Atmos. Chem. Phys., 13, 8643–8650, 2013
www.atmos-chem-phys.net/13/8643/2013/
doi:10.5194/acp-13-8643-2013

Atmospheric Chemistry and Physics Open Access

Re-analysis of ground-based microwave ClO measurements from Mauna Kea, 1992 to early 2012

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Received: 12 October 2012 – Published in Atmos. Chem. Phys. Discuss.: 26 November 2012
Revised: 22 July 2013 – Accepted: 25 July 2013 – Published: 2 September 2013

Abstract. We present a re-analysis of upper stratospheric ClO measurements from the ground-based millimeter-wave instrument from January 1992 to February 2012. These measurements are made as part of the Network for the Detection of Atmospheric Composition Change (NDACC) from Mauna Kea, Hawaii, (19.8° N, 204.5° E). Here, we use daytime and nighttime measurements together to form a day–night spectrum, from which the difference in the day and night profiles is retrieved. These results are then compared to the day–night difference profiles from the Upper Atmosphere Research Satellite (UARS) and Aura Microwave Limb Sounder (MLS) instruments. We also compare them to our previous analyses of the same data, in which we retrieved the daytime ClO profile. The major focus will be on comparing the year-to-year and long-term changes in ClO derived by the two analysis methods, and comparing these results to the long-term changes reported by others. We conclude that the re-analyzed data set has less short-term variability and exhibits a more constant long-term trend that is more consistent with other observations. Data from 1995 to 2012 indicate a linear decline of mid-stratospheric ClO of $0.64 \pm 0.15\,\%\,\mathrm{yr}^{-1}$ (2σ).

1 Introduction

Chlorine monoxide (ClO) in the stratosphere is the product of catalytic destruction of ozone by chlorine released from anthropogenic compounds, especially CFCs (chlorofluorocarbons). Remote measurements of ClO were begun from Mauna Kea, Hawaii, in 1982 and have been made near-continuously since 1992, barring interruptions for instrument repair (Solomon et al., 1984, 2006). The measurements are made by ground-based observation of thermally excited spectral emission lines at 278.6 GHz, by a small telescope and radiometer designed for the purpose. This instrument and its sibling at Scott Base, Antarctica (Solomon et al., 2000, Connor et al., 2007), have made the longest continuous records of stratospheric ClO in existence.

The calibrated spectra are prepared for analysis by subtraction of nighttime spectra from daytime ones, to remove interfering spectral lines and instrument artifacts. This technique was justified by Solomon et al. (1984) with a detailed discussion of ClO diurnal variation. More recently, Ricaud et al. (2000) have shown that mid-stratospheric ClO rises rapidly after sunrise from low nighttime values, while upper stratospheric ClO shows little diurnal variation. Mesospheric ClO, which has a nighttime maximum (Sato et al., 2012), will augment the relatively narrow upper stratospheric signal (see Sect. 3.2 for further discussion).

All diurnal effects combine to produce a ClO emission line that is narrow and very weak during night (except in polar spring). Previously published data from Mauna Kea (but not from Scott Base) were processed after interpolation of the nighttime spectra over a narrow region around the ClO frequency, which effectively removed the nighttime signal and produced a background spectrum. This was subtracted from the full daytime spectrum, to produce an estimate of the ClO

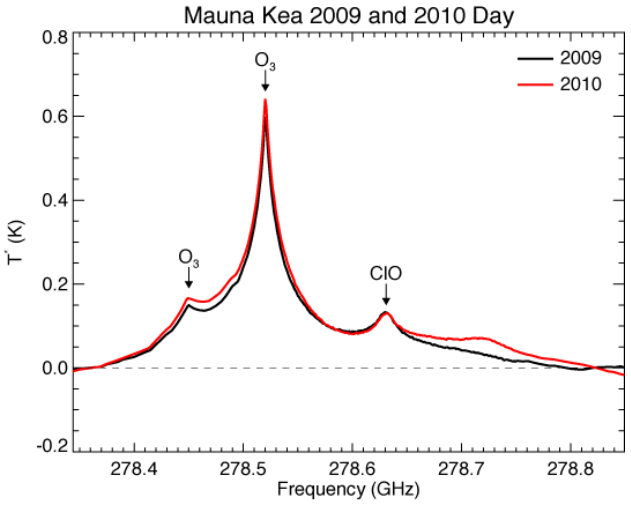

Fig. 1. Average daytime spectra in 2009 and 2010.

daytime signal, which was then used to retrieve the full daytime ClO amount.

We show here that interpolation of the nighttime spectra introduces small, variable errors, which result in increased uncertainty in the long-term trend of stratospheric ClO. Consequently, we now simply subtract night spectra from day. We have re-analyzed the entire data set from January 1992 to February 2012, and present those results here.

2 Atmospheric spectra

In June 2009, the millimeter-wavelength receiver failed. It was repaired and rebuilt at the University of Massachusetts, and returned to service in December 2009. Figure 1 shows the average daytime spectrum measured in January–June 2009 compared to that measured in the same period of 2010. In 2010, a broad spectral artifact, of apparent amplitude roughly half the ClO signal, is visible at higher frequencies, with a small but broad peak near ∼278.72 GHz. Such an artifact can easily be rationalized as the product of millimeter-wavelength standing waves internal to the receiver, and is part of what is called the instrument's spectral "baseline". The subtraction of nighttime spectra from daytime ones is intended to remove the baseline, as well as the ozone emission lines that dominate the spectral window observed.

In Fig. 2 we enlarge the spectral region near the ClO signal, for 2009 (a) and 2010 (b). In each is shown the daytime and nighttime average, and the nighttime spectrum after interpolation over the 50 MHz wide region around the ClO line frequency (which we call "interpolated night" for brevity). This "interpolated night" spectrum provides a good estimate of all spectral components besides those contributed by the ClO emission. Thus, as described in the last section, subtracting "interpolated night" from the daytime spectrum produces

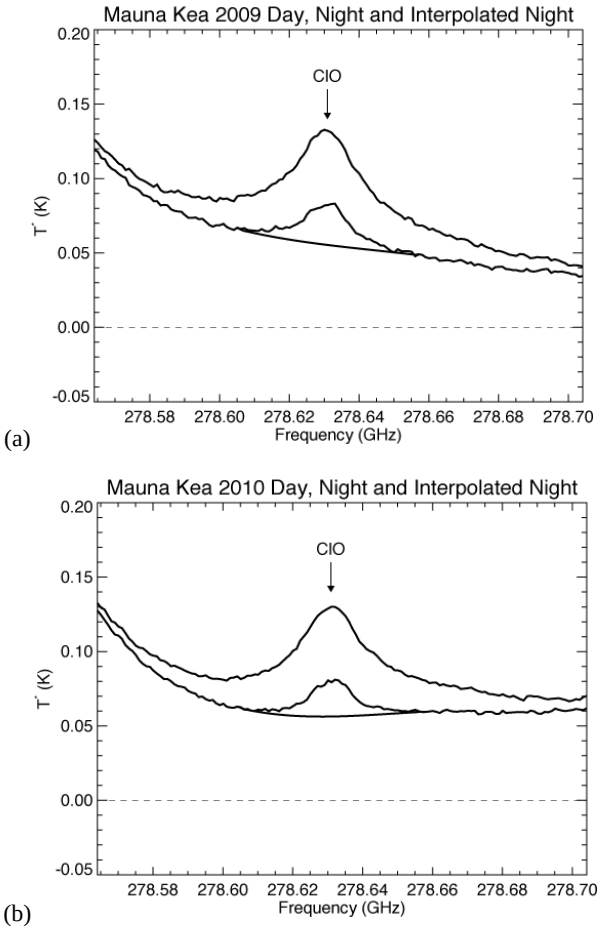

Fig. 2. Daytime, nighttime, and "interpolated nighttime" average spectra in 2009 **(a)** and 2010 **(b)**.

an approximation to the daytime ClO emission independent of other atmospheric and all instrumental signals. This technique has served its purpose well, and has been applied in numerous publications over nearly 30 years, most recently by Nedoluha et al. (2011). However, the interpolation function produces an imperfect estimate of the instrument baseline, and so when the baseline character changes, a differential error is introduced. From Fig. 2 it is clear that the background curvature in the vicinity of the ClO line is very different in 2009 and 2010, due to the spectral artifact (change in instrument baseline) seen in Fig. 1. Thus one would expect somewhat different errors in the retrieved ClO in the 2 yr.

In Fig. 3 we show the spectra for 2009 and 2010 after subtraction of the nighttime spectra. In Fig. 3a we show the spectra of day minus "interpolated night", while in Fig. 3b is seen the spectrum of day minus night, without interpolation over the nighttime signal. Examination of Fig. 3a reveals an apparent difference of approximately 5 mK, or 7 %, in the amplitude of the "day–interpolated night" ClO signal observed in 2009 and 2010. Figure 3b on the other hand shows that there

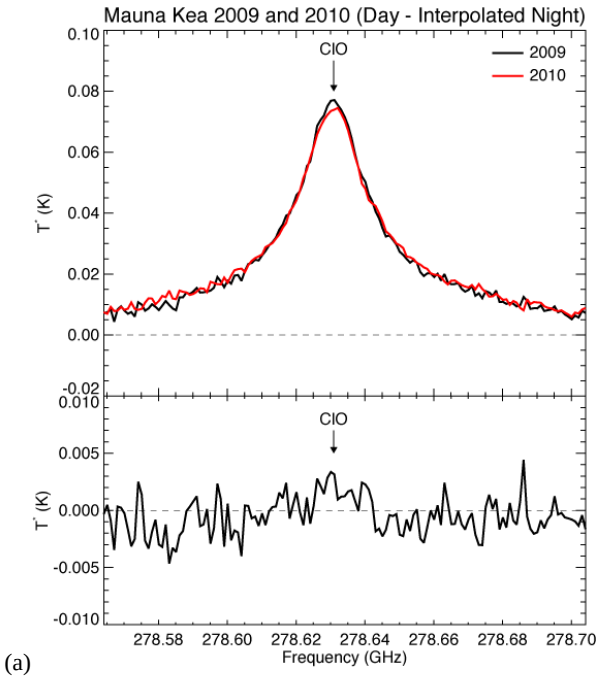

Fig. 3. Day–"interpolated night" **(a)** and day–night **(b)** average spectra for 2009 and 2010, and the difference of the 2 years.

is no difference in the "day–night" ClO signal between the 2 yr large enough to be visible relative to the spectral noise.

Comparison of Fig. 3a and b illustrates that the procedure of simply subtracting night spectra from day is less sensitive to instrumental baseline artifacts than the more sophisticated interpolation procedure heretofore used in analysis of the Mauna Kea ClO data. Therefore, we have adopted the simpler procedure, and now analyze the day–night difference spectrum, rather than attempting to estimate the daytime spectrum by subtracting "interpolated night". It is worth noting that this change to the algorithm makes it effectively identical to that used for the Scott Base ClO data. In Antarctic spring, the nighttime ClO line is highly variable and very broad in frequency; therefore we have never attempted to remove it by interpolation, and have always analyzed day–night spectra.

Further, since "day–interpolated night" spectra and altitude profiles are approximations to the daytime values, we will hereafter refer to them simply as "daytime".

3 ClO profiles

3.1 2009 vs. 2010

Retrievals from the daytime spectra of Fig. 3a suggest a significant decrease in ClO in 2010. On the other hand, retrievals from the day–night spectra in Fig. 3b show almost no change between 2009 and 2010. The difference between years is illustrated for both methods in Fig. 4. The significance of the change in daytime retrievals in 2010 is shown by the error bar in Fig. 7a, discussed below.

It would seem clear that the baseline artifact illustrated in Fig. 1 introduced a distortion of the nighttime spectrum seen in Fig. 2, resulting in a spurious difference between spectra for 2009 and 2010, as seen in Fig. 3a. That difference, and thus the spectral artifact, is the cause of the significant difference in ClO profiles in the 2 yr (Fig. 4).

3.2 Altitude sensitivity

The averaging kernels for the Mauna Kea ClO retrieval are shown in Fig. 5. These are effectively the same as shown in Nedoluha et al. (2011). They indicate that the sensitivity of the retrieval to the ClO profile is reasonably good from 15 to 45 km; at higher altitudes it decreases rapidly. This occurs because the observed ClO signal is in fact a group of hyperfine transitions, concentrated in a $\sim$ 10 MHz wide spectral band. The pressure broadened linewidth is 10 MHz at $\sim$ 4 hPa pressure, which is effectively the highest level where the spectrum has any sensitivity to altitude. The altitude sensitivity of the retrieval is also controlled by the linewidth, and is thus $\sim$ 4 hPa, since the measured spectral resolution is much less than the linewidth (it is 1 MHz).

The averaging kernels are identically the same for both analysis methods, because the absolute spectral sensitivity is the same, to both the daytime and day–night mixing ratios, because the spectral line observed is optically thin. But of course, the daytime and day–night difference profiles are not the same thing, and to assess the effect of switching analysis methods, there are two key questions. First, how different are the measured quantities, in particular the peak mixing ratios on which we base our trend determination? Second, does

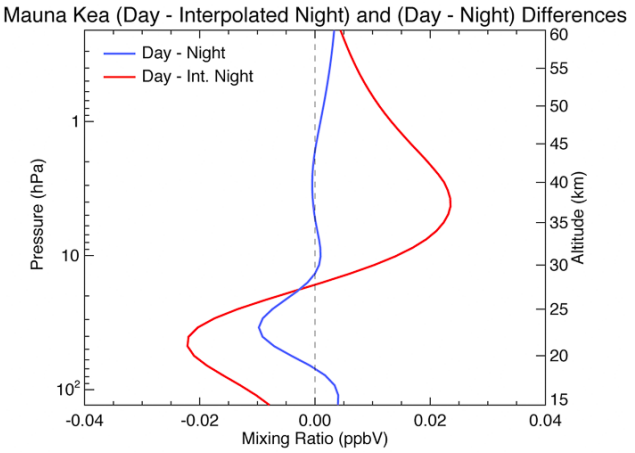

Fig. 4. Change in ClO between 2009 and 2010, with and without the revised analysis.

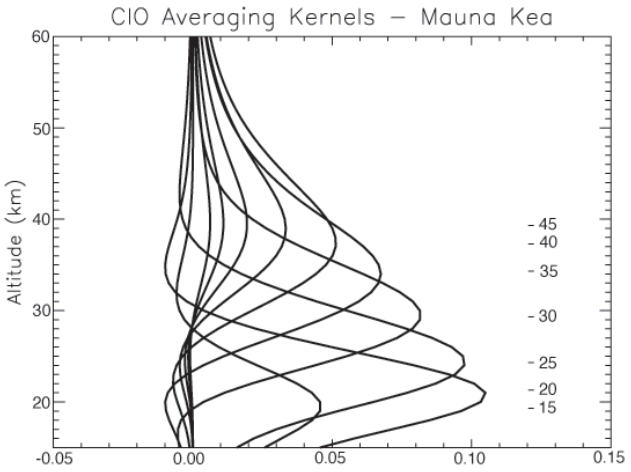

Fig. 5. The averaging kernels of the Mauna Kea ClO retrieval for selected altitudes from 15 to 60 km. The labels to the right indicate the actual peak altitude of the function for the specified altitude. The functions for 50, 55, and 60 km are also shown; their amplitudes progressively decline, while the maximum value of all 3 is at about 40 km. They are not labeled to avoid clutter.

using the day–night value improve or detract from our ability for scientific interpretation of the data record? We will address the first question in the next few paragraphs, and the second question in Sect. 3.4, below.

The effect of the nighttime profile on the day–night measurement was assessed as follows. First, a subset of the Aura Microwave Limb Sounder (MLS) data was created, including all the ClO profiles measured within ±5° latitude and ±30° longitude of Mauna Kea, between 2004 and early 2013. These are unambiguously day (local time near 13:30) or night (local time near 01:55). Daytime, nighttime, and day–night mean profiles were calculated from these data.

Second, Livesey et al. (2011) recommend using Aura ClO data at pressures of 1 hPa and more (altitudes less than about 50 km, near the stratopause). However ClO is non-negligible in the mesosphere, and as seen in Fig. 5, the microwave measurement has some sensitivity to the mesosphere. In addition, the Aura MLS mean profiles (described in the previous paragraph) have an anomalous oscillatory feature at and just above 1 hPa. For these reasons we have used the mean MLS profiles at pressures of 1.5 hPa and above, have taken representative values for ClO at lower pressures from Sato et al. (2012), and formed composite daytime and nighttime profiles extending to 0.1 hPa by interpolation.

The mean daytime and day–night profiles were convolved with the microwave averaging kernels and a priori profiles, and the mean value between 33 and 37 km was calculated from each convolved profile. This mean is defined as the "peak mixing ratio" of the day–night Mauna Kea ClO (MKO) measurements. We use it for both daytime and day–night profiles here so we can isolate the effect of subtracting the nighttime profile. The daytime and day–night mean values differ by 13 %, and we conclude that this is the extent of the effect of nighttime ClO, in the upper stratosphere and mesosphere, on the retrieved day–night values.

ClO in the mesosphere has a very different diurnal variation from that in the stratosphere, since the chemical processes operating in the two regions are rather different. To quantify that effect, we tested how the day–night measurement would change if the mixing ratio of nighttime ClO was equal to zero at all altitudes in the mesosphere. In that case, the derived day–night peak mixing ratio value would increase by ∼ 3 %.

It is also worth considering the implications for trend determination. Mesospheric ClO could well have a different trend from stratospheric ClO. It is influenced by CH4 and total chlorine, while nighttime upper stratospheric ClO depends on NO2 as well. (Sato et al., 2012). However the fact that complete removal of mesospheric ClO affects our day–night peak mixing ratio by only ∼ 3 % indicates that any differential trend in the mesosphere would have a similarly small impact on our trend computation.

We note in passing that for comparison of our results to models and to other available measurements, in particular MLS and SMILES, it is a simple matter to compute the convolved day–night profile from the other data set, and the said convolved profile is directly equivalent to the ground-based measurement. In the following section we make such a comparison to MLS.

3.3 Comparison to MLS Instruments

The revised Mauna Kea ClO profiles (MKO) agree reasonably well with measurements in a similar geographic region by Upper Atmosphere Research Satellite (UARS) and Aura MLS instruments. The following uses v5 of the UARS MLS data, and v3.3 of Aura MLS. In Fig. 6a is shown the mean

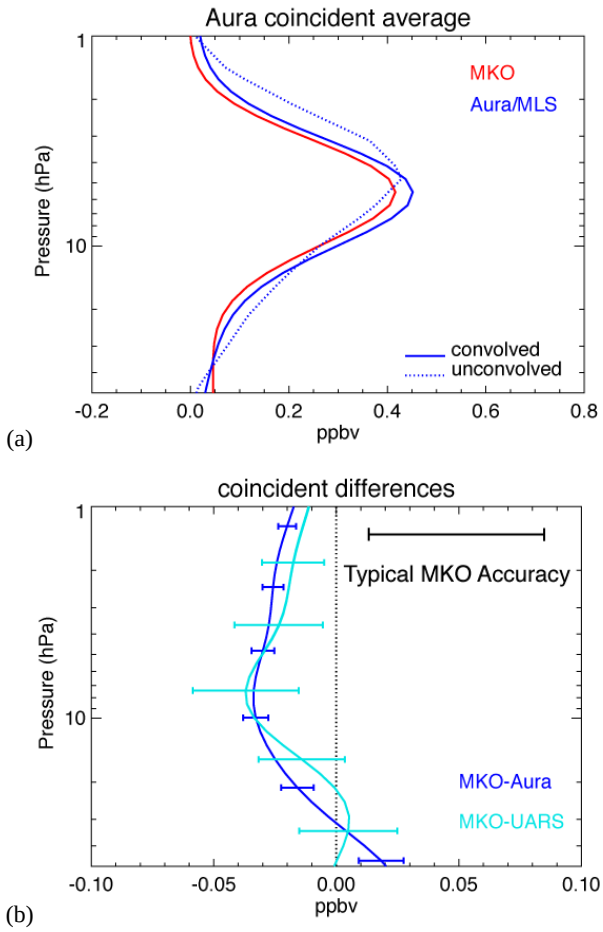

Fig. 6. Comparison of revised Mauna Kea ClO to that from Aura and UARS MLS.

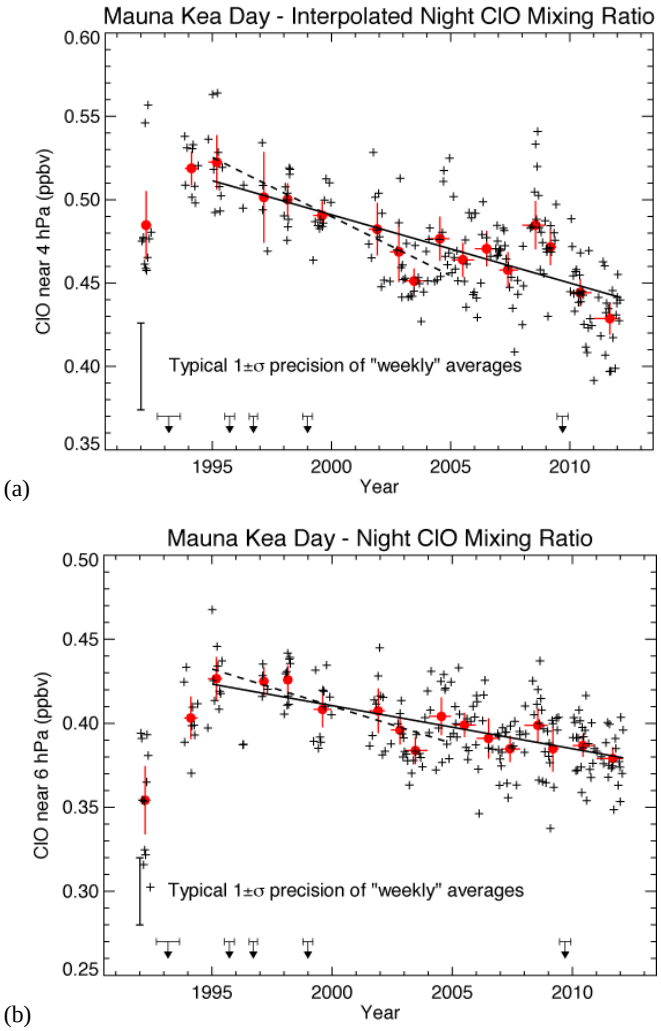

Fig. 7. Time series of peak ClO mixing ratios over Mauna Kea, 1992–2012. Data in **(a)** are derived from daytime spectra, while for data in **(b)** simple day–night spectra are used. The dashed lines show regression fits to data from 1995 to 2004, while solid lines are regression for 1995–2012.

of 118 coincident Aura and Mauna Kea ClO profiles. For Aura, the original and "convolved" profiles (combined with the ground-based averaging kernels and a priori profiles; e.g., Connor et al., 2007) are shown. In Fig. 6b, the MLS–MKO mean difference is shown, for the 118 Aura profiles and for 24 UARS profiles. The smaller number of UARS coincidences is the result of both sparser UARS data and sparser ground-based data during the UARS years (see Fig. 4 of Nedoluha et al., 2011).

The error bars reflect the observed scatter in the actual comparisons; the estimated typical accuracy for MKO (Solomon et al., 2006) is shown separately; the MLS–MKO difference is everywhere less than the MKO accuracy. These comparisons of MLS to the revised MKO data are slightly improved relative to the previous version of the MKO data, which made use of daytime MKO profiles (Nedoluha et al., 2011). The scatter is very similar, while the mean difference is marginally smaller and also less oscillatory. The uncertainty in the UARS–MKO vs. Aura–MKO comparison is essentially unchanged from that discussed in Nedoluha et al. (2011); thus their conclusion stands that the Mauna Kea observations suggest that UARS and Aura ClO can be used as a single data set without correction.

3.4 Secular trend

In Figure 7, we show time series of the ClO peak mixing ratio, from 1992 to 2012. Values shown are averages over a 5 km wide range near the peak of their altitude profile, for daytime (Fig. 7a) and day–night (Fig. 7b). Seasonal variations, of 3- to 12-month periods, have been separately derived and subtracted from each data set. Measurements averaged over periods of about one week are shown as "+" symbols, while large red dots are averages over periods of about one year. For convenience these are called "weekly" and "annual" averages, respectively.

Table 1. Linear trend of Mauna Kea ClO observations.

Day–night	(33–37 km)*
1995–2004	-1.08 ± 0.40 % yr^{-1}
2001[1]–2008	-0.32 ± 0.48
2004[2]–2012	-0.79 ± 0.40
1995–2012[3]	-0.64 ± 0.15
Daytime	(35–39 km)
1995–2004	-1.45 ± 0.38 % yr^{-1}
2001[1]–2008	$+0.22 \pm 0.44$
2004[2]–2012	-1.45 ± 0.48
1995–2012[3]	-0.86[4] ± 0.17

* Uncertainties are 2σ (twice the standard deviation of the derived trend); [1] July 2001 following extended instrument repair; [2] August 2004 to coincide with Aura operations; [3] last date is 18 February 2012; [4] a value for 1995–2010 (-0.73) was incorrectly reported in the discussion paper.

Both Fig. 7a and b show the values near the peak of the relevant ClO mixing ratio profile; however the pressure level of this peak is different for the two data sets because the nighttime ClO peak is at higher altitudes than the daytime ClO peak. Hence, the daytime ClO peak is at higher altitudes than the daytime ClO minus nighttime ClO peak. It is clear by inspection that the day–night time series is less scattered and more self-consistent, on both short and longer timescales, and appears consistent with a steady linear decrease in annual-average ClO mixing ratio over the period 1995–2012.

Table 1 shows trends in the MKO time series, derived as follows. Measurements are averaged over periods of about one week, as described above. The mixing ratio profiles retrieved from these measurements are then averaged between 33–37 km for day–night and 35–39 km for daytime. These two ranges were chosen to represent the typical altitude of the peak mixing ratio of the day–night and daytime profiles, ± 2 km. The resulting values are the "peak mixing ratios" plotted with symbol + in Fig 7. The time series of weekly peak mixing ratios is then fit, by simple linear regression, over a selected time period, with a function including a constant, linear slope, and sine and cosine functions with periods of 3, 4, 6, and 12 months. The linear slope is then quoted as the "trend", and its standard deviation is the basis for the trend's uncertainty. In previous publications, we followed our past standard practice of reporting the 1σ uncertainty. However, for ease of comparison to other published estimates, we have quoted a 2σ uncertainty in the following tables, and will adopt that practice uniformly in this paper.

Questions have been raised as to whether the transition from estimated daytime retrievals to day–night does in fact improve the trend determination. To address them, we will first compare trends over several time periods from the two analysis methods, then compare those to Aura measurements in the vicinity of Hawaii, and finally to other selected published values.

Three of the time periods selected were chosen for comparison to other results. The period 1995–2004 was reported in Solomon et al. (2006); 2001–2008 is used by Jones et al. (2011), and August 2004 to February 2012 is for comparison to Aura data. Several notable features are worthy of comment.

Daytime annual averages would suggest that there have been significant variations (as shown by the error bars) in ClO on timescales of a few years, for example the higher values in 2008–2009. The day–night annual averages, on the other hand, have both smaller error bars, reflecting less short-term variability, and show no significant departure from a linear decrease over the full period. Values slightly lower than average in 2003–2004 cause trends derived from the 1995-2004 period to be larger (more negative) than trends from the full period, for both data sets. The values in Table 1, from both time series, are consistent with the value of -0.9 ± 0.2 % yr^{-1} for 1995–2007, which was based on the daytime data, and reported in WMO (2011).

We note in passing that the "annual" point in mid-2003 is the only one from the day–night series that is more than 1σ from the linear trend line. It is slightly more than 2σ from the line and may indicate a real, relatively short-lived variation in mid-stratospheric ClO.

The daytime retrievals produce trend estimates that vary much more than the corresponding day–night retrievals. Also, while the error bars of the two analyses overlap for the full period (1995–2012), the uncertainty of the fit to the day–night data is somewhat smaller. Both of these factors suggest the day–night analysis is more stable than using the estimated daytime values. On the other hand, as discussed in Sect. 3.2, if the day–night trend is taken as a proxy for the daytime trend, a small bias, of up to 3 %, may be introduced. For this reason, we compare our results to Aura day–night trends in Sect. 3.5, below.

3.5 Comparisons to Aura inferred trend

The linear trend inferred by Aura has been calculated from the original, unmodified Aura profiles, selected in the region of Hawaii, by convolving them with the ground-based averaging kernels and a priori, and averaging them over altitude to derive the "peak mixing ratio" as described above. They are shown in Table 2 and are directly comparable to the "2004–2012" figures of Table 1.

The day–night values agree within their uncertainties ($-0.48\,4 \pm 0.08$ % yr^{-1} vs. -0.79 ± 0.40 % yr^{-1}), while the daytime values suggest a significant discrepancy (-0.56 ± 0.08 % yr^{-1} vs. -1.45 ± 0.48 % yr^{-1}). It is our view that this apparent discrepancy is an artifact of the Mauna Kea "daytime" estimate, and we take the better agreement of the day–night values between Mauna Kea and Aura

Table 2. Linear trend of Aura ClO convolved with Mauna Kea averaging kernels*.

Day–night	$-0.48 \pm 0.08\,\%\,\mathrm{yr}^{-1}$
Daytime	$-0.56 \pm 0.08\,\%\,\mathrm{yr}^{-1}$

* August 2004 to 18 February 2012; uncertainties are 2σ.

Table 3. HCl trends.

Mauna Loa[1]	Total column	2000–2009	$-0.39 \pm 0.19\,\%\,\mathrm{yr}^{-1}$
Izana[1]	Total column	2000–2009	-0.66 ± 0.15
Tropics[2]	35–45 km	1997–2008	-0.58 ± 0.17
Global[3]	Upper stratosphere	2004–2010	-0.6 ± 0.1

[1] Kohlhepp, 2012; [2] Jones et al., 2011; [3] MLS, WMO 2010, Table 1–13

as further validation of the analysis procedure introduced in this paper.

3.6 Comparisons to published results

There is a fairly limited selection of published results that are directly comparable to the Mauna Kea ClO measurements. To the best of our knowledge, the only published long-term ClO trend estimate is in Jones et al. (2011). The authors have performed a sophisticated analysis that allows them to combine different satellite data sets. In the case of ClO, they have published a trend in the 35–45 km altitude region using a data set that combines Aura MLS with the Odin Sub-millimeter Radiometer. We note that our 45 km averaging kernel peaks near 40 km (Fig. 5); hence a direct comparison with the Jones et al. (2011) altitude ranges is not possible. For the period 2001–2008, they find a trend of -0.71 ± 0.78 $\%\,\mathrm{yr}^{-1}$. The comparable number for day–night at Mauna Kea, from Table 1, is -0.32 ± 0.48; thus the two central values are consistent. The daytime estimate of $+0.22 \pm 0.44$ from Table 1 differs from the Jones et al. (2011) value by approximately 3σ. We believe this is another point validating our day–night analysis. As an aside, we note that it is difficult to assess the significance to the fact that the Jones et al. (2011) trend uncertainty is formally larger than the Mauna Kea uncertainty. They have used two independent data sets measured at different local times, and applied a model-based homogenization to the data sets, before deriving the trend and its uncertainty. The difference in the data sets and the technique used to combine them will no doubt affect the trend uncertainty, but we cannot assess its quantitative effect.

Comparison of the ClO trend to other chlorine species is certainly possible (e.g., WMO 2010, Table 1–13), and informative about the evolution of stratospheric chemistry, but complicated by dependencies on the concentration of CH4 and NO2 (Jones et al., 2011). Thus its value as validation for a given technique is debatable.

In the interest of rounding out the context of our results, we show recent published trends in stratospheric HCl in Table 3. It is beyond the scope of this study to compare these HCl trends to our measured ClO trends quantitatively. We note that "measured ClO trends are not directly comparable to changes in total stratospheric chlorine" (WMO, 2011). Nedoluha et al. (2011) used HALOE CH4 to estimate differences in HCl and ClO trends prior to 2005. It would be interesting in a future study to combine HALOE and subsequent observed changes in CH4, which has continued to vary substantially (Kohlhepp, 2012), to compute the chlorine change implied by the observed ClO changes at Mauna Kea over selected time periods, and in turn compare that to total stratospheric chlorine, represented by the sum HCl + ClONO2.

4 Summary and conclusions

The failure and repair, in 2009, of the ClO millimeter-wave receiver at Mauna Kea, Hawaii, has led to a refined understanding of the long-term ClO record at that site. After repair, we observed what seemed to be an unexpected decrease in mid-stratospheric ClO. We have shown that this apparent change was due to a change in the instrument's spectral baseline, which was not fully corrected by the analysis procedure heretofore used routinely. The procedure in question attempted to retrieve the daytime ClO mixing ratio profile by subtracting a spectral baseline from which the nighttime ClO signal had been removed. The removal process caused small errors whose character changed as the system baseline itself changed. We have further shown that direct subtraction of the nighttime spectrum from the daytime one is largely free of this error.

In response to our new understanding of the effects of the analysis procedures, we reprocessed the entire Mauna Kea ClO data set, extending from 1991 to 2012. The new data set has less scatter than the earlier one, and the ClO exhibits a more constant and linear decrease over the entire period. Comparison of the reprocessed data to Aura MLS shows substantially better agreement in the 2004–2012 trend, and a marginal improvement in the profile comparisons. Consequently we have adopted the "day–night" subtraction as the new standard procedure, and have replaced the Mauna Kea ClO in the Network for the Detection of Atmospheric Composition Change (NDACC) database with the reprocessed version.

Using the reprocessed data set and the full 17 yr of measurements from early 1995 to early 2012, we report the long-term trend in stratospheric ClO of $-0.64\,\% \pm 0.15\,\%\,\mathrm{yr}^{-1}$ (a 2σ uncertainty in the derived trend), as shown in Table 1 and discussed further in the relevant text.

Acknowledgements. We thank the Caltech Submillimeter Observatory staff and management, and the NASA Upper Atmosphere Research Program, for their support of our measurements over

more than 20 yr. Research at Stony Brook University was supported by NASA Grant NNX09AF40G. Work at the Jet Propulsion Laboratory, California Institute of Technology, was done under contract with the National Aeronautics and Space Administration.

Edited by: G. Stiller

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