS). From August onwards we observe a strong increase in lower stratospheric ClO, with a peak column amount usually occurring in early September. From mid-September onwards we observe a strong decrease in ClO. In order to study interannual differences, we focus on a 3-week period from August 28 to September 17 for each year, and compare the average column ClO anomalies. These column ClO anomalies are shown to be highly correlated with the average ozone mass deficit for September and October of each year. We also show that anomalies in column ClO are strongly anti-correlated with 30 hPa temperature anomalies, both on a daily and an interannual timescale. Making use of this anti-correlation we calculate the linear dependence of the interannual variations in column ClO on interannual variations in temperature. By making use of this relationship, we can better estimate the underlying trend in the total chlorine (Cl_y = HCl + $ClONO_2$ + HOCl + 2 × Cl_2 + 2×Cl_2O_2 + ClO + Cl). The resultant trends in Cl_y, which determine the long-term trend in ClO, are estimated to be -0.5±0.2% yr^{-1}, -1.4±0.9% yr^{-1}, and -0.6±0.4%

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yr^{-1}, for zonal MLS, Scott Base MLS (both 2004-2015), and ChlOE (1996-2015) respectively. These trends are within 1σ of trends in stratospheric Cl_y previously found at other latitudes. The decrease in ClO is consistent with the trend expected from regulations enacted under the Montreal Protocol.

1. Introduction

Chlorine monoxide (ClO) is central to the formation of the Antarctic ozone hole. It is both the direct product of the reaction between chlorine (Cl) and ozone (O_3) and the catalytic agent in the most important ozone-depleting chemical cycle ($Cl + O_3 \rightarrow ClO + O_2$; $ClO + O \rightarrow Cl + O_2$;Waters et al., 1993, Salawitch et al., 1993). Understanding trends in ClO is therefore critical to our understanding of polar ozone recovery. The Antarctic spring is unusual in that, in the lower stratosphere, most of the available total chlorine (Cl_y) is present in its reactive forms (ClO_x= ClO + $2xCl_2O_2$). The amount of ClO in the Antarctic vortex is dependent upon both the available Cl_y and on the prevalence of polar stratospheric clouds (PSCs), which provide the surfaces for heterogeneous processes that convert unreactive chlorine species into ClO_x. While Cl_y will vary from year-to-year the due to dynamical effects (Strahan et al., 2014), it will vary much more slowly than ClO, which, because of its sensitivity to the prevalence of PSCs, is very sensitive to interannual variations in temperature. The primary goal of this study is to estimate the trend in Cl_y in the Antarctic lower stratosphere, during the annual formation of the ozone hole, over the period 1996 to 2015.

The ClO molecule has emission lines at microwave frequencies, and the first ground-based measurements of stratospheric ClO were made using a microwave radiometer in 1980 (Parrish et al., 1981). High concentrations of ClO in the lower stratosphere over Antarctica were first measured using this technique in 1986 (de Zafra et al., 1987; Solomon et al., 1987). The Chlorine Oxide Experiment (ChlOE1) ground-based millimeter wave spectrometer was permanently deployed at Scott Base, Antarctica (77.85° S, 166.77° E), by Stony Brook University and the National Institute of Water and Atmospheric Research (NIWA) in February 1996. Details of the measurement technique, data analysis, and error analysis were presented in Solomon et al. (2000). ChlOE1 is currently jointly operated by the Naval Research Laboratory (NRL) and NIWA. Both this instrument and the ChlOE3 instrument, which operated at Mauna

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Kea, Hawaii, until 2015, are part of the Network for the Detection of Atmospheric Composition Change (NDACC). A new ChlOE4 instrument has now been deployed at Mauna Loa, Hawaii.

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In this paper, we describe the measurement technique and present results from the ChlOE1 time series from 1996-2015. Measurements are only shown from mid-August to mid-October, when ClO daytime mixing ratios can reach up to ~2 ppbv. We show, from 2004 onwards, ClO measurements from the Aura Microwave Limb Sounder (MLS), both coincident with Scott Base and zonally averaged at the latitude of Scott Base. The ChlOE1 measurements were previously compared with the v1.5 MLS retrievals for the austral spring of 2005 (Connor et al., 2007). Comparisons of measurements taken within ±30 minutes of the MLS ascending orbit overpass showed agreement of 11±8% in the peak mixing ratios.

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We also show annual anomalies for measurements during the 3 weeks when the stratospheric ClO column densities generally reach their maximum values, and compare these anomalies with interannual anomalies in temperature, as provided by the Modern Era Reanalysis for Research and Applications (MERRA) (Rienecker et al., 2011). We use the 20 years of ChlOE measurements and 12 years of MLS measurements to derive a relationship between the interannual anomalies in ClO column and those in 30 hPa temperature. We then make use of this relationship to derive an estimate of Cl_y trends in the Antarctic vortex.

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2. ClO Measurements

The ChlOE ground-based radiometer measures the thermally excited rotational emission lines near 278.63 GHz. The spectrometer bandwidth permits measurement of the pressure-broadened lineshape from which ClO altitude profiles are retrieved. The instrument is a cryogenically cooled (~20 K) heterodyne receiver, tuned to observe the ClO transition by adjustment of a phase-locked local oscillator. It is coupled to a spectrometer with 506 MHz total bandwidth, which is approximately the width of the ClO line at 15 km altitude.

At night, the ClO emission is much weaker and narrower, because nearly all ClO in the lower stratosphere rapidly converts to chlorine peroxide (Cl_2O_2) after sunset (Solomon et al., 2002). This allows us, in the ground-based measurements, to remove the instrumental baseline and a small number of interfering atmospheric spectral lines in the instrument bandpass (primarily the ozone line at 278.521 GHz) by subtracting the nighttime spectrum from the daytime one. For these measurements we have defined day as the period from 3 hours after

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sunrise to 1 hour before sunset, and night as the period from 4 hours after sunset to 1 hour before sunrise. Sunrise and sunset are defined to occur when the solar zenith angle is 94.5° at the surface (equivalent to 90° near ~20km altitude). These day and night variations were chosen empirically, in an attempt to estimate daytime and nighttime equilibrium concentrations of ClO, by excluding times near sunrise and sunset when we observed rapid diurnal changes in ClO. If more observations are included in either daytime or nighttime integrations, these periods of rapid change are partially included, significantly changing the mean values. If fewer observations are included, the integrated spectra are noisier and the mean ClO is correspondingly more uncertain.

A retrieval of the ‘‘day minus night’’ spectrum, and thus of the day ClO mixing ratio less the night mixing ratio, is performed by a three-stage process, described in detail by Solomon et al. (2000). The first stage determines the altitude of the peak of the lower stratospheric distribution as a function of date, by performing retrievals on a full season of data, using an a priori profile without a separate lower stratospheric component. In the second stage, the a priori ClO distribution consists of a climatological profile having a peak in the lower stratosphere determined by stage 1 at ~30 hPa (22 km) in mid-August to ~48 hPa (19 km) by late September, with a secondary peak in the upper stratosphere at ~5 hPa. The second-stage retrieval is simply a nonlinear least-squares-fit of a single multiplier applied to the lower stratospheric distribution. The climatological distribution, modified by the retrieved multiplier, is used as the a priori distribution for the third stage, which is a maximum a posteriori solution as given by Rodgers (2000, e.g., equation (4.5)). Figure 1 shows a ChlOE1 day minus night retrieval for September 4, 2011. The retrieved and a priori profiles both have two mixing ratio peaks, one peak in the upper stratosphere and a much larger peak in the lower stratosphere. The lower stratospheric peak is only present when inactive chlorine is converted to active chlorine on the surface of PSCs, and is therefore only observed under the extremely cold Arctic and Antarctic conditions where PSC formation is possible (temperatures below ~195K). The upper stratospheric ClO peak can be observed at any location, but because of the weak signal, the best ground-based measurements require extended integrations (~1 week) from a high-altitude site. Measurements of this ClO peak, as made from the ChlOE3 instrument at Mauna Kea, have been shown in Nedoluha et al. (2011) and Connor et al. (2013). Since all of the measurements shown here are from the ChlOE1 instrument, we will henceforth refer to this instrument simply as ChlOE.

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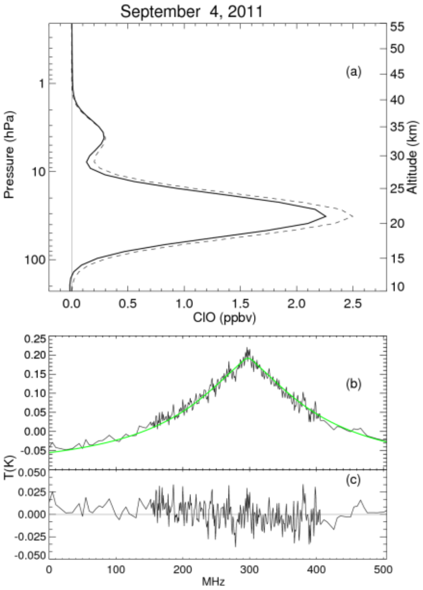
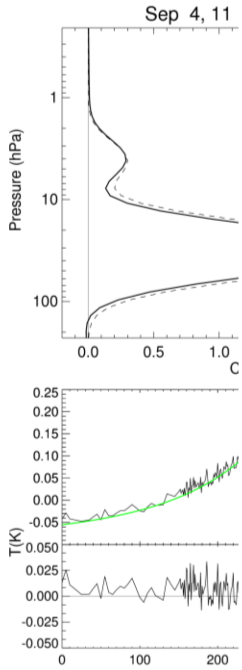


Figure 1 –a: The retrieved day minus night ClO mixing ratio profile for September 4, 2011 (solid line), and the a priori profile for that day (dashed line), as a function of pressure (left y-axis) and geometric altitude (right y-axis) . b: The measured (black line) and modeled (green line) spectra. c: The measured minus modelled residual spectrum.

As is clear in Figure 1, and has previously been shown by ground-based microwave (de Zafra et al., 1987; Solomon et al., 1987, 2000) and satellite measurements (e.g., Waters et al., 1993, Santee et al. 2005), ClO in the Antarctic spring is overwhelmingly concentrated in the lower stratosphere. We shall, throughout this study, make use of the column ClO at altitudes above 100 hPa. Any variations in this ClO column during the polar PSC season are dominated by changes in the lower stratospheric peak of ClO.

Aura MLS measurements of ClO are available since 2004. The version 2.2 ClO measurements were validated in Santee et al. (2008). Here, we use the v4.2 retrievals (Livesey et al., 2015). The Aura measurement overpasses near the latitude of Scott Base occur at ~1630 and ~2300 LST. The times for these measurements remain consistent within ±4 minutes throughout the entire Aura mission. Although it is not possible to replicate the ChlOE diurnal sampling with the twice daily MLS overpass sampling, we shall nevertheless in this study show exclusively MLS daytime (~1630 LST) minus nighttime (~2300 LST) measurements. We note that at the



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78°S latitude of Scott Base the sun actually sets before 1630 at 23 km (i.e., near the ClO peak) until August 24.

The typical seasonal evolution of ClO, as measured by MLS, is shown in Figure 2. This figure shows zonal average day minus night MLS measurements within ±2° latitude of Scott Base for 2006. As in Figure 1 both the upper and lower stratospheric peaks in ClO are apparent. The ClO begins to increase in the sunlit portions of the vortex in late May/early June as the reservoir gases, hydrogen chlorine (HCl) and chlorine nitrate ($ClONO_2$), are converted to ClO. The lower-altitude ClO continues to show a gradual increase until mid-September, and then experiences a sharp decline, as it converts back to the reservoir gases. Santee et al. (2008) present a detailed study of the seasonal evolution of the partitioning between ClO, HCl (from MLS), and $ClONO_2$ (from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer; ACE-FTS) in the Arctic and Antarctic.

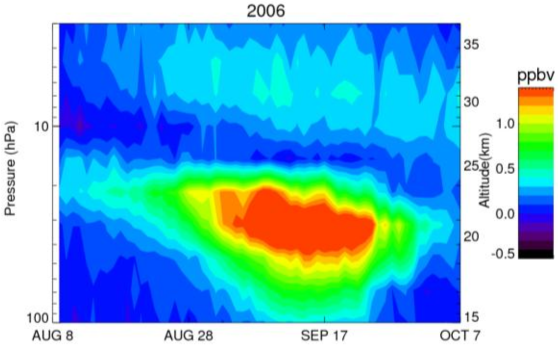

Figure 2 – The zonally averaged daily ClO mixing ratio (day minus night) as measured by Aura MLS for 2006.

Figure 3 shows measurements of day minus night ClO column from ChlOE during 2006 together with those from the coincident (within ±2° latitude and ±15° longitude of Scott Base) MLS measurements. ChlOE measurements are missing for some days because poor tropospheric weather made it impossible to obtain both the daytime and nighttime spectra required for the retrieval, but the general temporal development is clear in both the MLS and ChlOE datasets. Essentially there is an increase in August, a maximum in mid-September, and then a rapid decrease at the end of winter.

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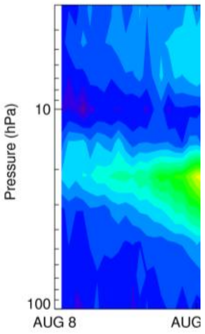

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In addition to the daily measurements for 2006, we also plot the ChlOE and MLS climatologies for these datasets. The ChlOE climatology is calculated from the daily average of all measurements taken from 1996-2015, while the MLS climatology is derived from measurements taken from 2004-2015. Calculating the ChlOE climatology using only measurements from 2004-2015 makes very little difference in the analysis. In both cases a 5-day smoothing has been applied. Both datasets show values of the 2006 ClO column that, at least until mid-September, are generally higher than their climatologies. Santee et al. (2011) previously noted that the 2006 Antarctic winter showed strong and prolonged chlorine activation in the lowermost vortex, and postulated that this was the cause of unusually low column ozone that year.

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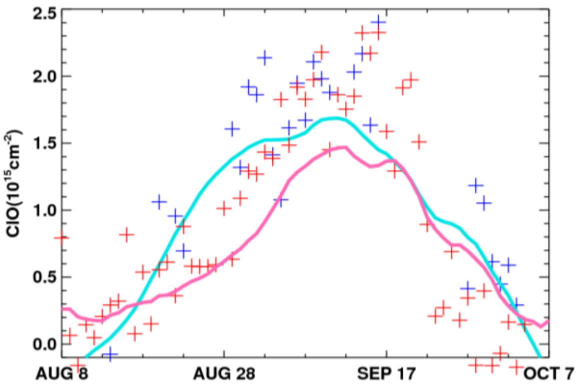

Figure 3 – Daily (day minus night) column density of ClO measurements at altitudes above 100 hPa for mid-August to mid-October 2006 from ChlOE measurements at Scott Base (blue crosses) and from MLS measurements within ±2° latitude and ±15° longitude of Scott Base (red crosses). Also shown are climatologies for this period based on the ChlOE measurements from 1996-2015 (light blue line) and MLS measurements from 2004-2015 (pink line).

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Figure 3 shows a clear difference in the seasonal development of the day minus night MLS and ChlOE climatologies. Since the MLS measurements in the vicinity of Scott Base only begin to see sunlit air near the altitude of the ClO peak near August 24, the fast increase in ClO measured by MLS between August 28 and September 7 is to some extent caused by the very large fractional increase in sunlight exposure during this period. Figure 4 shows the diurnal variation of ChlOE ClO column density for measurements at Scott Base on days when hourly measurements were possible. As is seen in the climatologies in Figure 3, the difference between the MLS and ChlOE measurements decreases as the length of daylight increases and the 1630 LST MLS measurement becomes more representative of a mid-day measurement.

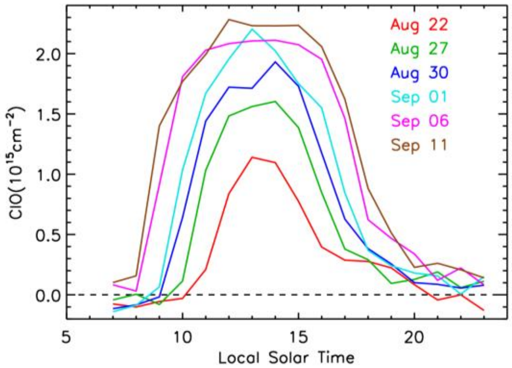

Figure 4 – The diurnal variation of ClO column density at altitudes above 100 hPa at Scott Base as measured from a series of measurement days in 2005. The date given for each curve is the middle date of a 3-day average of hourly measurements.

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We note that Upper Atmosphere Research Satellite (UARS) MLS ClO measurements are available for the years 1991-1993. Using the ground-based ChlOE measurements from Mauna Kea, we previously showed that the UARS MLS ClO measurements in the upper stratosphere were consistent with the Aura MLS ClO measurements (agreeing to within ~1±4%(2σ); Nedoluha et al., 2011). However, unlike Aura MLS, UARS MLS was in a precessing orbit, and therefore the local solar times of measurements varied from day to day. Given the large diurnal variability of lower stratospheric ClO in the vortex we would require an extremely accurate model in order to be able to usefully compare the UARS MLS ClO measurements with other measurements in this study.

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3. Annual ClO Anomalies

On the basis of 20 years of springtime lower stratospheric ClO measurements from Scott Base, and 12 years of MLS measurement near this latitude, we will provide an estimate of the trend in Cl_y in this region which underlies the trend in ClO. To calculate this trend, we need a period over which we have an adequate number of elevated ClO measurements, and during which the year-to-year differences resulting from meteorological variations are minimized. As was shown in Figures 2 and 3, there is a gradual increase in ClO at the latitude of Scott Base

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from ~August 28 to September 17. At the latitude of Scott Base (77.85ºS), we do not expect that during these dates any of the measurements have occurred outside of the vortex (except possibly in 2002, when the Antarctic stratosphere exhibited an unusual major warming). In the weeks after September 17, there is generally a very sharp drop in ClO, with the exact timing of this drop differing from year to year. As a measure of this increased variation, we note that the standard deviation of the 12 years of MLS measurements near Scott Base increases from 29% of the ClO column on September 17 to 58% of the ClO column on September 22. We therefore choose September 17 as the final day of the period for which we will compare interannual variations. On August 28, the climatological ClO from the ChlOE measurements is similar to that on September 17, and the ChlOE measurements show a steep increase up to this date. The choice of August 28 as the first day for the comparisons provides us a 3-week period with an average of 16.4 daily measurements from ChlOE for each year.

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We calculate, for the chosen 3-week period, an annual anomaly for each measurement dataset by taking the average difference from the climatology. We then add back the climatological average column ClO for the period so that we can express changes in ClO both in absolute and in fractional terms. So for each year we plot in Figure 5

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$$Y(\mathrm{year}) = Y_{\mathrm{climo}} + [1/n(\mathrm{year})]\ \Sigma\ [D(\mathrm{year,day}) - D_{\mathrm{climo}}(\mathrm{day})] \qquad (1)$$

where Y_{climo} is the dataset-specific climatological average column ClO for the 3-week period, n(year) is the number of measurements during that period for a specific year, D(year,day) is the measured column ClO for that day, and D_{climo}(day) is the dataset-specific climatological average column ClO for that day of the year. MLS values are shown both for the zonal average (within 77.85ºS±2º) and with a further restriction to within ±15º longitude of Scott Base. As expected from the difference in diurnal sampling, the MLS average column day minus night ClO values are somewhat smaller than those from the ChlOE measurements.

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We also show in Figure 5 the average ozone mass deficit (in 10^9 kg of ozone relative to the 220 DU value) for September and October of each year. This data was obtained from NASA Ozone Watch (ozonewatch.gsfc.nasa.gov) and is based upon data from the Total Ozone Mapping Spectrometer (TOMS) and the Ozone Monitoring Instrument (OMI), with missing data filled in by the Goddard Earth Observing System Model (GEOS-5).

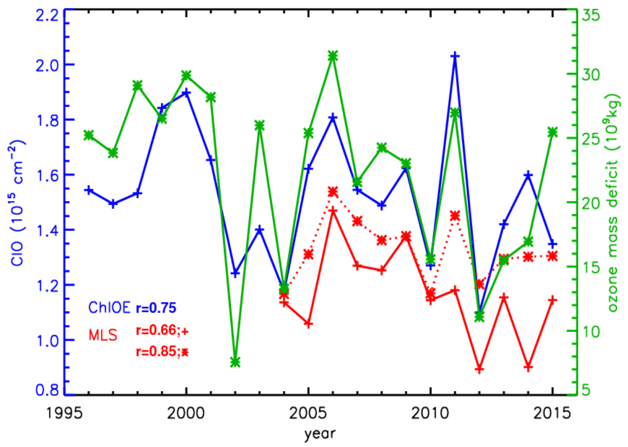
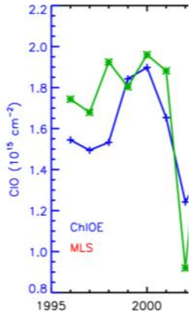


Figure 5 – The climatology plus annual average anomaly (1) day minus night ClO column density for August 28 to September 17 of each year, calculated as described in text. Averages are shown for ChlOE at Scott Base (blue), MLS coincident with Scott Base (solid red) and MLS at the latitude of Scott Base (dashed red line). Also shown is the ozone mass deficit in 10^9 kg of ozone relative to the 220 DU value (green line, with right-hand axis). The correlation coefficients between the ClO measurements and the ozone mass deficit are indicated.

Figure 5 shows a strong correlation between the annual average ClO column and the ozone mass deficit. For the 20 years of ChlOE measurements the correlation coefficient between these is 0.75. The correlation coefficient increases to 0.78 if we do not include 2014, for which there are only 6 ChlOE measurement days out of a possible 21 between August 28 and September 17, as opposed to the annual average of 16.4 measurement days. For the 12 years of Aura MLS ClO column measurements, the correlation coefficient is 0.66 for the measurements coincident with Scott Base, and 0.85 for the zonally averaged measurements at the Scott Base latitude.

4. Temperature and ClO

The fraction of Cl$_y$ that is in the form of ClO is sensitive to the availability of PSCs, which require low temperatures (below ~195K). As was noted by Santee et al. (2011) for the 2006 winter, the chlorine deactivation and the dissipation of PSCs as observed by the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO; Pitts et al., 2009) both occur in mid-October. While the variation in ClO measured at any one place and time is



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dependent not just upon the local temperature but also upon the temperature history of the measured parcel, we do find that in many cases sudden changes in local temperatures coincide with changes in measured ClO. An example of the sensitivity of ClO to changes in temperature is seen very clearly in Figure 1 of Kremser et al. (2011), where a sudden increase in temperature in early September over Scott Base resulted in a sudden decrease in measured ClO.

Figure 6 shows daily (day minus night) ChlOE column measurements for mid-August to mid-October 2000. This is the same as Figure 3, but for a different (pre-Aura MLS) year. Also shown in Figure 6 are MERRA temperatures at 30 hPa (the pressure level nearest to the ClO mixing ratio peak) within ±2º latitude and ±15º longitude of Scott Base. In addition, we show the climatological temperature from the same 20-year time period as the ChlOE measurements. While the temperatures in late August are clearly colder than those in September, the ClO during this period remains low because of a lack of the sunlight required for the activation of chlorine. Once sunlight becomes available, the ClO column begins to increase and, in this particular year, increases to levels well above the climatology. As Figure 5 shows, this year is second only to 2011 in the annual average ClO column for August 28 to September 17. At the same time, Figure 6 shows that the temperatures are colder than the climatology from August 28 to September 15, and are then warmer than the climatology for 6 of the next 7 days. The date when the temperature crosses from below to above the climatological value is the same date on which the ClO column density crosses from above to below the climatology.

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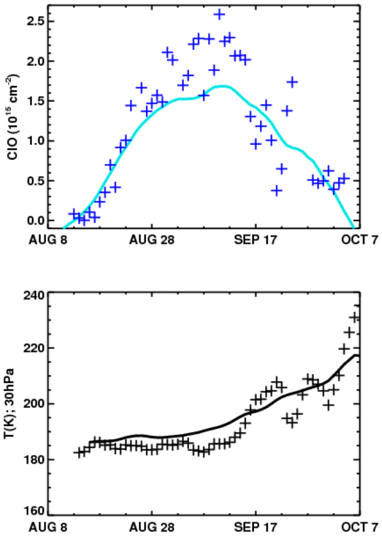

Figure 6 – Top panel: Daily (day minus night) ChlOE1 column density measurements for mid-August to mid-October 2000 (crosses) and a climatology for that period based on the ChlOE measurements from 1996-2015 (solid line). Bottom panel: Daily 30 hPa temperature from MERRA within ±2° latitude and ±15° longitude of Scott Base (crosses) and a 1996-2015 climatology for this location (solid line).

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The annual average MERRA temperature anomalies at three pressure levels (20, 30, and 40 hPa) are plotted in Figure 7. As in Figure 5, these are calculated by taking the average difference between the daily temperature and the temperature climatology for that day over the 3-week period of August 28 to September 17, but here we do not add back the climatological temperature average. We find that the relationship between temperatures at these three levels changed between 1998 and 1999. The 20 hPa and 30 hPa temperature anomalies suggested extremely cold years from 1996-1998 (at 20 hPa 1996 and 1997 were the coldest years), while at 40 hPa none of these three years was the coldest in the 20-year record.

In between the 1998 and 1999 periods that we analyzed, data from the Advanced TIROS (Television Infrared Observation Satellite) Operational Vertical Sounder (ATOVS, on NOAA15) began to be assimilated into the MERRA analysis, and it has been shown that this causes some inhomogeneities in the reanalysis (Pawson, 2012). We therefore compared the MERRA temperatures with sondes launched from Scott Base from 1996-1998, and with sondes launched from 1999-2010. We found that the cold bias of MERRA relative to the sondes at 20 hPa was much reduced in the 1999-2010 MERRA temperatures. Due to the biases in MERRA

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temperatures indicated by the sonde data, we added 4.0K to the 20 hPa 1996-1998 MERRA temperatures and 2.1K to the 30 hPa temperatures to account for this temperature bias. We also subtracted 0.3K from the 40 hPa 1996-1998 MERRA temperatures. When we estimate chlorine trends in Section 5, it will be particularly important to have temperatures during these first three years of ChlOE measurements that are consistent with temperatures in later years.

Given the anti-correlation between column ClO and temperature, we would expect an anti-correlation between temperature and ozone loss. Just as in Figure 5, we therefore also show in Figure 7 the ozone mass deficit, although in this case with the scale inverted. The magnitude of the anti-correlation between the 30 hPa temperature anomalies over Scott Base and the ozone mass deficit shown in Figure 7 is comparable to that of the correlation between ClO and ozone mass deficit shown in Figure 5, with a correlation coefficient of -0.82, while for the zonal average temperatures the correlation drops to -0.78. The temperature and ozone mass deficit have a slightly weaker correlation at 40 hPa, while at 20 hPa the correlation is slightly stronger for the local temperatures and slightly weaker for the zonal average temperatures.

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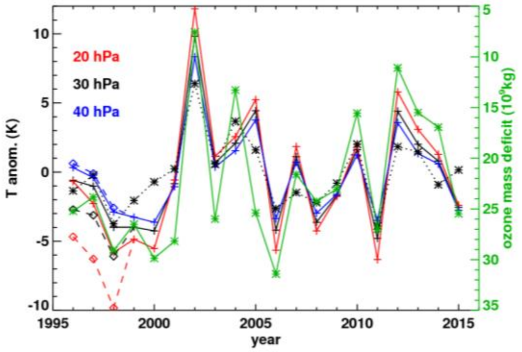

Figure 7 – Annual average temperature anomalies for August 28 to September 17 within ±2° latitude and ±15° longitude of Scott Base. Results are shown at 20 hPa (red), 30 hPa (black), and 40 hPa (blue). The dashed lines for 1996-1998 show the anomalies before applying the bias correction to the MERRA temperatures (see text). Also shown (dotted black line) is the zonal temperature anomaly for this latitude range. The green line shows the ozone mass deficit, with values given on the right-hand axis (as in Figure 5, but with the axis reversed).

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Figure 8 presents scatter plots of the annual average column ClO and the 30 hPa temperature anomalies for August 28 to September 17. Since, after the bias correction, the temperature anomalies are very similar for all three pressure levels, the anomalies shown in Figure 8 are nearly independent of the pressure level chosen for the temperatures. We chose 30 hPa since, among the three pressure levels shown in Figure 7, zonally averaged temperatures at this level showed the highest correlation with MLS zonally averaged column ClO measurements (correlation coefficients of -0.862, -0.863, and -0.781 at 20, 30, and 40 hPa respectively). For the local MLS measurements and ChlOE, the correlation with local temperatures was slightly higher at 20 hPa. Results are shown for ChlOE measurements, as well as for MLS measurements both zonally averaged (with corresponding zonally averaged temperatures) and restricted to within ±2° latitude and ±15° longitude of Scott Base.

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To establish a linear fit for the annual average anomalies shown in Figure 8, we need to estimate uncertainties in the temperature and ClO measurements. We estimate these uncertainties by calculating the standard error of the mean for the daily anomalies for each year. This will tend to weight years that have consistently high (or low) ClO column and temperature anomalies, as well as, for ChlOE, years when there are a large number of measurements (MLS almost always has measurements for every day). The uncertainties for each year are generally similar, but in 2014 there were very few ChlOE measurements and these measurements were particularly variable.

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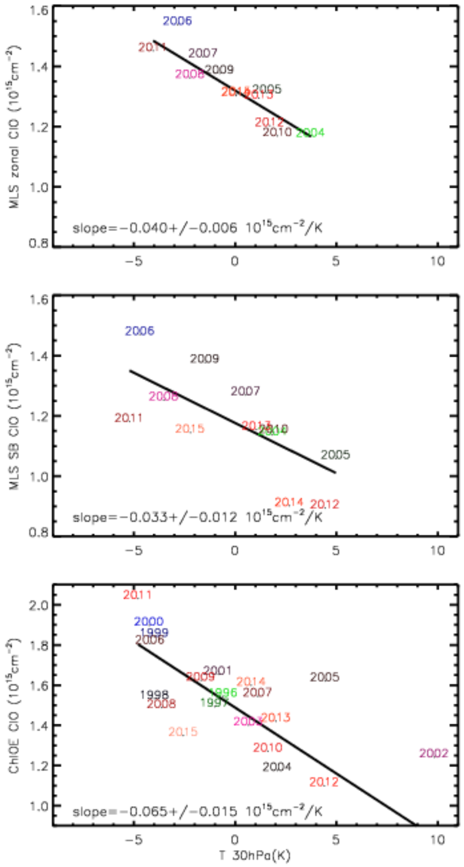

Figure 8 – The climatology plus annual average anomaly for the August 28 to September 17 column ClO (shown in Figure 5) plotted against the temperature anomalies (shown in Figure 7). Also shown are linear fits with a 1σ error estimate. Results are shown for the zonally average MLS ClO column measurements and 30 hPa MERRA temperatures within ±2° latitude of Scott Base (top), for MLS ClO and MERRA temperatures with a further restriction to within ±15° longitude of Scott Base (middle), and for ChlOE ClO measurements and MERRA temperatures with this tighter restriction (bottom).

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We calculated linear fits and found that the slopes were -0.040±0.006 10^{15}cm^{-2}/K, -0.033±0.012 10^{15}cm^{-2}/K, and -0.065±0.015 10^{15}cm^{-2}/K for the zonal MLS, Scott Base MLS, and ChlOE ClO measurements, respectively. We attribute the difference in the linear fits between the ChlOE and MLS ClO measurements to the different diurnal sampling of the ChlOE and MLS measurements. The slopes for the MLS measurements near Scott Base and for the zonally averaged MLS measurements at this latitude are not statistically different.

As a consistency check, we repeated this study using temperatures from the NCEP Reanalysis (REAN2) (Kistler et al., 2001), and calculated fits that were nearly identical to those shown in Figure 8. The slopes were very close to those calculated with MERRA: -0.040±0.006 $10^{15}cm^{-2}/K$, -0.032±0.012 $10^{15}cm^{-2}/K$, and -0.068±0.015 $10^{15}cm^{-2}/K$ for the zonal MLS, Scott Base MLS, and ChlOE ClO measurements, respectively.

5. Estimating a Chlorine Trend

There have been a number of studies attempting to quantify the temporal trend in Cl_y using measurements of either HCl or ClO. HCl is the primary reservoir species for chlorine, and measurements of HCl in the upper stratosphere show a decline since around 1997 (Anderson et al., 2000; Froidevaux et al. (2006); Jones et al., 2011; Nedoluha et al., 2011). Jones et al. (2011) showed, for a range of latitude bands, a decrease in HCl measured by the Halogen Occultation Experiment (HALOE) of between 0.4 and 0.6% yr^{-1} from 1997-2005. Froidevaux et al. (2006) estimated a decrease of ~0.8±0.1% yr^{-1} from Aura MLS HCl measurements over a very brief August 2004 to January 2006 period, but unfortunately the MLS channel measuring HCl near the stratopause experienced rapid deterioration so no extended HCl trend study from the MLS dataset has been possible. Jones et al. (2011) produced a combined ODIN/SMR and MLS ClO dataset for 2001-2008, and calculated a trend of -0.7±0.8% yr^{-1} (2σ) in tropical ClO from 35-45km. Nedoluha et al. (2011) used ground-based measurements of ClO from Mauna Kea to show a clear decrease since 1996, and validated the relative consistency of the UARS MLS (1991-1998) and Aura MLS (2004-present) ClO measurements. Finally, Connor et al. (2013) used a reanalyzed version of the ground-based ClO measurements from Mauna Kea and calculated a trend of -0.64±0.15% yr^{-1} (2σ) from 1995-2012.

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The linear trend in the annual average ChlOE ClO columns from August 28 to September 17 (those shown in Figure 5 from 1996-2015) is -1.1±0.4% yr^{-1}. However, since the first ChlOE measurement years were colder than average, this trend is almost certainly to some extent the result of increased processing on PSC particles during these years, and is therefore not representative of the trend in Cl_y. In addition, interannual variations in dynamics will cause interannual variations in Cl_y which will in turn affect the measured ClO. Strahan et al. (2014)

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used the compact relationship between nitrous oxide (N_2O) and Cl_y, as established by Schauffler et al. (2003), to estimate the variability in Antarctic Cl_y for the years 2004-2012 based upon MLS measurements of N_2O. They found year-to-year variations of Cl_y in the vortex on the 500K potential temperature surface of as much as ~7%.

Accounting for the dynamical variations over the entire 20-year ChlOE measurement dataset is problematic, and we will not attempt to do so here, but it is certainly possible to account for the interannual temperature variations over this period. Making use of the annual temperature anomalies, we calculate an adjusted annual column ClO, which is given for each year by $ClO_{adj}(year) = ClO(year) - \alpha\, \Delta T(year)$, where α is the temperature dependence of ClO shown in Figure 8 and $\Delta T(year)$ is the temperature anomaly for that year. To the extent that we have successfully removed the effect of temperature variations, the variations in the adjusted ClO columns should better represent the variation in Cl_y.

The column ClO values, adjusted for interannual temperature variations, are shown in Figure 9. We then calculate a linear trend using these modified column ClO values and express the trends as a function of the average column values. The resultant trends calculated for zonal MLS, Scott Base MLS (2004-2015), and ChlOE (1996-2015) are $-0.5\pm0.2\%\ yr^{-1}$, $-1.4\pm0.9\%\ yr^{-1}$, and $-0.6\pm0.4\%\ yr^{-1}$, respectively. Note that the 1σ error bars shown in this plot are the same as the error estimates used in establishing the ClO column vs. temperature relationship in Figure 8. The fraction of points falling within 1σ of the trend line is approximately what would be expected given a Gaussian distribution, so our uncertainty estimate seems reasonable. If we use the REAN2 temperatures both to establish the relationship between temperature and ClO column and subsequently to calculate trends then we find trends almost identical to those found with the MERRA temperatures. The calculated trends in adjusted ClO column, as calculated using REAN2 temperatures, are $-0.6\pm0.2\%\ yr^{-1}$, $-1.4\pm0.9\%\ yr^{-1}$, and $-0.5\pm0.4\%\ yr^{-1}$ for zonal MLS, Scott Base MLS, and ChlOE, respectively.

While the trends are almost insensitive to the choice of temperature dataset, they are somewhat sensitive to the precise choice of dates from which the annual average is determined. Although we believe that we have made an optimal choice for these dates, it is nevertheless instructive to examine this sensitivity. If we add or subtract 5 days from the beginning or end of the comparison periods and repeat our calculations for these four additional cases we find trends

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in adjust ClO columns in the range -0.3 to -0.6% yr^{-1} for the zonal MLS measurements, -0.9 to -1.8% yr^{-1} for the local MLS measurements, -0.2 to -0.7% yr^{-1} for the ChlOE measurements.

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Finally, we have also performed the entire analysis using daytime zonal average MLS measurements without subtracting the nighttime measurements. The results are very similar to the results from day minus night measurements, agreeing to within 1σ in both the sensitivity of the ClO column to temperature, and in the calculated trend.

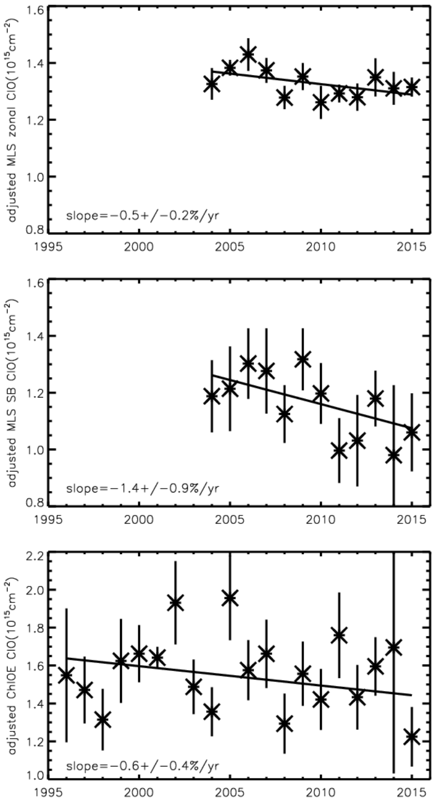

Figure 9 – The annual average temperature adjusted ClO columns (see text) for August 28 to September 17. The adjustment is based upon the annual average temperature and the relationship shown in Figure 8 (see text). Results are shown for the zonally averaged MLS measurements within ±2° latitude of Scott Base (top), for MLS measurements within ±2° latitude and ±15° longitude of Scott Base (middle), and for ChlOE measurements at Scott Base (bottom). Also shown is a linear fit to the data. Uncertainties are 1σ.

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6. Summary

We have shown column ClO from 20 years of ChlOE measurements over Scott Base, Antarctica, as well as from 12 years of Aura MLS measurements near Scott Base and zonally averaged around 78°S. Interannual variations in column ClO over the 3-week period from August 28 to September 17 were correlated with the average ozone mass deficit for September and October (r=0.75 for ChlOE). Such a correlation is to be expected, given that ClO is the catalytic agent in the most important ozone-destroying cycle.

We have also shown that the interannual variation in column ClO is anti-correlated with interannual variations in 30 hPa temperature. This is physically reasonable since colder temperatures increase the availability of polar stratospheric clouds, and these will in turn provide the heterogeneous surfaces for the production of ClO (Molina and Molina, 1987; Solomon, 1999).

The multi-year ChlOE and Aura MLS datasets provided the opportunity to study trends. While there have been a number of studies of trends in Cl_y, this is to our knowledge the first study that addresses the question of stratospheric Cl_y trends in the Antarctic region. Since the ozone hole represents the most extreme manifestation of ozone depletion, it is of particular interest to determine whether the trends in Antarctic stratospheric Cl_y, which underlie the trends in ClO that causes this destruction, are similar to those measured elsewhere.

Because of the strong dependence of ClO on temperature, any calculated trend in ClO could misrepresent the trend in Cl_y, particularly if there were unusually warm or cold temperatures near the beginning or end of the timeseries. We therefore used the calculated relationship between interannual variations in column ClO and 30 hPa temperature to account for the effect of variations in column ClO caused by changes in temperature. We then calculated trends in temperature-adjusted ClO. The resultant trends for zonal MLS, Scott Base MLS (2004-2015), and ChlOE (1996-2015) were -0.5±0.2% yr^{-1}, -1.4±0.9% yr^{-1}, and -0.6±0.4% yr^{-1}, respectively. While our temperature regression does not account for dynamical effects that might influence ClO trends (e.g. changes in the Brewer-Dobson circulation), these trends are within 1σ of trends in Cl_y previously found at other latitudes (WMO, 2014). The decrease in ClO is consistent with the trend expected from regulations enacted under the Montreal Protocol.

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