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# Global OZone Chemistry And Related Datasets for the Stratosphere (GOZCARDS): methodology and sample results with a focus on HCl, $H_2O$, and $O_3$

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## Abstract

We describe the publicly available dataset from the Global OZone Chemistry And Related Datasets for the Stratosphere (GOZCARDS) project, and provide some results, with a focus on hydrogen chloride (HCl), water vapor ($H_2O$), and ozone ($O_3$). This dataset is a global long-term stratospheric Earth System Data Record (ESDR), consisting of monthly zonal mean time series starting as early as 1979. The data records are based on high quality measurements from several NASA satellite instruments and ACE-FTS on SCISAT. We examine consistency aspects between the various datasets. To merge ozone records, the time series are debiased by calculating average offsets with respect to SAGE II during periods of measurement overlap, whereas for other species, the merging derives from an averaging procedure based on overlap periods. The GOZCARDS files contain mixing ratios on a common pressure/latitude grid, as well as standard errors and other diagnostics; we also present estimates of systematic uncertainties in the merged products. Monthly mean temperatures for GOZCARDS were also produced, based directly on data from the Modern-Era Retrospective analysis for Research and Applications (MERRA).

The GOZCARDS HCl merged product comes from HALOE, ACE-FTS and (for the lower stratosphere) Aura MLS data. After a rapid rise in upper stratospheric HCl in the early 1990s, the rate of decrease in this region for 1997–2010 was between 0.4 and 0.7 $\%\,yr^{-1}$. On shorter timescales (6 to 8 years), the rate of decrease peaked in 2004–2005 at about 1 $\%\,yr^{-1}$, and has since levelled off, at $\sim 0.5\,\%\,yr^{-1}$. With a delay of 6–7 years, these changes roughly follow total surface chlorine, whose behavior vs. time arises from inhomogeneous changes in the source gases. Since the late 1990s, HCl decreases in the lower stratosphere have occurred with pronounced latitudinal variability at rates sometimes exceeding 1–2 $\%\,yr^{-1}$. There has been a significant reversal in the changes of lower stratospheric HCl abundances and columns for 2005–2010, in particular at northern midlatitudes and in the deep tropics, where short-term increases are observed. However, lower stratospheric HCl tendencies appear to be

reversing after about 2011, with (short-term) decreases at northern midlatitudes and some increasing tendencies at southern midlatitudes.

For GOZCARDS $H_2O$, covering the stratosphere and mesosphere, the same instruments as for HCl are used, along with UARS MLS stratospheric $H_2O$ data (1991–1993). We display seasonal to decadal-type variability in $H_2O$ from 22 years of data. In the upper mesosphere, the anti-correlation between $H_2O$ and solar flux is now clearly visible over two full solar cycles. Lower stratospheric tropical $H_2O$ has exhibited two periods of increasing values, followed by fairly sharp drops, the well-documented 2000–2001 decrease, and another recent decrease in 2011–2013. Tropical decadal variability peaks just above the tropopause. Between 1991 and 2013, both in the tropics and on a near-global basis, $H_2O$ has decreased by ~5–10 % in the lower stratosphere, but about a 10 % increase is observed in the upper stratosphere and lower mesosphere. However, recent tendencies may not hold for the long-term, and the addition of a few years of data can significantly modify trend results.

For ozone, we used SAGE I, SAGE II, HALOE, UARS and Aura MLS, and ACE-FTS data to produce a merged record from late 1979 onward, using SAGE II as the primary reference for aligning (debiasing) the other datasets. Other adjustments were needed in the upper stratosphere to circumvent temporal drifts in SAGE II $O_3$ after June 2000, as a result of the (temperature-dependent) data conversion from a density/altitude to a mixing ratio/pressure grid. Unlike the 2 to 3 % increase in near-global column ozone after the late 1990s reported by some, GOZCARDS stratospheric column $O_3$ values do not show a recent upturn of more than 0.5 to 1 %; continuing studies of changes in global ozone profiles, as well as ozone columns, are warranted.

A brief mention is also made of other currently available, commonly-formatted GOZCARDS satellite data records for stratospheric composition, namely those for $N_2O$ and $HNO_3$.





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# 1 Introduction

The negative impact of anthropogenic chlorofluorocarbon emissions on the ozone layer, following the early predictions of Molina and Rowland (1974), stimulated interest in the trends and variability of stratospheric ozone, a key absorber of harmful ultraviolet radiation. The discovery of the ozone hole in ground-based data records (Farman et al., 1985) and the associated dramatic ozone changes during Southern Hemisphere winter and spring raised the level of research and understanding regarding the existence of new photochemical processes (see Solomon, 1999). This research was corroborated by analyses of aircraft and satellite datasets (e.g., Anderson et al., 1989; Waters et al., 1993), and by independent ground-based data. Global total column ozone averages in 2006–2009 were measured to be smaller than during 1964–1980 by $\sim 3\,\%$, and larger more localized decreases over the same periods reached $\sim 6\,\%$ in the Southern Hemisphere midlatitudes (WMO, 2011). Halogen source gas emissions have continued to decrease as a result of the Montreal Protocol and its amendments. Surface loading of total chlorine peaked in the early 1990s (WMO, 2011), and subsequent decreases in global stratospheric HCl and ClO have been measured from satellite-based sensors (Anderson et al., 2000; Froidevaux et al., 2006; Jones et al., 2011) as well as from the ground (e.g., Solomon et al., 2006; Kohlhepp et al., 2012). A slow recovery of the ozone layer is expected between the late 1990s and several decades from now, towards pre-1985 levels (WMO, 2011); the robust determination of a long-term global trend requires a sufficiently long and accurate data record. It is desirable to use high quality datasets for ozone and related stratospheric species for a robust documentation of past variations and as constraints for global atmospheric models.

The history of global stratospheric observations includes a large suite of satellite-based instruments, generally well-suited for the elucidation of long-term global change. A review of differences between past and ongoing satellite measurements of atmospheric composition has been the focus of the Stratosphere–troposphere Processes And their Role in Climate (SPARC) Data Initiative (DI); results for stratospheric water



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vapor and ozone intercomparisons have been published by Hegglin et al. (2013) and Tegtmeier et al. (2013), respectively, to be followed by a larger report on intercomparisons of multiple species. Systematic biases reported in these recent papers tend to mirror past validation work. However, these investigations have not pursued data merging aspects or the creation of long-term records.

Under the Global OZone Chemistry And Related Datasets for the Stratosphere (GOZCARDS) project, we have created monthly zonally averaged datasets of stratospheric composition on a common latitude/pressure grid, using satellite-based limb viewing instruments launched as early as 1979 (for ozone data in particular) and now continuing with instruments launched about a decade ago. The creation of this Earth System Data Record stays close to the data values themselves. Therefore, spatial or temporal gaps are typically not filled in; various methods can be used to try to produce continuous fits to time series, but we viewed this as being outside the scope of this data record creation. The GOZCARDS products arise from several high quality satellite datasets, namely from Stratospheric Aerosol and Gas Experiment instruments (SAGE I and SAGE II), the Halogen Occultation Experiment (HALOE) which flew aboard the Upper Atmosphere Research Satellite (UARS), the UARS Microwave Limb Sounder (MLS), the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) on SCISAT, and the Aura MLS experiment. Table 1 provides characteristics of the original datasets; validation papers from the instrument teams and other related studies give a certain degree of confidence in these datasets. However, the existence of validation references does not imply that there are no caveats or issues with a particular measurement suite. In this project, we have strived to optimize data screening and mitigate some undesirable features, such as the impact of outlier values or the effects of clouds or aerosols. All source datasets still have shortcomings or imperfections, but we have refrained from arbitrarily removing specific monthly means.

Based on original profiles from the various instruments, GOZCARDS “source” monthly zonal mean values were derived. After data screening, monthly average profiles were created by vertical interpolation onto the GOZCARDS pressure levels, followed by binning and averaging into monthly sets. In order to accomodate the lower vertical resolution of some limb viewers, such as UARS MLS, the GOZCARDS pressure grid was chosen as

$$p(i) = 1000 \times 10^{-i/6} \text{ (hPa)} \tag{1}$$

with $i$ varying from 0 to a product-dependent top; this grid width corresponds to $\sim 2.7$ km. The high resolution SAGE $O_3$ profiles were smoothed vertically onto this grid (see Sect. 5). Given the sampling of solar occultation instruments, which typically provide 15 sunrise (SR) and 15 sunset (SS) profiles every day (vs. the emission-based sampling from MLS), we used latitude bins of width 10° (18 bins from 80–90° S to 80–90° N) to construct the monthly zonal means.

After the production of GOZCARDS source data files on the above grid, merged (combined) products were created. This involves the calculation of average biases between monthly zonal means from different source data during periods of overlap, followed by an adjustment (using calculated average offsets) of the time series. Non-zero biases always exist between datasets from different instruments for various reasons, such as systematic errors arising from Level 1 (radiances) or Level 2 (retrievals), different vertical resolutions, or sampling effects. A useful reference regarding the sampling effects, which can arise spatially (within a latitude bin) or temporally (within a month), is the recent work by Toohey et al. (2013). They studied sampling biases from a large suite of satellite-based stratospheric profiling instruments, based on simulations using fully-sampled model abundance averages vs. averaged sampled results from suborbital track locations. The magnitude of such sampling errors is typically inversely related to the number of available profiles routinely sampled, so that larger sampling errors arise from occultation than from emission measurements, which often sample thousands of profiles per day. Toohey et al. (2013) found that sampling-related biases can reach 10–15 % in some regions/periods, notably at high latitudes when larger atmospheric variability exists. Sofieva et al. (2014) have also discussed sampling uncertainty issues for satellite ozone datasets.





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We have observed very good correlations between GOZCARDS ozone and other long-term ozone datasets, such as the Stratospheric Water vapor and OzOne Satellite Homogenized (SWOOSH) dataset (Davis et al., personal communication, 2012) and homogenized Solar Backscatter Ultraviolet (SBUV) data; these analyses (along with related work on $H_2O$) will be discussed elsewhere. Results from GOZCARDS and other data relating to midlatitude ozone trends have appeared (e.g., Nair et al., 2013). Dissemination of trend results arising from analyses of GOZCARDS and other ozone profile data is planned as part of the $SI^2N$ initiative, which stands for Stratospheric Processes And their Role in Climate (SPARC), International Ozone Commission (IOC), Integrated Global Atmospheric Chemistry Observations (IGACO-$O_3$), and the Network for the Detection of Atmospheric Composition Change (NDACC). Recent results on such ozone profile trend comparisons can be found in Tummon et al. (2014) and Harris et al. (2015).

This paper starts with a discussion of general data screening issues (Sect. 2), and then describes the GOZCARDS data production approach and methodology, followed by some atmospheric results for HCl (Sect. 3), $H_2O$ (Sect. 4), and $O_3$ (Sect. 5). We provide specific diagnostics that indicate generally good correlations and small relative drifts between the main datasets being used to create the longer-term GOZCARDS merged time series. Section 6 briefly mentions the availability of a few other GOZCARDS products, namely $N_2O$, $HNO_3$, and temperatures derived from MERRA fields. The version of GOZCARDS described here is referred to as ESDR version 1.01 or ev1.01. Each product's public GOZCARDS data record has an associated digital object identifier (DOI) along with a relevant dataset reference.

## 2 GOZCARDS source data and data screening

Data provenance information regarding the various measurements used as inputs for GOZCARDS is provided in Appendix A.1.

## 2.1 GOZCARDS data screening and binning

The screening of profiles for GOZCARDS has largely followed guidelines recommended by the various instrument teams and/or relevant publications; such screening procedures are rarely all described in one convenient location, so we review this briefly here for the various data sets. Data screening can reduce the total number of good profiles below our chosen threshold for flagging zonal monthly means; unless otherwise noted, we only provide monthly means constructed from 15 or more values in a given latitude/pressure bin.

For HALOE, cloud contamination may add retrieval artifacts and HALOE profiles were screened for clouds, following procedures described in Hervig and McHugh (1999); values at and below the cloud level (found in the netCDF files) were excluded. Also, HALOE profiles that may occasionally contain artifacts associated with either a faulty trip angle or constant lockdown angle registration were screened out, per recommendations from the HALOE data processing team (see http://haloe.gats-inc.com/user_docs/index.php for details).

For UARS MLS, we used screening recommendations documented by Livesey et al. (2003). In particular, MLS data points whose retrieved precisions are flagged with a negative sign were discarded, ensuring only a negligible contribution to the retrieval from a priori. Also, our data filtering followed the recommendations regarding the "MMAF_STAT" flag for operational status (we only used values of "G", "t", or "T" for this flag) and the product-specific "QUALITY" flag (for which we only used values equal to 4, thus eliminating bad radiance fits).

For Aura MLS data screening, the procedures are generally as follows: we only use profiles with even values of the Status field, Quality values larger than documented thresholds (indicating good radiance fits), Convergence values smaller than documented thresholds (indicating good convergence), and positive (unflagged) values of the estimated precisions. Species-specific validation papers give data screening rec-



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ommendations, with appropriate flag values for Quality and Convergence; see Livesey et al. (2013) for such references and v3.3 data screening updates.

For ACE-FTS, a list of profiles with data issues is provided by the ACE-FTS team (see https://databace.scisat.ca/validation/data_issues_table.php) and these have been removed from our database. However, we also found it necessary to remove occasional large outlier values that could significantly impact monthly zonal means by adding a bias (or "noise") to the time series if such screening were not performed; such outliers are not otherwise routinely removed from the original ACE-FTS profiles. Our outlier screening procedure removed values outside 2.5 times the SD, as measured from the median values in each latitude/pressure bin, for each year of data. This was deemed close to optimum by comparing the results to Aura MLS time series (which usually are not impacted by such outliers), as well as to independent zonal means (using 5° latitude bins) provided by the ACE-FTS instrument team. Up to 5 % of the profile values in each bin in any given month were typically discarded as a result of this procedure, but the maximum percentage of discarded values can be close to 10 % for a few months of ACE-FTS version 2.2 data, depending on year and species. Moreover, because of poor ACE-FTS sampling in the tropics, the threshold value for minimum number of (good) ACE-FTS profiles determining a monthly zonal average was allowed to be as low as 10 for mid- to high latitudes, and as low as 6 for low latitudes (bins centered from 25° S to 25° N). Our zonal mean datasets for ACE-FTS would become too sparse in some years if such lower threshold values were not used; such data sparseness can introduce limitations (larger error bars) in the determination of trends, for example, although we also performed comparisons vs. Aura MLS monthly means to provide some level of confidence in the results. In addition, ACE-FTS single profile values were discarded if the associated error was larger than the mixing ratio or smaller than $10^{-4}$ times the mixing ratio, following recommendations from the ACE-FTS team.

The binning of profiles occurs after the screened values are averaged (in each latitude/pressure bin). Negative monthly means have been flagged (set to −999.0) in the GOZCARDS files; while a negative mixing ratio that is smaller (in absolute value) than



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its associated standard error (or a few times this standard error) can in theory be meaningful, we deem that occasional small negative monthly means are unlikely to be very useful, scientifically.

The organization of profiles on a common pressure grid is straightforward when pressure values are present in the original files, as is the case for most data used here. Also, the vertical resolutions are similar for most of the instruments used for GOZCARDS (see more details in each species-specific section). The UARS MLS, HALOE, and Aura MLS native pressure grids are either the same as or a superset of the GOZCARDS pressure grid, so these datasets were readily sampled for the construction of the GOZCARDS monthly means. For ACE-FTS profiles, pressures are provided along with the fixed altitude grid, and we used linear interpolation vs. log(pressure) to convert profiles to the GOZCARDS grid. More details are provided in the $O_3$ section for SAGE I and SAGE II, for which density vs. altitude is the native representation.

## 3 GOZCARDS HCl

### 3.1 GOZCARDS HCl source data records

We used HCl datasets from HALOE, ACE-FTS and Aura MLS to generate the monthly zonal mean source products for GOZCARDS HCl.

For the screening of HALOE HCl profiles, in addition to the procedures mentioned in Sect. 2, a first-order aerosol screening was applied: all HCl values at and below a level where the 5.26 µm aerosol extinction exceeds $10^{-3}$ km$^{-1}$ were excluded.

For Aura MLS, the ongoing standard HCl product is retrieved using band 14 rather than band 13, which was used to measure HCl for the first 1.5 years after launch, but started deteriorating rapidly after February 2006. Validation and error characterization for the Aura MLS HCl product (version 2.2) were provided by Froidevaux et al. (2008). The MLS version 3.3/3.4 HCl data used here (see Livesey et al., 2013) compare quite well with v2.2, with average biases within 5 % in general. A high bias exists in MLS





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HCl vs. aircraft data at 147 hPa at low latitudes (Froidevaux et al., 2008). Such regions with large uncertainties and biases are avoided (flagged) for the production of the GOZCARDS merged HCl dataset.

The use of the GOZCARDS source files for Aura MLS HCl, like the use of original Level 2 MLS HCl files, is not recommended for obtaining realistic trends in the upper stratosphere (at pressures < 10 hPa), even if monthly mean MLS HCl in this region displays reasonable values in comparison to other satellite-based measurements. Aura MLS switched to a backup band (band 14) to retrieve the daily HCl measurements after band 13 (originally targeted specifically for HCl) showed signs of rapid degradation in early 2006; as the remaining lifetime for band 13 is expected to be very short (days as opposed to weeks), this band has only been turned on for a few days since February 2006. However, for pressures ≥ 10 hPa, the long-term (band 14) HCl data now being routinely produced is deemed to be robust (because of the broader emission line in this region, in comparison to the measurement bandwidth). These considerations have implications for how we treat MLS HCl upper stratospheric data in terms of the merging process.

Past validation studies have compared MLS HCl (v2.2), ACE-FTS (v2.2) and HALOE (v19) datasets using coincident pairs of profiles; such work was described by Froidevaux et al. (2008) for MLS HCl validation and by Mahieu et al. (2008) for ACE-FTS HCl validation. HALOE HCl values were found to be biased low by ∼ 10–15 % relative to both MLS and ACE-FTS, especially in the upper stratosphere; this low bias vs. other (balloon- and space-based) measurements had been noted in past HALOE validation studies (Russell et al., 1996). Also, HALOE (v19) and ACE-FTS (v2.2) HCl data tend to lose sensitivity and reliability for pressures less than ∼ 0.4 hPa.

## 3.2 GOZCARDS HCl merged data records

HCl is a good candidate for merging the main satellite data that have provided this measurement since 1991. Indeed, one can benefit from the strengths of all datasets in the lower stratosphere, but rely on HALOE and ACE-FTS for upper stratospheric trends, because of the Aura MLS HCl trend detection issue mentioned above. Aura MLS HCl time series were not included in the merging at pressures less than 10 hPa, so after November 2005, the GOZCARDS HCl upper stratospheric trends are dictated only by changes in ACE-FTS abundances. However, in order to derive the systematic offsets needed to adjust the time series from these three instruments in a continuous way (in the pressure dimension), we used the absolute Aura MLS HCl measurements at all pressure levels in 2004 and 2005, during the overlap period between the three instruments.

Figure 1 illustrates the merging process for HCl at 32 hPa for the 45° S latitude bin (which covers 40 to 50° S). Given that there exists very little overlap between the three sets of measurements in the same months in 2004 and 2005, especially in the tropics, a simple 3-way averaging of the datasets is not practical and would lead to significant data gaps. Our methodology is equivalent to averaging all three datasets during this period (if one had full coverage from all datasets), but we use Aura MLS as a transfer dataset. This was done by first averaging ACE-FTS and Aura MLS data, where the datasets overlap, and then including the third dataset (HALOE) into the merging process with the intermediate (temporary) merged data. Although HALOE HCl is believed to be biased too low, modifying the HALOE values to somehow match ACE-FTS or Aura MLS values or a combination of these two datasets was deemed to be too subjective. The combined weight of the other two datasets leads to a merged HCl dataset that is generally further away from HALOE than it is from either ACE-FTS or Aura MLS. The top left panel in Fig. 1 shows monthly zonal average GOZCARDS source data for HALOE, ACE-FTS, and Aura MLS during the overlap period, from August 2004 (when Aura MLS data started) through November 2005 (when HALOE data ended). The top right panel illustrates the result of step 1 in the merging procedure, with the temporary merged data values (orange) resulting from the adjustment of ACE-FTS and Aura MLS values to the mean reference indicated by the black dashed line; this reference is simply the average (over the overlap period) of these two datasets, formed from the average of the points which overlap during the same months, meaning whenever



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ACE-FTS obtained monthly data (since Aura MLS HCl means exist every month). The middle left panel shows the result of step 2, namely the merged values (brown) that arise from merging HALOE values with the temporary merged values (orange) from step 1. In this second averaging step, we weigh the intermediate merged values by 2/3 and HALOE values by 1/3 (leading to a mean reference illustrated by the dashed black line), in order for this process to be equivalent to averaging all three datasets, each with a weight of 1/3. The middle right panel shows the source data along with the final merged values during the overlap period. A simple mathematical description of the above procedure is provided in Appendix A. The bottom panel shows the same datasets but for 1991 through 2012, after the calculated additive offsets are applied to the whole source series, thus debiasing the datasets; these adjusted time series are then merged (averaged) together wherever overlap exists. We tested this procedure by using one or the other of the two occultation datasets as the initial one in step 1, and results were not found to differ appreciably. This methodology is used as well for the same three datasets for the $H_2O$ merging; we have also checked our procedures and results by using two independent calculations from different institutions. We also found that the use of multiplicative adjustments generally produces very similar results as additive offsets. Some issues were found on occasion with multiplicative offsets, when combining very low mixing ratios, but additive offsets can also have drawbacks if the merged values end up being slightly negative, notably as a result of changes that modify the already low HCl values during Antarctic polar winter. This occurs on occasion as additive offsets tend to be weighted more heavily by the larger mixing ratios found during non-winter seasons; as a result, we decided not to offset the lower stratospheric HCl source datasets in the polar winter seasons at high latitudes for any of the years (for interannual consistency). Procedural details regarding the merging of HCl data are summarized in the Supplement.

In Fig. 2, we display the offsets that were applied to the three HCl source datasets as a result of the merging process in each latitude/pressure bin; a positive value means that a dataset is biased low and needs to be increased (on average) by the offset value.

These offsets show that in general, ACE-FTS and Aura MLS HCl values were adjusted down by 0.1–0.2 ppbv (a decrease of about 2–10 %), while HALOE HCl was adjusted upward by 0.2–0.4 ppbv. Offset values tend to be fairly constant with latitude and the sum of the offsets equals zero. The generally homogeneous behaviour vs. latitude is a good sign, as large discontinuities would signal potential issues in the merging (e.g., arising from large variability or lack of sufficient statistics). Figure S1 provides more detailed examples of some of these (upper and lower stratospheric) offsets vs. latitude, including standard errors based on the variability in the offsets from month to month during the overlap period. Error bars in the offsets provide an indication of the results' robustness. Another indication of first-order compatibility between datasets is provided by a comparison of annual cycles. Figure S2 provides average annual cycle amplitudes obtained from simple regression model fits to HALOE, ACE-FTS, and Aura MLS series over their respective periods. While there are a few regions where noise or spikes exist (mainly for ACE-FTS), large annual amplitudes in the polar regions occur in all the time series; this arises from HCl decreases in polar winter, followed by springtime increases.

A more detailed analysis of interannual variability and trend consistency is provided from results in Fig. 3, which shows an example of ACE-FTS and Aura MLS time series. We note that no v2.2 ACE-FTS data (for any species) are used after September 2010, because of a data processing problem; a fully updated version of ACE-FTS data was not available when the GOZCARDS data records were constructed. We have used coincident points from these time series to compare the deseasonalized anomalies (middle panel in Fig. 3) from both instrument series; correlation coefficient values ($R$ values) are also computed. In the Fig. 3 example, very good correlations are obtained and no significant trend difference between the anomalies (bottom panel) is found for ACE-FTS and Aura MLS HCl. A global view for all latitude/pressure bins of these correlations and drifts is provided in Fig. 4, where the top panel gives $R$ values for deseasonalized anomalies, and the bottom panel gives the ratio of the difference trends over the error in these trends. The results in Fig. 4 confirm that there are significant trend differences between the upper stratospheric HCl time series from ACE-FTS and





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that of Aura MLS (as a reminder, we did not use Aura MLS HCl for pressures less than 10 hPa). Figure 4 also shows very low correlation coefficients from the deseasonalized HCl series in the uppermost stratosphere, because Aura MLS HCl exhibits unrealistically flat temporal behavior, whereas ACE-FTS HCl varies more. In the lower stratosphere, there is generally good agreement between the ACE-FTS and Aura MLS HCl time series, with $R$ values typically larger than 0.7 and difference trend to error ratios smaller than 1.5. The few low $R$ values for 100 hPa at low latitudes likely reflect more infrequent ACE-FTS sampling and some (possibly related) outlier data screening issues.

Figure S3 illustrates GOZCARDS merged 46 hPa HCl variations vs. time; there is clearly a much more complete global view (with no monthly gaps) after the launch of Aura MLS. Gaps at low latitudes in 1991 and 1992 are caused by post-Pinatubo aerosol-related issues in the HALOE record, and gaps in later years arise from the decrease in coverage from UARS. In the upper stratosphere, there are more gaps compared to 10 hPa and below, as a result of the much poorer tropical coverage from ACE-FTS and the elimination of MLS data in this region.

An indication of systematic errors in the merged values can be obtained by providing estimates of the range of available monthly mean source data. We have made such a calculation, although these error values are not part of the public GOZCARDS data files. For each bin, we computed the ranges of monthly means above and below the merged values that include 95 % of the available source data monthly means. These error bars are not usually symmetric about the merged values, especially if one dataset is biased significantly in relation to merged values. We did not have enough datasets here to consider a more statistical approach (such as actual SD among source datasets). Figure 5 shows the result of such a systematic error calculation at 46 hPa for the 35° S latitude bin. The lower shaded region range gives the lower bound, determined by HALOE data, and the upper limit of the grey shading originates from ACE-FTS data. Figure 6 shows contour plots of these estimated systematic errors in HCl for all latitudes and pressures. These are fairly conservative error bars; however, even the source data





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averages at the 95 % boundaries have their own systematic errors (rarely smaller than 5 %), so our estimates do not really encompass all error sources. Error bars representing a range within which 95 % of the source data values reside (see Figs. 5 and 6) can be a useful guide for data users or model comparisons; users can readily calculate such ranges (or we can provide these values).

Other quantities are provided in the netCDF GOZCARDS files, which are composed of one set of individual yearly files for all source datasets, and one set of yearly files for the merged products. The main data quantities are monthly averages, plus SD and standard errors for these means. The GOZCARDS source files also provide the number of days sampled each month as well as minimum and maximum values for the source datasets. Other information includes average solar zenith angles and local solar times for individual sources. Note that for the species discussed here, sunset and sunrise occultation values in the same latitude bin during a given month are averaged together. Finally, formulae for monthly SD of the merged data are given in Appendix A, where sample time series of the SD and standard errors (not systematic errors) for both source and merged HCl data are also shown.

### 3.3 GOZCARDS HCl sample results and discussion

Stratospheric HCl is important because it is the main reservoir of gaseous chlorine and it can be used to follow the chlorine budget evolution over the past decades. This includes a significant increase before the mid-1990s as a result of anthropogenic chlorofluorocarbon (CFC) production, followed by a slower decrease as a result of the Montreal Protocol and subsequent international agreements to limit surface emissions that were correctly predicted to be harmful to the ozone layer (Molina and Rowland, 1974; Farman et al., 1985).

In Fig. 7, we provide an overview of the HCl evolution since 1991, based on GOZCARDS average merged HCl for 3 different latitude regions at 4 pressure levels, from the upper stratosphere to the lower stratosphere. In the upper stratosphere (at 0.7 hPa shown here), the rapid early rise in HCl was followed by a period of stabilization (1997–



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2000) and subsequent decreases. The GOZCARDS HCl time series for pressures less than 10 hPa stop in September 2010 because after this, v2.2 ACE-FTS data were halted, due to technical retrieval issues with that data version. Rates of decrease for stratospheric HCl and total chlorine have been documented based on such satellite-based upper stratospheric abundances, which tend to follow tropospheric source gas trends with a time delay of order 6 years, with some uncertainties in the modeling of this time delay and related age of air issues (Waugh et al., 2001; Engel et al., 2002; Froidevaux et al., 2006). As summarized in WMO (2011), the average rate of decrease in stratospheric HCl has typically been measured at −0.6 to −0.9 % $yr^{-1}$, in reasonable agreement with estimated rates of change in surface total chlorine; see also the HCl upper stratospheric results provided by Anderson et al. (2000) for HALOE, Froidevaux et al. (2006) for the one and a half year Aura MLS data record (from the initially used primary band), and Jones et al. (2009) and Brown et al. (2011) for a combination of HALOE and ACE-FTS datasets. The WMO (2011) summary of trends also includes results from column HCl data at various NDACC Fourier transform infrared (FTIR) measurement sites; see Kohlhepp et al. (2012) for a comprehensive discussion of ground-based results, showing some scatter as a function of latitude. Figure 7 demonstrates that a global-scale decline in mid- to lower stratospheric HCl is visible since about 1997. We also notice that at 68 hPa in the tropics, the long-term rate of change appears to be near-zero or slightly positive. In addition, there are shorter-term periods in recent years when an average increasing "trend" would be inferred rather than a decrease, in particular, see the Northern Hemisphere data from 2005 through 2012 at 32 hPa.

To quantify the rates of change further, we created deseasonalized GOZCARDS merged monthly zonal mean HCl data for the different latitudes, and we show in Fig. 8 the linear rate of change that results from simple fits through such series (averaged into 20° wide latitude bins). The long-term trends (1997 through 2013 for the lower stratosphere, and 1997 through 2010 for the upper stratosphere) are generally negative and between about −0.5 % $yr^{-1}$ in the upper stratosphere and −1 % $yr^{-1}$ in the lower





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stratosphere, depending on latitude. Some separation between northern and Southern Hemisphere results is observed in the lower stratosphere, with smaller trends in the Northern Hemisphere. Also, the scatter increases for 68 to 100 hPa and some positive (or essentially zero) trends occur at low latitudes in this region; however, we have less confidence in the results at 100 hPa, given the larger scatter and error bars in that region (and the smaller abundances). Results at more polar latitudes (not shown here) tend to follow the adjacent midlatitude bin results, but with more scatter (and larger error bars), especially for shorter time periods. To explore these rates of change in the lower stratosphere in more detail, Fig. 9 shows the same type of analysis as Fig. 8 for three other time periods and for pressures of 10 hPa or more: (Fig. 9a) for a decade of data from 2003 through 2012, (Fig. 9b) for a shorter 6 year period from 2006 through 2011, and (Fig. 9c) for the most recent 6 year period from 2008 through 2013. For the results in (Fig. 9a), a decadal decrease is still observed for the Southern Hemisphere bins and some of the tropics in the upper region, but increases can be detected in the Northern Hemisphere and at the higher pressures in the tropics. In (Fig. 9b), we see an accentuation of this hemispheric asymmetry in the short-term rates of change, with large positive changes in the Northern Hemisphere, and values on both negative and positive sides between 1 and 3 % $yr^{-1}$ in many cases; during this past decade, this 6 year period (2006–2011) is near the temporal peak of this asymmetric lower stratospheric behavior. In the most recent 6 year period, however (see Fig. 9c), the rates of change have decreased for all the latitude bins shown, with all results from 10 to 68 hPa under 0.5 to 1 % $yr^{-1}$ (absolute value). Without assigning an exact linear "trend" from these simple analyses, we illustrate here that there is considerable variability in lower stratospheric HCl short-term behavior, especially after 2005. Such lower stratospheric changes in HCl have been captured in column HCl FTIR data, as demonstrated by Mahieu et al. (2013, 2014). In the latter reference, it is shown that total column (FTIR) results and GOZCARDS lower stratospheric HCl trends agree quite well; also, these authors' analyses imply that a relative slowdown in the northern hemispheric circulation is responsible for these observed recent changes in the lower stratosphere. However,





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we note from Fig. 7, that such changes in lower stratospheric HCl appear to be fairly short-term in nature, with an apparent reversal in behavior occurring at both northern and southern midlatitudes since 2011 (e.g., at 32 hPa). The lower stratospheric changes are distinct from the upper stratospheric long-term decrease, which we expect to continue, as long as the Montreal Protocol agreements are fulfilled worldwide and total surface chlorine emissions keep decreasing.

The rate of change analyses above were repeated and shown in Fig. 10 for sliding time periods centered on different years (e.g., a 6 year average for 2004 means an average from 2001 through 2006) in the upper and lower stratosphere for various latitude bins (covering 50° S to 50° N in 10° steps). As observed in Fig. 10a, the sliding 6 year results indicate that there has been an acceleration in the rate of decrease of upper stratospheric HCl between 2000 and 2004, followed by a flatter period until 2010 (this being the last year of GOZCARDS data available for the upper stratosphere, with a 6 year period centered at the start of 2008). The rate of upper stratospheric HCl change reached a maximum close to $-1\,\%\,yr^{-1}$, and has retreated to values near $-0.5\,\%\,yr^{-1}$ in more recent years. This is roughly in agreement with time-shifted curves showing the rates of change for surface total chlorine based on National Oceanic and Atmospheric Administration (NOAA) surface data (Montzka et al., 1999), as shown in Fig. 10 (upper panels, green and purple curves) with the Earth System Research Laboratory Global Monitoring Division (website) data, time shifted by 6 or 7 years to approximately account for transport delays into the upper stratosphere. The tropospheric source gases for chlorine have also shown a reduction in the rate of decrease during the 2nd half of the past decade, as discussed by Montzka et al. (1999) and summarized more recently in WMO (2011). As discussed in the latter report, this arises from a combination of factors, including the initial rapid decrease in methyl chloroform (which now plays a much smaller role), slower rates of decrease from the sum of CFCs in more recent years, and increases in hydrochlorofluorocarbons (HCFCs), along with small contributions from very short-lived species, all of which requires continued monitoring. In Fig. 10, the lower stratospheric response is summarized (Fig. 10b)





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by considering the rates of change in partial column density between 68 and 10 hPa. The lower stratospheric rates of change show more variability with latitude than in the upper stratosphere for short (6 yr) time periods, and a hemispheric asymmetry exists, peaking in 2009, when positive tendencies are seen in the Northern Hemisphere, as opposed to decreases in the south. Figure 10c and d also displays the sensitivity to the time period chosen, as we average the different latitudinal results (from the left panels) and add 8 year sliding periods to this analysis of HCl changes. The near-global results are not too dependent on whether 6 or 8 year periods are used, but longer periods tend to smooth out the rates of change; interannual changes, including those arising from the quasi-biennial oscillation (QBO), will affect short-term results, especially in the lower stratosphere. It is worth noting (Fig. 10) that the patterns in the upper and lower stratosphere are qualitatively similar, and that rates of change in surface emissions will impact both regions, but carefully disentangling this from changes in the dynamics and in other constituents (e.g., $CH_4$) that can affect the partitioning of chlorine species will require more analyses and modeling.

## 4 GOZCARDS $H_2O$

### 4.1 GOZCARDS $H_2O$ source data records

We used water vapor datasets from HALOE, UARS MLS, ACE-FTS, and Aura MLS to generate the monthly zonal mean source products for GOZCARDS $H_2O$.

In addition to the data screening procedures mentioned in Sect. 2, screening of HALOE $H_2O$ data for high aerosol extinction values was performed, in a way very similar to the method used for the creation of merged $H_2O$ for the Stratospheric Water vapor and OzOne Satellite Homogenized (SWOOSH) dataset (S. Davis, personal communication, 2012). This method (see Fig. S4) screens out anomalous HALOE $H_2O$ values that occurred mainly in 1991–1992, when the aerosol extinction near 22 hPa exceeded $5 \times 10^{-4}$ km$^{-1}$; for pressure levels at and below 22 hPa, we have excluded the



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corresponding $H_2O$ values. Also, for upper mesospheric HALOE data used here, care should be taken during high latitude summer months, as no screening was applied for the effect of polar mesospheric clouds (PMCs). High biases (by tens of percent) in $H_2O$ above ~ 70 km have been shown to occur as a result of PMCs in the HALOE field of view (McHugh et al., 2003). Indeed, monthly mean values larger than 8–10 ppmv are observed in GOZCARDS $H_2O$ merged data and in HALOE source data for pressures less than ~ 0.03 hPa. A more recent HALOE data version (version 20), or the version labeled VPMC based on the above reference, could be used to largely correct such PMC-related effects, although this was not implemented for GOZCARDS $H_2O$. The Aura MLS and ACE-FTS measurements, obtained at longer wavelengths than those from HALOE, do not yield such large $H_2O$ values; a rough threshold value of 8.5 ppmv could also be used (by GOZCARDS data users) to flag the pre-2005 merged dataset.

UARS MLS stratospheric $H_2O$ for GOZCARDS was obtained from V6 (or V600) $H_2O$ data. This data version is identical to the original prototype (named V0104) from Pumphrey (1999), who noted that UARS MLS $H_2O$ often exhibits drier values (by 5–10 %) than HALOE $H_2O$ (see also Pumphrey et al., 2000). The resulting GOZCARDS $H_2O$ monthly zonal means span the period from September 1991 through April 1993. We note that a significant fraction of UARS MLS tropical data values at 100 hPa are flagged bad (as a result of diminishing sensitivity).

Summarizing briefly past validation results, SPARC WAVAS (2000) analyses pointed out the existence of a small low bias in HALOE stratospheric data vs. most other measurements (from satellites or other means), except for UARS MLS. Lambert et al. (2007) showed agreement within 5–10 % between Aura MLS version 2.2 stratospheric $H_2O$ and other satellite data, including ACE-FTS $H_2O$ (see also Carleer et al., 2008), as well as for comparisons between Aura MLS and balloon data; Aura MLS $H_2O$ values are slightly larger than HALOE $H_2O$ in the stratosphere, with differences increasing to 10–15 % in the mesosphere. Changes from MLS v2.2 to v3.3 led to an increase of 0.2–0.3 ppmv in stratospheric values (Livesey et al., 2013). Past disagreements between aircraft water vapor measurements have made those datasets some-





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what difficult to use as absolute validation of satellite-derived $H_2O$ in the upper troposphere and lower stratosphere (UTLS) (Read et al., 2007; Weinstock et al., 2009). An intercomparison of measurements under controlled chamber conditions has helped to better constrain this issue (Fahey et al., 2014). Very good agreement exists between Aura MLS UTLS $H_2O$ and measurements from Cryogenic Frost point Hygrometers (CFH), as discussed by Read et al. (2007) and Voemel et al. (2007) for MLS v2.2 data. At the lowest level (147 hPa) used here for merged $H_2O$, the latter study showed a dry bias (by ~ 10 %) in the MLS v2.2 data vs. CFH. Recent comparisons by Hurst et al. (2014) of MLS v3.3 $H_2O$ data vs. Boulder CFH time series show excellent overall agreement, and no significant trend differences between coincident profile sets. There is therefore support for systematic uncertainties as low as 5 % for lower stratospheric MLS data. Aura MLS stratospheric $H_2O$ v3.3 values are slightly larger (by up to ~ 5 %) than the multi-instrument average from a number of satellite datasets, as discussed in SPARC Data Initiative comparisons by Hegglin et al. (2013). No large disagreements in interannual variations were noted by these authors for the GOZCARDS datasets ($p < 150$ hPa). From the mid-stratosphere to the upper mesosphere, excellent agreement between ground-based data from the Water Vapor Millimiter-wave Spectrometer (WVMS) and $H_2O$ profiles from Aura MLS and ACE-FTS has been demonstrated by Nedoluha et al. (2007, 2009, 2011).

### 4.2 GOZCARDS $H_2O$ merged data records

The merging process for $H_2O$ is nearly identical to the method used for HCl. The main difference is an additional step that merges UARS MLS data with the already combined datasets from HALOE, ACE-FTS, and Aura MLS, by simply adjusting UARS MLS values to the average of the previously merged series during the early (1991–1993) overlap period; see Fig. S5 for an illustration at 22 hPa for the 5° N latitude bin. Typically, this requires an upward adjustment of the UARS MLS $H_2O$ data, as these values are biased low vs. most other datasets; nevertheless, the fairly short but global record from UARS MLS helps to fill the time series. After considering the channel drift issues for SAGE II





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$H_2O$ (and following past advice from the SAGE II team itself), we decided to use caution and did not include that dataset for GOZCARDS merging, as some trend results could be affected to an unknown extent. Also, there is probably some remaining retrieval contamination from volcanic aerosol effects for some time after the volcanic eruptions of El Chichon (1982) and Mt. Pinatubo (1991), as well as after several smaller eruptions; see Bauman et al. (2003) for a review of stratospheric aerosol climatology (1984–1999) and Thomason et al. (2008) for the SAGE II stratospheric aerosol dataset.

Minor procedural merging details or issues for $H_2O$ are included in the Supplement. Also, data users should be aware of effects from unequal latitudinal sampling when no MLS data exist, for regions where large latitudinal variations occur, as for $H_2O$ at 147 hPa (the largest pressure value). Indeed, global or latitudinal averages can be significantly biased in certain months and month-to-month variability for such averages increases. This is because of poor sampling of the full latitudinal variability, prior to August 2004; after this, regular sampling exists from MLS every month. Such variations in sampling can become an issue for temporal analyses of latitudinal or global averages, unless additional fits or interpolations to mitigate such effects are undertaken.

In Fig. 11, we display the average offsets that were applied to the four $H_2O$ source datasets; these offsets follow previously known relative data biases (mentioned earlier). For example, low biases in UARS MLS $H_2O$, especially in the mesosphere, were discussed by Pumphrey (1999) and the UARS MLS offsets (see Fig. 11) correct that dataset upward. The application of offsets derived for HALOE and UARS MLS raises the $H_2O$ time series from these instruments, whereas negative offsets lower the $H_2O$ source data from ACE-FTS and Aura MLS. As we found for HCl, the offset values generally display small variations vs. latitude and are therefore fairly stable systematic adjustments to the time series. Figure S6 displays the amplitudes of the fitted annual cycles for HALOE, ACE-FTS, and Aura MLS. As for HCl, similar patterns emerge for these datasets. Wintertime descent into the polar vortex regions is responsible for large annual cycles at high latitudes, especially in the mesosphere; also, the seasonal impact of dehydration in the lower stratospheric Antarctic region causes a large annual cycle in





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Aura MLS high southern latitude data. Figure 12 provides some statistical information, as done for HCl in Sect. 3.2, regarding the correlations and trend differences between ACE-FTS and Aura MLS. There are a few regions with noisier relationships. While slow increases in $H_2O$ are generally observed by both instruments in the stratosphere and mesosphere, the tropical region near 0.1 hPa shows a slight decreasing trend for the ACE-FTS points, thus leading to larger discrepancies; it is not clear what the source of these discrepancies is. While the tropical ACE-FTS data are generally sampled with a significantly lower temporal frequency, the same applies for all pressure levels; however, a few outlier points can have a much larger impact when sampling is poorer. There are also a few other spots, such as near 65° S and 65° N and near 5 hPa with a poor trend value for the difference series, in comparison to the errors; this may be caused by a combination of poorer sampling by ACE-FTS and higher atmospheric variability, which can lead to more scatter. At the highest latitudes in the lower stratosphere, the observed slope differences are more within error bars, but the larger variability means that a longer record is needed to determine if two time series really trend differently. The main point here is to show the dataset characteristics and to point out where the agreement is better or worse than typical. The merged dataset tends to be much closer to Aura MLS in terms of trends because there are usually many more months of Aura MLS data than ACE-FTS data, including the fact that the ACE-FTS time series (data version 2.2) used here was halted for data after late 2010 due to technical retrieval issues. Therefore, the overall impact of ACE-FTS data on the merged $H_2O$ series is fairly small.

Figure S7 provides a visual representation of the merged GOZCARDS $H_2O$ fields at 3 and 68 hPa, respectively. Well-known features are displayed in these plots, given the good global coverage in the post-2004 period in particular. In the upper stratosphere, descent at high latitudes during the winter months leads to larger $H_2O$ values, and low latitude QBO features are also observed. In the lower stratosphere, one observes dehydration evidence at high southern latitudes in the winter months, as well as a low latitude seasonal “tape recorder” signal; this phenomenon is driven by tropopause tem-





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peratures and has been measured in satellite data since the early 1990s (Mote et al., 1996; Pumphrey, 1999). A vertical cross-section of this lower stratospheric tropical (20° S to 20° N) tape recorder in GOZCARDS merged $H_2O$ for 1991–2013 is shown in Fig. 13; periods of positive anomalies alternate with negative anomalies, including the post-2000 lows, as well as the most recent decreases in 2012–2013 (see also next section).

As we discussed for HCl, we have estimated systematic errors for the merged $H_2O$ product. This is illustrated by the contour plots in Fig. 14; these ranges encompass at least 95 % of the monthly mean source data values from HALOE, UARS MLS, ACE-FTS, and Aura MLS above or below the merged series. These errors typically span 5 to 15 % of the mean between 100 and 0.1 hPa; errors larger than 30 % exist in the tropical upper troposphere (147 hPa), and similarly, large values in the upper mesosphere arise from the low bias in UARS MLS $H_2O$.

### 4.3 GOZCARDS $H_2O$ sample results and discussion

Stratospheric $H_2O$ variations have garnered attention in the past two decades, because of the radiative impacts of water vapor in the UTLS and the connection to climate change, as well as the stratospheric chemical significance of $H_2O$ oxidation products. $H_2O$ can influence changes in stratospheric and mesospheric ozone via the $HO_x$ catalytic cycles. $H_2O$ in the UTLS has a significant radiative impact (e.g., Forster and Shine, 2002) and has the potential to influence surface temperature changes in ways that could mitigate surface warming (Solomon et al., 2010) if $H_2O$ exhibits a significant drop, as was observed right after 2000. A decrease of about 1 ppmv was also observed in in situ data (Fujiwara et al., 2010; Hurst et al., 2011; Kunz et al., 2013). Randel et al. (2004, 2006) correlated this post-2000 decrease with a decline in tropical cold point temperatures. An increasing trend in stratospheric $H_2O$ since the 1950s (see Rosenlof et al., 2001) will have a surface warming tendency. We expect to see continued studies of the influence of cold point temperatures on stratospheric $H_2O$ and the possible connections to changes in sea surface temperatures (see Rosenlof and





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Reid, 2008; Garfinkel et al., 2013). Efforts to better understand past changes in $H_2O$, and their causes and expected impacts, include the references above, and (among others) Dvortsov and Solomon (2001), Shindell (2001), Nedoluha et al. (2003), Urban et al. (2007), Dhomse et al. (2008), Scherer et al. (2008), Read et al. (2008), Tian et al. (2009), Schoeberl et al. (2012), Fueglistaler (2012), Fueglistaler et al. (2013), and the recent review of the tropical tropopause layer by Randel and Jensen (2013). The reconciliation of long-term trends in tropopause temperatures with changes in lower stratospheric water vapor is a task worthy of continued study, using additional datasets as well as model studies.

Individual water vapor datasets have been used here to produce a merged record now spanning more than two decades. Linear trend estimates can be quite sensitive to the starting and ending points of the time series, even for 22 years of data, and simple linear trends do not best describe the variations in stratospheric $H_2O$ over the past two decades. We do not attempt here to characterize trends or to imply that recent tendencies will carry into the next decade or two. Rather, as variability is also of interest to climate modelers, we provide information below regarding observed decadal-type (longer-term) variability in stratospheric water vapor. Figure 15 illustrates monthly, annual, and longer-term changes in stratospheric water vapor, based on the global GOZCARDS merged $H_2O$ series; this shows the well-known $H_2O$ minimum in the lower tropical stratosphere as well as an increasing vertical gradient in the upper stratosphere (as a result of methane oxidation). As we know from past studies (e.g., Randel et al., 2004), medium- to long-term changes in $H_2O$ are large-scale in nature. However, lower stratospheric $H_2O$ variations are more accentuated at low latitudes, in comparison to near-global (60° S–60° N) results. It has long been known (e.g., from the in situ balloon-borne measurements of Kley et al., 1979) that the hygropause is typically located a few km higher than the thermal tropopause; this is consistent with the tape recorder and Brewer–Dobson circulation concepts. We observe low water vapor mixing ratios at 68 hPa in the tropics, in comparison to 100 hPa values (near the tropopause). According to the 22 year GOZCARDS data record, annually-averaged $H_2O$ values in



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the tropics (20° S–20° N) have varied between about 3.2 and 4.2 ppmv at 68 hPa. The rapid drop between 2000 and 2001 is observed at 100 and 68 hPa, with some dilution of this effect at higher altitudes. There is a clear difference in long-term behavior between the upper stratosphere, where changes in methane should have the clearest influence, and the lower stratosphere, especially in a narrow vertical region above the tropopause, where cold point temperatures and dynamical changes have a significant impact. To first-order, the last few years show ~ 10 % larger values in the upper stratosphere than in the early 1990s, while the opposite holds in the lowest stratospheric region, where a decrease of order 10 % is observed over the same period. The long-term upper stratospheric increase carries into the mesosphere (see below). Figure 15 also shows that month-to-month and seasonal variations (thin lines) are usually somewhat larger than the long-term changes in the lower stratosphere, most notably at 100 hPa.

In order to provide longer-term variability diagnostics for water vapor, we show in Fig. 16 the minimum to maximum spread in annual averages (tropics and mid-latitudes) from Fig. 15. These variability diagnostics are provided for the 22 year period (1992 to 2013) and also separated into the two 11 year periods (thin and dashed lines); as expected, the 22 year variability is always larger than the variability in either of the two decadal period subsets. We also note that the tropical variability is largest just above the tropopause (here this means at the 68 hPa GOZCARDS level), where it reaches 20–28 % (or 0.8 to 1 ppmv) depending on the time period. Such variability diagnostics should be useful for comparisons to various chemistry climate models.

The longer-term variability in water vapor increases above the stratopause and reaches close to 30 % in the uppermost mesosphere, as seen in Fig. 17a; this plot shows the monthly and annual near-global (60° S–60° N) $H_2O$ variations at 0.01 hPa. Large seasonal changes in this region are driven by vertical advection associated with the mesospheric circulation, with each hemisphere's summertime peaks contributing to the maxima (two per year) in these near-global averages; such seasonal variations were compared to model results by Chandra et al. (1997), based on the first few years of HALOE $H_2O$ data. The strong upper mesospheric variability in annual-mean $H_2O$ is





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known from previous studies of ground-based and satellite $H_2O$ data (Chandra et al., 1997; Nedoluha et al., 2009; Remsberg, 2010), and this region is where the solar (Lyman $\alpha$) influence on $H_2O$ is strongest. Figure 17b displays the near-global variations in annual upper mesospheric $H_2O$ from 0.1 to 0.01 hPa. We clearly see increased variability in the uppermost mesosphere, and decreases in the mixing ratios as a result of $H_2O$ photodissociation.

## 5 GOZCARDS ozone

A number of discussions relating to signs of ozone recovery have been presented before (Newchurch et al., 2003; Wohltmann et al., 2007; Yang et al., 2008; Jones et al., 2009; Hassler et al., 2011; Salby et al., 2011, 2012; Ziemke and Chandra, 2012; Gebhardt et al., 2013; Kuttipurath et al., 2013; Kirgis et al., 2013; Nair et al., 2013, 2015; Shepherd et al., 2014; Frith et al., 2014). While there are some indications of small increases in $O_3$ in the past 10–15 years, further confirmation of an increase in global $O_3$ and its correlation with column increases, is needed, in order to more clearly distinguish between long-term forcings, notably from the 11 year solar cycle, slow changes in halogen source gases, temperature changes, and shorter-term variability. Continuing, good long-term ozone datasets are clearly needed for such studies.

### 5.1 GOZCARDS ozone source data records

We used ozone datasets from SAGE I, SAGE II, HALOE, UARS MLS, ACE-FTS, and Aura MLS to generate the monthly zonal mean source products for GOZCARDS. Due to time constraints, we did not use the newer SAGE II version 7 ozone (see Damadeo et al., 2013) as part of the GOZCARDS merged dataset. Our studies indicate that there are systematic differences of a few percent between SAGE II V6.2 and V7 $O_3$ on their native coordinates (number density vs. altitude). However, these 2 versions will exhibit different trends, mainly in the upper stratosphere, after the data are converted to mix-



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ing ratios on pressure surfaces (as shown later). These differences result mainly from different temperature trends between MERRA and analyses from the National Centers for Environmental Prediction (NCEP), which are used by the SAGE II V7 and V6.2 retrievals, respectively; the main differences between MERRA and NCEP temperatures occur in the upper stratosphere for time periods before 1989 and after mid-2000. After June 2000, SAGE II V6.2 $O_3$ at upper stratospheric pressures ($\leq$ 3 hPa) is not included in our merged data (see discussions in Sect. 5.2). In addition to the general data screening methods (Sect. 2), HALOE $O_3$ was screened for aerosols based on recommendations from Bhatt et al. (1999). Specifically, the $O_3$ profiles were screened for instances when either the 5.26 µm aerosol extinction exceeded $10^{-3}$ km$^{-1}$ or a local aerosol extinction minimum was present near the tropopause; all $O_3$ values at or below the identified levels were flagged bad.

#### 5.1.1 Treatment of SAGE ozone profiles

Both SAGE I and SAGE II used solar occultations during satellite sunrise and sunset to measure vertical profiles of ozone, along with other composition data and aerosol extinction (McCormick et al., 1989; Cunnold et al., 1989). It takes about 1 month for SAGE I and II to provide near global coverage (about 80° N to 80° S), with some dependence on season. The SAGE I measurements started in February 1979 and stopped in November 1981, while SAGE II provided data between October 1984 and August 2005. In the middle of July 2000, SAGE II had a problem in its azimuth gimbal system. Although this was corrected by November 2000, the instrument operation was switched to a 50 % duty cycle, with either sunrise or sunset occultations occurring in monthly alternating periods, until the end of the mission.

It has been known that there were altitude registration errors in SAGE I (V5.9) data (Veiga et al., 1995; Wang et al., 1996). To correct this problem, an empirical altitude correction method based on Wang et al. (1996) had been applied to SAGE I (V5.9) data; these corrected SAGE I V5.9 profiles, which had been evaluated in previous trend studies (e.g. SPARC Report, 1998; WMO, 2003), were used to create the GOZCARDS





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SAGE I product (denoted as version V5.9_rev). We did not use reprocessed version 6.1 SAGE I data (L. W. Thomason, personal communication, 2012) because the altitude registration problems had not been completely fixed and new altitude correction criteria should be derived and validated.

Ozone data screening details for the original SAGE I and SAGE II datasets are provided in the Supplement. The number density profiles were converted to mixing ratios on pressure levels by using NCEP temperature and pressure data provided with each profile. Derived ozone profiles were then interpolated to fixed pressure levels on the following grid:

$$p(i) = 1000 \times 10^{-i/30}\ (\mathrm{hPa}),\ i = 0, 1, 2, \ldots \tag{2}$$

Ozone values at each of the 5 levels centered on every GOZCARDS pressure level were then averaged (weighted by pressure) to derive mixing ratios at each GOZCARDS pressure level. By doing this, the SAGE profiles were smoothed to a vertical resolution comparable to that of the other satellite instruments used in this GOZCARDS work. Monthly zonal means were then computed for the SAGE ozone datasets on the GOZCARDS-compatible grid.

#### 5.1.2 Comparisons of ozone zonal means

$O_3$ differences between SAGE II and other satellites are shown in Fig. S8. Zonal mean differences between SAGE II and HALOE are generally within 5 % for 1.5 to 68 hPa at mid-latitudes, and for 1.5 to 46 hPa in the tropics. The relative biases are larger outside those ranges and increase to ∼ 10 % near the tropopause and also near 1 hPa. SAGE II data show better agreement with UARS and Aura MLS in the upper stratosphere and lower mesosphere, within 5 % up to 0.68 hPa and for latitudes outside the polar regions. Aura MLS $O_3$ compares better with SAGE II data than does UARS MLS in the tropics for pressures larger than 68 hPa; the high bias in UARS MLS $O_3$ at 100 hPa has been discussed previously (Livesey et al., 2003). There are no months that include





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both SAGE II and ACE-FTS data in the Northern Hemisphere tropics (see the gap in Fig. S8, bottom right panel), largely due to the poorer coverage from ACE-FTS in the tropics. ACE-FTS $O_3$ shows the largest positive bias (greater than 10 %) with respect to SAGE II, for pressures less than 1.5 hPa. The high bias in upper stratospheric ACE-FTS ozone has been mentioned in past validation work using ACE-FTS data (e.g., Froidevaux et al., 2008; Dupuy et al., 2008). The biases shown here are also consistent with recent $O_3$ intercomparison studies from a comprehensive array of satellite instruments by Tegtmeier et al. (2013). It has been known for some time that the HALOE and SAGE II ozone datasets, which govern the main variations of the GOZCARDS merged ozone values before 2005, agree quite well (within 5 %) in absolute value, and also in terms of temporal trends (Nazaryan et al., 2005), and vs. ozonesondes (mostly above $\sim$ 20 km or $\sim$ 50 hPa). Larger percentage differences occur in the lowest region of the stratosphere at low latitudes, and especially in the upper troposphere, where HALOE values become significantly smaller than SAGE II data, which are already biased low (by $\sim$ 50 %) vs. sondes (Wang et al., 2002); see also Morris et al. (2002), as well as results of SAGE II and HALOE comparisons vs. solar occultation UV-Visible spectrometer measurements from long duration balloons (Borchi et al., 2005). We should note here that in this GOZCARDS merging work, we have largely avoided the upper tropospheric region.

Zonal mean differences between SAGE II and Aura MLS show some latitudinal structure between 1 and 3 hPa, with larger (5–10 %) biases in the Southern Hemisphere, especially for 0 to 30° S (see Fig. S8). There are no such features between SAGE II and HALOE or UARS MLS. We found that this results from anomalous NCEP temperatures after 2000, which affect SAGE II data converted from number density/altitude to GOZCARDS VMR/pressure coordinates. Figure 18 shows an example of the ozone series from SAGE II and other satellite data for 10 to 20° S from 1 to 6.8 hPa. At 1 hPa, SAGE II ozone drifts and is elevated after mid-2000, when compared to HALOE. Similar features are found down to pressures near 3 hPa. These anomalous values can be attributed to abnormal NCEP temperature trends compared to MERRA and HALOE



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during the same time period (for detailed views, see Figs. S9 and S10). Issues relating to anomalous upper stratospheric NCEP temperature trends were noted by McLinden et al. (2009). Because such NCEP-related artifacts are confirmed by both MERRA and HALOE, we decided not to include in the merging process any SAGE II $O_3$ values after 30 June 2000 for pressures equal to or less than 3.2 hPa. SAGE II ozone is not significantly affected by the conversion to mixing ratio/pressure coordinates at 4.6 and 6.8 hPa (Fig. 18).

## 5.2 GOZCARDS ozone merged data records

### 5.2.1 Methodology for GOZCARDS merged ozone

Ozone measurements from SAGE I, SAGE II, HALOE, UARS MLS, Aura MLS and ACE-FTS, as described in Sect. 5.1, were used to establish a near-continuous monthly zonal mean record from late 1979 through 2012 for the GOZCARDS merged $O_3$ product (ESDR version 1.01). The monthly means from each instrument were produced after applying the screening described in Sect. 5.1. The SAGE II dataset was used as a reference standard, since it has the longest period of measurements and has been extensively validated. A GOZCARDS ozone merged data record is constructed by combining these measurements after removing systematic biases with respect to SAGE II. This is done by applying additive offsets to all other instrument series, as determined from average differences between monthly zonal means and SAGE II during overlap time periods. The merged data are then derived by averaging all available adjusted datasets. Because there are gaps in overlap between SAGE II and ACE-FTS monthly mean data in some latitudes (Fig. S7), and as SAGE II ozone VMRs obtained from the vertical grid transformation were affected by anomalous NCEP temperatures after mid-2000 for pressures smaller than or equal to 3.2 hPa, a two-step approach is used to generate the merged product. First, SAGE II data are used as reference for pressures larger than 3.2 hPa to adjust HALOE, UARS MLS and Aura MLS based on overlapping months between 1991 and November 2005; see the method overview





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schematic in Fig. 19. For $p \leq 3.2$ hPa, SAGE II $O_3$ is still used as a reference through June 2000, and HALOE and UARS MLS data are adjusted accordingly. This eliminates the effect of anomalous NCEP temperatures on SAGE II ozone and leads to more accurate offsets based on HALOE values, after they have been adjusted to SAGE II. The adjusted HALOE data (denoted as HALOE* in Fig. 19) are then used as a reference to derive estimated offsets for Aura MLS $O_3$, using the overlap period with HALOE from August 2004 to November 2005. In step 2, a new reference value is derived by averaging all available data from SAGE II, HALOE*, UARS MLS* and Aura MLS*. This new reference value is then used to adjust the ACE-FTS ozone values based on all overlapping months between March 2004 and November 2005. By including Aura MLS in the dataset created in step 1, we obtain more complete spatial and temporal coverage than possible with SAGE II and HALOE, and ensure that there are overlapping months between this combined dataset and ACE-FTS source data. At the end of step 2, the final merged ozone is derived by averaging the temporary merged dataset from step 1 with the adjusted ACE-FTS data values.

### 5.2.2 Further considerations regarding GOZCARDS merged ozone data

Diurnal changes in ozone can affect measurement comparisons and could impact data merging. Measurements and models of the ozone diurnal variation from the lower stratosphere to the mesosphere have been discussed previously (Ricaud et al., 1996; Haefele et al., 2008; Huang et al., 2010). Sakazaki et al. (2013) presented diurnal changes measured by the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), and Parrish et al. (2014) have analyzed ground-based microwave ozone profile variations vs. local time in conjunction with satellite datasets. These studies indicate that ozone diurnal variations range from a few percent in the lower stratosphere to more than 10 % in the upper stratosphere and lower mesosphere. SAGE II and other occultation instruments observe ozone at local sunrise or sunset, and the retrieved values are generally closer to nighttime values in the upper stratosphere and mesosphere. To characterize systematic differences between satellite data, coincident





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profiles with small differences in space and time are most often used; an example of mean differences and SD between SAGE II and Aura MLS using both coincident profile and zonal mean methods is provided in Fig. S11. SAGE II and coincident Aura MLS nighttime $O_3$ values agree within ~ 5 % between 0.46 and 100 hPa, except in the tropical lower stratosphere where comparisons are noisier due to weak $O_3$ signals and strong dynamical variability. Differences between zonal mean SAGE II and Aura MLS data are very close to the differences from averaged coincident values, except for pressures less than 2 hPa, where differences between the methods increase from a few % to ~ 10 % at 0.3 hPa, consistent with what one expects from the diurnal cycle. Biases between SAGE II and Aura MLS based on coincident profiles vs. zonal means could be different by more than 5 % in the upper stratosphere and above. Zonal mean differences are likely to be less representative of "true" differences between the two instruments. By combining SAGE II with Aura MLS data adjusted by zonal mean biases, we provide a series adjusted to the average of sunrise and sunset, as measured by SAGE II. However, if Aura MLS data were adjusted by biases obtained using the coincident method, an upper stratospheric offset of several percent and artificial trends due to such a diurnal cycle effect could be introduced.

Even in the absence of diurnal variations, measurements from occultation sensors can yield larger sampling errors than those from more densely-sampled emission measurements (Toohey et al., 2013). The use of long-term data with consistent sampling should be an advantage for trend detection. Avoiding SAGE II data after mid-2000 also mitigates potential artifacts arising from different SAGE II sunrise/sunset sampling patterns vs. time. The HALOE sampling remained fairly balanced between SR and SS events over its mission duration, although there were also more data gaps in the later years. Similarly, the Aura MLS ozone data generated here are averaged from local times roughly in the middle of the day and the middle of the night, with repeatable and stable patterns over the years; ACE-FTS sampling patterns are also quite stable.

Figure 20 displays the average ozone offsets obtained from the calculated biases vs. SAGE II data. The effect of a high bias in upper stratospheric and lower mesospheric





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ozone from ACE-FTS relative to other datasets is made evident by the need to apply a negative offset as large as 25 % to the ACE-FTS series. Most of the stratospheric offsets applied to the other instrument datasets are in the 5–10 % range; a lowering of $O_3$ from UARS MLS, HALOE, and Aura MLS in the lower mesosphere is generally required to match the SAGE II values. Sampling differences and data sparseness may be responsible for larger offsets at the highest latitudes. In these regions, more caution is required with the merged data, which is also less amenable to long-term analyses because of data gaps and larger variability (especially prior to 2004).

As shown in the Supplement (Fig. S12), we observe strong similarities in the ozone annual cycle amplitude patterns from SAGE II, HALOE, ACE-FTS, and Aura MLS over their respective measurement periods (e.g., peaks at midlatitudes near 10 and 1.5 hPa). The middle stratospheric peaks are a result of the annual cycle in oxygen photolysis, whereas temperature variations drive the annual cycle in the upper stratosphere (Perliski et al., 1989). This sort of comparison provides some (first-order) reassurance regarding the consistency of the various datasets. For further details, Fig. 21 provides diagnostics similar to those presented for HCl and $H_2O$, namely the correlation coefficients and significance ratios for the slopes of the deseasonalized anomaly time series from SAGE II vs. HALOE as well as from ACE-FTS vs. Aura MLS (for 1992 through 1999, and 2005 through 2009, respectively). These diagnostic results for ACE-FTS and Aura MLS are of a quality that is comparable to the HALOE/SAGE II results; poorer fits occur mostly at high latitudes and in the upper stratosphere. Poorer correlations at upper altitude appear largely tied to a decrease in the amount of valid data in this region (especially at high latitudes), coupled with a relatively small variability. For regions with poorer agreement between ACE-FTS and Aura MLS, we often see small variability in the series from Aura MLS but larger changes (scatter) in the ACE-FTS series. Larger differences in trends between SAGE II and HALOE were noted by Nazaryan et al. (2005) at low latitudes near 50 km; this is also indicated by our simple linear fits (not shown here) to the GOZCARDS source datasets from these two instruments and the existence of poorer agreements in Fig. 21 (2nd panel from top) for the





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slope of the differenced (anomaly) series in that region. The existence of good correlations in interannual ozone variations between a large number of satellite measurements was discussed by Tegtmeier at el. (2013). Regarding temporal drifts, Nair et al. (2012) have shown that small drifts (mostly within about ±0.5 % $yr^{-1}$ for the 20–35 km region) exist between most of the datasets from six ozone lidar sites and coincident HALOE, SAGE II, and Aura MLS measurements; similar results were obtained at two of these sites by Kirgis et al. (2013). Other recent or ongoing studies (in particular, the comprehensive study by Hubert et al., 2015) corroborate the very good stability of the longer-term ozone datasets used for GOZCARDS, which relies most heavily on data from SAGE II and Aura MLS. Jones et al. (2009) also studied the consistency of various satellite datasets (including SAGE II and HALOE) for 1979–2008; they concluded that small relative drifts existed between their averaged series and individual instrument series, although they did find that larger inter-instrument drifts exist in the extra-tropical upper stratosphere. While we feel justified in the use of the longer-term time series and generally robust datasets chosen for GOZCARDS $O_3$, data users should still note the existence of a few regions with poorer correlations or trend agreement (and, therefore, larger uncertainties) between different satellite ozone datasets, as indicated in Fig. 21. Long-term merged datasets from GOZCARDS and other sources should undergo continued scrutiny from the community, as done recently for trends by Tummon et al. (2014) and Harris et al. (2015). Sample cross-sectional views of two slices through the GOZCARDS merged $O_3$ field are provided in the Supplement (Fig. S13). Figure 22 shows estimated systematic errors from our calculation of the 95 % ranges for the monthly mean source data used here, both above and below the merged values. In this case, as SAGE II is used as a reference dataset, the applied offsets (Fig. 20) correlate quite well with this plot depicting the ranges about SAGE II values. Minimum error bars can be slightly lower than 5 % for the middle stratosphere at low latitudes, where ozone values are largest. This view of systematic error bars is generally consistent with results by Tegtmeier et al. (2013), who used a SD measure, based on the larger set of satellite datasets analyzed for the SPARC Data Initiative. They also found that the regions





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with lowest errors (scatter) are in the middle stratosphere at low to mid-latitudes, where most monthly mean satellite data fit within ±5 % of the multi-instrument mean.

### 5.3 GOZCARDS ozone sample results and discussion

Nair et al. (2013) used regression analyses to compare profile trend results from GOZCARDS merged $O_3$ at northern midlatitudes vs. a combined $O_3$ dataset from lidar and coincident satellite data at the Observatoire de Haute Provence (OHP), France. They showed that good consistency exists for the decreasing ozone time period, from the early 1980s to 1997, and for the upper stratospheric increase since 1997, but some differences exist in the lower stratosphere during this second time period, when the GOZCARDS results show a near-zero trend in comparison to small positive trends from the combined (and more localized) dataset. The above results for the declining time period agree broadly with earlier work (for the 1979–1997 period) by Jones et al. (2009), who averaged various satellite ozone datasets and produced trend estimates; however, these authors obtained only a small positive, but statistically insignificant, linear trend for the post-1997 phase, most likely because of too short a time series at that time. Gebhardt et al. (2013) analyzed ozone profile trends from SCIAMACHY on ENVISAT, and compared this to trends from Aura MLS, Optical Spectrograph and InfraRed Imager System (OSIRIS) on the Odin satellite, and sondes; their results include the detection of localized ozone increases in the mid-stratosphere at low latitudes; see also Bourassa et al. (2014), who analyzed merged SAGE II and (OSIRIS) observations for 1984–2013, as well as results from Kyrölä et al. (2013) on combined SAGE II and Global Ozone Monitoring by Occultation of Stars (GOMOS) records for 1984–2012, and Eckert et al. (2014), who investigated ENVISAT MIPAS trends for 2002–2012. The shortness of data records since 1997, coupled with relative variability and potential drifts between various measurements may explain some differences in recent trend results, notably for the post-1997 period. More comprehensive analyses from the $SI^2N$ initiative have focused on an intercomparison of profile changes from a variety





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of datasets, including GOZCARDS and other merged records (Tummon et al., 2014; Harris et al., 2015).

Here, we investigate ozone column results based on the global GOZCARDS dataset, given the work by Ziemke and Chandra (2012), hereafter generally referenced to as ZC12; these authors analyzed total column and stratospheric column data from satellite measurements, and their analyses yielded a rather strong near-global (60° S–60° N) average ozone increase since 1998. Their stratospheric column measurements depend on the convective-cloud differential (CCD) method, which uses Total Ozone Mapping Spectrometer (TOMS) and Ozone Monitoring Instrument (OMI) column ozone data over convective clouds near the tropopause; below the clouds, little sensitivity exists, so the method can lead to stratospheric column estimates, especially in the tropics, where good cloud-related data of this kind exist. For midlatitudes, their methodology has focused on ozone data over the Pacific, along with a few assumptions relating to cloud heights and longitudinal invariance, in order to try to represent zonal mean stratospheric columns (see also Ziemke et al., 2005). In Fig. 23, we show the near-global total and stratospheric column values from ZC12 (J. Ziemke, personal communication, 2013), along with (unscaled) GOZCARDS column densities down to three pressure levels (68, 100, and 215 hPa) for mid-latitudes (30–60° S and 30–60° N), low latitudes (30° S–30° N), and a near-global range (60° S–60° N). In an absolute sense, the GOZCARDS near-global columns above 215 hPa are larger than the ZC12 stratospheric columns, but quite close to the total column amounts from ZC12. Not too surprisingly, the GOZCARDS column values above 100 hPa are slightly lower than the stratospheric columns from ZC12, as the latter columns (estimated down to cloud tops) will capture more of the lower stratosphere in the extra-tropics. Most of the near-global decrease comes from the midlatitudes, as more of the lower stratospheric column resides in these regions, in an absolute sense. Randel and Thompson (2011) obtained small decreases (−2 to −4 % decade$^{-1}$) in tropical lower stratospheric ozone for 1985–2009, from a combination of SAGE II ozone and sonde data from the Southern Hemisphere Additional Ozonesondes (SHADOZ) network. A fairly strong increase is observed from



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2008 to 2010 at northern midlatitudes (orange curves); Steinbrecht et al. (2011) attributed large ozone enhancements in that region in 2010 to a coupling between the quasi-biennial and Arctic oscillations (and the North Atlantic oscillation). Long-term halogen source gas reductions that have occurred since the mid-1990s should only lead to an ozone increase of a few DU since 1997 (Steinbrecht et al., 2011).

In Fig. 24, we compare the changes in 60° S–60° N ZC12 column ozone data to the GOZCARDS column amounts above 68 hPa for that region; note that GOZCARDS values do not provide for a continuous long-term time series down to pressures of 100 hPa (or more) in the SAGE I years (1979–1981). To eliminate biases between stratospheric columns as calculated using the CCD methodology and the GOZCARDS fixed bottom pressure approach, we reference all stratospheric columns to the 1980 total column value. These column series include the SAGE I data record and are linearly interpolated between 1981 and 1984, when no GOZCARDS source datasets exist. We observe that the relative changes in GOZCARDS columns follow the ZC12 curves within a few DU in the downward phase until about 1992, but the 1992–1997 decrease in total columns does not compare very well. Some of this discrepancy may be because total columns capture a stronger decrease from levels below 68 hPa, not fully represented in GOZCARDS columns; there are also gaps in ZC12 stratospheric columns in 1993–1996 and other years. Focusing on the late period for GOZCARDS data from Aura MLS and ACE-FTS, we also show the GOZCARDS columns above 68 hPa, referenced to 2007 instead of 1980. There is a good match in the variations between GOZCARDS and ZC12 columns during 2005–2010, in agreement with the fact that very good correlations were obtained by ZC12 between Aura MLS column variations and stratospheric column data from the CCD technique. The ZC12 values for stratospheric and total columns are in good agreement, although the stratospheric values have gaps when not enough data were present for near-global estimates. Also, the large increase in ZC12 data from 1997 to 1998 is not matched very well by GOZCARDS column data.

We removed the solar cycle from the deseasonalized anomalies, as was done in the ZC12 study, namely via a regression fit and subtraction of that component from the



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time series. In the resulting plots (see Fig. 25), a 3 year smoothing is also applied (as done by ZC12). We focus in these plots on the total time period from 1979 onward, and therefore, on the GOZCARDS columns above 68 hPa. While agreement exists at the few DU level between the ZC12 relative changes and the GOZCARDS columns, the apparent "recovery" in the ZC12 datasets is quite large ($\sim 3\,\%$) and is not matched by the changes in GOZCARDS columns. The latter columns show an increase of less than 0.8 % between 2001 and 2011, with some decrease (by $\sim 0.5\,\%$) from 2011 to 2013. We note that the recent analyses by Sheperd et al. (2014), who used a chemistry-climate model constrained by observed meteorology to investigate potential causes of long-term total column ozone variations, show a partial return, in 2010, towards the 1980 ozone levels (for 60° S–60° N), but not nearly as much as implied by ZC12, neither in the model, nor in the observations. It is possible that the discrepancies lie in the various datasets and their merging; for example, it would be worthwhile to check if homogenized SBUV column $O_3$ data show results that are substantially different from those of ZC12. Alternatively, the discrepancies could mainly reflect differences between the coverage or meaning of the different ozone columns used here (because of different methodologies, grids and/or sampling to properly determine a near-global result). Although most column discrepancies are not that large as a percent of the total column values, a better consensus regarding the recovery of near-global ozone columns (and profile values discussed in other recent references) will be desirable in the future.

## 6 Other GOZCARDS data records

We now briefly mention the three other datasets that were part of the delivery of GOZCARDS records for public dissemination in 2013, namely $N_2O$, $HNO_3$, and temperature. For $N_2O$ and $HNO_3$, the somewhat simpler merging procedure consisted of averaging the source datasets from ACE-FTS and Aura MLS over the overlap time period (August 2004 through September 2010) to obtain the additive offsets for each of the





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two individual records. We then simply used the correspondingly-adjusted and averaged series to create the merged results; this procedure is the same as we described for the first step in the HCl (or $H_2O$) merging process.

### 6.1 $N_2O$

This data set starts in August 2004, when the Aura MLS data record began; the only dataset after September 2010 is the Aura MLS $N_2O$ (version 3.3) data record, because we no longer had ACE-FTS version 2.2 data after that time due to ACE-FTS data processing issues mentioned earlier. Because of degradation in the main target MLS $N_2O$ band (near 640 GHz) after the first few months of 2013, the $N_2O$ standard MLS product will be reprocessed for the whole Aura MLS period using an alternate measurement band and an updated software version. As discontinuities in the version 3.3 MLS data are introduced after mid-2013, when the standard $N_2O$ product was replaced with results from the 190 GHz band, there are currently no GOZCARDS $N_2O$ zonal mean data after 2012 based on the original (640 GHz) MLS $N_2O$ measurement band.

Validation results for the first few years of Aura MLS and ACE-FTS $N_2O$ data were provided by Lambert et al. (2007) and Strong et al. (2008), respectively. Livesey et al. (2013) provided a minor update regarding the v3.3 Aura MLS $N_2O$ data used here, which show typically small differences (within ±5 %) in comparison to v2.2 data. The references mentioned above showed that excellent agreement (mostly within 5 %) exists between the stratospheric ACE-FTS and Aura MLS $N_2O$ profiles. Plots showing the average offsets applied to both MLS and ACE-FTS $N_2O$ series as a function of latitude and pressure are provided in Fig. S14. These plots are in agreement (in magnitude and in sign) with the above-referenced studies; the two datasets yield typical offsets (one half of the average differences) of less than 5 %. Also, very good temporal agreement between these two time series (for 2004–2010) is illustrated by the quality of the $N_2O$ diagnostic information displayed in Fig. S15 (computed as for other MLS and ACE-FTS comparisons discussed in this work). This generally shows very highly correlated fields, with insignificant drifts between the two separate time series of





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deseasonalized $N_2O$ data; the poorest correlations are obtained near 100 hPa in the tropics.

Figure 26 shows sample contour plots for the $N_2O$ merged field (2004–2012); as seen from the bottom panel (100 hPa), wintertime descent brings low $N_2O$ values down at high latitudes (inside the polar vortices). $N_2O$ is a conserved tracer in the lower stratosphere and its variations near the tropopause have implications regarding age of air. Variations in upper stratospheric $N_2O$ are clearly affected by seasonal and dynamical effects; this is evident from the striking semi-annual, annual and QBO-related patterns displayed in Fig. 26 for the 6.8 hPa level (top panel).

## 6.2 $HNO_3$

As for $N_2O$, we merged the $HNO_3$ data from ACE-FTS (version 2.2) and Aura MLS (version 3.3) from August 2004 onward, and included only the adjusted MLS dataset after September 2010. The average offsets applied to MLS and ACE-FTS time series as a function of latitude and pressure for $HNO_3$ are provided in Fig. S16. The typical offsets (one half of the average differences) for $HNO_3$ are less than $\sim 10\,\%$ (and less than 0.5 ppbv). Despite somewhat larger percent absolute differences than for $N_2O$ between Aura MLS and ACE-FTS $HNO_3$, there is very good agreement as a function of time between these two datasets in the stratosphere. This is illustrated by the quality of the $HNO_3$ diagnostic information provided in Fig. S17; the poorest correlations are obtained at or below the tropical tropopause.

Comparisons of v3.3 Aura MLS and v2.2 ACE-FTS nitric acid profiles have shown good agreement (see also Livesey et al., 2013), as the MLS $HNO_3$ v3.3 values are now generally larger than in v2.2, for which validation results were provided by Santee et al. (2007). Wolff et al. (2008) also compared MLS (v2.2) and ACE-FTS (v2.2) coincident profiles, and obtained similar results; in addition, they demonstrated that very good agreement exists between the $HNO_3$ profiles from ACE-FTS and coincident profiles from MIPAS on Envisat. Also, comparisons between Aura MLS $HNO_3$ (v3.3) profiles and wintertime $HNO_3$ profiles retrieved by a Ground-based Millimeter-wave





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Spectrometer (GBMS) in Thule, Greenland, during the first 3 months of 2010, 2011, and 2012 have shown good agreement, mostly within 10–15 % (Fiorucci et al., 2013).

Figure 27 (top two panels) displays the $HNO_3$ fields at 46 hPa from the UARS MLS period (1991–1997) as well as from the 2004–2013 period, for which a merged GOZCARDS product was produced, based on Aura MLS and ACE-FTS source datasets. Also shown (bottom two panels) are time series for 45° N and 32 hPa from both these periods; the bottom right panel includes the source and merged time series. We have performed additional investigations (not shown here) which lead us to believe that small upward adjustments to the UARS MLS $HNO_3$ values (by about 10 %) are needed to better cross-correlate these datasets across the two distinct time periods; such relative biases are within the expected systematic errors. This is based on a consideration of ground-based Fourier Transform infrared column $HNO_3$ data covering the full time period, as well as past GBMS $HNO_3$ profile retrievals. Also, Aura MLS and ACE-FTS $HNO_3$ data match ground-based and other correlative data quite well, and typically better than the (intrinsically poorer quality) UARS MLS $HNO_3$ data. However, obtaining an optimum global set of adjustments for the UARS MLS nitric acid field will be limited by the number of sites with such ground-based data (as well as by the different vertical resolutions for these datasets vs. MLS). More collaborative work regarding such analyses is needed in order to find the optimum adjustments to help tie together these two time periods for this species. Although we did not deliver the UARS MLS $HNO_3$ source data files for GOZCARDS, we could provide these monthly zonal mean series upon request, keeping the above caveats in mind.

### 6.3 Temperature

Finally, in terms of the initial set of delivered GOZCARDS products, and for the convenience of stratospheric composition data users, we have used temperatures ($T$) from the Modern-Era Retrospective Analysis for Research and Applications (MERRA) to produce a monthly mean GOZCARDS temperature data set from 1979 onward. MERRA is a NASA Goddard reanalysis (Rienecker et al., 2011) for the





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satellite era using Goddard Earth Observing System Data Assimilation System version 5 (GEOS-5); $T$ is from the DAS 3d analyzed state MAI6NVANA, version 5.2 files (such as MERRA300.prod.assim.inst6_3d_ana_Nv.20110227.hdf). Data from four daily MERRA files (for 00:00, 06:00, 12:00, and 18:00 h UT) were averaged to provide daily mean temperature fields (appropriate for a mean time of 09:00 h). Vertical interpolation was performed onto the GOZCARDS pressure grid, which, for temperature, covers 30 pressures levels from 1000 to 0.0147 hPa. Averaged values were stored for the 10° GOZCARDS latitude bins, and daily results were binned to create the GOZCARDS monthly temperature data set (version 1.0).

## 7 Summary and conclusions

We have reviewed the MEaSUREs GOZCARDS project's production of merged data records of stratospheric composition using carefully screened satellite data, starting in 1979 with SAGE I $O_3$ and continuing with Aura MLS and ACE-FTS data. The source datasets have a high degree of maturity, and we have reinforced our confidence in their usefulness through investigations of various diagnostics (offsets, annual cycle amplitudes, temporal correlations and trend differences of deseasonalized series). We have focused here on the relatively long-term data records for HCl, $H_2O$, and $O_3$. These records are publicly available as GOZCARDS ESDR version 1.01 and can be referenced using DOI numbers (Froidevaux et al., 2013b; Anderson et al., 2013, and Wang et al., 2013, for the above species, respectively). The other GOZCARDS data records mentioned here also have dataset references, namely Schwartz et al. (2013) for the MERRA-based temperature records, and Froidevaux et al. (2013c, d) for $N_2O$ and $HNO_3$, respectively. Table 2 provides a summary of the monthly mean datasets produced for GOZCARDS. Yearly netCDF files were delivered to the GES DISC for public access (see http://mirador.gsfc.nasa.gov, where a README document is also available, see Froidevaux et al., 2013a). Temperature records based on MERRA are also included on the GOZCARDS grid (see Sect. 1). The merging methodology follows from



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the determination of mean biases (for each pressure level and 10° latitude bin) between satellite instrument datasets, based on the overlap periods between the various series. Each species is treated separately: for ozone, SAGE II data are the chosen reference, whereas for other species, the approach is equivalent to an average of the datasets during the periods of overlap. The merged data files contain the average offset values that were applied to each source data time series, along with SD and standard errors. The GOZCARDS README document provides more details about the GOZCARDS data file quantities, including local time and solar zenith angle information, and a list of days with available data for each month. We have also presented here a compilation of systematic error estimates about the merged values. While it is difficult to identify error sources specifically, we find that typical estimated systematic errors (ranging from ~5 to 15 % for composition data) are consistent with the magnitude of observed relative biases.

The GOZCARDS HCl merged record in the upper stratosphere enables us to track long-term changes in this reservoir for stratospheric chlorine, and by implication, total stratospheric chlorine. The long-term increase in HCl prior to the late 1990s, and the subsequent gentler decrease in the 21st century, are delayed manifestations of changes in the sum of the surface source gas abundances as a result of regulations from the Montreal Protocol (and its amendments). From 1997 to 2010, the average rate of change in upper stratospheric HCl (50° S to 50° N) was about −0.4 to −0.7 % $yr^{-1}$. There are smaller rates of decrease and a flattening or slight turn-around after 2003. In the lower stratosphere, where Aura MLS data weigh in heavily, recent short-term variations have shown a flattening out and, in particular for northern midlatitudes and at 50–70 hPa for the deep tropics, a significant reversal and increasing trend, compared to the decrease from the late 1990s to about 2004. Mahieu et al. (2014) have discussed the reversal in total column HCl trends for 2007–2012 (for northern, not southern midlatitudes), based on ground-based FTIR series at various sites; they also showed that column trends agree with those from lower stratospheric GOZCARDS abundances for the appropriate latitude bands. However, lower stratospheric HCl tendencies appear to





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be reversing in recent years (2011–2014), with decreases at northern midlatitudes (see Fig. 7 for 32 hPa) and some increasing tendencies at southern midlatitudes (Fig. 7). Continued data will be needed to track such short-term changes in HCl, but we expect to see long-term global HCl decreases in both upper and lower stratosphere, as long as the Montreal Protocol and its amendments are adhered to; also, surface chlorine shows smaller rates of decrease in recent years, for reasons that are largely understood (Sect. 3).

For water vapor, we have merged monthly mean datasets for 1991–2013 from the same satellite instruments as for HCl using the same basic methodology, except for the addition of 1991–1993 UARS MLS data, and the inclusion of Aura MLS $H_2O$ data for all pressures. Mostly at the uppermost (mesospheric) altitudes, large variations that are anti-correlated with solar flux are clearly observed over the past two 11 year solar cycles, as discussed previously by others, using shorter data records. Net long-term trends in lower stratospheric water vapor are quite small if one considers the past 22 years, but there has been considerable interannual change, as mentioned also in past work using satellite data and Boulder sonde records. Notably, the steep drop (by 0.5–0.8 ppmv depending on latitude and pressure from 46 to 100 hPa) from 2000 to 2001 is clearly visible in the GOZCARDS record. While the trends have been generally positive in the decade following 2001 (see Sect. 3), the 68 and 100 hPa levels show equally steep decreases again from 2011 to 2013 (from $\sim 0.5$ ppmv for 60° S to 60° N averages to $\sim 0.8$ ppmv in the 20° S–20° N bin at 68 hPa); see also Urban et al. (2014). Long-term stratospheric trends may be observable most readily in the upper stratosphere. In the past 22 years, long-term global $H_2O$ increases of order 10 % are observed in the upper stratosphere and lower mesosphere, whereas a decrease of nearly 10 % has occurred in the lower stratosphere (near 70–100 hPa). However, there is no regular monotonic change on decadal timescales, especially in the tropical lower stratosphere, where fairly sharp decreases followed by steadier increases may be a recurrent pattern. This remains to be better understood, with ongoing global datasets; Fueglistaler (2012) recently discussed the possibility of stepwise changes in





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water vapor. We have displayed the seasonal and decadal variability in stratospheric and mesospheric $H_2O$ based on the GOZCARDS records for the past 22 years. As one might expect from the well-documented temperature influence on the tropical lower stratosphere, the $H_2O$ variability based on maximum minus minimum yearly averages, is largest in the tropics and just above the tropopause. The elucidation of lower stratospheric water vapor changes over multiple decades is complicated by the significant low frequency variability in this region, and the occurrence of sudden changes; the addition of a few years of data can significantly modify trend results. More accurate studies of seasonal to decadal water vapor variability will be enabled by continuing merged $H_2O$ data from the lower stratosphere to the upper mesosphere. A reduction in model spread for stratospheric $H_2O$ is likely easier to achieve than tighter model results for water vapor (and ice water content) in the upper troposphere (for this region, see the data/model comparisons by Jiang et al., 2012). We should continue to improve model comparison results for $H_2O$ (see, for example, Gettelman et al., 2010), although some studies should still focus on non-zonal aspects such as the Asian monsoon or Western Pacific regions, rather than zonal means like GOZCARDS.

For ozone, we have used measurements from SAGE I, SAGE II, HALOE, UARS MLS, Aura MLS and ACE-FTS to produce a merged data record from late 1979 onward, after adjusting the monthly zonal mean series to SAGE II averages. Some complications arose because of the conversion of the original SAGE II profiles from a density/altitude grid to the GOZCARDS mixing ratio/pressure grid. In particular, we observed temperature-related temporal drifts in the converted SAGE II series, as a result of the NCEP temperature data used in this conversion, mostly in the upper stratosphere after June 2000 (see also McLinden et al., 2009). To circumvent this issue, we used HALOE upper stratospheric $O_3$ ($p < 4$ hPa) as a reference for July 2000 to November 2005, after applying offsets to the HALOE series. Aura MLS and ACE-FTS provide the continuing data past 2004. The resulting GOZCARDS merged ozone profiles for northern midlatitudes have recently been used in regression analyses (Nair et al., 2013) to reveal significant decreases in the whole stratosphere during 1984–





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1996. Nair et al. (2015) extended this work to a broader range of latitudes; they found significant increasing trends in upper stratospheric GOZCARDS ozone since 1997, but no significant positive trends in the lower stratosphere. Studies of GOZCARDS (and other) $O_3$ profile trends have been recently discussed as part of the $SI^2N$ analyses (Tummon et al., 2014; Harris et al., 2015), among other efforts.

Here, we looked into the consistency of column ozone data between stratospheric GOZCARDS results and the study by Ziemke and Chandra (2012), who noted, using a simple regression model, that a fairly rapid change ("recovery") in near-global ozone columns from TOMS and OMI data could be inferred since the mid-1990s. We show here that, unlike the 2 to 3 % net increase in near-global column ozone reported by ZC12 after the late 1990s, the similarly analyzed GOZCARDS record does not show such a strong reversal (or an upturn of more than 0.5–1 % since that period). Reasons for these differences could include data coverage or merging-related issues in either dataset, as well as differences in column sensitivities, as column ozone data down to certain pressures (as used in GOZCARDS analyses) cannot be exactly equivalent to stratospheric or total column estimates from ZC12; for example, changes that occur at the lowest altitudes may not be that well captured (for all latitudes) by GOZCARDS-derived columns. Further studies regarding the consistency of various column ozone results and a recovery tendency are warranted; a recent global total ozone study (Shepherd et al., 2014) also points to less of a return towards the 1980 levels than implied by ZC12.

We also briefly described the creation of $N_2O$ and $HNO_3$ GOZCARDS data records, based on Aura MLS and ACE-FTS monthly mean time series. The agreement between these two instruments' datasets for these species was shown to be generally very good. For $HNO_3$, UARS MLS $HNO_3$ source datasets in the GOZCARDS format are available from the authors. However, a small upward adjustment (of order 10 %) to the UARS MLS values is most likely needed based on our preliminary work comparing these time series to $HNO_3$ column results from FTIR measurements. More detailed work should help determine if adjustments can indeed be made in a more generalized way to the





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global UARS MLS $HNO_3$ dataset; lacking this, one should ensure that the error bars reflect the likely biases that can affect the continuity between satellite $HNO_3$ datasets before and after 2000, given the multi-year gap in satellite coverage for this species.

## Appendix A:

### A1 GOZCARDS data provenance

The general origin of the datasets is summarized here. Data coverage from limb sounders (including the instruments used here) is displayed nicely in the work by Toohey et al. (2013).

#### SAGE I

SAGE I was launched 18 February 1979, aboard the Applications Explorer Mission-B (AEM-B) satellite. SAGE I was a sun photometer using solar occultation (Chu and McCormick, 1979), and it collected a global database for nearly three years on stratospheric aerosol, $O_3$, and $NO_2$. For more information, the reader is referred to http://sage.nasa.gov/SAGE1.

#### SAGE II

SAGE II was launched aboard the Earth Radiation Budget Satellite (ERBS) in October 1984 and its data gathering period ended in August 2005. During each sunrise and sunset, SAGE II measured stratospheric aerosols, $O_3$, $NO_2$, and $H_2O$ via solar occultation. This long dataset has proven very valuable in determining past ozone trends. For more information on and data access to the (V6.2) dataset used for GOZCARDS, the reader is referred to http://sage.nasa.gov/SAGE2.





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## HALOE

Since its launch on 12 September 1991 from the Space Shuttle Discovery until November 2005, UARS HALOE collected profiles of atmospheric composition and temperature. HALOE (Russell et al., 1993) used solar occultation to measure vertical profiles of $O_3$, HCl, HF, $CH_4$, $H_2O$, NO, $NO_2$, temperature, aerosol extinction, and aerosol composition and size distribution. More information and access to the HALOE data can be obtained from http://haloe.gats-inc.com and http://disc.sci.gsfc.nasa.gov/UARS/data-holdings/HALOE. For GOZCARDS purposes, we have used Version 19 HALOE netCDF data files available at http://haloe.gats-inc.com.

## UARS MLS

This instrument observed the Earth's limb in microwave emission using three radiometers, at frequencies near 63, 183 and 205 GHz (Waters, 1993; Barath et al., 1993), providing unique daily global information on stratospheric ClO, along with other profiles, including $O_3$, $H_2O$, $HNO_3$, temperature, and cloud ice water content. The stratospheric $H_2O$ data ceased on 15 April 1993, after the failure of the 183 GHz radiometer. After 15 March 1994, measurements became increasingly sparse in order to conserve the life of the MLS antenna scan mechanism and UARS power. Data exist until 28 July 1999, although for GOZCARDS, only data through mid-June 1997 are used, as data sparseness and degradation of the 63 GHz radiometer led to less "trend-quality" data after this. Sampling patterns follow the alternating yaw cycles imposed on MLS by the precessing UARS orbit; MLS measurements were obtained continuously for all latitudes between 34° S and 34° N, with higher latitudes covered in either the northern or Southern Hemisphere with a roughly 36 day cycle. Livesey et al. (2003) provide more information on the UARS MLS instrument, retrievals, and results. For data access, the reader is directed to the relevant Goddard Earth Sciences and Information Services Center (GES DISC) data holdings at





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http://disc.sci.gsfc.nasa.gov/UARS/data-holdings/MLS. L3AT data files were used as the basis for the production of the GOZCARDS UARS MLS monthly source datasets.

### ACE-FTS

ACE-FTS is the primary instrument onboard the SCISAT satellite, launched on 12 August 2003. It is a high spectral resolution ($0.02\,cm^{-1}$) Michelson interferometer operating from 2.2 to 13.3 µm ($750–4400\,cm^{-1}$); see Bernath et al. (2005) for an overview of the ACE mission. The instrument can simultaneously measure temperature and many trace gases (including all the species mentioned here for GOZCARDS), thin clouds, and aerosols, using the solar occultation technique. ACE-FTS data version 2.2, along with the version 2.2 update for ozone, were used here for GOZCARDS. For access to the public ACE-FTS datasets, with a routine measurement start date of March 2004, the reader is directed to http://www.ace.uwaterloo.ca.

### Aura MLS

MLS is one of four instruments on NASA's Aura satellite, launched on 15 July 2004. Aura MLS is a greatly enhanced version of the UARS MLS experiment, providing better spatial coverage, vertical resolution, and vertical range, along with more continuous data over its lifetime (and with ongoing measurements at the time of writing). The instrument includes radiometers at 118, 190, 240, and 640 GHz, and a 2.5 THz module (Waters et al., 2006). Aura MLS provides measurements of many chemical species, cloud ice, temperature and geopotential height. Continuous measurements have been obtained since August 2004, with the exception of OH, for which sparser measurements exist since August 2010, in order to preserve the life of the THz module. For more information and access to the Aura MLS datasets, the reader is referred to http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/MLS. For GOZCARDS, we use the currently recommended Aura MLS data versions (version 2.2/2.3 for ozone and 3.3/3.4 for other species).





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## A2 Calculation details for the iterative merging procedure

Given three time series, the merging procedure that we use first combines two out of the three time series, $y_1(i)$ and $y_2(i)$ (where index $i$ represents time for each monthly mean value in a given latitude/pressure bin). We first obtain the temporary merged series $m_1(i)$ via:

$$m_1(i) = 0.5(y_1(i) + y_2(i)) \tag{A1}$$

with the average offsets for $y_1(i)$ and $y_2(i)$ being $(1/(2n_{12}))\ \Sigma(y_1(i) - y_2(i))$ and $-1$ times this value, respectively; $n_{12}$ is the number of overlapping data points between the two time series. Then, we merge together the time series $m_1(i)$ and $y_3(i)$, keeping the weightings equal for all 3 time series (1/3 for each), so that we calculate the new merged time series $m(i)$ via:

$$m(i) = w_m m_1(i) + w_3 y_3(i) = (1/3)(y_1(i) + y_2(i) + y_3(i)) \tag{A2}$$

which will hold if the weights are $w_m = 2/3$ and $w_3 = 1/3$ (given Eq. A1) for $m_1(i)$). The average reference value (to which the adjustments of $m_1(i)$ and $y_3(i)$ in the 2nd step are made) is given by $(1/n_m)\Sigma((2/3)m_1(i) + (1/3)y_3(i))$, where $n_m$ represents the number of (overlapping pairs of) data values used in step 2. For the HCl and $H_2O$ data merging procedure, we always use the Aura MLS time series as one of the first two series involved in the initial merging step, for example as $y_1(i)$, in order to maximize the overlap between the first two series and obtain more robust offset values. Then, we use the 3rd time series; the order used for HALOE and ACE-FTS (i.e., whether we use HALOE or ACE-FTS for $y_2$ or $y_3$) makes very little difference.





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### Calculation of the SD for the merged data values

The average and SD (square root of variance) for each $y_k$ value (i.e. for each monthly zonal mean in a particular lat/p bin) are calculated from Eqs. (A3) and (A4) below:

$$\overline{y_k} = \frac{1}{n_{yk}} \sum_j y_{kj} \tag{A3}$$

and, for the variance,

$$\sigma^2_{yk} = \frac{1}{n_{yk} - 1} \sum_j (y_{kj} - \overline{y_k})^2 \tag{A4}$$

where index "$j$" corresponds to individual data values within a month, index $k$ represents a given instrument (data source), and $n$ is the total number of data values for a given bin and source (instrument) time series point in time (or month). Each value $\overline{y_k}$ above is a monthly average (although we also use instead the simpler notation $y_k$), with SD about the mean $\sigma_{yk}$. Now, given the merged series $u(i)$ (where index $i$ runs over a large number of months), the SD of each merged data point (for a given month) can be obtained by considering the original datasets $y_{kj}$ that were used to construct $u$. Specifically, we have the variance for the merged dataset

$$\sigma^2_u = \frac{1}{n_u - 1} \sum_j (u_j - u_{\text{ref}})^2 \tag{A5}$$

where $u_{\text{ref}}$ is the merged value (which is not necessarily chosen to be the average value $\overline{u}$) and the $u_j$ values represent the union of adjusted data values that make up the merged product, with the index $j$ for this combined dataset covering all values (up to the total $n_u$) obtained from the original source values $y_{kj}$. In practice, we do not keep track of the individual data values that went into making the averages for the series $y_k$ that are being merged, and we need to obtain $\sigma_u$ based solely on the values $\overline{y_k}$, $\sigma_{yk}$,



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and the original number of points for each dataset $y_k$, namely $n_{yk}$. If we consider all the original values, we have a combined dataset with $n_u$ points, such that $n_u = \sum_k n_{yk}$. Now, expanding Eq. (A5), we get

$$(n_u - 1)\sigma_u^2 = \sum_j \left( u_j^2 + u_{\text{ref}}^2 - 2u_{\text{ref}} u_j \right) \tag{A6}$$

or

$$(n_u - 1)\sigma_u^2 = \sum_j u_j^2 + n_u u_{\text{ref}}^2 - 2u_{\text{ref}} \sum_j u_j \tag{A7}$$

Expanding Eq. (A4) for each individual dataset $y_k$, we get

$$(n_{yk} - 1)\sigma_{yk}^2 = \sum_j y_{kj}^2 + \overline{y_k}^2 - 2\overline{y_k} \sum_j y_{kj} \tag{A8}$$

which leads to

$$\sum_j u_j^2 = \sum_{\text{k, j}} y_{kj}^2 = \sum_k (n_{yk} - 1)\sigma_{yk}^2 + \sum_k n_{yk} \overline{y_k}^2, \tag{A9}$$

so that extracting the variance from Eq. (A7) now leads to

$$\sigma_u^2 = \frac{1}{(n_u - 1)} \left( \sum_k (n_{yk} - 1)\sigma_{yk}^2 + \sum_k n_{yk} \overline{y_k}^2 + n_u u_{\text{ref}}^2 - 2u_{\text{ref}} \sum_k n_{yk} \overline{y_k} \right) \tag{A10}$$

The adjusted time series are obtained from the original series $y_k$ as $Y_k$, and we can write Eq. (A4) in the same manner for the $Y_k$ data values, namely

$$\sigma_{\text{Yk}}^2 = \frac{1}{n_{yk} - 1} \sum_j (Y_{kj} - \overline{Y_k})^2 \tag{A11}$$





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with $\sigma_{Yk} = \sigma_{yk}$ as the adjustments (offsets) are performed in an additive manner; if these adjustments were performed using multiplicative factors, those factors would also have to be considered in a multiplicative way to get the new $\sigma_{Yk}$ values. We can thus write Eq. (A10) for the adjusted datasets as:

$$\sigma_u^2 = \frac{1}{(n_u - 1)} \left( \sum_k (n_{yk} - 1)\sigma_{yk}^2 + \sum_k n_{yk} \overline{Y_k}^2 + n_u U_{\mathrm{ref}}^2 - 2U_{\mathrm{ref}} \sum_k n_{yk} \overline{Y_k} \right) \tag{A12}$$

Equation (A12) for the SD of the merged dataset simplifies if the original datasets are adjusted to exactly the same reference value ref ($\overline{Y_k}$ = ref) and the merged value $U_{\mathrm{ref}}$ is also equal to that value, as the sum of the last 3 terms in Eq. (A10) (with $Y_k$ replacing $y_k$) then reduces to $n_u \mathrm{ref}^2 + n_u \mathrm{ref}^2 - 2n_u \mathrm{ref}^2$, which is zero. In this case, one obtains

$$\sigma_u^2 = \frac{1}{(n_u - 1)} \left( \sum_k (n_{yk} - 1)\sigma_{yk}^2 \right) \tag{A13}$$

However, in general, one should use Eq. (A12) for the SD of the merged dataset, given the adjusted datasets $\overline{Y_k}$ and the merged (or reference) value $U_{\mathrm{ref}}$. Also, we often use a merged value equal to the average of the original data (over a given overlap period), so that

$$U_{\mathrm{ref}} = \frac{1}{n_y} \sum_k \overline{y_k} \tag{A14}$$

where $n_y$ is the total number of datasets ($y_k$), as opposed to having the merged value place more weight on the larger datasets (e.g., for emission-type measurements vs. occultation-type), in which case one would consider using $U_{\mathrm{ref}} = \frac{1}{n_u} \sum_k n_{yk} \overline{y_k}$. For ozone, we use a particular dataset (SAGE II ozone) as the primary reference, but Eq. (A12) can be used to obtain the SD for the merged dataset (about the SAGE II reference) in that case also. While it is useful to have the formalism above for obtaining the



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merged dataset SD $\sigma_u$, we often find significant differences between the SD of various datasets, so that this effect will have the greatest influence on the results, as opposed to the impact of the last 3 terms in the summation (in Eq. A12). Finally, it is easy to test Eq. (A12) (and we have done so) by using synthetic series and calculating the SD of the combined set. In reality, the SD of the time series monthly mean values are typically larger for MLS than for ACE-FTS, mainly because of the more complete sampling of variability from the daily global measurements acquired by MLS. Sample plots for SD and standard errors in the case of HCl are shown in Fig. A1. As expected, merged SD follow the SD from HALOE HCl before August 2004 and those from MLS HCl after this time. However, the merged standard errors for the MLS time period follow the smaller MLS standard errors, because these values vary inversely with the square root of the number of values sampled, and are therefore made smaller by the significantly larger daily and monthly MLS sampling rate and coverage.

## A3 Procedural merging details for GOZCARDS HCl, $H_2O$, and $O_3$

### A3.1 HCl

- The vertical data range for valid HCl merged values is between 0.46 and 147 hPa (inclusive), as a result of data sparseness or data quality issues outside these ranges.
- At 147 hPa, no merged HCl values exist for latitude bins from 35° S to 35° N inclusive, because of unrealistically large Aura MLS HCl values in this region; also, there is not enough data at this level to provide a meaningful product from HALOE and ACE-FTS data alone.
- Because of occasional small negative merged values during Southern Hemisphere polar winter, we did not apply HCl data offsets in the lower stratosphere for the 65° S through 85° S bins from June through September and for pressures larger than or equal to 15 hPa. For vertical continuity purposes, we applied this





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method to all lower stratospheric pressure levels, although the small negative merged values only occurred in a small fraction of cases and the impact on the merged values is not large. Seasonal variations in other bins are milder and did not lead to such an Antarctic winter issue; also, this issue did not affect other species.

- As Aura MLS and ACE-FTS data exist in the 85° N and 85° S bins, but there are no HALOE measurements, we could not use our standard merging procedure there. We simply extended the offsets from the adjacent bins (at 75° N and 75° S) to these two bins to obtain a merged record after 2004 that exhibits continuity vs. latitude.

- At 100 hPa, we used HCl offsets from the 5° S bin for the 5° N bin, as there was insufficient data from the three combined datasets in the latter bin to calculate meaningful offsets and merge the datasets. This procedure seems reasonable, given that the time series in these two adjacent tropical latitude bins (during years outside the 2004/05 overlap period) look continuous and stable enough to justify identical adjustments in both bins and to avoid a data gap in the merged series at 5° N, although this does imply somewhat larger error bars at 5° N.

### A3.2 $H_2O$

- The vertical data range for valid $H_2O$ merged values is between 0.01 and 147 hPa (inclusive). Some $H_2O$ data exist at 147 hPa for low latitudes, but more careful work would be needed to extend the merged data globally in such a region.

- Users should keep in mind the PMC-related caveats mentioned in Sect. 4 for summer at high latitudes in the upper mesosphere, prior to the end of the HALOE dataset (November 2005).

- As for HCl, we could not use our standard merging procedure at the two most poleward latitude bins; we simply extended the offsets from the adjacent bins (at





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75° N and 75° S) to these polar bins to obtain a merged record after 2004 that exhibits continuity vs. latitude.

– Also as for HCl, at 100 hPa, we used $H_2O$ offsets from the 5° S bin for the 5° N bin, as there was insufficient data from the combined datasets in the latter bin to calculate meaningful offsets and merge the datasets. This procedure avoids a data gap in the merged series at 5° N.

### A3.3 $O_3$

**Screening of SAGE $O_3$ data**

For SAGE I $O_3$, the main uncertainty is aerosol interference, especially below 15 to 20 km. All SAGE I values below (in altitude) where the aerosol extinction at 1.0 µm reaches a value larger than $1.0 \times 10^{-3}\,km^{-1}$ are removed from the analysis (L. W. Thomason, personal communication, 2012).

For SAGE II ozone, the screening steps are based on Wang et al. (2002) as follows:

– We removed the entire ozone profile when any reported error bar value exceeded 10 % between 30 and 50 km, in order to filter out outliers affected by "short events" (Wang et al., 2002), which mainly occurred between mid-1993 and mid-1994, when SAGE II had a battery problem. In order to preserve power, sunset measurements were started later than normal while sunrise measurements were ended earlier. These "short events" had fewer extraterrestrial solar irradiance measurements for calibration and normalization.

– We used aerosol extinctions and extinction ratios to remove data affected by clouds, and aerosols from the June 1991 Mt. Pinatubo eruption. $O_3$ data were removed when the aerosol extinction at 0.525 µm exceeded $6 \times 10^{-3}\,km^{-1}$, thus removing data affected by this eruption for months and even years, in the lower stratosphere. For cases with extinctions less than $6 \times 10^{-3}\,km^{-1}$ but greater than



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$1 \times 10^{-3}\,km^{-1}$, and extinction ratios ($0.525\,\mu m/1.02\,\mu m$) $\leq 1.4$, the corresponding data were removed for additional filtering. Although more stringent criteria could be used to remove a few more outliers, this would also remove many more "good" ozone data that are not affected by aerosol/cloud. Fortunately, any artifacts from these few unfiltered data values are greatly reduced after binning the data into monthly zonal means.

- We removed anomalously low $O_3$ values resulting from very small SAGE II transmittances; $O_3$ error values in these cases were capped at 300 % by the algorithm. Such low $O_3$ values (sometimes low by 2–3 orders of magnitude) generally occur close to the tropopause and in the troposphere, and can be identified by using this 300 % error flag (Wang et al., 2002).

- It was found that SAGE II ozone data could be affected during high sun-orbit beta angle conditions (Wang et al., 1996). SAGE II profiles immediately following fully sunlit orbits with absolute values of beta greater than 40° are eliminated from monthly zonal means.

**Other merging details for $O_3$**

- SAGE I monthly mean source data are used for the merged dataset in the tropical bins (25° S to 25° N) from 1 through 68 hPa only and, at higher latitudes, from 1 through 100 hPa only.

- The vertical range for valid $O_3$ merged values is between 0.2 and 215 hPa (inclusive), with the lower altitude bound varying with latitude. The merged product at 147 and 215 hPa has valid data only for the 35 to 85° latitude bins. Indeed, we limited merged data mostly to stratospheric values (larger than ∼ 0.1 ppmv); the upper troposphere is more of a challenge for such a merging activity, given smaller abundances, more challenging measurements, and a larger impact from different instrument resolutions. The upper range limit was chosen to enable studies of the





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upper stratosphere and lower mesosphere, even if this is a region where diurnal ozone change occurs; arguments we have presented (se main text) suggest that the GOZCARDS merged ozone time series variations should not be subject to a large impact from diurnal variations, although high altitude regions should still be treated with caution.

- We omitted the use of UARS MLS at 100 hPa for low latitudes (from 25° S to 25° N), as these monthly values are biased quite high and also exhibit too large a seasonal cycle amplitude, in comparison to HALOE and SAGE II data; this appears to relate to a UARS MLS artifact.

- Since there is no (monthly) overlap between SAGE II and HALOE vs. UARS MLS or Aura MLS in the 85° N and 85° S latitude bins, the same offsets as for 75° N and 75° S (respectively) are applied to the datasets at these two extreme latitude bins, in order to minimize latitudinal discontinuities in the merged data record.

- Because of discontinuities that appeared in merged $O_3$ at high latitudes above the stratopause, particularly in the 75° S bin, we flagged merged values for 75 and 85° (N and S) as bad, for pressures less than 1 hPa. This issue could be the result of a few bad data points or not enough data overlap. To minimize artifacts, we left the resolution of this issue for future investigations; also, the reduced amount of occultation data at these high latitudes makes the usefulness of a merged product with poorly sampled seasonal changes somewhat marginal (for certain years at least, the number of monthly values drops significantly at high latitudes).

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**Table 1.** Characteristics of instrument datasets used to create GOZCARDS ESDRs (version ev1.01).

| Instrument and Data Versions | Platform | Type of measurement | Time period (GOZCARDS source files) | Vertical Resolution (km) | Retrieved quantity and stratospheric vertical grid spacing |
|---|---|---|---|---|---|
| SAGE I<br>V5.9_rev $O_3$ | AEM-2 | Solar occultation VIS/UV and near-IR | Feb 1979–Nov 1981 | 1 | Density on altitude grid<br>1 km spacing |
| SAGE II<br>V6.2 O3 | ERBS | Solar occultation VIS/UV and near-IR | Oct 1984–Aug 2005 | 0.5–1 | Density on altitude grid<br>0.5 km spacing |
| HALOE<br>V19 | UARS | Solar occultation mid-IR | Oct 1991–Nov 2005 | 2.5 | Volume Mixing Ratio on pressure grid with 30 levels per decade (LPD) change in $p$ |
| MLS<br>V5 $O_3$<br>V6 $H_2O$ | UARS | Limb emission microwave/sub-mm | Oct 1991–Jun 1997 (May 1993 end for strat. $H_2O$) | **$H_2O$**<br>3–4 (strat.)<br>5–12 (mes.) | Volume Mixing Ratio on pressure grid with 6 LPD in stratosphere |
| | | | | **$O_3$**<br>3.5–5 (strat.)<br>5–8 (mes.) | 6 LPD in stratosphere |
| ACE-FTS<br>V2.2<br>(V2.2 update for $O_3$) | SCISAT | Solar occultation mid-IR | Mar 2004 through Sep 2010 (2009 for $O_3$) | 3–4 | Volume Mixing Ratio on 1 km grid spacing (height and $p$ provided) |
| MLS<br>V3.3<br>V2.2 $O_3$ | Aura | Limb emission microwave/sub-mm | Aug 2004 through 2012 | **HCl**<br>3–5 | Volume Mixing Ratio on pressure grid with 6 LPD |
| | | | | **$H_2O$**<br>3–4<br>($p > 0.1$ hPa)<br>5–9<br>(0.1–0.01 hPa) | 12 LPD |
| | | | | **$O_3$**<br>3 | 6 LPD |





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**Table 2.** Products and instrument source data making up the available GOZCARDS data records.

| **Merged Products** and pressure range | **Source Datasets** (and years used) |
|---|---|
| **HCl** 147–0.5 hPa | HALOE (1991–2005), ACE-FTS (2004–2010), Aura MLS (2004 onward) |
| **$H_2O$** 147–0.01 hPa | HALOE (1991–2005), UARS MLS (1991–1993), ACE-FTS (2004–2010), Aura MLS (2004 onward) |
| **$O_3$** 215–0.2 hPa | SAGE I (1979–1981), SAGE II (1984–2005), HALOE (1991–2005), UARS MLS (1991–1997), ACE-FTS (2004–2009), Aura MLS (2004 onward) |
| **$HNO_3$** 215–1 hPa | ACE-FTS (2004–2010), Aura MLS (2004 onward) |
| **$N_2O$** 100–0.5 hPa | ACE-FTS (2004–2010), Aura MLS (2004 onward) |
| **Temperature** 1000–0.015 hPa | GMAO MERRA (1979 onward) |





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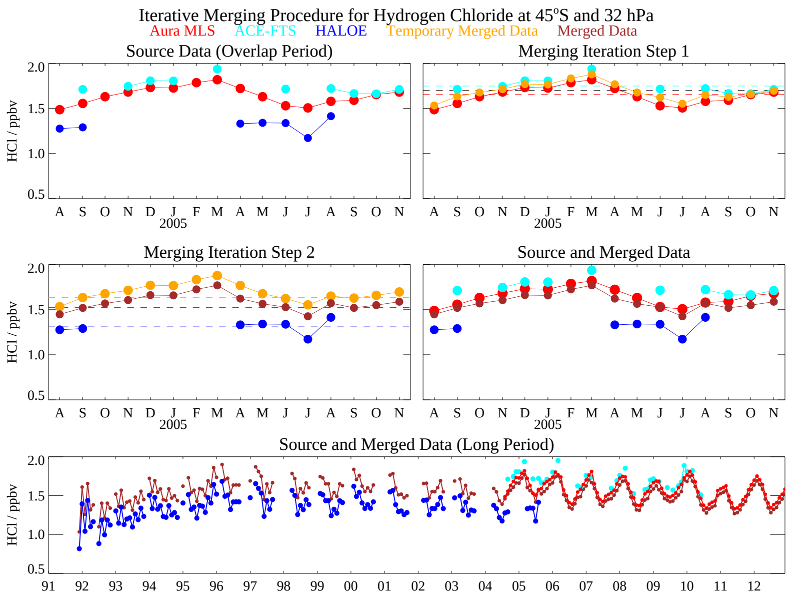


**Figure 1.** Merging procedure illustration for HCl. Top left panel shows the HCl monthly mean source data during the overlap period (August 2004–November 2005) for HALOE, ACE-FTS, and Aura MLS. Top right panel illustrates step 1 in the merging procedure, with the temporary merged data values (orange) resulting from the adjustment of ACE-FTS and Aura MLS values to the reference indicated by the black dashed line. Middle left panel shows the result of step 2, namely the merged values arising from merging HALOE data with the temporary merged values from step 1; the black dashed line is the new average reference value, obtained from a 2/3 and 1/3 weighting of the dashed orange and dashed blue line values, respectively (see text). Middle right panel shows all the source data and the final merged values during the overlap period. Bottom panel shows the source and merged time series from 1991 through 2012 after the calculated additive offsets are applied to the whole source datasets, which are then merged (averaged) together wherever overlap between instruments exists.





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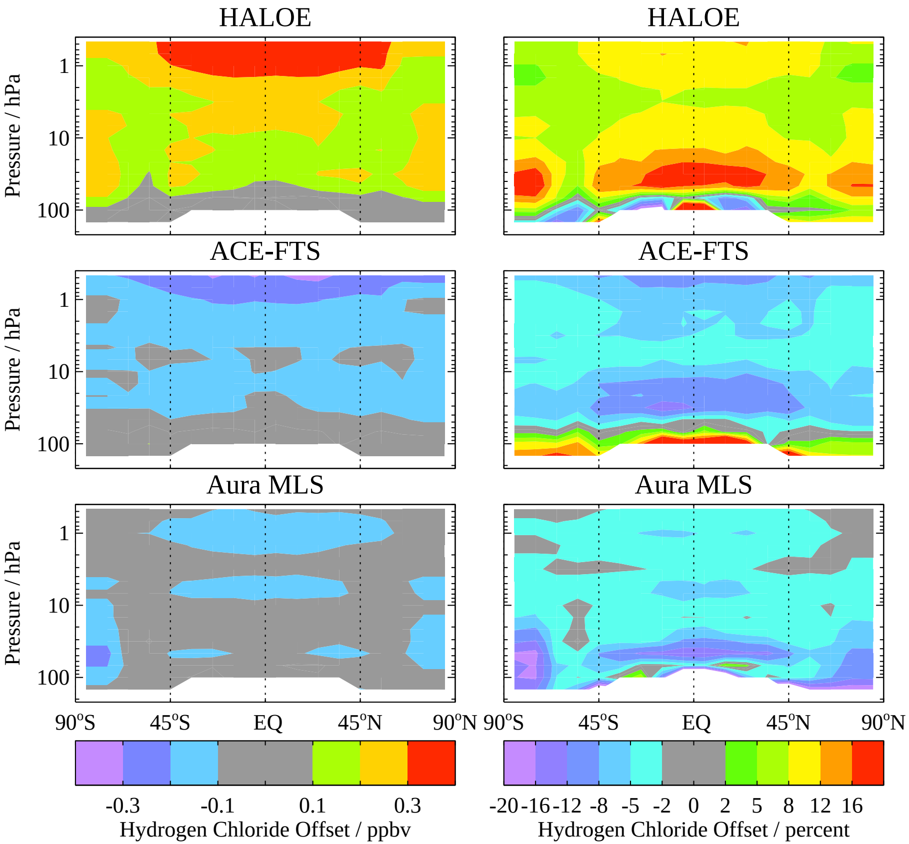


**Figure 2.** Offsets applied to the HCl source datasets (top panels for HALOE, middle panels for ACE-FTS, bottom panels for Aura MLS) as a function of latitude and pressure. The left column gives offsets in ppbv and the right column provides offsets as a percent of the zonal average merged mixing ratios during the overlap period (August 2004–November 2005) used here to compute the average offsets.





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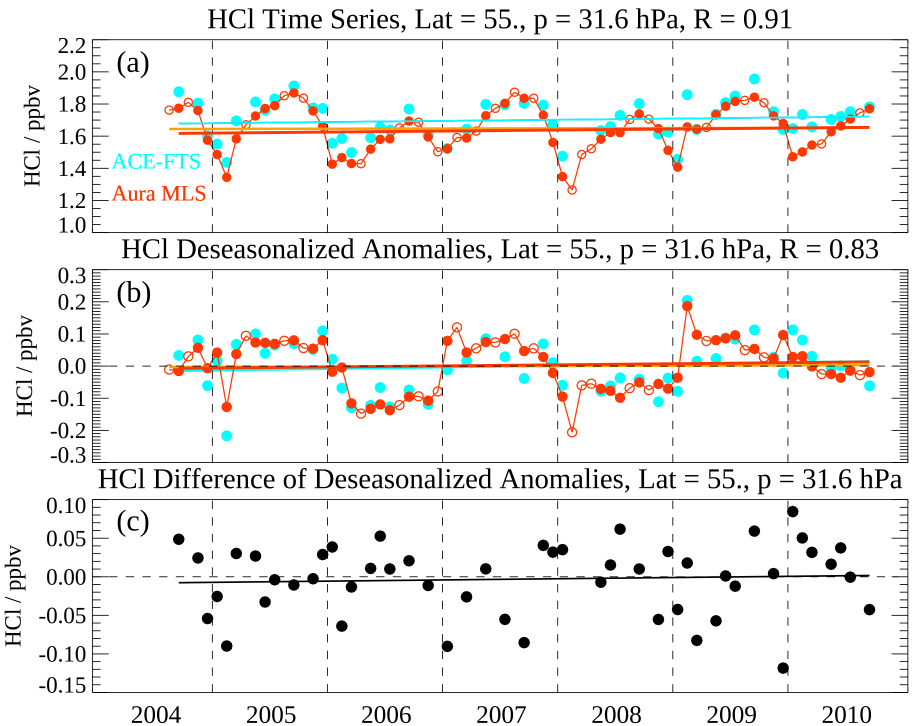


**Figure 3.** Example of HCl time series analyses for 50–60° N and 32 hPa. **(a)** HCl monthly mean source data from ACE-FTS and Aura MLS; the MLS dots are filled when there is time overlap with ACE-FTS, and open if no such overlap exists. Simple linear fits are shown as colored lines for ACE-FTS and for Aura MLS (orange line for all red dots and red line for filled red dots only). Correlation coefficient values ($R$ values) for the two time series are provided in the title. **(b)** Deseasonalized anomalies for both ACE-FTS and Aura MLS, with corresponding linear fits (and $R$ values). **(c)** Difference of deseasonalized anomalies (ACE-FTS minus Aura MLS), with linear fit.





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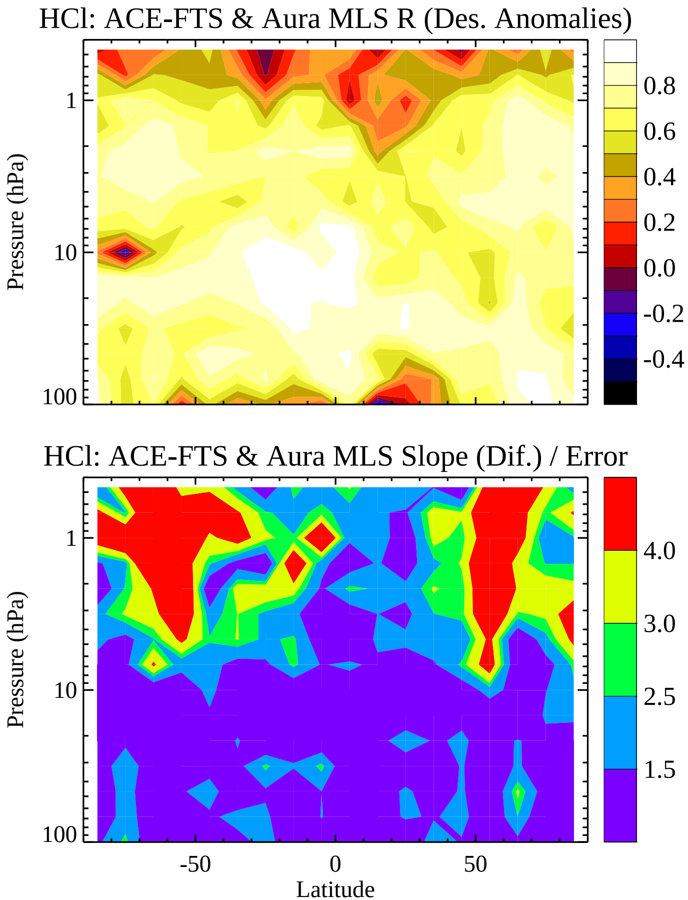


**Figure 4.** Latitude/pressure contours of time series diagnostics obtained from analyses illustrated in for HCl from Aura MLS and ACE-FTS. Top panel: correlation coefficient for the deseasonalized time series. Bottom panel: ratio of the slope of the difference between deseasonalized series over the error in this slope.

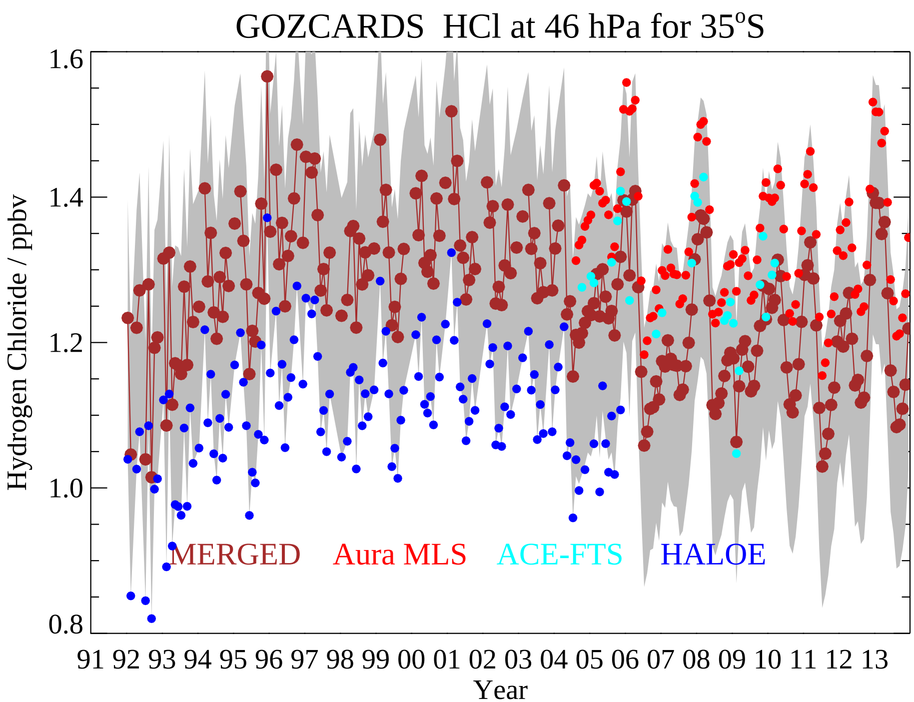


**Figure 5.** Illustration of GOZCARDS HCl monthly averages with systematic error estimates (shown as grey shading) at 46 hPa for 30–40° S; see text for the meaning of this shaded region. The source data from HALOE, Aura MLS, and ACE-FTS are shown in different colors (see legend), along with the merged values.

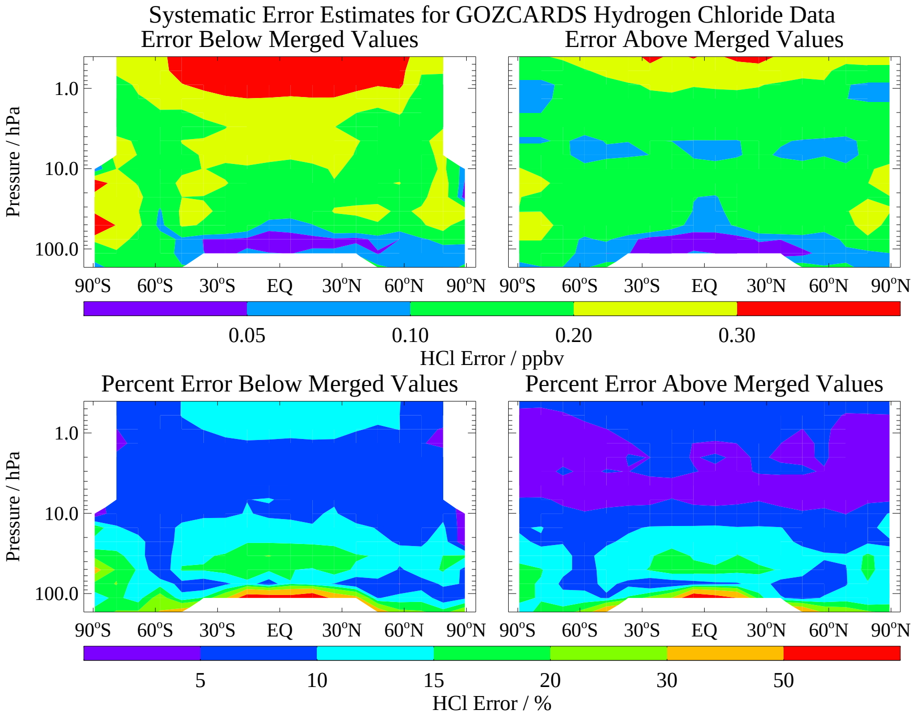


**Figure 6.** Systematic error estimates for GOZCARDS HCl. One error (left panels) is relevant for values lower than (below) the merged values, and one (right panels) for values larger than the merged values; the top panels give the error estimates in ppbv, and the bottom panel errors are expressed as percent of the average merged values over the relevant time periods (see text). These error bars provide a range within which 95 % of the source data values lie.



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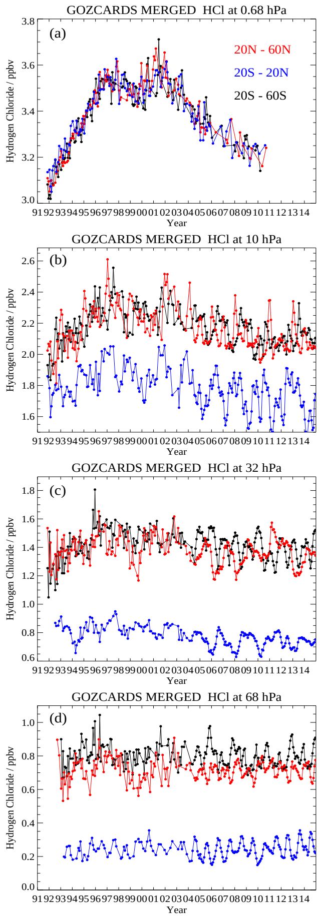


**Figure 7.** Time series of the GOZCARDS monthly-averaged merged HCl abundance for 3 different latitude bin averages (see color legend in **a**) for **(a)** 0.7 hPa, **(b)** 10 hPa, **(c)** 32 hPa, and **(d)** 68 hPa.





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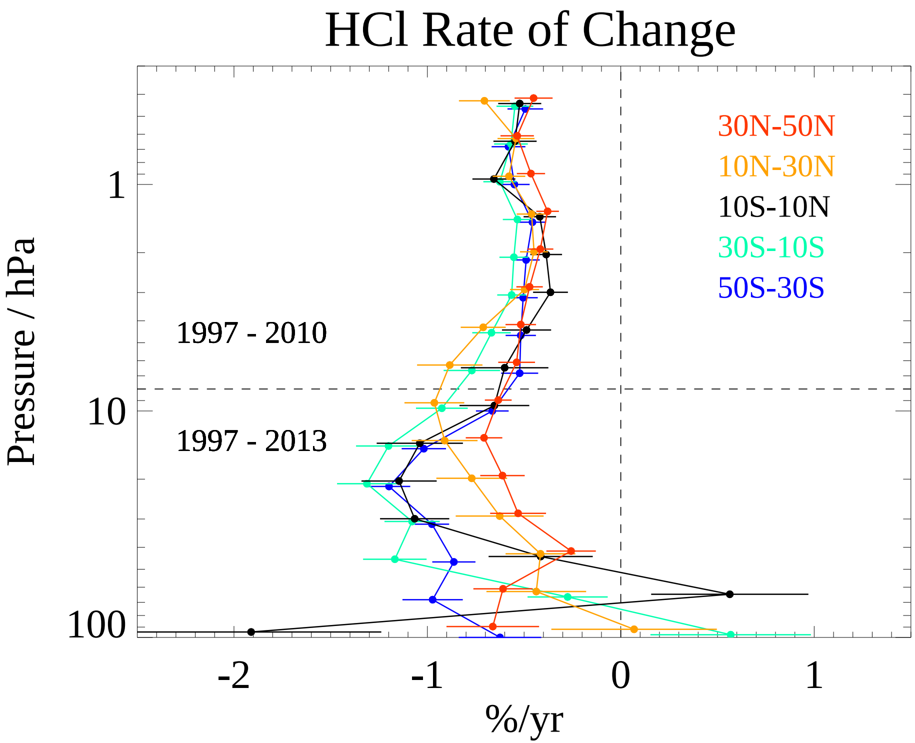


**Figure 8.** The average rate of change (percent per year) for HCl as a function of pressure for different latitude bin averages (see legend) for time periods corresponding to the appropriate GOZCARDS HCl values (see text) in the upper stratosphere (January 1997–September 2010) and lower stratosphere (January 1997–December 2012). Deseasonalized monthly data were used to obtain a long-term trend for these time periods; two-sigma error bars are shown.





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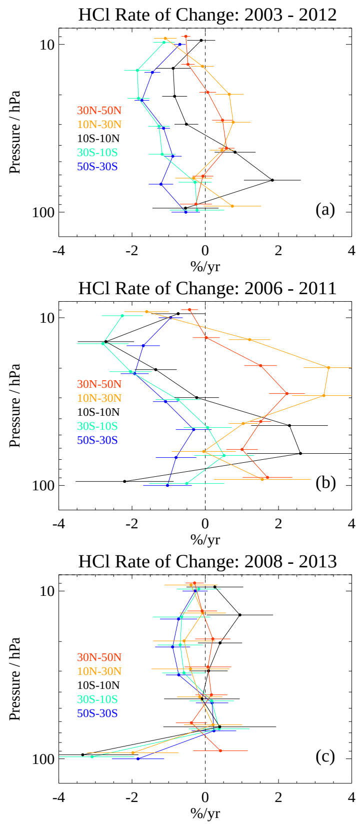


**Figure 9.** Same as Fig. 8, but for the lower stratosphere only and using different time periods to illustrate shorter-term changes in this region. Average rates of change in HCl are given for **(a)** 2003 through 2012, a decade exhibiting significant differences between northern and southern hemispheric change, **(b)** the 6 year period 2006 through 2011, when the largest changes occurred, and **(c)** the most recent 6 year period 2008 through 2013, when a significant decrease in the variability took place.





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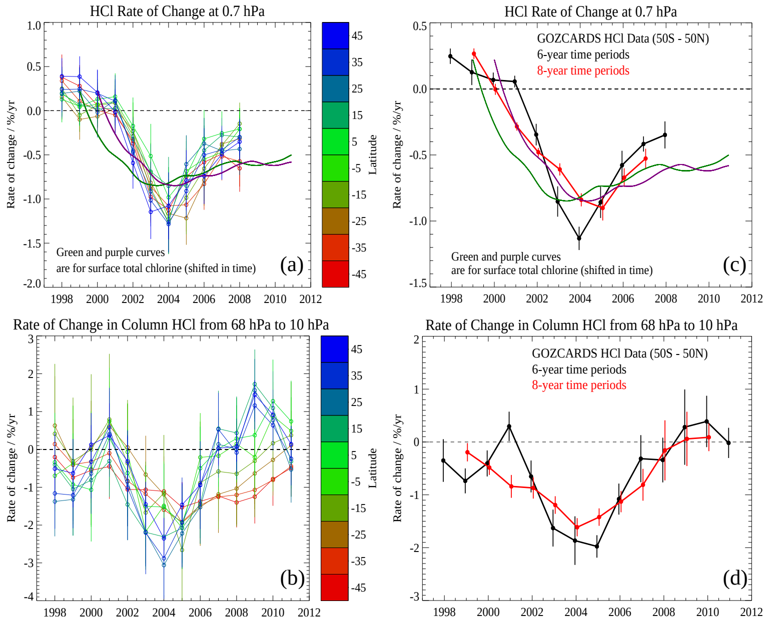


**Figure 10.** Rates of change for GOZCARDS HCl (connected open circles) are given as a function of latitude in 10° latitude bins for sliding 6 year periods centered on 1 January of each year (e.g., the 1998 point is an average for data from 1995 through 2000, and the 2011 point is for data from 2008 through 2013). **(a)** is for changes in upper stratospheric HCl at 0.7 hPa and **(b)** is for the change in the integrated HCl column between 68 and 10 hPa. The two additional curves in **(a)** represent the rates of change in the estimated surface total chlorine from NOAA data (green is for a 6 year time shift, and purple for a 7 year time shift, to account for transport time to the upper stratosphere); see text for more details. (**c** and **d**) are similar to (**a** and **b**), respectively, in terms of the pressure levels used, but the rates of change are averaged over all latitude bins covering 50° S to 50° N for two sets of sliding time periods (black is for 6 year periods, red is for 8 year periods). As in **(a)**, surface total chlorine variations are also displayed in **(c)**. Error bars indicate twice the standard errors in the means.





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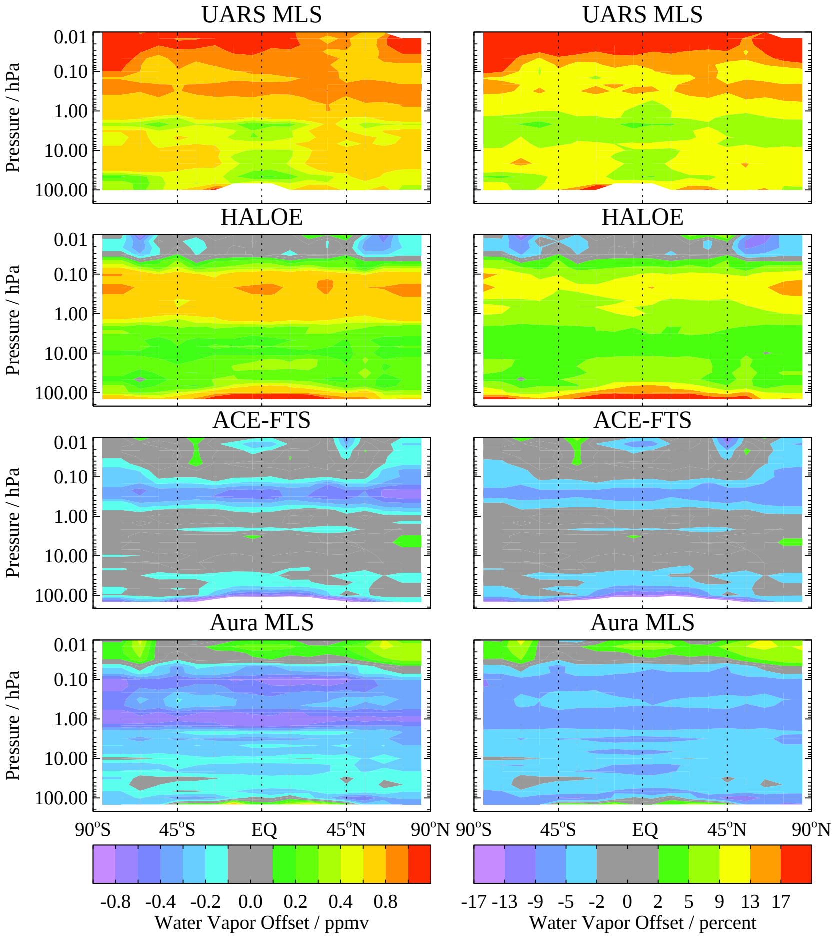


**Figure 11.** Offsets applied to the $H_2O$ source datasets as a function of latitude and pressure, similar to Fig. 2 for HCl.





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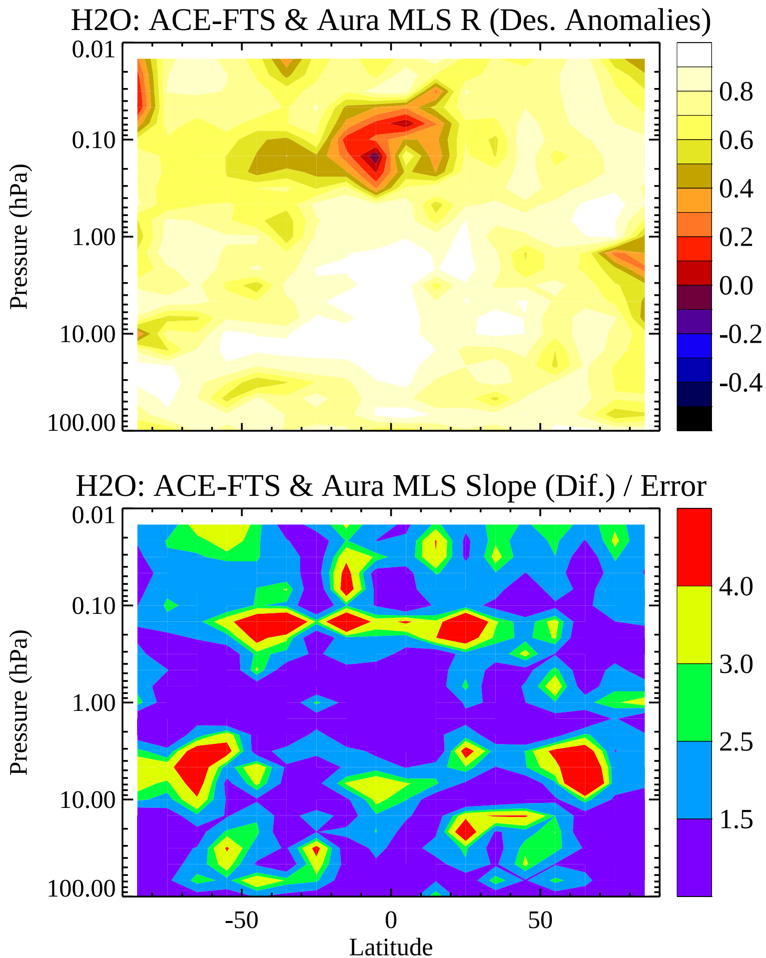


**Figure 12.** Latitude/pressure contours of time series diagnostics for $H_2O$ from Aura MLS and ACE-FTS; this is similar to Fig. 4 for HCl.





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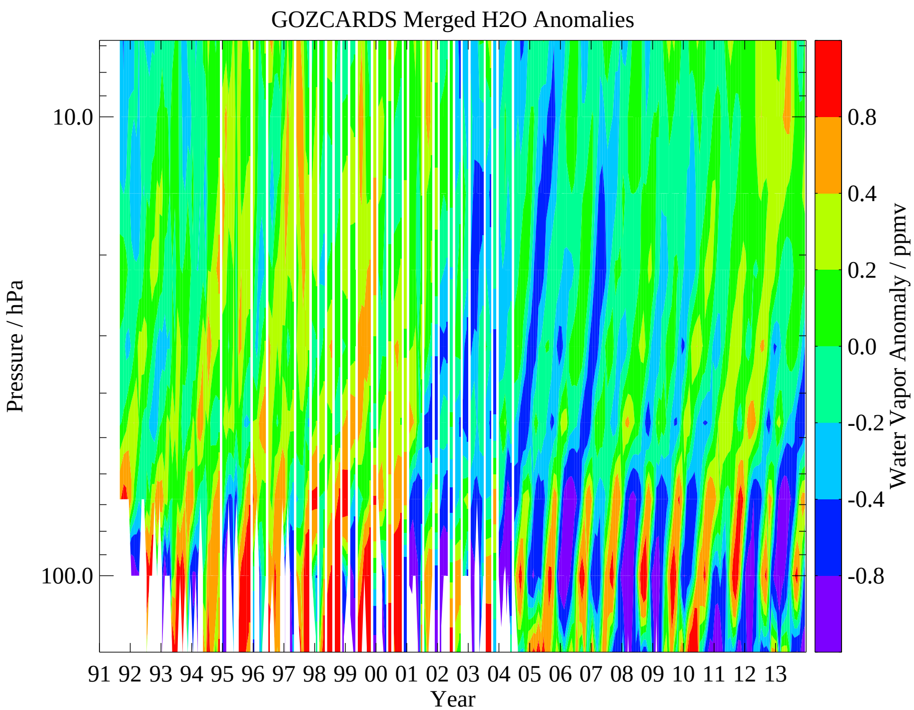


**Figure 13.** A depiction of the "tape recorder" evolution for tropical water vapor abundances from 147 to 10 hPa for October 1991 through December 2013. This plot was produced from GOZCARDS merged $H_2O$ time series anomalies (differences from the long-term means) for the average of the 4 tropical bins covering 20° S to 20° N.





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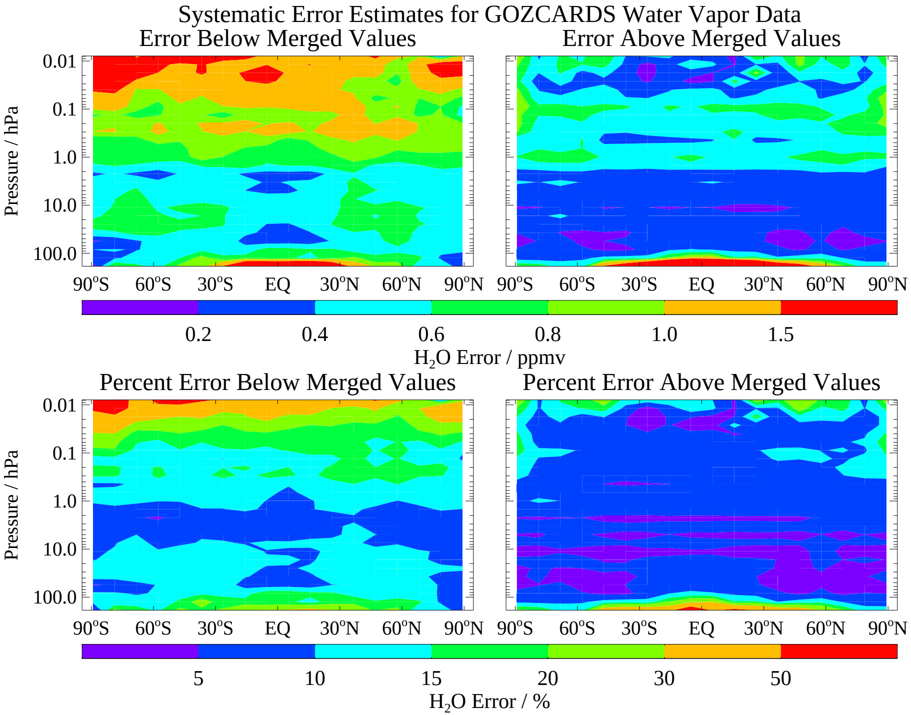


**Figure 14.** Systematic error estimates for GOZCARDS $H_2O$ (similar to Fig. 6 for HCl).





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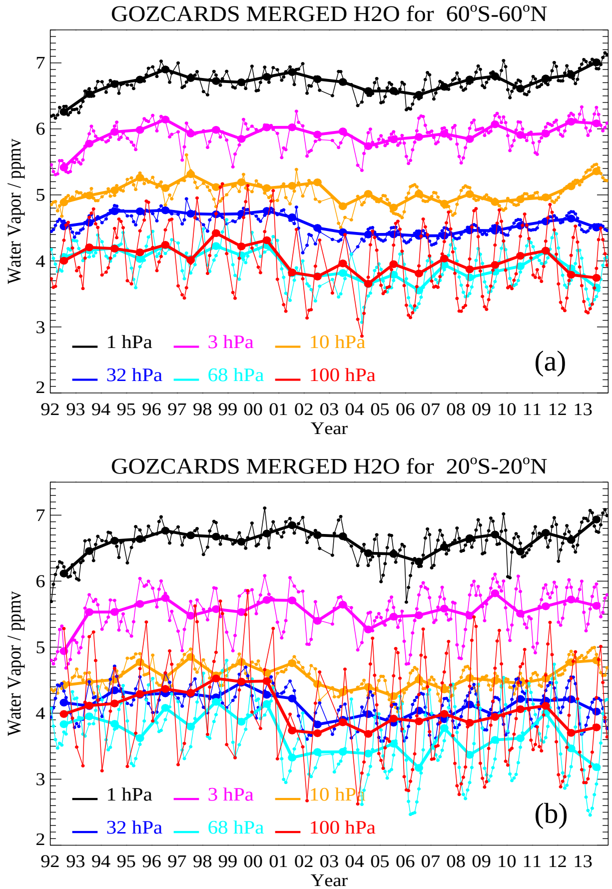


**Figure 15.** Variations in stratospheric water vapor from the GOZCARDS $H_2O$ merged data records (1992 through 2013) averaged from **(a)** 60° S to 60° N and **(b)** 20° S to 20° N. Monthly average values and annual averages are shown by thin and thick lines (connecting similarly-colored dots), respectively, for the pressure levels indicated in the plot legend.





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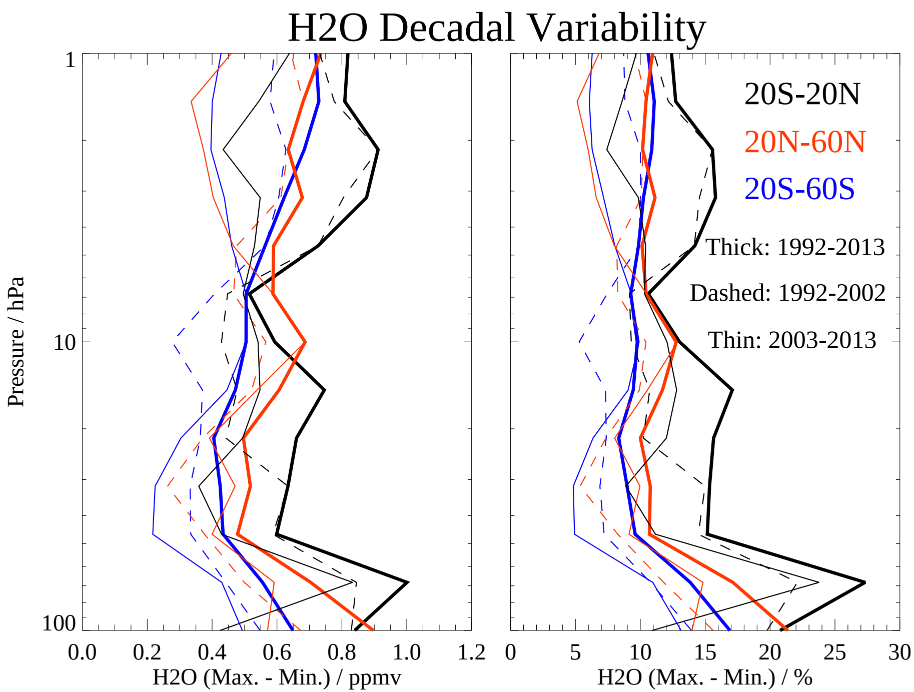


**Figure 16.** Stratospheric water vapor variability on decadal timescales for 1992 through 2013 (thick lines) for tropical (20° S–20° N in black) and mid-latitude (20–60° N in red and 20–60° S in blue) zonal means, based on the GOZCARDS merged $H_2O$ data record. The variability is expressed here as the difference between maximum and minimum annual average abundances, from 100 to 1 hPa, in ppmv (left panel) and percent (right panel). The 22 year period is broken up into two 11 year periods to illustrate how the variability changes from the 1st period (dashed lines) to the 2nd period (thin solid lines).





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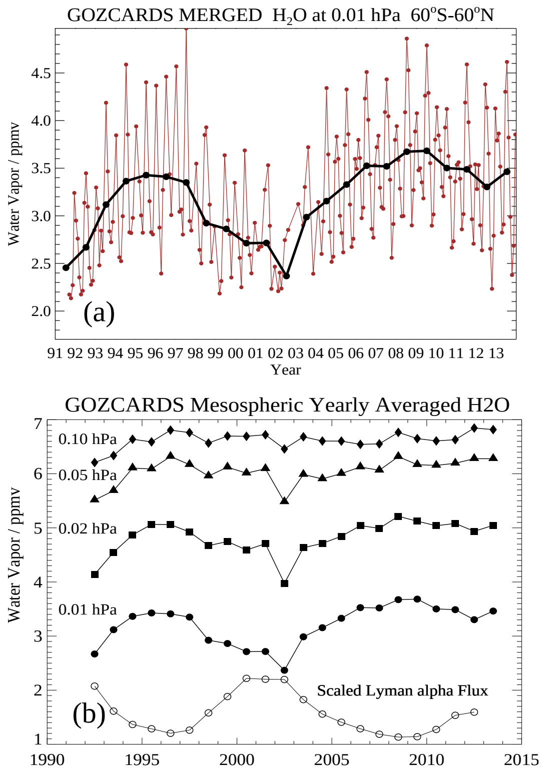


**Figure 17. (a)** Variations in upper mesospheric (0.01 hPa) water vapor mixing ratios averaged from 60° S to 60° N for October 1991 through December 2013, based on the GOZCARDS merged $H_2O$ data records. Monthly average values and annual averages are shown by connected brown dots and connected black dots, respectively. **(b)** GOZCARDS merged $H_2O$ annual averages (connected filled symbols) from 60° S to 60° N for 1992 through 2013 at pressure levels between 0.1 and 0.01 hPa. A time series of annually-averaged Lyman $\alpha$ solar flux values (open circles), scaled to arbitrary units, is also displayed (see text).





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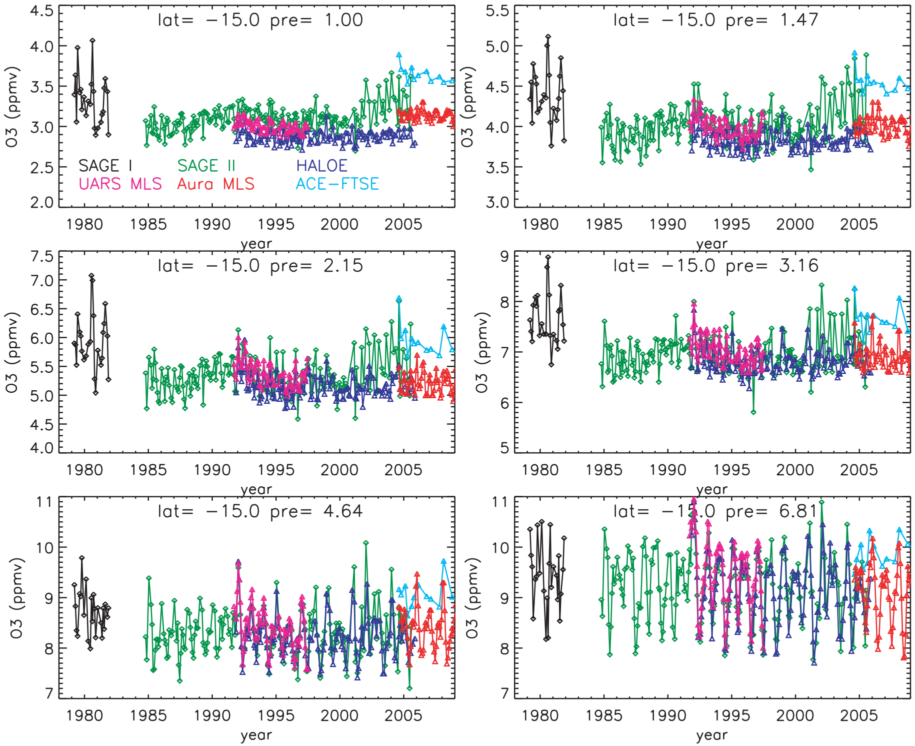


**Figure 18.** Time series of monthly zonal mean $O_3$ for 10° S–20° S between 1 and 6.8 hPa (with pressure values given by "pre") from SAGE I, SAGE II, HALOE, UARS MLS, Aura MLS, and ACE-FTS, all color-coded following the legend in top left panel.





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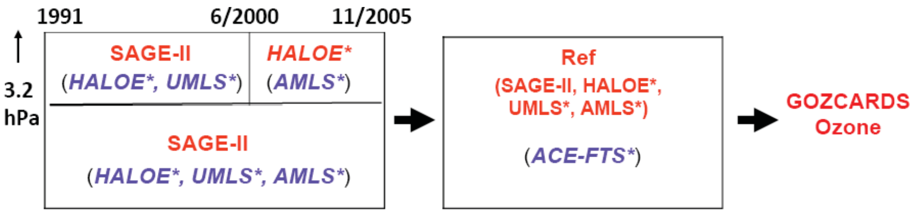


**Figure 19.** Schematic diagram describing the creation of the merged GOZCARDS monthly zonal mean ozone data record from various satellite datasets. Instruments represented in red inside the boxes are used as a reference. Instruments whose measurements have already been adjusted to a reference are indicated with a "$*$" superscript. AMLS refers to Aura MLS and UMLS to UARS MLS. See text for more details.



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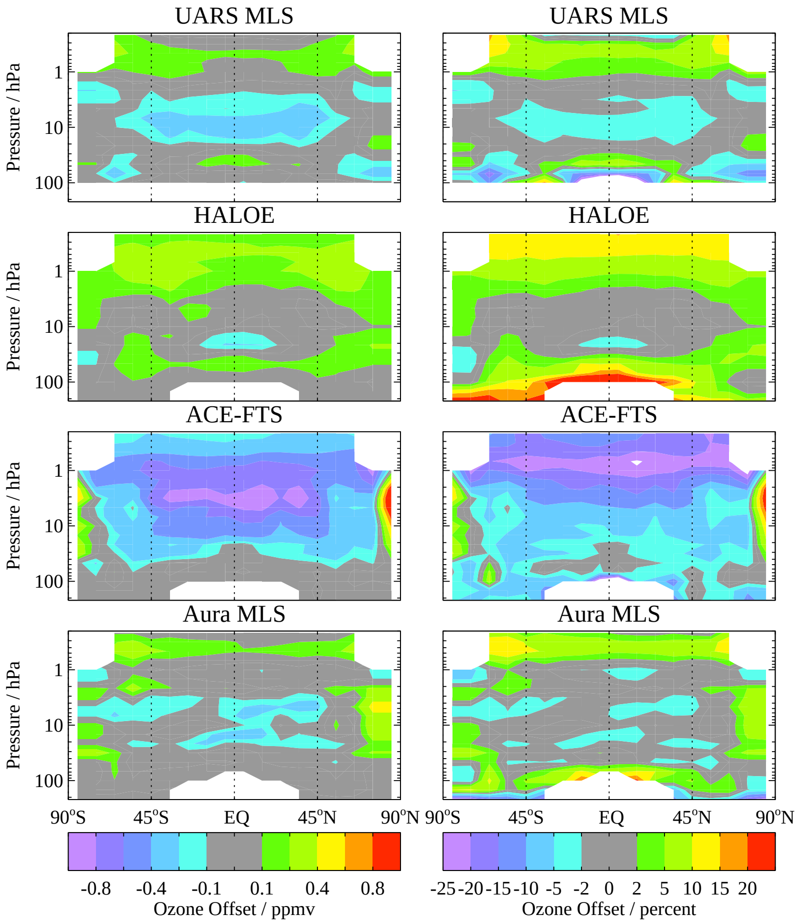


**Figure 20.** Offsets applied to the $O_3$ source datasets, similar to Fig. 2 for HCl.





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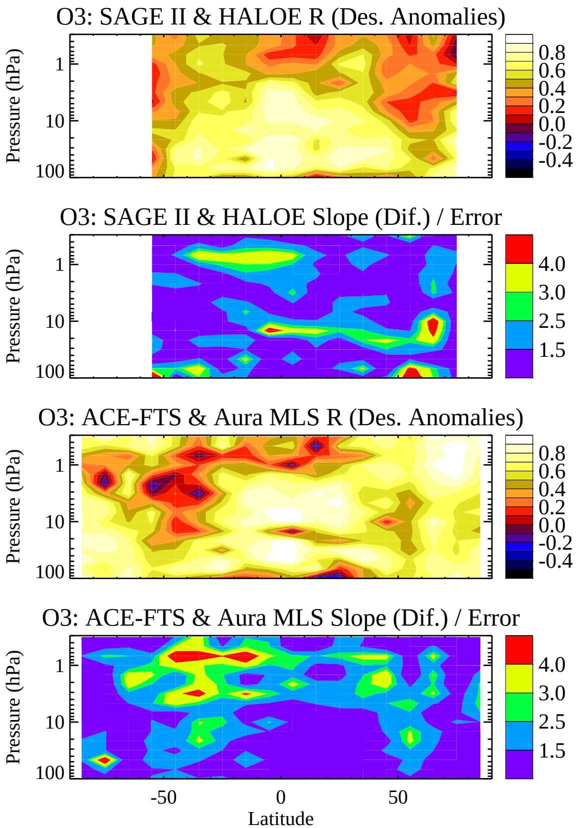


**Figure 21.** Latitude/pressure contours of time series diagnostics for $O_3$ from Aura MLS and ACE-FTS; this is similar to Fig. 4 for HCl. The correlation coefficients ($R$ values) and slope trend diagnostics are provided for HALOE vs. SAGE II in the top two panels (for 1993–1999 as the trend issue for converted SAGE II data occurs after mid-2000 and to avoid Pinatubo-related data gaps before 1993) and for ACE-FTS vs. Aura MLS in the bottom two panels (for 2005–2009).





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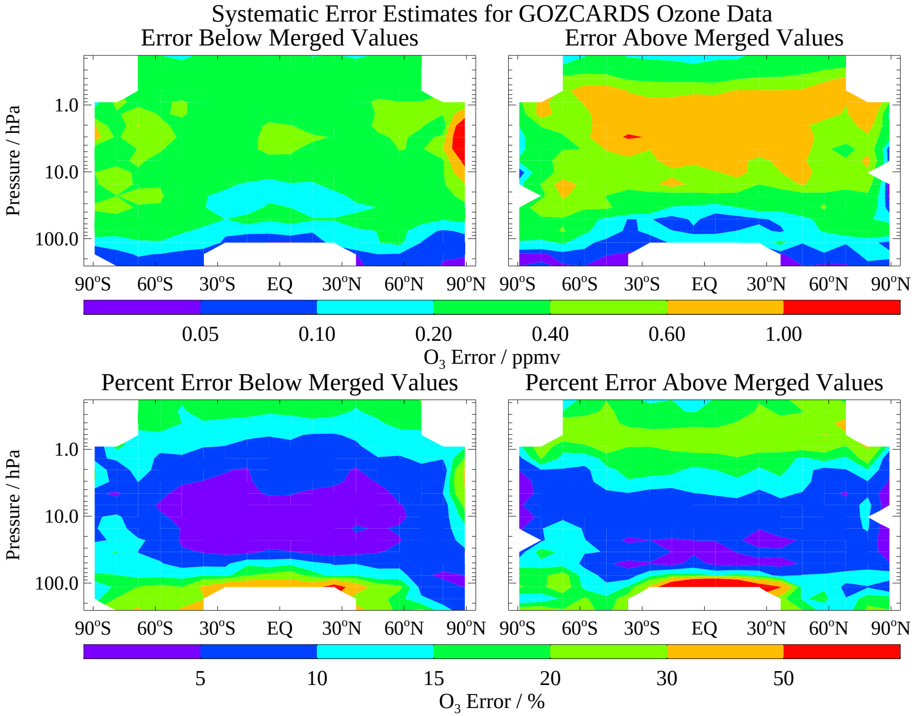


**Figure 22.** Systematic error estimates for GOZCARDS $O_3$ (similar to Fig. 6 for HCl).





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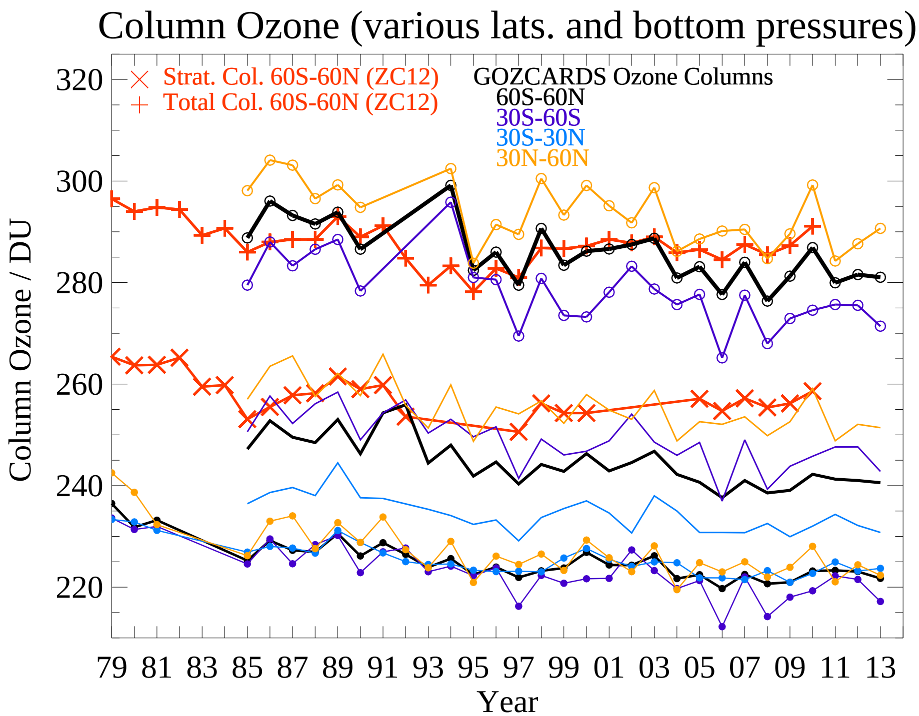


**Figure 23.** Column ozone values (DU) from ZC12 (in red) for 60° S–60° N and from GOZCARDS averages in different latitude bins (see legend). The connected dots are for GOZCARDS column ozone densities above 68 hPa from 1979 through 2013. The lines with no symbols and the connected open dots are also for GOZCARDS columns, but for values above 100 hPa and above 215 hPa, respectively, for 1985 through 2013; there are no blue open dots because of the lack of GOZCARDS merged ozone data in the tropics for pressures larger than 100 hPa, during 1985–2013.





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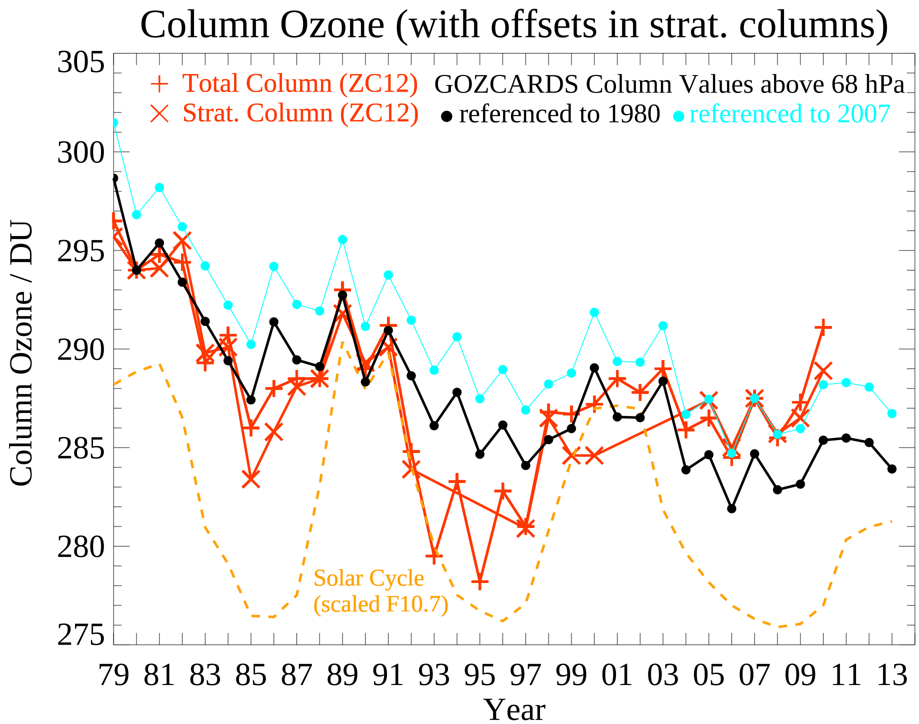


**Figure 24.** Near-global (60° S to 60° N) results for average column ozone (total and stratospheric, from Ziemke and Chandra, 2012) compared to GOZCARDS $O_3$ columns above 68 hPa. Stratospheric columns are offset in order to more easily compare relative variations vs. time; the black dots and red crosses are referenced to the 1980 total column values, while the cyan curves are referenced to 2007 to better illustrate the fits in the later years.





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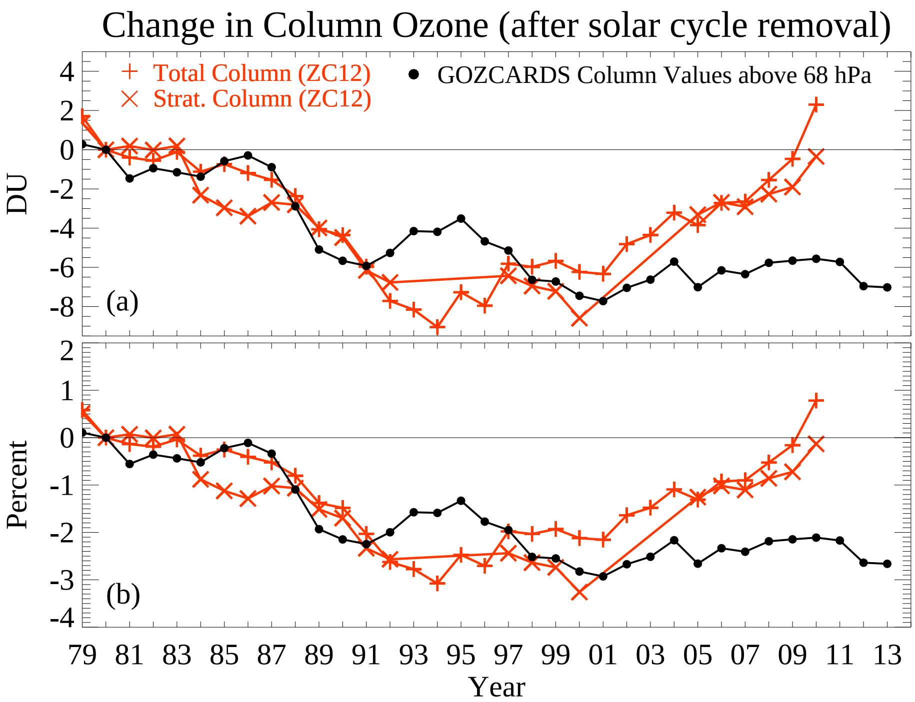


**Figure 25.** Change in column ozone after removal of solar cycle signal and with 3 year smoothing applied; **(a)** gives relative changes in DU, and **(b)** shows percent changes relative to the average GOZCARDS stratospheric columns in 1979. Black symbols are from the GOZCARDS column values above 68 hPa, averaged over 60° S–60° N. Red symbols, for comparison, are from the ZC12 data over the same latitude range (after removal of the fitted solar cycle signal).





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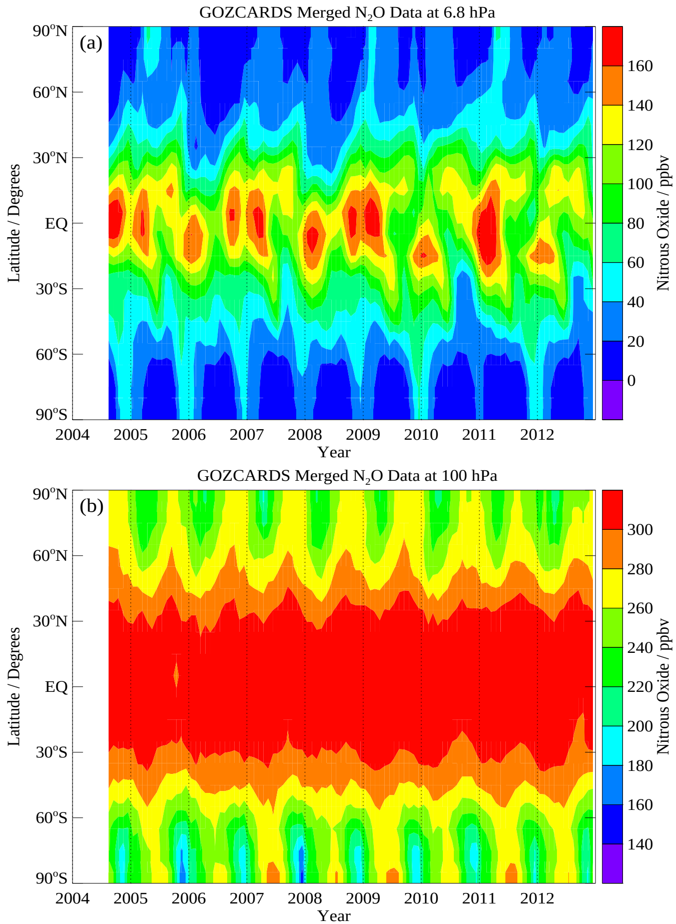


**Figure 26.** Time evolution (August 2004 through 2012) vs. latitude of GOZCARDS merged $N_2O$ (ppbv) at **(a)** 6.8 hPa and **(b)** 100 hPa.





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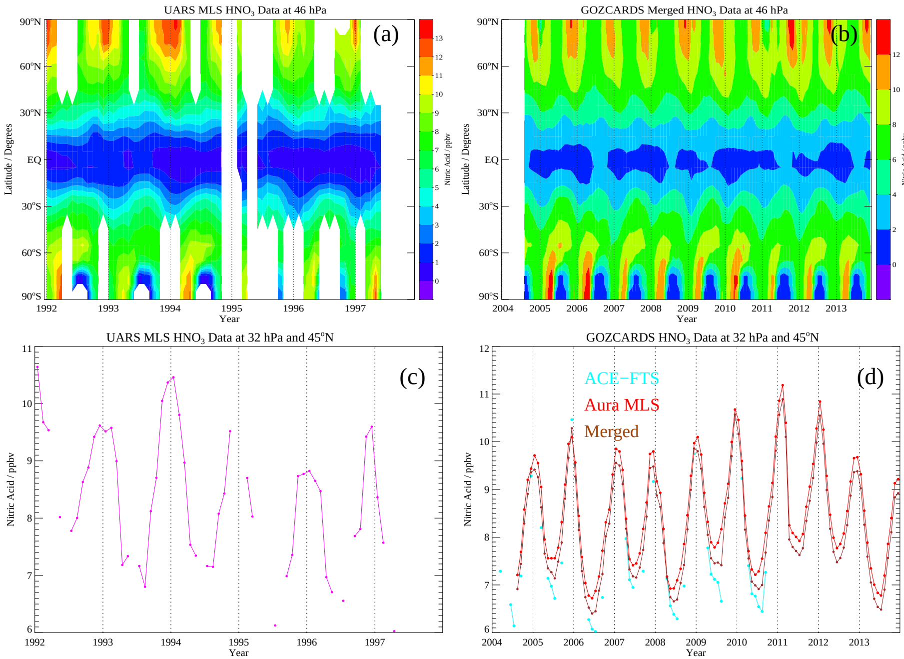


**Figure 27.** Sample results display the time evolution of satellite-retrieved $HNO_3$ (ppbv) for two different periods, 1992–1997 in (**a** and **c**) vs. 2004–2013 in (**b** and **d**). (**a** and **b**) are contour plots at 46 hPa from UARS MLS global data and the merged GOZCARDS global data after 2004, respectively; (**c** and **d**) show time series at 32 hPa and for the 40–50° N latitude bin, with **(a)** from UARS MLS data, and **(d)** from ACE-FTS, Aura MLS, and the merged combination (between the two source data sets).

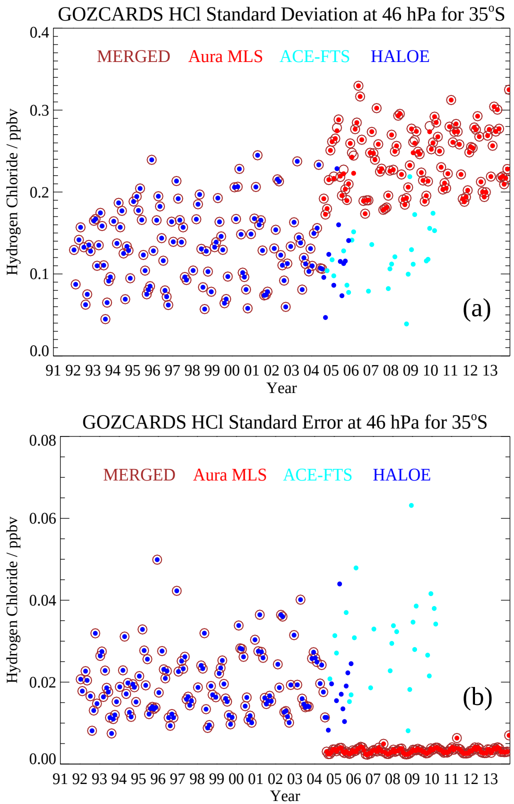


**Figure A1.** Illustration of the SD (in **a**) and standard errors (in **b**) for monthly mean GOZCARDS HCl (source and merged records) at 46 hPa for 30–40° S. Source data from HALOE, Aura MLS, and ACE-FTS are given by the filled colored dots (see legend); each SD is simply obtained from the range of values measured during the month. The large open brown circles give SD for the merged HCl product; this Appendix provides the formulae to calculate these quantities.





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