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Comparison of ClO measurements from the Aura Microwave Limb Sounder to ground-based microwave measurements at Scott Base, Antarctica, in spring 2005

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[1] We have examined measurements of chlorine monoxide, ClO, in the lower stratosphere by the Microwave Limb Sounder (MLS) on the Aura satellite in the austral spring of 2005. These measurements have been compared to those of a ground-based spectrometer at Scott Base, Antarctica. The data analysis is performed in both cases by subtracting nighttime measurements from daytime ones. The transition from full darkness to full daylight at the high latitude of Scott Base limits the time during which both day and night measurements are made. After further selection for good observing conditions and the position of the polar vortex, 16 valid profile comparisons are made. The day-to-day variability of ClO is observed to be large, ~30% of its peak value. The daily column densities of the two instruments are correlated with a significance of 3σ with most of the mean difference arising from 2 days. The statistical agreement between MLS and the Scott Base instrument is good. Scott Base values are on average marginally but not significantly larger, by 0.10 ± 0.07 ppb, or 11 ± 8% (1σ), in peak mixing ratio, than the MLS values.

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1. Introduction

[2] It is well established that formation of the Antarctic ozone hole is driven by the presence of "active chlorine" in the lower stratosphere in austral spring. Active chlorine is defined as $ClO_x = Cl + ClO + 2[Cl_2O_2]$. Chlorine monoxide, ClO, has been observed there at mixing ratios up to 2.2 ppb, and plays a unique, central role in the formation of the Antarctic ozone hole [*Barrett et al.*, 1988; *Brune et al.*, 1989; *Waters et al.*, 1993; *Solomon et al.*, 2002]. It is both the direct product of the reaction between Cl and ozone and the catalytic agent in the most important ozone-depleting chemical cycle [*Solomon et al.*, 1986; *Salawitch et al.*, 1993]. Total active chlorine, ClO_x, is thought to be present at concentrations of up to 2.7 ppb only because heterogeneous reactions on the surface of Polar Stratospheric Cloud particles release Cl from the reservoir species HCl and $ClONO_2$ [*Chipperfield*, 1999].

[3] To provide long-term monitoring of ClO over Antarctica, SUNY-Stony Brook and NIWA jointly operate an automated ground-based millimeter wave spectrometer at Scott Base, Antarctica (77.85°S). Operating year-round, the instrument observes the development of the ozone hole starting with its onset at polar sunrise. The instrument started measurement in February 1996, and has made the only routine, near continuous ClO measurements in Antarctica since then [*Solomon et al.*, 2002; *Connor et al.*, 2007]. A predecessor instrument made the first observation of enhanced lower stratospheric ClO in the ozone hole [*Solomon et al.*, 1987; *de Zafra et al.*, 1987, 1989; *Barrett et al.*, 1988].

[4] We observe a thermally excited emission line of ClO at 278.6 GHz; observations are carried out around the clock throughout the year. The spectrometer bandwidth permits measurement of the pressure-broadened line shape from which ClO altitude profiles between 15 and 40 km are retrieved using a variant of the methods of *Rodgers* [2000]. These results are derived from daytime spectra from which nighttime spectra (in which the ClO line is weak and narrow) have been subtracted. This subtraction is done to cancel out interfering ozone lines and instrumental artifacts in the spectra.

[5] Measurements of ClO from space have been made by the MLS instruments carried on both the UARS [*Waters et al.*, 1993] and Aura satellites (M. L. Santee et al., Validation of the Aura Microwave Limb Sounder ClO measurements,

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submitted to *Journal of Geophysical Research*, 2007, hereinafter referred to as Santee et al., submitted manuscript, 2007). In this paper, we focus on a limited subset of MLS ClO measurements in the vicinity of Scott Base, made during the ozone hole in September 2005. These MLS data are compared to measurements made at Scott Base by the SUNY/NIWA microwave spectrometer. We have examined both of the ClO data sets discussed by Santee et al. (submitted manuscript, 2007), namely v1.5 and the fewer profiles available (as of the date of writing) in v2.2. We find small, insignificant differences in ClO between these versions for this subset of MLS data, and will focus exclusively on v1.5 data in this paper. The outline of the paper is as follows. In the next section we describe the specific data used for comparison, providing some details of the ground-based measurement and the selection criteria applied to both data sets. We next discuss quantitative comparison of these two remote measurements employing very different geometry; how it is done and how interpreted. We then present comparisons made between MLS and the ground-based data. Finally, we assess the comparisons.

2. Measurements

[6] The spectra measured with the ground-based instrument at Scott Base, their calibration, retrieval of ClO from them, and their error analysis, are described in detail by *Solomon et al.* [2000]. The first 5 years of results are compared to model calculations, and used to set limits on the ratio j/k_f, of the photolysis rate of the ClO dimer (j), and rate of the forward reaction forming the dimer (k_f), in the work by *Solomon et al.* [2002]. Peak daytime mixing ratios of ClO in the lower stratosphere in the Antarctic vortex reach 2.2 ppbv during late August and September [*Solomon et al.*, 2002]. A notionally identical instrument has been in operation since 1992 at Mauna Kea, Hawaii, and has observed a significant decrease in active chlorine in the upper stratosphere, starting in the mid-1990s [*Solomon et al.*, 2006].

2.1. Ground-Based Measurements

[7] In brief, the instrument is a cryogenically cooled ($\sim$20 K) heterodyne receiver, tuned to observe the ClO transition at 278.631 GHz by adjustment of a phase-locked local oscillator. It is coupled to a spectrometer with 506 MHz total bandwidth, and observes an emission line which is purely rotational, so has low excitation energy and consequently modest temperature sensitivity. As with other atmospheric lines of the same approximate frequency, the ClO line is pressure broadened throughout the lower and middle stratosphere. Its width is then approximately an exponential function of altitude, and the shape of the observed emission (integrated over altitude) is a sensitive function of the ClO altitude distribution. This sensitivity allows the ClO altitude distribution to be retrieved from the spectra.

[8] Before performing this retrieval, we make use of the fact that at night, the ClO emission is much weaker and narrower, because nearly all ClO_x rapidly converts to Cl_2O_2 after sunset in the lower stratosphere [*de Zafra et al.*, 1987; *Rodriguez et al.*, 1989; *Solomon et al.*, 2002]. This allows us to remove the instrumental baseline and a small number of interfering atmospheric spectral lines in the instrument band pass (primarily the ozone line at 278.521 GHz), by subtracting the nighttime spectrum from the daytime one.

[9] A retrieval of the "day-night" spectrum, and thus of the day ClO mixing ratio less the night mixing ratio, is then 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 examining a full season's data. 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 $\sim$30 mbar (22 km) in mid August to $\sim$48 mbar (19 km) by late September, with a secondary peak in the upper stratosphere at $\sim$5 mbar. 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)], and usually described as "optimal estimation."

2.2. Selection of Data for Comparison

[10] We will compare the ground-based measurements to MLS daytime overpass less MLS nighttime overpass measurements. Thus the available days are limited to those for which the sun is up during the MLS ascending overpass at approximately 1700 LST but down during the descending overpass at approximately 2230. More specifically, we require the full solar disk to be clear of the horizon at the precise time of each overpass, as seen from $\sim$20 km altitude, after making an approximate correction for refraction. These appropriate day numbers are 245–270. Fortunately, these days correspond to the period with the greatest amount of lower stratospheric ClO. We note that the peak daytime ClO measured at Scott Base is about a factor of 3 higher than that at 1700 LST on day 245; this ratio decreases rapidly as the sun sets later, becoming near 1 by day 263.

[11] MLS measurements are closely spaced at high latitudes. Thus there are frequent close overpasses, both for ascending and descending orbits, and even a choice of close overpasses on the same day. We have selected the MLS measurements geographically closest to 78.85°S, 166.77°E, which is a point 1° of latitude south of Scott Base, approximately where the ground-based spectrometer's beam intercepts the enhanced ClO layer at $\sim$20 km altitude. The nominal distance of both day and nighttime overpasses from this point ranges from 4 to 194 km, averaging just over 100 km. The horizontal coincidence is somewhat degraded by the MLS line of sight; *Livesey et al.* [2005] imply the width of the MLS horizontal averaging kernels is about 300 km in the polar lower stratosphere.

[12] Ground-based measurements were selected within a half-hour of the nominal time of the MLS ascending orbit overpass (1653 LST, or 1746 NZST, ±30 min). Spectra are recorded for a nominal integration time of 20 min, and then the spectra are averaged over the desired time interval. The nighttime spectrum is subtracted from the average, and a single ClO profile is retrieved from the result. Only spectra taken when the zenith optical depth was <0.3 were used (this is the same quality filter applied to all data taken at Scott Base). The corresponding nighttime spectrum is an

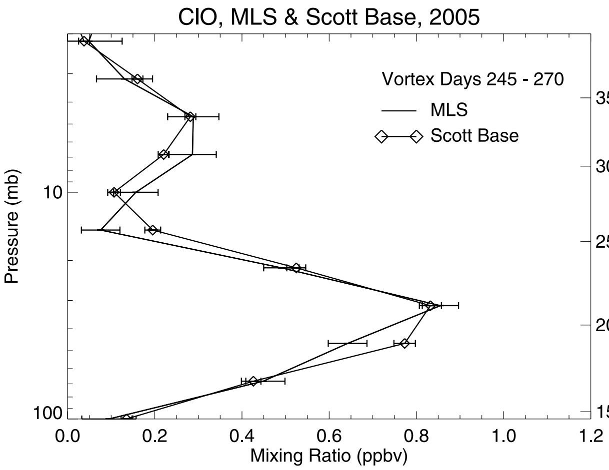

Figure 1. Mean profiles, from 16 measurements between 2 and 27 September 2005. The Scott Base profiles have been linearly interpolated onto the MLS retrieval grid, in log pressure. The error bars represent the precision of the mean profiles.

integration over the entire night, since ClO is expected to be near constant overnight.

[13] We tested our sensitivity to the nighttime integration period by comparing the nighttime spectra integrated over the entire night, to spectra from a shorter integration time, within one half-hour of the MLS descending overpass time. The shorter integration time caused increased scatter due to spectral noise, but there was otherwise no significant difference between the two sets of nighttime spectra. Therefore we have used the full night integration time exclusively.

[14] Of 26 possible days in 2005 (245–270) which have both day and night overpasses by MLS, 6 are eliminated by poor observing weather at Scott Base, and on 4 of the remaining 20 days, Scott Base was in a region of strong ozone gradient, and thus presumably near the vortex edge, as judged by inspection of OMI O_3 maps. Since ClO is likely to be highly spatially variable at such times, we have excluded them from the comparisons. Thus we include 16 days in the MLS comparisons, namely 245–250, 253–255, 260–264, 269, and 270.

3. Comparisons

[15] Remote measurements are not necessarily directly comparable, in the sense that they measure somewhat different functions of a desired geophysical quantity as the best estimates of that quantity. In particular, the horizontal and vertical sampling, expressed in the averaging kernels, and the correlations of the error between locations sampled, may vary from one measurement technique to another. To compare retrieved altitude profiles from different types of remote measurements, it is customary to account for these factors using a formal methodology, e.g., of *Rodgers and Connor* [2003]. Unless these factors are accounted for by that or a comparable procedure, quantitative comparison between remote measurements is severely compromised.

[16] However, the same result can be achieved by directly simulating the value of one measurement given another. The simulation approach is conceptually straightforward, though it requires significant computation for each comparison; we have adopted this procedure since we are concerned with a relatively small number of comparisons. Whether explicitly or by simulation, one forms effectively the same profile, which we use for comparison and which we refer to as the "convolved profile" [*Connor et al.*, 1994; *Rodgers and Connor*, 2003]. Conceptually, start with the ground-based retrieved profile expressed by *Rodgers and Connor* [2003, equation (3)] as

$$\hat{\mathbf{x}}_{SB} = \mathbf{x}_a + \mathbf{A}_{SB}(\mathbf{x} - \mathbf{x_a}) + \varepsilon_{SB} \tag{1}$$

Then the convolved MLS profile is

$$\mathbf{x}_c = \mathbf{x}_a + \mathbf{A}_{SB}(\mathbf{x}_{MLS} - \mathbf{x}_a) + \mathbf{A}_{SB}\ \varepsilon_{MLS} \tag{2}$$

where $\mathbf{x}_a$ and $\mathbf{A}_{SB}$ are the ground-based a priori and averaging kernel matrix, respectively, $\mathbf{x}$ is the real atmospheric profile, $\mathbf{x}_{MLS}$ is the MLS profile, and ε_{SB} and ε_{MLS} are the errors in the Scott Base and MLS profiles.

[17] If we ignore the last term, then $\mathbf{x}_c$ is what the ground-based measurement would find if the MLS profile were the true atmospheric profile, in the absence of other error. If it is compared to the ground-based retrieval $\hat{\mathbf{x}}$, the difference is given by

$$\hat{\mathbf{x}}_{SB} - \mathbf{x}_c = \mathbf{A}_{SB}(\mathbf{x} - \mathbf{x}_{MLS}) + \varepsilon_{SB} - \mathbf{A}_{SB}\ \varepsilon_{MLS} \tag{3}$$

with error covariance

$$\mathbf{S}_\delta = \mathbf{S}_{SB} + \mathbf{A}_{SB}\mathbf{S}_{MLS}\mathbf{A}_{SB}^T \tag{4}$$

where S_{SB} and S_{MLS} are the error covariances of the Scott Base and MLS measurements, respectively. Note that equation (3) is independent of the ground-based a priori profile, and thus such comparisons are unbiased by the use of a priori.

[18] Strictly, the last three equations are only applicable if the vertical resolution of the MLS measurements is much less than the ground-based measurements. For MLS, resolution in the lower stratosphere is 3–3.5 km [*Livesey et al.*, 2005] while for the 1-h ground-based integrations it is ~12 km, so we adopt equations (2)–(4) without modification.

3.1. Interpolated Profile Comparison

[19] We will start, however, with a naïve comparison of reported results. For this, we simply interpolate the ground-based retrieved profile onto the MLS pressure grid, and compare directly. The mean comparison of 16 profiles is shown in Figure 1. The agreement is striking. The amplitude of both peaks in ClO mixing ratio is nearly the same, the altitude of the lower stratospheric peak agrees well, and the altitude of the upper stratospheric peak agrees within approximately 2 km.

[20] Figure 2 shows some statistics relevant to the comparison, including the mean difference and the RMS difference. Also shown are the combined formal uncertainty (single measurement precision) of the 2 instruments. The main contributor to this formal uncertainty is the stated MLS precision; near the lower stratospheric peak the MLS precision is about 1.6 times larger than the Scott Base

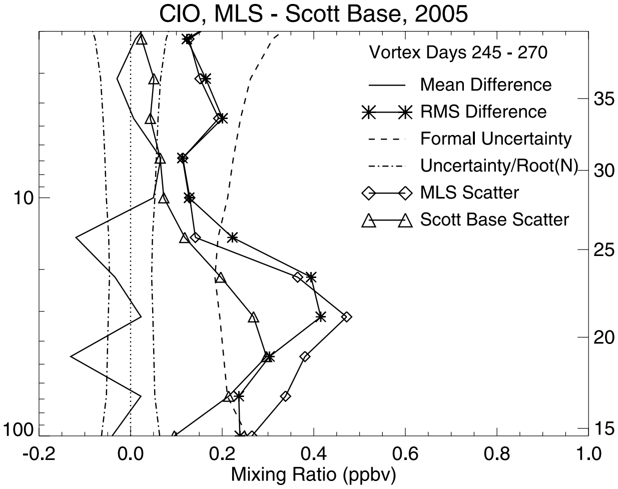

Figure 2. Statistics for the 16 profiles used to produce Figure 1. The line labeled "Formal Uncertainty" is the quadratic sum of the single-profile formal errors for the MLS and Scott Base data. That labeled "Uncertainty/Root(N)" shows the 1σ bounds of the expected mean difference. "MLS Scatter" and "Scott Base Scatter" show the RMS variation of the individual data sets.

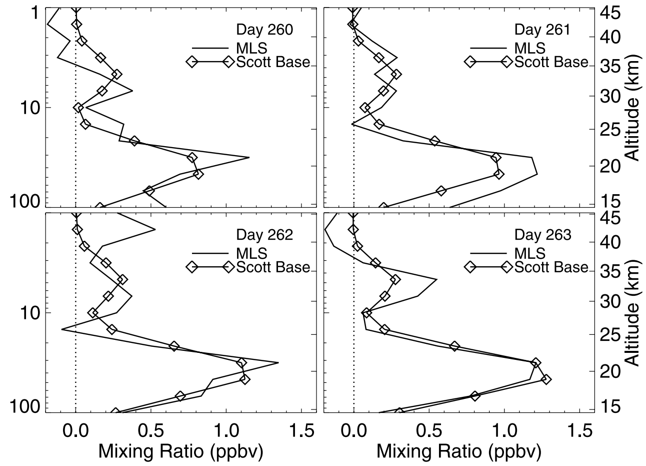

Figure 3. Profiles for days 260–263 from the Scott Base and MLS data.

instrument. This formal uncertainty is an estimate of the precision of the agreement between the instruments, or an estimate of the RMS difference in the comparison. There is rough agreement between this estimate and the actual RMS; the actual RMS exceeds the estimate by ~0.2 ppb near the peak, and is ~0.1 ppb less than the estimate at altitudes above 10 mbar. Finally, the formal uncertainty divided by the square root of the number of comparisons in the mean (16) is used to estimate the 1σ bounds on the mean difference. The agreement of the actual mean difference with this estimate is good, though on average the Scott Base values are slightly greater than MLS below ~15 mbar.

[21] Also shown in Figure 2 is the RMS variability of the two data sets. Both instruments measure highly variable ClO in the lower stratosphere; this is an important factor in quantitative interpretation of the comparison, and will be discussed further in the next section. The scatter of MLS is substantially larger than that of Scott Base. A significant factor is the lower estimated precision in single MLS profiles. This may also reflect variability in real vertical structure that the ground-based instrument cannot measure. Finally, in Figure 3 we show 4 individual days, 260–263, to illustrate some of the variability, both in each data set and in the comparison.

3.2. Convolved MLS Profiles

[22] To account for the effects of the limited vertical resolution of the ground-based measurements, and their a priori information, the convolved MLS profiles were derived by simulation of the ground-based measurements, as discussed above. First, the MLS profiles were assumed to represent the real atmosphere and microwave spectra were calculated from them. The separate day and night spectra were then processed by the same software used for the real ground-based microwave spectral measurements. Note that the result of this procedure is identical to use of equation (2), ignoring the error term, if one makes the very good approximation of linearity of the Scott Base retrieval in the vicinity of its a priori. These results are referred to as "Convolved MLS" and are shown in Figures 4 and 5.

[23] Agreement in the lower stratosphere is very good considering the extreme variability of ClO and the limited number of comparisons which can be made. On the 16 available days, the peak ClO mixing ratio at the time of MLS observation was measured by the Scott Base instrument to be 0.87 ± 0.29 ppb (1 RMS scatter), while the Scott Base–MLS difference was 0.10 ± 0.29 ppb. The mean difference between MLS and the ground-based instrument is not significant (0.10 ± 0.07 ppb, or ~1.5σ).

[24] In naïve comparison (section 3.1), both instruments see a peak value of ClO in the mid-to-upper stratosphere of ~0.3 ppb, with a small vertical offset (Figure 1). Simulating ground-based measurement of the MLS profiles eliminates

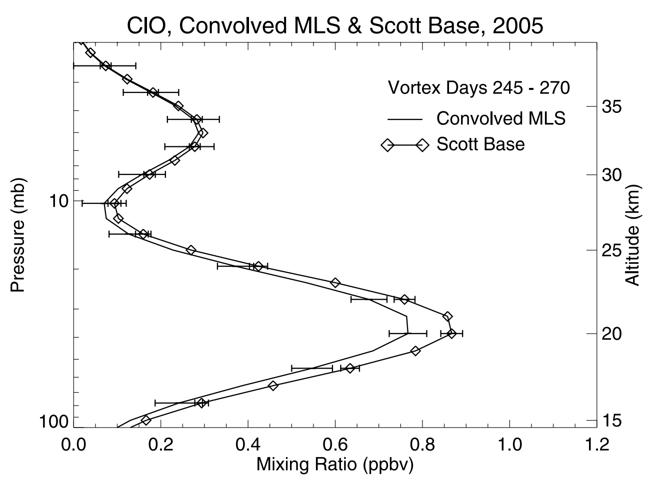

Figure 4. Mean profiles, from 16 measurements between 2 and 27 September 2005. The Scott Base profiles are shown on their native retrieval grid, converted to pressure. The error bars represent the precision of the mean profiles. Convolved MLS profiles have been produced by using MLS data to simulate the ground-based measurements (see text).

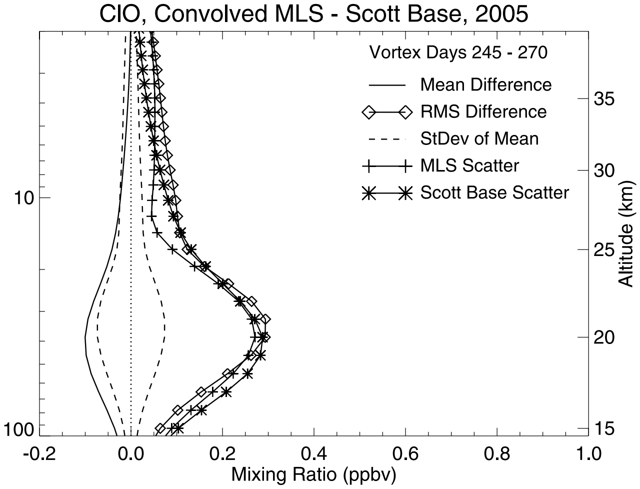

Figure 5. Statistics for the 16 profiles used to produce Figure 4. Those labeled "StDev of Mean" show the 1σ bounds of the expected mean difference. The MLS scatter and Scott Base scatter are the RMS variation of those two data sets about their means.

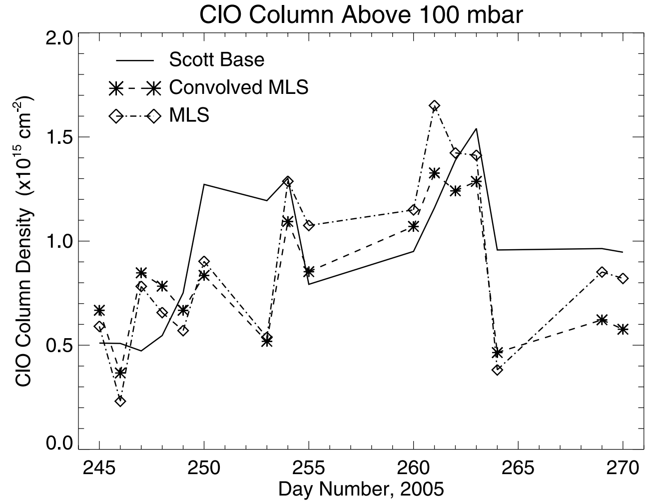

Figure 6. Column densities for the interpolated Scott Base data and MLS data, before and after convolution to simulate the ground-based measurement. The mean difference for the data before convolution (Scott Base-MLS) is 5.2 ± 5.1%. After convolution, the mean difference is 6.9 ± 4.7%.

the vertical offset between them, as seen in Figure 4. This implies the offset is an artifact of the measurement and retrieval process, presumably due to the a priori profile used for the Scott Base instrument.

[25] The ClO column density in the lower stratosphere is a good indicator of the total active chlorine present. Both instrument teams recommend use of their results for altitudes at and above about 100 mbar, and the column above that level dominates the lower stratospheric column, since the ClO amount at lower altitudes [*Brune et al.*, 1989; *Anderson et al.*, 1989] and in the middle and upper stratosphere is small. In Figure 6 we show the time series of ClO column above 100 mbar for the ground-based data as well as for MLS, before and after convolution. Detailed numerical results are shown in Table 1. As seen in Figure 6, the ground-based and MLS instruments track each other reasonably well. The correlation coefficients between the 2 instruments imply a 99% probability of coherent variation.

[26] The large day-to-day variations seen in both data sets (see Table 1) are thought to be primarily due to real changes in ClO, as noted in the discussion of Figure 2. For comparison, the estimated precision of the column measurement is $\pm 1.2 \times 10^{14}$ cm^{-2} for Scott Base and $\pm 1.9 \times 10^{14}$ cm^{-2} for MLS.

[27] The corresponding precision of the difference between the two column measurements is $\pm 2.3 \times 10^{14}$ cm^{-2}. The scatter observed is slightly larger (3.0×10^{14} cm^{-2}). We have considered the possibility that this marginal discrepancy may be due in part to real horizontal variability. The distance to the nominal overpass points and the horizontal averaging by MLS will both contribute to this effect. Considering these effects and typical wind speeds in the polar vortex, we estimate the resulting variability in the full set of comparisons at 5–10%. However, we have examined the difference in column density compared to overpass distance and find no correlation. Thus we have no direct evidence to support the importance of horizontal variability to the statistics of the comparison.

[28] Two of the 16 days, 253 and 264, are observed to have particularly large differences between the two column measurements. If we exclude those two days, the difference between the data sets is $0.6 \pm 2.5 \times 10^{14}$ cm^{-2}. Thus if we treat those days as outliers, the scatter in the column difference is almost fully accounted for by the estimated measurement precision.

[29] The effect of broad vertical resolution and the use of an a priori profile on the ground-based results may be seen by the comparison between the MLS columns before and after convolution. The differences are important but not large, approaching 10%. Fortuitously, the columns before convolution agree better on average with the ground-based data than do the columns after convolution (albeit with slightly larger scatter), even though the convolution corrects known errors in the comparison.

4. Conclusions

[30] The relatively brief time of year when ClO is enhanced and both daytime and nighttime measurements

t1.1 **Table 1.** ClO Column Density, $p < 100$ mbar[a]

		Mean Column 10^{14}, cm^{-2}	SB-MLS 10^{14}, cm^{-2}	SB-MLS(%), Std of Mean	Correlation	Probability
t1.2						
t1.3	SB	9.5 ± 3.4	–	–	–	–
t1.4	MLS as reported	9.0 ± 4.1	0.6 ± 3.1	6.3 ± 8.5	0.66	0.996
t1.5	MLS convolved	8.3 ± 3.0	1.3 ± 3.0	14.3 ± 8.3	0.58	0.990

t1.6 [a]Unit is cm^{-2}.

can be made, making comparisons possible, limits the number of profiles available for this validation study. With that proviso, the mean agreement between MLS and ground-based data is very good. Both the mean and RMS differences compare reasonably well to uncertainties stated with the data sets.

[31] In order to eliminate the influence of the ground-based a priori profiles and to minimize the effect of limited vertical resolution, the convolved MLS data have been produced by simulation of the ground-based measurements. Lower stratospheric ClO is highly variable during this time, which complicates the interpretation. The RMS variability of the peak mixing ratios of convolved MLS and of the Scott Base data is very similar, and is more than 30% of the mean peak value. Nevertheless, the time series of ClO column densities measured by the two instruments are consistent and coherent at a 3σ level. The convolved MLS ClO values are on average slightly less than the Scott Base values. The peak mixing ratios are less by 11 ± 8%, and the column densities by 14 ± 8% (1σ uncertainties in the mean). Note, however, that these differences are less than the 2σ uncertainty, and thus not statistically significant.

[32] Most of the mean difference between MLS and Scott Base results from 2 specific days (numbers 253 and 264; see Figure 6). If we remove these 2 days, the convolved MLS mean difference decreases from $1.3 \pm 3.0 \times 10^{14}$ cm^{-2} to $0.6 \pm 2.5 \times 10^{14}$ cm^{-2}, and the correlation coefficient of the two data sets improves from 0.58 to 0.72. The scatter in the comparison with days 253 and 264 removed is close to the estimated precision of the comparison.

[33] Finally, we note that the Scott Base data are normally reported as midday averages [*Solomon et al.*, 2000, 2002]. The agreement between MLS and the Scott Base measurements at the time of MLS overpasses (~1700 LST) may be viewed as indirectly validating the midday Scott Base measurements relative to the approximately midday measurements made by MLS at other latitudes.

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