

Editor Decision (revised manuscript):

Reconsider after minor revisions (Editor review) (14 Aug 2015)

- by Dr. Michel Van Roozendael.

Comments to the Author:

The main criticism on this paper was related to its size. I think that the authors made a good job addressing this issue by removing unnecessary or duplicated text/material. Also the number of figures has been reduced while maintaining the same level of information. However there is one point where I think the recommendations from both referees have not been followed. This concerns the ozone discussion in section 5.3. On this issue, I tend to agree with the referees. The comparison with Ziemke and Chandra is certainly outdated, and do not really make sense as such considering the fact that long-term column data sets of much better quality have been recently published. If the authors really want to keep the column comparison (which I can understand), I suggest they also include data from SBUV MOD V8.6. E.g. a comparison of Figure 3 in Frith et al. (2014, <http://onlinelibrary.wiley.com/doi/10.1002/2014JD021889/abstract>) and Fig.22 of this manuscript clearly suggests a much better agreement with GOZCARDS.

Author Response:

We have reconsidered this aspect of the column ozone comparisons and agree that the addition of data from the SBUV Merged Ozone Dataset (V8.6) helps to put the ZC12 results in perspective, and the SBUV results generally agree significantly better with GOZCARDS column data, for averages over 60S-60N. A new Figure is shown below (with modified caption) as our response. Brief text changes were also introduced, as shown below (Abstract for ozone section and ozone section itself). The full manuscript has been uploaded with the changes shown in detail (including a few very minor changes having to do with reference updates).

We thank the editor for his useful response and for the reconsideration of these minor changes.

Abstract

We describe the publicly available data 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 (H₂O), and ozone (O₃). This dataset is a global long-term stratospheric Earth System Data Record, 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 relative to SAGE II values by calculating average offsets versus SAGE II during measurement overlap periods, whereas for other species, the merging derives from an averaging procedure during 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.

The GOZCARDS HCl merged product comes from HALOE, ACE-FTS and lower stratospheric 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. On 6-8 yr timescales,

the rate of decrease peaked in 2004-2005 at about 1%/yr, and has since levelled off, at ~0.5%/yr. With a delay of 6-7 years, these changes roughly follow total surface chlorine, whose behavior versus 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. Recent short-term tendencies of lower stratospheric and column HCl vary substantially, with increases from 2005-2010 for northern mid-latitudes and deep tropics, but decreases (increases) after 2011 at northern (southern) mid-latitudes.

For H₂O, the GOZCARDS product covers both stratosphere and mesosphere, and the same instruments as for HCl are used, along with UARS MLS stratospheric H₂O data (1991-1993). We display seasonal to decadal-type variability in H₂O from 22 years of data. In the upper mesosphere, the anti-correlation between H₂O and solar flux is now clearly visible over two full solar cycles. Lower stratospheric tropical H₂O has exhibited two periods of increasing values, followed by fairly sharp drops (the well-documented 2000-2001 decrease and a recent drop 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, H₂O has decreased by ~5-10% in the lower stratosphere, but about a 10% increase is observed in the upper stratosphere and lower mesosphere. However, such tendencies may not represent longer-term trends.

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. Unlike the 2 to 3% increase in near-global column ozone after the late 1990s reported by some, GOZCARDS stratospheric column O₃ values do not show a recent upturn of more than 0.5 to 1%; **long-term interannual column ozone variations from GOZCARDS are generally in very good agreement with interannual changes in merged total column ozone (Version 8.6) data from SBUV instruments** ~~continuing studies of changes in global ozone profiles, as well as 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 N₂O and HNO₃.

5.3 GOZCARDS ozone sample results and discussion

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Here, we investigate ozone column results for the stratosphere, based on the global GOZCARDS data, **in light of other column ozone datasets, including the work by Ziemke and Chandra (2012)**, hereafter referenced as ZC12. These authors analyzed total column and stratospheric column data from satellites, and their analyses yielded a rather strong near-global (60°S-60°N) average ozone increase since 1998. Their stratospheric columns depend on the convective-cloud differential (CCD) method and use Total Ozone Mapping Spectrometer (TOMS) and Ozone

Monitoring Instrument (OMI) column data over convective clouds near the tropopause (see also Ziemke et al., 2005). In Fig. 22, we compare changes in 60°S-60°N ZC12 column ozone data (J. Ziemke, personal communication, 2013) to changes in GOZCARDS O₃ columns 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 SAGE I data and are linearly interpolated between 1981 and 1984, when no GOZCARDS source datasets exist. We observe that 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 occur because total columns capture a stronger decrease from levels below 68 hPa, not fully represented in GOZCARDS. Focusing on the late period (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 columns and stratospheric column data from the CCD technique. 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. The increase in ZC12 data from 1997 to 1998 is not matched very well by GOZCARDS; this is also true if we remove the 11-yr solar cycle from both datasets (not shown here), as done by ZC12. However, the interannual changes in GOZCARDS columns are in better agreement with near-global total column variations in the Merged Ozone (Version 8.6) Dataset obtained from the suite of SBUV instruments (McPeters et al., 2013, Frith et al., 2014), as shown in Fig. 22. Discrepancies between the GOZCARDS and SBUV column data are largest between 1992 and 1997; this could be related to the somewhat less robust SBUV datasets in this period (resulting from SBUV satellite orbits closer to the terminator, e.g., see Frith et al., 2014), and/or to some issues in this portion of the GOZCARDS ozone data record. Discrepancies between the various column results in Fig. 22 could also arise from differences in ozone column calculations or coverage because of different methodologies, grids, or sampling to properly determine near-global results. We note that recent analyses by Shepherd et al. (2014), who used a chemistry-

climate model constrained by meteorology to investigate causes of long-term total column O_3 variations, show a partial return, in 2010, towards 1980 ozone column values, but not nearly as much as implied by ZC12. Long-term halogen source gas reductions that have occurred since the mid-1990s should only lead to column ozone increases of a few DU since 1997 (Steinbrecht et al., 2011). ~~It is likely that the discrepancies seen here 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. Discrepancies could also arise from differences between the coverage or the meaning of ozone columns, because of different methodologies, grids or sampling to properly determine near-global results. A better consensus regarding the recovery of near-global ozone columns (and profile values) is desirable in the future.~~

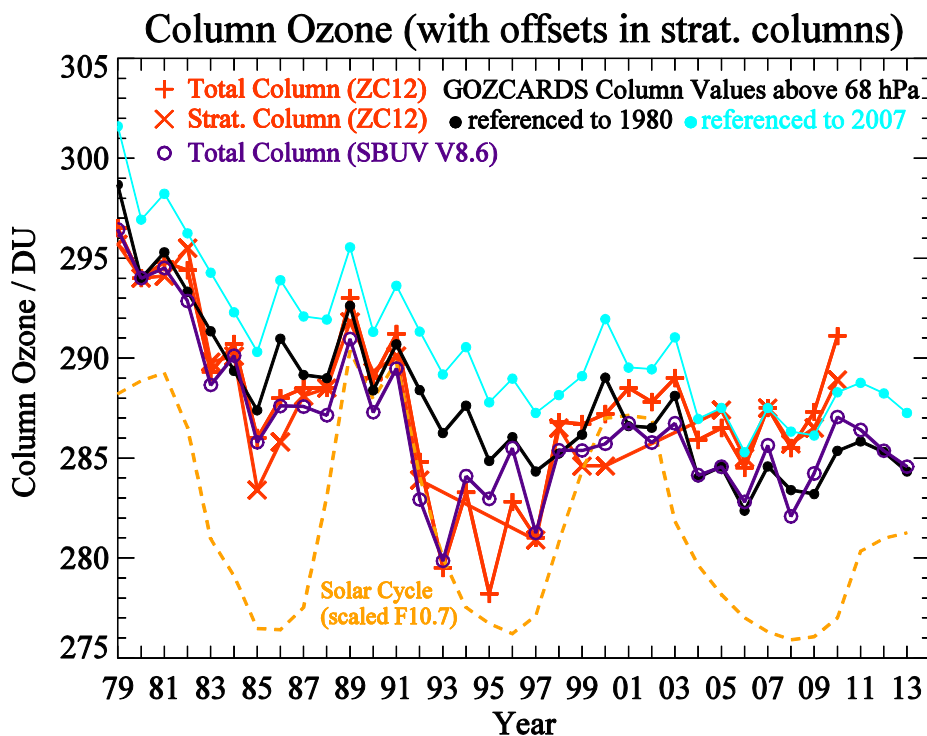


Fig. 22. Near-global (60°S to 60°N) results for average column ozone (total and stratospheric, from Ziemke and Chandra, 2012) compared to GOZCARDS O_3 columns above 68 hPa. Stratospheric columns are offset to better match the total column values, in order to more easily compare relative variations versus 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. Also shown (as purple open circles) are yearly-averaged total column data (60°S to 60°N) from the SBUV Merged Ozone (V8.6) Dataset (see text); these values were adjusted upward slightly (by 0.8 DU) to match the ZC12 total column values in 1980.